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ANNUAL PROGRESS REPORT
FOR PERIOD ENDING JUNE 30, 1969

IDAHO NUCLEAR CORPORATION
NATIONAL REACTOR TESTING STATION
IDAHO FALLS, IDAHO

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NUCLEAR TECHNOLOGY BRANCH ANNUAL PROGRESS REPORT FOR PERIOD ENDING JUNE 30, 1969

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FOREWORD

This document is a report of progress on technical programs of the Nuclear Technology Branch of Idaho Nuclear Corporation for the period ending June 30, 1969. It contains abstracts or expansions of abstracts of papers which have been published within the year. In these cases preprints or reprints of the articles are available. Results of work in progress are also reported; since this work is of a preliminary nature, the authors should be contacted before including any reference to these works in other publications.

On June 1, the research and development functions of the Analytical Chemistry Branch were transferred to the Nuclear Technology Branch. Since the Analytical Chemistry Section tenure with the Nuclear Technology Branch covers only one month of this annual report, their accomplishments will not be reported in this document this year. See IN-1316 for the report of their work. Sixteen members of the Analytical Chemistry Section were transferred to the Nuclear Technology Branch.

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A. NEUTRON INTERACTIONS WITH NUCLEI

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TOTAL NEUTRON CROSS SECTIONS OF ^{94}Nb AND ^{95}Nb

M. R. Serpa, T. E. Young, R. P. Schuman

The low energy neutron radiative capture cross sections of ^{94}Nb and ^{95}Nb are desired for thermal reactor calculations^[1]. Large amounts of both isotopes are produced from the fission of either ^{235}U or ^{239}Pu , and ^{95}Nb decays to an important fission product poison, ^{95}Mo , with a half-life of 35 days. In addition, the resonance structure of the odd-odd isotope, ^{94}Nb , is of interest because of the information it may furnish for the extension of nuclear systematics of odd-odd isotopes.

The total neutron cross sections of ^{94}Nb and ^{95}Nb have been measured using the MTR fast chopper. Samples were made by irradiating solid metallic ^{93}Nb in the MTR to produce a ^{94}Nb component of 4.1% and a ^{95}Nb component of 0.1%. The thickest sample available had original $1/n$ values of 185 barns/atom for ^{94}Nb and 7400 barns/atom for ^{95}Nb . The data have been analyzed and the resonance parameters in ^{94}Nb below 50 eV are shown in Table I. A transmission versus delay plot of the two large resonances in ^{94}Nb , at 11.63 and 22.87 eV, is shown in Figure 1. The solid curves were obtained using the parameters from Table I. A value of 105 ± 10 barns was calculated for the resonance absorption integral of ^{94}Nb using the parameters of Table I. The resonance at 11.63 eV contributes 86 barns of this amount.

Transmission measurements were made shortly after the sample was removed from the reactor and again after two half-lives decay of the ^{95}Nb component. No change was observed in the transmission spectrum below 50 eV, indicating the absence of any large resonances in ^{95}Nb in this energy region.

A resonance would have been observed in ^{95}Nb , in the region of 1 to 50 eV, if Γ_n° had been larger than that given by the following equation, $\Gamma_n^\circ = 0.0003E^2 + 0.02E + 0.03$. Resonances having values of Γ_n° greater than those quoted from the above equation would have appeared in the transmission data having a dip less than 0.95. If one assumes a level spacing in ^{95}Nb similar to that of ^{93}Nb or ^{94}Nb it has been shown that ^{95}Nb will have a resonance absorption integral of less than 150 barns. Preliminary analysis of thermal data shows no contribution from ^{95}Nb , which allows an upper limit of 100 barns to be placed on the thermal cross section of this isotope.

[1] A. B. Smith, Compilation of Requests for Nuclear Cross-Section Measurements, WASH-1078 (June 1967).

TABLE I
 RESONANCE PARAMETERS OF ^{94}Nb

E_{γ} (eV)	Γ_n^o (meV)	Γ_{γ} (meV)
11.63 ± 0.05	1.72 ± 0.20	162 ± 20
22.63 ± 0.20	0.20 ± 0.02	213 ± 25
[a] (10.74 ± 0.05)	0.010 ± 0.003	180)

[a] Assignment of this resonance to ^{94}Nb is uncertain. Γ_{γ} was assumed to be 180 meV by reference to Γ_{γ} 's observed for resonances at 10.63 and 22.87 eV.

Fig. 1 Resonances observed in ^{94}Nb for E_n less than 50 eV. These two resonances contribute 90 barns to the resonance integral and 4.4 barns to the thermal absorption cross section of ^{94}Nb .

THE TOTAL NEUTRON CROSS SECTION OF ^{170}Tm IN THE RESONANCE REGION

G. E. Stokes, R. P. Schuman, F. B. Simpson

The total neutron cross section of ^{170}Tm from 1 to 100 eV has been measured on the Materials Testing Reactor fast chopper. The sample material irradiated was natural thulium purchased in metallic form from Dresser Products, Great Barrington, Massachusetts. It was in the form of a foil 0.025 x 5.08 x 15.24 cm. The metal thulium sample has a quoted purity of 99.99%. An emission spectrograph analysis of the sample after it was received confirmed the purity. A list of the results of the emission spectrograph is shown in Table I.

Additional analysis for impurities was carried out by making transmission measurements with the MTR fast chopper on the natural thulium sample. These measurements revealed a tantalum impurity of 0.7 atom percent in the sample. The cross section data were corrected for this contaminant.

The metal foil was cut into strips 0.13 x 0.0025 cm, and these strips were placed around a spool to allow a uniform irradiation of the sample. The spool was slipped in a stainless steel container and the container was sealed for irradiation in the Engineering Test Reactor (ETR). The capsule was irradiated in the ETR at a flux of $\approx 4 \times 10^{14}$ neutrons/cm²-sec for 56 days. At the end of the irradiation the foils were removed from the capsule and prepared for a sample to be used on the MTR fast chopper. The irradiation converted approximately 8.2% of the ^{169}Tm to ^{170}Tm .

Transmission measurements were made on the sample using the MTR fast chopper system. The sample was not isotopically separated but it was run as a composite sample with all the irradiation products present in the sample. ^{169}Tm was used as a comparison or "open" in the ratio technique of determining the transmission of ^{170}Tm .

The data were analyzed using the Breit-Wigner single-level equation in conjunction with shape fitting and area analysis. A transmission plot of data taken soon after irradiation and data taken after nearly one half-life of decay is shown in Figures 1 and 2. The solid line represents a theoretical fit to the data obtained from the Breit-Wigner equation. The fit to the data of the run after 111 days of decay was generated using the parameters derived from the early run. This comparison is shown to illustrate the consistency in the identification and quantities of the various isotopes derived from the analysis of the data. There are a number of places where large resonances in the ^{169}Tm have removed all the neutrons from the beam. These places are left blank in the data plot. These were obscured areas that could not be analyzed. The analysis of the data above 20 eV was carried out by both area analysis and peak fitting. The peak fitting is governed by the Γ_n 's and the resolution function. The measured widths of the resonances above this energy are due mostly to the resolution function. If the resolution function is well known this technique of fitting to the peaks of the resonances in the region where the resolution function dominates the widths is equivalent to the area analysis with some limitations. The results from these two methods agreed within 5%. The advantage

of using the Breit-Wigner generated curve instead of ordinary area analysis is that it is easier to pick out two resonances that are close together and appear as one. This is shown in the resonance at 78.8 eV; this resonance should probably be two resonances. It has been quoted as one for this analysis. The parameters derived from the analysis are given in Table II. A cross section curve was generated from the derived parameters and is shown in Figure 3. This cross section was calculated using the Breit-Wigner single level equation and Doppler broadened for a temperature of 300°K.

Using an average Γ_n° [Γ_n° is the neutron width converted to its value at 1 eV ($\Gamma_n^\circ = \Gamma_n/\sqrt{E_0}$)] obtained from the analyzed resonances and calculating an average level spacing per spin state D from the observed resonances, the neutron strength function $\Gamma_n^\circ/D = 1.5 \pm 0.2 \times 10^{-4}$. A resonance absorption integral was calculated using the measured parameters below 100 eV and average parameters for the higher energies. The value for the resonance absorption integral obtained from these data is 460 ± 50 barns.

TABLE I

SPECTROCHEMICAL ANALYSIS
OF THULIUM SAMPLE

Element	Weight Percent
Thulium	Major part of sample
Copper	0.0001
Iron	0.01
Magnesium	0.001
Manganese	0.001
Silicon	0.001
Ytterbium	0.0005

TABLE II

^{170}Tm RESONANCE PARAMETERS

E_0 (eV)	Γ_n° (mV)	Γ_Y (mV)
2.82 ± 0.015	0.13 ± 0.02	104 ± 11
9.44 ± 0.02	0.32 ± 0.03	105 ± 10
12.38 ± 0.02	4.1 ± 0.5	110 ± 12
17.51 ± 0.05	2.6 ± 0.6	210 ± 60
20.38 ± 0.05	0.21 ± 0.03	80 ± 15
21.4 ± 0.1	0.62 ± 0.07	$100^{[a]}$
25.5 ± 0.1	0.30 ± 0.04	$100^{[a]}$
57.2 ± 0.2	2.0 ± 0.3	$100^{[a]}$
70.9 ± 0.3	0.63 ± 0.07	$100^{[a]}$
75.2 ± 0.3	1.5 ± 0.2	$100^{[a]}$
78.9 ± 0.3	4.5 ± 0.5	$100^{[a]}$
85.3 ± 0.4	5.1 ± 0.6	$100^{[a]}$
89.6 ± 0.4	1.1 ± 0.2	$100^{[a]}$
[a] Assumed values.

Fig. 1 A transmission versus neutron energy curve for an irradiated sample of ^{169}Tm from 1 to 22 eV. A comparison is shown between a fit at $t = 111$ days.

Fig. 2 A transmission versus neutron energy curve for an irradiated sample of ^{169}Tm from 22 to 100 eV. A comparison is shown between a fit at $t = 29$ days and $t = 111$ days.

Fig. 3 The total neutron cross section of ^{170}Tm from 1 to 90 eV. This curve was generated from the descriptive parameters of the isotope.

TOTAL NEUTRON CROSS SECTION PARAMETERS OF ^{236}U

R. A. Harlan

The total neutron cross section of ^{236}U has been measured from 0.01 to 1000 eV using the MTR fast chopper. The resonances have been analyzed up to 400 eV and are listed in Table I. These parameters are in good agreement with those quoted by Carlson in the October 1968 NCSAC Report.

TABLE I

RESONANCE PARAMETERS FOR ^{236}U (Preliminary Values)

E_0 (eV)	Γ_n° (meV) and Estimated Error	$\Gamma_Y^{[a]}$ (meV)
- 8.0 ^[b]	2.4 ^[b]	30 ^[b]
5.46	0.89 $\pm$ 0.05	25.6
29.8	0.104 $\pm$ 0.05	(25)
34.1	0.38 $\pm$ 0.04	(25)
43.9	1.71 $\pm$ 0.17	(25)
71.5	2.1 $\pm$ 0.2	(25)
86.4	3.1 $\pm$ 0.3	(25)
121	5.2 $\pm$ 0.6	(25)
125.5	1.2 $\pm$ 0.2	(25)
193	5.3 $\pm$ 0.6	(25)
212	6.9 $\pm$ 0.7	(25)
273	1.8 $\pm$ 0.5	(25)
298	4.0 $\pm$ 1.0	(25)
376	10.5 $\pm$ 2.5	(25)

[a] Values in parentheses were assumed for the area analysis.

[b] The negative energy resonance parameters were assumed to be those in BNL-325.

LOW ENERGY TOTAL CROSS SECTION OF ^{241}Pu

J. R. Smith, T. E. Young

The analytical fits[1] to the low energy absorption and fission data for ^{241}Pu gave the clearest picture then available of the behavior of these cross sections. This clarity is nevertheless far from adequate. The analytical fits indicate a very strong variation in η . For example, they predict a variation in η more than three times the variation observed in the manganese bath measurements at 0.0253 and 0.06 eV. Either the fission or the total cross section could be at fault. The availability of the metal foil samples used in the manganese bath η measurement made it possible to bypass the errors of water contaminant and particle size that might have been present in the previous measurements with oxide samples. The high purity of the metal samples also promised to make possible a better determination of the absolute value of σ_T at 0.0253 eV.

Measurements of the total cross section of ^{241}Pu were extended down in energy to 5.1×10^{-4} eV. The MTR crystal spectrometer used a mica crystal, in conjunction with a mechanical neutron filter, to achieve this low energy. Measurements were also made on the MTR fast chopper, the latter data extending down to 0.001 eV.

Four metal foils prepared for the manganese bath η measurement were the samples. These are the first metallic ^{241}Pu samples upon which total cross sections measurements have been made. The use of metal foils eliminates the problems of water contamination and small angle scattering that are encountered in the use of oxide samples. On the other hand there is an uncertainty in the $1/N$ values of the foils due to irregularities in the scissor-trimmed, crack-infested contours of the foils. Weights and thicknesses of the samples were measured before the foils were sealed in their aluminum containers, but an area or density determination is also needed for determination of cross sections from sample transmissions. The samples were X-rayed and area measurements made upon the images. The images are shown in Figure 1, along with photographs of the sealed samples. Areas were both measured directly using a planimeter and calculated using the average value of 12-diameter measurement. The average of the two area determinations was used. In the case of sample IV only the planimeter measurement was used, because of the severe irregularities in the contour of the foil.

Transmission measurements were made using all four samples on the crystal spectrometer and samples I and II only on the fast chopper. Below 0.01 eV only the thinnest sample (II) was used. On both instruments measurements were made several months apart as a check on the accuracy of the ^{241}Am contaminant correction. The corrected cross sections agreed within statistical errors.

The ^{241}Pu total cross section, including data from both spectrometers, is shown in Figure 2. Table I shows the determination of the 0.0253 eV value. For each data run made on each instrument using each sample, the value of $\sigma_T \sqrt{E}$ was averaged over 9 to 16 data points centered on 0.0253 eV. The average of the determination yields 1389 ± 15 barns.

The shape of the total cross section is very close to that indicated by analytical fit[1], so the low energy anomaly in ^{241}Pu persists. Additional measurements of σ_f and η as a function of energy will be required to eliminate the discrepancy.

[1] R. G. Fluharty, N. H. Marshall, and O. D. Simpson, Conf. on Neutron Cross Section Technology, Conf-660303, 2 985 (1966).

TABLE I
SUMMARY OF σ_T VALUES FOR ^{241}Pu AT 0.0253 eV

<u>Sample and Transmission</u>	<u>Crystal Spectrometer</u>			<u>Fast Chopper</u>	
	<u>Crystal Planes</u>	<u>1967</u>	<u>1968</u>	<u>1967</u>	<u>1968</u>
I	Be(10 $\bar{1}$ 1)	1379		1378	
0.21	Be(0002)	1375	1377		
II	Be(10 $\bar{1}$ 1)	1405		1394	1400
0.58	Be(0002)	1409	1413		
III	Be(0002)	1368	1374		
0.015					
IV	Be(0002)		1399		
0.35					
Average Value 1389 $\pm$ 15 b

Fig. 1 Samples used in the ^{241}Pu experiment. Below the photograph of the clad samples are X-ray images of the foils, showing their irregular contour. From left to right the samples are: IV, II, I, III.

Fig. 2 Total cross section ($\sigma_T \sqrt{E}$) below 0.1 eV for ^{241}Pu , as measured by the crystal spectrometer and fast chopper using the thinnest ^{241}Pu sample only. The thickness of this sample (Foil II) was 2680 barns/atom.

RESONANCE PARAMETER DETERMINATIONS OF ^{243}Am FROM 0.04 TO 7 eV

J. R. Berreth, F. B. Simpson

Americium 243 is a stepping stone in the production of certain trans-plutonium elements. Knowledge of its cross section is, therefore, important. Transmission measurements as a function of neutron energy have been made on the MTR fast chopper from 0.01 to over 100 eV. The transmission data have been analyzed from 0.4 to 7 eV and resonance parameters assigned to observed resonances in this energy region.

Figure 1 shows a Breit-Wigner single level fit to the transmission data up to 7 eV for a sample containing 279.3 b/a Am. Shown in Figure 2 is a Breit-Wigner single level fit to the transmission data for three of the lower energy resonances taken with three different thicknesses of Americium. A list of the energies of the resonances observed and the resonance parameters determined by fitting the transmission data is shown in Table I. Analysis of the data from 0.01 to 100 eV is continuing.

Fig. 1 Transmission as a function of time of flight for ^{243}Am . A sample inverse thickness of 279.3 b/a was used. The solid line is the single level fit to the data using the parameters in Table I.

TABLE I

RESONANCE PARAMETERS FROM
0.4 TO 7 eV FOR ^{243}Am

E_o (eV)	Γ_n^o meV	Γ_γ meV
0.420	0.0013	39
0.983	0.0151	40
1.356	0.950	43
1.746	0.181	40
3.144	0.0072	45
3.433	0.125	48
3.849	0.0064	46
5.128	0.127	52
6.560	0.240	70
7.076	0.0264	41

Fig. 2 Transmission as a function of time of flight for ^{243}Am . Three sample inverse thicknesses of 2793, 1288, and 6018 b/a were used. The solid line is a single level fit to the data using the parameters in Table I.

TOTAL NEUTRON CROSS SECTIONS OF $^{243,245,246}\text{Cm}$

J. R. Berreth, F. B. Simpson

A knowledge of the cross sections of the curium isotopes is important in the production of ^{252}Cf . The total neutron cross sections of $^{243,244,245,246}\text{Cm}$ as a function of neutron energy using the Materials Testing Reactor fast chopper have been measured from 0.01 to 100 eV using a 20 meter flight path and resolutions from 0.10 to 1.8 $\mu\text{sec}/\text{meter}$.

Three thick samples of different curium isotopic contents were prepared, each containing approximately 2×10^{22} atoms/cm² ^{244}Cm . These samples varied from 1×10^{19} to 8×10^{20} atoms/cm for the various curium isotopes. Samples of approximately 1/6 the above thickness were also prepared. Table I shows the percentage isotopic content of the three samples. The values shown under the column "determined" are the amount determined to ideally fit the data for all three samples.

A preliminary evaluation of the data has been made from 0.01 to 10 eV. The neutron transmission curves for the three thick samples from 1 to 10 eV are shown in Figure 1. From these data three resonances at 1.969, 4.696, and 9.168 eV have been identified as belonging to ^{245}Cm , six resonances at 2.263, 3.098, 3.341, 3.748, 5.411, and 6.005 eV to ^{243}Cm , and one resonance each to ^{246}Cm at 4.316 eV and ^{244}Cm at 7.678 eV. The resonances assigned to ^{243}Am and ^{240}Pu are based upon prior measurements and their resonance parameters are assumed known. The broad resonances of $^{243,245}\text{Cm}$ indicate the presence of a fission component. The Reich-Moore multilevel formula in conjunction with an automatic fitting program was used in determining the resonance parameters by a theoretical fit to the composite transmission data.

Resonance parameters have been determined for the $^{243,244,245,246}\text{Cm}$ resonances from these data. The solid line in Figure 1 shows the theoretical fit to the data as determined from the measured neutron resonance parameters. It was expedient in fitting the data to postulate two channels of fission for the 2.263 and 6.005 eV ^{243}Cm resonances. The error in the fit just above 1 eV in Sample No. 1 could be due to the effect of a negative energy resonance in ^{245}Cm which is not accounted for here.

Table II lists the resonance parameters determined from the fit to the transmission data. A neutron capture width of 40 millivolts was assumed in fitting the $^{243,245}\text{Cm}$ data. The sign given to the parameters for each fission resonance is not unique but the interference between resonances of the particular isotope would appear to indicate the proposed sign assignments. The parameters as determined appear to be accurate to within 10%. In the case of the parameters for resonances with two channels of fission, Γ_{f1} and Γ_{f2} may vary considerably from the assigned values, although the sum of the two fission channels should accurately describe the total fission width. Figure 2 shows the total, fission, capture, and scattering cross sections as a function of neutron energy from 1.0 to 10 eV for ^{243}Cm as calculated from the resonance parameters. Figure 3 shows the same cross sections for ^{245}Cm . For these data the potential scattering cross section was calculated to be 10.3 barns.

Resonance absorption integrals were calculated using the measured resonance parameters in the energy region from 1 to 10 eV. In the case of ^{245}Cm , the resonance absorption integral includes the effect of a negative energy at -1.0 eV. The contribution above 10 eV was determined from an average of the measured parameters. This may or may not be a good representation for the average effect above 10 eV because of the small number of known resonances. However, the contribution above 10 eV is only 15% for ^{245}Cm and 30% for ^{243}Cm . The resonance absorption integrals as determined by the above method are 805 ± 80 for ^{245}Cm and 2270 ± 200 for ^{243}Cm . Plans are to continue the analysis of the data above 10 eV.

Determination of the thermal cross sections for these isotopes is a more difficult problem. One in essence has 6 unknowns and 5 equations. Some assumptions can be made, though, which should give reasonable cross sections at thermal for $^{244,245}\text{Cm}$. The ^{246}Cm cross section is small as is its content in the samples. If one assumes a σ_{tot} of 26 b for ^{246}Cm at thermal, a small error in the product of $n\sigma$ for ^{246}Cm compared to the other isotopes is a negligible error in determining the cross sections for $^{244,245}\text{Cm}$. A more serious error is introduced due to a bias required to fit the data from 1 to 10 eV. This bias may be due to an error in ϕ potential or, more likely, a contaminant such as water present in the oxide material. At low energies, this bias may have a significant effect in determining the thermal cross section of $^{243,245}\text{Cm}$ in particular, but also on ^{244}Cm due to its small section.

Attempts to measure the thermal cross sections were made by solving a series of simultaneous equations. Five samples are available for determining the unknowns present. It was assumed the bias observed in the 1 to 10 eV data was due to H_2O but the amount was unknown. Figure 4 shows the determined total cross section for ^{245}Cm from 0.01 to 1 eV. From the appearance of this curve, one must postulate a resonance perhaps at slightly negative energy, since the resonance parameter data from 1 to 10 eV account for less than half of the thermal cross section. The cross section of 2900 barns determined for ^{245}Cm at thermal energy should be accurate to $\pm 15\%$.

The cross section curve for ^{244}Cm (Figure 5) appears to closely match a $1/v$ spectrum in the low energy range. A preliminary thermal value obtained for ^{244}Cm is 23.5 ± 3 barns. Due to the large ^{245}Cm content which is present with the sample containing ^{243}Cm , an internally consistent number for the thermal cross section of ^{243}Cm has not yet been arrived at.

TABLE I

PERCENTAGE COMPOSITION OF CURIUM SAMPLES

Sample No.	^{244}Cm %		^{245}Cm %		^{246}Cm %	
	<u>Stated</u>	<u>Determined</u>	<u>Stated</u>	<u>Determined</u>	<u>Stated</u>	<u>Determined</u>
1	93.69	--	4.07	--	0.666	--
2	94.61	--	1.79	1.62	3.51	--
3	95.11	--	0.720	--	3.97	--

	^{243}Cm %		^{240}Pu %		^{243}Am %	
	<u>Stated</u>	<u>Determined</u>	<u>Stated</u>	<u>Determined</u>	<u>Stated</u>	<u>Determined</u>
1	1.517	--	0.40	0.64	0.02	--
2	0.02	0.053	1.30	1.65	0.23	0.30
3	0.024	0.054	1.07	1.79	0.15	0.18

TABLE II

RESONANCE PARAMETERS FROM 1 TO 10 eV FOR
 ^{243}Cm , ^{244}Cm , ^{245}Cm , ^{246}Cm

Isotope	E_o (eV)	Γ_n^o meV	Γ_{f1} meV	Γ_{f2} meV	Γ_γ meV
^{243}Cm	2.263	1.335	250	300	40[a]
	3.098	0.353	-115	---	40[a]
	3.341	0.144	80	---	40[a]
	3.748	0.334	-90	---	40[a]
	5.411	0.359	125	---	40[a]
	6.005	1.029	-150	750	40[a]
^{245}Cm	1.969	0.170	-230	---	40[a]
	4.696	0.902	350	---	40[a]
	9.168	0.240	-110	---	40[a]
^{246}Cm	4.316	0.153	---	---	35
^{244}Cm	7.678	3.57	---	---	35

[a] Assumed value.

Fig. 1. Transmission curves of the three different curium samples from 1 to 10 eV. Solid line shows the theoretical fit to the curves.

Fig. 2. Cross section curves from 1 to 10 eV for ^{243}Cm as calculated from the resonance parameters.

Fig. 3. Cross section curves from 1 to 10 eV for ^{245}Cm as calculated from the resonance parameters.

Fig. 4. Total cross section of ^{245}Cm from 0.01 to 1 eV.

Fig. 5. Total cross section of ^{244}Cm from 0.01 to 1 eV.

ETA MEASUREMENTS OF ^{233}U AT 0.1 eV

J. R. Smith, S. D. Reeder

The development of the Molten Salt Breeder Reactor depends strongly upon the behavior of eta for ^{233}U in the vicinity of 0.1 eV. The importance of eta is such that concern was expressed over even the small discrepancies between the old MTR crystal spectrometer data and the newer data of Weston et al[1]. Figure 1, showing a comparison of the data, is from the Weston report. It was felt that the best way to shed further light on the problem would be to make a series of manganese bath measurements of eta in the region of interest. The corrections to this type of experiment vary much more slowly with energy than do those for the direct neutron counting experiment. For example, the counting efficiency is constant for the manganese bath, as both the low energy and fission neutrons are absorbed in thick absorbers and counted with essentially the same efficiency, and the effects of second order in the Bragg beam are less troublesome.

Two problems faced the use of the manganese bath for these higher energy measurements. The beam strength falls off as the energy increases and the background would become unbearable if the tank were to get into the path of the main beam. To minimize these problems we decided to make relative measurements with the small (21-inch diameter) manganese bath tank, normalizing these to the previous results using the large tank. To minimize the considerable time and effort expended to move and align the tank when changing energy points, we constructed a short spectrometer arm. This arm is not rigidly connected to the tank, and the two are moved separately between energy points. A dowel pin through facing plates on the arm and tank establishes the realignment unequivocally. This system made it possible to change tank positions while a sample was being counted, with no loss of effective running time for the experiment. When the dowel pin was inserted the alignment of the sample in the beam was checked and found to be true.

There was time for measurement of only one energy point in addition to a 0.06 eV normalization point before the long reactor shutdown prior to the Phoenix experiment. Accordingly, data were taken at 0.06 and 0.095 eV, each set consisting of three runs each of sample and open beam data and two background determinations. The mechanical neutron filter was not used, partly because its mount for use on the short arm was not ready, but mainly because measurement and calculation indicated that the second order fraction in both these points was low enough to have negligible effect on the experiment.

The results of the measurement showed that η at 0.095 eV is about half a percent lower than it is at 0.060 eV, as shown by the square data points in Figure 1. This includes a correction for the difference in sample transmission between the two points. The error on this relative measurement is about 0.4%, evaluated at a 75% confidence level.

[1] L. W. Weston et al, Neutron Fission and Capture Cross Section Measurements for ^{233}U in the Energy Region 0.02 to 1 eV, ORNL-TM-2353 (February 1, 1969).

Fig. 1 η for ^{233}U . The figure is from Weston et al^[1] showing comparison of MTR and ORNL data. The square points represent the recent MTR manganese bath comparison of N at 0.06 and 0.095 eV, normalized to the ENDF/B curve,

η MEASUREMENTS OF ^{239}Pu AND ^{235}U AT 2 keV

J. R. Smith, S. D. Reeder

The manganese bath has been used to measure the value of η for ^{239}Pu and ^{235}U in the 2 keV beam of the MTR. The techniques and equipment used were the same as those previously used in making η measurements on the four fissile nuclei at low energies.

The manganese bath η experiment is basically a comparison of ^{56}Mn activities produced in a totally absorbing solution of MnSO_4 . When the incident beam is allowed to enter the solution directly it produces a ^{56}Mn activity

proportional to the neutron beam intensity. When the beam strikes a fissile sample the resulting fission neutrons are absorbed in the solution and produce a ^{56}Mn activity proportional to the number of fission neutrons produced. As the eta measurement depends only upon the ratio of these activities, it is not necessary to have an accurate calibration of the sampling and counting efficiencies of the system. If the system is calibrated, however, it is possible to determine accurately the beam strength of the irradiation facility. A preliminary calibration of the counting system made it possible to determine the 2 keV beam strengths shown in Table I for the available collimation systems. These figures have an accuracy of approximately 5%. As there was no absolute cross section measurement or standard detector calibration directly associated with and depending upon these beam measurements, there was no point in determining the beam strength to higher accuracy at that time. The beam strength varies several percent over the course of a reactor cycle.

Included in the beam strength table are values for the non-2-keV neutron background in the beam. This background has been shown from proton-recoil counter studies to be due to several minor neutron windows in the vicinity of 350 keV. Discrimination against this background is accomplished through the use of a filter of cerium and manganese, which removes essentially all of the 2 keV neutrons from the beam. The transmission of this filter to the higher energy neutrons was determined in separate experiments to be 0.42. Application of this factor to the ratio of solution activities observed with and without the cerium-manganese filter in the beam allowed the high energy fraction of the beam to be determined as 38% of the total intensity.

With the beam collimated to 1/2-inch diameter to confine it to the fissile sample area, the solution counting rates were greater than those encountered at 0.06 eV. Monitor foil counting rate represented the statistical limitation to the experiment. In order to have a suitable monitor foil activity, it was necessary to use a thicker foil than that used at the lower energies. The monitor used at 2 keV had a transmission of about 85%.

A fundamental assumption in the manganese bath eta measurement is that the fissile sample absorbs all the neutrons in the beam. At 0.025 eV this assumption is close enough to the truth that corrections for sample transmission and neutron scattering are small. At 2 keV both corrections are considerable. The sample transmission is easy enough to measure, but the scattering cross section is difficult. This correction remains the limiting factor in the accuracy of the experiment.

The experimental portion of the experiment was time-consuming but straight-forward. Two ^{239}Pu sample thicknesses were used. These were found to have transmissions of 0.802 and 0.396. The transmissions of individual foils were measured, as well as those of combinations of foils. The indicated total cross section varied from about 22 barns for the thinnest sample used to about 16 barns for the thickest. These measurements are in good agreement with a set of measurements made by O. D. Simpson on a separate set of ^{239}Pu samples. The decrease is due to self-shielding of the resonances within the sample. This variation poses additional complications to the scattering corrections. The determining parameter in these corrections is the ratio τ_s/τ_t . There are uncertainties in both the value of this ratio at 2 keV and in its variation with sample thickness. The ultimate solution will require data derived from a scattering experiment to be done in the 2 keV beam. Mean-

while a relatively high error must be assigned because of the differences indicated between several plausible scattering assumptions.

The experimental results showed that irradiations of the thicker sample yielded ^{56}Mn solution activities 1.115 times that of the open beam. For the thinner sample this ratio was 1.024. Even the thin sample is expected to have some self-shielding effect, and ideally the experiment should have used a still thinner sample, such as a single 0.020-inch plutonium foil. Such a foil, however, would produce only about a half percent effect, which could not be determined with sufficient accuracy to be definitive.

Table II shows values of eta and alpha derived from the experimental solution activity ratios under various assumptions concerning the scattering. Most evaluated data files show a scattering cross section of about 13.5 barns for ^{239}Pu near 2 keV. The first data reduction assumed this value to be constant through the sample, allowing the ratio σ_s/σ_t to rise from 0.62 to 0.82 as the total cross section decreased. The second assumption was that σ_s/σ_t was constant at the lower of these values, while the third kept σ_s/σ_t constant at 0.82, the higher of the two limits. The fourth assumption was that the scattering cross section consisted of a constant term due to a potential scattering cross section of 10.5 barns plus a variable fraction of the 3 barn difference between σ_p and the 13.5 barn value previously used.

The results of calculations under these various assumptions are indicated in Figure 1, where alpha is plotted as a function of sample thickness. It can be seen that three of the four scattering assumptions lead to values of alpha in fair agreement with each other. The maverick points are given by the

assumption $\frac{\sigma_s}{\sigma_t} = 0.82$, which would have the thin sample scattering cross

section as high as 18 barns. This seems quite an unlikely value. It is felt that the most likely values are given by assumption number 4. A line connecting the two points for this assumption extrapolates to a value $\alpha = 1.3$ at zero sample thickness. This is in good agreement with a set of preliminary measurements made by O. D. Simpson and described in this report, page 38. The Simpson points are also shown on the curve.

As a comparison measurement the value of eta for ^{235}U was measured on a sample whose transmission was 0.74. Again a question arises as to the correct scattering cross section to use. There seems to be less sample thickness effect in ^{235}U than in ^{239}Pu , due no doubt to the smaller value of $\bar{\Gamma}_n^0$ for ^{235}U . Transmission measurements on various samples showed the sample thickness effect to be smaller for ^{235}U . Again there is a question as to what is the proper scattering cross section to assume for ^{235}U . Three sets of 2 keV cross section values were tried. One of these was a set of zero-thickness values assembled by O. D. Simpson, partly measured and partly evaluated. The second set is the current ENDF set, which adopted J. J. Schmidt's evaluation, vintage 1962. The third set was from Schmidt's 1968 reevaluation. These three values of σ_s/σ_t , combined with the manganese bath data, lead to the values of eta and alpha shown in Table III. The results are in reasonably good agreement with both measured and evaluated values. This agreement supports the determination for ^{239}Pu .

The values determined here are provisional, pending the acquisition of better values of σ_s/σ_t plus an understanding of sample thickness variations in alpha and σ_s/σ_t appropriate to the 2 keV beam.

TABLE I
SCANDIUM-FILTERED BEAM INTENSITIES

Total Beam Intensities (Per Second)			
<u>Collimator Insert</u>	<u>2 keV Neutrons</u>	<u>Fast Bkg. Neutrons (~ 350 keV)</u>	<u>Total</u>
1/2 in.	1.84×10^6	1.13×10^6	2.97×10^6
3/4 in.	4.67×10^6	2.88×10^6	7.55×10^6
None (Max. Beam)	7.66×10^6	4.72×10^6	1.24×10^7

Fig. 1 Alpha for ^{239}Pu measured as a function of sample thickness in the 2 keV beam. The circles represent the Mn bath data for two sample thicknesses, the data having been reduced under four different assumptions as to the behavior of the scattering cross section. The diamond and triangles are from measurements made by O. D. Simpson and described in another section of this report.

TABLE II
 η AND α VALUES FOR ^{239}Pu

Mn Bath	Thick Sample:		Thin Sample:	
	$T_{[a]} = 0.396$ $\eta = 1.115$		$T_{[a]} = 0.802$ $\eta = 1.024$	
Scattering Assumption	<u>Eta</u>	<u>Alpha</u> ^[a]	<u>Eta</u>	<u>Alpha</u> ^[a]
(1) $\sigma_s = 13.5 \text{ b.}$ $(0.62 \leq \sigma_s/\sigma_t \leq 0.82)$	1.62	0.77	1.32	1.18
(2) $\sigma_s/\sigma_t = 0.62$	1.46	0.97	1.27	1.26
(3) $\sigma_s/\sigma_t = 0.82$	2.04	0.41	1.61	0.79
(4) $\sigma_s = 10.5 + f_{[a]}^3$	1.50	0.92	1.29	1.23
Avg. of 4:		0.77		1.12
Avg. wt.% #3:		0.89		1.22

[a] Assuming $\bar{v} = 2.87$.

TABLE III
 η AND α FOR ^{235}U

$T = 0.738$ $\eta^* = 1.0$		
<u>Correction Data Used</u>	<u>η</u>	<u>α</u>
KFK 120, 1968	1.65	0.48
ENDF (Schmidt 1962)	1.69	0.44
ODS set	1.81	0.35

RESONANCE ENERGY AND PARAMETER ASSIGNMENTS FOR

^{147}Pm AND ^{147}Sm

R. L. Tromp, J. W. Coddington, F. B. Simpson

Neutron resonance parameter determinations for ^{147}Pm (and decay product ^{147}Sm) have been extended to cover the range between 60 and 250 eV. Details of sample preparation, MTR fast chopper operation, and data analysis of the lower energy range were described previously[1].

Inclusion and consideration of ^{147}Sm in the ^{147}Pm resonance analysis (which is of principal interest) was necessary because of the finite amount of this daughter present at even the earliest chopper runs and thus, also, to the sketchy ^{147}Sm resonance data for this energy region available in the literature. Since the transmission measurements were made on three different sample thicknesses, and at various times over a two year span, preliminary visual examination of resonance peak locations and magnitudes in appropriate groups of chopper runs permitted tentative assignment of resonances to the proper nuclide.

The final numerical treatment used the area analysis program of Harvey and Atta[2], recorded for the CDC-1604 computer. Assumed constant values for Γ_γ were used from averaged values at low energy resonances. The wide range of isotopic ratios created by the time factor allowed verification of most of the previous resonance assignments. However, at a few energies, a systematic variation of Γ_n° with time was resolved by postulating coincident resonances; individual parameter values were then estimated by averaging ^{147}Pm and ^{147}Sm Γ_n° contributions obtained from simultaneous equation treatment of all combinations of chopper run pairs with significantly differing isotopic ratios.

The Γ_n° values listed in Table I, below, have a confidence level ranging between $\pm 10\%$ at lowest energies and $\pm 50\%$ at the highest. The 65.5 eV ^{147}Pm resonance is believed, from shape-fit analysis, to be a doublet separated by 0.4 eV; since this is unresolvable in area analysis, the Γ_n° value obtained is without exact physical meaning. The ^{147}Pm level spacing plot exhibits a tailing off above 100 eV; thus either additional doublets or very weak unobserved resonances, or both, may exist.

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- [1] J. W. Coddington, R. L. Tromp, F. B. Simpson, "Total Neutron Cross Section of ^{147}Pm ", in R. L. Heath (ed.), Nuclear Technology Branch Yearly Progress Report for Period Ending June 30, 1968, IN-1218 (December 1968), pp 31-32.
- [2] S. E. Atta and J. A. Harvey, Numerical Analysis of Neutron Resonances, ORNL-3205 (January 1962).

TABLE I

RESONANCE ASSIGNMENTS AND PARAMETERS FOR ^{147}Pm , 60 eV TO 250 eV:
45 METER AREA ANALYSIS

(Assumed Γ_Y : $^{147}\text{Pm} = 80 \text{ mV}$, $^{147}\text{Sm} = 65 \text{ mV}$)					
$E_o \text{ (eV)}$	Isotope	$\Gamma_n^o \text{ (mV)}$	$E_o \text{ (eV)}$	Isotope	$\Gamma_n^o \text{ (mV)}$
65.5	^{147}Pm (doublet)	(9)	145.1	^{147}Pm	3
76.4	^{147}Sm	2	148.5	^{147}Pm	8
80.1	^{147}Sm	0.3	152.2	^{147}Sm	8
82.6	^{247}Pm	3.5	154.3	^{147}Pm	4
83.9	^{147}Sm	6	161.4	^{147}Sm	4.5
85.5	^{147}Pm	4	164.5	^{147}Sm	7
89.8	^{147}Pm	0.12	174.0	^{147}Pm	20
91.5	^{147}Pm	0.10	179.1	^{147}Pm	0.9
95.4	^{147}Pm	2.7	185.3	^{147}Sm	15
99.8	^{147}Pm	3.0	190.3	^{147}Pm	8
	^{147}Sm	24	193.1	^{147}Pm	9
103.1	^{147}Sm	10	208.9	^{147}Sm	20
107.4	^{147}Sm	4	211.1	^{147}Pm	4
115.2	^{147}Pm	5	222.1	^{147}Pm	3
117.7	^{147}Pm	0.24		^{147}Sm	4.5
124.6	^{147}Sm	15	224.0	^{147}Sm	7
131.5	^{147}Pm	15	231.7	^{147}Pm	2.5
140.1	^{147}Pm	0.27	241.2	^{147}Pm	12
	^{147}Sm	64	249.0	^{147}Sm	10

A SEARCH FOR THE (n, γ f) PROCESS IN SLOW-NEUTRON

FISSION OF ^{238}Pu

L. A. Kroger^[1], R. R. Spencer

There has been much renewed interest in the fission process in the last several years due to the discovery of a number of short lived, spontaneous-fission isomers in the actinide region. In addition, there have been observations first made by Michaudon et al at Saclay of intermediate structure in the form of isolated groupings of resonances in the fission cross section of some of these same nuclei. Strutinsky^[2] calculated shell-energy corrections to the liquid drop model and postulated a second minimum in the nuclear potential as a function of deformation to explain these two phenomena. The energy and half-life of these isomers indicate that they fission from or near the ground state of this second minimum^[3].

In light of these recent developments it is necessary to reexamine the (n, γ f) reaction. The probability of this reaction in fissionable nuclei has been discussed by Stravinsky and Shaker^[4] who estimate a width for this process of the order of the radiative capture width, and by Lynn^[5] who estimated a width of 5 to 10% of the radiative capture width. Experimentally there has been no direct evidence for the (n, γ f) process in normally fissionable nuclei. However, in the case of ^{238}Pu , which exhibits rather strong sub-threshold fission, Bowman et al^[6] have suggested the (n, γ f) mechanism may predominate over ordinary fission. Bowman's suggestion is based primarily on an analysis by Vorotnikov et al^[7] who have measured the fission fragment angular distribution of fast neutron-induced fission of ^{238}Pu . Bowman states that for this nucleus, fission from states with $J\pi = 1/2^+$ may be strongly forbidden compared to fission from states with $J\pi = 1/2^-$ or $3/2^-$. A recent investigation^[8] of the angular anisotropy of fission of ^{238}Pu by neutrons confirms the earlier work of Vorotnikov et al for the $K\pi$ values at the saddle point. If these are the only open channels at thermal energies, then the relatively large thermal fission cross section of ^{238}Pu (~ 18 barns) may be the result of the emission of pre-fission $\text{E1 } \gamma$ rays from the initial $1/2^+$ state formed by S-wave neutrons to some $1/2^-$ or $3/2^-$ final state which can fission through the open channel.

These previous estimates of the width of the (n, γ f) process^[4,5,6] were made on the assumption that the nucleus after the emission of a pre-fission gamma ray is still in a highly excited state and fission through a single barrier can compete with further gamma ray emission. The existence of the double hump potential suggests that the (n, γ f) reaction may take place via one or more states in the second well. Observation of monochromatic γ rays associated with the fissioning isomer would give a direct measurement of the energy level structure in the second well.

A search for monochromatic, pre-fission γ rays was carried out with targets of ^{238}Pu , ^{235}U , and ^{239}Pu . The known 300 and 105 n sec half-lives of the fissioning isomers in ^{239}Pu and ^{236}U are particularly amenable to observation by standard delayed coincidence techniques. The ^{239}Pu target was investigated since the ^{238}Pu target contained approximately 0.5% ^{239}Pu .

The neutron source was a beam of neutrons from the HT-I South beam hole of the MTR filtered through a 3-inch quartz single crystal. The beam at the target was approximately 2×10^8 neutrons $\text{cm}^{-2} \text{sec}^{-1}$ with a cadmium ratio (gold activation) of about 200. The target was a 1-1/2-inch diameter deposit of ^{238}Pu electroplated to approximately $300 \mu\text{g}/\text{cm}^2$ onto a 0.003-inch-thick nickel backing. A gas scintillation chamber was used to detect the fission events. A Ge(Li) detector was used to detect gamma rays in coincidence with fission and also with 80 μsec delay in the fission line. The time resolution of the gas scintillator, Ge(Li) detector system, was 70 μsec (FWHM). The purpose of the delayed coincidence run was to look at those gamma rays immediately preceding fission and eliminate most of the post-fission gamma rays. Similar coincidence measurements were made with ^{239}Pu and ^{235}U targets for comparison. An analysis of the spectra (from 140 keV to the neutron binding energy) revealed no strong monochromatic pre-fission gamma rays. Within the statistics of the present spectra, the ^{238}Pu spectra are identical with similar spectra taken with the ^{239}Pu and ^{235}U targets. For thermal neutron fission of ^{238}Pu , a pre-fission γ ray occurring with greater than 10% of the total fissions could have been observed. For the ^{235}U and ^{239}Pu targets, the observable limits were 5 and 3%, respectively.

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 - [8] D. L. Shpak and G. N. Smirenkin, "Angular Anisotropy of Fission of Pu^{238} by Neutrons", JETP Letters, 8 n 10 (November 1968) pp 332-334.

^{241}Pu DATA EVALUATION FOR ENDF/B

J. R. Smith, O. D. Simpson

The Automatic Cross Section Analysis Program ACSAP is being used to obtain a set of single-level resonance parameters for ^{241}Pu . These parameters will be used in a revised ENDF/B data file for the fissile and fertile materials. The current ENDF/B does not contain resonance parameters for ^{241}Pu .

The basis of the resonance portion of the file will be total cross section measurements made on the MTR fast chopper and fission cross section measurements made on both the RPI linear accelerator and the Petrel nuclear explosion. The first step is to obtain a single set of multilevel parameters to describe both the MTR total cross section and the RPI fission cross section data in the energy region 1 to 12 eV. A provisional fit to these data is shown in Figure 1. The resonance parameters used are shown in Table I. To obtain this fit, it has been necessary to use values of Γ_V in the vicinity of 29 meV. Previous fits of total and fission cross sections separately assumed $\Gamma_V = 40$ meV. The lower values for Γ_V will lead to lower values of α in the resonance region.

The second step in the evaluation is to obtain a single-level fit to the curve generated by the multilevel parameters.

Fig. 1 Total and fission cross sections for ^{241}Pu . The curves were calculated from the multilevel parameters listed in Table I.

TABLE I
MULTILEVEL RESONANCE PARAMETERS FOR ^{241}Pu

E_o (eV)	Γ_n^o (meV)	Γ_γ (meV)	Γ_{f1} (meV)	Γ_{f2} (meV)
- 0.160 ^[a]	0.0725	40	0	60
0.264 ^[a]	0.101	40	0	- 72
4.278	0.270	35	0	42
4.560	0.191	29 ^[b]	190	0
5.910	1.02	29 ^[b]	-1350	0
6.922	0.235	18	0	-123
8.600	0.285	34	0	80
9.50	0.045	29 ^[b]	0	-100
10.20	0.453	29 ^[b]	1000	0

[a] Resonance parameters assumed from previous analysis.

[b] Assumed.

Note: The fission widths are always positive; the sign is used to define the type of interference. Resonances having fission widths of the same sign interfere destructively between resonances while resonances with opposite signs interfere constructively.

NEUTRON TRANSMISSION MEASUREMENTS FOR SHIELDING

J. W. Rogers, R. M. Brugger, O. D. Simpson

It is well known that an accurate description of the deep penetration of neutrons is vital to design and optimization of reactor shielding. These neutron effects are evident in radiation damage, energy deposition, and biological exposure. Such descriptions depend on an accurate knowledge of the cross sections, particularly for those cross sections (called windows) that are minimal. The measurements of these minimal cross sections are the most difficult to make by the conventional transmission methods.

While developing the iron filtered-beam[1] it was realized that the measurements of the spectra passed by the filters are closely related to cross section measurements of these windows. In fact, the proton recoil measurements of the spectra passed by the windows has the potential for determining the "parameters" of the window much more precisely than does a conventional transmission measurement. Exploitative runs were started to develop this capability.

Neutron spectra transmitted through iron ranging in thicknesses between 7.98 and 40.62 inches were measured. Two proton recoil detectors of different sensitivities were used because of the increase in intensity as the filter thickness was reduced. A spherical methane-filled (200 cm mercury) detector was used for iron thicknesses ranging from 40.62 to 18.62 inches, and a small cylindrical hydrogen-filled (380 cm mercury) detector was used for iron thicknesses ranging from 23.94 to 7.98 inches. The spectra obtained from these measurements are shown in Figures 1 and 2.

The measurements were made by placing the detectors in the beam and adjusting the gas amplification of each detector to the energy range between 100 and 400 keV. In this energy range the detectors are insensitive to gamma rays; neutron windows at six discrete and resolvable energies (136, 167, 218, 271, 307, and 350 keV) [2] were observed.

The relative change of magnitude of the transmission peaks as the thickness of the filter was changed indicates that each window has a different minimum cross section and window width. An area analysis, similar to that used in the "area analysis" of resonances, is being developed to determine these parameters. Once these parameters have been measured, measurements of inelastic scattering[3] in the last few inches of the iron will also be pursued.

[1] B. W. Howes, R. M. Brugger, J. W. Rogers, R. P. Schuman, and O. D. Simpson, "The Materials Testing Reactor 25 $\pm$ keV Neutron Beam", ANS Trans., 12 n 1 (June 1969) pp 195-196.

[2] J. Rainwater, private communication (April 1969).

[3] W. E. Preeg and H. Goldstein, "Effects of Cross-Section Minima on Deep Penetration of Neutrons", ANS Trans., 12 n 1 (June 1969) p 390.

Fig. 1 The neutron spectra through various thicknesses of iron. The data were taken using a proton-recoil spectrometer with a methane spherical (200 cm Hg) detector.

Fig. 2 The neutron spectra through various thicknesses of iron. The data were taken using a proton-recoil spectrometer with a small cylindrical hydrogen filled (380 cm Hg) detector.

²³⁹Pu PARTIAL CROSS SECTIONS AND ALPHA AT 2 keV

O. D. Simpson, L. G. Miller

A set of pilot experiments has been performed to study the feasibility of making partial cross section measurements in the 2 keV scandium-filtered beam. From these experiments preliminary values of the partial cross sections and alpha for ²³⁹Pu have been derived. Alpha has been determined using two different techniques: (a) measuring partial cross section measurements to get $\alpha = \sigma_{nv}/\sigma_{nf}$ and (b) measuring eta directly and calculating alpha by $\alpha = \bar{\nu}/\eta - 1$. Table I lists the partial cross sections and describes how each value was determined.

In another method of measuring eta directly, three special detectors were used; first a total detector BF₃ counter records the number of neutrons that were removed from the beam by the fissile sample. Second a scattering detector determines the number of neutrons that were scattered by the sample. This detector was calibrated relative to the scattering cross section of lead. And third, a fission detector records the number of fission neutrons. This detector was calibrated using a ²⁵²Cf neutron source. The efficiencies of the scattering detector to fission neutrons and the fission detector to scattered neutrons were also determined.

The absolute value of eta could not be determined because the flux of the 2 keV beam was not known accurately enough. Therefore, alpha for ²³⁹Pu was determined relative to an assumed alpha value of 0.42 for ²³⁵U. Table II lists the measured value of alpha for ²³⁹Pu relative to ²³⁵U.

TABLE I

PARTIAL CROSS SECTIONS AND ALPHA FOR ^{239}Pu AT 2 keV

$\sigma_{nT} = 23.0 \pm 0.5$ b	Determined from the ^{239}Pu total cross section measurements as a function of sample thickness, extrapolated to zero thickness.
$\sigma_{nn} = 15.1 \pm 0.8$	Measured relative to the scattering cross section of lead.
$\sigma_{nx} = 7.9 \pm 0.9$	$\sigma_{nx} = \sigma_{nT} - \sigma_{nn}$
$\sigma_{nf} = 3.5 \pm 0.5$	Measured with a fission chamber in the 2 keV beam relative to a fission cross section of 6.4[a] barns for ^{235}U . The ^{239}Pu and ^{235}U fission chambers had been previously intercalibrated at 0.025 eV on the MTR crystal spectrometer.
$\sigma_{n\gamma} = 4.4 \pm 1.0$	$\sigma_{n\gamma} = \sigma_{nx} - \sigma_{nf}$
$\alpha = 1.26 \pm 0.33$	$\alpha = \sigma_{n\gamma} / \sigma_{nf}$

[a] This number was obtained from the evaluation of ^{235}U as measured by several laboratories. The values below were obtained by weighting the differential fission cross sections with the 2 keV beam spectrum.

W. K. Brown, Nuclear Explosions (1966)	6.26
ENDF/B (1967)	6.24
Wang-Shi-De (1966)	6.36
Yeater (1966)	6.64
Parker (1966)	6.48
Michaudon (1966)	6.31
Average	6.4

TABLE II

ALPHA FOR ^{239}Pu RELATIVE TO AN ALPHA OF 0.42 for ^{235}U AT 2 keV [a]

Sample Thickness (inch)	Alpha
0.020	1.46
0.040	1.20

[a] Errors have not been determined for the above values but are probably of the order of $\pm 25\%$.

THE TOTAL NEUTRON CROSS SECTION OF ^{235}U AND ^{239}Pu AT 2 keV AS A FUNCTION OF SAMPLE THICKNESS

O. D. Simpson, L. G. Miller

It is of interest to know how the self-shielding of neutron resonances effects the cross sections of ^{239}Pu and ^{235}U in the low keV unresolved resonance region. Ideally one desires to know how all the partial cross sections change as a function of sample thickness. However, in this particular measurement only the variation in the total cross section was measured. With luck, the others will be measured later. The total cross section was calculated from transmission data which were determined using the sample-in sample-out technique. The data were taken using the 2 keV filtered beam facilities.

The total cross sections for ^{235}U and ^{239}Pu are shown in Figure 1. The error bars are standard deviations due to counting statistics only. For sample thicknesses greater than 0.100 inch the counting statistics are smaller than the experimental data points.

It is interesting to note that the variation in cross sections as a function of sample thickness for ^{239}Pu is more pronounced than for ^{235}U . This is expected because the reduced neutron scattering widths on the average are much larger for ^{239}Pu than ^{235}U . It is the reduced neutron scattering width (Γ_n^0) that determines the strength of a resonance which reflects directly into the self-shielding problem.

For reactor calculation it is important to know the cross section for infinitely thin samples. Most total cross section measurements are not made with these types of samples and should therefore be corrected for sample thickness effects.

Future plans are scheduled not only to measure partial cross section as a function of sample thickness but also to see how they vary as a function of sample temperature.

Fig. 1 Total neutron cross sections of ^{235}U and ^{239}Pu as a function of sample thickness at 2 keV.

ACTIVATION CROSS SECTION MEASUREMENTS USING THE 2.0 keV Sc
FILTERED AND 25 keV Fe FILTERED NEUTRON BEAMS

R. P. Schuman, R. L. Tromp

Accurate absolute neutron cross sections are needed in the keV region because (a) present accuracy in this region is $\sim 30\%$, (b) the neutron energy spectra of the proposed fast breeder power reactors contain a considerable component of neutrons in this region, and (c) these cross sections are needed to help explain nucleosynthesis. Because of the importance of keV neutron cross section data and because of the advantageous fluxes from the filtered neutron beams of the MTR, a program has been started to measure 2 and 25 keV activation cross sections.

The scandium filter passes a beam of neutrons of 2.0 keV energy with a full width at half maximum of about 700 eV[1]. The iron filter passes a beam of neutrons of 25.0 keV energy with a full width at half maximum of about 2 keV[2]. The iron beam is backed up with sulfur and aluminum so that there are very few neutrons passed with energies outside of the 25 ± 1 keV region. The scandium beam, however, also passes neutrons in the 300 keV region with an intensity of about $1/3$ that in the 2.0 ± 0.35 keV region. Because of the $1/v$ dependence of most activation cross sections, the neutrons in the 300 keV region will normally account for no more than a few percent of the activation. No activation runs have yet been made using a "cleaned up" scandium beam. No activity could be detected in scandium beam irradiated rhodium due to the $^{103}\text{Rh}(n,n^*)$ 57 min $^{103\text{m}}\text{Rh}$ reaction. This reaction has a threshold of 40 keV and will occur with the neutrons in the 300 keV region.

The cross sections determined in the filtered beams are actually resonance integrals with the integration limits (rather indistinctly defined) about 1.65 to 2.35 keV for scandium (plus the minor component in the 300 keV region) and 24 to 26 keV for iron. Thus, the problems of sample self-shielding will be similar to those for resonance integral measurements[3]. Because the activation in the beams is low, the best counting statistics require thick samples; on the other hand, sample self-shielding and neutron degradation require thin samples for accurate cross section results. In order to check the self-shielding effects in both beams, a series of stacks of foils were irradiated and the specific activation measured versus sample thickness. In the scandium beam, the stacks were made up of squares about 1 cm on a side with the squares nearest the beam having a slightly larger area than those farther from the beam. The stacks were irradiated in several positions, usually inside either a 1.27 cm or a 1.90 cm diameter iron collimator and next to the scandium in order to get maximum activation. It could be seen that the collimator or other backing caused some extra activation of the backmost foils due to backscatter and flux softening. The iron filtered beam is less intense and the cross sections lower so the stacks were made up of squares about 3 cm on a side, again with the largest area squares next to the beam. The foils were irradiated next to the iron of the filter which was just outside of the reactor shielding where there was minimum scattering material, and the activation showed no indication of backscatter or flux softening.

Stacks of copper, gold, indium, and tantalum foils have been activated in the scandium beam. The nuclides counted were 12.8 hr ^{64}Cu (count 511 keV annihilation gammas, 38% abundant), 2.697 day ^{198}Au (count 412 keV

gamma, 96% abundant), 54 min ^{116m}In (count 1293 keV gamma, 80% abundant) and 115 day ^{182}Ta (count all gammas above 1 MeV, 100% abundant)[4]. The activity was determined by absolute NaI(Tl) counting. The specific activation versus sample thickness is given in Figures 1, 2, 3, and 4. Stacks of copper and gold foils were irradiated in the iron beam and counted in the same way. The specific activation of foils versus sample thickness is shown in Figures 5 and 6.

From the standpoint of activity formed and ease of counting, gold would make the best flux monitor in the beams. However, the activation versus thickness shows that gold has sharp resonances in the energy ranges covered by the beams and so self-shielding corrections will be large and gold will deplete the neutron flux in narrow energy regions of the filtered beams. Recent cross section measurements made at Saclay have resolved the resonances in the scandium beam energy region and both energies and parameters have been measured (see Table I). The only usable gold flux monitor would be dilute 0.5% Au-Al alloy (first point Figure 2), but the activation is too low for good counting statistics. This thin gold activated about twice as much (per unit mass) as 0.025 mm foils in the scandium beam. Even in the iron beam, gold shows its sharp resonance structure. Indium and tantalum did not show nearly the self-shielding of gold even though they show sharp resonances in the low energy region[5]. Copper activates to 12.8 hr ^{64}Cu which can be determined by the absolute counting of its annihilation radiation (must add sufficient absorber to stop all positrons). Figures 1 and 6 show that 0.05 mm copper foil shows relatively little self-absorption. Copper has a resonance at 2.05 keV (see Figure 7) with a maximum σ_T of about 300 barns, and it also has a σ_T as low as 4 barns in the scandium beam energy region[5]. The curved log activation versus thickness curve indicates there is structure in the scandium beam energy region. The stack of foils activated inside the 1.27 cm diameter iron collimator also showed the effect of neutron scattering or moderation which caused the additional activation of the backmost foils. The stack irradiated outside the collimator (about 3.7 cm diameter for this run), and reactor shield showed no indication of significant neutron scattering or energy degradation. The log activation versus thickness curve in the 25 ± 1 keV iron beam is nearly a straight line indicating $\sigma_T = 16$ b. Copper has structure in this energy region (see Figure 7) but the stack was too thin to show a definite curve. The stack of foils irradiated in the iron beam, again outside of the reactor, also showed no indication of significant neutron scattering or energy degradation.

In order to obtain actual activation cross sections in the scandium and iron filtered beams it is necessary to use a standard. Probably the best standard would be thin ^{10}B [neutrons determined by counting the alpha particles for the $^{10}\text{B}(n,\alpha)$ reaction] or possibly thin ^{235}U (neutrons determined by counting fission fragments). These standards in turn could be calibrated by simultaneously determining the intensity of the beams with the manganese bath[6]. Since no $^{10}\text{B}(n,\alpha)$ or $^{235}\text{U}(n,f)$ counters were available, a number of preliminary measurements were made in both the scandium and iron beams using gold (0.125 mm foils and later 0.5% Au-Al alloy foils) and later copper (0.05 mm foils). The 0.125 mm gold proved to be a poor absolute standard because of the large self-absorption correction, but it could be used as a relative standard. The 0.05 mm copper foils showed relatively little self-absorption, and so made fairly good flux monitors although the counting rates were small. The 0.50% Au-Al alloy foils gave too small counting rates for good counting statistics.

The cross sections measured in the iron beam, relative to the activation of copper extrapolated to zero thickness, are listed in Table II. There are such widely divergent values of ≈ 25 keV cross sections in the literature[5] that it is impossible to choose a value for either gold or copper as a standard.

A number of cross sections have been measured relative to either 0.125 mm gold or 0.05 mm copper in the 2 keV scandium filtered beam. As for 25 keV neutrons, there are widely divergent 2 keV neutron cross section values for both gold and copper. The cross sections measured in the scandium beam, relative to the activation of 0.126 mm gold foils = 4.8 barns, are given in Table III. The value of 4.8 barns for 0.126 mm gold is consistent with the value given in BNL-325 for 0.508 mm gold and the estimated difference in activation for the two thicknesses of samples[5].

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- [1] O. D. Simpson, "Experimental Facilities, Filtered Sc Beams", this report.
 - [2] B. W. Howes, "Experimental Facilities, Filtered Fe Beams", this report.
 - [3] R. P. Schuman and J. R. Berreth, Resonance Integral Measurements, IN-1296 (May 1969).
 - [4] C. M. Lederer, J. M. Hollander, I. Perlman, Table of Isotopes, 6th ed. New York: John Wiley & Sons, Inc., 1967.
 - [5] M. D. Goldberg et al, Neutron Cross Sections. Volume IIC, Z equals 61 to 87, Volume IIB, Z equals 41 to 60, Volume IIA, Z equals 21 to 40, BNL-325 (2nd ed.) (Suppl. 2) (Vol. IIC, IIB and IIA) (1966).
 - [6] J. R. Smith and S. D. Reeder, " η Measurements of ^{239}Pu and ^{235}U at 2 keV", this report.

TABLE I

RESONANCE ENERGIES AND PARAMETERS FOR ^{197}Au IN THE
SCANDIUM BEAM ENERGY REGION, DATA FROM
ALVES, JULIEN, MORGENSTERN, AND SAMOUR^[a]

<u>E_o (eV)</u>	<u>J</u>	<u>$g\Gamma_n$ (meV)</u>	<u>Γ_γ (meV)</u>
1578.2	1	180	~ 160
1592.7	2	37	
1614.4	2	104	~ 112
1634.5		2.7	
1641.1	1?	45	
1645.9	2?	60	
1659.8		2.3	
1693	2	84	
1706	2	169	
1721	2	23	
1734.2	2	197	
1754	2	200	
1756.3	1	213	
1811.4	1	36	
1821.1		9	
1831.6	1	33	
1856	1	520	~ 140
1860	2	54	
1883.4	1	85	
1893		2.3	
1913.6	1	920	~ 150
1939.7	1	195	~ 130
1960.9	2	459	~ 170
2021.1		7	
2028.4	1	100	
2032.8	1	160	
2035.5	2	150	
2059.2		9	

TABLE I (Contd.)

RESONANCE ENERGIES AND PARAMETERS FOR ^{197}Au IN THE
SCANDIUM BEAM ENERGY REGION, DATA FROM
ALVES, JULIEN, MORGENSTERN, AND SAMOUR^[a]

<u>E_o (eV)</u>	<u>J</u>	<u>$g\Gamma_n$ (meV)</u>	<u>Γ_Y (meV)</u>
2074.7	2	676	~ 120
2082.0	2	296	
2088.9	1	177	
2112.4		45	
2131	1	518	~ 140
2147.8	2	307	
2154		125	
2194		120	
2224		20	
2241		75	
2278		10	
2287		80	
2333		90	
2367		60	
2380		3	
2406		75	
2416	1?	400	
2420	2?	700	

[a] R. N. Alves et al, "Determination du Spin et des Parameters des Resonances Pour $^{197}\text{Au} + n$ DE 1000 eV A 2100 eV", Nucl. Phys., A131 (June 1969) pp 450-480.

TABLE II

PRELIMINARY ACTIVATION CROSS SECTION FOR 25 keV
IRON FILTERED NEUTRONS^[b]

Target	Thickness Of Normal Metal mg/cm ²	Reaction Product	Gamma Counted	Cross Section Relative To Thin Copper	
				Thick Target	Thin Target
⁶³ Cu	653	12.8 hr ⁶⁴ Cu	511(38%)	0.90	1 ^[a]
⁶⁵ Cu	653	5.1 min ⁶⁶ Cu	1039(9%)	0.63	--
¹¹⁴ Cd	443	53.5 hr ¹¹⁵ Cd	490+535(36%)	2.50	--
¹¹⁰ Cd	443	48.6 min ^{111m} Cd	247(94%)	0.43	--
¹¹⁵ In	101	54 min ^{116m} In	1293(80%)	13	--
¹⁹⁷ Au	246	2.697 d ¹⁹⁸ Au	412(96%)	14	22

[a] Assumed as standard.

[b] J. F. Vervier, "Section Efficace de Capture Radiative Pour des Neutrons d'une Source Sb-Be", Nucl. Phys., 9 n 4 (1958/59) pp 569-576.

TABLE III

PRELIMINARY ACTIVATION CROSS SECTION FOR 2 keV
SCANDIUM FILTERED NEUTRONS

Target	Thickness of Normal Material mg/cm ²	Reaction Product	Gamma Counted keV and Abundance[4]	"Thick Sample" Cross Section Assuming 0.126 mm Au = 4.8 barns barns
²³ Na	610 (NaBr)	14.96 hr ²⁴ Na	2754 (100%)	0.015
⁴¹ K	425 (KCl)	12.36 hr ⁴² K	1524 (18%)	0.30
⁵⁵ Mn	567 (Mn)	2.576 hr ⁵⁶ Mn	847 (99%)	0.36
⁶⁰ Co	885 (Co)	5.263 yr ⁶⁰ Co	1332 (100%)	0.13
⁶⁰ Co	885 (Co)	10.47 m ^{60m} Co	59 (2.1%)	0.096
⁶³ Cu	266 (Cu)	12.8 hr ⁶⁴ Cu	511 (38%)	0.48
⁶⁴ Zn	259 (Zn)	245 d ⁶⁵ Zn	1115 (49%)	0.10
⁶⁵ Cu	266 (Cu)	5.1 m ⁶⁶ Cu	1039 (9%)	0.10
⁶⁸ Zn	259 (Zn)	13.8 hr ^{69m} Zn	439 (95%)	0.022
⁷⁹ Br	610 (NaBr)	4.38 hr ^{80m} Br	37 (36%)	1.1
⁸¹ Br	610 (NaBr)	35.34 hr ⁸² Br	777 (83%)	1.1
⁹⁴ Zr	438 (Zr)	65.5 d ⁹⁵ Zr	724+756 (98%)	0.095
⁹⁶ Zr	438 (Zr)	17.0 hr ⁹⁷ Zr	665 (98%)	0.079
⁹⁶ Ru	344 (Ru)	2.88 d ⁹⁷ Ru	215 (91%)	0.48
¹⁰² Ru	344 (Ru)	39.5 d ¹⁰³ Ru	497 (88%)	0.79
¹⁰³ Rh	301 (Rh)	4.41 m ^{104m} Rh	51 (47%)	0.13
¹⁰⁴ Ru	344 (Ru)	4.44 hr ¹⁰⁵ Ru	726 (48%)	0.84
¹¹⁰ Cd	280 (Cd)	48.6 m ^{111m} Cd	247 (94%)	0.11
¹¹⁴ Cd	280 (Cd)	53.5 hr ¹¹⁵ Cd	535 (26%)	0.76
¹¹⁵ In	211 (In)	54.0 m ^{116m} In	1293 (80%)	1.7
¹²¹ Sb	654 (Sb)	2.77 d ¹²² Sb	564 (66%)	2.0

TABLE III (Contd.)

PRELIMINARY ACTIVATION CROSS SECTION FOR 2 keV
SCANDIUM FILTERED NEUTRONS

Target	Thickness of Normal Material mg/cm ²	Reaction Product	Gamma Counted keV and Abundance[4]	"Thick Sample" Cross Section Assuming 0.126 mm Au = 4.8 barns barns
¹²³ Sb	654 (Sb)	60.4 d ¹²⁴ Sb	1692 (50%)	1.6
¹³³ Cs	501 (CsCl)	2.05 yr ¹³⁴ Cs	796+802 (98%)	2.4
¹³³ Cs	501 (CsCl)	2.90 hr ^{134m} Cs	128 (14%)	0.32
¹³⁹ La	213 (La ₂ O ₃)	40.2 hr ¹⁴⁰ La	1596 (96%)	0.23
¹⁵⁹ Tb	241 (Tb)	72.3 d ¹⁶⁰ Tb	920 (62%)	12
¹⁸¹ Ta	274 (Ta)	115 d ¹⁸² Ta	>1 MeV (100%)	6.2
¹⁹¹ Ir	754 (Ir)	74.2 d ¹⁹² Ir	317 (81%)	10
¹⁹³ Ir	754 (Ir)	17.4 hr ¹⁹⁴ Ir	328 (19%)	7.6
¹⁹⁷ Au	245 (Au)	2.70 d ¹⁹⁸ Au	412 (96%)	[4.8 assumed]
²³² Th	552 (Th)	27.2 d ²³³ Pa	311 (34%)	2.0
²³⁵ U	1539 (93% enr.)	fission products		6.4
²³⁸ U	330 (U ₃ O ₈)	2.35 d ²³⁹ Np	228 (11.8%) 278 (14%)	1.6

Fig. 1 Scandium beam activation of stack copper foils.

Fig. 2 Scandium beam activation of gold foils.

Fig. 3 Scandium beam activation of thick indium foils.

Fig. 4 Scandium beam activation of tantalum foils.

Fig. 5 Activation of Au foils in 25 keV beam.

Fig. 6 Activation of Cu foils in 25 keV beam.

Fig. 7 Total cross section of copper in the kilovolt energy region.

FAST NEUTRON ACTIVATION CROSS SECTIONS

R. P. Schuman, D. K. Oestreich

It appears that breeder reactors will soon become the most economical source of nuclear power. Thermal breeder reactors will effectively use ^{232}Th while fast breeder reactors will utilize ^{283}U .

The prompt fission neutrons range in energy up to tens of MeV with a most probable energy of about 1 MeV. The highest energy neutrons are capable of causing a large number of endoergic nuclear reactions, and thus the activation of materials irradiated in fast reactors will be quite different from that of materials irradiated in thermal reactors. The activation of materials by threshold reactions will often pose problems of accessibility during fueling and repairing of the reactors. For example, titanium in a pure thermal flux will become activated only to $5.1 \text{ min } ^{51}\text{Ti}$ (and that with only a small yield). However, in a typical fast reactor the titanium will be activated, due to the fast flux, by a number of scandium isotopes, especially $84 \text{ d } ^{46}\text{Sc}$, which are produced to n,p reactions, and also to a smaller extent by $3 \text{ hr } ^{45}\text{Ti}$, produced by a $n,2n$ reaction, and $163 \text{ d } ^{45}\text{Ca}$, produced by a n,α reaction. Thus, titanium will be highly activated in a reactor even though Ti has no long-lived neutron capture product. In a fast reactor, the n,p , n,α and $n,2n$ reactions will be much more prevalent than in a thermal reactor. Other (and much more important) materials in which threshold reactions activation is very important are aluminum where the $^{27}\text{Al} (n,\alpha)^{24}\text{Na}$ contributes to a serious radiation hazard in maintenance

of reactors, and stainless steel where $^{56}\text{Fe}(n,p)^{56}\text{Mn}$, $^{54}\text{Fe}(n,p)^{54}\text{Mn}$, and $^{58}\text{Ni}(n,p)^{58}\text{Co}$ produce as much activity as $^{58}\text{Fe}(n,\gamma)^{59}\text{Fe}$ even in quite highly moderated reactors like the MTR[1].

The use of fast flux activation could greatly extend the utility of activation analysis, especially if a fast reactor or a receptacle slug were used so the usual n,γ activation was minimized. For example, aluminum could be analyzed by activation (n,γ) to ^{24}Na , and in a fast reactor, the interference due to $\text{Na}(n,\gamma)$ would be minimized.

Fast reactors could be used to produce certain isotopes by threshold reactions. For example, ^{58}Co could be produced very efficiently by $^{58}\text{Ni}(n,p)^{58}\text{Co}$ in a fast reactor; in a thermal reactor, not only is the yield smaller due to the lower fast flux, but also the product is burned out by thermal neutron capture. Fast reactors could produce ^{231}Pa by the $^{232}\text{Th}(n,2n)^{231}\text{Th} \xrightarrow{\beta^-} ^{231}\text{Pa}$ reaction. The protactinium could be chemically separated after the ^{233}Pa had decayed. A small amount of protactinium has been isolated from thorium irradiated in a thermal production reactor; however, the yield would be greater in a fast reactor.

Because of the interest in fast breeder power reactors and because of the possibilities of doing fast activation analysis and of producing isotopes by fast flux irradiation, a program to measure fast neutron activation cross sections was started.

In order to check the feasibility of using a spontaneous fission neutron source for measuring fission spectrum threshold reaction cross sections, samples of aluminum, iron, indium, and gold were placed next to a 2 g ^{244}Cm source (2.2×10^7 n/sec). Activation products due to $\text{Al}(n,\alpha)$, $\text{Al}(n,p)$, $\text{Fe}(n,p)$, $\text{In}(n,n')$ as well as $\text{In}(n,\gamma)$ and $\text{Au}(n,\gamma)$ were detected. The activation was then repeated using a 50 μg ^{252}Cf ($\approx 1.2 \times 10^8$ n/sec) fast neutron source. The californium was stored in a moderating environment so the samples were wrapped in 0.5 mm Cd to minimize thermal neutron capture. The following threshold reaction activities were produced in sufficient yield to detect easily by Ge(Li) and NaI(Tl) counting: 14.96 hr ^{24}Na and 9.46 min ^{27}Mg from Al, 3.43 d ^{47}Sc from Ti, 71.3 d ^{58}Co from Ni, 4.50 hr $^{115\text{m}}\text{In}$ from In, and 6.18 d ^{196}Au from Au. These preliminary experiments showed that a large, ≈ 1 mg, ^{252}Cf source, emitting 2.4×10^9 n/sec and placed in an environment where scattering materials tailored the proper flux, could be used to determine typical fast reactor activation cross sections.

In order to obtain some quantitative integral cross sections, a number of targets were irradiated for one hour in the Argonne AFSR reactor operating at 500 watts. Titanium, nickel, zinc, gold, thorium, and aluminum were irradiated and the aluminum used as the fast flux monitor. The irradiated foils were counted with standardized Ge(Li) and NaI(Tl) counters. The cross sections determined, based on absolute gamma counting with standardized lithium-drifted germanium and scintillation counters and on the gamma abundances listed in the Table of Isotopes[2], are given in Table I. The errors given are the estimated errors assuming that the gamma abundances and $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$ cross section are correct. If more than one gamma ray was used, the errors include any relative error in the gamma abundances. The thorium fission cross section determinations showed that the fission yields in the Reactor Handbook[3] are in error for several fission products. The cross sections

determined are those for the AFSR glory hole, and the n,γ cross sections will vary considerably from those for other fast reactors; the threshold cross section will vary less.

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- [2] C. M. Lederer, J. M. Hollander, I. Perlman, Table of Isotopes, 6th ed. New York: John Wiley & Sons, Inc., 1967.
- [3] C. R. Tipton, Jr. (ed.), Reactor Handbook. Volume I, Materials, New York: Interscience Publishers, Inc., 1960.
- [4] W. N. McElroy, S. Berg, T. B. Crockett, "Measurement of Neutron Flux Spectra by a Multiple-Foil Activation Iterative Method and Comparison with Reactor Physics Calculations and Spectrometer Measurements", ANS Trans., 10 n 2 (November 1967) p 577.

TABLE I

FAST ACTIVATION CROSS SECTIONS IN THE AFSR GLORY HOLE FLUX

Target	Product	Gamma Counted		Cross Section (Millibarns)	
Aluminum	(n, α)	^{24}Na	1369	(100%)	0.767 (standard) ^[4]
			2754	(100%)	
Thorium	(n,2n)	^{131}Th	84	(7.4%)	~ 10
	(n,f)	^{140}Ba	537	(27%)	
		^{132}Te	773	(89%)	84
	(n, γ)	^{233}Pa	311	(34%)	262 $\pm$ 15
Gold	(n,2n)	^{196}Au	356	(94%)	3.14 $\pm$ 0.2
			333	(25%)	
	(n, γ)	^{198}Au	412	(96%)	250 $\pm$ 15
Zinc	(n, γ)	^{65}Zn	1115	(49%)	40 $\pm$ 2
	(n,p)	^{64}Cu	511	(38%)	32 $\pm$ 2
	(n,p)	^{67}Cu	184	(40%)	1.11 $\pm$ 0.08
	(n, α)	^{65}Ni	1481	(25%)	0.077 $\pm$ 0.01
	(n, α)	^{71m}Zn	385	(94%)	4.0 $\pm$ 0.8
			495	(75%)	$\pm$
	(n, γ)	^{69m}Zn	439	(95%)	3.6 $\pm$ 0.3
Nickel	(n,p)	^{58}Co	810	(99%)	114 $\pm$ 7
	(n,p)	^{61}Co	67	(89%)	1.3 $\pm$ 0.1
	(n, γ)	^{65}Ni	1481	(25%)	3.3 $\pm$ 0.2
Titanium	(n,p)	^{46}Sc	1120	(100%)	10.9 $\pm$ 0.7
	(n,p)	^{47}Sc	150	(73%)	19.8 $\pm$ 1.2
	(n,p)	^{48}Sc	1314	(100%)	0.334 $\pm$ 0.02
	(n,2n)	^{45}Ti	511	(170%)	0.0087 $\pm$ 0.001
	(n, γ)	^{51}Ti	320	(95%)	2.6 $\pm$ 0.4

THERMAL CROSS SECTIONS AND RESONANCE INTEGRALS OF ^{93}Nb and ^{94}Nb

R. P. Schuman

The thermal capture cross section and resonance integral of niobium, 100% ^{93}Nb , were measured by irradiating ≈ 1 -mg samples of niobium metal, both unshielded and inside a 2-mm-thick cadmium shield, in the core of the MTR. The 20,300 year ^{94}Nb formed was determined by absolute gamma counting of the 871.1 keV (100% abundant) [1] gamma ray of ^{94}Nb on a standardized NaI(Tl) scintillation spectrometer using the counting efficiencies given by Heath [2]. The resonance and thermal fluxes were determined by 0.5% Co-Al activation monitors assuming σ_{th} of $^{59}\text{Co} = 38$ b and R.I. of $^{59}\text{Co} = 74.6$ b [3]. The cross sections obtained for ^{93}Nb are: Thermal (subcadmium) cross section, 1.0 ± 0.1 b, and resonance integral (including $1/\nu$ contribution), 8.5 ± 0.5 b.

Since the irradiation of niobium also produced ^{95}Nb by second order capture, samples of niobium metal that had been irradiated to an nvt of 1.2×10^{22} n/cm² in the MTR and which had stood long enough for all the 35.3 d ^{95}Nb to decay were irradiated, along with dilute Au-Al flux monitors, for one hour, both unshielded and in a 1-mm-thick cadmium shield, in a pneumatic rabbit in the MTR. The irradiated samples were counted, after $^{95\text{m}}\text{Nb}$ had decayed, with a standardized Ge(Li) gamma spectrometer and the cross section determined assuming the 765 keV gamma ray is 100% abundant [4]. Thermal, subcadmium, cross section of ^{94}Nb is 13.6 ± 1.5 b and the resonance integral, 125 ± 8 b (versus gold = 98.8 b thermal and 1558 b resonance integral) [5]. Our values can be compared with ^{93}Nb , $\sigma_{\text{th}} = 1.1 \pm 0.1$ b, R.I. = 9.2 ± 0.5 b and ^{94}Nb , $\sigma_{\text{th}} = 16.8 \pm 1.5$ b, R.I. = 122 ± 10 b [6].

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- [5] R. L. Macklin and H. S. Pomerance, "Resonance Capture Integrals", Prog. in Nucl. Energy. Series I, Physics and Mathematics, 1 (1956) p 187.
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REACTOR THERMAL-NEUTRON CROSS SECTIONS OF ^{64}Zn ,

^{68}Zn , ^{121}Sb , ^{123}Sb , ^{55}Mn , AND ^{186}W

C. H. Hogg, W. L. Wilson^[1]

Reactor thermal-neutron (n, γ) cross sections for the isotopes ^{64}Zn , ^{68}Zn , ^{121}Sb , ^{123}Sb , ^{55}Mn , and ^{186}W have been measured relative to the 2200 m/sec value of 37 b for ^{59}Co [2]. It was assumed that the cross section for each isotope followed the 1/V law in the neutron energy range from 0 up to the cadmium cut-off energy, ie, ~ 0.5 eV for 40 mil cadmium thickness. The cross sections quoted below are therefore assumed to be 2200 m/sec cross sections.

These measurements were made by irradiating infinitely dilute samples of each isotope in the VH-2, VH-4, and T₂H₃ (thermal column) irradiation facilities of the MTR, measuring the gamma-ray emission rate of the reaction products by procedures described in Reference 3 and calculating the disintegration rates using parameters listed in Reference 4. The activities produced by resonance neutrons having energies greater than 0.5 eV were determined from cadmium ratio measurements and corrections made when necessary to obtain the isotope production due to thermal neutrons.

Table I summarizes the results. Each cross section is the total cross section for the formation of all isomers. The values for ^{64}Zn , ^{68}Zn , and ^{122}Sb differ from those reported in BNL-325.

- [1] Present address: The American School of Kinshasa Democratic Republic of Conga.
- [2] M. D. Goldberg, S. F. Mughabghab, B. A. Maguro, V. M. May, Neutron Cross Sections, BNL-325, 2nd Edition, Supplement No. 2.
- [3] R. L. Health, Scintillation Spectrometry Gamma-Ray Spectrum Catalogue, IDO-16880 (1964).
- [4] C. M. Lederer, J. M. Hollander, I. Perlman, Table of Isotopes, 6th Edition (1967).

TABLE I

CROSS SECTION SUMMARY

<u>Isotope</u>	<u>Cross Section</u>	<u>Reaction</u>
^{64}Zn	0.75 ± 0.04 b	$^{64}\text{Zn}(n,\gamma)^{65}\text{Zn}$ 245 day
^{68}Zn	0.073 ± 0.004 mb	$^{68}\text{Zn}(n,\gamma)^{69}\text{Zn}$ 13.8 hr
^{121}Sb	6.1 ± 0.3 b	$^{121}\text{Sb}(n,\gamma)^{122}\text{Sb}$ 2.80 day
^{123}Sb	4.3 ± 0.4 b	$^{123}\text{Sb}(n,\gamma)^{124}\text{Sb}$ 60 day
^{55}Mn	13.5 ± 0.5 b	$^{55}\text{Mn}(n,\gamma)^{56}\text{Mn}$ 2.58 hr
^{186}W	40 ± 1.5 b	$^{186}\text{W}(n,\gamma)^{187}\text{W}$ 23.9 hr

THERMAL (SUBCADMIUM) CAPTURE CROSS SECTION AND
RESONANCE INTEGRAL OF ^{244}Pu

R. P. Schuman

Plutonium 244 is the longest lived plutonium isotope. It is produced in small yields during the high flux irradiation of ^{242}Pu by the neutron capture sequence $^{242}\text{Pu}(n,\gamma) ^{243}\text{Pu}(n,\gamma) ^{244}\text{Pu}$; however, because of the short ^{243}Pu beta half-life, most of the chain goes to ^{243}Am by beta decay rather than to ^{244}Pu by neutron capture or to fission. However, ^{244}Pu has a very low cross section for a heavy isotope and so it builds up to appreciable concentrations in very highly irradiated TRU plutonium.

Because of its very long half-life $(8.28 \pm 0.10) \times 10^7$ years [1], ^{244}Pu is an extremely valuable isotope for chemical studies (essentially no radiolysis or radiation damage) and biological studies (chemical toxicity probably greater than radioactive). Small amounts of enriched ^{244}Pu have been produced by electromagnetic enrichment of highly irradiated transuranium program plutonium, and a production of gram quantities is feasible. The buildup of ^{244}Pu depends strongly upon its cross section; consequently, a determination was made of the thermal (subcadmium) capture cross section and resonance capture integral. Previously, only "pile" cross sections had been measured.

The cross sections were measured by irradiation of $\approx 200 \mu\text{g}$ samples of highly enriched ^{244}Pu , both in a 1-mm-thick cadmium shield and unshielded in the MTR VP-7 pneumatic rabbit facility. The enriched plutonium (Pu-244-239A) was obtained from Oak Ridge National Laboratory and has been mass analyzed by them. It contained: $^{238}\text{Pu} < 0.006\%$, $^{239}\text{Pu} 0.018\%$, $^{240}\text{Pu} 0.007\%$, $^{241}\text{Pu} 0.002\%$, $^{242}\text{Pu} 0.904\%$, and $^{244}\text{Pu} 99.06\%$. Alpha spectra on fairly thick samples of the plutonium showed four peaks, ^{238}Pu (plus any ^{241}Am daughter of ^{241}Pu), $^{239}+^{240}\text{Pu}$, ^{242}Pu , and ^{244}Pu ; the largest fraction of the alpha activity was due to ^{238}Pu . The plutonium, obtained as oxide, was dissolved in concentrated HI, then converted to chloride and aliquots evaporated to dryness and sealed in quartz ampoules. The flux was monitored with very dilute 0.5% gold in aluminum foil.

After irradiation, the quartz ampoules were opened and the plutonium from the unshielded sample was purified by adsorption of the plutonium [as Pu(IV)] on a Dowex 1 column from 8 N HCl and subsequent elution of the Pu [as Pu(III)] from the column with 8 N HCl + 2% HI. Since the plutonium showed very little radioactive impurity, the cadmium shielded plutonium sample was counted without chemical purification. The amount of 10.48 hr ^{245}Pu produced was determined by absolute Ge(Li) gamma counting assuming the major 327 keV ^{245}Pu gamma ray has a photon abundance of $\approx 24\%$ [2] and the major 252 keV ^{245}Am daughter gamma ray has a photon abundance of $\approx 6.1\%$ [3]. The ^{245}Am gamma was used only after the ^{245}Am had reached secular equilibrium. The ^{245}Pu plus ^{245}Am gamma spectrum obtained on the purified plutonium is shown in Figure 1. The amount of ^{245}Pu was determined by absolute Ge(Li) gamma counting of the ^{240}Np in secular equilibrium with the ^{244}Pu after all the 10.48 hr ^{245}Pu had decayed. The ^{240}Np gamma-ray abundances were taken as 21.4% for the 554 keV gamma ray and 12.6% for the 597 keV gamma ray [4]. The fluxes of the irradiation positions were determined

from the thin Au-Al alloy monitors assuming that the thermal capture cross section of ^{197}Au is 98.8 barns and the resonance capture integral 1558 b [5]. The amount of ^{198}Au was determined by absolute Ge(Li) gamma counting assuming an abundance of 96% for the 412 keV gamma ray [6].

The thermal (subcadmium) activation cross section of ^{244}Pu was measured as 1.6 ± 0.3 barns and the resonance integral as 35 ± 7 barns. The uncertainty in the gamma abundances is the largest contributor to the errors. The counting and experimental errors are about $\pm 10\%$. The values can be compared with "pile" cross sections of 2.1 ± 0.3 barns [7] and 1.4 ± 0.5 barns [8] previously measured.

- [1] C. E. Bemis, Jr., J. Halperin, R. Eby, "The Alpha Decay Half Lives of ^{242}Pu and ^{244}Pu ", J. Inorg. Nucl. Chem., 31 (March 1969) pp 599-604.
- [2] W. R. Daniels et al, "Decay of ^{245}Pu ", Nucl. Phys., A107 (1968) pp 569-580.
- [3] M. E. Bunker et al, "Decay of ^{245}Am ", Nucl. Phys., A97 (1967) pp 593-599.
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- [5] R. L. Macklin and H. S. Pomerance, "Resonance Capture Integrals", Prog. in Nucl. Energy. Series I, Physics and Mathematics, 1 (1956) p 187.
- [6] C. M. Lederer, J. M. Hollander, I. Perlman, Table of Isotopes, 6th ed. New York: John Wiley & Sons, Inc., 1967.
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Fig. 1. Gamma Spectrum of ^{245}Pu plus ^{245}Am .

RESONANCE INTEGRALS FROM THE CADMIUM SHIELDED IRRADIATION OF ^{241}Am

R. P. Schuman

The neutron cross section of the 152 year isomer of ^{242}Am is of interest both in the production of isotopic heat sources and in the systematics of odd-odd nuclide cross sections. The thermal pile irradiation of ^{241}Am will normally give a little more than 1% ^{242m}Am and considerable ^{243}Am , and so irradiated americium will make a poor ^{242m}Am cross section sample unless the ^{242m}Am is electromagnetically enriched.

In order to investigate the possibility of producing higher enrichment ^{242m}Am cross section samples, two samples of pure ^{241}Am were irradiated in high flux ETR core positions to different integrated fluxes in 3- and 7-mm-thick cadmium shields, respectively. The irradiations showed that a somewhat higher concentration of ^{242m}Am could be produced with relatively little ^{243}Am , and allowed resonance integrals to be obtained for ^{241}Am , 152 yr ^{242m}Am , and ^{242}Cm . In each irradiation about $100\mu\text{g}$ of ^{241}Am as AmO_2 were sealed in quartz ampoules and irradiated along with 0.5% Co-Al alloy flux monitors. After the irradiation and several months cooling, the capsules were opened in a hot cell and the americium ampoules and the Co-Al flux monitors removed and processed. The amount of ^{60}Co activity was determined by a standardized high pressure gamma iron chamber. The irradiated americium was dissolved in dilute HCl, and Pu, Am, and Cm separated by a heated Dowex 50 column eluted with α hydroxyisobutyric acid. The yields of Pu, Am, and Cm, activity were obtained by 2π alpha counting and alpha spectroscopy using a silicon surface-barrier counter. The americium from one sample and the Pu, Am, and Cm from the other were mass analyzed. Relative yields of Pu, Am, and Cm isotopes, obtained from the alpha counting and mass analyses, are given in Table I.

The resonance flux of the irradiations was determined assuming a ^{59}Co resonance integral of 74.6 barns[1]. The resonance integrals of ^{241}Am , ^{242m}Am , and ^{242}Cm were obtained by assuming values of resonance integrals and calculating isotopic ratios using Bateman equations, and then taking the values that give the best fit. The calculations were made on a PDP-8 computer using the ESI language. The resonance integrals are given in Table II. The resonance integrals given in Table II are the values measured and are not corrected for variable cadmium shield thickness. The second resonance (at 0.576 eV)[2] in ^{241}Am occurs at the cadmium cutoff energy; as expected, the measured ^{241}Am resonance integral was less (810 barns to 16 hr ^{242}Am) for the thicker shield (initially 7 mm) than (875 barns to 16 hr ^{242}Am) for the thinner shield (initially 3mm). The ^{241}Am resonance integral was calculated assuming that the total resonance integral of 16 hr ^{242}Am is $<20,000$ b. The ^{242}Cm resonance capture integral was calculated assuming the total resonance integral of ^{243}Cm is <3000 b.

The purified curium fraction from the irradiated americium was counted with a standardized lithium-drifted germanium gamma spectrometer, and the photon abundances of the ^{242}Cm and ^{243}Cm gamma rays determined by comparing gamma intensities with alpha counting rates. The results of the gamma intensity measurements are given in Table III.

[1] R. P. Schuman and J. R. Berreth, Resonance Integral Measurements, IN-1296 (May 1969).

[2] J. R. Stehn et al, Neutron Cross Sections. Volume 3, Z equals 88 to 98, BNL-325 (2nd ed.) (Suppl. 2) (Vol. 3) (February 1965).

TABLE I

RESONANCE FLUX IRRADIATION OF ^{241}Am
COMPOSITION AT END OF IRRADIATION

<u>Irradiation</u>	<u>No. 1</u>	<u>No. 2</u>
Time in seconds	2.42×10^6	8.64×10^6
Resonance Flux $n/\text{cm}^2/\text{sec}$	2.20×10^{13}	2.95×10^{13}
^{241}Am amount	1,000,000 ^[a]	1,000,000 ^[a]
^{238}Pu	--	10,600
^{242}Pu	--	27,000
$^{242\text{m}}\text{Am}$ (152 yr)	10,940	33,880
^{243}Am	415	5,400
^{242}Cm	37,400	152,900
^{243}Cm	--	2,484
^{244}Cm	--	806

[a] Composition normalized to 10^6 parts ^{241}Am .

TABLE II

RESONANCE INTEGRALS OF ^{241}Am , $^{242\text{m}}\text{Am}$, and ^{242}Cm

<u>Target Nucleus</u>	<u>Product Nucleus</u>	<u>Resonance Integrals (barns)</u>
458 yr ^{241}Am	16 hr ^{242}Am	850 ± 60 b
458 yr ^{241}Am	152 yr $^{242\text{m}}\text{Am}$	250 ± 40 b
152 yr $^{242\text{m}}\text{Am}$	total destruction	7000 ± 2000 b
162.5 d ^{242}Cm	32 yr ^{243}Cm	150 ± 40 b

TABLE III
PHOTON INTENSITIES OF ^{242}Cm AND ^{243}Cm GAMMA RAYS

Nuclide	Gamma-Ray Energy (keV)	Photon Intensity (%)
162.5 d ^{242}Cm	44.1	0.027
	102.0	0.0023
	157.7	0.00155
32 yr ^{243}Cm	209.9	3.5
	228.4	11.8
	277.8	16.6

RESONANCE INTEGRALS FROM THE CADMIUM SHIELDED
IRRADIATION OF ^{244}Cm

R. P. Schuman

The neutron cross section of ^{245}Cm is of interest both to the large scale production of ^{252}Cf and in the systematics of fission cross sections. The thermal pile irradiation of ^{244}Cm (or ^{232}Pu) usually produces ^{245}Cm with an abundance of about 1% of that of ^{244}Cm ; the 245/244 abundance ratio also varies greatly with reactor spectrum.

In order to investigate the possibility of producing curium with much higher percentages of ^{245}Cm by resonance flux irradiation, a $\approx 1 \mu\text{g}$ sample of ^{244}Cm was included with the americium sample during the second high integrated flux irradiation in an ETR core position. The irradiation resulted in an even greater buildup of ^{245}Cm than expected. The isotopic composition of the curium before and after irradiation is given in Table I. The irradiated curium was purified by elution from a heated Dowex 50 column with α hydroxyisobutyric acid. The purified fractions were analyzed by 2π alpha counting, alpha spectroscopy using silicon surface-barrier counters, lithium-drifted germanium gamma spectrum analysis, and mass analysis of the curium sample, then the relative composition at the end of irradiation determined, see Table II. The irradiated curium was found to contain heavy nuclides up to ^{252}Cf .

The resonance flux was determined using a 0.5% Co-Al flux monitor and assuming a resonance integral of 74.6 barns for ^{59}Co [1]. The resonance integrals of a number of curium isotopes were estimated by assuming resonance integral values and calculating abundances using Bateman equations. The calculations were made using a PDP-8 computer and the ESI language. Since only one sample was irradiated, and abundances depend upon a number of cross sections, only approximate values of resonance integrals could be obtained. The values that give the best fit and seem most likely are listed in Table III. Resonance integrals of ^{244}Cm and ^{246}Cm were also estimated from resonance parameters[2] and are also given in Table III.

The purified curium fraction was counted with a standardized lithium-drifted germanium gamma spectrometer and photon abundances and energies of the major ^{244}Cm and ^{245}Cm gamma rays determined (see Table IV). Alpha spectrum measurements on the purified curium showed the major alpha group of ^{245}Cm , but the uncertainty in the abundance measurement was quite large.

- [1] R. P. Schuman and J. R. Berreth, Resonance Integral Measurements, IN-1296 (May 1969).
- [2] J. R. Stehn et al, Neutron Cross Sections. Volume 3, Z equals 88 to 98, BNL-325 (2nd ed.) (Suppl. 2) (Vol. 3) (February 1965).

TABLE I

ISOTOPIC COMPOSITION OF RESONANCE FLUX IRRADIATED CURIUM

Average Resonance Flux $2.95 \times 10^{13} \text{ n/cm}^2 \text{ sec}$		
Time $8.64 \times 10^6 \text{ sec}$		
	<u>Percent Abundance</u>	
<u>Isotope</u>	<u>Before Irradiation</u>	<u>After Irradiation</u>
243	0.037	0.02
244	95.26	81.00
245	1.66	15.50
246	2.97	3.31
247	0.051	0.12
248	0.021	0.05

TABLE II

RELATIVE COMPOSITION OF RESONANCE FLUX IRRADIATED CURIUM

	<u>Before Irradiation</u>	<u>At End of Irradiation</u>
Time, seconds	---	8.64×10^6
Resonance flux, $n/cm^2 \text{ sec}$	---	2.95×10^{13}
^{238}Pu	---	437
^{240}Pu	---	16,000
^{242}Cm	---	315
^{243}Cm	390	480
^{244}Cm	1,000,000	1,000,000
^{245}Cm	17,400	186,000
^{246}Cm	31,200	39,800
^{247}Cm	530	1,440
^{248}Cm	220	604
^{250}Cf	---	1.71
^{252}Cf	---	0.28

TABLE III
 RESONANCE INTEGRALS OF Cm ISOTOPES
 7 mm Cd Shield Initially

Target Nucleus	Product Nucleus	Resonance Integral-Barns [a]	Notes
^{244}Cm	^{245}Cm	$650 \pm 50 \text{ b}$	Also calculated 650 b from resonance parameters[19]
^{245}Cm	Total Destruction	$680 \pm 300 \text{ b}$	Assume R.I. $^{244}\text{Cm} =$ 650 b
^{246}Cm	^{247}Cm	$110 \pm 40 \text{ b}$	Assume ^{247}Cm , R.I. < 2500 b. Estimated R.I. = 140 b from resonance parameters[19]

[a] Values as measured; no correction made for the effects of resonances near cadmium cutoff.

TABLE IV
 ENERGIES AND PHOTON INTENSITIES OF ^{244}Cm AND ^{245}Cm GAMMA RAYS

Nuclide	Gamma-Ray Energy (keV)	Photon Intensity (%)
18.0 yr ^{244}Cm	42.85	0.018
	152.6	0.0012
9300 yr ^{245}Cm	133.0	2.6
	175.1	9.7

REACTIVITY MEASUREMENT OF ^{238}U DOPPLER EFFECT

J. J. Scoville

A measurement of the Doppler effect in ^{238}U was undertaken in support of the Power Burst Facility (PBF) project. The experiment was necessary in order to provide a check against the mathematical model employed for the calculation of the Doppler shutdown characteristics of PBF.

Samples for the experiment were pressed and sintered $\text{UO}_2\text{-ZrO}$ pellets of the PBF type, but contained depleted uranium. Reactivity measurements were made in the cadmium lined dry tube facility of the ARMF-II in a heated capsule over a temperature range from 20 to 1000°C . The data were then corrected for fast fission in ^{238}U and for fission in ^{235}U . The fractional change in reactivity as a function of temperature is shown in Figure 1. The calculated values shown are the results of a Doppler broadened effective resonance integral calculation using the RAVEN code. Although it is not striking, the agreement between calculation and experiment is nevertheless reasonable for this kind of comparison. Of importance is the fact that the theoretical curve underestimates the Doppler shutdown and, hence, is a conservative one for safety considerations. The experiment also demonstrates the ability of performing Doppler studies on single rod samples by reactivity techniques as a first step in developing mathematical models to be applied to entire reactors.

Fig. 1 Doppler effect in a depleted uranium PBF pellet.

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B. NEUTRON INTERACTIONS WITH ATOMS

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SCATTERING LAW DATA FOR THE ENDF/A FILE

R. E. Schmunk, P. D. Randolph

Experimental scattering law $S(\alpha, \beta)$ data for liquid sodium at 100°C which were obtained in previous experiments [1,2] have been tabulated and submitted to the Sigma Center at Brookhaven National Laboratory for inclusion in the ENDF/A file. The range of variables of these data ($0.0003 \leq \alpha \leq 0.3$, $0.0 \leq \beta \leq 2.0$) covers the main range for which the microscopic dynamics of the sodium atom is influenced by being bound in a liquid.

Experimental scattering law data for beryllium at 22°C^[3] with parameters in the range $0.02 \leq \alpha \leq 2.14$, $0.6 \leq \beta \leq 4.0$ have also been submitted to the Sigma Center.

From recent phonon dispersion relation studies of zirconium at room temperature and the accompanying model fitting of these data (modified axially symmetric model) a frequency distribution was obtained and used as input data for the LEAP program to calculate a scattering law for Zr in the incoherent approximation. These calculations have been completed with parameters limited to the range $0.001 \leq \alpha \leq 10.0$, $0.10 \leq \beta \leq 5.0$. A plot of the calculated scattering law for Zr is shown in Figure 1. These data are now being prepared in the "B" file format for submission to the Sigma Center for inclusion in the ENDF/A file.

Following the same approach as was used for zirconium, a ten parameter shell model has been least-squares fitted to the dispersion relation of UO_2 ^[4] and work is progressing on programming the calculation of the frequency distribution for this material. Upon completion of the programming, the frequency distribution of UO_2 will be used as input data for LEAP to evaluate scattering law data for UO_2 for inclusion in the ENDF/A file.

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- [1] P. D. Randolph, "Slow Neutron Inelastic Scattering from Liquid Sodium", Phys. Rev., **134** (June 1964) pp A1238-A1248.
 - [2] P. D. Randolph, "Inelastic Scattering in Liquid Sodium at Small Momentum Transfer", in Neutron Inelastic Scattering. Proceedings of a Symposium on Inelastic Scattering Held by the International Atomic Energy Agency in Copenhagen, 20-25 May, 1968. Volume I, Vienna: International Atomic Energy Agency, 1968, pp 449-456.
 - [3] R. E. Schmunk, "Slow-Neutron Inelastic Scattering from Beryllium Powder", Phys. Rev., **136** (November 1964) pp A1303-A1311.
 - [4] G. Dolling, R. A. Cowley, A. D. B. Woods, "The Crystal Dynamics of Uranium Dioxide", Can. J. Phys., **43** (1965) pp 1397-1413.

Fig. 1 Scattering law $S(\alpha, \beta)$ for zirconium plotted as a function of momentum transfer for a given constant energy transfer. The scattering law was obtained by calculation using the frequency distribution of zirconium as input data for the LEAP program to evaluate the scattering law in the incoherent approximation.

C. NUCLEAR PROPERTIES

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THE DECAY OF ^{56}Co

R. J. Gehrke

The decay of 77.3-day ^{56}Co has been studied with a Ge(Li) spectrometer system. Gamma-ray energy and relative intensity measurements have been made and are reported in Table I. The intensities are in agreement with previous results[1-3]. The gamma-ray energies are in good agreement with those measured by Gunnink et al[4]. These measurements are consistent with the decay scheme proposed by Sher and Pate[1]. Six previously unreported gamma rays at 263, 896, 1159, 1335, 1442, and 1460 keV have been identified by half-life and placed in the decay scheme (see Figure 1). A spectrum of the gamma rays resulting from the decay of ^{56}Co is given in Figure 2. Three very weak gamma rays with energies of 1199, 1352, and 2275 keV may also belong to the decay of ^{56}Co . Shown in Figures 3, 4, and 5 are the gamma-ray spectra of these three energy regions on an expanded scale. These gamma rays were located with the help of a peak-finding method[5]. Two of these transitions are indicated by dashed lines in the decay scheme proposed in Figure 1. The gamma rays at 1335 and 1352 keV have not been placed in the ^{59}Fe level scheme. The gamma-ray transition of 3598 keV reported by Sher and Pate was not observed.

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TABLE I
GAMMA-RAY ENERGY AND INTENSITY MEASUREMENTS ON ^{56}Co

Present Work		Gunnink et al ^[4]	Sher and Pate ^[1]		Chasman and Ristinen ^[2]		Auble et al ^[3]	
E_Y (keV)	Relative intensity	E_Y (keV)	E_Y (keV)	Relative intensity	E_Y (keV)	Relative intensity	E_Y (keV)	Relative intensity
263.3 $\pm$ 0.3	0.05 $\pm$ 0.04							
733.4 $\pm$ 0.2	0.19 $\pm$ 0.10		732.5 $\pm$ 0.5	0.13 $\pm$ 0.06			732.2 $\pm$ 2.0	0.1 $\pm$ 0.05
788.06 $\pm$ 0.12	0.34 $\pm$ 0.10		788.6 $\pm$ 0.5	0.2 $\pm$ 0.1	788.6 $\pm$ 1.5	0.36	788.2 $\pm$ 2.0	0.4 $\pm$ 0.2
846.76 $\pm$ 0.05	100	846.782 $\pm$ 0.060	846.6 $\pm$ 0.5	100	846.64 $\pm$ 0.10	100	846.6 $\pm$ 0.5	100
896.7 $\pm$ 0.3	0.05 $\pm$ 0.01							
977.4 $\pm$ 0.1	1.25 $\pm$ 0.18		977.7 $\pm$ 0.5	1.01 $\pm$ 0.3	977.5 $\pm$ 0.5	1.5	977.8 $\pm$ 1.0	1.36 $\pm$ 0.36
1037.34 $\pm$ 0.05	12.8 $\pm$ 1.0	1037.851 $\pm$ 0.060	1037.6 $\pm$ 0.5	12.1 $\pm$ 0.8	1037.87 $\pm$ 0.20	14.0	1038.9 $\pm$ 1.0	12.8 $\pm$ 0.9
1140.2 $\pm$ 0.3	0.28 $\pm$ 0.08				1141.2 $\pm$ 1.0	0.17		
1159.7 $\pm$ 0.3	0.15 $\pm$ 0.05							
1175.1 $\pm$ 0.1	2.25 $\pm$ 0.20	1175.085 $\pm$ 0.070	1175.3 $\pm$ 0.5	2.2 $\pm$ 0.1	1175.5 $\pm$ 0.5	1.6	1175.2 $\pm$ 1.0	2.4 $\pm$ 0.2
1199 $\pm$ 2	0.04 $\pm$ 0.04							
1238.28 $\pm$ 0.06	68.1 $\pm$ 3.5	1238.290 $\pm$ 0.040	1238.0 $\pm$ 0.5	70.2 $\pm$ 2.5	1238.4 $\pm$ 0.2	64	1238.2 $\pm$ 0.5	69.5 $\pm$ 3.5
1335.5 $\pm$ 0.3	4.15 $\pm$ 0.10							
1352 $\pm$ 2	0.07 $\pm$ 0.07							
1360.26 $\pm$ 0.04	4.5 $\pm$ 0.6	1360.219 $\pm$ 0.040	1359.8 $\pm$ 0.5	4.2 $\pm$ 0.4	1360.2 $\pm$ 0.15	4.0	1359.2 $\pm$ 1.0	4.5 $\pm$ 0.3
1442.9 $\pm$ 0.4	0.16 $\pm$ 0.04							
1460 $\pm$ 2	0.05 $\pm$ 0.05							
1771.38 $\pm$ 0.15	15.0 $\pm$ 1.5	1771.33 $\pm$ 0.06	1771.4 $\pm$ 0.5	16.7 $\pm$ 1.0	1771.4 $\pm$ 0.15	14.0	1771.2 $\pm$ 1.0	16.1 $\pm$ 0.8
1810.6 $\pm$ 0.2	0.57 $\pm$ 0.08		1810.7 $\pm$ 0.5	0.5 $\pm$ 0.3			1811 $\pm$ 2.0	0.4 $\pm$ 0.2
1963.8 $\pm$ 0.3	0.65 $\pm$ 0.08		1963.4 $\pm$ 0.5	0.63 $\pm$ 0.2	1964.6 $\pm$ 1.2	0.68	1965.4 $\pm$ 1.0	0.59 $\pm$ 0.09
2015.24 $\pm$ 0.12	2.38 $\pm$ 0.40	2015.33 $\pm$ 0.07	2015.3 $\pm$ 0.5	2.9 $\pm$ 0.4	2015.4 $\pm$ 0.3	2.6	2015.3 $\pm$ 1.0	2.7 $\pm$ 0.2
2034.82 $\pm$ 0.10	7.08 $\pm$ 1.0	2034.90 $\pm$ 0.06	2034.9 $\pm$ 0.5	7.7 $\pm$ 0.5	2034.9 $\pm$ 0.3	6.6	2035.2 $\pm$ 0.5	7.4 $\pm$ 0.6
2113.1 $\pm$ 0.3	0.35 $\pm$ 0.05		2112.8 $\pm$ 0.5	0.32 $\pm$ 0.15	2113.0 $\pm$ 0.1	0.56	2113.2 $\pm$ 2.0	0.29 $\pm$ 0.05
2213.0 $\pm$ 0.3	0.30 $\pm$ 0.05		2213.0 $\pm$ 0.5	0.2 $\pm$ 0.02	2213.0 $\pm$ 1.5	0.60		
2275 $\pm$ 2	0.07 $\pm$ 0.07							
2598.53 $\pm$ 0.06	17.72 $\pm$ 0.9	2598.52 $\pm$ 0.05	2598.4 $\pm$ 0.5	17.0 $\pm$ 0.6	2598.70 $\pm$ 0.15	14.0	2598.5 $\pm$ 0.5	17.3 $\pm$ 0.9
3009.8 $\pm$ 0.2	0.54 $\pm$ 0.20		3009.8 $\pm$ 0.5	1.0 $\pm$ 0.1	3010.0 $\pm$ 0.4	0.6	3009.9 $\pm$ 1.0	0.9 $\pm$ 0.2
3202.1 $\pm$ 0.2	3.35 $\pm$ 0.60	3202.18 $\pm$ 0.07 ^[a]	3202.3 $\pm$ 0.5	2.8 $\pm$ 0.4	3201.9 $\pm$ 0.3	2.9	3202.6 $\pm$ 1.0	3.4 $\pm$ 0.2
3253.5 $\pm$ 0.2	8.30 $\pm$ 1.2	3253.61 $\pm$ 0.06 ^[a]	3253.6 $\pm$ 0.5	7.3 $\pm$ 0.5	3253.7 $\pm$ 0.2	7.2	3254.5 $\pm$ 1.0	7.8 $\pm$ 0.4
3273.08 $\pm$ 0.10	1.90 $\pm$ 0.40	3273.16 $\pm$ 0.07 ^[a]	3273.1 $\pm$ 0.5	1.5 $\pm$ 0.4	3273.2 $\pm$ 0.3	1.6	3274.1 $\pm$ 1.0	1.5 $\pm$ 0.3
3451.5 $\pm$ 0.3	1.08 $\pm$ 0.20	3451.29 $\pm$ 0.10 ^[a]	3451.3 $\pm$ 0.5	0.83 $\pm$ 0.1	3452.1 $\pm$ 0.3	0.72	3451.1 $\pm$ 1.0	0.87 $\pm$ 0.09
3548.3 $\pm$ 0.4	0.20 $\pm$ 0.10		3548.2 $\pm$ 0.5	0.15 $\pm$ 0.5	3548.1 $\pm$ 0.3	0.2	3547.6 $\pm$ 2.0	0.15 $\pm$ 0.03
			3598.7 $\pm$ 0.5	0.02 $\pm$ 0.01				

^[a] These gamma-ray energies were obtained by measuring the double escape peak and adding 2 m_0c^2 .

Fig. 1 Proposed decay scheme for ^{56}Co .

Fig. 2 Gamma-ray spectrum of ^{56}Co on Ge(Li) spectrometer.

Fig. 3 Expanded plot of spectrum showing 1199 keV gamma ray.

Fig. 4 Expanded plot of region showing 1352 keV gamma ray.

Fig. 5 Expanded plot of region showing 2275 keV gamma ray.

ENERGY AND INTENSITY MEASUREMENTS OF GAMMA RAYS IN THE DECAY OF ^{69}As , ^{101}Pd , ^{94}Tc , AND ^{94m}Tc

R. J. Gehrke

The following is a list of gamma-ray energies and intensities of a few of the isotopes studied during this fiscal year with Ge(Li) spectrometers. Gamma rays which have not been previously reported are preceded by an asterisk.

<u>15-min ^{69}As</u>		<u>52-min ^{94m}Tc</u>	
<u>E_γ</u>	<u>I_{rel}</u>	<u>E_γ</u>	<u>I_{rel}</u>
86.9 $\pm$ 0.3	35.4 $\pm$ 4	871.01 $\pm$ 0.08	100
*146.10 $\pm$ 0.2	50.9 $\pm$ 5	*875.45 $\pm$ 0.12	1.6 $\pm$ 0.1
232.73 $\pm$ 0.10	100	993.01 $\pm$ 0.10	2.3 $\pm$ 0.8
*287.4 $\pm$ 0.4	13.3 $\pm$ 1.5	1195.8 $\pm$ 0.4	0.9 $\pm$ 0.2
		1521.52 $\pm$ 0.17	5.6 $\pm$ 1.1
		1868.50 $\pm$ 0.25	6.2 $\pm$ 0.4
		2393 $\pm$ 1	0.5 $\pm$ 0.2
		2739.7 $\pm$ 0.3	4.5 $\pm$ 0.5
		3128.2 $\pm$ 0.4	2.7 $\pm$ 0.7

8.5-hr ^{101}Pd

E_γ	I_{rel}
269.6 ± 0.2	39.03
296.1 ± 0.2	100
320.6 ± 0.2	3.28
*355.2 ± 0.3	1.10
*428.2 ± 0.4	1.13
453.5 ± 0.25	0.25
566.0 ± 0.2	20.30
590.54 ± 0.15	69.66
723.8 ± 0.4	14.40
748.6 ± 0.4	2.58
*854.0 ± 1.0	0.67
*881.0 ± 1.0	0.84
992.8 ± 0.4	4.90
1177.7 ± 0.4	3.01
1201.8 ± 0.4	9.02
1218.3 ± 0.4	3.42
1288.7 ± 0.4	13.28
1311.4 ± 0.6	1.37
*1638.0 ± 2.5	0.45

4.8-hr ^{94}Tc

E_γ	I_{rel}
449.07 ± 0.26	2.6 ± 1.0
532.13 ± 0.10	2.6 ± 1.0
702.60 ± 0.08	99.8 ± 6
742.2 ± 0.3	1.2 ± 0.6
849.65 ± 0.08	97.7 ± 6
871.01 ± 0.08	100
916.12 ± 0.10	7.4 ± 1.5
*1362.7 ± 0.6	0.9 ± 0.4
1508.6 ± 0.7	0.66 ± 0.3
1591.7 ± 0.3	2.41 ± 1.0

ENERGIES OF THE GAMMA RAYS FROM ^{94}Nb AND ^{95}Zr

R. P. Schuman

The long-lived ground state of ^{94}Nb is an ideal nuclide for gamma standards. Its long half-life of $(2.03 \pm 0.16) \times 10^4$ years [1] and the fact that it emits two gamma rays in cascade means its decay rate can be precisely determined by γ - γ coincidence counting and that the rate will show essentially no change with time.

Since we have been using purified ^{94}Nb as a routine gamma counting standard, it seemed useful to determine the gamma-ray energies more precisely. The ^{94}Nb was first counted with ^{207}Bi , ^{60}Co , ^{197}Au , ^{137}Cs , and ^{54}Mn standards using a Ge(Li) counter and a 1024 channel analyzer. The nonlinearity of the analyzer was determined from the positions of the standard peaks. Then a window amplifier was used to expand the portion of the spectrum from about 600 to 1200 keV and the purified ^{94}Nb again counted with ^{137}Cs , ^{95}Nb (from a ^{95}Zr - ^{95}Nb sample), ^{88}Y , ^{54}Mn , and ^{207}Bi . The ^{94}Nb energies, as determined versus ^{198}Au (411.795 ± 0.009) [2], ^{207}Bi (569.63 ± 0.08 keV, 1063.63 ± 0.07) [2,3], ^{137}Cs (661.595 ± 0.076) [2], ^{95}Nb (765.83 ± 0.07) [3], ^{54}Mn (834.84 ± 0.07) [3], ^{88}Y (898.01 ± 0.07) [3], and ^{60}Co (1173.226 ± 0.040 , 1332.483 ± 0.046) [2], are: 702.72 ± 0.10 and 871.10 ± 0.10 keV. The energies of the ^{95}Zr gamma rays were determined as 724.36 ± 0.10 and 756.92 ± 0.10 keV.

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DECAY OF ^{102}Rh AND $^{102\text{m}}\text{Rh}$ [1]

R. J. Gehrke, R. G. Helmer

The decay of the 206-day and 2.9-yr electron-capture activities of ^{102}Rh have been studied with a Ge(Li) spectrometer and with a γ - γ coincidence system. From the analysis of γ -ray spectra, measured over a period of ≈ 1.5 years, the γ rays associated with each activity were identified. Twenty-four transitions, nine previously unreported, are assigned to the decay of 206-day ^{102}Rh , and fifteen, six previously unreported, to 2.9-yr ^{102}Rh . All observed transitions except one are placed in a consistent level scheme for ^{102}Ru with levels at 0, 475, 944, 1103, 1106, 1522, 1581, 1837, 2037, and 2261 keV populated by the 206-day activity; and 0, 475, 1103, 1106, 1522, 1799, 1873, and 2219 keV from the 2.9-yr activity. The levels at 944, 1581, and 1799 keV have not been previously reported. The log ft values of the various electron-capture branches are consistent with a spin of 1 for the 206-day level of ^{102}Rh and with a spin ≥ 4 for the 2.9-yr state. A reasonable assignment for the new 944-keV level is $0+$, which suggests that it is associated with the $2+$ and $4+$ two-phonon states reported at 1103 and 1106 keV, respectively.

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- [1] Published in The Physical Review, Vol. 177, 1792-1797.

NUMERICAL CONSTRUCTION OF LEVEL SCHEMES -- ^{149}Pm [1]

R. G. Helmer, A. Bäcklin^[2]

A method of constructing a more complete level scheme from the energies of the associated gamma-ray transitions and a few known levels is illustrated. The number of possible new levels obtained in an energy region is compared with the number of levels expected from a set of random transition energies. In certain cases, this allows one to conclude, with a known probability of being right, that the possible level does exist. The method is illustrated by construction of the level scheme of ^{149}Pm from the 155 transitions observed in the decay of ^{149}Nd .

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- [1] Published in Nuclear Instruments and Methods **65** (1968) pp 31-44.
 - [2] Institute of Physics, University of Uppsala and The Swedish Research Council's Laboratory, Studsvik, Nyköping, Sweden.

LEVELS IN ^{156}Gd FROM $^{155}\text{Gd}(n,\gamma)$

R. C. Greenwood, C. W. Reich

Information about the level structure of ^{156}Gd has been obtained from measurements of the prompt gamma rays which are emitted as a result of neutron capture in ^{155}Gd . Various Ge(Li) detectors were used for these measurements.

Capture gamma-ray spectra below 3 MeV were principally measured using a $6\text{ cm}^2 \times 8\text{ mm}$ Ge(Li) detector which had an energy resolution of 1 keV at 100 keV and 1.7 keV at 1 MeV. Some measurements were also made with a 35 cm^3 coaxial detector. Extensive energy calibrations were made. From these measurements some 145 gamma rays were observed below 2.5 MeV.

Measurements of the high energy portion of the capture gamma-ray spectrum were made using a thermal neutron beam and a 2-keV neutron beam. Because the thermal neutron cross section of ^{155}Gd is overwhelmingly contributed by a 2^- resonance the primary capture gamma-ray transitions will populate levels with spins of 1, 2, and 3 by dipole transitions. The energy spread of the 2-keV neutron beam has a FWHM of 700 eV. Thus the resultant capture gamma-ray spectra are an average over many neutron 1^- and 2^- resonances. Because of this the statistical fluctuations in the partial radiation width are averaged out so that E1 transitions are significantly stronger than M1 transitions. Furthermore, final states with spins and parities of 1^+ or 2^+ were observed to be populated about twice as strongly as final states which had spins of 0^+ or 3^+ .

The ^{155}Gd level scheme shown in Figure 1 was obtained by combining the primary and the low energy capture gamma-ray data. All of the strongest low energy gamma-ray transitions have been accounted for as de-excitations of these levels. A schematic representation of this level scheme, from which the de-excitation modes of the levels are omitted, is shown in Figure 2. A heavy arrow into the level indicates that it is populated by primary transitions, while the number associated with the arrow out of each level gives the total intensity (relative to an intensity of 100 for the 1257-keV gamma-ray transition) of the de-excitation gamma-ray transitions.

Considering now some of the specific features of this level scheme:

The 0^+ and 2^+ members of the beta-vibrational band have previously been identified both in (d,d') measurements and in the decay of ^{156}Eu as levels at 1049 keV and 1129 keV. These levels are strongly populated in the $^{155}\text{Gd}(n,\gamma)$ reaction. By combining the prompt internal conversion intensities measured by Prokof'ev et al[1] with our gamma-ray intensities we find that the 1040-keV transition between the 1129-keV level and the 2^+ member of the ground-state band contains a significant E0 component, further confirming that the 1129-keV level has $K^\pi = 0^+$. The 1297-keV level is assigned as the 4^+ member of this band. This assignment is strengthened by the fact that the 1009-keV transition contains a significant E0 component. Ratios of the reduced transition probabilities from levels in the beta-vibrational band to levels in the ground-state band are given in Table I. As is observed for other beta vibrational bands in this region, a single value of the band mixing parameter Z_0 cannot fit these ratios.

A second $K^\pi = 0^+$ band has been established at 1168 keV. The 1462-keV level is tentatively assigned here as the 4^+ member of this band. There are some problems with this assignment; principally that a 1462-keV level was observed in the (d,d') measurements of Bloch et al[2] whereas the 0^+ and 2^+ members are apparently not seen. [Although a case might be made for the fact that they are masked by other stronger lines in the (d,d') spectrum.] However, comparison of the gamma-ray intensities and the internal conversion intensities[1] suggests that the level has positive parity, and the fact that a primary capture gamma-ray transition is not observed to it in the 2-keV neutron capture data requires that its spin be ≥ 4 . Hence, any other reasonable spin and parity assignment for this level is even more difficult to reconcile with the (d,d') data. Ratios of the reduced transition probabilities from levels in this $K^\pi = 0^+$ band to levels in the ground-state band are given in Table I. These ratios are not fitted well by a pure rotational model; however, neither are they fitted with a single value of the band mixing parameter Z_0 .

The 2^+ , 3^+ , and 4^+ members of the gamma vibrational band are well established. The ratio of the reduced transition probabilities for decay from the 4^+ member of this band to members of the ground-state band is fitted with a band mixing parameter $Z_2 \sim 0.047$. We have tentatively assigned the 1506-keV level as the 5^+ member of this band. (Transition intensities from this level would require $Z_2 \sim 0.024$.) With this assignment we obtain a rather significant odd-even shift in the band.

The situation with the negative parity states is presently not well understood. The data seem to indicate that these states are members of strongly mixed (Coriolis coupled) $K^\pi = 0^-$ and $K^\pi = 1^-$ bands. However, it is not clear which of these bands is the lowest in energy.

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TABLE I

RATIOS OF REDUCED TRANSITION PROBABILITIES FOR $K^\pi = 0^+$ BANDS

Level Energy (keV)	Ratio RTP	Experimental Ratios	Pure Rotation	Z_0 ($\times 10^3$)
Beta vibration band				
1129	$\frac{B(2;20 \rightarrow 00)}{B(2;20 \rightarrow 20)}$	0.22 (2)	0.70	74 ± 4
	$\frac{B(2;20 \rightarrow 40)}{B(2;20 \rightarrow 20)}$	1.12 (9)	1.80	-15 ± 2
	$\frac{B(2;20 \rightarrow 00)}{B(2;20 \rightarrow 40)}$	0.19 (2)	0.39	18 ± 2
1297	$\frac{B(2;40 \rightarrow 20)}{B(2;40 \rightarrow 40)}$	0.35 (4)	1.10	31 ± 2
	$\frac{B(2;40 \rightarrow 60)}{B(2;40 \rightarrow 40)}$	1.18 (48)	1.75	-8 ± 8
	$\frac{B(2;40 \rightarrow 20)}{B(2;40 \rightarrow 60)}$	0.30 (12)	0.63	11 ± 5
Other $K^\pi = 0^+$ band				
1257	$\frac{B(2;20 \rightarrow 00)}{B(2;20 \rightarrow 20)}$	0.21 (3)	0.70	77 ± 5
	$\frac{B(2;20 \rightarrow 40)}{B(2;20 \rightarrow 20)}$	2.8 (2)	1.80	18 ± 4
	$\frac{B(2;20 \rightarrow 00)}{B(2;20 \rightarrow 40)}$	0.07 (1)	0.39	47 ± 4
1462	$\frac{B(2;40 \rightarrow 20)}{B(2;40 \rightarrow 40)}$	<0.05	1.10	>56
	$\frac{B(2;40 \rightarrow 60)}{B(2;40 \rightarrow 40)}$	1.37 (21)	1.75	-5 ± 3

Fig. 1 Levels in ^{156}Gd populated by the $^{155}\text{Gd}(n,\gamma)$ reaction.

Fig. 2 Level scheme of ^{156}Gd with the de-excitation modes of the levels omitted.

LEVELS IN ^{158}Gd FROM $^{157}\text{Gd}(n,\gamma)$

R. C. Greenwood, C. W. Reich

Analysis of the $^{157}\text{Gd}(n,\gamma)$ data continued during the past year. A preliminary analysis of these data together with details of the experimental techniques used to obtain the data were given in Reference 1.

The emphasis of our more recent analysis of the $^{157}\text{Gd}(n,\gamma)$ data was to obtain information about the collective states which exist in ^{158}Gd below 2 MeV. The results of this analysis are illustrated in the level scheme for ^{158}Gd shown in Figure 1. A schematic presentation of this level scheme, omitting details of the level decays, is shown in Figure 2. In Figure 2, a solid arrow into a level indicates that it is populated by primary transitions in thermal neutron capture, and the numbers associated with the arrows out of each level give the total intensities (relative to an intensity of 100 for the 1107-keV gamma-ray transition) of the de-excitation gamma-ray transitions. These latter intensities have proven to be quite useful in helping us construct this level scheme.

Considering now some of the specific features of this level scheme: Bloch et al[2] suggested, based upon their (d,d') experiments, that collective 3^- states existed in ^{158}Gd at 1042, 1402, and 1856 keV. The existence of these 3^- states is confirmed in this present work and they are assigned here as members of the $K^\pi = 1^-, 0^-,$ and 2^- octupole vibration bands, respectively. The assignment of the 977-keV band as $K^\pi = 1^-$ is suggested both by the presence of even-spin odd-parity levels which appear to be members of the band and the log foot values of the electron capture branches from ^{158}Tb to the 1^- , 3^- , and 5^- states[3] are much too low to correspond to K-forbidden transitions.

The assignment of the 1451-keV band as the beta-vibrational band[2] is supported by comparison of our gamma-ray intensities with the conversion-electron intensities measured by Bäcklin[4], which indicates that the $\Delta I = 0$ transitions have an EO component. As noted in Table I, the ratios of reduced transition probabilities from the beta-vibrational band into the ground-state band cannot be fitted with a single value of the band mixing parameter Z_0 . This observation is consistent with the data available on such bands in other nuclei in this region.

In addition to the beta-vibrational band a second $K^\pi = 0^+$ band is observed at 1198 keV. The (d,p) data of Shelton[5] provided us with an understanding of the makeup of this band. In the (d,p) experiments, the 1198- and 1259-keV states in the $K^\pi = 0^+$ band are unresolved from the 1187- and 1265-keV states, respectively, in the gamma-vibrational band. If allowance is made for the expected (calculated) contribution of proton groups from the gamma-vibrational band members to the observed groups, the remaining intensity pattern into the $K^\pi = 0^+$ band is very similar to that into the ground-state band. This suggests that there is a significant contribution from the $(3/2^-[521], 3/2^-[521])_{0^+}$ configuration in this band. The ratios reduced transition probabilities from this $K^\pi = 0^+$ band into the ground state band are shown in Table I. These ratios are not fitted well by a pure rotational model, but neither are they fitted with a single value of the band mixing parameter Z_0 .

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TABLE I

RATIOS OF REDUCED TRANSITION PROBABILITIES FOR $K^\pi = 0^+$ BANDS

Level Energy (keV)	Ratio RTP	Experimental Ratios	Pure Rotation	Z_0 ($\times 10^3$)
Beta Vibration Band				
1517	$\frac{B(2;20 \rightarrow 00)}{B(2;20 \rightarrow 20)}$	0.49 (8)	0.70	28 (12)
	$\frac{B(2;20 \rightarrow 40)}{B(2;20 \rightarrow 20)}$	0.82 (25)	1.80	-23 (7)
	$\frac{B(2;20 \rightarrow 00)}{B(2;20 \rightarrow 40)}$	0.59 (20)	0.39	-10 (10)
1667	$\frac{B(2;40 \rightarrow 20)}{B(2;40 \rightarrow 40)}$	1.05 (44)	1.10	2 (14)
Other $K^\pi = 0^+$ Band				
1259	$\frac{B(2;20 \rightarrow 00)}{B(2;20 \rightarrow 20)}$	0.84 (10)	0.70	-15 (11)
	$\frac{B(2;20 \rightarrow 40)}{B(2;20 \rightarrow 20)}$	3.50 (48)	1.80	28 (7)
	$\frac{B(2;20 \rightarrow 00)}{B(2;20 \rightarrow 40)}$	0.24 (2)	0.39	13 (2)
1406	$\frac{B(2;40 \rightarrow 20)}{B(2;40 \rightarrow 40)}$	1.5 (5)	1.10	-12 (15)
	$\frac{B(2;40 \rightarrow 60)}{B(2;40 \rightarrow 40)}$	1.9 (1.1)	1.75	2 (12)

Fig. 1 Levels in ¹⁵⁸Gd populated by the ¹⁵⁷Gd(n,γ) reaction.

Fig. 2 Schematic representation of the level scheme in Figure 1.

LEVELS IN ^{164}Dy FROM 2-keV NEUTRON CAPTURE

R. A. Harlan

The scandium-filtered 2-keV neutron beam has been used to populate final states in ^{164}Dy . The utility of this beam as a tool in neutron capture gamma-ray spectroscopy has been discussed[1]. Because of the finite energy spread in this beam (≈ 700 eV), capture should occur in ≈ 70 resonances. One should observe essentially all primary transitions to spin 1, 2, 3, and 4 final states, subject to limitations imposed by the increasing level density at higher excitation energies and by the lower efficiency of the Ge(Li) detector at intermediate (≈ 3 -4 MeV) gamma-ray energies. The results are summarized in Table I.

Figure 1 shows a plot of the relative gamma-ray intensities for the first 37 observed levels divided by $E\gamma^5$. The $E\gamma^5$ energy dependence was used since Bollinger and Thomas[2] empirically determined the $E\gamma^5$ dependence was approximately followed at least for the first 2 MeV of excitation in ^{168}Er . There is a clear separation of E1 and M1 transition intensities below ≈ 2 MeV in excitation. The experimental conditions were such that one is confident of having observed essentially all E1 transitions to positive parity states. The M1 transitions to negative parity final states, however, were barely above the Compton scattering background. Consequently, only seven M1 transitions were identified. There are three E1 transitions which are significantly stronger than the others, which may indicate they are unresolved doublets. These are indicated in a footnote to Table I. Unfortunately, the data in Figure 1 indicate the Porter-Thomas fluctuations have not been averaged to the point that one can separate the spin 1 and 4 final states from the spin 2 and 3 final states. To do so would apparently require an average over a larger energy interval than provided by the scandium-filtered beam, for example in the 24 keV beam being developed at the MTR.

In summary, we have established the parity and limited the spin on a large number of final states in ^{164}Dy and discovered a number of levels which were not populated by thermal neutron capture[3].

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TABLE I

LEVELS IN ^{164}Dy POPULATED BY 2 keV NEUTRON CAPTURE ON ^{163}Dy

<u>Gamma-Ray Energy</u>	<u>Level Energy</u>	<u>Relative Intensity</u>	<u>I_{π}</u>
7586.3	73.4	1.00	2+
7417.4	242.4	0.88	4+
6898.3	761.4	0.69	2+
6832.1	827.6	0.83	3+
6744.4	915.3	0.41	4+
6682.4	977.3	0.10	2-
6072.8	1586.9	0.048	1 to 4-
5944.3	1715.5	0.32	1 to 4+
5922.1	1737.6	0.076	1 to 4-
5901.9	1757.8	0.045	1 to 4-
5883.9	1775.8	0.22	1 to 4+
5863.7	1796.1	0.31	1 to 4+
5851.6	1808.2	0.080	1 to 4-
5818.3	1841.5	0.083	1 to 4-
5808.5	1851.2	0.21	1 to 4+
5768.1	1891.7	0.17	1 to 4+
5738.8	1920.9	0.135	1 to 4+
5727.5	1932.3	0.20	1 to 4+
5681.6 ^[a]	1977.8	0.43	1 to 4+
5676.0?	1983.7?	0.060	1 to 4-?
5608.8 ^[a]	2050.9	0.40	1 to 4+
5582.4	2077.3	0.15	1 to 4+
5558.7	2101.0	0.21	1 to 4+
5547.1	2112.6	0.18	1 to 4+
5536.6	2123.1	0.21	1 to 4+
5506.9	2152.8	0.23	1 to 4+
5466.3	2193.4	0.14	1 to 4+
5453.8	2205.9	0.21	1 to 4+
5412.4	2247.3	0.15	1 to 4+

TABLE I (Contd.)

LEVELS IN ^{164}Dy POPULATED BY 2 keV NEUTRON CAPTURE ON ^{163}Dy

<u>Gamma-Ray Energy</u>	<u>Level Energy</u>	<u>Relative Intensity</u>	<u>I_{π}</u>
5399.2	2260.6	0.13	1 to 4+
5357.1	2302.6	0.26	1 to 4+
5311.9 ^[a]	2347.8	0.32	1 to 4+
5269.4	2390.3	0.10	1 to 4+
5250.1	2409.7	0.10	1 to 4+
5230.7	2428.6	0.11	1 to 4+
5224.0	2435.5	0.14	1 to 4+
5218.1	2442.1	0.15	1 to 4+
5202.0		0.10	
5190.2		0.13	
5166.9		0.19	
5144.0		0.13	
5127.2		0.12	
5107.4		0.091	
5092.2		0.13	
5077.2		0.14	
5067.1		0.16	
4951.5		0.11	
4925.3		0.14	
4909.2		0.15	
4873.5		0.16	
4861.4		0.19	
4838.3		0.21	
4774.7		0.19	
4742.3		0.18	
4716.0		0.12	
4669.9		0.11	
4658.2		0.12	
4586.4		0.15	

TABLE I (Contd.)

LEVELS IN ^{164}Dy POPULATED BY 2 keV NEUTRON CAPTURE ON ^{163}Dy

<u>Gamma-Ray Energy</u>	<u>Level Energy</u>	<u>Relative Intensity</u>	<u>I_{π}</u>
4548.3		0.15	
4536.2		0.088	
4083.3		0.16	

[a] Possible unresolved doublets.

Fig. 1 Plot of relative gamma-ray intensity $\div E_{\gamma}^5$ versus energy for the $^{163}\text{Dy}(n, \gamma)^{164}\text{Dy}$.

MIXING OF THE GAMMA-VIBRATIONAL AND GROUND-STATE BANDS IN ^{166}Er

C. W. Reich, J. E. Cline

Investigations of the properties of the gamma-vibrational band in ^{166}Er continue. The relative intensities of the gamma rays emitted following the decay of the 27-hr and 1.2×10^3 -yr activities of ^{166}Ho have been measured with improved precision to study the effects of band mixing on the E2 transitions between the gamma-vibrational and ground-state bands. The emphasis of this aspect of the study centered around the question of the equality of the intrinsic quadrupole moments of the two bands. One recently reported analysis in ^{166}Er [1] indicated that the intrinsic quadrupole moment of the gamma band was $\approx 40\%$ larger than that of the ground-state band. Since the band-mixing equations upon which this analysis was based assumed equality of the two moments, we felt that a further investigation of the problem was of interest.

The analysis was made using two different, but completely equivalent, approaches. The first is the conventional one, in which the mixing is characterized by a parameter z [2], although we have generalized it to include the situation where the intrinsic quadrupole moments of the two bands are allowed to differ. The second is the generalized procedure first presented by Mikhailov[3] and applied to ^{166}Er by Mottelson[4].

In the first approach, the reduced transition probabilities for the interband E2 transitions are related to the "unmixed" ones through the relationship[2]

$$B(E2; I, K=2 \rightarrow I', K=0)_{\text{mixed}} = B(E2; I, 2 \rightarrow I', 0)_{\text{unmixed}} \times [1 + f(I, I', z)]^2.$$

When the intrinsic quadrupole moments of the two bands are assumed to be unequal, the general functional form remains the same, but one now has two z -parameters. The relevant equations are summarized in Table I. An inspection of Table I reveals that the $B(E2)$ branching ratios from the odd-spin states in the gamma band do not depend on $z_2(0)$ and hence may be used to determine $z_2(2)$. Using the value thus obtained, one can determine $z_2(0)$ from the branching ratio data involving the even-spin states. With these values thus determined, one can use the ratio of intraband to interband transition probabilities to determine the ratio of intraband to interband transition matrix elements. The results of these three stages of the analysis are summarized in Table II. It is apparent from these data that there is no significant difference in $z_2(2)$ and $z_2(0)$ and, hence, in the intrinsic quadrupole moments of the two bands. From the data in Table II, and the recently reported[5] value of $(0.128 \pm 0.013) e^2 \times 10^{-48} \text{ cm}^4$ for $B(E2; 00 \rightarrow 22)$, we obtain a value of 7.42 ± 0.36 barns for $Q_0(2)$, the intrinsic quadrupole moment in the gamma band of ^{166}Er . This is in good agreement with the value of (7.62 ± 0.14) barns, obtained from various experiments (see Reference 6), for the intrinsic quadrupole moment, $Q_0(0)$ of the ground state band.

The fact that our analysis yields a different result from that of Reference 1 we attribute to the following:

- (1) The $B(E2;00-22)$ value used in our analysis is smaller than the value which was available when the analysis[1] was carried out.
- (2) Our values for the relative intensities of the relevant gamma-ray transitions differ significantly and systematically from those in Reference 1.

The effect of both is to lower the value of $Q_0(2)$ relative to that of $Q_0(0)$.

In the approach of Mikhailov[3], the interband $B(E2)$ values are expressed in the form

$$B(E2;I,K=2 \rightarrow I',K=0) = 2 \langle I \ 2 \ 2 \ 2 | I'0 \rangle^2 |M_1 + M_2 [I'(I'+1) - I(I+1)]|^2,$$

where it has been assumed that the two intrinsic quadrupole moments are equal. On this basis, a plot of the square root of a given $B(E2)$ value divided by the respective Clebsch-Gordan coefficient should exhibit a linear dependence on the quantity $I'(I'+1) - I(I+1)$. If the two moments are unequal, additional terms appear in the above expression and the linear dependence no longer obtains. Our gamma-ray relative intensity data, expressed in this format, are presented in Figure 1. It is evident that the straight line shown, which results from a least-squares analysis, provides a quite excellent fit to the data. Our data provide no evidence for a departure from the linear dependence and, hence, no evidence for a difference in the two moments. (This merely restates in a different form the results of our alternate analysis above.) The band-mixing parameter, z , is related to the quantities M_1 and M_2 in the following manner^[3,4]

$$z = \frac{2 M_2}{M_1 - 4 M_2}.$$

From our data, we obtain a value of 0.0415 for z .

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TABLE I

SUMMARY OF RELEVANT EQUATIONS

$B(E2; I2 \rightarrow I'0) = B(E2)_0 \{1 + f(I, I')\}^2$		
Initial State (K = 2)	Final State (K = 0)	f(I, I')
I-2	I	$\frac{1}{2} \{(I+1)(I+2)z_2(2) - I(I-1)z_2(0)\}$
I-1	I	$(I+2)z_2(2)$
I	I	$\frac{1}{6} \{(12-I(I+1))z_2(2) + I(I+1)z_2(0)\}$
I+1	I	$-(I-1)z_2(2)$
I+2	I	$\frac{1}{2} \{I(I-1)z_2(2) - (I+1)(I+2)z_2(0)\}$
$z_2(0) = \epsilon \sqrt{24} \langle 0 M'(E2; 0) 0 \rangle / \langle 0 M'(E2; -2) 2 \rangle$		
$z_2(2) = \epsilon \sqrt{24} \langle 2 M'(E2; 0) 2 \rangle / \langle 0 M'(E2; -2) 2 \rangle$		

TABLE II
ANALYSIS OF MIXING OF GROUND-STATE AND
GAMMA-VIBRATIONAL BANDS IN ^{166}Er

Transitions ($IK \rightarrow I'K'$)	B(E2) ratio (experimental)	Mixing parameter
(72+80)/(72+60)	2.11 ± 0.22	$z_2(2) = 0.041 \pm 0.004$
(52+60)/(52+40)	1.32 ± 0.03	$z_2(2) = 0.042 \pm 0.001$
(32+40)/(32+20)	0.75 ± 0.05	$z_2(2) = 0.050 \pm 0.006$
$\langle z_2(2) \rangle = 0.042 \pm 0.002$		
(82+80)/(82+60)	18.2 ± 3.6	$z_2(0) = 0.040 \pm 0.002$
(62+60)/(62+40)	10.8 ± 0.3	$z_2(0) = 0.0413 \pm 0.0004$
(42+40)/(42+20)	6.14 ± 0.12	$z_2(0) = 0.047 \pm 0.001$
(22+20)/(22+00)	1.86 ± 0.04	$z_2(0) = 0.046 \pm 0.005$
$\langle z_2(0) \rangle = 0.042 \pm 0.002$		
(82+62)/(82+60)	519 ± 100	$R^{2*} = 34 \pm 7$
(72+52)/(72+60)	56.0 ± 1.5	$R^2 = 39.6 \pm 0.8$
(62+42)/(62+40)	233 ± 9	$R^2 = 38.6 \pm 1.6$
(52+32)/(52+40)	31.0 ± 2.0	$R^2 = 39.6 \pm 2.4$
$\langle R^2 \rangle = 39.4$		

$$* R \equiv \langle 2 | M'(E2; 0) | 2 \rangle / \sqrt{2} \langle 0 | M'(E2; -2) | 2 \rangle$$

Fig. 1 "Mikhailov" [3] plot of the interband E2 transition data for ^{166}Er .

EXCITED $K^\pi = 0^+$ BANDS IN ^{172}Yb

R. C. Greenwood, C. W. Reich, R. A. Harlan

Recent investigations of the energy levels of several deformed even rare-earth nuclides have revealed the existence of numerous (3 to 5) bands having $K^\pi = 0^+$ in the energy region between about 1 and 2 MeV. These studies suggest that the occurrence of these bands may be a systematic feature of all such nuclides. In favorable cases the prompt gamma-ray spectrum resulting from neutron capture in an odd-N target nucleus can provide a direct measure of the number and distribution of such 0^+ states. The $^{171}\text{Yb}(n,\gamma)$ reaction is such a case. Here, s-wave neutron capture populates compound-nucleus states having $I^\pi = 0^-$ and 1^- . Subsequent de-excitation of these states via primary E1 transitions will excite 0^+ , 1^+ , and 2^+ final states. If the neutron capture reaction proceeds via one or a few such compound-nucleus resonances (as generally occurs in thermal-neutron capture), large statistical fluctuations are observed in the intensities of the primary transitions to final states of the same spin-parity. Hence primary E1 and M1 transitions cannot be unambiguously distinguished, and some of the weaker primary E1 transitions may not be observed. We have used the 2-keV neutron beam from the scandium-filter facility of the MTR^[1] to investigate the $^{171}\text{Yb}(n,\gamma)$ reaction. Since this beam has a measured energy spread of ≈ 0.7 keV (FWHM) the resultant capture gamma-ray spectrum, shown in Figure 1, represents an average over ≈ 100 compound-nucleus resonances. Hence, the statistical fluctuations in the primary gamma-ray intensities should be averaged out. Since the E1 transitions are intrinsically stronger than M1 transitions, the multipolarities of the primary gamma rays may be inferred from their intensities, as illustrated in Figure 2. Below 1.7 MeV the states excited by the E1 primary transitions may be grouped into bands with the following K^π assignments and energies: 0^+ , ground state; 0^+ , 1043.0 keV; 0^+ , 1405.1 keV; 2^+ , 1465.9 keV; 2^+ , 1608.4 keV. The two $K^\pi = 2^+$ states and the 0^+ band based at 1043.0 keV have previously been observed in decay-scheme studies^[2,3] and (d,p) and (d,t) reactions^[4]. The 0^+ state at 1405.1 keV has not been previously observed. The 0^+-2^+ energy spacings of the $K = 0$ bands at 0, 1043, and 1405 keV are 78.8, 75.0, and 71.7 keV, respectively. Above 1.7 MeV, the ordering of the observed positive-parity states into rotational bands is not apparent using only the data shown here, although the $K^\pi = 1^+$ assignment proposed for a band at 2010 keV by Burke and Elbek^[4] appears to be substantiated. Definite band assignments for the positive-parity states above 1.7 MeV must await a detailed analysis of the gamma-ray transitions de-exciting them, which is currently in progress. These data should indicate the number of excited $K^\pi = 0^+$ bands occurring between 1.0 and ≈ 2.4 MeV in ^{172}Yb .

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Fig. 1 A portion of the prompt gamma-ray spectrum resulting from 2-keV neutron capture in ^{171}Yb .

Fig. 2 A plot of $I_\gamma E_\gamma^{-5}$ versus the energy of the prompt gamma-ray transitions observed from 2-keV neutron capture in ^{171}Yb . (The I_γ represents the relative intensity of the prompt gamma-ray transitions.)

RESONANCE-NEUTRON CAPTURE TO LEVELS IN ^{171}Yb , ^{172}Yb , AND ^{173}Yb

R. A. Harlan, R. R. Spencer, K. T. Faler

Data on capture of resonance-energy neutrons on enriched targets of ^{170}Yb , ^{171}Yb , and ^{173}Yb have been analyzed. Two types of neutron sources were used. First, a beryllium-crystal neutron spectrometer was used to provide monoenergetic beams for energies below ≈ 20 eV; the energies were selected to correspond to known resonances in the selected target isotopes. The second technique involved filtering the raw beam from the tangential hole in the MTR with 1/16- and 1/8-inch $^{10}\text{B}_4\text{C}$ filters; these beams covered a broad spectrum with a width of several hundred eV.

The first technique offered the possibilities of restricting the final state spin assignments from capture in states with known spin or of determining the spin of the capturing state by observing transitions to known final states. The second (filtered beam) technique allows the determination, in some cases,

of the multipolarity of the radiation, by averaging over the Porter-Thomas fluctuations in individual gamma-ray transition widths. A similar experiment has been described by Bollinger and Thomas[1]. The results should be a useful complement to information determined by other reactions.

^{171}Yb Levels

The data obtained from the $^{170}\text{Yb}(n,\gamma)^{171}\text{Yb}$ reaction are summarized in Table I. The symbol "X" is used in this and succeeding tables to indicate observation of a transition. The capturing states spin and parity must be $1/2+$ for s-wave capture, which limits the final states to $I = 1/2$ or $3/2$. Transitions believed to be E1 transitions as shown in Table I fix the final-state parity as odd. Where no multipolarity is given, the data were insufficient to decide between E1 and M1 assignments.

^{172}Yb Levels

Information on ^{172}Yb levels is given in Table II. Final states fed by these transitions as listed in Table II will have $I = 0, 1$, or 2 ; E1 transitions will feed even parity states and M1 transitions will feed odd parity states. In this case, we have been able to assign the spin and parity for the 7.93 eV resonances as 1^- , since primary transitions are observed to known $0+$ and $2+$ states. Although the ground and first excited $0+$ states were not directly populated, a recently established[2] $0+$ state at 1405 keV is populated from this resonance which eliminates the possibility of a $0-$ assignment because $0 \rightarrow 0$ transitions are completely forbidden.

^{174}Yb Levels

Table III lists the primary gamma-ray transitions to levels in ^{174}Yb . In this case it was necessary to deduce the neutron binding energy from the known excitation energy of the first excited state at 73.5 keV, since the $0+$ ground state cannot be populated from the $2-$ and $3-$ capturing states. The result is 7465.0 ± 1.0 keV. Final state spins may range from 1 to 4. The filtered beam experiments allow a classification of the E1 transitions and establish the final state parity as even for those states fed by these transitions. The two M1 transitions observed would feed odd parity states. Unfortunately, the data on the de-excitation of the 17.7 eV resonance in ^{173}Yb are insufficient to establish its spin.

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TABLE I
 ^{171}Yb LEVELS

Peak No.	Energy (keV)	Excitation Energy (keV)	Neutron Beam Used for Observation		Probable Type of Transition
			$1/16''$ $^{10}\text{B}_4\text{C}$ $J\pi = 1/2^+$	8.0 eV beam $J\pi = 1/2^+$	
1	6615 $\pm$ 2	0	X	X	E1
2	6549 $\pm$ 2	66	X	X	E1
3	5706 $\pm$ 3	909	X	X	E1
4	5626 $\pm$ 3	989	X	X	E1
5	5577 $\pm$ 3	1038	X	X	E1
6	5075 $\pm$ 4	1540		X	
7	4911 $\pm$ 4	1704	X?	X	E1?
8	4895 $\pm$ 4	1720		X	
9	4725 $\pm$ 5	1890		X	
10	4702 $\pm$ 5	1913		X	
11	4475 $\pm$ 5	2140		X	

TABLE II
LEVELS IN ^{172}Yb

Peak No.	Gamma-Ray		Excitation Energy (keV)	Neutron Beam Used For Observation			Probable Type of Transition
	Energy (keV)	Error		$1/16''$ $^{10}\text{B}_4\text{C}$ Filter $J\pi = 0, 1^-$	7.83 eV $J\pi = 1^-$	13.07 eV $J\pi = 1^-$	
1	8021.6	$\pm$ 1.5	0.0	X		X	E1
2	7941.8	$\pm$ 1.5	79.8	X	X	X	E1
3	6977.0	$\pm$ 1.5	1044.6	X			E1
4	6901.6	$\pm$ 2.0	1120.0	X	X		E1
5	6863.8	$\pm$ 3.0	1157.8	X			M1

TABLE II (Contd.)

LEVELS IN ^{172}Yb

Peak No.	Gamma-Ray Energy (keV)	Error	Excitation Energy (keV)	Neutron Beam Used for Observation			Probable Type of Transition
				1/16" $^{10}\text{B}_4\text{C}$ Filter $J\pi = 0, 1-$	7.83 eV	13.07 eV	
					$J\pi = 1-$	$J\pi = 1-$	
6	6821.9	± 3.0	1199.7	X		X	M1
7	6615.9	± 2.0	1405.7	X	X	X	E1
8	6554.2	± 2.0	1467.4	X	X		E1
9	6592.5	± 2.0	1479.1	X	X		E1
10	6411.2	± 2.0	1610.4	X	X		E1
11	6224.7	± 3.0	1796.0	X	X		E1
12	6201.9	± 3.0	1819.7		X		M1
13	6170.2	± 2.0	1851.4	X	X		E1
14	6126.7	± 4.0	1894.9	X	X	X	E1
15	(6102.9)	± 3.0	(1918.7)			(X)	(M1)
16	6061.0	± 3.0	1960.6	X			E1
17	6009.1	± 3.0	2012.5	X		X	E1
18	5967.2	± 3.0	2054.4	X			E1
19	5915.0	± 3.0	2105.8	X	X	X	E1
20	5826.6	± 3.0	2095.0	X	X		E1
21	5791.1	± 3.0	2230.5	X	X		E1
22	5703.6	± 3.0	2318.0	X	X		E1
23	5690.2	± 4.0	2331.4	X	X		E1
24	5673.6	± 4.0	2348.0	X			E1
25	5644.3	± 3.0	2377.3	X	X		E1
26	5631.6	± 4.0	2390.0	X	X		E1
27	5579.6	± 4.0	2442.0	X	X		E1
28	5555.1	± 4.0	2466.5	X	?		E1
29	5536.5	± 4.0	2485.1	X	?		E1
30	5528.4	± 4.0	2493.2	X	?		E1
31	5503.7	± 4.0	2517.9	X	?		E1
32	5492.3	± 4.0	2529.3	X	X		E1

TABLE II (Contd.)

LEVELS IN ^{172}Yb

Peak No.	Gamma-Ray Energy (keV)	Error	Excitation Energy (keV)	Neutron Beam Used for Observation			Probable Type of Transition
				1/16" $^{10}\text{B}_4\text{C}$ Filter $J\pi = 0, 1-$	7.83 eV	13.07 eV	
					$J\pi = 1-$	$J\pi = 1-$	
33	5484.0	± 5.0	2537.6	[a]	X	[a]	[a]
34	5443.2	± 5.0	2578.4		X		
35	5433.6	± 5.0	2588.0		X		
36	5285.5	± 5.0	2736.1		X		
37	5221.2	± 5.0	2800.4		X		
38	5210.0	± 5.0	2811.6		X		
39	5197.6	± 5.0	2824.0		X		
40	4918.6	± 5.0	3103.0		X		
41	4982.9	± 5.0	3038.7		X		
42	4918.5	± 5.0	3103.1		X		
43	4835.6	± 5.0	3186.0		X		
44	4761.9	± 5.0	3259.7		X		
45	4702.0	± 5.0	3319.6		X		
46	4621.9	± 5.0	3399.7		X		

[a] Only the 7.93 eV resonance was studied for lower energy transitions for comparison with a similar energy range in the $^{170}\text{Yb} (n, \sigma) ^{171}\text{Yb}$ reaction data, since the 8.0 eV resonance in ^{170}Yb nearly coincides with the 7.93 eV resonance in ^{171}Yb .

TABLE III
LEVELS IN ^{174}Yb

Peak No.	Gamma-Ray Energy (keV)	Error	Excitation Energy (keV)	Neutron Beam Used for Observation				Probable Type of Transition
				$1/8"^{10}\text{B}_4\text{C}$ Filter $J\pi = 2,3-$	$1/16"^{10}\text{B}_4\text{C}$ Filter $J\pi = 2,3-$	4.53 eV $J\pi = 3-$	17.7 eV $J\pi = 3-$	
--	(7465.0) ^[a]	± 1.0	0.0	[a]	[a]	[a]	[a]	
1	7388.5	± 1.0	76.5	X	X	X	X	E1
2	7212.1	± 1.0	252.9	X	X	X		E1
3	6144.6	± 2.0	1320.4			X		M1
4	5903.7	± 1.0	1561.3	X	X	X	X	E1
5	5857.0	± 2.0	1608.0	X	X	X	X	E1
6	5836.5 ^[b]	± 2.0	1628.5	X	X	X	X	E1
7	5828.7 ^[b]	± 2.0	1636.3		X	X	X	E1
8	5788.1	± 2.0	1676.9	X	X	X	X	E1
9	5753.4 ^[c]	± 3.0	1711.6	X	X	X	X	E1
10	5748.4 ^[c]	± 3.0	1716.6			X		M1?
11	5730.7	± 2.0	1734.3	X	X	X		E1
12	5659.0	± 2.0	1806.0	X	X	?		E1
13	5606.2	± 2.0	1858.8	X	X	X		E1
14	5521.7	± 3.0	1943.3	X	X	X		E1
15	5505.6	± 2.0	1959.4	X	X	X		E1
16	5447.9 ^[a]	± 3.0	2017.1			X		

[a] Deduced neutron binding energy using 76.5 keV as the excitation energy of the first excited state.

[b] Probable doublet.

[c] Possible triplet. The lowest energy number was obtained from the 4.53 eV data.

[d] Possible doublet observed only in the 4.53 eV beam; other runs did not extend to this energy.

LEVELS IN ^{174}Yb FROM $^{173}\text{Yb}(n,\gamma)$

J. J. H. Berlijn^[1], R. C. Greenwood, C. W. Reich

The level scheme of ^{174}Yb has been studied from measurements of the prompt gamma rays from thermal neutron capture in ^{173}Yb . Measurements on the high energy prompt gamma-ray spectra were made using two different Ge(Li) detectors, and energy calibrations were obtained with a mixed source technique using lead, sodium, and hydrogen capture gamma-ray lines as calibrations. Both a 6 cm² x 8 mm Ge(Li) detector (which had a resolution of 1 keV at 100 keV and 1.7 keV at 1 MeV) and a 35 cm³ coaxial Ge(Li) detector were used to measure that portion of the capture gamma-ray spectrum below 3 MeV. Extensive energy calibration measurements were made using several different gain scales.

A preliminary analysis of this data has established that portion of the ^{174}Yb level scheme which is shown in Figure 1. This level scheme principally shows details of the ground-state band, the $K^\pi = 2^-$ octupole vibration band, and the gamma-vibration band. The observed ground-state band level energies are fitted with parameters $A = 12.801$ keV, $B = -7.9 \times 10^{-3}$ keV, and $C = 32 \times 10^{-6}$ keV in the expression $E_I = A I(I+1) + B I^2(I+1)^2 + C I^3(I+1)^3$.

Work is continuing in an attempt to fit the $^{173}\text{Yb}(n,\gamma)$ data into a consistent ^{174}Yb level scheme. The 2-keV neutron capture gamma-ray spectrum which we recently obtained with a ^{173}Yb sample is expected to be particularly helpful to us in this effort.

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Fig. 1 A preliminary level scheme for ^{174}Yb .

LEVELS IN ^{182}Ta

R. G. Helmer, R. C. Greenwood, C. W. Reich

The study of the level structure of the odd-odd deformed nuclide ^{182}Ta has been continued. As previously reported[1] these levels have been studied from the decay of ^{182}Hf , the decay of the 16-min isomeric state of ^{182}Ta , and the prompt gamma rays emitted as a result of thermal neutron capture in ^{181}Ta .

Recently spectra of the primary gamma rays resulting from neutron capture in tantalum were obtained using the external 2-keV neutron beam from the Materials Testing Reactor. Since the neutron energy distribution in the 2-keV beam is approximately Gaussian with 700 eV FWHM, the resultant capture gamma-ray spectrum of tantalum is an average over about 150 neutron resonances with spins and parities of 3+ and 4+ (the ground state of ^{181}Ta being 7/2+). The statistical fluctuations in the partial radiation widths are thus averaged out. The primary E1 transitions in this spectrum are observed to be about a factor of 8 stronger than the corresponding M1 transitions at the same energy.

Furthermore, transitions to final states with spins of 3 or 4 are approximately twice as strong as transitions to those with spins of 2 or 5. Figure 1 shows a plot of the relative gamma-ray intensities divided by the transition energy to the third power (I_γ/E_γ^3) for the observed transitions. Thus, this plot allows a differentiation between primary capture gamma-ray transitions which populate final states with spins and parities of 3- or 4-, 2- or 5-, 3+ or 4+, and 2+ or 5+.

All of the experimental data have been combined to form a preliminary level scheme for ^{182}Ta which is shown in Figure 2. This includes 24 levels in 11 rotational bands. Unassigned ^{182}Ta levels below 800 keV are shown on the right-hand side of Figure 2. For clarity of presentation, transitions between levels in this scheme are omitted from this figure.

The two lowest lying odd-proton Nilsson states in ^{181}Ta are $7/2+[404]$ (the ground state) and $9/2-[514]$, and the four lowest lying odd-neutron states in ^{183}W (which like ^{182}Ta has 109 neutrons) are $1/2-[510]$ (the ground state), $3/2-[512]$, $7/2-[503]$, and $11/2+[615]$. Combinations of these odd-proton and odd-neutron states are expected to constitute the lowest energy states in ^{182}Ta . The $^{181}\text{Ta}(d,p)^{182}\text{Ta}$ data of Erskine and Buechner[2] have been of particular value in helping us make configuration assignments because the only states which are expected to be excited directly in this (d,p) reaction are those containing the proton orbital $7/2+[404]$.

These two proton and four neutron levels can produce eight combinations (or sixteen levels). Of these eight, seven have been identified as shown in Figure 2. In most cases some of the associated rotational states are observed above these base states. Although only 11 of the possible 16 base states are observed, most of the remaining are of high spin and would not be expected to be observed in these experiments.

While the analysis and interpretation of the properties of these levels in ^{182}Ta is only in an early stage, the following features can be pointed out.

It has been suggested[3-5] that proton states are not populated by primary neutron capture gamma-ray transitions in the $^{169}\text{Tm}(n,\gamma)$ and $^{165}\text{Ho}(n,\gamma)$ reactions. We find no evidence for such an effect; the proton states which we have definitely identified are indeed populated by primary capture gamma-ray transitions.

As pointed out by Erskine and Buechner[2] the $^{181}\text{Ta}(d,p)$ cross sections for exciting the four bands arising from the configurations $\{7/2+[404]$, $1/2-[510]\}$ and $\{7/2+[404]$, $3/2-[512]\}$ require a band-mixing picture for their understanding. This is also true for some of the gamma-ray branching results and the level energies. A detailed Coriolis-coupling analysis of these levels is in process. This analysis is complicated by the fact that in the odd-A nuclei, Coriolis coupling of the $1/2-[510]$ and $3/2-[512]$ orbitals is not well understood at the present time[6,7]. These calculations are continuing, however, and promise to give information of a fundamental nature concerning the coupling of these bands.

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Fig. 1 Plot of relative reduced gamma-ray intensities for transitions from the $^{181}Ta(n, \gamma)$ capture state to the levels indicated.

Fig. 2 Partial level scheme for ^{182}Ta with levels grouped into rotational bands. Arrow into level indicates population by primary gamma ray from neutron capture and triangle below level indicates population by (d,p) reaction. The quantum numbers from the Nilsson model are given below the rotational bands for the odd proton and neutron. The unassigned levels below 800 keV observed in the (n, γ) process are shown at the right.

LEVELS IN ^{183}Ta FROM THE DECAY OF ^{183}Hf [1]

L. D. McIsaac, R. G. Helmer, C. W. Reich

For this study, ^{183}Hf activity was produced by neutron irradiation of samples of 9×10^6 -yr ^{182}Hf for 1 to 1-1/2 hours in a flux of 2×10^{14} n-cm $^{-2}$ -sec $^{-1}$. These ^{182}Hf samples were obtained by neutron irradiation of about 100 mg of hafnium for one to two years in the Materials Testing Reactor at a flux of 5×10^{14} neutrons-cm $^{-2}$ -sec $^{-1}$. The resulting material was then isotopically separated by the Isotope Separation Group, Chemistry Division, Argonne National Laboratory, to obtain the ^{182}Hf .

This small amount of hafnium (< 20 micrograms) was to be irradiated several times with intervening chemical purification. Therefore, a purification procedure was developed which (1) provided essentially 100% yield for hafnium, (2) did not require addition of any hafnium or zirconium carriers, and (3) left the sample in a form in which it could be recovered.

Gamma-ray spectra were measured with Ge(Li) detectors. Successive spectra obtained from a chemically pure source were used to verify that all of the gamma rays assigned to the ^{183}Hf activity decay with the same half-life and to determine the absolute gamma-ray intensities. From the growth of the

^{183}Ta daughter, and the absolute ^{183}Ta intensities, the absolute intensities were found. The resulting gamma-ray energies and intensities are given in Table I.

Gamma-gamma coincidence relationships were observed in a gamma-gamma coincidence measurement using two NaI(Tl) detectors. A beta-ray spectrum of ^{183}Hf was obtained with an anthracene detector. Also, the beta spectra in coincidence with the intense gamma rays of 459 and 783 keV were measured.

The proposed decay scheme is shown in Figure 1. The gamma-gamma and beta-gamma coincidence results and the transition energies and intensities uniquely determine the existence of levels at 73, 459, 856, and 1543 keV. The transitions definitely associated with these levels are shown in the left section of Figure 1. In the right section, several other transitions are shown for which there is less definitive information about their placement.

The 856-keV gamma is shown de-populating the 856-keV level although energetically this transition could also represent the crossover of a 165- and 691-keV cascade elsewhere in the scheme. The coincidences observed between gamma rays at ≈ 225 and 735 keV could involve either or both of the gamma rays at 223 and 225 keV.

There are four pairs of gamma rays whose energies differ by 4.1 to 4.4 keV. These transitions are probably associated, but there is no definitive information about their positions in the decay scheme or their relations to each other.

The $\log ft$ values have been computed from the gamma-ray intensities and the measured beta branch energies.

No measurements have been made of the excited-state spins and parities for ^{183}Ta . However, the $\log ft$ values of the various beta branches and the observed decay modes of the levels enable one to make plausible configuration assignments to most of the proposed levels.

The ground state of ^{183}Hf corresponds to the Nilsson neutron orbital $3/2^- [512]$. The ^{183}Ta ground state and 73-keV level correspond to the Nilsson proton orbitals $7/2^+ [404]$ and $9/2^- [514]$. The 143-keV level is interpreted as the spin-9/2 member of the ground-state rotational band. From its mode of de-excitation, the state at 459 keV is assigned as the Nilsson state $5/2^+ [402]$ and the 573-keV state is interpreted as the spin-7/2 member of the associated rotational band.

The most reasonable interpretation of the nature of the 856-keV state is that it is a K-2 gamma vibration built on the $9/2^- [514]$ state at 73 keV. Nuclear excitations of the K-2 gamma-vibrational type are a well established feature of the level structure of the odd-A deformed nuclei in the rare-earth region. However, in the vast majority of cases, they are observed to be strongly mixed with one-quasiparticle states. We believe that this state is free of large admixtures of one-quasiparticle excitations. Therefore, it is of interest to investigate the extent to which the microscopic description of vibrations accounts for the observed properties of the gamma-vibrational state. The beta- and gamma-ray processes involving this level are illustrated schematically in Figure 2. At the upper left, the proton and neutron systems which correspond to the ^{183}Hf

ground state are shown. The beta decay is presumed to proceed via the au transition $7/2[514]_n \rightarrow 9/2[514]_p$, leading to the three-quasiparticle configuration $\{9/2[514]_p, 7/2[514]_n, 3/2[512]_n\}$, with $K^\pi = 5/2^-$, as shown at the top in Figure 2. The two neutron orbitals coupled to $K^\pi = 2^+$ may be thought of as one particular two-quasiparticle component of the gamma-vibrational phonon in the corresponding doubly-even core. This vibrational phonon consists of a coherent superposition of two-quasiparticle states as shown in the center of Figure 2.

The 856-keV state de-excites predominantly via the intense 783-keV gamma ray to its base state, to the $5/2^+$ and $7/2^+$ members of the $5/2^+[402]$ band and, much more weakly if at all, to the ground state. The preference of the 856-keV state for decay to the $5/2^+[402]$ band, rather than the ground state, finds a ready explanation in terms of the microscopic structure illustrated in the center of Figure 2. This decay may take place via the E1 transition $9/2[514] \approx 9/2[404]$, as indicated at the bottom of Figure 2.

Examination of the three-quasiparticle makeup of the 856-keV state reveals no components which can decay to ground state by an E1 transition. Hence, one expects that the ground-state transition will be nonexistent or at least much weaker than those to the $5/2^+[402]$ band, in agreement with observation.

[1] Published in Nucl. Phys., A132, 28 (1969).

Fig. 1 Decay scheme proposed for ^{183}Hf . The energies are in keV and the intensities in percent of decays. The left section is well established while the right-hand portion is tentative.

TABLE I

GAMMA-RAY ENERGIES AND INTENSITIES FROM ^{183}Hf [a]

Energy (keV)	Intensity (% of Decays)	Energy (keV)	Intensity (% of Decays)
73.160 (15)	38.0	500.7 (3)	0.08
84.69 (6)	0.03 ± 0.006	536.8 (2)	0.15
113.72 (3)	0.14	594.8 (3)	0.21 ± 0.1
124.1 (2)	0.025 ± 0.008	686.50 (10)	$0.24 \pm$
131.0 (3)	0.018 ± 0.012	691.73 (10)	0.30
139.0 (2)	0.025 ± 0.012	715.3 (10)	0.044 ± 0.02
143.19 (2)	0.47	717.2 (10)	0.071 ± 0.03
149.1 (2)	0.019 ± 0.01	727.3 (3)	0.059 ± 0.03
153.75 (15)	0.034 ± 0.007	735.05 (8)	0.88
157.90 (15)	0.033 ± 0.012	762.9 (3)	0.065 ± 0.03
160.78 (2)	0.52	769.3 (3)	0.14 ± 0.05
165.20 (15)	0.036 ± 0.012	783.73 (3)	65.0
180.4 (2)	0.031 ± 0.012	797.2 (5)	0.02
223.40 (15)	0.078 ± 0.03	806.5 (2)	0.14 ± 0.03
225.00 (10)	0.15	856.78 (15)	0.11 ± 0.03
284.10 (5)	0.36	868.5 (2)	0.04
295.21 (8)	0.17	875.5 (3)	0.062 ± 0.03
315.855 (20)	1.22	959.6 (10)	0.026 ± 0.02
375.78 (15)	0.088 ± 0.02	987.1 (3)	0.04
397.86 (2)	2.9	1045.5 (3)	0.07 ± 0.02
459.073 (20)	27.3	1112.7 (3)	0.04
466.7 (2)	0.085 ± 0.04	1470.2 (1)	2.7
471.1 (2)	0.16 ± 0.03	1543.0 (10)	0.02 ± 0.02
476.90 (18)	0.72	1784.3 (2)	0.07 ± 0.03
481.31 (5)	0.7 ± 0.15		

[a] The uncertainties are in the last digit(s) given. The estimated uncertainties in the intensities are 10% unless otherwise stated.

- (2) The low gamma-ray background which is present in this beam because of the thick filter (< 2 mR/hr).

In order to explore the utility of this 2-keV neutron beam facility for neutron capture gamma-ray spectroscopy we will discuss as illustrative examples the data obtained with the following three reactions: $^{181}\text{Ta}(n,\gamma)$, $^{180}\text{Hf}(n,\gamma)$, and $^{55}\text{Mn}(n,\gamma)$. These three reactions have been chosen because they include the following three types of neutron capture reaction: (1) with a high Z , odd- A target nucleus (where the neutron resonance spacing is a few eV), (2) with a high Z , even-even target nucleus (where the resonance spacing is not small, typically ranging from a few tens of eV upward), and (3) with a low- Z target nucleus (where the resonance spacing is typically a few keV).

Before discussing these data we will briefly comment on the distribution of intensities (partial radiation widths) which are observed for primary capture gamma rays. Wide statistical fluctuations are expected in the distribution of partial radiation widths of transitions from individual neutron resonance states to a lower energy state. Porter and Thomas[4] have suggested that this distribution of partial radiation widths can be described by a chi-squared distribution with one degree of freedom. That is, the distribution function is of the form

$$P(x) dx = \frac{1}{2} \Gamma\left(\frac{1}{2}\right)^{-1} \left(\frac{x}{2}\right)^{-\frac{1}{2}} e^{-\frac{x}{2}} dx$$

where $P(x)$ is the probability at amplitude x , $\Gamma(1/2)$ is the gamma function, and where x is defined as

$$x = \frac{\Gamma_{\gamma i}}{\langle \Gamma_{\gamma i} \rangle_{Av}}$$

This distribution is illustrated in Figure 1. Experimental data on the distributions of partial radiation widths tend to support this hypothesis[5], although Chrien[6] has suggested that the distribution of partial radiation widths for at least some nuclei is better fitted with a chi-squared distribution with greater than one degree of freedom. In the following discussion we will assume for simplicity that the distribution of partial radiation widths is described by a chi-squared distribution with one degree of freedom. (The conclusions we will reach, however, are not critically dependent on this assumption.) Then, if the neutron capture is an average over ν resonances, instead of merely resulting from one resonance, the total partial radiation width to a particular final state is given by

$$\Gamma_{\gamma i} = \sum_{j=1}^{\nu} \Gamma_{\gamma ij} .$$

For this case it is reasonable to suggest that this averaged partial radiation width has a chi-squared distribution with ν degrees of freedom[4]. That is a distribution with the following form:

$$P(x) dx = \Gamma\left(\frac{\nu}{2}\right)^{-1} \left(\frac{\nu x}{2}\right)^{\frac{\nu}{2}-1} e^{-\frac{\nu x}{2}} \left(\frac{\nu}{2}\right) dx.$$

Additional chi-squared distributions with 5 and 25 degrees of freedom are illustrated for purposes of comparison in Figure 1.

We now consider the three specific cases mentioned above.

$^{181}\text{Ta}(n,\gamma)$. The $^{181}\text{Ta}(n,\gamma)$ spectrum obtained using the 2-keV neutron beam provides an example of a capture gamma-ray spectrum which is averaged over many resonances. This spectrum is shown in Figure 2. For comparison, the corresponding portion of the thermal-neutron capture gamma-ray spectrum is included in Figure 2. Figure 3 shows a plot of $I_\gamma E_\gamma^{-3}$ versus the energy, E_γ , of the primary capture gamma-rays observed from 2-keV neutron capture in tantalum. It is seen from Figure 3 that values of $I_\gamma E_\gamma^{-3}$ are significantly different for E1 and M1 transitions. Also, as expected[7], there is approximately a factor of two between the intensities of primary transitions to spin 3 or 4 and spin 2 or 5 states. (If only s-wave neutron capture is considered, the compound states in ^{182}Ta will have spins of 3 or 4.) The $J = 2,5$ lines in Figure 3 are drawn in at one-half the intensity of the $J = 3,4$ lines. Each of these lines is drawn with an E_γ^{-5} energy dependence. As noted by Bollinger[5,8] this E_γ^{-5} energy dependence often seems to represent a better fit to the averaged intensity data than does the E_γ^{-3} function. This also appears to be the case for $^{181}\text{Ta}(n,\gamma)$, at least for the E1 transitions.

If we assume an average resonance spacing of 5 eV for the $^{181}\text{Ta}(n,\gamma)$ reaction[9] we would estimate that the 2-keV neutron capture gamma-ray spectrum is an average over approximately 170 resonances (with both $J = 3$ and 4). An estimate of the number of resonances that this spectrum is an average over can also be obtained from the distribution of primary capture gamma-ray intensities. Figure 4 shows a plot of the experimental distribution of the E1 transitions, grouped into intervals $\Delta x = 0.1$, plotted against x . The parallel lines drawn for $J = 3,4$ - and $J = 2,5$ - in Figure 3 are used to define $x = 1.0$ and $x = 0.5$, respectively. For comparison, the expected probability distributions for a spectrum averaged over 50, 100, and 200 total (both $J = 3$ and 4 resonances) are also shown in Figure 4. The variance computed from the transition intensities to final states assigned as 3- or 4- suggests that we are averaging over approximately 140 resonances. This is in rather good agreement with the approximately 170 resonances which we would expect to average over. The transition intensities to final states assigned as 2- or 5- show considerably more statistical spread, and the computed variance in these experimental data suggests that we are only averaging over approximately 40 resonances, compared to an expected 85 resonances. However, it should be noted that the transition intensities to 2- or 5- final states, being weaker, would be affected proportionately more by unresolved M1 transitions within these peaks. Also, a pair of unresolved M1 transitions could be incorrectly assigned as an E1 transition to a final 2- or 5- state. Thus it is not surprising that we have an apparently poorer averaging of the E1 transition intensities to final states with spins 2- or 5-.

Actually, because the density of states in ^{182}Ta is becoming so high, any spin assignments based solely on the 2-keV neutron capture data must be considered as tentative above about 700 keV.

$^{180}\text{Hf}(n,\gamma)$. The $^{180}\text{Hf}(n,\gamma)$ spectrum obtained with the 2-keV neutron beam contains features of interest to this discussion. In this spectrum we observe strong transitions to the 1/2- and 3/2- members of the 1/2-[510] ground-state band of ^{181}Hf , but the primary transition intensity to the 3/2-[512] state at 255 keV is very weak. This fluctuation in primary E1 transition intensities may be explained by the wide resonance spacing for the $^{180}\text{Hf}(n,\gamma)$ reaction.

Resonances in the $^{180}\text{Hf}(n,\gamma)$ reaction have spacings > 100 eV[9] so that our 2-keV neutron capture spectrum represents an average over about five resonances. As we note from Figure 1, wide fluctuations in primary gamma-ray transition intensities are to be expected if the averaging is over so few resonances. In fact, in such a case it will be impossible to reliably distinguish primary E1 transitions from primary M1 transitions. This example perhaps serves to point up a limitation on an averaging technique using the 2-keV neutrons; namely that, even for the deformed rare-earth nuclei, many of the even-even stable isotopes will not have a sufficiently small neutron resonance spacing to permit good averaging. In these cases then the best that can be hoped for is that one gets a spectrum which is somewhat different than the thermal neutron capture spectrum, and which reveals some added features of the level scheme.

$^{55}\text{Mn}(n,\gamma)$. For the low-Z nuclei, the neutron resonance spacing is generally too wide to permit any kind of an averaging. In some cases though the 2-keV neutron capture spectrum is dominated by a different resonance than that which dominates the thermal neutron capture. Such is the case for $^{55}\text{Mn}(n,\gamma)$. The prompt gamma-ray spectra resulting from thermal and 2-keV neutron capture in manganese are shown in Figure 5. Here the 2-keV neutron capture spectrum is principally contributed by the 2.375-keV resonance in the $^{55}\text{Mn}(n,\gamma)$ reaction.

However, a more important use of a 2-keV neutron capture gamma-ray spectrum obtained from a low-Z target nucleus should be that, when used in conjunction with the corresponding thermal neutron capture gamma-ray spectrum, it should allow primary capture gamma rays (those decaying from the capturing state) to be unambiguously distinguished from secondary capture gamma rays (which are emitted in the de-excitation of the lower energy states). This is because the primary capture gamma rays will have energies which are 2 keV greater than those measured from the corresponding thermal neutron capture spectrum, while the energies of the secondary prompt gamma rays will remain unchanged. We have demonstrated this effect with the $^{181}\text{Ta}(n,\gamma)$ spectra shown in Figure 1. The 2-keV neutron capture gamma-ray line energies were measured relative to the ^{16}O line (which resulted from the decay of 7-sec ^{16}N) and were found to have energies of 1.8 ± 0.3 keV greater than the corresponding line energies in the thermal spectrum.

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Fig. 1 Chi-squared probability distributions with 1, 5, and 25 degrees of freedom.

Fig. 2 Comparison of the high energy portions of the prompt gamma-ray spectra resulting from 2-keV and thermal neutron capture in tantalum. The energies associated with the peaks are the double-escape peak energies measured for thermal neutron capture. The 2-keV neutron capture gamma-ray spectrum was obtained with an 80-hr run using a 15-g tantalum sample.

Fig. 3 A plot of $L_\gamma E_\gamma^{-3}$ versus the energy of the prompt gamma-ray transitions observed from 2-keV neutron capture in tantalum. The L_γ represents the relative intensity of the prompt gamma-ray transitions.

Fig. 4 The observed distribution of the E1 transition intensities from 2-keV neutron capture in tantalum. For comparison, expected probability distributions for a spectrum averaged over 50, 100, and 200 total neutron resonances are also shown.

Fig. 5 Comparison of the high energy portions of the prompt gamma-ray spectra resulting from 2-keV and thermal neutron capture in manganese. The 2-keV neutron capture gamma-ray spectrum was obtained with a 15-hr run using a 25-g manganese sample.

MEASUREMENT OF FAST NEUTRON CAPTURE GAMMA-RAY SPECTRA FOR FAST REACTOR SHIELDING CALCULATIONS

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Neutron capture gamma-ray production will contribute a major part of the hard gamma-ray component of a fast reactor. These neutron capture gamma rays will be produced by neutrons having a wide range of energies. A portion of the fast neutrons produced in the reactor core will be captured in the core or blanket, while those neutrons which escape from these regions will be captured in the biological shield as they slow down to thermal energies. Thus, in making gamma-ray shielding calculations for a fast reactor, one is interested in neutron capture gamma-ray spectra produced by neutrons with energies ranging from a few hundred keV to thermal.

Because the magnitude of the partial radiation widths vary greatly from resonance to resonance and in a random fashion, neutron capture gamma-ray spectra exhibit dramatic changes as the energy of the captured neutron is changed. At the present time, however, the only substantial body of capture gamma-ray data which exists is for thermal neutron capture. Thus, because fast neutron capture gamma-ray spectra from various elements are not simply related to their corresponding thermal neutron capture gamma-ray spectra, the necessary input data to make reliable gamma-ray shielding calculations for a fast reactor are not presently available.

The objective of this present work is to investigate the feasibility of obtaining such fast neutron capture gamma-ray data using specially tailored beams from the Materials Testing Reactor. For example, essentially gamma-free monoenergetic neutron beams having energies of approximately 2, 25, and 144 keV have been extracted from the MTR with good efficiency by using thick filters of scandium, iron, and silicon, respectively.

To date, neutron capture gamma-ray measurements have been made using 2 keV neutrons from the scandium-filtered beam facility at the MTR. A 6 cm³ Ge(Li) coaxial detector coupled to a 4096 channel analyzer is being used for these experiments. Because of the low background at the reactor face, the Ge(Li) detector requires only minimal shielding. As an illustration of the data which have been obtained using this facility, the high energy portion of the prompt gamma-ray spectra resulting from capture of 2 keV neutrons by manganese and tantalum are shown in Figures 1 and 2, respectively. For comparison, the corresponding portions of the thermal neutron capture gamma-ray spectra are also included. Inspection of these figures show that the 2 keV neutron capture gamma-ray spectra are of very high statistical quality (comparable in quality to the thermal neutron capture gamma-ray spectra) and, most importantly, that they contain only small contributions from background contaminants (principally from the 7-sec ¹⁶N decay and from neutron capture in iron).

In Figures 1 and 2 it is observed that the 2 keV and thermal neutron capture gamma-ray spectra show significant differences. The relative intensities obtained from the 2 keV and thermal neutron capture gamma-ray spectra of manganese are compared in Table I. For tantalum, because of the finite energy spread

in the 2 keV neutron beam (0.7 keV FWHM), the resultant 2 keV spectrum represents an average over many resonances. This effectively removes the statistical fluctuations in the partial radiation widths, so that the reduced transition intensities to final states of the same spin and parity tend to be equal.

Other elements for which 2 keV neutron capture gamma-ray spectra have been measured include: sodium, potassium, chromium, iron, cobalt, nickel, copper, zirconium, niobium, molybdenum, terbium, dysprosium, lutetium, hafnium, tungsten, thorium and depleted uranium together with a number of separated isotope samples.

The dramatic differences which are observed between thermal and 2-keV neutron capture gamma-ray spectra for many elements are of particular relevance to gamma-ray shielding calculations for fast reactors. In particular, from comparison of thermal and 2-keV neutron capture gamma-ray spectra one can hope to gain insight into how capture gamma-ray spectra vary as a function of neutron energy.

Fig. 1 A comparison of the high energy portions of the prompt gamma-ray spectra resulting from 2 keV and thermal neutron capture in manganese. The 2-keV spectrum represents 15 hours of running time on a 25-g sample of manganese metal. The energies shown for the peaks in this spectrum are the double-escape peak energies obtained for the thermal neutron capture data.

Fig. 2 A comparison of the high energy portions of the prompt gamma-ray spectra resulting from 2 keV and thermal neutron capture in tantalum. The 2-keV spectrum represents 80 hours of running time on a 15-g sample of tantalum metal. The energies shown for the peak in the spectrum are the double-escape peak energies obtained with the thermal neutron capture data.

TABLE I

COMPARISON OF GAMMA-RAY INTENSITIES FOR THE $\text{Mn}(n, \gamma)$ REACTION
WITH THERMAL AND 2 keV NEUTRON BEAMS

<u>Gamma-Ray Energy</u>	<u>Thermal Capture Relative Intensity</u>	<u>2 keV Capture Relative Intensity</u>
7270.3	32.9	6.4
7243.6	109.9	1.9
7159.7	51.5	---
7057.9	100.0	100.0
6941.0	(2.5)	---
6929.0	21.6	13.7
6815.9	----	23.5
6784.0	28.8	18.2
6724.6	----	3.1

TABLE I (Contd.)

<u>Gamma-Ray Energy</u>	<u>Thermal Capture Relative Intensity</u>	<u>2 keV Capture Relative Intensity</u>
6549.4	3.8	4.4
6430.2	6.7	13.9
6104.4	16.8	---
6031.1	5.7	6.7
5921.0	7.1	3.2
5761.4	16.4	9.7
5527.1	62.5	35.5
5435.1	17.5	6.3
5404.0	3.2	3.3
5254.0	11.3	11.9
5198.6	8.4	5.2
5181.1	32.2	4.0
5111.4	2.9	6.4
5067.7	24.6	---
5034.6	8.2	4.0
5014.6	51.2	7.2
4949.6	15.5	---
4907.6	6.9	2.8
9875.0	5.4	---
4841.4	5.8	---
4833.8	7.7	---
4827.8	8.2	---
4725.0	23.3	3.0
4689.4	7.9	5.0
4643.7	7.1	---
4588.7	3.6	---
4566.7	16.9	---
4550.3	5.7	4.6
4445.5	4.3	8.1
4267.8	4.2	---
4251.6	----	3.2
4230.0	2.9	---
4222.8	5.5	---

THE DECAYS OF ^{99}Rh AND $^{99\text{m}}\text{Rh}$

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The decays of 16-day ^{99}Rh and 4.7-hr $^{99\text{m}}\text{Rh}$ have been investigated using the techniques of Ge(Li) spectroscopy. The activity was produced by the ^{100}Ru (p,2n) ^{99}Rh reaction using the cyclotron at the University of Colorado. Both activities were produced. Intensities from both decay modes of ^{99}Rh could be uniquely determined because the mixed source contained the 353-keV line which is assumed to result only from the 16-day activity. The energies and intensities of the gamma rays in 16-day ^{99}Rh and 4.7-hr $^{99\text{m}}\text{Rh}$ are given in Table I. These measurements are in good agreement with previous results [1,2].

The previously reported transitions [1] at 850 and 1505 keV in the decay of ^{99}Rh are not observed. However, a gamma ray at 1009.6 keV has been identified by half-life as belonging to ^{99}Rh . The energy and intensity measurements are consistent with the decay scheme proposed by Moss and McDaniels (see Figure 1).

Many unreported gamma rays were observed in the decay of 4.7-hr $^{99\text{m}}\text{Rh}$. Previous studies [2] report only 5 gamma-ray transitions. A decay scheme of $^{99\text{m}}\text{Rh}$ is proposed on the basis of energy and intensity measurements (see Figure 1). The 1.41-MeV gamma ray reported by Katcoff and Abrash was not observed.

- [1] G. A. Moss and D. K. McDaniels, "Gamma-Ray Spectrum of Ruthenium-99", *Phys. Rev.*, **162** n 4 (October 1967) pp 1087-1097.
- [2] S. Katcoff and H. Abrash, "Decay of Neutron-Deficient Isotopes of Pd and Rh", *Phys. Rev.*, **103** n 4 (August 1956) pp 966-970.

Fig. 1 Proposed decay scheme for ^{99}Rh after Moss and McDaniels. Decay scheme for $^{99\text{m}}\text{Rh}$ shown is proposed from present work.

TABLE I
GAMMA-RAY ENERGY AND INTENSITY MEASUREMENTS
ON A 16-DAY ^{99}Rh and 4.7-hr $^{99\text{m}}\text{Rh}$

Energy (keV)	Relative Intensity 16-day Activity	Relative Intensity 4.7-hr Activity
89.6 $\pm$ 0.2	100	1.98
120.3 $\pm$ 0.2	0.22	
175.45 $\pm$ 0.05	6.58	
232.75 $\pm$ 0.05	1.64	
251.0 $\pm$ 0.5		1.14
277.2 $\pm$ 0.5		2.57
295.76 $\pm$ 0.08	4.04	
322.32 $\pm$ 0.05	20.4	3.15
340.6 $\pm$ 0.5		100
353.06 $\pm$ 0.05	115	
442.59 $\pm$ 0.05	6.42	
486.28 $\pm$ 0.08	1.44	1.18
528.5 $\pm$ 0.5		2.88
528.53 $\pm$ 0.07	132	
558 $\pm$ 1		0.4
575.7 $\pm$ 0.3	0.65	0.88
617.9 $\pm$ 0.5		21.9
618.08 $\pm$ 0.05	14.4	
643.9 $\pm$ 0.6		0.4
659.8 $\pm$ 0.4		1.13
719.9 $\pm$ 0.3		2.36
733.93 $\pm$ 0.06	0.93	1.45
764.90 $\pm$ 0.07	1.25	
807.07 $\pm$ 0.05	4.1	
850	Not observed	
896.65 $\pm$ 0.15	2.7	
921.0 $\pm$ 0.5		1.64
938.0 $\pm$ 0.5		4.18
940.38 $\pm$ 0.12	4.9	
998.51 $\pm$ 0.10	2.8	
1009.6 $\pm$ 0.2	0.19	
1060.6 $\pm$ 0.2	0.86	
1088.88 $\pm$ 0.15	1.0	
1209.1 $\pm$ 0.4	0.80	
1261.3 $\pm$ 0.2		19.97
1293.34 $\pm$ 0.15	0.95	
1325.2 $\pm$ 0.4	0.60	
1382.9 $\pm$ 0.4	1.7	
1.41 $\pm$		Not observed
1441.3 $\pm$ 0.5	0.1	
1482.8 $\pm$ 0.3	0.42	
1505.1	Not observed	
1531.5 $\pm$ 0.4	1.62	
1571.9 $\pm$ 0.6	0.74	
1616.5 $\pm$ 0.8	0.77	
1660.9 $\pm$ 0.6	0.2	
1749.2 $\pm$ 0.8	0.2	
1970.2 $\pm$ 0.7	0.34	
2058.6 $\pm$ 3	0.05	

NUCLEAR LEVELS OF ^{103}Rh

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The nuclear levels of ^{103}Rh are of interest because of the potential use of this mono-isotopic element as a neutron flux monitor. The inconsistencies in the existing decay scheme for $A = 103$ [1] motivated the study of the decay of these isobars.

Shown in Figure 1 is the gamma-ray spectrum from ^{103}Ru as detected by a Ge(Li) detector $2.5\text{ cm}^2 \times 8\text{ mm}$. Portions of this spectrum have been previously discussed[2]. Figure 2 shows the spectrum of gamma rays from a source of ^{103}Pd as detected by a Ge(Li) spectrometer. From these data were determined the energies and relative intensities of the various gamma rays through the use of the computer program GAUSS III[3]. The K-shell internal conversion coefficient of the 39.75-keV transition in ^{103m}Rh was determined from the data shown in Figure 3 which is the spectrum of X-rays and gamma rays from a purified source of ^{103m}Rh detected by a Si(Li) spectrometer $30\text{ mm}^2 \times 3\text{ mm}$.

The results of these experiments are summarized in Tables I and II and Figures 4 and 5. Theoretical conversion coefficients given here are from Hager and Seltzer[4]. A decay scheme for ^{103}Ru and ^{103}Pd consistent with these data is shown in Figure 6. This level scheme is in agreement with that recently published by W. H. Zoller et al[5].

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- [1] C. M. Lederer, J. M. Hollander, I. Perlman, Table of Isotopes, 6th ed. New York: John Wiley & Sons, Inc., 1967.
 - [2] E. B. Nieschmidt, "Nuclear Levels of ^{103}Rh ", in R. L. Heath (ed.), Nuclear Technology Branch Yearly Progress Report for Period Ending June 30, 1968, IN-1218 (December 1968) pp 87-90.
 - [3] R. G. Helmer et al, "Photopeak Analysis Program for Photon Energy and Intensity Determinations Ge(Li) and NaI(Tl) Spectrometers", Nucl. Instr. & Methods, 57 (1967) pp 46-57.
 - [4] R. S. Hager and E. C. Seltzer, "Internal Conversion Tables. Part I: K-, L-, M-Shell Conversion Coefficients for $Z=30$ to $Z=103$ ", Nucl. Data, A4 (February 1968) pp 1-11.
 - [5] W. H. Zoller et al, "Decay of 40 d ^{103}Ru and 17 d ^{103}Pd to Levels of ^{103}Rh ", Nucl. Phys., A130 (1969) pp 293-304.

Fig. 1 Spectrum of higher energy gamma rays from ^{103}Ru .

Fig. 2 Spectrum of gamma rays from ^{103}Pd .

Fig. 3 X-rays and gamma rays observed from $^{103\text{m}}\text{Rh}$ with a Si(Li) detector.

TABLE I
GAMMA RAYS IN ^{103}Rh

E_γ (keV)	I_{rel} (Ru)	I_{rel} (Pd)	α_K	α_K (Theory)
39.75 ± 0.07	$0.072 \pm 0.008^{[a]}$	$1100 \pm 90^{[a]}$	138 ± 5	$140(\text{E3})$
53.3 ± 0.09	0.30 ± 0.03	0.23	1.77 ± 0.03	$1.82(\text{M1})$
62.5 ± 0.1	N.O.	20 ± 3		
113.2 ± 0.5	V.W.	N.O.		
241.9 ± 0.5	V.W.	N.O.		
295.0 ± 0.1	0.21 ± 0.02	72 ± 8	0.020 ± 0.005	$0.016(\text{M1}), 0.026(\text{E2})$
317.4 ± 0.3	N.O.	0.41 ± 0.08		
357.5 ± 0.1	N.O.	531 ± 5		
443.9 ± 0.1	0.31 ± 0.03	0.23 ± 0.06	0.011 ± 0.003	$0.007(\text{E2})$
497.1 ± 0.1	100 ± 10	100 ± 10	0.005 ± 0.001	$0.0045(\text{M1}), 0.005(\text{E2})$
557.0 ± 0.1	0.76 ± 0.07	N.O.	0.007 ± 0.001	$0.0035(\text{M1}), 0.0036(\text{E2})$
610.3 ± 0.1	4.8 ± 0.4	N.O.	0.0041 ± 0.0008	$0.0028(\text{M1}), 0.0028(\text{E2})$

[a] Uncertainties in intensity of the 39.75 keV gamma ray are statistical only.
Uncertainties in efficiency at this energy are considerable.

TABLE II
BETA DECAYS LEADING TO
LEVELS IN ^{103}Rh

E_β (keV)	% decays	log ft
$^{103}\text{Ru} \beta_1$ 90	4.4	6.0
β_2 203	81	6.0
β_3 383	<0.01	>10
β_4 445	0.2	9.5
β_5 700	15	8.2
$^{103}\text{Pd} \beta_1$ 23 (EC)	0.05	--
β_2 200	0.3	6.3
β_3 265	<0.02	>8
β_4 520	99.6	5.3

Fig. 5 ^{103}Rh K-shell internal conversion coefficients (low energy).

Fig. 4 ^{103}Rh K-shell internal conversion coefficients (high energy).

Fig. 6 Nuclear level scheme of ^{103}Rh .

THE DECAY OF 18-MIN ^{141}Ba AND 11-MIN ^{142}Ba

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As a continuation of the effort to investigate properties of fission product activities, the decays of 18-min ^{141}Ba and 11-min ^{142}Ba have been studied. These activities were produced by irradiating ^{235}U in the HR-2 pneumatic rabbit system of the MTR. Gamma-ray energies and intensities were measured using Ge(Li) and Si(Li) detectors. Coincidence measurements were made using two NaI(Tl) spectrometers and a 256- x 256-channel coincidence analyzer.

It was possible to prepare ^{141}Ba free of ^{142}Ba by rapidly removing all Ba activities from freshly prepared fission products then allowing ^{141}Ba to grow in from its 24-sec ^{141}Cs parent. Activity due to ^{142}Ba , which has a 2.3-sec ^{142}Cs parent, was eliminated in this manner. Since it is not possible to prepare ^{142}Ba free of ^{141}Ba , decay measurements were made on sources of mixed Ba activities. Energies and intensities of gamma rays of ^{141}Ba and ^{142}Ba are shown in Tables I and II, respectively. From coincidence data and energy combinations the majority of the transitions identified with these activities are incorporated in the proposed decay schemes shown in Figures 1 and 2.

TABLE I

TRANSITION ENERGIES AND RELATIVE GAMMA-RAY INTENSITIES FOR ^{141}Ba .

THE QUOTED UNCERTAINTIES IN ENERGY ARE ESTIMATED STANDARD DEVIATIONS.

THE ESTIMATED UNCERTAINTIES IN INTENSITIES ARE 10% UNLESS OTHERWISE STATED.

E_{γ} (keV)	Relative Intensity	E_{γ} (keV)	Relative Intensity
112.96 $\pm$ 0.09	2.2	929.4 $\pm$ 0.2	1.8 $\pm$ 0.3
162.9 $\pm$ 0.2	1.7	943.2 $\pm$ 0.2	2.4 $\pm$ 0.4
180.7 $\pm$ 0.1	0.80	981.5 $\pm$ 0.2	1.9 $\pm$ 0.3
190.33 $\pm$ 0.06	100	1160.7 $\pm$ 0.2	3.3
276.95 $\pm$ 0.05	54	1197.3 $\pm$ 0.2	12.0 $\pm$ 2
304.18 $\pm$ 0.05	60	1224.7 $\pm$ 0.2	0.7 $\pm$ 0.1
343.66 $\pm$ 0.05	35	1264.0 $\pm$ 0.2	1.7 $\pm$ 0.2
349.1 $\pm$ 0.2	0.9 $\pm$ 0.1	1273.4 $\pm$ 0.3	1.5 $\pm$ 0.2
364.7 $\pm$ 0.2	1.0	1278.1 $\pm$ 0.3	1.7 $\pm$ 0.2
389.71 $\pm$ 0.08	3.3	1310.9 $\pm$ 0.2	1.5 $\pm$ 0.2
457.59 $\pm$ 0.06	12	1323.7 $\pm$ 0.2	2.2 $\pm$ 0.3
462.13 $\pm$ 0.06	12	1346.1 $\pm$ 0.4	<1
467.26 $\pm$ 0.06	15	1377.8 $\pm$ 0.3	2.1 $\pm$ 0.3
522.1 $\pm$ 0.3	1.2	1405.2 $\pm$ 0.3	0.5 $\pm$ 0.1

TABLE I (Contd.)

E_{γ} (keV)	Relative Intensity	E_{γ} (keV)	Relative Intensity
524.1 $\pm$ 0.3	1.0	1436.7 $\pm$ 0.2	1.8 $\pm$ 0.3
527.5 $\pm$ 0.3	1.0	1458.6 $\pm$ 0.2	2.0 $\pm$ 0.3
572.0 $\pm$ 0.3	0.6 $\pm$ 0.1	1501.4 $\pm$ 0.3	0.8 $\pm$ 0.1
625.21 $\pm$ 0.06	8.0	1551.0 $\pm$ 0.4	0.5 $\pm$ 0.2
635.9 $\pm$ 0.2	<1	1568.8 $\pm$ 0.4	1.0 $\pm$ 0.2
647.89 $\pm$ 0.09	15	1653.7 $\pm$ 0.2	1.8 $\pm$ 0.3
660.8 $\pm$ 0.3	0.6 $\pm$ 0.1	1682.3 $\pm$ 0.2	2.8 $\pm$ 0.4
675.1 $\pm$ 0.3	1.1 $\pm$ 0.2	1712.9 $\pm$ 0.4	0.8 $\pm$ 0.2
698.5 $\pm$ 0.2	1.2 $\pm$ 0.2	1735.3 $\pm$ 0.4	<1
704.5 $\pm$ 0.2	<1	1740.8 $\pm$ 0.4	1.0 $\pm$ 0.2
739.0 $\pm$ 0.1	11	1795.4 $\pm$ 0.3	1.3 $\pm$ 0.2
826.3 $\pm$ 0.3	1.0 $\pm$ 0.2	1989.6 $\pm$ 0.3	<1
831.6 $\pm$ 0.1	3.8	2026.4 $\pm$ 0.2	1.0 $\pm$ 0.2
876.1 $\pm$ 0.1	9.2		

TABLE II

TRANSITION ENERGIES AND RELATIVE GAMMA-RAY
INTENSITIES FOR ^{142}Ba . UNCERTAINTIES ARE AS IN TABLE I.

E_{γ} (keV)	Relative Intensity	E_{γ} (keV)	Relative Intensity
69.7 $\pm$ 0.2	1.0 $\pm$ 0.2	417.6 $\pm$ 0.2	2.2 $\pm$ 0.3
77.59 $\pm$ 0.02	42	424.97 $\pm$ 0.06	30
84.4 $\pm$ 0.2	<1	432.22 $\pm$ 0.08	6.0
123.05 $\pm$ 0.07	4.2	473.3 $\pm$ 0.1	2.4 $\pm$ 0.4
154.5 $\pm$ 0.1	2.7 $\pm$ 0.4	590.3 $\pm$ 0.3	3.3 $\pm$ 0.5
176.97 $\pm$ 0.06	6.5	599.71 $\pm$ 0.08	12
216.3 $\pm$ 0.1	1.2 $\pm$ 0.2	608.9 $\pm$ 0.2	2.0 $\pm$ 0.4
222.9 $\pm$ 0.1	1.7 $\pm$ 0.3	769.5 $\pm$ 0.2	3.8 $\pm$ 0.7
231.63 $\pm$ 0.06	57	840.3 $\pm$ 0.1	18
242.80 $\pm$ 0.08	1.5 $\pm$ 0.2	895.0 $\pm$ 0.1	69

TABLE II(Contd.)

E	E_{γ} (keV)	Relative Intensity	E_{γ} (keV)	Relative Intensity
255.29 $\pm$ 0.06		100	948.9 $\pm$ 0.1	58
269.65 $\pm$ 0.07		4.6	1001.0 $\pm$ 0.1	52
286.30 $\pm$ 0.06		5.9	1078.6 $\pm$ 0.1	59
309.16 $\pm$ 0.07		15	1093.7 $\pm$ 0.1	16
334.97 $\pm$ 0.05		7.6	1123.1 $\pm$ 0.2	2.3 $\pm$ 0.6
337.4 $\pm$ 0.2		1.6 $\pm$ 0.3	1126.6 $\pm$ 0.1	8.9
363.94 $\pm$ 0.05		28	1148.5 $\pm$ 0.2	2.3 $\pm$ 0.5
379.2 $\pm$ 0.2		3.5 $\pm$ 0.5	1203.9 doublet	97
381 $\pm$ 1		$\sim$ 1	1380.1 $\pm$ 0.1	15

Fig. 1 Proposed decay scheme for ^{141}Ba . Gamma-ray energies are in keV. The intensities shown (numbers printed diagonally in light characters) are relative values for the gamma-ray de-excitation of the particular level above which they appear. Beta-ray intensities have been calculated from relative gamma-ray intensities in and out of each level. The 190.33-keV transition was assumed to have M1 character for beta-branch calculations.

Fig. 2 Proposed decay scheme for ¹⁴²Ba. See Figure 1 for a discussion of the format used. The 77.59-keV transition was assumed to have M1 character for beta-branch calculations.

MEASUREMENT OF SPALLATION CROSS SECTIONS FOR 600-MeV
PROTONS INCIDENT ON THIN COPPER, NICKEL, AND ALUMINUM TARGETS

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As part of a continuing program to determine the abundance and species of radionuclides resulting from the long-term bombardment of bulk materials by solar and cosmic protons, we have measured the cross sections for the production of radionuclides by 600 MeV protons on copper, nickel, and aluminum targets. Production cross sections for over 50 nuclides having half-lives greater than 20 seconds have been obtained. It is anticipated that, eventually, cross sections for incident protons of energies from 20 to 3000 MeV will be obtained for the common elements such as calcium, silicon, sodium, aluminum, chlorine, etc, and structural materials such as iron, vanadium, and copper.

Samples of copper, nickel, and aluminum foil, 2 to 5 mils thick, were irradiated in the internal beam of the 600-MeV proton synchrocyclotron at the Space Radiation Effects Laboratory at Newport News, Virginia. Irradiation times for these targets were 5 minutes, 30 minutes, and 11 hours. The three target materials were irradiated simultaneously so that the integrated proton flux could be determined through the well known cross section for the reaction $^{27}\text{Al}(p;3p,n)^{24}\text{Na}$, 10.8 ± 0.4 mb. The gamma rays from these samples were counted with a high-resolution Ge(Li) detector at times following irradiation of 1, 4, 10, 17, 32, 125, 230, 490, 970, 2000, and 4000 hours in order to obtain data on all nuclides having half-lives greater than 30 minutes. Samples were irradiated for times of 1, 5, and 30 minutes using the external beam of the synchrocyclotron. A pneumatic rabbit system using 0.375-inch diameter polyethylene tubing was used in these irradiations. The transit time for the rabbit was about five seconds. Samples from these irradiations were counted at periods of 15 seconds, 1 minute, 5 minutes, 10 minutes, 20 minutes, 40 minutes, 1 hour, 2 hours, and 4 hours. From all of these measurements, data on nuclides having half-lives greater than 20 seconds were obtained.

A typical set of data is shown in Figures 1 through 6. These show gamma-ray pulse-height spectra from the activation of the copper targets. The spectra, taken at various times following irradiation, from several of the targets, have been normalized in intensity to one another so that they may be viewed as the decay of a single sample, counted at different times for the same counting period and geometry. One can easily see in these spectra, the decay of the shorter lived species and the relative build up of the longer lived nuclides. All of the spectra taken in all of the experiments were analyzed to obtain relative gamma-ray intensities and energies for each of the spectra. The energies which were obtained have uncertainties of about 0.2 keV. No extraordinary techniques were used to extract more precise energies since they served only to identify the nuclide from which they were emitted. The gamma-ray intensities are felt to be accurate, at least for the more intense gamma rays in each spectra, to about 5%. These values were used to obtain relative intensities for the various nuclides that were observed in the data. The techniques for the analysis of these data have been developed over the past few years at this laboratory. They currently involve the use of a PDP-9 digital processor, and a 4096-channel spectrum can be completely analyzed for gamma-ray energies and intensities in approximately 30 minutes. Table I presents a listing of the nuclides identified

in these studies and the gamma-ray energies which have been identified as being associated with their decay. This identification was made on the basis of half-life data on the various gamma rays and on previously determined decay data for the various nuclides. In some cases the nuclidic assignment was ambiguous since the nuclear level giving rise to the transition was excited through the decay of different nuclide isobars or isomers. As a more pertinent example of these problems, consider the 1173-keV transition, present in the spectra as a result of the decay of 1.6-min ^{62m}Co , 13.9-min ^{62}Co , 9.8-min ^{62}Cu , and 9.3-hour ^{62}Zn , which has ^{62}Cu as a daughter. At longer times an 1173-keV gamma ray is also present as a result of the decay of 5.3-year ^{60}Co . Thus, in the spectra taken at early times, although the intensity of the 1173-keV transition was well determined, the division of this intensity into the components corresponding to the decay of the various nuclides has somewhat less accuracy. In many cases the division is assisted by the presence of other gamma rays in the decay of one of the nuclides that is not present in the decays of the others.

Table II lists the production cross sections which have been obtained from these measurements. These values were obtained by (a) extrapolating the relative decay curves for the particular gamma rays to the irradiation time, correcting for the effects of finite irradiation and counting periods, (b) calculating the relative number of atoms of each species present at this time, using the known branching ratio for the particular transition, and (c) converting to absolute cross section using the measured total proton intensity from the abundance of ^{24}Na in the aluminum samples and the absolute intensity of a major gamma ray in one of the spectra from each of the target materials. The table also lists nuclides which were not observed, including the stable isotopes, for the sake of completeness. It has been observed that the cross section as a function of A for constant Z or as a function of Z for constant A is smoothly varying. It is, therefore, possible to interpolate with limited degree of accuracy values for cross sections for those nuclides not observed which lie between nuclides for whom cross section values were obtained.

The next measurements will be of the production of nuclides having half-lives less than 20 seconds. This group of nuclides includes most of the products resulting from the bombardment of aluminum. To make these measurements, it will be necessary to count the gamma rays emitted by the targets between the proton pulses in the external beam of the accelerator (200 μs pulses, 54 pulses/sec). Since the prompt gamma rays emitted during the reactions with the protons are so very intense, it is necessary in these experiments to gate off the preamplifier of the Ge(Li) spectrometer as well as the pulse-height analyzer during the burst. Preliminary measurements have indicated that the counting rate during the burst results in a detector current of about 1 mA. This current results in a voltage at the input to the FET of sufficient magnitude to bias the FET off. The high values of the feed-back resistor (1000 meg) prevents the charge from leaking off sufficiently fast so that the circuit is able to be operated during the interval between the beam pulses. To alleviate this problem, we have constructed a preamplifier which contains a transistor used as a high-resistance diode in series with 1 meg resistor shunting the 1000 meg feed-back resistor. This diode can then be gated on during the beam in order to drain the charge off the input of the FET.

Fig. 1 Gamma-ray spectra from copper targets irradiated with 600-MeV protons: channels 0 to 1023 at decay times of 30 sec, 5 min, 15 min, 1 hr, and 4 hr.

Fig. 2 Gamma-ray spectra from copper targets irradiated with 600-MeV protons: channels 1024 to 2047 at decay times of 30 sec, 5 min, 15 min, 1 hr, and 4 hr.

Fig. 3 Gamma-ray spectra from copper targets irradiated with 600-MeV protons: channels 2048 to 3071 at decay times of 30 sec, 5 min, 15 min, 1 hr, and 4 hr.

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Fig. 4 Gamma-ray spectra from copper targets irradiated with 600-MeV protons: channels 0 to 1023 at decay times of 5 hr, 16 hr, 31 hr, 124 hr, and 480 hr.

Fig. 5 Gamma-ray spectra from copper targets irradiated with 600-MeV protons: channels 1024 to 2047 at decay times of 5 hr, 16 hr, 31 hr, 124 hr, and 480 hr.

Fig. 6 Gamma-ray spectra from copper targets irradiated with 600-MeV protons: channels 2048 to 3071 at decay times of 5 hr, 16 hr, 31 hr, 124 hr, and 480 hr.

TABLE I

NUCLIDE AND GAMMA-RAY ENERGY OBSERVED FROM THE VARIOUS TARGETS

Nuclide	$T_{1/2}$	Gamma-ray Energies Observed	Target
^{63}Zn	38 m	669 keV	Cu
^{62}Zn	9 h	962 548 596	
^{64}Cu	13 h	1345	Cu
^{62}Cu	10 h	875 1173	Cu Ni
^{61}Cu	3 h	67 282 589 656 908 1185	Cu Ni
^{60}Cu	24 m	467 826 953 1332 1791 1861 3123 3193	Cu Ni
^{57}Ni	36 h	126 1377 1757 1919	Cu Ni
^{56}Ni	6.4 d	163 748 812 1560	Cu Ni
$^{62\text{m}}\text{Co}$	2 m	1172	Cu Ni
^{62}Co	14 m	1163 1172	Cu Ni
^{61}Co	99 m	67	Cu Ni
^{60}Co	5 y	1172 1332	Cu Ni
^{58}Co	71 d	810	Cu Ni Fe
^{57}Co	270 d	122 136	Cu Ni Fe
^{56}Co	77 d	846 1037 1238 1359 1771 2014 2034 2178	Cu Ni Fe

TABLE I (Cont.)

Nuclide	$T_{1/2}$		Gamma-ray Energies Observed	Target
^{56}Co (Cont)			2249 keV	
			2597	
			3252	
			3449	
^{55}Co	18	h	476	Cu Ni Fe
			931	
^{59}Fe	45	d	1099	Cu Ni Fe
			1291	
^{53}Fe	9	m	377	Cu Ni Fe
^{52}Fe	8	h	169	Cu Ni Fe
^{58}Mn	1	m	810	Cu Ni Fe
^{57}Mn	2	m	122	Cu Ni Fe
^{56}Mn	2.6	h	846	Cu Ni Fe
			1810	
			2112	
^{54}Mn	303	d	834	Cu Ni Fe
$^{52\text{m}}\text{Mn}$	21	m	1434	Cu Ni Fe
^{52}Mn	6	d	744	Cu Ni Fe
			935	
			1246	
			1332	
^{50}Mn	2	m	783	Cu Ni Fe
^{56}Cr	6	m	84	Cu Ni Fe
^{51}Cr	28	d	320	Cu Ni Fe
^{49}Cr	42	m	62	Cu Ni Fe
			90	
			152	
^{48}Cr	24	h	308	Cu Ni Fe
^{53}V	2	m	1011	Cu Ni Fe
^{52}V	4	m	1434	Cu Ni Fe
^{48}V	16	d	944	Cu Ni Fe
			983	
			1312	
			2240	
^{44}Ti	48	y	78	Cu Ni Fe
^{48}Sc	1.8	d	178	Cu Ni Fe
			983	
			1037	
			1312	
^{47}Sc	3.4	d	159	Cu Ni Fe
$^{46\text{m}}\text{Sc}$	20	s	142	Cu Ni Fe
^{46}Sc	84	d	889	Cu Ni Fe
			1120	

TABLE I (Cont.)

Nuclide	$T_{1/2}$	Gamma-ray Energies Observed	Target
^{44m}Sc	2.4 d	271 keV	Cu Ni Fe
^{44}Sc	4 h	1157	Cu Ni Fe
^{43}Sc	4 h	372	Cu Ni Fe
^{42m}Sc	61 s	438	Cu Ni Fe
		1228	
		1525	
^{44}K	22 m	1157	Cu Ni Fe
^{43}K	22 h	372	Cu Ni Fe
		617	
^{42}K	12 h	1525	Cu Ni Fe
^{38}K	8 m	2168	Cu Ni Fe
^{41}Ar	2 h	1293	Cu Ni Fe
^{40}Cl	1.4 m	1461	Cu Ni Fe
^{39}Cl	55 m	249	Cu Ni Fe
^{38}Cl	37 m	1642	Cu Ni Fe
		2168	
^{29}Al	7 m	1273	Fe
^{28}Al	2.3 m	1778	Fe
^{27}Mg	10 m	843	Al
		1014	
^{25}Na	1 m	584	Al
		974	
^{24}Na	15 h	1368	Cu Ni Fe Al
		2754	
^{22}Na	2.6 y	1275	Al
^{24}Ne	3.4 m	472	Al
^{23}Ne	38 s	440	Al
^{20}F	11 s	1633	Al
^{16}N	7 s	2750	Al
		6134	
^7Be	53 d	477	Al

TABLE II

CROSS SECTIONS FOR PRODUCTION OF RADIONUCLIDES WITH HALF LIVES
LONGER THAN 20 SECONDS BY IRRADIATIONS WITH 600 MeV PROTONS

Nuclide	T _{1/2}	γ-Ray Energy	γ-Ray Branching	Cu	σ (mb)		
					Ni	Fe	Al
⁶⁵ Zn	245 d	1114	46 %		--	--	--
⁶⁴ Zn	stable	----	----		--	--	--
⁶³ Zn	38.4 m	669	9 %				
		962	7 %	3.7 ±1.0	--	--	--
⁶² Zn	9.3 h	596	22 %	0.97±0.2	--	--	--
⁶¹ Zn	1.5 m	480	11 %				
⁶⁴ Cu	12.8 h	1346	0.5 %	30.5 ±2.5		--	--
⁶² Cu	9.8 m	1172	1 %	47.2 ±3.5		--	--
⁶¹ Cu	3.32h	284	12 %	24.4 ±2.0		--	--
⁶⁰ Cu	24 m	1332	80 %	5.4 ±1.0		--	--
⁵⁹ Cu	81 s	343	45 %			--	--
		460	40 %				
⁶² Ni	stable	----	----			--	--
⁶¹ Ni	stable	----	----			--	--
⁶⁰ Ni	stable	----	----			--	--
⁵⁹ Ni	10 ⁵ y	no γ	----			--	--
⁵⁸ Ni	stable	----	----			--	--
⁵⁷ Ni	36 y	1377	80 %	2.1 ±0.7		--	--
⁵⁶ Ni	6.4 d	163	99 %	1.1 ±0.3		--	--
⁶² Co	13.9 m	1172	100 %	0.5 ±0.3			--
^{62m} Co	1.6 m	1172	----	0.4/x ±0.3/x			--
⁶¹ Co	99 m	67	100 %	1.5 ±0.4	35.0 ±2.0		--
⁶⁰ Co	5.26y	1332	100 %	31.7 ±2.0	41.4 ±2.0		--
^{60m} Co	10.5 m	59	99 %				--
⁵⁹ Co	stable	----	----				--
⁵⁸ Co	71.3 d	810	99.5 %	59.0 ±3.5	34.7 ±2.0		--
⁵⁷ Co	270 d	121	100 %	38.2 ±3.0	56.5 ±3.0		--
⁵⁶ Co	77 d	1238	66 %	55.8 ±3.5	43.5 ±2.5		--
⁵⁵ Co	18.2 h	477	12 %	4.7 ±1.0	11.3 ±1.3		--

TABLE II (Cont.)

Nuclide	$T_{1/2}$	γ -Ray Energy	γ -Ray Branching	Cu	σ (mb)		Al
					Ni	Fe	
^{61}Fe	6.0 m	1210	100 %				--
^{60}Fe	stable	----	----				--
^{59}Fe	45 d	1099 1291	56 %	3.0 ± 0.8			--
^{58}Fe	stable	----	----				--
^{57}Fe	stable	----	----				--
^{56}Fe	stable	----	----				--
^{55}Fe	2.94y	No γ	----				--
^{54}Fe	stable	----	----				--
^{53}Fe	8.5 m	378	49 %	2.4 ± 0.7			--
^{52}Fe	8.2 h	169	100 %	0.3 ± 0.2	1.2 ± 0.3		--
^{58}Mn	1.1 m	810	X	$0.15/x \pm 0.1/x$			--
^{57}Mn	1.7 m	121	X	$0.07/x \pm 0.06/x$			--
^{56}Mn	2.58h	846	100 %	6.5 ± 1.0	0.7 ± 0.2		--
^{55}Mn	stable	----	----				--
^{54}Mn	303 d	834	100 %	37.7 ± 3.0	16.5 ± 1.6		--
^{53}Mn	10^6 y	No γ	----				--
^{52}Mn	21 m	1434	98 %	5.9 ± 1.0	18.1 ± 1.7		--
^{52}Mn	5.7 d	744	100 %	30.2 ± 2.5	19.2 ± 1.7		--
^{51}Mn	45 m	1560	X				--
^{50}Mn	2 m	783	100 %	0.18 ± 0.13			--
^{56}Cr	5.9 m	84	100 %	0.07 ± 0.04			--
^{55}Cr	3.5 m	No γ	----				--
^{54}Cr	stable	----	----				--
^{53}Cr	stable	----	----				--
^{52}Cr	stable	----	----				--
^{51}Cr	27.8 d	320	9 %	34.3 ± 2.5	51.8 ± 3.0		--
^{50}Cr	stable	----	----				--
^{49}Cr	42 m	90	30 %	3.8 ± 0.8	4.6 ± 0.8		--
^{48}Cr	23.5 h	308	100 %	0.6 ± 0.4	1.8 ± 0.6		--

TABLE II (Cont.)

Nuclide	T _{1/2}	γ -Ray Energy	γ -Ray Branching	σ (mb)			Al
				Cu	Ni	Fe	
⁵⁴ V	55 s	835	100 %				--
⁵³ V	2.0 m	1011	100 %	0.6 $\pm$ 0.4			--
⁵² V	3.76m	1434	100 %	0.7 $\pm$ 0.4			--
⁵¹ V	stable	----	----				--
⁵⁰ V	10 ¹⁴ y	----	----				--
⁴⁹ V	330 d	No γ	----				--
⁴⁸ V	16.0 d	983	100 %	10.3 $\pm$ 1.5	21.7 $\pm$ 1.8		--
⁴⁷ V	33 m	1800	0.8%				--
⁵¹ Ti	5.8 m	320	100 %				--
⁵⁰ Ti	stable	----	----				--
⁴⁹ Ti	stable	----	----				--
⁴⁸ Ti	stable	----	----				--
⁴⁷ Ti	stable	----	----				--
⁴⁶ Ti	stable	----	----				--
⁴⁵ Ti	3.09h	718	0.4%				--
⁴⁴ Ti	48 y	78	100 %				--
⁵⁰ Sc	1.72m	1550	100 %				--
⁴⁹ Sc	57.5	1780	0.03%				--
⁴⁸ Sc	1.83d	983	100 %	1.3 $\pm$ 0.4	3.4 $\pm$ 0.7		--
⁴⁷ Sc	3.4 d	159	73 %	1.7 $\pm$ 0.5	2.9 $\pm$ 0.6		--
^{46m} Sc	20 s	142	100 %	0.05 $\pm$ 0.05			--
⁴⁶ Sc	83.9 d	889	100 %	3.8 $\pm$ 0.9	4.1 $\pm$ 0.8		--
⁴⁵ Sc	stable	----	----				--
^{44m} Sc	2.44d	271	98.6%	2.1 $\pm$ 0.6	10.6 $\pm$ 1.3		--
⁴⁴ Sc	3.92h	1157	100 %	4.6 $\pm$ 1.0	4.1 $\pm$ 0.8		--
⁴³ Sc	3.92h	372	22 %	6.4 $\pm$ 1.0			--
^{42m} Sc	61 s	437	100 %	0.09 $\pm$ 0.07			--
⁴⁹ Ca	8.8 m	3100	89 %				--
⁴⁸ Ca	10 ¹⁶ y	----	----				--
⁴⁷ Ca	4.53d	1300	71 %				--

TABLE II (Cont.)

Nuclide	T _{1/2}	γ-Ray Energy	γ-Ray Branching	Cu	σ (mb)		Al
					Ni	Fe	
⁴⁶ Ca	stable	-----	-----				--
⁴⁵ Ca	164 d	No γ	-----				--
⁴⁴ Ca	stable	-----	-----				--
⁴³ Ca	stable	-----	-----				--
⁴² Ca	stable	-----	-----				--
⁴¹ Ca	10 ⁵ y	No γ	-----				--
⁴⁰ Ca	stable	-----	-----				--
⁴⁵ K	16 m	1710	60 %				--
⁴⁴ K	22 m	1157	65 %	0.7 ±0.4			--
⁴³ K	22.4 h	617	81 %	0.8 ±0.5	1.5 ±0.5		--
⁴² K	12.4 h	1525	18 %	1.8 ±0.6	1.8 ±0.5		--
⁴¹ K	stable	-----	-----				--
⁴⁰ K	10 ⁹ y	-----	-----				--
³⁹ K	stable	-----	-----				--
³⁸ K	7.71m	2168	100 %	0.07±0.07			--
⁴¹ Ar	1.83h	1293	99.2%	0.4 ±0.2			--
⁴⁰ Ar	stable	-----	-----				--
³⁹ Ar	3x10 ² y	No γ	-----				--
³⁸ Ar	stable	-----	-----				--
³⁷ Ar	34.1 d	No γ	-----				--
³⁶ Ar	stable	-----	-----				--
⁴⁰ Cl	1.4 m	1460	X	0.13/x ±0.09/x			--
³⁹ Cl	55.5 m	249	90 %	0.17±0.10			--
³⁸ Cl	37.3 m	2168	100 %	0.20±0.10			--
³⁷ Cl	stable	-----	-----				--
³⁶ Cl	10 ⁵ y	-----	-----				--
³⁵ Cl	stable	-----	-----				--
³⁴ mCl	32.4 m	2140	40 %		0.09±0.04		--

TABLE II (Cont.)

Nuclide	$T_{1/2}$	γ -Ray Energy	γ -Ray Branching	Cu	σ (mb)		Al
					Ni	Fe	
^{38}S	2.87h	1880	95 %				--
^{37}S	5.06m	3090	90 %				--
^{36}S	stable	----	----				--
^{35}S	87.1 d	No γ	----				--
^{34}S	stable	----	----				--
^{33}S	stable	----	----				--
^{32}S	stable	----	----				--
^{31}S	2.6 s	No γ	----				--
^{30}S	1.4 s	687	80 %				--
^{31}P	12.4 s	2130	25 %				--
^{33}P	24.4 d	No γ	----				--
^{32}P	14.3 d	No γ	----				--
^{31}P	stable	----	----				--
^{30}P	2.6 m	2230	0.5%				--
^{29}P	4.4 s	1280	0.8%				--
^{31}Si	2.62h	1270	0.07%				
^{30}Si	stable	----	----				
^{29}Si	stable	----	----				
^{28}Si	stable	----	----				
^{27}Si	4.2 s	No	----				
^{26}Si	2.1 s	820	34 %				
^{30}Al	3.3 s	2260	60 %				
^{29}Al	6.56m	1280	94 %				
^{28}Al	2.31m	1780	100 %				
^{27}Al	stable	----	----				
$^{26\text{m}}\text{Al}$	6.4 s	No γ	----				
^{26}Al	$7.4 \times 10^5 \text{y}$	1810	99 %				
^{25}Al	7.8 s	No γ	----				
^{24}Al	2.1 s	1390	40 %				

TABLE II (Cont.)

Nuclide	T _{1/2}	γ-Ray Energy	γ-Ray Branching	Cu	σ (mb)		Al
					Ni	Fe	
²⁸ Mg	21 h	1350	.70 %				--
²⁷ Mg	9.5 m	843	70 %				--
²⁶ Mg	stable	----	----				
²⁵ Mg	stable	----	----				
²⁴ Mg	stable	----	----				
²³ Mg	12.1 s	439	9 %				
²² Mg	4 s	590					
²⁶ Na	1.0 s	1810	100 %				
²⁵ Na	60 s	974	15 %				0.62±0.25
²⁴ Na	15 h	1368	100 %				*10.8
²³ Na	stable	----	----				
²² Na	2.6 y	1275	100 %				15.0 ±1.5
²¹ Na	23 s	350	2.3%				
²⁴ Ne	3.38m	472	100 %				0.15±0.07
²³ Ne	37.6 s	439	33 %				
²² Ne	stable	----	----				
²¹ Ne	stable	----	----				
²⁰ Ne	stable	----	----				
¹⁹ Ne	17.5 s	No γ	----				
¹⁸ Ne	1.5 s	No γ	----				
²¹ F	4.4 s	350	100 %				
²⁰ F	11.4 s	1627					
¹⁹ F	stable	----	----				
¹⁸ F	110 m	No γ	----				
¹⁷ F	66 s	No γ	----				
²⁰ O	14 s	1070	100 %				
¹⁹ O	29 s	1370	60 %				
¹⁸ O	stable	----	----				
¹⁷ O	stable	----	----				

* Used to determine flux of protons.

TABLE II (Cont.)

Nuclide	$T_{1/2}$	γ -Ray Energy	γ -Ray Branching	Cu	σ (mb)		Al
					Ni	Fe	
^{16}O	stable	----	----				
^{15}O	2 m	No γ	----				
^{14}O	71 s	2311	99.4%				
^{17}N	4.2 s	871	98 %				
^{16}N	7.2 s	6131	70 %				
^{15}N	stable	----	----				
^{14}N	stable	----	----				
^{13}N	10 m	No γ	----				
^{15}C	2.4 s	5299	68 %				
^{14}C	5700 y	No γ	----				
^{13}C	stable	----	----				
^{12}C	stable	----	----				
^{11}C	20.3 m	No γ	----				
^{10}C	19.4 s	717	100 %				
^{11}B	stable	----	----				
^{10}B	stable	----	----				
^{11}Be	13.6 s	2140	29 %				
^{10}Be	2.5×10^6 y	No γ	----				
^9Be	stable	----	----				
^8Be	2×10^{-16} s	No γ	----				
^7Be	53 d	477	10.3%				1.6 ± 0.4

EVIDENCE FOR THE EXISTENCE
OF AN ISOMERIC STATE IN ^{241}Pu ^[1]

R. G. Nisle, I. E. Stepan

Recent analysis of data suggests the existence of a previously unreported isomeric state in ^{241}Pu . This evidence is the outcome of work undertaken to obtain an accurate value of the beta-decay half-life of ^{241}Pu . Previously reported values varied from 13.0 ± 0.2 years[2] to 14.98 ± 0.33 years[3].

The technique we employed[4] makes use of the fact that neutron absorption produces a negative response in a nuclear reactor, while fission has a positive response. The use of two reactors having different relative responses to absorption and fission enables one to measure both absorber and fissile-element content of a sample[5]. We have used two such precision reactivity-measuring reactors[6] to measure the change in ^{241}Pu content of a sample over a period of time. Variations in content of 0.2 mg in 200 mg of ^{241}Pu are readily detectable by this technique. In cases where the results of our experiments have been compared with the results of chemical analyses on a given sample, the sample contents as obtained by the two methods have always agreed to within the quoted uncertainties of the two methods. For the sample used in this work the results of such a comparison are

Initial Sample Content:

Total Plutonium $\left\{ \begin{array}{l} \text{by this technique} = 0.2108 \pm 0.0002 \text{ g} \\ \text{by chemical analysis} = 0.2120 \pm 0.0021 \text{ g} \end{array} \right.$

Final Sample Content:

Total Plutonium $\left\{ \begin{array}{l} \text{by this technique} = 0.1916 \pm 0.0002 \text{ g} \\ \text{by chemical analysis} = 0.1920 \pm 0.0019 \text{ g} \end{array} \right.$

The sample used for these measurements consisted of an alloy of 3% Pu, enriched to 94.01% ^{241}Pu , in high purity aluminum (>99.99% Al). The plutonium source material was of indeterminate age and could have been as old as three or four years. The enrichment, by calutron, was made about one year prior to the start of our measurements. About four months before starting these measurements this calutron-enriched material was chemically purified by ion-exchange techniques to remove ^{241}Am .

The results of such measurements, made over a period of two and a half years, are shown in Figure 1. The logarithm of the ^{241}Pu content of the sample is plotted on the Y-axis, and time is plotted on the X-axis. The dashed straight line is the expected decay for a single component having a half-life of 14.56 years, which value is based on a fit to the last ten points. The curved line is a least squares fit of a two-component equation to the data. The standard deviation of the latter fit is ± 0.17 mg which agrees well with the standard deviation of an individual data point of ± 0.16 mg obtained from repeated measurements on a stable sample. The indicated deviation of ± 1.2 mg of the data

from the predicted single-component decay curve at early times is about seven times the uncertainty in our measurements. Furthermore, the data deviate in a systematic manner from the single-component line. The resulting half-life for ^{241}Pu is 14.63 ± 0.27 years, and for the second component the half-life is 0.34 ± 0.11 year. The uncertainties quoted are standard deviations derived by the usual methods from the standard deviation of the fit of the two-component equation to the data.

The reactivity results require that the shorter lived component be fissionable by thermal neutrons. Such behavior could arise either from sample impurities or from an isomeric state in ^{241}Pu . We believe the first explanation is not likely. Based on the requirements of half-life and fissionability, the isotopes ^{241}Cm and ^{237}Pu might appear to be possible candidates for this second component. These isotopes, however, should have been removed by the sample purification processes described above. Consequently, we believe that the second alternative is the correct explanation of the effect we observed.

It is obviously of great importance to know what nucleonic configuration gives rise to the proposed isomeric state in ^{241}Pu , but aside from half-life, our data provide no information of that sort.

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Fig. 1 Decay data on the ^{241}Pu sample. The time of the first reactivity measurements is taken as $t = 0$, and the ^{241}Pu content is plotted on a logarithmic scale. The standard deviation of the measured values of the ^{241}Pu content of the sample is ± 0.17 mg. This value is comparable to the size of the data points shown.

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D. MATERIALS

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MICROSCOPIC DYNAMICS OF LIQUID ^{36}A BY SLOW
NEUTRON SCATTERING

P. D. Randolph, K. Sköld^[1], J. M. Rowe^[2], G. O. Ostrowski^[3]

One of the most effective methods for studying the nature of the liquid state is that of slow neutron inelastic scattering. In particular, information can be obtained about the microscopic dynamics of the atoms of the liquid. For monatomic systems the neutron-nuclear scattering cross section determines what aspects of the atomic motion can be observed. For materials having a coherent scattering cross section, the correlated motion of atom pairs determines the scattered neutron spectra, while for an incoherent cross section, only the single atom motions are seen. Most monatomic materials have cross sections that are all coherent, or else a mixture of coherent and incoherent. Thus in a neutron scattering experiment only the pair motions are seen or else an unseparated mixture of the two types of atomic motion. As yet no adequate first principles theory exists for those motions in liquids, and only phenomenological mathematical models are available to describe them. In addition there are ad hoc models to describe the relation between single particle and pair motions. Comparisons of experiments on purely coherent scatterers with models for pair motion have been made [4]. These indicate that current models which treat the liquid as a kind of quasisolid predict some of the qualitative features observed, though they are by no means exact. Most theoretical models have dealt with the single particle or diffusive motions of atoms, since these are somewhat simpler to deal with mathematically. However experiments to observe only the diffusive motion of single particles are difficult primarily because only one naturally occurring monatomic completely incoherent scatterer is known -- vanadium metal. This measurement is intractable because of the high melting point of vanadium. An ideal monatomic substance for studying liquid dynamics is liquid argon which has the advantage of a low melting point. In addition, it is one of the simplest of liquids being an insulator with Van der Waals type interatomic forces. Natural argon is a mixture of argon-36 and argon-40 and its cross section is partly coherent and partly incoherent. Separation of the two kinds of motions which occur can only be done by theories which themselves are either inadequate or have not been tested.

In order to overcome these difficulties an experiment has been performed on two different samples of liquid argon. The first was a sample of pure argon-36 which has a completely coherent cross section, and the second was a mixture of argon-36 and argon-40 with the proportions chosen to give maximal incoherent scattering (7.7% ^{36}A , 92.2% ^{40}A). Making the reasonable assumption that the atomic motions are not significantly different in the two samples, this experiment allows for the first time an experimental separation of the single particle and two particle motions without recourse to a theoretical model. The purpose of the experiment is to do the separation to make detailed comparisons with current theories of single particle motions, pair motions, and models relating the two. This type of comparison has not been done before.

The experiment was performed at the NRTS using the MTR phased-chopper velocity selector. The samples and cryostat were supplied by Argonne National Laboratory. Data runs of 80 to 100 hours duration were obtained on each sample at incident neutron energies of 15 and 20 meV for each and at 15 scattering

angles between 18 and 90 degrees. A helium filled scattered neutron flight path was used to reduce air scattering[4]. Figure 1 shows representative raw sample and background time-of-flight spectra for the liquid argon-36 sample at 20 meV incident energy and data at the same angles and energy for the mixture sample.

This experiment was a cooperative effort by Idaho Nuclear Corporation and the Solid State Sciences Division of Argonne National Laboratory. As of this writing, data acquisition has just been completed. Preliminary data analysis is being done at the NRTS and the final analysis will be done at ANL.

- [1] Swedish scientist from Studsvik on assignment to ANL.
- [2] Senior scientist, ANL, Chicago, Illinois.
- [3] Staff scientist, ANL, Chicago, Illinois.
- [4] P. D. Randolph and K. S. Singwi, "Slow-Neutron Scattering and Collective Motions in Liquid Lead", Phys. Rev., **152** n 1 (December 1966) pp 99-112.

Fig. 1. Raw scattered neutron and background spectra at 20 meV incident energy for liquid argon-36 and a maximally incoherent mixture of argon-36 and -40.

BISMUTH III DIFFRACTION DATA AT 30 KBAR

R. B. Bennion, R. M. Brugger

In a continuation of the study of the polymorphs of bismuth metal at high pressures, neutron diffraction patterns were obtained of bismuth above the II-III transition at 27 kbar. Figure 1 shows a typical pattern obtained comparing Bi-III with a pattern representing Bi-II. A complete change is noted in the structure; the structure is complicated and about 25 lines of the Bi-III pattern are distinct. The reason for the difference in the general pattern below 1.0 Å is that a new chopper which eliminates the faster neutrons was used for the Bi-III data. This chopper allows the use of the "overlap" neutrons where the large d-values appear in the 90 degree scattering data (not shown). The d-values listed in Table I are a compilation of best values obtained from several runs using both the 60 and 90 degree scattering systems.

The efforts to index the Bi-III diffraction data and determine the resulting space group have thus far been unsuccessful; however, this effort is continuing[1]. The similarity in the two diffraction patterns shown indicate a possible correlation between Bi-II and -III exists. Note that the $(11\bar{1})$ and $(20\bar{1})$ of the monoclinic Bi-II structure seem to increase intensity while the (020) , (001) , and (200) almost disappear in transforming to Bi-III. Below 2.5 Å there seems to be no significant similarity in the patterns. The question arises whether the Bi-II pattern is completely eliminated in the Bi-III pattern. The alumina cylinders consistently rupture at about 28 or 30 kbar with only bismuth in the sample chamber. However, the pattern was not significantly different in any of more than 10 runs on Bi-III.

The use of Ito's method that was so successful in indexing Bi-II has not been successful on Bi-III because of the uncertainty in the low intensity peaks of Bi-III above 3.0 Å. One must use the largest d-values as basis vectors in Ito's method. It is not certain whether there are 0, 1, 2, or even 3 peaks in the 3.0 to 3.1 Å region and so the structure of Bi-III remains unsolved. It is evident that the structure is not a highly symmetric system and is probably monoclinic or triclinic.

[1] W. H. Zachariasen, private communication.

Fig. 1 Comparison of the diffraction patterns of Bi-II and -III.

TABLE I

Bi-III OBSERVED d VALUES

Peak No.	d(A)	Peak No.	d(A)	Peak No.	d(A)	Peak No.	d(A)
1	3.072	11	2.068	21	1.526	31	1.223
2	3.034	12	2.015	22	1.509	32	1.196
3	2.865	13	1.935	23	1.471	33	1.175
4	2.745	14	1.908	24	1.444	34	1.142
5	2.492	15	1.885	25	1.393	35	1.050
6	2.189	16	1.762	26	1.360	36	1.043
7	2.170	17	1.725	27	1.328	37	1.002
8	2.156	18	1.679	28	1.295	38	0.978
9	2.121	19	1.602	29	1.287	39	0.957
10	2.087	20	1.584	30	1.247	40	0.940

PRESSURE DEPENDENCE OF THE DEBYE TEMPERATURE
OF NaCl TO 26 KBAR

W. R. Myers, R. B. Bennion, R. M. Brugger

The Debye-Waller coefficients of sodium chloride were measured as a function of pressure up to 26 kbar at room temperature using the MTR high-pressure neutron diffractometer. These Debye-Waller coefficients may be converted to a characteristic Debye temperature (as a function of pressure) to test various empirical equations of state for the sample. Time-of-flight diffraction patterns were measured for fixed scattering angles of 60 and 90 degrees at pressures in increments of 5 kbar and the Debye-Waller coefficients determined for each pressure.

The sample was 0.25 inch in diameter, contained in a 1.25-inch diameter Al_2O_3 pressure cell. A slotted binding ring, used to allow entry and exit of the neutron beam, provides collimation sufficiently fine that the sample holder does not contribute to the scattered beam.

Analysis of the data indicates a Debye temperature of $301 \pm 2^\circ\text{K}$ at standard conditions, which is in good agreement with currently accepted values. The pressure dependence is $0.9 \pm 0.2^\circ\text{K/kbar}$. This compares with a theoretically calculated slope [1] of 1.5°K/kbar .

[1] D. L. Decker, private communication.

THERMAL EXPANSION AND COMPRESSIBILITY MEASUREMENTS ON V_2O_5
BY NEUTRON DIFFRACTION

R. J. Riedner^[1,2], L. Cartz^[3], W. R. Myers, R. B. Bennion

Measurements of the diffraction patterns of V_2O_5 were made for several pressures between 0 and 23 kbar at 300°K , and at several temperatures from 25 up to 600°C at room pressure to determine the relationship of the thermal expansion coefficients to the anisotropic compressibility coefficients. Preliminary analysis shows that the thermal expansion coefficients are linear in the range 25 to 600°C at room pressure and are equal to 4, 5.9, and $29.3 \times 10^{-6}/^\circ\text{C}$ for the a , b , and c axes, respectively.

The coefficients of compressibility are not linear at 25°C . In the pressure range from 0 to 7 kbar, the compressibility coefficients are 0.31, 0.01, and $0.36 \times 10^{-6} \text{ cm}^2/\text{kg}$ along the a , b , and c axes; they are 1.75, 0.55, and -0.72 in the range 7 to 14 kbar, and 2.16, 0.90, -1.34 in the range 14 to 23 kbar.

[1] Graduate Student, Dept. of Mechanical Engineering, Marquette University.

[2] Partially supported by Associated Western Universities.

[3] Professor, Dept. of Mechanical Engineering, Marquette University.

CATALOGUE OF NEUTRON
MOLECULAR SPECTRA

K. A. Strong

A catalogue of neutron molecular spectra has been assembled [1]. It contains all spectra obtained to date using the small κ method of neutron molecular spectroscopy [2] at the MTR-WBRR. Spectra of 16 molecules are included under three headings: alkanes, alkane derivatives, and aromatic compounds.

The catalogue represents the combined contributions of several people: Dr. R. M. Brugger, Dr. H. L. McMurry, and K. A. Strong of Idaho Nuclear Corporation and Prof. R. J. Pugmire, Prof. D. M. Grant, and R. C. Livingston of the University of Utah. Its purpose is to provide a preliminary compilation of available spectra so that those interested may view them without waiting for final analysis and journal publication.

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- [1] K. A. Strong, Catalogue of Neutron Molecular Spectra, IN-1273 (to be published).
- [2] R. M. Brugger, K. A. Strong, D. M. Grant, "The 'Small K' Method of Neutron Molecular Spectroscopy", in Proceedings of a Symposium on Neutron Inelastic Scattering Held by the International Atomic Energy Agency in Copenhagen, 20-25 May, 1968, Vienna: International Atomic Energy Agency, 1968. pp 323-329.

THE C-C TORSIONAL VIBRATION IN LIQUID ETHANE MEASURED
BY NEUTRON MOLECULAR SPECTROSCOPY

K. A. Strong, R. M. Brugger, R. J. Pugmire^[1], D. M. Grant^[1], H. L. McMurry

Ethane has been studied in the liquid state at a temperature of -180°C (see Figure 1). The $0 \rightarrow 1$ transition of the torsional vibration is measured to be 36.4 meV (294 cm^{-1}) which is a shift of 16 cm^{-1} from the value measured in the room temperature gas phase reported previously [2]. Similar shifts have been observed in the torsional spectra of neopentane and other simple hydrocarbons. Data for ethane in the solid phase will soon be obtained to complete the study.

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- [1] Chemistry Dept., University of Utah.
- [2] K. A. Strong and R. M. Brugger, "Measurement of the Frequency of Torsional Vibration in the Ethane Molecule", J. Chem. Phys. **47**, 421 (1967).

Fig. 1 Reduced partial differential scattering cross section versus neutron energy change for liquid ethane. The arrow indicates the position of the torsional libration mode.

METHYL LIBRATIONS IN PROPANE MEASURED WITH NEUTRON INELASTIC SCATTERING

R. J. Pugmire^[1,2], D. M. Grant^[1,2], R. C. Livingston^[2,3], K. A. Strong,
H. L. McMurry, R. M. Brugger

Propane has been studied in the gaseous, liquid, and solid states. The two methyl librational frequencies of symmetry A_2 and B_2 have now been observed by means of neutron molecular spectroscopy (NMS) whereas numerous attempts to observe them in the optical spectroscopies have not been successful. The spectra for the three states of matter are presented in Figures 1, 2, and 3, and torsional frequencies are given in Table I. Recently, the torsional frequencies have been indirectly measured by analysis of the fine structure in the microwave spectra^[4]. The splitting patterns in question are the results of coupling of the librational motion to the overall molecular rotation.

Considerable effort has been expended during the year to obtain accurate values for the potential function in propane. The potential energy governing internal rotation of the two methyl groups in propane may be expressed with a two dimensional Fourier expansion in the internal coordinates. With the expansion of the potential written in exponential form, the resulting torsional wave equation can be written in the general form of Hill's modification to the Mathieu equation for which one can obtain accurate solutions. The three-fold and negative six-fold barriers restricting internal rotation are given in Table I along with the results of the perturbation calculation of Reference 2.

[1] Dept. of Chemistry, University of Utah.

[2] Partially supported by Associated Western Universities.

[3] Predoctoral student, Dept. of Chemistry, University of Utah.

[4] E. Hirota, C. Matsumura, and Y. Morino, Bull. Chem. Soc. Japan, 40, 1124 (1967).

TABLE I

METHYL TORSIONAL FREQUENCIES IN PROPANE

State	Temperature	Energy Change (meV)		Barrier Potentials (cal/mole)	
		A_2	B_2	V_3	V_6
Gas	298°K	25.4±8	31.9±8	3481±183	-357±119
Liquid	195°K	26.8 ^[a]	33.6 ^[a]	3325±20 ^[a]	-170±20 ^[a]
	92°K	28.0±0.4	33.5±0.4		
Solid	83°K	29.1±2	34.3±0.2		

[a] Reference 4.

Fig. 1 Reduced partial differential scattering cross sections versus neutron energy change for gaseous propane. The arrows point to the approximate position of the torsional libration modes.

Fig. 2 Reduced partial differential scattering cross sections versus neutron energy change for liquid propane. The arrows indicate the positions of the torsional libration modes.

Fig. 3 Reduced partial differential scattering cross sections versus neutron energy change for solid propane. The arrows indicate the positions of the torsional libration modes.

A STUDY OF ISOBUTANE WITH NEUTRON MOLECULAR SPECTROSCOPY

R. J. Pugmire^[1,2], D. M. Grant^[1], R. C. Livingston^[2,3], K. A. Strong,
H. L. McMurry, R. M. Brugger

Isobutane represents a three-top molecule with a doubly degenerate methyl torsional mode of symmetry E and the single degenerate A mode. Once again, these torsional vibrations have not been observed with optical spectroscopy but are readily observable with Neutron Molecular Spectroscopy, as portrayed in Figures 1, 2, and 3 for the three states of matter. Although the two torsional frequencies are not well resolved in the gas, reasonable ΔE values can be obtained from curve fitting techniques after completion of the analysis of the frequencies and intensities in the condensed states. The resulting vibrational frequencies are presented in Table I.

Theoretical work is in progress to calculate the barrier potential in a manner similar to that of propane, utilizing a three-dimensional Fourier expansion in methyl rotational coordinates.

[1] Dept. of Chemistry, University of Utah.

[2] Partially supported by Associated Western Universities.

[3] Predoctoral Student, Dept. of Chemistry, University of Utah.

TABLE I
METHYL TORSIONAL FREQUENCIES IN ISOBUTANE

<u>State</u>	<u>Temperature</u>	<u>Energy Change (meV)</u>	
		<u>E</u>	<u>A</u>
Gas	298°K	25.0±1	29.5±1
Liquid	148°K	27.1±0.3	31.8±0.3
Solid	103°K	28.3±0.3	33.4±0.3

Fig. 1 Reduced partial differential scattering cross sections versus neutron energy change for gaseous isobutane. The arrows point to the approximate position of the torsional libration modes.

Fig. 2 Reduced partial differential scattering cross sections versus neutron energy change for liquid isobutane. The arrows indicate the positions of the torsional libration modes.

Fig. 3 Reduced partial differential scattering cross sections versus neutron energy change for solid isobutane. The arrows indicate the positions of the torsional libration modes.

A STUDY OF NEOPENTANE WITH NEUTRON MOLECULAR SPECTROSCOPY

K. A. Strong, R. M. Brugger, R. J. Pugmire^[1], D. M. Grant^[1],
H. L. McMurry

Data have now been obtained for gas, liquid^[2], and two solid phases of neopentane. The librational modes A_2 and F_2 , which are inactive for other spectroscopic methods, have been measured in each case and the results are tabulated in Table I. Figures 1, 2, 3, and 4 present the spectra. It is noted that the energy change associated with the librational modes increases with the change in phase from gas to liquid, from liquid to solid, and again from solid phase II to solid phase I. This shift is due to an increase in the torsional barrier caused by intermolecular forces. Also apparent in the spectra is the improvement in resolution with phase change. This is due to decreased sample temperature and increased effective mass and is characteristic of the small κ method. Evidence of three skeletal vibration modes, two of which are well resolved in the solid phase data, is observed in the gaseous and liquid phase data.

[1] Chemistry Dept., University of Utah.

[2] D. M. Grant, K. A. Strong, R. M. Brugger, "Direct Observation of Methyl Librations in Neopentane", Phys. Rev. Letters, 20 n 18 (April 1968) pp 983-986.

TABLE I
NEOPENTANE

Sample			Energy Change, meV	
State	Temperature	Transition	A_2	F_2
Gas	297°K	0→1	24.5	31.6
Liquid	297°K	0→1	26	33
Solid Phase II	150°K	0→1	26.3	34
Solid Phase I	103°K	0→1	27	34.8

Fig. 1 Reduced partial differential scattering cross section versus neutron energy change for gaseous neopentane. The arrows point to torsional libration modes while hash marks indicate skeletal vibration modes.

Fig. 2 Reduced partial differential scattering cross section versus neutron energy change for liquid neopentane. The arrows point to torsional libration modes while hash marks indicate skeletal vibration modes.

Fig. 3 Reduced partial differential scattering cross section versus neutron energy change for solid (phase II) neopentane. The arrows point to torsional libration modes while hash marks indicate skeletal vibration modes.

Fig. 4 Reduced partial differential scattering cross section versus neutron energy change for solid (phase I) neopentane. The arrows point to torsional libration modes while hash marks indicate skeletal vibration modes.

RING BENDING MODES IN BENZENE BY NEUTRON MOLECULAR SPECTROSCOPY

R. J. Pugmire^[1,2], D. M. Grant^[1], R. C. Livingston^[2,3], K. A. Strong,
H. L. McMurry, R. M. Brugger

Benzene, with its low frequency out-of-plane ring bending modes, lends itself well to investigation by means of Neutron Molecular Spectroscopy. The resulting spectra shown in Figure 1 display three prominent peaks in the energy range obtainable with the velocity selector. The B_{2g} ring bending mode is Raman and infrared forbidden and is predicted to lie in the energy region in question. Data analysis is presently underway to assign the observed peaks and to determine if the forbidden transition can be resolved.

[1] Dept. of Chemistry, University of Utah.

[2] Partially supported by Associated Western Universities.

[3] Predoctoral Student, Dept. of Chemistry, University of Utah.

Fig. 1 Reduced partial differential scattering cross sections versus neutron energy change for solid benzene. The hash marks indicate the positions of ring bending modes.

VIBRATIONAL MODES IN ETHYL CHLORIDE MEASURED
BY NEUTRON SCATTERING

K. A. Strong, R. M. Brugger, R. J. Pugmire^[1]

Neutron molecular spectroscopy has been used to observe the C-C-Cl deformation and CH₃ torsional modes of ethyl chloride in the liquid and solid states (Figures 1 and 2). The neutron data for the torsional mode have been compared with results obtained with infrared spectroscopy^[2] and Schottky-barrier tunneling^[3] in an attempt to resolve an ambiguity between the intensities of the 0→1 and 1→2 transitions measured by the latter two techniques. The results are shown in Table I. The neutron molecular spectroscopy (NMS) data agree with infrared data within the limit of experimental resolution. However, the shift in the 0→1 transition of $\approx 24 \text{ cm}^{-1}$ for the cold solid phase seen by NMS is not seen by the Schottky-barrier method. Also it is not understood how that method could detect the 1→2 transition with intensity equal to the 0→1 transition when the sample is at a temperature of 1.3°K. It was concluded that not enough was known about the sample environment in the Schottky-barrier tunneling experiment to properly evaluate the technique in the light of the neutron data.

The ethyl chloride work has been submitted for publication to the Journal of Chemical Physics.

- [1] Dept. of Chemistry, University of Utah.
- [2] W. G. Fateley and F. A. Miller, "Torsional Frequencies in the Far Infrared, III. The Form of the Potential Curve for Hindered Internal Rotation of a Methyl Group", Spectrochim. Acta, **19** (1963) pp 611-628.
- [3] W. A. Thompson, "Molecular Spectroscopy Using Schottky-Barrier Tunneling", Phys. Rev. Letters, **20** n 20 (May 1968) pp 1085-'086.
- [4] G. Allen and H. J. Bernstein, "Intensity in the Raman Effect. II. Standard Raman Intensities and Vibrational Assignments in the Chloroethanes", Can. J. Chem., **32** (1954) pp 1124-1141.

TABLE I

COMPARISON OF NEUTRON DATA FOR TORSIONAL MODE WITH INFRARED
SPECTROSCOPY AND SCHOTTKY-BARRIER TUNNELING

ETHYL CHLORIDE $\text{CH}_3\text{CH}_2\text{Cl}$			METHOD		
			Neutrons	Schottky-barrier Tunneling	Infrared
Phase	Mode	Excitation	$\Delta E, \text{cm}^{-1}$	$\Delta E, \text{cm}^{-1}$	$\Delta E, \text{cm}^{-1}$
Gas	C-C Tors.	0 $\rightarrow$ 1			251.5
		1 $\rightarrow$ 2			235.5
	C-C-Cl Def.				336
Liquid	C-C Tors.		245 $\pm$ 4		
	C-C-Cl Def.		331 $\pm$ 4		335 ^[a]
Solid	C-C Tors.	0 $\rightarrow$ 1	275 $\pm$ 2	250 ^[b]	
		1 $\rightarrow$ 2		235 ^[b]	
	C-C-Cl Def.		331 $\pm$ 2	329 ^[c] , 373 ^[c]	

[a] Raman data from Reference 4.

[b] Values not quoted in text of article (Reference 4) but taken from curve of data.

[c] Both attributed to bending modes, but there is nothing in the ethyl chloride frequency spectrum close to the latter value of 373 cm^{-1} .

Fig. 1 Reduced partial differential scattering cross sections versus neutron energy change for liquid ethyl chloride. The arrow points to the torsional libration mode while the hash mark indicates a skeletal deformation mode.

Fig. 2 Reduced partial differential scattering cross sections versus neutron energy change for solid ethyl chloride. The arrow points to the torsional libration mode while the hash mark indicates a skeletal deformation mode.

NEUTRON MOLECULAR SPECTROSCOPY OF MISCELLANEOUS MOLECULAR SYSTEMS

R. J. Pugmire^[1], K. A. Strong, R. M. Brugger, D. M. Grant^[1],
R. Livingston^[1]

In addition to the other organic species reported separately, a number of other molecular species are currently under study and at various stages of completion of analysis. These additional systems studied to date include:

Two Top Molecules

The propane analogs dimethyl ether (gas, liquid, and solid), dimethyl sulfide (liquid and solid), dimethyl amene (gas, liquid, and solid) and isobutylene have been studied by means of NMS, and the data are presently being analyzed. The theoretical considerations developed for propane can be appropriately applied to these molecular species.

Methyl Benzenes

Ortho-xylene (1-, 2-dimethyl benzene) and durene (1-, 2-, 4-, 5-tetramethyl benzene) have been studied at 40, 50, and 60 meV incident energy during the past year in an effort to identify the low frequency ring bending modes as well as the methyl torsional vibrations. Analysis of the data indicates that the infrared data contain some anomalies with respect to ring bending vibrational assignments made by means of combination bands. The analysis of the methyl torsional modes are analogous to the simple two-top problem and should provide three- and six-fold barriers for hindered methyl rotation in aromatic compounds.

N-Butane

Data have been taken on liquid and two solid phases of n-butane. Three torsional (two CH₃ and one CH₂-CH₂) modes have been observed and, following acquisition of the gas data, a theoretical analysis will be made.

[1] Dept. of Chemistry, University of Utah.

CRYSTAL VIBRATIONS OF CNBr

H. L. McMurry, J. B. Bates^[1]

In anticipation of Raman and IR measurements on the molecular crystal CNBr, the dispersion relations for CNBr were calculated. Although the intermolecular force interactions are not known and crude guesses were used for these force constants, the calculations do provide a guide for interpreting the spectroscopic data. They also show the importance of obtaining dispersion relations and compressibility data in order to better define the intermolecular force interactions.

Calculations were done with and without including Coulomb interactions between charges using a rigid ion approximation. The frequencies were not much different and this suggests that Coulombic interactions are not of large significance in the intermolecular force field. Some pertinent details about the calculations are given below.

Crystal structure

This is Pmmn ($D_{2h}^{(13)}$) with two molecules per unit cell. Cell vectors have the magnitude $a = 6.02 \text{ \AA}$, $b = 4.12 \text{ \AA}$, $c = 5.80 \text{ \AA}$. The atom positions and numberings are:

	<u>x(a)</u>	<u>y(b)</u>	<u>z(c)</u>	<u>Estimated Charge Z e </u>
Br(1)	a/4	b/4	0.145 c	+0.24
C (2)	a/4	b/4	0.450 c	-0.09
N (3)	a/4	b/4	0.650 c	-0.15
Br(4)	3/4a	3/4a	0.856 c	0.24
C (5)	3/4a	3/4a	0.550 c	-0.09
N (6)	3/4a	3/4a	0.350 c	-0.15

The force constants used in the calculations are defined in Table I.

In this table subscripts I and II refer to the molecules in a primitive cell. The symbol R denotes rotation with an x or y subscript telling the axis of rotation. Symbols X, Y, and Z denote translations. The forms of the modes were determined from the calculated eigenvectors.

The calculated dispersion in the lowest frequency modes along $[00\xi]$ is shown in the next table. The near agreement in the optic and acoustic branches at $[0 \ 0 \ 1]$ arises because the coupling between the bromine and carbon atoms in adjacent collinear molecules dominates over the interchain couplings in determining this frequency. When ξ is at $[0 \ 0 \ 1]$ the molecules in adjacent cells along the c axis move in opposite phase.

Measurements of dispersion relations along $[00\xi]$ and $[\xi 00]$ would be most helpful for determining the along-chain and cross-chain inter-molecular interactions. Compressibilities along the c axis and perpendicular to this axis would also be helpful.

The calculations were made before the Raman data had been measured. Table IV lists the low frequency Raman and IR frequencies and compares with plausible calculated values for these modes.

The force constants for the intermolecular interactions are guesses. For this reason, it is not possible to make strong statements about how to associate the observed frequencies with normal modes. However, the results do suggest that the modes between 60 cm^{-1} and 108 cm^{-1} may arise from coupling of rotational motions of the molecules, while the single mode at 39.5 cm^{-1} involves interaction between molecular translations. However, there is considerable mixing of rotational and translational motions in the B_{2g} and B_{3g} normal modes.

[1] Chemistry Dept., University of Maryland (now at ORNL).

TABLE I
FORCE CONSTANT FOR CNBr

<u>Symbol</u>	<u>Description</u>	<u>Value^o (millidynes/A)</u>
$F_{2,3}$	Intramolecular C-N stretch	17.71
$F_{1,2}$	Intramolecular C-Br stretch	4.00
$F_{1',3}$	Intermolecular N-Br stretch between collinear molecules	0.50
$f_{12,34}$	Intermolecular C-N, C-Br stretch-stretch interaction	0.42
$f_{1'3,23}$	Intramolecular C-N, C-Br stretch-stretch interaction	0.01
$\Delta\phi_{1,2,3}$	Intramolecular bending of Br-C-N angle in the z,x and z,y planes (taken to be the same)	0.20
$\Delta\phi_{1',3,2}$	Br-N-C angle in either the z,x or z,y plane	0.05
$F_{3,4}$	Intramolecular N-Br stretch between atoms on non-collinear molecules	0.015
$F_{3,5}$	Intramolecular N-C stretch between atoms on non-collinear molecules	0.025
$F_{2,5}$	Intramolecular C-C stretch between atoms on non-collinear molecules	0.05

TABLE II

CALCULATED FREQUENCIES AT $\kappa = 0$ (cm^{-1})

Without Coulomb Interactions	With Coulomb Interactions	Description
A_{1g} Modes (Raman)		
2207.7	2209.6	C-N stretch intramolecular
615.4	612.9	Br-(CN) stretch intramolecular
26.2	24.9	$Z_I - Z_{II}$ intermolecular. Molecules within a cell translate oppositely along c axis
B_{1u} Modes (IR)		
2207.7	2209.5	C-N stretch intramolecular
615.0	614.2	Br-(CN) stretch intramolecular
B_{2g} Modes (Raman)		
359.5	355.2	Br-C-N angle in x,z plane. Intramolecular
177.4	179.5	$[R_{yI} - R_{yII}] + [X_I - X_{II}]$ Intermolecular. Ro-
41.7	46.8	$[X_I - X_{II}] - [R_{yI} - R_{yII}]$ tations dominate in high frequency mode.
B_{3u} Modes (IR)		
341.5	335.9	Br-C-N angle in x,z plane. Intramolecular
91.0	92.5	$R_{yI} + R_{yII}$ intramolecular
B_{3g} Modes (Raman)		
343.4	338.1	Br-C-N angle in y,z plane. Intramolecular.
133.3	135.0	$[R_{xI} + R_{xII}] - [Y_I - Y_{II}]$ Intermolecular. Ro-
37.9	39.9	$[Y_I - Y_{II}] + [R_{xI} + R_{xII}]$ tations dominate in high frequency mode.
B_{2u} (IR)		
336.2	330.4	Br-C-N angle in y,z plane. Intramolecular
80.7	83.5	$R_{xI} - R_{xII}$ intermolecular

TABLE III

DISPERSION OF LOW FREQUENCY MODES ALONG $[00\xi]$

k	Optic		Acoustic	
	<u>No Coulomb</u>	<u>Coulomb</u>	<u>No Coulomb</u>	<u>Coulomb</u>
	(cm^{-1})	(cm^{-1})	(cm^{-1})	(cm^{-1})
0	26.2	24.9	0.0	0.0
[0 0 .1]	58.0		51.9	
[0 0 .2]	102.9		99.7	
[0 0 .3]	141.3		139.1	
[0 0 .4]	167.3		165.5	
[0 0 .5]	176.5	179.1	174.8	178.4

TABLE IV

OBSERVED SPECTROSCOPIC FREQUENCIES (cm^{-1})

Raman (g mode)		
<u>Observed</u>	<u>Calculated</u>	
373.0	359.5	B_{2g} ; Br-C-N angle bend in x,z plane
366.5	343.4	B_{3g} ; Br-C-N angle bend in y,z plane
108.0		Thought to be an A_{1g} mode. No calculated frequency is near this.
86.0		The calculated frequencies for molecular rotational motions come nearest to these.
62.0		
39.5		
		This is closest to the frequencies calculated for the molecular translation modes.
IR modes (u modes)		
114	83.5	Possibly interaction of molecular rotations in the x,z plane.
93	91.0	Possibly interaction of molecular rotations in the x,z plane.

COUPLING OF CH₃ TORSIONAL MOTIONS TO INTERNAL
MOTIONS IN HYDROCARBONS

H. L. McMurry

The determination of barrier heights inhibiting torsional rotation of the CH₃ groups in propane and neopentane uses the "rigid frame" approximation for the torsional frequencies. This approximation assumes the molecule is rigid except for the possibility of torsional motions of the CH₃ groups about their symmetry axes. The work outlined below indicates that the errors arising from the rigid frame approximation are probably too small to affect seriously the barrier height analysis.

Coupling between the torsional motions and other degrees of freedom can arise through rotation-vibration type interactions and through the potential energy. This work investigated the interactions arising in the potential energy. The vibrations of propane and o-xylene were calculated in the rigid frame approximation and without making this approximation (which requires calculating all vibrations of the same symmetry species as are exhibited by the torsional motions).

Propane and o-xylene both have C_{2v} symmetry. The CH₃ torsional motions provide symmetry coordinates belonging to species A₂ (anti-symmetric for reflection in both symmetry planes) and B₂ (anti-symmetric for 180 degree rotation about the symmetry axis and for reflection in the plane of the molecular skeleton). Table I compares the frequencies for these modes as calculated in the rigid frame approximation, and without any approximation other than those needed in a harmonic analysis.

The interactions cause negligible changes in the A₂ frequencies. The small changes in the B₂ modes have been assumed to be unimportant for determining the barrier heights.

TABLE I
CALCULATION OF TORSIONAL FREQUENCIES

	<u>A₂ Rigid</u>	<u>A₂ General</u>	<u>B₂ Rigid</u>	<u>B₂ General</u>
Propane	218.8 cm ⁻¹	217.6 cm ⁻¹	267.9 cm ⁻¹	258.2 cm ⁻¹
O-xylene	183.3 cm ⁻¹	182.7 cm ⁻¹	186.0 cm ⁻¹	173.4 cm ⁻¹

COORDINATES FOR GROUP ROTATIONS IN MOLECULES AND CRYSTALS

H. L. McMurphy

The group vibration method^[1-3] for calculating the vibrations of large molecules or molecular crystals expresses the motion of any atom as the sum of three parts. The first gives its displacement within some local group (intra group motion) of which it is a part (like CH₃, CH₂, etc). The second is the displacement due to a pure translation of this group; the third is the displacement due to a pure group rotation. The transformation expressing the intra group and pure translational motions in terms of general Cartesian displacements are usually not hard to obtain. The transformation expressing the group rotations is more laborious to find.

This work presents a systematic way of finding the transformation for the rigid rotations. Application to propane, isobutane, o-xylene, and the molecular crystal CNBr have reduced considerably the work of setting up vibrational calculations on these systems.

The transformation is obtained from the expression for a general displacement pattern for atom v in group J . It is convenient to express any displacement as a three element column vector. The equation is

$$\Delta \tilde{r}_{vJ} = A_J [\Delta \tilde{r}_J + \Delta \tilde{\rho}_{vJ} + R_{vJ}^T \tilde{\theta}_J]. \quad (1)$$

Here

$\Delta \tilde{r}_{vJ}$ = the displacement of atom v in group J as observed from axes fixed in the molecule.

$\Delta \tilde{r}_J$ = displacement of the center of mass of group J . For convenience in later manipulations $\Delta \tilde{r}_J$ is expressed in terms of components parallel to the axis for group J .

$\Delta \tilde{\rho}_{vJ}$ = displacement of atom v in group J as observed from principal axes for group J with origin at the CM of the group. These axes translate and rotate with the group so an observer in this axis system sees no linear or angular momentum within the group.

$\tilde{\theta}_J$ = a vector containing as components small rotations of group J about each principal axis for the group.

In Equation (1), $\Delta \tilde{r}_{vJ}$, $\Delta \tilde{r}_J$, $\Delta \tilde{\rho}_{vJ}$, and $\tilde{\theta}_J$ are written as column vectors as follows:

$$\Delta \tilde{r}_{vJ} = \begin{bmatrix} \Delta X_{vJ} \\ \Delta Y_{vJ} \\ \Delta Z_{vJ} \end{bmatrix} ; \quad \Delta \tilde{r}_J = \begin{bmatrix} \Delta X_J \\ \Delta Y_J \\ \Delta Z_J \end{bmatrix} ; \quad \Delta \tilde{\rho}_{vJ} = \begin{bmatrix} \Delta x_{vJ} \\ \Delta y_{vJ} \\ \Delta z_{vJ} \end{bmatrix} ; \quad \tilde{\theta}_J = \begin{bmatrix} \Delta \theta_{xJ} \\ \Delta \theta_{yJ} \\ \Delta \theta_{zJ} \end{bmatrix} .$$

Then the displacement of atom v along the group axes is given by

$$\theta_{Jxr}^o_{vJ} = R_{vJ}^T \theta_J = \begin{bmatrix} 0 & z_{vJ} & -y_{vJ} \\ -z_{vJ} & 0 & x_{vJ} \\ y_{vJ} & -x_{vJ} & 0 \end{bmatrix} \begin{bmatrix} \Delta\theta_{xJ} \\ \Delta\theta_{yJ} \\ \Delta\theta_{zJ} \end{bmatrix}. \quad (2)$$

Here x_{vJ} , y_{vJ} , z_{vJ} are the equilibrium positions of atom v in group J . Finally, in Equation (1) A_J is the 3×3 matrix containing as elements the direction cosines of the group axes relative to the molecular axes.

$$A_J = \begin{bmatrix} \underline{I} \cdot \underline{i}_J & \underline{I} \cdot \underline{j}_J & \underline{I} \cdot \underline{k}_J \\ \underline{J} \cdot \underline{i}_J & \underline{J} \cdot \underline{j}_J & \underline{J} \cdot \underline{k}_J \\ \underline{K} \cdot \underline{i}_J & \underline{K} \cdot \underline{j}_J & \underline{K} \cdot \underline{k}_J \end{bmatrix} \quad (3)$$

Here $\underline{I}$, $\underline{J}$, and $\underline{K}$ are unit vectors along the molecular axes, and $\underline{i}_J$, $\underline{j}_J$, and $\underline{k}_J$ are unit vectors along principal axes for group J .

It is useful to employ mass adjusted coordinates $q_{vJ} = \sqrt{m_v} \Delta r_{vJ}$. The kinetic energy is then just a sum of squares

$$T = \frac{1}{2} \dot{q}^T \dot{q}; \quad q = \begin{bmatrix} q_i \\ \vdots \\ q_J \\ \vdots \end{bmatrix}; \quad q_J = \begin{bmatrix} q_{iJ} \\ \vdots \\ q_{vJ} \\ \vdots \end{bmatrix}.$$

In terms of mass adjusted coordinate Equation (1) becomes

$$q_{vJ} = A_J \left[\left(\frac{m_v}{M_J} \right)^{1/2} Q_J + q_{vJ} + \sqrt{m_v} R_{vJ}^T \theta_J \right] \quad (4)$$

where

$$Q_J = \sqrt{M_J} \Delta r_J$$

$$q_{vJ} = \sqrt{m_v} \Delta r_{vJ}$$

$$M_J = \sum m_{vJ} = \text{mass of all atoms in group } J.$$

If Equation (4) is multiplied from the left by $\sqrt{m_v} R_{vJ} A_J^T$ and then summed over v the result is

$$\sum_v \sqrt{m_v} R_{vJ} A_J^T q_{vJ} = \underline{I} \cdot \underline{\theta}_J. \quad (5)$$

The term involving Q_J drops out because the group axes are at the center of mass of the group. Those involving Δr_{vJ} drop out because these intra group displacements conserve angular momentum within the group.

In Equation (5), $\tilde{I}_J$ is a 3 x 3 diagonal matrix with the principal moments of inertia for group J as elements. The kinetic energy expression is simpler if $\tilde{I}_J^{1/2} \tilde{\theta}_J$ appears on the right side of Equation (5). Here $\tilde{I}_J^{1/2}$ is like $\tilde{I}_J$ except for using the $I_{J\alpha}^{1/2}$ ($\alpha = x, y, z$) as the diagonal elements. This leads to

$$[\tilde{I}_J^{-1/2}] \sum_v \sqrt{m_{vJ}} R_{vJ} A_{vJ}^T q_{vJ} = [\tilde{I}_J^{1/2}] \tilde{\theta}_J = \tilde{\Omega}_J \quad (6)$$

or

$$[\gamma_{1J} \cdots \gamma_{vJ} \cdots] q_{vJ} = \gamma_J q_J = \tilde{\Omega}_J$$

where

$$\gamma_{vJ} = \left[\tilde{I}_J^{-1/2} \right] \sqrt{m_{vJ}} R_{vJ} A_{vJ}^T; \quad q_J = \begin{bmatrix} q_{1J} \\ \vdots \\ q_{2J} \\ \vdots \\ q_{vJ} \end{bmatrix}.$$

The $\tilde{\Omega}_J$ is a three element column vector with "moment of inertia" adjusted elements $\tilde{\Omega}_{J\alpha}$ ($\alpha = x, y, z$). Each $\tilde{\Omega}_{J\alpha}$ corresponds to a rigid rotation of group J about its α principal axis. When symmetry exists projection operator methods can be used to obtain symmetry coordinates from any $\tilde{\Omega}_{J\alpha}$. The γ_{vJ} is almost the same as the B_{vJ} defined in Reference 3. It differs in not using the normalization constant which appears in B_{vJ} and because the matrix A_J^T appears as a right hand multiplier.

The procedure described in Reference 3 uses a transformation,

$$\tilde{S} = \begin{bmatrix} A_q \\ A_T \\ A_R \end{bmatrix} q.$$

The rows in A_q define the coordinates associated with motions within groups. The rows in A_T are obtained from group translations. The rows in A_R define coordinates associated with rigid group rotations. These are conveniently expressed using the rows in the γ_J , or suitable combinations when symmetry exists. The general method is like that described in some early work on C_3H_8 [4], except that the present method of writing the rotation coordinates appears to have advantages.

An existing computer program has been modified to calculate the γ_J [5].

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COMPUTER PROGRAMS FOR USE IN CALCULATING MOLECULAR
AND CRYSTAL VIBRATIONS -- INCLUSION OF LONG-RANGE COULOMB
INTERACTIONS IN CRYSTALS

J. K. Boyter^[1], H. L. McMurry, A. W. Solbrig, Jr., Richard Martin^[2]

A series defect of our earlier computer programs^[3,4] for calculating crystal vibrations was the limitation to short-range force interactions. This defect has been removed by modifying an earlier program to allow inclusion of long-range Coulomb interactions^[4]. The new program also permits calculating Jacobian matrices whose elements are the changes $\partial v_i / \partial f_k$ of frequencies with force constants. The change in frequency for a change in the square of the charge on one of the ions is also calculated. These Jacobians are used in determining how to alter force constants so the calculated frequencies provide a least squares fit to experimental data^[5]. The pertinent theory for the program and the necessary input information are described in report IN-1280^[6].

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A PROGRAM FOR SETTING UP VALENCE FORCE MATRICES FOR MOLECULES AND CRYSTALS

J. K. Boyter^[1], H. L. McMurry

The potential function for a molecule expressed in terms of valence force coordinates (changes in bond distances, interbond angles, etc) has the form

$$V = \frac{1}{2} \tilde{H}^T \tilde{F} \tilde{H} . \quad (1)$$

In Equation (1), $\tilde{H}$ is a column vector containing the valence coordinates and $\tilde{F}$ is a square matrix which contains the primary (diagonal) and interaction (off diagonal) force constants. The analogous equation for crystals contain force matrices $\tilde{F}_{0\ell}$ which tell how the coordinates in the origin cell (denoted by 0) interact with coordinates associated with neighboring cells (indexed by ℓ)^[2].

The construction of the $\tilde{F}$ matrices for a molecule is not too difficult. For a crystal, it is a tedious process and it is easy to make erroneous insertions or omissions of elements in the $\tilde{F}_{0\ell}$. Therefore, a computer program was written which uses a minimum of input data (cell indices, atom positions in the cells, etc)^[3]. It constructs the $\tilde{F}_{0\ell}$ matrices and punches cards suited for use in MACS C, or in program 40*0206 (used only for molecules).

This program is now working and has greatly reduced the labor of setting up valence force matrices for UO_2 . It may be almost indispensable for treating crystals with larger numbers of atoms per primitive cell.

[1] Summer employee, now at Biology Dept., University of Colorado.

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APPLICATION OF GROUP THEORY TO THE LATTICE DYNAMICS OF MAGNESIUM STANNIDE^[1]

R. J. Kearney^[2], T. G. Worlton^[3], R. E. Schmunk

Group theoretical techniques have been applied to an analysis of the lattice dynamics of Mg_2Sn along directions of high symmetry and at certain points in the Brillouin zone. Matrix representations have been constructed for the symmetry elements in the group of the wave vector. These representations have been used to:

- (1) Determine the classification of the phonon modes for a given $\vec{q}$
- (2) Block diagonalize the dynamical matrix $\underline{D}$
- (3) Construct projection operators to determine the form of the eigenvectors for the atomic motions.

In addition, a comparison is made of the methods of Chen and of Maradudin and Vosko[4]. The results obtained are generally applicable to other fluorite structure crystals as well as Mg_2Sn .

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 - [2] Physics Dept., University of Idaho.
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VALENCE FORCE POTENTIALS FOR CALCULATING CRYSTAL VIBRATIONS IN SILICON

A. W. Solbrig, Jr., H. L. McMurtry

This work continues an effort summarized in Reference 1 aimed at using a valence force potential to fit the dispersion relations of silicon.

The theory of directed valence should be applicable to silicon and this implies that the forces resisting changes in bonds between near neighbors will be particularly strong, as will be forces resisting changes of the angles between these bonds. Furthermore, the alteration of one bond distance or a bond angle should disturb the electron distributions in neighboring bonds, and this leads naturally to the expectation of interaction force constants.

These inferences are supported by the excellent agreement between calculated and observed dispersion relations for diamond[2]. Only six valence force constants are required. They were obtained from an initial set which has been used to calculate vibration frequencies in n-paraffins.

In the earlier work on silicon[1], the six diamond force constants were "scaled" to obtain the "initial set" for silicon. These constants were then adjusted by a least squares fitting procedure. However, with only six constants it was not possible to get close agreement with experimental data; hence more constants are used in the work described here. In addition, minor changes have been made in some frequencies to be fitted. For example, a slightly shifted but much more accurate value of the Raman optical intercept[3] has been used. It was necessary to increase the number of points to be fitted by the least squares procedure. To the 13 points used in Reference 1, 12 new points were added: 5 points along Δ (LA and TA at $q/q_{\text{max}} = 0.4$ and all 3 points at $q/q_{\text{max}} = 1$), 6 points along Λ (LA and TA at $q/q_{\text{max}} = 0.4$ and all 4 points at q/q_{max}

= 1), and 1 point along Σ (the acoustic I point at $q/q_{\max} = 3/4$). An attempt was made to improve the fit of Reference 1 by inclusion of (a) more valence-force interaction constants and/or (b) a one-parameter electrostatic interaction based on the "simple bond charge" model[4].

The simple bond charge contribution was not useful in improving the fit. The results (as well as unpublished results of R. M. Martin) indicate that there may be too much duplication, or similarity, between the contribution to the potential energy from the valence forces and the contribution from the simple bond charge.

The best fit to data by a valence force field is depicted in Figures 1, 2, and 3. This fit is based on the "Nine Constant Set" listed in Table I. The numbers adjoined to some of the experimental points are values of

$$\left| \frac{\text{calculated energy} - \text{experimental energy}}{\text{experimental error}} \right|$$

where experimental results are in the form "experimental energy $\pm$ experimental error". This number is not given if its value is less than unity. The major remaining discrepancies are on the transverse acoustic branches along $\Delta[100]$ and $\Lambda[111]$. It may be that there exists a set of ten or fewer valence force constants which yields a completely satisfactory fit to the dispersion relations for silicon, but we have not yet found such a set. A valence force field containing nine (possibly fewer) force constants does yield an almost satisfactory fit. The effects not included in this valence force field are apparently small; these small effects may involve electrostatic interactions, omitted valence force constants, or other mechanisms[5].

Tables II and III list some characteristics of 14 of the multitude of other valence force constants which might be used in the diamond structure. Of these 14, only the first 11 were actually tried in our calculations. The 14 constants contain three sets of redundant force constants: (1) $F_{\bar{\tau}}, f_{\bar{\tau}\bar{\tau}}, f_{\bar{\tau}\bar{\tau}}^{(2)}$; (2) $f_{r\bar{\tau}}, f_{r\bar{\tau}}^{(2)}$; (3) $F_r, F_{\bar{\tau}}, f_{rr}, f_{r\bar{\tau}}, F_R$. We believe that there are no other redundant sets in the tabulated constants. Redundant sets (1) and (2) have the properties

$$\frac{\partial \omega_i^2(\underline{\kappa})}{\partial F_{\bar{\tau}}} + \frac{\partial \omega_i^2(\underline{\kappa})}{\partial f_{\bar{\tau}\bar{\tau}}} + \frac{\partial \omega_i^2(\underline{\kappa})}{\partial f_{\bar{\tau}\bar{\tau}}^{(2)}} = 0$$

and

$$\frac{\partial \omega_i^2(\underline{\kappa})}{\partial f_{r\bar{\tau}}} + \frac{\partial \omega_i^2(\underline{\kappa})}{\partial f_{r\bar{\tau}}^{(2)}} = 0$$

for all branches i and all wave vectors $\underline{\kappa}$. (The ω_i^2 are eigenvalues which are proportional to energy squared.) A similar sort of relation holds among the partial derivatives with respect to the force constants of redundant set (3),

but we have not calculated the coefficients of the partial derivatives. Once the redundancy of sets (1) and (2) was verified, no further use was made of constants $f_{\Phi\Phi}^{(2)}$ or $f_{r\Phi}^{(2)}$ in the fitting. The constant F_R (for the change in second neighbor distance) has not been inserted in our force constant matrices, and because of the redundancy there is no reason ever to insert F_R . The dihedral angle constants F_τ and $F_\tau^{(1)}$ appear to be worth trying in further work^[6]. To date, our fitting of silicon has made use of selected subsets of the following 18 valence force constants: The nine constants of Table I plus the nine constants $f_{rr}^{(1)}$, $f_{rr}^{(2)}$, $f_{rr}^{(3)}$, $f_{r\Phi}^{(1)}$, $f_{r\Phi}^{(3)}$, $f_{\Phi\Phi}^{(1)}$, $f_{\Phi\Phi}^{(3)}$, $f_{\Phi\Phi}^{(6)}$ and $f_{\Phi\Phi}^{(8)}$ from Tables II and III. The procedure of searching for a fit was roughly as follows: (a) Evaluate a Jacobian matrix with 25 eigenvalue columns and 18 force-constant rows at the "Third Iteration Set" of Table I. (b) Run a single 25×6 "Adjuster"^[7] problem using only columns in the Jacobian corresponding to the first six constants in Table I. (c) Run $\binom{12}{1} = 12$ of 25×7 "Adjuster" problems using the first six columns plus all possible single columns from the remaining 12 columns. (d) Run $\binom{12}{2} = 66$ of 25×8 "Adjuster" problems using the first six columns plus all possible sets of two columns from the remaining 12 columns. (e) Run $\binom{12}{3} = 220$ "Adjuster" problems using the first six columns plus all possible sets of three columns from the remaining 12 columns. (On the IBM 360/75, "Adjuster" problems run at a rate of about 100/minute.) (f) Examine the predicted values of the weighted sum of squared differences between observed and calculated eigenvalues. (g) Try promising sets of force constants by substitution in and solution of the secular equation. (h) Further improve useful sets of force constants by employing "Adjuster" and iteration. Often the matrix inverted by "Adjuster" was "ill-conditioned", with large predicted changes in force constants and large disagreement between predicted eigenvalues and eigenvalues calculated by solution of the secular equation. In some of these cases some use was made of the "scaling" or "fractional step" technique^[8], but convergence appeared to be very slow. It should be worthwhile to apply the "damped least squares" method, for it is reported to be more rapidly convergent^[8,9]. Attempts to use more than nine constants in the fit have not yet been successful, mainly because of (a) problems with "ill-conditioning" and (b) failure of the linearity assumption of "Adjuster" as revealed by lack of agreement between predicted eigenvalues and eigenvalues calculated by solution of the secular equation.

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TABLE I

VALENCE FORCE CONSTANTS FOR SILICON^[a]

Force Constant				Neighbor Order ^[b]	Initial Set	Third Iteration Set	Nine Constant Set
F_r	$(\Delta r)^2$;	Δr = change in nearest neighbor distance		1	0.65471	1.4850	1.47543
F_ϕ	$r_o^2(\Delta\phi)^2$;	$\Delta\phi$ = change in angle		1,2	0.06054	0.02944	0.02984
f_{rr}	$\Delta r \Delta r'$;	r and r' have atom in common		1,2	0.02320	0.02697	0.02305
$f_{r\phi}$	$r_o \Delta\phi \Delta r$;	r is along a leg of ϕ		1,2	0.04737	0.03997	0.04050
$f_{\phi\phi}$	$r_o^2 \Delta\phi \Delta\phi'$;	ϕ and ϕ' have common leg and apex		1,2	-0.00135	-0.00358	0.00306
$f_{\phi\phi}^*$	$r_o^2 \Delta\phi \Delta\phi'$;	all legs coplanar		1,2,5	0.01513	0.02922	0.03564
$f_{\phi\phi}^{(4)}$	$r_o^2 \Delta\phi \Delta\phi'$;	legs 1,2,3 coplanar leg 4 out of plane		1,2,5,6	--	--	0.00423
$f_{\phi\phi}^{(5)}$	$r_o^2 \Delta\phi \Delta\phi'$;	plane of legs 1,2 $\perp$ plane of legs 3,4		1,2,3,4	--	--	0.00717
$f_{\phi\phi}^{(7)}$	$r_o^2 \Delta\phi \Delta\phi'$;	plane of legs 1,2 $\perp$ plane of legs 3,4		1,2,3	--	--	-0.00401

[a] All units are millidynes/ $\text{\AA}$.

[b] The first-order neighbor is the nearest neighbor, second-order is next nearest, etc.

TABLE II

VALENCE FORCE CONSTANTS FOR DIAMOND STRUCTURE^[a]

$f_{rr}^{(1)}$	$\Delta r \Delta r'$	; all legs coplanar
$f_{rr}^{(2)}$	$\Delta r \Delta r'$	; leg 3 out of plane
$f_{rr}^{(3)}$	$\Delta r \Delta r'$	; legs 1 and 4 coplanar
$f_{r\phi}^{(1)}$	$r_o \Delta r \Delta \phi$	; all legs coplanar
$f_{r\phi}^{(2)}$	$r_o \Delta r \Delta \phi$	; r and ϕ have apex only in common	
$f_{r\phi}^{(3)}$	$r_o \Delta r \Delta \phi$	; leg 3 out of plane
$f_{\phi\phi}^{(1)}$	$r_o^2 \Delta \phi \Delta \phi'$	; all legs coplanar
$f_{\phi\phi}^{(2)}$	$r_o^2 \Delta \phi \Delta \phi'$	; ϕ and ϕ' have apex atom only in common	
$f_{\phi\phi}^{(3)}$	$r_o^2 \Delta \phi \Delta \phi'$	; leg 3 out of plane
$f_{\phi\phi}^{(6)}$	$r_o^2 \Delta \phi \Delta \phi'$	; legs 1 and 4 coplanar
$f_{\phi\phi}^{(8)}$	$r_o^2 \Delta \phi \Delta \phi'$	; legs 2,3,4 coplanar, leg 1 out of plane
F_R	$(\Delta R)^2$	; ΔR = change in second neighbor distance
F_τ	$(r_o \Delta \tau)^2$	; $\Delta \tau$ = change in dihedral angle, all legs coplanar at equilibrium	} [b]
$F_\tau^{(1)}$	$(r_o \Delta \tau')^2$	; $\Delta \tau'$ = change in dihedral angle, one leg out of plane	

[a] These force constants are all zero in the sets listed in Table I. See Table III and accompanying text for further discussion.

[b] E. B. Wilson, Jr., J. C. Decius, and P. C. Cross, Molecular Vibrations, McGraw-Hill, New York, (1955) pp 60-61.

TABLE III
NEIGHBOR ORDER^[a] IN VALENCE FORCE CONSTANTS
FOR DIAMOND STRUCTURE

Force Constant	Neighbor Order	Force Constant	Neighbor Order
$f_{rr}^{(1)}$	1,2,5	$f_{\phi\phi}^{(2)}[b]$	1,2
$f_{rr}^{(2)}$	1,2,3	$f_{\phi\phi}^{(3)}$	1,2,3
$f_{rr}^{(3)}$	1,2,3	$f_{\phi\phi}^{(6)}$	1,2,3
$f_{r\phi}^{(1)}$	1,2,5	$f_{\phi\phi}^{(8)}$	1,2,3,5
$f_{r\phi}^{(2)}[b]$	1,2	$F_R[b]$	2
$f_{r\phi}^{(3)}$	1,2,3	F_τ	1,2,5
$f_{\phi\phi}^{(1)}$	1,2,5,8	$F_\tau^{(1)}$	1,2,3

[a] The first-order neighbor is the nearest neighbor, second-order neighbor is next nearest, etc.

[b] These constants belong to sets of redundant constants. See accompanying text for further discussion.

Fig. 1. Dispersion relations for silicon along the [111] direction. (See accompanying text.)

Fig. 2. Dispersion relations for silicon along the [100] direction. (See accompanying text.)

Fig. 3. Dispersion relations for silicon along the [110] direction. (See accompanying text.)

VALENCE FORCE MODEL WITH A "CELL DIPOLE" APPROXIMATION FOR LONG-RANGE FORCES

A. W. Solbrig, Jr., W. J. Suitt

This work is an attempt to modify the valence-force model^[1] to include long-range electrostatic forces from ionic charges, bond charges, and electronic polarizability in a "cell dipole" approximation. (A modification of the valence-force model to include long-range electrostatic forces from ionic charges in the "rigid ion" approximation is described in Reference 2.) To the valence-force potential energy we have added a long-range term

$$2V_L = \sum_{ll'} [\underline{p}(l) + \underline{G}(l)]^T \underline{T}_{ll'} [\underline{p}(l') + \underline{G}(l')] + \frac{1}{\alpha} \sum_l \underline{G}^T(l) \underline{G}(l)$$

where

$$\underline{T}_{ll'} = \frac{\underline{I} - 3\hat{r}_{ll'}\hat{r}_{ll'}}{r_{ll'}^3} \text{ is a } 3 \times 3 \text{ matrix}$$

$$\underline{T}_{ll} = 0, \quad \underline{I} \text{ is the } 3 \times 3 \text{ identity matrix}$$

$$\tilde{r}_{\ell\ell'} = \tilde{r}_{\ell} - \tilde{r}_{\ell'}, \quad \hat{\tilde{r}}_{\ell\ell'} = \frac{\tilde{r}_{\ell\ell'}}{r_{\ell\ell'}}$$

$$\mathbf{p}(\ell) = \sum_j \mathbf{K}_{\ell j} \mathbf{q}_j$$

and

$$\mathbf{G}(\ell) = \alpha \mathbf{E}_{\ell} = -\alpha \sum_{\ell'} \mathbf{T}_{\ell\ell'} [\mathbf{p}(\ell') + \mathbf{G}(\ell')].$$

The $\mathbf{p}(\ell)$ is a vector "cell dipole" coordinate which depends on the positions of ionic and bond charges in cell ℓ . Only a small number of $\mathbf{K}_{\ell j}$ appear in the sum which defines $\mathbf{p}(\ell)$ in terms of the mass-adjusted Cartesian displacement coordinates $\mathbf{q}_j$. The $\mathbf{K}_{\ell j}$ have the same properties as the $\mathbf{D}_{\ell j}$ of Reference 1. The α is a "cell electronic polarizability", and the term involving $1/\alpha$ is the energy required to create the "cell electronic dipole" $\mathbf{G}(\ell)$. The above long-range potential energy can be treated in a fairly straightforward fashion by the Fourier expansion technique of lattice dynamics. Inversion of a real, symmetric 3×3 matrix and use of a "Dipole Sum" program [3] are required for evaluation of the long-range contribution to the dynamical matrix. The program MACS J [4] has been modified to include the above long-range contribution. The modified program is being checked on the IBM 360/75 by application to BeO [5]. Numerical results display the expected effects of the long-range term [5] as the wave vector approaches zero along or perpendicular to the c -axis, but they display unexpected splitting of degeneracy for wave vector parallel to the c -axis and terminating on the Brillouin zone boundary. We hope to find the cause of this unexpected splitting and to obtain correct degeneracies [a]. In the diamond structure (diamond, Si, Ge, α -Sn) [6], the above long-range term appears to vanish. We hope to consider an analogous "cell quadrupole" term [7] for the diamond structure.

[a] Note added in proof: A modified form of the equations now yields correct degeneracies.

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 - [4] J. K. Boyter and A. W. Solbrig, Jr., MACS J -- A Program for Computing Crystal Phonon Dispersion Relations, Structure Factors for Neutron Inelastic Scattering, and Jacobian Matrices, IN-1147 (January 1968).

- [5] A. W. Solbrig, Jr., "Application of the Valence-Force Model to BeO: The Need for Long-Range Forces". G. L. Ostheller, R. E. Schmunk, R. M. Brugger, R. J. Kearney, "Lattice Dynamics of Beryllium Oxide". Both in R. L. Heath (ed.), Nuclear Technology Branch Yearly Progress Report for Period Ending June 30, 1968, IN-1218 (December 1968) pp 149-151.
- [6] A. W. Solbrig, Jr., "Valence Force Potentials for Calculating Crystal Vibrations in Silicon", this report.
- [7] M. Lax, "Comments on the Shell Model for Lattice Vibrations", in R. F. Wallis (ed.), Lattice Dynamics. Proceedings of the International Conference held at Copenhagen, Denmark, August 5-9, 1963, New York: Pergamon Press, Inc., 1965, pp 179-187.

LATTICE DYNAMICS OF SODIUM CHLORIDE AT ROOM TEMPERATURE ^[1]

R. E. Schmunk, D. R. Winder ^[2]

The phonon dispersion relation has been measured for sodium chloride at room temperature for waves propagating along the symmetry directions $[00\bar{1}]$, $[\bar{1}\bar{1}0]$, and $[\bar{1}\bar{1}\bar{1}]$ by slow neutron inelastic scattering using the Materials Testing Reactor three axis spectrometer. These data are in reasonable agreement with the low temperature (80°K) data obtained by Almqvist et al and with the X-ray scattering data of Buyers and Smith. The shell model has been fit to the present data using a nonlinear least squares routine with the 14 parameter version giving the best overall agreement when the fitting to the elastic constants is also included. In the text, comparison is made with a seven parameter rigid-ion model along with 11 and 14 parameter versions of the shell model. The 14 parameter shell model has been applied in calculating a frequency distribution from which the Debye temperature was evaluated for comparison with thermodynamic data, and the combined density-of-states distribution was computed and compared with second order Raman scattering data, both with good agreement. The data from this experiment and the shell model calculations are displayed in Figure 1.

[1] Accepted for publication in J. Phys. Chem. Solids.

[2] Physics Depart., Colorado State University.

Fig. 1 Phonon dispersion relation for sodium chloride along the symmetry directions indicated in the figure. The solid (dashed) curves represent a 14 (11) parameter shell model which was fit to the data.

LATTICE DYNAMICS OF MAGNESIUM STANNIDE AT ROOM TEMPERATURE^[1]

R. J. Kearney^[2], T. G. Worlton^[3], R. E. Schmunk

Neutron inelastic scattering has been used to determine the frequency-wave vector phonon dispersion relation of Mg_2Sn at room temperature along the $[00\xi]$, $[\xi\xi0]$, and $[\xi\xi\xi]$ symmetry directions. The frequency of the infrared active optical mode was found to be $5.56 \times 10^{12} \text{ sec}^{-1}$ in excellent agreement with infrared absorption measurements. Slopes of the acoustic modes were in good agreement with elastic constant data. Both rigid ion and shell models have been least squares fit to the experimentally measured frequencies. A satisfactory fit to the experimental data was obtained with 9 and 10 parameter shell models, but no satisfactory fit was found for the seven parameter rigid ion model. The best fitting model was used to calculate the vibration frequencies along the $[1\xi0]$ direction in addition to the directions for which measurements were made. Figure 1 shows the experimental data obtained for Mg_2Sn and in addition the results of fitting the 10 parameter shell model to the data.

[1] Submitted for publication in J. Phys. Chem. Solids.

[2] Physics Dept., University of Idaho

[3] Los Alamos Scientific Laboratory.

Fig. 1 Phonon dispersion relation of Mg_2Sn for the three symmetry directions $[00\xi]$, $[\xi\xi0]$, and $[\xi\xi\xi]$. Diamonds and round points indicate branches having transverse and longitudinal polarizations, respectively, for the Δ and Λ branches, whereas the Σ branches of this experiment do not have symmetry-determined polarizations. The curves in the figure are the result of a 10 parameter shell model which was least squares fit to the data.

LATTICE DYNAMICS OF THALLIUM

T. G. Worlton^[1], R. E. Schmunk, R. J. Kearney^[2]

Dispersion relation data have been obtained for the symmetry directions $[0001]$ and $[01\bar{1}0]$ in thallium at 300°K for comparison with data at 80°K which were obtained previously. The measurements on thallium are part of a study being made of the temperature dependence of the vibrations of hexagonal close packed (HCP) crystals which undergo a transformation from HCP to body centered cubic (BCC). This particular transformation takes place in thallium at 507°K. Data for 300 and 80°K are displayed, respectively, in Figures 1 and 2. Diamonds (circles) indicate modes having transverse (longitudinal) polarization and, along $[01\bar{1}0]$ the modes indicated by the open (closed) diamonds, having their polarization parallel (perpendicular) to the hexagonal plane. Temperature effects in the present data which are particularly noticeable include: (a) the lowering of the Δ_6 optical branch with increased temperature, (b) the lowering of the point Γ_5^+ which relates directly to a, and (c) the lowering of the point M_3^+ . The linearity of the transverse acoustic branches at small q is obscured by the poor quality of the data that were very difficult to obtain because of high backgrounds. The smooth curves which are displayed in the data figures are the results of least-squares fitting twelve parameter self-consistent version of the modified axially symmetric lattice dynamics model to the data at each temperature. Differences in the model parameters for the two temperatures do not show any significant trend.

Fig. 1 Dispersion relation for thallium at 300°K for the symmetry directions $[0001]$ and $[01\bar{1}0]$. The smooth curves in the figure are the result of least squares fitting the twelve parameter modified axially symmetric model to the data.

Resolution calculations are being made to improve our understanding of the TA data at low q , and a final report on the thallium studies is being prepared.

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[2] Professor of Physics, University of Idaho.

Fig. 2 Dispersion relation for thallium at 80°K for the symmetry directions [0001] and [0110]. The smooth curves in the figure are the result of least squares fitting the twelve parameter modified axially symmetric model to the data.

LATTICE DYNAMICS OF ZIRCONIUM

H. F. Bezdek^[1], R. E. Schmunk, L. X. Finegold^[2]

Additional phonon data have been obtained for zirconium for the transverse acoustic (TA) branches at low q using improved angular resolution in the experiment. These measurements were deemed necessary due to the large discrepancies between the initial slopes of the TA branches as determined by neutron scattering and by ultrasonics. The latest measurements indicate that the discrepancies mentioned above were partly, but not entirely, due to resolution effects. Our neutron scattering data predict higher initial slopes for the TA branches than given by ultrasonics; Cowley et al^[3] suggest that the difference may be due in part to the difference between zero and first sound, ie, ultrasonic measurements are made in a regime where the waves

propagate in a collision-dominated mode whereas the neutron scattering samples waves of higher frequencies which propagate in a collision-free mode.

A least-squares fitting routine has been applied in comparing the modified axially symmetric (MAS) lattice dynamics model[4] with the present data. Results of the fitting are displayed by the smooth curves in Figure 1 which are compared with the latest data.

In addition, the MAS model has been used to calculate a frequency distribution for zirconium which is shown in Figure 2. A final report on the room temperature measurements is now being written.

Fig. 1 Phonon dispersion relation for zirconium at room temperature along the symmetry directions [0001] and [0110] with longitudinal modes indicated by round points and transverse modes by diamonds. The modified axially symmetric lattice dynamics model (13 parameters) was least-squares fit to the data and is displayed as the curves in the figure.

- [1] Predoctoral student, Dept. of Physics, University of Colorado.
- [2] Professor of Physics, University of Colorado.
- [3] R. A. Cowley, W. J. L. Buyers, E. C. Svensson, G. L. Paul, "Neutron Scattering and Elastic Constants", in Neutron Inelastic Scattering. Proceedings of a Symposium on Neutron Inelastic Scattering Held by the International Atomic Energy Agency in Copenhagen, 20-25 May, 1968. Volume I, Vienna: International Atomic Energy Agency, 1968, pp 281-288.
- [4] R. E. De Wames, T. Walfram, G. W. Lehman, "Lattice Dynamics, Heat Capacities, and Debye-Waller Factors for Be and Zn Using a Modified Axially Symmetric Model", Phys. Rev., 138 (May 1965) pp A717-A728.

Fig. 2 Frequency distribution of zirconium which was calculated for the modified axially symmetric model using the extrapolation technique.

DISPERSION RELATIONS OF SMALL CRYSTALS USING A STATISTICAL CHOPPER

P. D. Randolph, R. E. Schmunk

The basic advantage of a statistical chopper for time-of-flight neutron experiments is its ability to distinguish sharp peaks in the presence of high background counting rates. A statistical chopper has a duty cycle of about 0.5 compared to ≤ 0.005 for conventional choppers. A chopper of this type has been constructed and is described in "Correlation Chopper for Neutron Time-of-Flight Experiments", of this report. It was used there to check out the operation of the chopper in a time-of-flight diffraction application. Such a chopper can also be used for inelastic scattering experiments, in particular for the measurement of the phonon dispersion relations of single crystals where the phonons show up as small peaks in the scattered neutron spectrum. Phonon experiments are usually done using a three-axis crystal spectrometer, which being a continuous beam device has a duty cycle of unity. Thus when crystals of sufficient size are available for use on a three-axis machine, the statistical chopper usually offers no advantages over the three-axis because of its lower duty cycle. An exception to this would be for samples which have a high absorption coefficient, such as cadmium. An additional possibility, for which the

statistical chopper may offer advantages, is that of measuring the phonon dispersion relations for very small crystals. In general the smaller the sample size, the smaller is the signal-to-background ratio. Thus for crystals of volume less than a few tenths of a cubic centimeter, the statistical chopper may be superior to the three-axis because of its ability to distinguish the phonon peaks in the presence of a large background. Examples of interest to reactor physics are UC, graphite, and other high melting point materials. In addition, most organic crystals, especially biological materials, can only be obtained as crystals in small sizes.

In order to test the feasibility of this approach, a preliminary experiment was performed with the statistical chopper to observe phonons from a small crystal of sodium chloride ($1/8$ in. $\times$ $1/8$ in. $\times$ $1/2$ in. = 0.13 cm³). The layout of the experiment is shown in Figure 1. Two of the three axes of the MTR triple axis spectrometer were used, one to mount the beryllium monochromator and the other to mount the NaCl sample crystal. This was set up inside the shielded room of the velocity selector. The choppers which normally are used with the velocity selector were removed, and the ones labeled A, B, C in the figure were replaced with stationary collimators. The statistical chopper was placed just upstream of the monochromator position. The chopper was run at 15,000 rpm, giving a channel width of 7.843 μ sec. The data were stored in the velocity selector analyzer as four quadrants of 255 channels each. The beryllium monochromator was set to give an incident beam on the sample of 55.68 meV using the (0002) reflection. As expected the count rate was quite low and data runs were 20 to 24 hours long for each phonon. Figure 2 shows a sample of the data for the transverse acoustic phonon having a wave vector $q = 0.4 q_{\text{max}}$ in the $[00\bar{c}]$ direction. Two peaks are seen in this figure;

INC-A-12697

Fig. 1. Plan view of the small crystal statistical chopper experiment.

the smaller one in channel 90 is due to diffraction from the cadmium sleeve used to mask the NaCl crystal to 1/2 inch in length. The larger peak is the phonon of interest. The inset of this figure shows a dispersion relation; the smooth curve being the results of three-axis measurements on a large sample [1] and the points, the three phonons observed in this experiment.

The results clearly indicate that the statistical chopper can be useful for small crystals. However, improvements can be made over the temporary setup used. In future experiments the chopper will be put near the sample rather than near the monochromator, and the monochromator shielding will be improved, reducing backgrounds.

- [1] R. E. Schmunk and D. R. Winder, "Lattice Dynamics of Sodium Chloride at Room Temperature", this report.

Fig. 2. Data obtained in the small crystal experiment after cross correlation with the chopper sequence. The large peak is a phonon at $q = 0.4 q_{\text{max}}$ and the smaller peak at the left is due to Bragg scattering from cadmium near the sample. The inset shows a comparison of this data with other measurements.

**E. TECHNIQUE DEVELOPMENT FOR PHYSICAL
AND BIOLOGICAL SCIENCES**

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PLUTONIUM DETECTION IN BONE SECTIONS

L. G. Miller, W. S. S. Jee^[1]

With the wide scale use of plutonium by industry and the AEC, it is becoming very important for medical people to know the location and concentrations of plutonium ingested into humans and animals. For high concentrations, autoradiographic techniques have been used very successfully. Film is layed adjacent to the thin bone sections, and the alphas from plutonium decay expose the film producing good resolution data on location and concentration. However, using this technique for very small concentrations (in the order of pico curies/cm²), years are required to obtain a satisfactory autoradiograph.

A technique initiated in England^[2] is further being developed at the MTR for detecting very minute amounts of plutonium in bone sections in a short time period. Very thin bone sections are attached to thin polycarbonate films and irradiated in a high neutron flux. The neutrons cause fission in the very small amounts of plutonium; the fission fragments enter the polycarbonate film. Using tract etch techniques, the fission tracks become visible in the film. With this system, position and concentrations of pico curies/cm² amounts of plutonium are being measured with high precision on thin bone sections in a few hours. The neutron irradiations are being made in the MTR thermal column because its high neutron flux permit these short time irradiations. In addition, the low gamma heating in this position is a necessity; high gamma heating anneals the polycarbonate film with subsequent loss of the fission tracks.

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[2] (Private reference), Betty Bleaney, Headington, Oxford, England.

BIOMEDICAL RESEARCH IN THE 25 keV BEAM

L. G. Miller, N. A. Frigerio^[1]

With the installation of the iron filter in a beam hole, a beam of 25 keV neutrons essentially free of gammas was achieved. It was apparent that this facility could be better for tumor irradiations than the 2 keV beam. A cooperative experiment with ANL in the tumor irradiation research confirmed our conclusions. Deeper penetration of the body is achieved with the higher neutron energy. The iron filtered beam has additional advantages over the scandium filtered beam because it is free of higher energy neutrons and because the filter is less expensive to construct.

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BIOMEDICAL RESEARCH IN 2 keV BEAM

R. M. Brugger, L. G. Miller

Much effort has been directed toward putting B^{10} and U into brain tumors and then irradiating the head with thermal neutrons to produce highly destructive localized radiation through the $B^{10}(n, \alpha)Li^7$ reaction or the fission process. Besides the problem of enriching the tumor with B^{10} or U, there is the problem of getting the maximum flux of thermal neutrons at the tumor with minimal damage to the peripheral areas. While working with the world's first scandium filter at the MTR, it was realized that because of the unique characteristics of its beam of neutrons, the scandium filtered beam should be a superior irradiation facility to a thermal beam. The advantages of the scandium filtered beam are: (a) almost pure 2 keV neutrons, (b) essentially no gamma rays, and (c) a well collimated beam. A simple experiment has been performed to demonstrate this. A phantom head was constructed with two polyethylene bottles (Figure 1). The inner bottle, which is about the size of a brain, was filled with urea, sugar, and 1500 cc of water. This mixture supposedly represents tissue for dosimetry. Plaster of paris was put between the inner and outer bottles to represent the skull. Then a B^{10} neutron hand monitor and a geiger dosimeter were used to detect the thermal neutrons and β - γ dose at different locations through the skull. The skull was radiated in the scandium beam and in a similar well thermalized beam from the MTR thermal column. Figure 2 shows the results. Because of the questionable conversion factors the absolute dose may be in question but the relative dose rates are representative.

In Figure 2, one notes that while the thermal beam delivers the maximum dose at the surface of the head and the intensity drops off rapidly with penetration, the scandium beam delivers a minimal thermal dose at the surface and a maximum thermal dose inside the skull where the tumor will be located. Since the tumor is to be destroyed by the B^{10} or U absorption of thermal neutrons, the maximum thermal dose in the area of the tumor is the desirable condition. The 2 keV beam is thermalized in the area of the tumor to produce the highly desired thermal dose. The contrast shown in Figure 2 is quite striking. In addition, the scandium beam does not diverge rapidly; this collimation will restrict the dose to near the tumor area. It is estimated that a scandium beam designed for medical irradiations can be 100 to 1000 times more intense than the one used in the above experiment.

Fig. 1 While the girl to the left is real, the dummy to the right is one which was used to show the effectiveness of a newly developed beam of 2 keV neutrons. The tests show that this beam is much more effective for radiating brain tumors that have been spiked with ^{10}B or U than are thermal neutrons. The local radiation produced when the ^{10}B or U absorbs the neutrons can destroy the cancers.

Fig. 2 Comparison of radiation dose for thermal neutrons, neutrons from a scandium filtered beam and gamma rays as a function of penetration depth into the phantom head.

EXAMINATION OF ZINC PHOSPHATE DENTAL CEMENTS BY NEUTRON DIFFRACTION

L. Cartz^[1], W. R. Myers

Zinc phosphate cement, which is a dental cement, contains a noncrystalline phosphate component which is responsible for the formation of the cement. This component gives rise to a broad diffuse diffraction band, and by careful studies of this diffuse scattering, information about the nature of the phosphate component can be obtained. With X-ray diffraction, only one band can be resolved (from $0.3 \text{ \AA}^{-1}$ to $0.5 \text{ \AA}^{-1}$) because the atomic scattering factor drops off rapidly. Using neutron diffraction, it should be possible to resolve further diffuse bands because the atomic scattering factor for neutrons does not decrease rapidly with increasing of scattering angle. An added advantage of neutrons is that they penetrate the sample and see the sample in bulk while X-rays only sample the surface.

Preliminary studies using the time-of-flight neutron diffractometer have shown that it is possible to obtain suitable neutron diffraction data, despite the presence of large amounts of hydrogen in the cement. Diffraction data obtained shortly after the cement had been mixed and approximately one month later indicate that the cement undergoes a transition from an initially highly crystalline phase to one that is almost entirely amorphous.

[1] Professor, Dept. of Mechanical Engineering, Marquette University.

ACTIVATION ANALYSIS OF GEOLOGICAL SAMPLES

M. K. Corbett^[1], K. T. Faler

Neutron activation analyses using the MTR thermal column are being performed on several selected rock suites collected from the Never Summer stock, Colorado; the Cornelia stock, Ajo, Arizona; and the Albion Range, Idaho.

The Never Summer and Cornelia stocks are both silicic late-kinematic zoned igneous plutons whose compositions range from gradiorite as a border facies to monzonite-quartz monzonite inner facies. They both tend to reflect mineralogic variation characteristic of magmatic differentiation during mountain building events within a major tectonic belt, the Cordilleran region of Western United States. Standard geologic studies have previously been conducted and the general compositional variation relating to petrogenetic history established for both intrusive bodies. As a result, they provide an ideal framework within which to study natural occurrence, concentration, and distribution of trace elements associated with late magmatic stages of igneous intrusive activity. Of particular interest in the present study are the rare earth elements and those metals subject to hydrothermal emplacement. Although not generally mineral formers themselves, the substitutional behavior of these elements within silicate lattices permits their use in a petrogenetic sense, either as indices of the character and extent of the magmatic differentiation process or as clues to degree of assimilative and/or metasomatic reaction.

Neutron activation analyses are also simultaneously being carried out on samples collected from gneiss-schist units of the 2.4 billion year Green Creek complex from the Albion Range, Idaho. These rocks represent a metamorphic assemblage currently existing as the marginal envelope to the mid-Tertiary Almo monzonite-grandiorite pluton. Neutron activation, in this case, is being used to "fingerprint" the metamorphic complex in order to determine initial origin as either an igneous or sedimentary body, based on differences in element concentration resulting from the initial differentiation process involved.

The unusually old age and complex metamorphic history of the Green Creek gneisses and schist preclude retention of features normally considered to be indicative of origin. Several documented metamorphic events have caused the destruction and transformation of original mineralogy, texture, and structure. Recent studies have shown, however, that despite marked internal rearrangement, element diffusion and migration tend to be extremely limited. The presence or absence and relative abundance of key trace elements, and perhaps their distribution pattern, should be useful environmental indicators of the initial mode of formation. Such determinations are essential for interpreting the early geologic history for the Albion Range. The neutron activation technique is thus being applied, in this case, as one aspect of a more extensive geologic study.

As the studies described are carried out, some effort and time are also being devoted to development of the application of neutron activation to geologic-geochemical investigations. In all geologic studies involving sample analysis,

special concern is necessarily given to ensuring that samples are representative of the rock unit sampled. Natural variability in coarseness and composition often necessitates sample sizes which are excessively large. Normal procedure dictates homogenizing collected samples by pulverizing or otherwise grinding them to fine powder with analyses made on selected splits. When conducting standard analyses of major rock-forming components such a procedure has been considered sufficiently accurate.

Neutron activation analysis is, however, a highly sensitive technique and when used for trace element analyses could quickly produce questionable data, either because of exceptionally low element concentration, unrepresentative samples, or contamination during sample handling and preparation. The availability of the MTR's thermal column has provided an opportunity for a comparative study of data reliability resulting from the use of different sample sizes and preparation methods. Neutron activation of large bulk rock samples, of smaller powdered rock samples, and of fused rock samples is planned. Plans are also made for a comparison of the reproducibility of data collected.

It is hoped that the studies currently being conducted at the MTR will both improve and extend the usefulness of neutron activation analysis as a research technique and its application to the solution of geologic-geochemical problems.

[1] Associate professor of Geology, Idaho State University.

FILTERED BEAMS AS A NEUTRON SOURCE FOR SEED IRRADIATION STUDIES

J. R. Berreth

The scandium filter produces a neutron beam of 2 keV and also of several hundred keV with essentially no gamma component [1]. The neutron intensity of the 2 keV peak next to the scandium is approximately 10^7 n/cm²/sec and 3×10^6 n/cm²/sec for the high-energy component. In the use of this beam for seed irradiations, there are several advantages over other methods of irradiations: (a) The filtered beam provides essentially "monoenergetic" or at least narrow-energy range neutrons. (b) There is no gamma component. (c) One need not go through a gamma field to perform the irradiation; therefore, the dose effect is all due to the neutrons the seeds see. (d) Accessibility is outside the reactor.

In order to test the feasibility of using this beam to study neutron effects on seeds such as mutations, growth inhibition, etc, a 24-hour irradiation of Peroline barley was made. Three replications of 10 seeds each (irradiated and unirradiated) were germinated and checked for growth reduction according to procedures recommended by the IAEA [2]. A dose of 750 rad, produced by fast neutrons, for barley is considered to produce a 50% reduction in seedling height. For the 24-hour irradiation in the scandium beam in this preliminary experiment, a growth reduction of 37% was observed.

Even though the higher energy component is approximately 1/3 of the 2 keV neutron intensity the dose effect on seeds is much greater for the higher energy component. In this irradiation the high energy component contributed in the range of 80% of the total dose imparted to the seeds.

A 12-hour irradiation was also performed. These seeds received about 20% of the dose of the 24-hour irradiation due to positioning in the beam. The germination of these seeds produced an interesting result. There was an apparent growth enhancement of the barley. The irradiated samples were uniformly 22% taller than the unirradiated seeds.

Seeds have also been irradiated in the thermal column of the MTR. This column can produce thermal neutron fluxes up to 10^{11} n/cm²/sec with a minimal gamma component. At such a high flux we can easily study dose rate effects as well as total dose effects. The addition of a converter to the thermal column could also provide a fast neutron spectrum of high intensity for such studies. Utilization of the various beams now available provide an effective tool for studying neutron effects on seeds. Relative biological effectiveness (RBE) is a function of many things such as neutron energy, the type of seed irradiated, its moisture content, etc. The type of radiation damage incurred by the seed is very likely a function of neutron energy in particular in the keV range of neutrons. The filtered beams, therefore, provide an excellent means for testing and standardizing dose effects due to pure neutrons of different energies.

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- [1] "Experimental Facilities-Filtered Beams", this report.
 - [2] C. F. Konzak et al, "Recommended Standard Procedures for Irradiating, Cultivating, and Measuring Cereal Seeds to Determine the Effects of Neutron Irradiation in the Neutron-Seed-Irradiation Program", Technical Report Series No. 76, pp 103-107, International Atomic Energy Agency, Vienna (1967).

ACTIVATION ANALYSIS OF BIOLOGICAL TISSUE

E. L. Murri, R. L. Heath, C. J. Maletskos^[1]

Recent investigation indicates that certain diseases and metabolic conditions might be characterized by the abundance or lack of specific trace elements in human tissue. Conventional activation analysis techniques for studying trace elements have been quite unsuccessful due to the large amount of sodium and potassium in the tissue.

Dr. C. J. Maletskos of Harvard Medical School developed a technique, utilizing stable ion exchange on NaCl and KCl columns, which quite effectively reduces the sodium and potassium activity from the tissue. A cooperative measurement exchange was established wherein samples prepared by Dr. Maletskos were irradiated in the MTR, separated by the column technique, and analyzed with high-resolution Ge(Li) spectrometers at this laboratory.

The four types of tissue, kidney, liver, lung, and muscle, were from the same human subject. As illustrated in Figures 1 through 4, respectively, some significant differences are apparent in the trace elements observed in the different types of tissue. Figures 5, 6, and 7 show the radioactive decay of the muscle sample from 37 hours to 15 days. The presence of longer lived activation products, such as ^{75}Se and ^{134}Cs , are observed.

Since the techniques applied in this study appear satisfactory, continued investigation is likely, expanding to different human subjects from varying environmental conditions, and including those with known diseases.

[1] Medical School, Harvard University.

Fig. 1 Human tissue, kidney -- 15-min irradiation, 2-1/2 hr decay.

Fig. 2 Human tissue, liver -- 15-min irradiation, 2-1/2 hr decay.

Fig. 3 Human tissue, lung: -- 15-min irradiation, 2-1/2 hr decay.

Fig. 4 Human tissue, muscle -- 2-hr irradiation, 2-1/2 hr decay.

Fig. 5 Human tissue, muscle -- 2-hr irradiation, 37-hr decay.

Fig. 6 Human tissue, muscle -- 2-hr irradiation, 150-hr decay.

Fig. 7 Human tissue, muscle -- 2-hr irradiation, 15-day decay.

NEUTRON ACTIVATION METHOD FOR INVESTIGATING QUESTIONABLE ARCHAEOLOGICAL SPECIMENS

K. T. Faler

During the past year a large number of carved bone figurines like those shown in Figure 1 have been sold to art collectors. They are reportedly collected along the Columbia River. Archaeologists were unable to find stylistic or other observable differences between these carvings and those of known authenticity previously obtained through professional excavation of archaeological sites. However, the large number and relatively unbroken condition of the carvings suggested that they might more likely be recently made. It is of great scientific importance to archaeologists to determine if these items are of recent origin instead of some hundreds of years old. If they are recent, major confusion could be introduced into the study of the cultural history of the Columbia River area.

If the carvings in question had indeed been made recently, they must have been treated after carving, since their surfaces were darkened and cracked, making them appear to be very old. Chemical treatment might leave evidence which could be analyzed, but the value of the items precluded destructive testing methods of chemical analysis. Neutron activation seemed an attractive possibility for checking chemical differences. Preliminary studies of a method which could be used for these and similar cases were undertaken.

The smallest of the items was about three inches by one inch and the largest (a whale-bone dagger) was about 18 inches long. Their large size and fragile condition dictated use of a beam facility, such as the thermal column of the Materials Testing Reactor, as a source of neutrons rather than in-pile irradiation. The samples were irradiated for a few minutes and counted shortly after the irradiation was completed using a 30 cm³ Ge(Li) detector and the usual associated electronics. In order to establish a reference system for comparison purposes, both a fresh bone from a modern animal and a series of known samples recovered from different archaeological sites in the area from which the carvings were said to have been found were irradiated and counted under the same conditions.

The elements detected in the study were Na, Cl, Ca, and Mn. The relative amounts of the elements were determined by integrating the areas under the measured gamma-ray photopeaks and, after the usual corrections for radioactive decay time differences, the relative concentrations were normalized to the concentrations found in the fresh bone for Na, Ca, and Cl. Manganese was found to be below the detection limits of the procedure in fresh bone, so the manganese concentrations are expressed in arbitrary units. Bar graphs of the data are shown in Figure 2. Typical pulse-height spectra are shown in Figure 3.

The results show that the manganese, commonly found in buried bone, is present in the carvings but to a much smaller extent than in the museum samples of known origin. This might raise the question as to whether the carvings had indeed been buried or if they had been more shallowly stained. The strongest evidence that they may have been chemically treated, however, was presented by the low calcium and high chlorine content of the carvings compared to the museum pieces. In the laboratory, treatment of fresh bone by dilute hydrochloric acid produced Ca and Cl concentrations similar to that noted in the carvings. In addition, the acid treatment softened the surface of the fresh bone making it

porous and easier to stain. A surface layer was formed which, after drying, could be peeled off, leaving white material showing underneath. Such a condition was noted on the carvings, where flaking of the surface, exposing a light under-layer, seemed to be about the only visual difference between the carvings and the museum samples. The stains on the latter appeared to penetrate much more deeply into the body of the sample. Furthermore, the abnormally high concentration of sodium compared to fresh bone and museum samples might also indicate the possibility of chemical treatment of some sort. It is of interest that one of the pieces in question, the large whale-bone dagger, was tested after the conclusion of the study reported here and was found to be the same in Cl, Na, and Ca concentrations as the museum samples although low in manganese concentration. The manganese concentration by itself is not felt to be significant and the sample is likely of the stated antiquity and origin.

While the results of a study such as this cannot prove conclusively whether or not an item of reported prehistoric origin is authentic, the results of this exploratory project indicate that differences are observable among bone samples and can be established through nondestructive testing using neutron activation technique when intense beams of neutrons are available. Such tests can be of major importance to archaeology in evaluating the possibility of fraudulent or accidental mixing of recent and ancient bone objects in a collection being studied by scientists investigating cultural history.

Fig. 1. Some of the bone carvings reportedly from the Columbia River area.

Fig. 2. Relative concentrations of Cl, Ca, Na, and Mn in the bone carbons and museum samples tested. The length of the bar is proportional to the concentration of each element. Where the concentration was below the detection limit for the element, the arrow point indicates the maximum concentration that could have been present in the sample. The line through the bars represents the concentration of the element found in fresh bone except for Mn which was not found in fresh bone.

INC-A-25026

Fig. 3. A portion of the gamma-ray pulse-height spectra for two of the samples showing the typical variation in gamma-ray intensities characteristic of the two groups.

NEUTRON MOLECULAR SPECTRA OF GLYCINE ($\text{NH}_3^+\text{CH}_2\text{COO}^-$)

K. A. Strong, R. M. Brugger, R. J. Pugmire^[1], D. M. Grant^[1]

Neutron molecular spectroscopy has been used to study the biologically interesting molecule glycine. The spectrum, which was obtained with 50 meV neutrons (Figure 1), reveals several peaks which correspond to known frequencies in glycine[2]. One peak, however, which is indicated in Figure 1 by arrows at $\pm 160 \text{ cm}^{-1}$ (20 meV), has not been observed by other spectroscopic methods, and is tentatively identified as due to the C-C torsion. Table I compares the results of neutron spectroscopy with those of infrared.

TABLE I

VIBRATIONAL TRANSITIONS IN GLYCINE

Neutrons	Infrared ^[2]	Assignment
cm^{-1}	cm^{-1}	
1118	1111	NH_3 - rock
903	911	CH_2 - rock
597	608	CO_2 - wag.
500	516	NH_3 - torsion
339	355	CCN Bend.
160	not obs.	C-C torsion

The above neutron results are encouraging in that they show the value of neutrons in measuring vibrational transitions which are inactive to infrared; these measurements thereby expand our overall knowledge of molecules which are of biological importance. When neutrons are again available, the work on glycine will continue and careful consideration will be given to questions of crystalline form and changes in vibrational frequencies due to changes in crystalline form.

[1] Dept. of Chemistry, University of Utah.

[2] S. Suzuki, T. Shimanouchi, M. Tsuboi, "Normal Vibrations of Glycine and Deuterated Glycine Molecules", Spectrochim. Acta, **19** (1963) pp 1195-1208.

Fig. 1 Reduced partial differential scattering cross section versus neutron energy change for a sample of glycine at room temperature.

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F. EXPERIMENTAL FACILITIES, TECHNIQUES, AND INSTRUMENTATION

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I. EXPERIMENTAL FACILITIES

VERY INTENSE NEUTRON SOURCE (VINS)^[1]

G. J. Russell, R. M. Brugger

The need for more intense thermal neutron beams^[2] has been established and several systems to produce such sources have been proposed^[3-9]. One system, the Accelerator Pulsed Fast Reactor (APFR), is capable of generating copious numbers of short-duration, fast neutron bursts which can be moderated and used for low-energy time-of-flight experiments. The Very Intense Neutron Source (VINS) outlined here can be categorized as an APFR. It can provide effective^[2] thermal neutron fluxes in the 10^{16} - 10^{17} n/cm²-sec range. Compared to other proposed APFR's, VINS is of simple design, matches the best in beam current, does not have any rapidly rotating components, can effectively use ²³⁵U fuel, and is also distinguished by an excellent signal-to-background ratio. While the aggregate average power of VINS is high, the average power per module is low (25 kW), implying a long fuel life.

VINS is schematically illustrated in Figure 1. It consists of a number (60) of fast reactor modules attached to the rim of a slowly rotating wheel. Each module is composed of solid fuel and the energy generated in a module is not sufficient to melt it. The basic idea is to inject neutrons from an external source into a module when its reactivity is a maximum, reduce its reactivity, and wait a sufficient time for the delayed neutron precursors to decay away before the process is repeated. As presently depicted, the modules are decoupled from one another, but, conceivably, this moving decoupler need not be present. The modules would be gas cooled and the system would be flexible and accessible with respect to module replacement and moderator modifications. Some preliminary design and performance characteristics are shown in Table I.

A scaled down version of VINS could provide a source for university use which would produce an equivalent flux of 10^{14} n/cm²-sec with a very good signal-to-noise ratio. A facility built around a VINS concept, like any installation with an accelerator, would have great experimental flexibility.

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- [1] G. J. Russell and R. M. Brugger, "Very Intense Neutron Source (VINS)", ANS Trans., 12 n 1 (June 1969) pp 285-286.
 - [2] R. M. Brugger, "We Need More Intense Thermal-Neutron Beams", Phys. Today, 21 n 12 (December 1968) pp 23-30.
 - [3] R. G. Fluharty, "Discussion of Proposed Booster", in J. Spaepen (ed.), Neutron Time-of-Flight Methods. Proceedings of a Symposium Organized by European-American Nuclear Data Committee in Collaboration with Centre D'Etudes Nucleaires De Saclay (France) and Institut National Des Sciences et Techniques Nucleaires, Saclay, 24-27 July, 1961, Brussels: European Atomic Energy Community, 1961.
 - [4] G. A. Kolstad and I. F. Zartman, Proceedings of a Symposium on Pulsed High Intensity Fission Neutron Sources held at USAEC, Washington, D. C., February 1965, CONF-650217 (February 1965).

- [5] IAEA, Symposium on Pulsed Neutron Research, Karlsruhe, Germany, 10-14 May, 1965, Vienna: International Atomic Energy Agency, 1965.
- [6] IAEA, Proceedings of a Panel on Research Applications of Nuclear Pulsed Systems, Dubna, USSR, 18-22 July, 1966, Vienna: International Atomic Energy Agency, 1966.
- [7] AEC-ENEA, Intense Neutron Sources. Proceedings of a United States Atomic Energy Commission/European Nuclear Energy Agency Seminar, Santa Fe, New Mexico, 19-23 September 1966, CONF-660925 (September 1966).
- [8] R. G. Fluharty, F. B. Simpson, G. J. Russell, R. H. Morris, A Proposal for a Repetitively Pulsed Test Facility (RPTF), IN-1149 (December 1967).
- [9] Proceedings of a Meeting on Fast Burst Reactors, Albuquerque, New Mexico (January 1969) to be published.

Fig. 1 Very intense neutron source (VINS).

TABLE I

PRELIMINARY OPERATING CHARACTERISTICS OF VINS WITH VARIOUS FUELS

(Source Strength 1×10^{18} n/sec, Source On-Time 4 μ sec, Prompt Neutron Lifetime 20 nsec, Pulse Production Rate 100 pps)

	Moveable Modules (VINS)			Stationary Module ^[a]
	^{235}U	^{233}U	^{239}Pu	^{239}Pu
Wheel RPM	1	1	1	2,000 ^[b]
Wheel or Rotor Radius (meters)	~ 4 ^[c]	~ 4 ^[c]	~ 4 ^[c]	1
Avg. Power Per Module (MW)	25×10^{-3}	25×10^{-3}	25×10^{-3}	1.5
Avg. Total Power (MW)	1.5	1.5	1.5	1.5
Peak Power (MW)	1,660	1,690	1,570	1,530
Peak Power/Max. Bkg. Power	42,000	88,000	93,000	1,600
Peak Power/Avg. Bkg. Power	340,000	800,000	940,000	7,300
Pulse Width-FWHM (μ sec)	5.9	6.4	7.0	6.6
% Energy in Prompt Pulse	80	92	93	86
Avg. Leakage Rate (n/sec)	5.5×10^{16}	6.9×10^{16}	8.8×10^{16}	8.1×10^{16}
Peak Reactivity (β)	+0.38	-0.27	-0.45	-0.58

[a] More standard APFR operation; reactivity is varied by passing fuel through center of reactor or fuel or reflector past its surface.

[b] Based on three reactivity pulses per revolution.

[c] Determined by amount of decoupling assumed -- would probably be less than this value.

2 keV SCANDIUM FILTERED BEAM FACILITY

O. D. Simpson, L. G. Miller, D. R. Staples

The 2 keV beam facility^[1] has not been changed during the past year. An interface has been designed and built for the PDP-8/S data acquisition machine that will automatically control and change the temperature of a

furnace. This interface will make it possible to investigate the variation in cross sections as a function of temperature at 2 keV. The facility has been used extensively and many of the beam characteristics have been investigated. The following experiments have been carried out during the past year using the 2 keV beam and are reported in other sections of this annual report:

- (1) Proton-Recoil Measurements in the MTR 2 keV Neutron Beam, J. W. Rogers and O. D. Simpson, page 283.
- (2) Proton-Recoil Calibration Studies in the MTR Neutron Beams With Hydrogen and Methane Detectors, J. W. Rogers, page 289.
- (3) Biomedical Research in the 2 and 25 keV Beam, R. M. Brugger and L. G. Miller, pages 211 and 212.
- (4) Eta Measurements of ^{239}Pu and ^{235}U at 2 keV, J. R. Smith and S. D. Reeder, page 24.
- (5) ^{239}Pu Partial Cross Section and Alpha Measurements at 2 keV, O. D. Simpson and L. G. Miller, page 36.
- (6) The Total Neutron Cross Section of ^{235}U and ^{239}Pu at 2 keV as a Function of Sample Thickness, O. D. Simpson and L. G. Miller, page 38.
- (7) Filtered Beams as a Neutron Source for Seed Irradiation Studies, J. R. Berreth, page 216.
- (8) Activation Cross Section Measurements using the 2 keV Sc Filtered and 25 keV Fe Filtered Neutron Beams, R. P. Schuman and R. L. Tromp, page 39.
- (9) Levels in ^{182}Ta , R. G. Helmer, R. C. Greenwood, C. W. Reich, page 102.
- (10) Excited $K^\pi = 0^+$ Bands in ^{172}Yb , R. C. Greenwood, C. W. Reich, R. A. Harlan, page 93.
- (11) Levels in ^{156}Gd from $^{155}\text{Gd} (n,\gamma)$, R. C. Greenwood and C. W. Reich, page 78.
- (12) Neutron Capture Gamma-Ray Measurements with a 2 keV Reactor Produced Neutron Beam, R. C. Greenwood, R. A. Harlan, R. G. Helmer, C. W. Reich, page 109.
- (13) Measurement of Fast Neutron Capture Gamma-Ray Spectra for Fast Reactor Shielding Calculations, R. C. Greenwood, R. A. Harlan, C. W. Reich, page 116.

[1] O. D. Simpson and L. G. Miller, "Scandium Beam Experiments", in R. L. Heath (ed.), Nuclear Technology Branch Yearly Progress Report for Period Ending June 30, 1968, IN-1218 (December 1968) pp 3-5.

25 keV IRON FILTERED BEAM FACILITY

O. D. Simpson, R. M. Brugger, B. W. Howes, J. W. Rogers,
R. P. Schuman, F. B. Simpson

A reactor produces one of the most intense sources of neutrons available; the limitation is selecting only those neutrons that are needed in an experiment. A development program which will produce some of these desired beams of neutrons is being pursued at MTR.

A monochromatic beam of 25 keV neutrons has been obtained^[1] from the HG-5 beam facility of the MTR by using a combination of natural iron, aluminum, and sulfur filters. This well-collimated beam has an initial diameter of 3.50 inches and a neutron flux of approximately 3×10^5 neutrons/cm²/sec. The gamma-ray field associated with the beams was determined to be less than 20 mR/hr. The neutron energy spectrum of the beam has been analyzed from 1 keV to 1 MeV with a proton-recoil spectrometer. The raw proton-recoil data which were obtained with a hydrogen-filled detector and multi-channel pulse-height analyzer were used to "optimize" the beam, ie, to obtain the maximum ratio between the magnitude of the 25 keV neutron peak and general "background" level. The "background" intensity has been reduced to less than 2% of the 25 keV neutron intensity.

Figure 1 shows the neutron energy spectra which were obtained from proton-recoil spectrometer data taken in the HG-5 beam with the following filters: (1) 40.62 inches iron; (2) 37.96 inches iron, 2.66 inches aluminum; (3) 30.96 inches iron, 6.16 inches aluminum, 2.75 inches sulfur. These three spectra have been normalized to the same peak intensity. The FWHM of the 25 keV neutron peak was found to be 4.3 keV which is in reasonable agreement with the predicted resolution of the detector (3.75 keV).

The results of this study indicate that a near monochromatic source of 25 keV neutrons can be obtained with a neutron flux of approximately 10^7 n/cm²/sec by utilizing one of the high intensity beam holes of the MTR.

The following experiments have been carried out using the 25 keV beam and are reported in other sections of this annual report:

- (1) Activation Cross Section Measurements Using the 2 keV Sc Filtered and 25 keV Fe Filtered Neutron Beams, R. P. Schuman and R. L. Tromp, page 39.
- (2) Proton-Recoil Calibration Studies in the MTR Neutron Beams with Hydrogen and Methane Detectors, J. W. Rogers, page 289.
- (3) Neutron Transmission Measurements for Shielding, J. W. Rogers, R. M. Brugger and O. D. Simpson, page 34.

[1] B. W. Howes, R. M. Brugger, J. W. Rogers, R. P. Schuman, O. D. Simpson, "The Materials Testing Reactor 25 ± 1 keV Neutron Beam", ANS Trans., 12 n 1 (June 1969) pp 195-196.

Fig. 1 MTR neutron beam spectra obtained with a proton-recoil spectrometer for various thicknesses of Fe, Al, and S filters. These spectra demonstrate how the 25 keV neutron beam was obtained.

144 keV SILICON FILTERED BEAM FACILITY

O. D. Simpson, J. W. Rogers, H. G. Miller

A third neutron filtering material that has been tested is silicon. The resonance at 300 keV in silicon has a strong interference dip in the cross section at 144 keV. This interference dip indicated that silicon would produce a 144 keV beam much in the same manner as scandium and iron produced the 2 and 25 keV beams. The relatively low cost of a good grade of silicon made it possible to investigate the feasibility of using silicon as a neutron filter.

For preliminary studies, 40 inches of silicon (99.999% purity) were purchased. The silicon pieces were ordered such that their dimensions were the same as the iron filtered pieces so that they could be tested in the iron test facilities.

A proton-recoil spectrometer^[1] using a hydrogen filled proportional counter was used to examine the spectrum between 50 and 300 keV. These measurements show the major transmission "window" at 144 ± 2 keV and a secondary "window" at approximately 55 keV as depicted in Figure 1. The measurements were not extended to lower energies due to the high gamma background, but gamma discrimination measurements will be made following beam modifications. Higher energy measurements were not necessary since no flux could be detected above the 144 keV "window". The gamma intensity was measured in the direct beam and was found to be approximately 8 R/hr. In order to reduce the high

Fig. 1 Neutron spectrum through 40 inches of silicon. The data were taken with a proton-recoil spectrometer using a hydrogen filled proportional counter. The fine structure below 80 keV (except for the 55 keV window) is due to the strong gamma component.

gamma background, lead shielding was placed in the beam. To check for spectrum distortion and softening by the lead several thicknesses were used. Figure 2 shows a comparison of spectra measured through four and six inches of lead after normalization at the primary peak. The lead does not distort the neutron spectrum significantly. The neutron intensity is reduced by 75% by increasing the lead thickness from four to six inches.

Titanium has a large cross section in the region of 55 keV and was therefore used to remove the peak which was observed at 55 keV with no detectable distortion in the primary beam. Figure 3 shows the effect of the titanium on the spectrum. These spectra have been normalized at the 144 keV peak. The magnitude of the peak flux is 10% higher with the titanium and two inches of lead than for no titanium and four inches of lead. The "tail up" for the titanium filtered spectrum is attributed to a nonconstant gamma background due either to the titanium filter or room background.

Fig. 2 Comparison of the spectra measured through 40 inches of silicon and 3/16 inch of ^{10}B powder, with the addition of four or six inches of lead. The spectrum through six inches of lead is degraded. This effect would not have been observed if the detector had been further away from the lead.

Because of a Bragg cutoff at 0.002 eV in silicon, a strong low energy neutron component was present in the beam and was detected by the $^{14}\text{N}(n,p)^{14}\text{C}$ reaction from the nitrogen in the counter. This was easily removed by a boron-10 filter with no significant spectrum distortion.

A study was made as to the best way of removing the strong gamma component in the beam, and it was concluded that the thickness of silicon needs to be increased. Therefore, an additional 24 inches of silicon has been ordered. It is expected that by increasing the filter thickness by 24 inches that the gamma background will be reduced to the order of a few hundred mR/hr. The optimization of the 144 keV beam will be accomplished by investigating the beam intensity for various thicknesses of silicon, titanium, and cadmium or ^{10}B .

Fig. 3 Neutron spectra through the silicon filter with and without four inches of titanium in the beam. The fine structures below 80 keV are due to the strong gamma component.

Another measurement of interest was to measure the scattered neutron spectrum emerging from the side of the silicon filter. This was done by extending the end section of silicon out about three inches and measuring the spectrum at the face and side of the extended section. Figure 4 shows these spectra. The spectrum taken at the side of the silicon is 10 keV lower (134 keV) at the peak, very low in intensity (about 1.5% of that at the face) and broadened by smearing on the low energy side of the peak. This is what is expected since most of the neutrons would be 144 keV after passing through the first 37 inches of silicon, in order to be scattered out the side they would have to undergo a rather hard collision, further reducing the energy. Simple considerations of energy change in scattering indicate that the maximum fractional loss of energy per collision would be 20 keV for silicon. Qualitatively, this is in agreement since all scattered neutrons would not suffer maximum energy loss nor would there be only single collisions. In order to more precisely predict the energy of this side scattering a much more sophisticated treatment of the energy change in scattering would be required.

[1] J. W. Rogers and L. O. Johnson, "Proton-Recoil Spectrometer", this report.

Fig. 4 Neutron spectra at the face of the silica in the main beam as compared to the scattered spectra at the side of the silicon out of the main beam.

100 KBAR PRESS FOR NEUTRON DIFFRACTION STUDIES

R. B. Bennion

Neutrons have very distinct advantages for crystallographic studies at high pressures. Their deeply penetrating characteristics make them very adaptable as a probe in complex high-pressure systems. With neutrons, one can determine the position of hydrogen atoms in compounds, the order-disorder effects in near Z alloys, the crystal structures of the very heavy elements and their compounds, and magnetic structures of ferro- and antiferromagnetic materials.

Other laboratories have stayed below 10 kbar with neutrons but developments in the last several years at the MTR have achieved 40 kbars. To extend this range still further, a Bridgman anvil apparatus has been constructed[1]. This 100 kbar press was completed and installed at the HG-6 beam hole, and two weeks of neutron diffraction data were taken with the system installed before the MTR beryllium change-out occurred[2]. This was done to check out the neutron scattering properties of the system. The pressure system was tested previously and shown to be capable of reaching pressures in excess of 85 kbar. A picture of the press after assembly is shown in Figure 1. One of the anvil and binding ring assemblies is removed to show its construction.

Figure 2 shows the NaCl diffraction pattern and the beam monitor pattern for the 1 m, 2 m flight paths, respectively taken at room pressure. The general "hump" in the middle of the short flight path (SFP) data is from multiple scattering through the press and support table. This can be reduced by improving the shielding and collimation. The long flight path (LFP) used in this experiment was borrowed from the 40 kbar experiment; the regular LFP was not completed at the time of this check-out run.

These data are encouraging in that only 60 hours were required to obtain them with three counters (18- by 1-inch diameter) in the LFP and 12 counters (10- by 5/8-inch diameter) in the SFP. The final system will have 110 and 40

Fig. 1 100 kbar press as assembled. One of the anvil and binding ring assemblies is removed to show its construction.

counters in the LFP and SFP, respectively. The diameter of these counters will be smaller (1/2 inch - SFP, 5/8 inch - LFP) which will increase the resolution.

The pressure system check-out was done using pyrophyllite as the compressible gasket material and the electrical resistance of bismuth as the pressure calibrant. Bismuth metal exhibits three polymorphic transitions as evidenced by discontinuities in electrical resistance at 25.4, 27, and 76 kbar. The 76 kbar transition was reached with a press load of 39 kbar which indicates an amplification effect similar to that reported by Myers et al^[3] for a diameter-to-thickness ratio of about 9.

Since pyrophyllite contains 2 hydrogen atoms ($\text{Al}_2\text{O}_3 \cdot 4\text{SiO}_2 \cdot \text{H}_2\text{O}$) per molecule it is unsatisfactory as a gasket material for neutron diffraction at high pressures. A search for a workable gasket material with the proper neutron characteristics was initiated. Mixtures of alundum cement and sulfur were tried without success. The sulfur reacted with oxygen quite rapidly in the high shear stress environment of the pressure gasket. A hard aluminum alloy (7075-T6) was also tried. It begins to "flow" quite readily at about 20 kbar and will not contain the sample.

The sample geometry and material also play an important role in determining the gasket material. For instance, even pyrophyllite was unsuccessful in containing a large thallium sample (1/4-inch diameter x 1/10-inch thick disk) above 15 kbar while the aluminum gasket was capable of supporting 40 to 50 kbar on a thin bismuth sheet held between two aluminum plates. There are many other gasket materials and ideas to try and during the beryllium change-out of the MTR this study will continue.

In summary, the 100 kbar neutron diffraction experiment is progressing quite well. The neutron capabilities are better than expected and the large alumina anvils seem to be able to produce pressures in the 80 to 100 kbar range. Some alterations of these anvils will be tried to increase the life of each anvil. The gasket material and sample geometry problems are not completely solved, but they are not formidable.

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- [1] R. B. Bennion, "Apparatus for Neutron Diffraction Studies in the 100 Kbar Pressure Range", in R. L. Heath (ed.), Nuclear Technology Branch Yearly Progress Report for Period Ending June 30, 1968, IN-1218 (December 1968).
 - [2] Some of the beryllium in the core of the MTR is to be changed before the Phoenix core experiment during July and August 1969.
 - [3] M. Myers, F. Dachille, R. Roy, "Contribution to Calibration of High-Pressure Systems from Studies in an Opposed-Anvil Apparatus", in A. A. Giardini and E. C. Lloyd (eds.), High-Pressure Measurement, Washington: Butterworth Inc., 1963, pp 17-33.

Fig. 2 NaCl diffraction patterns for the 1- and 2-meter flight paths and the beam monitor taken in the 100 kbar press. The general "hump" in the middle of the short flight path can be reduced by adding acid or Li-salt to the water in the support table.

CORRELATION CHOPPER FOR NEUTRON TIME-OF-FLIGHT EXPERIMENTS

P. D. Randolph, W. R. Myers, H. G. Miller

The possibility of increasing the duty cycle for time-of-flight experiments to nearly 50% has resulted from development of statistical chopping techniques [1]. A correlation chopper which uses a pseudorandom array of slots to modulate the beam incident on the target has been built and tested. The scattered beam is time-analyzed and the resulting data are cross-correlated with the chopper slot sequence to obtain the actual time-of-flight spectrum.

The MTR chopper (Figure 1) is a 12-inch diameter disk of aluminum, 1/2 inch thick, with two identical pseudorandom sequences of 255 slots around the periphery. The slots are filled with gadolinium oxide-loaded epoxy in the specified sequence, such that 128 slots will pass neutrons and 127 are neutron absorbing. The figure shows the chopper prior to filling the slots. The duty cycle of the chopper is 128/225, or approximately 50% as compared to 0.1 to 0.5% for conventional choppers. The basic slot width is 1/16 inch, providing a neutron burst width of 7.8 μ sec when the chopper is rotating at 15,000 rpm. The small slots around the periphery pass light pulses to provide a phased analyzer clock.

The chopper was tested at the High Pressure Neutron Diffraction facility by obtaining diffraction patterns of NaCl, graphite, and aluminum. Figure 2 shows the diffraction pattern of graphite at a scattering angle of 60 degrees by the conventional method (top) and obtained using the correlation chopper (bottom). The pattern in the center of Figure 2 is the raw data obtained in the time-of-flight analyzer using the correlation chopper.

The mathematical analysis of the correlation method shows it to be statistically superior to the conventional method for points that are more than 33% above the average counting rate. This level is indicated by the dashed line in the bottom pattern.

While the duty cycle is actually increased to $\approx 50\%$, this improvement in efficiency is not always realized. The conventional method is superior if small singularities must be resolved in the presence of large peaks; the correlation method is preferred if all peaks are of the same order of magnitude and the time-independent background is large.

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Fig. 1 Photograph of the statistical chopper before the slots were filled with epoxy. The integral rotor for the motor drive is also shown.

Fig. 2 Time-of-flight patterns for: (a) the conventional method and (b) the correlation method. The correlation method diffraction pattern derived from (b) is shown in the lower section (c).

$\pi/2$ ELECTRON SPECTROMETER

R. G. Helmer

As reported previously[1] a high-resolution $\pi/2$ electron spectrometer has been constructed at the NRTS. During the past year this spectrometer has been put into operation, many of the necessary checks have been completed, and the first experiments have been started. As noted in the following, the operational parameters agree with the design characteristics.

After the six spectrometer coils were completed (by the Oak Ridge facility of Union Carbide), the desired distance between the coil pairs was computed in order to give the necessary magnetic field shape. From these spacings and the measurements of the support castings, the thicknesses of the various shims were calculated. In September 1968 the top half of the spectrometer was put in place and the separations of the coils were measured. It was found that the spacing of the large coils was correct, but the medium and small coils were 0.020 inch too far apart. Further, the lower coils were in the proper locations and the top ones were too high. These two coils were lowered. The final measurements show that the spacings between the coil pairs agree with the desired values within 0.001 inch -- large, 0.003 inch -- medium, and 0.009 inch -- small. If one defines the midplane as the plane half way between the large coils, the other two pairs of coils are symmetrically placed to within 0.001 and 0.002 inch.

One measurement was made of the shape -- or radial dependence -- of the magnetic field. With the spectrometer current set to give about 800 milligauss (mg) at the central orbit, the differences between the field at the central orbit (35 cm radius) and that at other radii were measured. For radii between 22 and 44 cm, the differences between the ideal field shape and the measured one were 1 to 15 parts in 10^4 , which is close to the limit of the accuracy of the measurement methods. From 45 to 50 cm the measured field diverges from the ideal form by 0.3 to 1%, but this deviation agrees with the field calculated for this coil configuration.

The vacuum system operates satisfactorily with a pressure of 1 to 3×10^{-6} torr at the vacuum tank. The drive systems for the detector slit and the baffles operated into the vacuum with no leaks. The source chamber, which allows two degrees of freedom in the source position, also operates without any leaks.

The cooling system for the spectrometer coils has been completed and the initial tests have not indicated any problems with it. However, the water lines have not been connected to the spectrometer. All the electron spectra taken so far have been taken without any cooling. For electrons of 300 keV, the focussing current is about 33 amperes. At this current the coil temperature rises about 5°C and then remains constant. This is expected to cause a change in the spectrometer of about 1 part in 10^5 , which is negligible in almost all cases. Lines as high as 400 keV have been observed without any difficulty.

Since the calibration of the spectrometer assumes that the only magnetic field that the electrons interact with is that of the coils, it is necessary to

cancel, as well as possible, the magnetic field of the earth and surrounding material. In the last check of the cancellation system, each of the three components of the residual field was measured at 15 places over the volume that the electrons travel within the vacuum tank and at one reference point outside the tank. With the cancellation system operating, the variation in the residual field components was 0.55 -- vertical, 0.80 -- north-south, and 1.00 milligauss (mg) -- east-west. (These are to be compared with the original components of about 550, 220, and 10 mg.) The vertical component is the most important since it affects the calibration, whereas the others have a defocussing effect. If one considers only those points on the optic circle (ie, these orbits used at high resolution), the range of the residuals of the vertical component is 0.40 mg. By setting the compensating field so that the average vertical component of the residual field is zero, the remaining effect of this component will be negligible.

The first electron spectrum was observed in November 1968. Since that time a series of sources has been made in order to experiment with various source-making techniques, in particular electroplating, electrospraying, and vacuum evaporation. The one electroplated source showed large tails on the low-energy side of the peaks due presumably to scattering of electrons in the source. The source gave a resolution of 0.2% although the source width corresponded to 0.1%. Several sources have been made by electrospraying. Of these one had excellent line shapes (no tailing) and gave a resolution of about 0.1%. The other sprayed sources have had undesirable amounts of tailing on the lines. Generally, the sources made by vacuum evaporation of cold material, with subsequent activation, have been good. One source has given a resolution of 0.016%.

For all spectra taken so far the electrons have been counted with a surface-barrier silicon detector. The noise from this detector and the associated electronics limit the resolution of the detector to about 25 keV. This means that electron lines below about 60 keV will be lost in the noise. Modifications of this detector and use of other detectors will extend the useful range to lower energies.

One series of measurements was undertaken to determine the problems involved in precision energy measurements and to develop computer techniques for analysis of such data. The K-internal-conversion lines from three strong transitions following the decay of ^{192}Ir were scanned four to six times over the period of 40 hours. In spite of the occurrence of a power outage during this period, it was possible to obtain some excellent measurements on the differences in the energies of these transitions. These results and comparable values from the measurements of the gamma-ray energies with Ge(Li) and crystal diffraction spectrometers are as follows:

<u>Method</u>	<u>Energy Differences (eV)</u>	
	<u>E(308)-E(295)</u>	<u>E(316)-E(308)</u>
electron spectrometer	12502	8050
crystal spectrometer ^[2]	12499	8050
Ge(Li) spectrometer	12493	8055

Previous studies of the level structure of ^{183}W from the decay of ^{183}Re and ^{183}Ta have produced some conflicting information and left some unanswered questions about the nuclear structure. Therefore, a study of the ^{183}Re decay has been initiated. As background, the electron spectrum from ^{183}Ta has been measured. These data were sufficient to resolve conversion lines from transitions of 313.0 and 313.3 keV for the first time. This is the first of the new experiments with the spectrometer.

As an illustration of about the best resolution that can be obtained with this spectrometer, the internal-conversion lines associated with the 316-keV transition from the decay of ^{192}Ir are shown in Figure 1. The resolution on the L shell lines is 0.016% (or about 50 eV). This value is limited primarily by the source width (0.006 inch), although resolutions better than 0.010% are not expected[3]. The K line resolution is only 0.025% (or about 60 eV) because the K shell of the atom has a natural width (ie, an effective uncertainty in the binding energy) of 50 eV which is larger than the instrumental width of ~ 35 eV.

Fig. 1 Internal-conversion-electron lines from 316-keV transition from the decay of ^{192}Ir . The atomic subshell from which the electrons come is given, along with the electron energy.

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- [3] R. L. Graham, G. T. Ewan, J. S. Geiger, "A One-Meter-Radius Iron-Free Double-Focusing $\pi\sqrt{2}$ Spectrometer for β -Ray Spectroscopy with a Precision of $1:10^5$ ", Nucl. Instr. & Methods, 9 (1960) pp 245-286.

LOW GEOMETRY ALPHA COUNTER

D. K. Oestreich, W. R. Beck

Fission experiments are done with neutron beams at the MTR and with nuclear explosions. Because of this, a definite need existed at the MTR for an instrument which could be used for low geometry alpha counting of fission foils to assay the amount and uniformity of material electro-deposited. Very often, these foils contain as much as $500 \mu\text{g}/\text{cm}^2$ of materials deposited over 10 to 15 cm^2 and exhibit high specific activity. It is possible that the sample may emit as many as $10^{13}\alpha/\text{min}$.

A device (see Figure 1) has been built consisting of a vacuum chamber and a silicon detector. A polyethylene cap with a $1/64$ -inch hole in the center can be placed over the detector to act as a collimator. The distance between source and detector is 7.75 inches. The efficiency of the system is $50 \times 10^{-3}\%$ without the collimator and $1.33 \times 10^{-4}\%$ with the collimator.

The amplified signal can be counted on a scaler, thereby determining the amount of material. If fed into a multichannel analyzer, information as to the quality of the foil can be obtained from the degradation in resolution of the alpha spectra due to poor plating or the presence of impurities. If extremely large quantities of material are present on the foil ($>1 \text{ mg}/\text{cm}^2$), the assay will not be quantitative due to self-absorption.

Fig. 1 Low geometry alpha counter.

THE COUPLED FAST REACTIVITY MEASUREMENT FACILITY

D. A. Millsap, J. J. Scoville

The Coupled Fast Reactivity Measurement Facility (CFRMF) was designed and built in support of the AEC's overall effort in fast reactor physics. A zoned core concept was employed in the modification of an existing reactivity measurement facility to provide a fast spectrum in the measurement region [1].

Criticality was achieved in the CFRMF December 31, 1968. Since that time operation has been directed primarily to measurements of the flux spectrum in the experimental region of the core. These measurements have utilized state-of-the-art techniques in proton-recoil spectroscopy and have been compared to reactor calculations [2]. Refinements in calculational techniques and input cross sections need to be explored in order that better agreement be achieved.

An experiment was undertaken which in the final analysis should yield a spectrum average value for the capture-to-fission ratio in ^{239}Pu . This experiment consisted of irradiating at 1000 watts power level a sample of ^{239}Pu in the experimental region of the CFRMF. The sample is now undergoing chemical analysis which includes both mass and gamma-ray spectroscopy.

Some reactivity measurements have been completed in the CFRMF; the data are shown in Table I. These data will be compared to perturbation theory calculations as soon as ENDF/B cross sections can be obtained. The comparison, when complete, will provide a useful check of the ENDF/B data.

Reactivities for $^{239}\text{PuO}_2$ were calculated in the spectra derived from both the PHROG and ENDF/B libraries and were compared with experimental results [2]. The calculations were made using the PERT-II perturbation code [3]. Table II shows the percentage reactivity contributions to various energy ranges for the calculations based on the two library sets. There is a significant shift to higher energies in the results based on ENDF/B cross sections. Examination shows that this is predominantly due to the differing central fluxes estimated by the two cross section sets and not due to significant differences in the plutonium cross sections. A reactivity measurement was made on a 12.01-g sample of high purity $^{239}\text{PuO}_2$ and gave a response of $1.62 \mu\text{k/g}$. The reactivities were $1.20 \mu\text{k/g}$ and $1.45 \mu\text{k/g}$ for the PHROG and ENDF/B library-based calculations, respectively. The improved agreement using ENDF/B cross sections is substantial.

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 - [3] H. P. Flatt, PERT -- A Perturbation Theory Code, AI-Memo-64-102 (May 1964).

TABLE I

CFRMF REACTIVITY DATA

<u>Sample</u>	<u>Mass</u>	<u>Reactivity (total)</u>	<u>Specific Reactivity</u>
Nb	210.65 g	- 44.00 μ k	-0.209 μ k/g
Ta	361.6	- 83.76	-0.232
Mo	228.0	- 35.33	-0.155
Au	43.3	- 9.92	-0.229
Bi	444.700	- 10.07	-0.023
Pb	505.070	- 10.69	-0.021
Sn	328.561	- 26.37	-0.080
Al	121.383	- 20.31	-0.167
Mg	78.147	- 22.22	-0.284
Zr	299.950	- 23.68	-0.079
Be	85.912	-103.02	-1.199
C(U-1)	77.511	- 49.84	-0.643
C(UF-4-5)	79.211	- 51.29	-0.648

TABLE II

CONTRIBUTION TO TOTAL REACTIVITY

<u>Energy Range</u>	<u>Percent of Total Absorption Reactivity</u>		<u>Percent of Total Fission Reactivity</u>	
	<u>PHROG Cross Sections</u>	<u>ENDF/B Cross Sections</u>	<u>PHROG Cross Sections</u>	<u>ENDF/B Cross Sections</u>
0.532 - 1.23 keV	2.65	1.27	4.02	1.90
1.23 - 15.0 keV	3.27	2.07	5.62	3.84
15.0 - 40.9 keV	6.24	3.25	8.45	4.74
40.9 - 183.0 keV	29.95	17.18	31.95	19.98
183.0 - 821.0 keV	29.76	45.43	27.23	42.57
0.821 - 3.68 MeV	23.87	25.89	19.62	23.04
3.68 - 10.0 MeV	4.23	4.91	3.35	3.91

FAST NEUTRON FLUX IRRADIATION FACILITY

G. J. Russell, E. Fast

A study is in progress to design a relatively inexpensive irradiation facility which would provide a high intensity filtered neutron spectrum comparable to that anticipated in a fast power reactor. This facility would be placed in the ETR or possibly the MTR. Previous studies concerned with performing fast fuel tests in thermal reactors^[1,2] focused on filtering the reactor spectra through different amounts of Cd or ^{10}B . The resulting spectra in the sample region would be essentially a fission spectrum shape (peaking at approximately 2 MeV) with various amounts of low energy neutron contamination. However the peak of the neutron spectrum in a fast power reactor should be in the 100 to 500 keV range. The present study has been concerned not only with filtering a thermal reactor spectrum but also degrading the fission distribution to one more closely matching a fast reactor. The feasibility calculations, which are summarized in the following paragraphs, are encouraging, and work is continuing on finalizing the design.

A model of the facility used in these preliminary calculations is shown in Figure 1. It consists primarily of sample, filter, and shaper regions as well as a mock-up of a critical ARMF^[3] core. Flux distributions, both spacial and energy, were obtained using the one-dimensional, diffusion theory code MONA with 26 energy groups. Cross section sets were obtained using the PHROG and INCITE codes. Cylindrical geometry was used with the irradiation facility confined to a three-inch diameter. Calculations were made with both ^{235}U , ^{239}Pu , and a void in the sample region. An example of what could ultimately be expected in the way of spectrum shaping and filtering compared to just filtering alone is shown in Figure 2. The curves are normalized to each other on an equal area basis. The effect of irradiating a ^{239}Pu sample in each of these spectra is shown in Table I. Both the percent absorptions in the sample and the percent of the spectra with no sample present are listed in the table for four energy regions.

The present study has also included investigations of different materials for use as "shapers", filters, and the effect of water channel thickness. Stainless steel was used wherever cladding or structural components were required. A summary of these studies is given below.

Spectrum Shaper

The heavy elements investigated for use as a shaper included ^{238}U , Ta, and W. A comparison of the relative spectra in a voided sample region for the same shaper thickness is shown in Figure 3. The relative normalization is based on a fixed reactor power. The results indicate that tungsten is preferable from nuclear considerations. In addition, from a fabrication viewpoint, it is desirable not to have to clad the shaper material. This again points toward using something like tungsten or tantalum. Tungsten appears to be several times cheaper than tantalum but is more difficult to fabricate. Other materials considerations may further delineate the choice between these two materials for this particular application.

The shaper performs two other desirable functions: (a) it acts as a gamma shield for the irradiation samples and (b) it can act as a low energy neutron absorber which negates the use of cadmium in the filter section.

Low Energy Neutron Filter

A previous study^[2] which investigated various materials for the role of a low and intermediate energy neutron absorber concluded that ^{10}B was the best of a number of filter materials tested. Therefore in the present study ^{10}B was assumed to be the primary filter material, and some calculations were done in combination with cadmium. The purpose of the cadmium is to lower the ^{10}B depletion rate. As indicated above, the shaper can effectively perform this function and it was concluded that the additional complication introduced to fabricate the facility with cadmium incorporated was not considered worthwhile. The density of ^{10}B was assumed to be 60% of that possible theoretically and, with this kind of density, an annular region of ^{10}B 1/8 inch thick appears to be sufficient. A plot of fraction of absorptions in ^{239}Pu for various energy regions and as a function of ^{10}B density is shown in Figure 4.

Inner Coolant Water Channel Thickness

The thickness of the inner coolant water channel (see Figure 1) was varied to see if some desirable spectrum shaping could be achieved. The spectrum for various inner water channel thicknesses is shown in Figure 5. Curves are for some spatial mesh point. The coolant channel plus shaper thickness was held constant. The results generally indicate that the thickness of the inner channel should be kept to a minimum determined by heat transfer considerations.

Preliminary heat transfer calculations indicate that more cooling than depicted in Figure 1 is necessary. Undoubtedly, the final design to be used will include compromises as a result of further heat transfer and mechanical design studies. These initial feasibility studies, however, indicate that this kind of facility would be a useful, inexpensive tool. Initially, it was conceived for use as an irradiation facility for samples to be used in conjunction with the CFRMF program^[4]. However, once built it should have a broader use.

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TABLE I

COMPARISON OF ABSORPTIONS IN A ^{239}Pu SAMPLE
AND SPECTRA FOR NO SAMPLE PRESENT

Energy Range	Filtering Only		Shaping and Filtering	
	% abs in ^{239}Pu	% spectrum void	% abs in ^{239}Pu	% spectrum void
$E > 1.35 \text{ MeV}$	26	31	17	19
$9.12 \text{ keV} \leq E < 1.35 \text{ MeV}$	43	53	59	72
$1.23 \text{ keV} \leq E < 9.12 \text{ keV}$	13	7	13	7
$50 \text{ eV} \leq E < 1.23 \text{ keV}$	18	9	11	2

Fig. 1. Preliminary calculational model for fast neutron flux irradiation facility.

Fig. 2. Comparison of a filtered spectrum to a "shaped" and filtered spectrum with and without tungsten spectral shaper.

Fig. 3. Comparison of several flux spectrum shapers for a fixed reactor power.

Fig. 4. Fraction of absorption in ^{239}Pu sample for various ^{10}B filter thicknesses.

Fig. 5. Comparison of spectra for various coolant channel thicknesses between shaper and filter.

RADIOGRAPHY FACILITIES

L. G. Miller, J. P. McNeece^[1], J. E. Fluegel

Because of the large volume of TRA irradiation work for which requests are received for neutron radiography inspection, a mock-up facility was constructed to determine the feasibility of using the MTR thermal column to do neutron radiographic work. The T2H6 thermal column hole with the three graphite stringers removed produces a 1.5×10^9 thermal neutrons/cm²/sec flux at the reactor face. By moving eight feet from the reactor face to a point with 4×10^6 neutron flux, the neutrons were collimated sufficiently to perform good resolution radiographic work.

The resolution was tested by using a cadmium resolution test object. The object consisted of a 0.020 inch thick cadmium piece with several 0.035 inch diameter holes drilled through. The holes were separated by continually decreasing distances from 0.031 to 0.00055 inch. Upper series of holes vary in size down to 0.002 inch diameter. The radiograph (Figure 1), shown as a positive, was taken using a gadolinium metal screen by the direct method and shows that the resolution is better than 0.001 inch. Also shown is a neutron radiograph of a stop watch showing the intricate internal parts.

Figure 2 illustrates a comparison of an X-radiograph and a neutron radiograph of a high-intensity fission neutron source. This comparison shows the value of having both methods of nondestructive testing, because one complements the other. In the X-radiograph the pyrex bottle is seen inside an aluminum container. In the neutron radiograph the bottle is emphasized because of the boron

in the pyrex process, and in addition, the polyethylene cap and the string attached to the source are also seen in detail.

Neutron radiography has proved effective for examining clad fuel specimens: our radiographs have shown quite clearly fuel cracking, swelling, relocation and pellet interfaces. With initial and interim neutron radiographs during irradiation programs, changes in fuel capsules are followed, such as fuel growth, chipping or cracking of pellets, cladding bulges, and void formation in the clad specimens.

Neutron radiographs have been taken of many irradiated fuel capsules for fuel development programs being carried on by Battelle Northwest Laboratories, Oak Ridge National Lab, Los Alamos Scientific Lab, and the Idaho Nuclear Corporation SPERT fuel testing program. Figure 3 is a neutron radiograph of two SPERT fuel rods after test clearly showing the cracking of pellet-type fuels and the sintering of granular fuel. In addition to the high resolution available, the ability to obtain periodic neutron radiographs of the fuel capsules during reactor refueling cuts in half the time needed to obtain the required irradiation.

When working with highly radioactive materials, neutron radiography becomes the only method of examining nondestructively the interior of a sample object. The film used in X-radiography would be exposed by the high gamma fields of the radioactive subject and thus no picture would be obtained. Using indium foil as the detection device for neutron radiographs, pictures of highly radioactive objects are obtained. A large lead shield was constructed to contain the gamma radiation so that neutron radiographs of irradiated fuel capsules could be taken at this facility.

The properties that make neutron radiography attractive to nondestructive testing engineering material also show promise as an investigative tool in such areas as biology and botany. In particular, any subject that possesses various concentrations of hydrogenous material has proved to be quite interesting.

In the field of biology we have used bones, a shrew, and small fish as subjects. Because of the lack of bone structure in the shrew, an X-ray radiograph failed to show much detail; however, neutron radiographs were able to show detail of the internal organs because of air cavities in the body (see Figure 4). The major disadvantage in biological applications is the limited sample thickness that can be radiographed. Due to excessive neutron scattering in samples greater than an inch thick, one is limited to much thinner samples for high-resolution work.

Probably the most rewarding study we have undertaken has been an attempt to develop a technique that would allow one to observe plant root growth without damaging the plant. The greater concentration of water in the roots than in the soil produced excellent contrast. To date we have been able to resolve roots on the order of 0.008 inch in diameter (see Figure 5). Further work is underway in an attempt to improve resolution and increase the soil thickness that may be radiographed.

[1] Idaho Operations Office, USAEC.

Fig. 1 Neutron radiograph of a cadmium test object used to measure resolution. Also shown is a neutron radiograph of a stop watch showing the intricate internal parts.

X-RAY

NEUTRON RADIOGRAPH

Fig. 2 X-radiograph (left) and neutron radiograph (right) of a ^{252}Cf neutron source.

Fig. 3 Neutron radiograph of two Spert test fuel rods after a reactor transient test.

Fig. 4 Neutron radiograph of shrew showing clearly the nasal passages, intestinal track, and air cavities.

Fig. 5 Neutron radiograph (left) and an X-radiograph (right) of a plant transplanted in sandy loam soil. In neutron radiograph roots of the order of 0.008 inch diameter are readily visible.

II. EXPERIMENTAL TECHNIQUES

PRECISE COMPARISON AND MEASUREMENT OF GAMMA-RAY ENERGIES WITH A Ge(Li) DETECTOR I: 50-420 keV^[1]

R. C. Greenwood, R. G. Helmer, R. J. Gehrke

A Ge(Li) gamma-ray spectrometer has been used to make precise comparisons of the reported energies of a number of gamma rays that are useful for energy calibration. The results indicate that in the energy region studied, below 420 keV, this spectrometer is capable of a relative precision of about 5 eV. The energies of the 40 lines compared appear to be consistent to within this precision or their quoted uncertainties (ie, 1 to 10 eV). Comparisons of energies reported relative to the X-ray energy scale (ie, the $K\alpha_1$ X-ray of tungsten) and to the $m\alpha c^2$ scale (ie, 511.006 keV) indicate that these scales are consistent to within (17 ± 28) ppm. Energies were also measured for gamma rays emitted in the decay of the isotopes ^{109}Cd , ^{133}Ba , ^{153}Gd , ^{57}Co , ^{75}Se , and ^{113}Sn .

[1] Accepted for publication in Nuclear Instruments and Methods.

PRECISION COMPARISON AND MEASUREMENT OF GAMMA-RAY ENERGIES WITH A Ge(Li) DETECTOR II: 420-1340 keV

R. G. Helmer, R. C. Greenwood, R. J. Gehrke

The work reported previously^[1] on the precision measurement of gamma-ray energies has been extended to the energy range of 420 to 1340 keV. In the first report the major emphasis was on the comparison of existing energies to determine their consistency, while in this one it is on the more precise measurement of energies. In this study four Ge(Li) detectors have been used with three multichannel analyzers. The comparison of these results will allow us to determine the validity of some of the techniques used in the data analysis.

In the energy region of interest only a few directly measured calibration energies exist of the desired accuracy. These are listed in Table I. To augment this set, we have used as calibrations several lines whose energies are equal to the sum of two lower energy transitions. Since the previous report^[1] indicates that the energies of the latter transitions can be determined with good accuracy, these calibration lines may be more accurate than those that are directly measured. An example of this method is given in Table II for the 604- and 612-keV lines from ^{192}Ir .

Additional calibration energies are generated by the measurement of small energy differences with the Ge(Li) detector system. This technique is illustrated in Table II where the 588-604 and 588-612 differences in ^{192}Ir have been used to obtain an accurate value for the 588-keV transition. Then from other energy sums and a measured 884-879 energy difference, the remaining ^{192}Ir and ^{160}Tb energies shown can be computed. In this method care must be taken to propagate properly both the errors in the energies due to the measurements and those due to the uncertainty in the reference energies.

From the calibration lines in Tables I and II, several other transition energies have been measured. Preliminary values of these energies are given in Table III. These measurements are continuing.

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TABLE I

DIRECTLY MEASURED CALIBRATION ENERGIES

<u>Parent Isotope</u>	<u>Reference</u>	<u>Energy (keV)</u>
¹⁹⁸ Au	2	411.795 ± 0.007
²⁰⁸ Pb	2	583.173 ± 0.013
¹³⁷ Cs	6	661.633 ± 0.012
¹⁹⁸ Au	7	675.378 ± 0.016
¹⁹⁸ Au	7	1087.692 ± 0.025
⁶⁰ Co	2	1173.23 ± 0.04
⁶⁰ Co	2	1332.48 ± 0.05

TABLE II
ENERGIES OF SOME ^{192}Ir AND ^{160}Tb GAMMA-RAY TRANSITIONS

Parent Isotope		
Energy (keV)	Method	Value (keV)
^{192}Ir		
295	Ref. 1	295.948 ± 0.006
308	Ref. 1	308.442 ± 0.007
316	Ref. 1	316.498 ± 0.006
468	Average of Ref. 2 ^[a] , 3 ^[b] , 4	468.063 ± 0.009
588	Average of Ref. 2 and measured (604-588) and (612-588) differences	588.567 ± 0.011
604	Average of Ref. 2, 3, and sum of 295 + 308	604.395 ± 0.011
612	Average of Ref. 2 and sum of 295 + 316	612.445 ± 0.011
884	Sum of 588 and 295	884.515 ± 0.016
^{160}Tb		
879	From measured (884-879) differences	879.356 ± 0.020
962	Sum 879 + (298-215) ^[c]	962.291 ± 0.022
966	Sum 879 + 86 ^[c]	966.143 ± 0.022
1177	Sum 879 + 298 ^[c]	1177.934 ± 0.023
1270	Sum 1177 + 93 ^[c]	1271.852 ± 0.025

[a] Only internal-conversion data from this reference.

[b] Published values were adjusted to give best agreement with the above energies for 295- and 316-keV transitions.

[c] See Reference 5.

TABLE III
MEASURED TRANSITION ENERGIES^[a]

Parent Isotope	Energy (keV)	Parent Isotope	Energy (keV)
⁷ Be	477.597	⁵⁹ Fe	1099.23
⁸⁵ Sr	514.001	⁶⁵ Zn	1115.52
²⁰⁷ Bi	569.684 ± 0.014	⁴⁶ Sc	1120.53
⁵⁴ Mn	834.828 ± 0.03	¹⁸² Ta	1121.27
⁴⁶ Sc	889.24	¹⁸² Ta	1189.05
⁸⁸ Y	898.005	²² Na	1274.52
²⁰⁷ Bi	1063.65 ± 0.03	⁵⁹ Fe	1291.57

[a] All values are preliminary. Where not shown the uncertainties are about 30 eV at 500 keV and 50 eV at 1 MeV for the data analyzed to date.

AUTOMATIC PEAK FINDING AND INTENSITY DETERMINATIONS

W. W. Black, A. L. Connelly

A new method has been found which will permit the automatic location of structure, such as peaks, in experimental data. The method has application in the automatic analysis of experimental data on computers, in preliminary searches of data for later inspection, in the areas of information retrieval, and several other possible applications. The method makes use of the cross-correlation function. In general the method is accomplished by making use of the fact that one form will tend to select itself from a more complicated form in the cross-correlation operation. Complete details of the computations are given in Reference 1.

Figure 1 shows the results of the application of this method. The data shown there were obtained with a lithium-drifted germanium detector and the spectrum from a source containing fission products. The dark points are the original data, and the spectrum plotted with open circles are the results of the cross-correlation operation and are referred to as the correlation spectrum. The correlation spectrum has the characteristic of identically returning to zero in those portions of the spectrum where there are no peaks. This characteristic makes it very easy to locate the positions of peaks with a digital computer.

To date the method has been successfully applied to data obtained with lithium-drifted germanium detectors, lithium-drifted silicon detectors, sodium iodide detectors, and neutron diffraction spectra from time-of-flight analyzers.

After the success that was obtained with the automatic locating of peaks, a search was started for a method of determining automatically and simultaneously the areas under the peaks. It would be highly desirable if the correlation spectrum could also be used to obtain the areas under the peaks in the original spectrum. If this can be accomplished the difficult details of locating backgrounds could be eliminated. Therefore, investigations were undertaken to determine if a relationship could be found between the areas under the correlation peaks and those of the corresponding photopeaks in the original data. Although a functional relationship cannot be obtained in which the desired variables can be extracted, it was found that a small table of parameters can be established from which the areas of the original photopeaks may be determined. Complete details of these computations may be found in Reference 2. The original goal was to be able to determine the areas under photopeaks within 20% of values found by other more sophisticated means such as least squares to Gaussian peaks. This goal can be easily accomplished with one important stipulation. A certain number of channels must occur between photopeaks before this accuracy is attained. The number of channels may be specified in terms of full widths at half maximum of the photopeaks. It has been found that a distance

Fig. 1 Results of the correlation method for automatic location of peaks in experimental data. The darkened points are the original data and the open points are the correlation spectrum.

of approximately six full widths must occur between photopeaks before their areas are accurately determined. As would be expected, the accuracy with which the areas may be determined depends in a direct manner on the peak-to-background ratio: the ratio of the peak height to the underlying background. Figure 2 vividly demonstrates this but shows that even down to very small peak-to-background ratios the areas can be determined within 20% of their values as determined by the Gaussian fitting method.

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Fig. 2 A comparison of the percentage difference between area found by the present method versus Gaussian fitting plotted against the peak-to-background ratios.

X-RAY SPECTROSCOPY USING Ge(Li) AND Si(Li) DETECTORS

R. J. Gehrke

Efficiency Measurements

Work is continuing in the development of the techniques used to calibrate the efficiency of Ge(Li) and Si(Li) detectors from 5 to 125 keV. The following radioactive sources have been used in the calibration of solid state X-ray detectors below 32 keV: ⁵¹Cr, ⁵⁴Mn, ⁶⁵Zn, ⁵⁷Co, ¹⁰⁹Pd, and ¹³⁷Cs. With the exception of ⁵⁷Co all these sources decay by a single gamma-ray transition and the ratios of gamma ray to K X-rays have been accurately determined[1-8]. These ratios are listed in Table I. The gamma rays are calibrated in the usual manner using a 3 x 3 inch NaI(Tl) scintillation spectrometer[9]. Above

32 keV the following gamma-ray sources have been used: ^{210}Pb , ^{153}Sm , ^{109}Cd , ^{241}Am , and ^{57}Co . The 122- and 136-keV gamma rays from ^{57}Co are summed together giving an efficiency point at 123.5 keV. Figure 1 illustrates a typical Si(Li) efficiency curve.

The efficiency curve of a 30-mm² x 3-mm Si(Li) detector so obtained has been used to measure the relative intensities of K X-rays and gamma rays emitted from radioactive nuclei. As an example, a spectrum of the decay of 57-min ^{103m}Rh is shown in Figure 2. In this case the K conversion coefficient (α_K) was determined from the K X-ray: gamma-ray ratio and the K fluorescence yield (ω_K).

In progress is the calibration of a newly purchased Kevex Si(Li) detector. The efficiency values are also being remeasured for the TMC Si(Li) detector. When this has been completed, the K X-ray: γ -ray ratio of several low-energy, monoenergetic, isomeric transitions which are highly converted and have short half-lives (≤ 15 min) will be measured and the α_K will be determined.

X-Ray Fluorescence Spectroscopy

Chromium, manganese, and iron are three of the more difficult elements to analyze using the techniques of X-ray fluorescence analysis. One source which has shown some promise is ^{238}Pu . However, its long half-life (89 yr) causing low specific activity and its potential hazard to health have discouraged its large scale use. A source developed at this laboratory to replace ^{238}Pu is 18-yr ^{244}Cm , but its advantages over ^{238}Pu are slight. To illustrate the high degree of sensitivity that potentially can be attained, a source of 11-day ^{71}Ge was produced in the MTR reactor and used for primary excitation on calibrated iron in oil samples. A plot of counts/min of Fe X-ray versus Fe concentration (ppm) is given in Figure 3. The minimum concentration observed was 25 ppm of Fe in oil. I am hopeful that with improved geometry, more intense sources and a better Si(Li) detector that lower concentrations can be determined. Other sources for the X-ray fluorescence of chromium, manganese, and iron are being investigated.

X-ray fluorescence spectroscopy has also been found useful in sorting fuel pins clad with 308 or 316 stainless steel. The fuel pins could be sorted by simply looking at the X-ray peak ratios on the oscilloscope display. A sorting rate of one per minute was attained.

A number of ore samples have been studied this past year using X-ray fluorescence. Elements ranging from sulphur to thorium have been identified. The techniques of X-ray fluorescence spectroscopy have been used to complement those of neutron activation analysis to determine the bulk and trace elements present in ores from the Purcell Sills and selected galena samples from mines in northern Idaho. The results of these analyses will be presented in a future report.

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TABLE I

K X-RAY/ γ -RAY INTENSITY RATIOS USED TO OBTAIN LOW ENERGY PORTION OF $\text{Si}(\text{Li})$ PEAK EFFICIENCY CURVE

Isotope	Average energy of X-rays in keV	X-ray / γ -ray	References
^{51}Cr	5.0	2.52	1, 3
^{54}Mn	5.5	0.257	1, 3
^{65}Zn	8.2	0.798	1, 3
^{109}Pd	22.4	9.27	4, 5, 6, 8
^{137}Cs	32.6	0.0778	7, 8

Fig. 1 Absolute photopeak efficiency curve of a 30 mm² x 3 mm Si(Li) detector.

Fig. 2 Si(Li) spectrum of the decay of 57-min 103^mRh.

Fig. 3 Curve of counting rate for Fe X-rays as a function of iron concentration in oil samples.

ENHANCING CONTRAST OF NEUTRON RADIOGRAPHS BY ENERGY TAILORING OF BEAMS

L. G. Miller, T. Watanabe

For many inspection problems, it is necessary to examine and measure an object contained within a variety of other materials. The reduction of the image contrast for the unwanted materials and the image contrast enhancement of the desired object increases the information which can be obtained from the neutron radiograph.

Development of a technique for contrast enhancement of the image on a neutron radiograph is being carried out using the high neutron flux beams available from the MTR. Cadmium as a beam filter is used for the enhancement in some radiographs, but it is only the first step toward attaining optimum contrast for specific materials. Optimum contrast may be achieved when the neutron energy of the beam and the energy response of the detector correspond to a high neutron cross section region of the object.

Many rare earth and other metal foils are being used as filters to remove many of the unwanted energy neutrons from the beams. Contrast enhancement has been achieved on several materials with this technique. As an example, the contrast as measured with an optical densitometer of a tantalum object was increased a factor of ten over a thermal neutron radiograph. This is shown in Figure 1. This technique can be used to enhance the contrast of a material in the presence of other high cross section materials. Nonuniformities of ^{240}Pu and ^{241}Pu in the presence of large amounts of ^{239}Pu are possible.

Although beam tailoring with filters does not produce the mono-energetic beam available from a crystal spectrometer, the technique with filters produces a beam not limited by the inherent characteristics of the diffracted beam, ie, nonuniformity, small size, poor diffraction efficiency, and limited energy range.

Fig. 1 Two neutron radiographs of a tantalum plate. The left radiograph was made with a normal thermalized neutron beam. The right radiograph was made with a neutron beam tailored to enhance the contrast for tantalum.

NEUTRON RADIOGRAPHIC APPLICATION TO FUEL DEVELOPMENT

L. G. Miller

In the area of fuel development, neutron radiography has been successfully used to study the construction of burnable poison plates made from B_4C in an aluminum matrix. To be effective the boron must be evenly distributed in the aluminum. Radiographs of samples (Figure 1) fabricated in the cold metallurgy

Fig. 1 Neutron radiograph of boron poisoned aluminum test plates showing the boron clumping of the poison.

laboratory at TRA showed clumping of boron. Armed with this information, the fabrication process was modified to produce more suitable plates. The convenient availability of the neutron radiography facility here at MTR made this modification possible with no long delay for shipping to some other facility. Testing of possible faulty fuel plates as they become available from the fuel fabrication is planned.

EPITHERMAL NEUTRON RADIOGRAPHY

L. G. Miller

For many materials such as boron, uranium, plutonium fuels, cadmium, etc, the thermal neutron cross section is sufficiently high as to prevent penetration of the object for inspection purposes. In such cases, a higher energy neutron beam with high intensity must be used. Higher energy neutrons penetrate these objects easier, and high resolution neutron radiographs are being made on a routine basis.

Neutron radiographs are being taken at MTR as a quality control measure of the FFTF simulated fuel rods. After several failures in the sodium test loop at BNW, rods were neutron radiographed elsewhere with thermal neutrons in an attempt to determine nonuniformities in the insulating materials. Since these rods are insulated with a material having a very high thermal neutron cross section, thermal neutrons were not able to penetrate the boron nitride insulator and no radiographs were obtained.

The rods were then sent to the MTR neutron radiographic facility where several techniques were tried. Radiographs with thermal neutrons were not successful, but high quality neutron radiographs of the simulated fuel rods were obtained using scanning techniques and the highest flux fast neutron beam hole available at the MTR. Success required a high flux of collimated fast neutrons to penetrate the boron nitride layer and a precision scanning system to cover large rod segments. Several nonuniformities and breaks in the boron nitride insulated rods were detected, as is apparent in Figure 1.

Techniques used at the MTR now permit routine examination of these rods with the epithermal beam when the reactor is operating. A complete set of neutron radiographs of five simulated fuel rods can be made in less than

Fig. 1 Neutron radiograph of boron nitride insulated rods showing several nonuniformities and breaks in the boron nitride insulator.

one hour. All the rods as they are fabricated are now being radiographed at the MTR. Pictures to date have shown variations in the boron density due to packing techniques in the manufacture of the rods and the fused sections in the rods after failure.

PWR simulated fuel rods similar to the FFTF rods mentioned above are now being tested to determine the growth rates of the boron nitride insulator and the Zircaloy cladding.

SPECTRUM DETERMINATIONS IN THE CFRMF

D. A. Millsap, J. W. Rogers

The interpretation of reactivity or activation measurements made in the CFRMF require that good agreement be obtained between the experiment and calculation. Agreement between calculated and experimentally determined flux spectra would give considerable confidence in both calculational procedures and the input cross section sets used in the reactor calculation. Some work of this type has already been done for the ARMF-I and -II reactors[1]. Indeed, correlations between calculations and experiments will be even more important for integral measurements in fast devices since there is no common reference spectrum to which experiment results can be related such as the Maxwellian or $1/E$ fluxes characteristic of the thermal reactors.

Calculational Results

The calculations made for the ARMF-I and -II reactors used diffusion theory, and in general these proved to be adequate. However, due to the presence of the boron liner, the large ^{238}U block itself, and the ^{10}B and ^{235}U sleeves in the CFRMF, it seemed entirely possible that diffusion theory calculations might be totally inadequate. In order to examine this possibility, eigenvalue and flux calculations were made using both diffusion and transport codes. The diffusion theory calculation was made using MONA[2]. The MONA calculation used 26 groups, 23 regions, and 124 mesh points. The transport theory calculation used SCAMP 2B[3], a double-precision cylindrical version of the SCAMP series of one-dimensional S_n codes. This was an S_6 calculation, with the same 26-group structure as the MONA calculation, but had 29 regions and 161 mesh points. These modifications were made in an attempt to obtain a more realistic flux solution in the sensitive regions. Specifically, for the transport calculation, two regions were added to the ^{10}B annulus and the number of mesh points increased from 8 to 30. One additional region was added to the ^{238}U block with a slight change in the mesh spacing; two regions were added to the boron liner and the number of mesh points increased from 8 to 30. The number of regions in the water reflector was increased by one and the mesh points increased from 18 to 25. Some mesh points were eliminated in regions where calculated flux gradients were low in MONA to save computer time. The SCAMP problem was not run to the same level of convergence but is close enough to warrant comparisons. Details of group structure, region locations, and mesh spacings are shown in Tables I and II. The cross section sets used for the two problems were essentially identical except for minor changes in the group 25 cross sections of certain elements and, of course, the addition of the P-1 scattering cross sections required for the transport calculations. The epithermal cross sections were obtained from PHROG[4] and

TABLE I
ENERGY AND LETHARGY GROUP STRUCTURE

<u>Group Number</u>	<u>Upper Limit Energy (eV)</u>	<u>Lower Limit Lethargy</u>	<u>Group Width Lethargy</u>
1	10.0×10^6	0.0	0.5
2	3.68×10^6	0.5	0.5
3	6.07×10^6	1.0	0.5
4	2.23×10^6	1.5	0.5
5	1.35×10^5	2.0	0.5
6	8.21×10^5	2.5	6.5
7	4.98×10^5	3.0	0.5
8	3.02×10^5	3.5	0.5
9	1.83×10^5	4.0	0.5
10	1.11×10^5	4.5	0.5
11	6.74×10^4	5.0	0.5
12	4.09×10^4	5.5	0.5
13	2.48×10^4	6.0	0.5
14	1.50×10^4	6.5	0.5
15	9.12×10^3	7.0	0.5
16	5.53×10^3	7.5	0.75
17	2.61×10^3	8.25	0.75
18	1.23×10^3	9.00	0.75
19	583.0	9.75	0.75
20	275.0	10.50	0.75
21	130.0	11.25	1.00
22	47.9	12.25	1.00
23	17.6	13.25	1.00
24	6.48	14.25	1.00
25	2.38	15.25	1.50
26	5.32	16.75	∞

TABLE II

STATISTICS FOR CFRMF CORE REPRESENTATIONS

Region Number		Material Represented	Outer Radius of Region (cm)		Number of Mesh Points in Region	
MONA	SCAMP	MONA AND SCAMP	MONA	SCAMP ^[a]	MONA	SCAMP
1	1	Sample or Void	1.1049		5	5
2	2	Sample Holder	1.91643		4	4
3	3	Dry Tube	2.21344		2	2
4	4	²³⁵ U Sleeve	2.21996		3	3
5	5	Cladding	2.29997		2	2
6	{ 6 7 8	¹⁰ B Sleeve	2.65049	{ 2.34997 2.60049 2.65049	8	{ 10 10 10
7	9	Cladding	2.69748		1	2
8	{ 10 11	²³⁸ U Block	8.19127	{ 2.92639 8.19127	24	{ 5 16
9	{ 12 13 14	Boral Liner	8.93931	{ 8.24127 8.88931 8.93931	8	{ 10 10 10
10	15	Cladding, water	9.40362		4	6
11	16	Fuel Element Side Plates	10.21748		4	4
12	17	Fuel Elements (Nominal 188 g)	16.21748		4	4
13	18	Fuel Elements (Nominal 205 g)	18.37780		5	2
14	19	Fuel Element Side Plates	19.36502		2	2
15	20	Fuel Elements (Nominal 179 g)	23.03407		7	2
16	21	Control rod followers	23.62273		2	2
17	22	Fuel Elements (Nominal 173 g)	25.59806		4	2
18	23	Fuel Elements (Nominal 177 g)	25.97511		1	1
19	24	Fuel Elements (Nominal 188 g)	26.71325		2	1
20	25	Fuel Element Side Plates	27.30449		2	1
21	26	Fuel Elements (Nominal 131 g)	28.69351		3	1
22	{ 27 28	Water reflector	35.0	{ 31.0 41.0	8	{ 10 10
23	29		50.0	51.0	10	5

[a] Where regions were split.

thermal cross sections from INCITE[5]. The cross section libraries used with these B-1 cross section averaging codes were not based on the ENDF/B cross sections at the time the calculations were made.

The results of the center-line flux calculations for the two problems are plotted in Figure 1. As can be seen, the flux shapes for the two calculations are nearly identical, at least in their main features. Relative differences are greatest below 5.53 keV (group 16 or greater), that is, primarily in the resonance region. Through group 15 (10 MeV to 5.53 keV) the differences are never greater than 4.26%. The eigenvalue from MONA was 1.0431 and from SCAMP, 1.0496. Again the agreement is good. From these results one can conclude that many preliminary investigations can be made for this type of system using diffusion theory codes.

Subsequent to the above calculations, ENDF/B-based cross section sets became available for many of the standard isotopes. The diffusion theory calculation was, therefore, repeated using these new cross sections. The resulting center-line flux spectrum is plotted in Figure 2. As can be seen from the graph the flux spectrum is considerably harder than that obtained from the previous calculations. The first calculations (both diffusion and transport) indicated that the flux in the center of the CFRMF would peak at group 9, 111 to 183 keV. The later calculation shows a peak at group 7 or between 302 and 498 keV. Also shown in Figure 2 is the spectrum as measured by a recoil-proton spectrometer. The spectrum has been coalesced from that of Figure 3 to provide a more suitable comparison. As is obvious, there are significant discrepancies between the two. This must result primarily from inaccuracies in the ^{238}U cross sections, but those used for ^{235}U , ^{10}B , and stainless may also be exerting some influence on the calculated fluxes. Most troublesome is the failure to predict even a hint of the "ripples" evident in the measured spectrum (shown much better in the uncoalesced spectrum of Figure 3). Additional modifications of cross sections may be required to obtain agreement. It is felt that the good correlation between the diffusion and transport theory fluxes indicates a lack of any gross transport effects in the spectrum shape at the reactor measurement position.

Experimental Results

The CFRMF neutron spectrum between 3 keV and 2.5 MeV was measured with a proton-recoil spectrometer. This experiment was performed with the reactor critical at 5×10^{-3} watts of power. The measurement was made in two-parameter mode with gamma discrimination by pulse shape analysis based on the rate of rise of the pulse.

The data were collected in a 4096 two-parameter pulse-height analyzer using a 64×64 channel array. The subtraction of the gamma-ray events was performed using a digital data processing computer (PDP-9)[6]. The final analysis of the proton spectrum was made using the analysis code of Bennett, Gold, and Olson[7]. Figure 3 shows a comparison of the spectrum with and without the energy-dependent response function corrections for finite detectors. Two detectors were used in this measurement, one filled with hydrogen (200 cm mercury) and the other with methane (380 cm mercury). The hydrogen-filled detector was used from 3 keV to about 250 keV and the methane detector from about 100 keV to 2.5 MeV. Both detectors were identical in dimensions and construction, being 0.9375 inch in diameter and 4.5 inches long with sensitive volumes of 29.73 cm^3 . Figure 4 is a photograph of the detector.

The spectrometer and each detector were calibrated using the $^{14}\text{N}(n,p)^{14}\text{C}$ reaction and the MTR neutron beams[8]. Good overlap of the hydrogen and methane data was obtained but depended on how the data were analyzed with regard to the energy dependence of the energy loss per ion pair in methane[3] and how the two-parameter rate-of-rise distributions were treated[1].

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Fig. 1 Comparison of transport and diffusion theory flux calculations.

Fig. 2 CFRMF spectra.

Fig. 3 Comparison of CFRMF neutron spectrum with and without corrections for finite size detector wall and end effects.

Fig. 1 Photograph of proportional counter.

PROTON-RECOIL MEASUREMENTS IN THE MTR 2 keV SCANDIUM-FILTERED NEUTRON BEAM

J. W. Rogers, O. D. Simpson

Neutron spectrum measurements have been made in the Materials Testing Reactor (MTR) 2 keV scandium-filtered neutron beam [1] with a proton-recoil spectrometer [2]. The 2 keV beam has been developed for use where an intense source of near monoenergetic neutrons are needed. The proton-recoil spectrometer has proven capable and useful for investigation of the neutron energy spectrum from this beam. The beam has conversely proven useful in the calibration and investigation of proton-recoil spectrometers [2].

The scandium-filtered beam has been discussed by Simpson and Miller [1] where neutron spectrum measurements were made within the limits of the time-of-flight spectrometer. These measurements went up to about 10 keV and accurately located the primary neutron transmission at 1.95 ± 0.05 keV and a smaller peak at 8 keV. Since it was desirable to know more about the energy spectrum at higher energies, a proton-recoil spectrometer was placed in the beam to measure the spectrum.

The spectrometer is the same as described in Reference 2. Two counters were used in these measurements, both being of identical geometry and dimensions. They are coaxial cylinders of stainless steel 2.38 cm in diameter and 11.43 cm in length. The sensitive section is 7.62 cm long with a volume of 29.73 cm³ and a 0.0025-cm diameter center wire. Figure 1 is a photograph of one of these detectors. Counter "H₂" was filled with 200-cm mercury pressure of hydrogen, 20-cm pressure of methane, and 10-cm mercury pressure of nitrogen. Counter "CH₄" was filled with 380-cm mercury pressure of methane and 19-cm mercury pressure of nitrogen. These counters were fabricated and filled commercially [3].

The spectrometer was calibrated using the thermal neutron induced $N^{14}(n,p)C^{14}$ reaction and the known energies in the iron and scandium filtered beams as discussed in Reference 2. The detector resolutions were 5% for " H_2 " and 8% for " CH_4 " as observed with the $N^{14}(n,p)C^{14}$ reaction.

The experimental setup is shown in Reference 2 where all measurements were made by placing the counter in the beam with its axis perpendicular to the beam axis about 90 cm from the beam opening and the beam centered on the sensitive volume. It was necessary to collimate the beam to 0.635 cm to make counting rates acceptable. Checks were made to assure that the measured spectra were not affected by back-scattered neutrons or gamma rays. To check for back-scattered neutrons the counters were placed to the sides of the beam and no counting rates could be detected. The gamma background was known to be very low from standard dosimetry measurements but another check was made by taking rate of rise pulse shape discrimination measurements. Figure 2 is a 0.08 keV wide rate of rise distribution at 2 keV in the beam with and without the presence of a 100 mR/hr ^{60}Co gamma source. Rate of rise distributions were taken throughout the energy range of the spectrum measurements with both counters and no significant gamma backgrounds could be found. This allowed single parameter measurements to be used eliminating the handling of two-parameter data. Consequently, a spectrum could be measured between 1 keV and 1 MeV in about six hours of counting time.

The proton-recoil data were unfolded using a computer code by Bennett, Gold, and Olson[4] but foregoing the W cutoff (energy loss per ion pair) corrections and energy-dependent wall and edge-effect corrections. Figure 3 is a proton spectrum from the scandium beam and is typical of those measured in this experiment. The y axis is counts per keV per hydrogen atom per second times $E, E[P(E)]$, where E is the energy of each proton data point. Figure 4 is the neutron spectrum emerging from a 106.7 cm thick scandium filtered beam showing transmission peaks where the cross section dips due to potential and resonance scattering interference. This illustrates the discernment that proton-recoil measurements can give where cross section measurements have not been made. There is considerable fluctuation in the cross section of scandium in the keV region and Table I lists the major transmission lines, "windows", measured between 1 keV and 1 MeV by this method.

Figure 5 is the neutron spectrum resulting after inserting a 1.43 cm slab of titanium in the beam. Here the effects of the titanium scattering resonances between 3 and 300 keV can be observed. The scandium-only spectrum is also included for comparison.

Figure 6 is the result of placing 5.08 cm of aluminum in the beam. The effect on the spectrum above 25 keV is as expected from knowledge of the aluminum cross sections.

Another measurement of interest was the spectrum with a cerium-manganese filter[1] used to eliminate the 2 keV component of the beam. Figure 7 shows the spectrum with 2.54 cm Mn powder, 1.59 cm Ce metal, and 1.43 cm of Ti in the beam. Comparison of the Ce-Mn filter in and filter out gave a transmission factor (J) for the higher energy neutrons transmitted by the filter of about 0.44 which is in agreement with that found by Simpson and Miller[1].

All of the measurements discussed thus far were made using counter "H₂". Seeing that the flux was down by three orders of magnitude at 1 MeV it was felt that a measurement with a detector of greater stopping power should be used to see if this was correct or if detector cutoff due to stopping power was being seen. Detector "CH₄" was calibrated and a measurement made between 2 MeV and 70 keV. Figure 8 is the Ti filtered spectrum taken with detector "CH₄" and is compared with that taken with detector "H₂". Good agreement was obtained above 300 keV with small differences in magnitude due to wall effects showing in counter "H₂". Below 300 keV the energy loss per ion pair effects in methane^[2] can be seen in counter "CH₄". These results show that the hydrogen counter gives satisfactory results over this energy range for locating the "windows".

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Fig. 1 Photograph of proportional counter.

Fig. 2 Pulse height rate of rise distribution at 2 keV in scandium beam taken with hydrogen counter.

TABLE I

WINDOW ENERGIES IN SCANDIUM
FROM PROTON-RECOIL SPECTROMETER
MEASUREMENTS^[2]

<u>E (keV)</u>	<u>E (keV)</u>
181 ± 2.0	1.96 ± 0.02
214 ± 2.0	7.62 ± 0.08
248 ± 2.5	16.1 ± 0.16
291 ± 3	29.5 ± 0.30
328 ± 3	40.2 ± 0.50
377 ± 4	48.0 ± 0.50
450 ± 5	55.0 ± 0.60
507 ± 5	65.5 ± 0.70
715 ± 10	76.4 ± 1.0
	131 ± 2.0

Fig. 3 Raw proton spectrum from scandium beam taken with hydrogen counter.

Fig. 4 Neutron spectrum from scandium beam taken with hydrogen counter.

Fig. 5 Neutron spectrum from scandium beam and neutron spectrum from scandium beam with 1.43 cm of titanium taken with hydrogen counter.

Fig. 6 Neutron spectrum from scandium beam and neutron spectrum from scandium beam with 5.08 cm of aluminum taken with hydrogen counter.

Fig. 7 Neutron spectrum from scandium beam and neutron spectrum from scandium beam with 1.43 cm of titanium, 2.54 cm of manganese powder, and 1.59 cm of cerium metal taken with hydrogen counter.

Fig. 8 Neutron spectrum from scandium beam with 1.43 cm of titanium taken with hydrogen counter and a portion of the same spectrum taken with a methane counter.

PROTON-RECOIL CALIBRATION STUDIES IN THE MTR NEUTRON BEAMS WITH HYDROGEN AND METHANE DETECTORS

J. W. Rogers

Proton-recoil spectrometers are becoming the standard differential flux measuring device for the LMFBR program. Measurements of the neutron spectra from the MTR scandium- and iron-filtered beams[1,2] have been made using a proton-recoil spectrometer with hydrogen- and methane-filled detectors. These spectra are compared to time-of-flight spectrometer measurements which have accurately located the discrete energy transmission "windows" in iron[3] up to about 1 MeV and in scandium[1] to about 10 keV. Through this comparison one can detect any variations in energy that may occur because of miscalibration of the proton-recoil counters.

The proton-recoil detectors used are, first, a spherical detector[4], 3.94 cm diameter with a 32 cm³ sensitive volume filled with 200 cm Hg pressure of methane and, second, a coaxial cylinder detector, 2.38 cm diameter, 11.43 cm in length, 29.73 cm³ sensitive volume filled with 200 cm Hg of hydrogen. Each detector has a 0.001 inch center electrode along the axis and a small amount of nitrogen for resolution and calibration purposes. The resolution of the detectors is 5% as observed from the $^{14}\text{N}(\text{n},\text{p})^{14}\text{C}$ reaction peak.

The spectrometer diagram is shown in Reference 5 and is similar to that of Bennett[6] except for modifications of the preamplifier and linear amplifier. The preamplifier is a charge sensitive, high gain, low-noise device connected directly to the detector for optimum performance. The linear amplifier is a delay line clipped type for use at high counting rates.

The experimental setups are shown in Figures 1 and 2 where the detector is positioned with its axis perpendicular to the beam and the sensitive volume is centered in each beam. The iron beam is collimated to 8.89 cm diameter and the scandium to 0.635 cm. One unique situation of this setup is the available thermal column in close proximity to the beams which allow calibration by $^{14}\text{N}(\text{n},\text{p})^{14}\text{C}$ reaction and beam exposure to be done without shutdown and moving of any equipment other than the detector.

An extremely advantageous feature of these beams is the very low gamma-ray background present in them. It is, in fact, so low that pulse shape discrimination against gamma rays is not necessary in the measurements. Figure 3 shows a 0.08 keV wide rate of rise distribution at 2 keV in the scandium beam with and without the presence of a 100 mR/hr ^{60}Co gamma source. This type of measurement was made at many energies throughout the spectrum of each beam, and no significant gamma backgrounds were found. Other attempts to measure the gamma background resulted in values of about 1 mR/hr. This feature makes it possible to take single parameter (energy) measurements thus eliminating the handling of two-parameter (rate of rise versus energy) data.

The spectrometer was first calibrated using the $^{14}\text{N}(\text{n},\text{p})^{14}\text{C}$ reaction. It was necessary to know the gas amplification much higher than could be measured due to saturation limitations; therefore, an extrapolation was made by the Diethorn relation[7] as shown in Figure 4. The accuracy of this extrapolation could then be tested by observing the distinct structure of the measured proton

spectra. Using these extrapolated gas amplifications unmatching sets of overlapping proton spectra at the different gas amplifications resulted in the extrapolated region (above 2800 V). Using sufficient (double) overlap and assuming all parameters affecting the gas amplification to be constant, the effective gas amplifications in the extrapolated region were found by fitting together the overlapping proton spectra; these are also shown in Figure 4. The proton spectra for the two beams are shown in Figures 5 and 6 using the effective gas amplifications. These data were taken with the spherical methane-filled detector. The values plotted are proton counts per kilovolt per hydrogen atom per second times E where E is the energy of each data point.

The proton data were unfolded by differentiation of the continuous spectrum using appropriate analysis considerations of the data and detectors[8]. The resulting neutron spectra are shown in Figures 7 and 8.

By comparing these results with the known energies of the "windows" in the energy region where the specific average energy loss per ion pair, W , is considered to have undistinguishable variations[9] and wall and edge effects are insignificant, agreement was obtained by assuming "effective ionization" for the $^{14}\text{N}(n,p)^{14}\text{C}$ reaction of 585 keV. It is known that the kinetic energy of the proton from the nitrogen reaction is 585 keV[10] and nuclear emulsion experiments[11] do not find detectable evidence of the ^{14}C track. Thus, it may be that the ^{14}C does not cause detectable ionization in proportional counters. Another explanation of the loss of total energy may be some electronics bias in the linear channel.

Table I is a listing of the "window" energies that are dominant and resolvable up to about 500 keV along with the energies at which they were found using the spectrometer. Also tabulated are the ratios of the proton-recoil measured energies to the "windows" energies. Figure 9 is a plot of this ratio at each "window" showing the manner in which the proton-recoil measured energies vary over this energy range. Figures 10 and 11 are comparisons of neutron spectra measured with the hydrogen and methane detectors showing how the methane spectra differ from the hydrogen measured spectra.

Assuming that all parameters of the gas amplification are constant, then this is a measurement of the effect of the energy dependance of W over this energy range. These measurements indicate no change in hydrogen and an increase as energy decreases in methane. The reciprocal of this ratio defines the manner in which W changes as a function of energy.

Errors on these results are very difficult to assess and only a "guesstimate" is attempted. All possible efforts were made to assure that problems in the accumulation of data as discussed by Wilkinson[12] were of no effect. The data-taking system did not drift more than 0.5% throughout the measurements. The errors on the "window" energies are well established, and the locations of the peaks in the measured data were easily established due to their Gaussian (resolution broadened) nature. The errors on the unfolded neutron spectra have been discussed by Bennett, Gold, and Olson[7] and will not be discussed further. Where the proton data were fitted together, the criterion used was that the overlapping data agree within a $\pm 1\%$ band. The systematic errors involved would be those in the measurement of electronic parameters and values of physical constants used. The random errors are expected to be $\pm 2\%$.

These measurements reveal an alternative method of calibration of proton-recoil spectrometers that can either complement or replace the internal source technique. If only the beams are used then it would be possible to omit the nitrogen gas in the detectors consequently reducing the heavy nuclei recoils. The advantage of this method is the discrete energy standards located throughout much of the applicable range of proton-recoil spectrometry. The analysis of these measurements has revealed the manner in which W for methane is energy dependent at lower energies than have been previously reported.

The uniqueness of these beam facilities lead to further proton-recoil studies in relation to mixtures of gamma and neutron irradiation, counter geometry effects, and resolution studies which are being pursued. These results should allow the application of methane counters at lower energies and help resolve some of the problems encountered when hydrogen and methane counters are used in conjunction for spectrum measurements. This use of scandium points to the need of differential cross section measurements on the element up to 1 MeV. However, it is seen that proton-recoil measurements have enough accuracy and resolution to give valuable insight to the structure of the cross section in this range.

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TABLE I

COMPARISON OF TIME-OF-FLIGHT AND PROTON-RECOIL MEASURED WINDOW ENERGIES

Window Energy E_{tof} (keV)	Beam Source	Proton-Recoil Measured Energies (keV)		$E_{\text{pr}}/E_{\text{tof}}$	
		CH_4	H_2	CH_4	H_2
2.00 $\pm$ 0.05	Sc	1.52 $\pm$ 0.02	1.96 $\pm$ 0.02	0.760	0.980
7.50 $\pm$ 0.40	Sc	6.76 $\pm$ 0.07	7.55 $\pm$ 0.08	0.901	1.006
24.74 $\pm$ 0.03	Fe	22.80 $\pm$ 0.22	24.80 $\pm$ 0.25	0.922	1.006
81.92 $\pm$ 0.08	Fe	78.00 $\pm$ 0.78	82.00 $\pm$ 0.82	0.952	1.007
136.85 $\pm$ 0.18	Fe	132.00 $\pm$ 1.30	137.00 $\pm$ 1.40	0.965	1.001
167.31 $\pm$ 0.24	Fe	164.00 $\pm$ 1.60	167.00 $\pm$ 1.70	0.980	0.998
218.33 $\pm$ 0.35	Fe	214.00 $\pm$ 2.20	218.00 $\pm$ 2.20	0.980	0.989
271.94 $\pm$ 0.50	Fe	269.00 $\pm$ 2.70	270.00 $\pm$ 2.70	0.989	0.985
307.90 $\pm$ 0.60	Fe	308.00 $\pm$ 3.10	305.00 $\pm$ 3.00	1.000	0.991
350.13 $\pm$ 0.70	Fe	350.00 $\pm$ 3.50	347.00 $\pm$ 3.50	0.999	0.991
465.43 $\pm$ 1.00	Fe	468.00 $\pm$ 4.70	449.00 $\pm$ 4.50	1.006	0.965
636.40 $\pm$ 2.00	Fe	649.00 $\pm$ 5.00	616.00 $\pm$ 5.00	1.020	0.968
947.00 $\pm$ 5.00	Fe	955.00 $\pm$ 6.00	952.00 $\pm$ 6.00	1.008	1.005

Fig. 1 Diagram of iron-filtered beam facility.

Fig. 2 Diagram of scandium-filtered beam facility.

Fig. 3 Pulse-height distribution of pulse rate of rise at 2 keV in scandium-filtered beam.

Fig. 4 Extrapolated and effective gas amplifications.

Fig. 5 Proton spectrum from iron-filtered beam taken with spherical methane-filled counter.

Fig. 6 Proton spectrum from scandium-filtered beam taken with spherical methane-filled counter.

Fig. 7 Neutron spectrum from iron-filtered beam taken with spherical methane-filled counter.

Fig. 8 Neutron spectrum from scandium-filtered beam taken with spherical methane-filled counter.

Fig. 9 Comparison of discrete energies measured with hydrogen- and methane-filled counters and time-of-flight technique.

Fig. 10 Neutron spectra from iron-filtered beam as taken with hydrogen- and methane-filled counters.

Fig. 11 Neutron spectra from scandium-filtered beam as taken with hydrogen- and methane-filled counters.

COMPARISON OF PROTON-RECOIL SPECTROMETER CALIBRATIONS

J. W. Rogers, J. E. Powell^[1]

A joint experiment between Idaho Nuclear Corporation and Argonne National Laboratory (Idaho Site) was performed to compare conventional proton-recoil spectrometer calibration techniques[2]. These comparisons were made with the discrete neutron energies of the MTR neutron beams[3,4]. The extended capability of the Argonne spectrometer[5,6] allowed lower energies to be measured and digital computer handling of gamma discriminated data. Spectra were measured between 185 eV and 1 MeV in the 2 keV (scandium filtered) and 25 keV (iron filtered) beams. Comparisons of the spectra measured with the Idaho Nuclear spectrometer[7] and the Argonne spectrometer were made.

Three co-axial cylinder proportional counter detectors were used in these measurements, two being hydrogen filled and the other methane filled. These detectors are described in Table I. The larger hydrogen counter was used with the Idaho Nuclear spectrometer and the smaller one with the Argonne spectrometer. The methane counter was used with both spectrometers.

The Argonne spectrometer was calibrated by the usual technique using the $^{14}\text{N}(n,p)^{14}\text{C}$ reaction. The MTR thermal column was used as a thermal neutron source for this calibration. The Diethorn relationship[8] used in calibration extrapolation was modified by using $\frac{\ln A}{V^{2/3}}$ versus V instead of $\frac{\ln A}{V}$ versus $\ln V$, where A is the gas amplification and V the applied voltage. The Idaho Nuclear spectrometer was calibrated as described elsewhere[9].

The proton-recoil data were analyzed using a computer code of Bennett, Gold, and Olson[10]. The resulting neutron spectra are shown in Figures 1 and 2. The Idaho Nuclear data were taken with no gamma discrimination and the Argonne data were taken with pulse shape (rate of rise) discrimination

of the gamma rays. The Idaho Nuclear data from the iron beam were taken using only the hydrogen detector. The gamma contribution in the nondiscriminated spectra can be seen clearly but does not cause serious problems from the standpoint of calibration investigation.

In order to get agreement between the proton-recoil measurements and time-of-flight measurements in the hydrogen data, it was necessary to assume 585 keV as the effective total ionization energy of the $^{14}\text{N}(n,p)^{14}\text{C}$ reaction and no W (energy loss per ion pair) cutoff in hydrogen. If a 0.3 keV threshold is assumed the hydrogen data yields energies 0.3 keV too high which contributes a 15% error at 2 keV.

The methane data from the Idaho Nuclear spectrometer were analyzed assuming 585 keV for the total effective ionization energy of the $^{14}\text{N}(n,p)^{14}\text{C}$ calibration reaction and no W cutoff energy. The Argonne data assumed 615 keV for the calibration reaction and 4 keV for the W cutoff for methane. These assumptions led to differences that can be seen in the spectra which are not yet resolved. The reasons for these different assumptions are based on the requirements necessary to get the best agreement between hydrogen and methane data where they overlap.

These results point to ambiguities concerning energy loss per ion pair in hydrogen and methane, the effective Q of the $^{14}\text{N}(n,p)^{14}\text{C}$ reaction, and the treatment of the energy loss per ion pair in the unfolding of the proton data. The results discussed in Reference 8 may help resolve some of the problems.

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Fig. 1 MTR iron-filtered neutron beam spectrum measured with different spectrometers, with and without gamma discrimination.

Fig. 2 MTR scandium-filtered neutron beam spectrum measured with different spectrometers, with and without gamma discrimination.

TABLE I
DESCRIPTION OF PROPORTIONAL COUNTER DETECTORS

	<u>H₂ Detectors</u>		<u>CH₄ Detector</u>
Diameter	0.625 in.	0.9375	0.9375 in.
Length	2.875 in.	4.50 in.	4.50 in.
Center Wire Diameter	0.7 mil	1 mil	1 mil
Sensitive Volume	5.657 cm ³	29.73 cm ³	29.73 cm ³
H ₂ Pressure	380 cm Hg	200 cm Hg	---
CH ₄ Pressure	19 cm Hg	10 cm Hg	380 cm Hg
N ₂ Pressure	19 cm Hg	10 cm Hg	19 cm Hg
Detector Material	stainless steel	stainless steel	stainless steel

RECOIL-PROTON SPECTROMETER DATA ANALYSIS

R. G. Nisle

The procedure necessary for successfully unfolding a neutron spectrum from the recoil-proton distribution measured in a proportional counter is very complex. A large part of the difficulty lies in the fact that the proton distribution, represented by experimental data points, needs to be differentiated. Since the points exhibit the usual random fluctuations of experimental data, it is necessary to give careful consideration to any scheme for numerically calculating the derivative. Various procedures are employed such as three- or five-point differentiation, or one of the standard data "smoothing" techniques of numerical calculus. In addition, when recoil-proton distributions are measured in the presence of gamma rays, their contribution to the ionization measured by the detector must be subtracted. New approaches to these problems have been demonstrated and are being employed in the analysis of recoil-proton spectrometer data.

The use of polynomial transforms (a digital approximation to the Laplace transform) for the reduction of a proton spectrum to a neutron spectrum was demonstrated [Nucl. Instr. and Methods 70, 169-172 (1969)]. The method uses the transform of the instrument transfer function derived from measurements in a 1/E flux in a standard graphite pile. A simple program for performing polynomial division is then used to convert the proton spectrum to a neutron spectrum by use of this transform of the transfer function. Figure 1 shows a neutron spectrum as obtained by the polynomial transform method compared with the same spectrum as obtained by differentiation.

Fig. 1 A comparison of spectra analyzed by polynomial transform method and a conventional 3-point differentiation.

A graphical method of analysis has been used successfully to reduce recoil-proton counter data to a neutron spectrum from a proton spectrum of discrete sources[1]. A computer-generated synthetic curve is matched visually on a cathode ray oscilloscope display to the experimental curve, and the parameters of the computer curve are taken to be those of the experimental curve when a match is obtained. The parameters used are source energy and intensity and instrument resolution. The instrument response to a mono-energetic source is assumed to be Gaussian and the source width is assumed to be negligible compared with the instrument resolution. Figures 2 and 3 illustrate the use of this method. Figure 2 shows an integral proton spectrum consisting of two line sources with a generated spectrum displaced below it. Figure 3 shows the two in register to illustrate the nature of the match. Both this approach and the polynomial transform approach emphasize the analysis of recoil-proton spectra by a direct treatment of the integral data without the use of differentiation.

The PDP-9 computer was programmed to use an oscilloscope display to assist in making a normalization of a gamma spectrum to the gamma background of raw two-parameter recoil-proton data. Both gamma and composite spectra are displayed concurrently for one energy channel, ie, counts versus rate-of-rise for one energy. The gamma spectrum is then matched visually with the gamma part of the composite spectrum by setting chosen values of

Fig. 2 Integral data from a line spectrum of two sources are shown with the generated integral displaced below the reference count level of the lower energy line.

the normalization factor into the computer. When an acceptable match is obtained, the gamma spectrum is subtracted from the composite spectrum and the channels in the neutron peak that remains are integrated. This process is repeated for each of the energy channels in a set of two-parameter recoil-proton data. The process permits a channel-by-channel examination of the agreement between the gamma distribution in the composite spectrum and the gamma spectrum used for making the background correction. The gamma spectrum used in the accompanying illustrations was that obtained from a cobalt-60 source. The figures show that the two spectra do not match, although such spectra are frequently used to make corrections for the gamma background in a recoil-proton spectrum. In the procedure described here the effects of such disagreement are minimized. The accompanying figures were prepared from photographs of the oscilloscope display. The direct photographs (positive prints) consist of white dots on a black background. It is difficult to indicate the various features on such prints. Therefore, negatives have been used and the pertinent features are indicated thereon. The first series of illustrations shows the 75.8 keV channel with various amounts of gamma compensation. Figure 4 shows the composite spectrum with an undercorrected gamma spectrum superposed to indicate more clearly the differences in spectral distributions. Figure 5 is a difference spectrum with the undercompensation shown in the previous figure. Figure 6 shows the difference spectrum of the same energy channel with correct compensation. The counts shown to the

Fig. 3 The generated integral is shown in register with the integral data of the lower energy line. The data of the higher energy line has been discarded.

left of the indicated rate-of-rise channel are due to the mismatch between composite and gamma spectra and are discarded in the summing operation. Figure 7 shows a difference spectrum with overcompensation. The second series of illustrations shows spectra in which gamma compensation is unnecessary. The variation of wall and end effects with increasing energy is shown. Figure 8 shows the counts versus rate-of-rise for the 151.6 keV channel; Figure 9 shows the same effects for the 214.0 keV channel; Figure 10 shows the 280.8 keV channel. Such a procedure is much slower than machine methods, but it permits an inspection of the data as the analysis processes. The information gained thereby can then be used to improve those operations that visual inspection has shown to be necessary. One such feature that has been indicated is the use of area-normalization of the gamma background in preference to point-scaling.

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Fig. 4 Composite spectrum shown with a gamma spectrum of reduced amplitude to illustrate shape differences.

Fig. 5 Difference of two spectra shown in Figure 4 to illustrate under-compensation.

Fig. 6 Difference spectrum with correct compensation. Left most channels indicated are the result of shape differences between composite gammas and the gamma spectrum of ^{60}Co . These latter are discarded in the summing operation.

Fig. 7 Difference spectrum with over-compensation. Note that counts in some channels are lost, while in others the counts are too small.

Fig. 8 Wall and end effects in the 151.6 keV channel illustrated.

Fig. 9 Wall and end effects in the 214.0 keV channel illustrated.

Fig. 10 Wall and end effects in the 280.8 keV channel illustrated.

A TECHNIQUE FOR DETERMINING NEUTRON FLUX SPECTRA FROM ACTIVATION MEASUREMENTS

D. A. Pearson

An effort is currently being made to utilize existing computer codes to determine neutron flux spectra from foil activation techniques. The method currently being used is a modified version of the SPECTRA code which provides a neutron flux spectrum that minimizes an error function^[1]. A trial spectrum is supplied as tabular input, and by requiring that the squared difference between the measured and calculated detector activities be a part of the function, it is possible to obtain a convergence fit to the new spectral shape. It can be shown that the technique does not force any specific characteristics of the trial spectrum upon the shape of the determined spectrum. In addition, through rigorous mathematical analysis, it can be shown that an exact solution to the activation equations can be obtained.

Fig. 1 Differential and integral results for CFRMF spectrum (critical).

The code is capable of computing up to 50 differential and integral flux points from a library of reactions taken from the compilation of McElroy et al[2].

The SPECTRA code is very input sensitive as certain combinations of detectors and errors can make the matrix equations unstable. If this occurs, the code will calculate unrealistic values. Spectral shape will be determined only to the extent that the cross sections or response functions of the detectors used are linearly independent, and careful interpretation is required when evaluating unexpected perturbations. The solution method was developed to utilize all of the physical information available to the user. The user must have adequate knowledge to select the proper detector foils and recognize reasonable trial solutions. For these, and other reasons, the code cannot be used as a "black box".

It is difficult to assess errors in differential flux since unknown errors can be introduced through the input cross section data. In a well controlled experiment, the differential and integral flux results can be expected to be accurate within 10 to 30% at all energies.

A trial run was made in the CFRMF facility (which has a spectrum typical of a fast reactor) utilizing the following activations: $^{23}\text{Na}(n,\gamma)^{24}\text{Na}$, $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$, $^{55}\text{Mn}(n,\gamma)^{56}\text{Mn}$, $^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$, $^{115}\text{In}(n,n')^{115\text{m}}\text{In}$, $^{58}\text{Ni}(n,p)^{58}\text{Co}$. The preliminary results obtained from the code are shown in Figure 1. Extensive evaluation of the experiment has not been completed. However, the results look extremely encouraging.

It is our opinion that exhaustive studies must be made utilizing the technique under a variety of circumstances before the code proves acceptable.

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USE OF CORRELATION TECHNIQUE FOR MEASURING FISSION
CROSS SECTIONS IN THE PRESENCE OF A LARGE TIME-
INDEPENDENT BACKGROUND

F. B. Simpson, T. Watanabe, W. R. Myers, P. D. Randolph

There is a vital need for fission cross section measurements for many of the transuranic isotopes. The samples available for making these measurements are limited and not of as high a quality as desired. Most of the material is a mixture of isotopes; some of these isotopes have high spontaneous fission rates. With the conventional techniques using the present available neutron sources it is very difficult to measure low rate signals in the presence of a high time-independent background.

With the advent of the correlation technique^[1,2], it seems that a correlation chopper could overcome the obstacles mentioned in the preceding paragraph. This method has the advantage of being able to measure low rate signals in the presence of high background noise. It has a duty cycle of 50% compared to the present fast chopper rotor of 0.1%, thus utilizing a much higher fraction of the neutrons available from the reactor. The correlation method coupled with a high intensity reactor should yield neutron intensities in the low neutron energy region which can successfully be used to measure these needed cross sections.

An experiment has been performed with a correlation chopper^[2] to determine the feasibility and advantages of using such a device to measure events in the presence of large time-independent backgrounds. A similar measurement was performed with a conventional chopper for comparison. Figure 1 is the fission cross section of ²³⁹Pu measured with the conventional 2-slot chopper rotating at 14,700 rpm. The data were taken using a 4-μsec channel width and a flight path of 2.7 meters. In this case the noise level was such that the signal-to-noise ratio was 100 to 1. The same type of data were also taken with a correlation chopper using the same fission chamber. However, the background noise was increased to demonstrate the ability of measuring the fission cross section in the presence of a large steady state background noise. This was accomplished by lowering the bias level of the alpha discriminator on the fission chamber.

In the correlation case, the signal-to-noise ratio was 1.6 to 1. The results of measuring induced fission events from ²³⁹Pu in the presence of the high level background with the correlation chopper is shown in Figure 2. The running times for collecting these data were 8 hours. Because of the high count rate, large dead times were introduced in the time-of-flight analyzer and it was necessary to make counting loss corrections to the data before processing them through the cross correlation program. Both sets of data

have been normalized to the data in BNL-325. Because the correlation chopper was constructed with Gd_2O_3 as the absorbing material, it has a cutoff at about 0.2 eV. Therefore, the data above 0.2 eV are partly time correlated and partly uncorrelated. The chopper is practically transparent to neutrons above 0.5 eV; therefore, these neutrons become a source of constant background. Because of the type of construction of the rotor, the data were only reliable to about 0.2 eV.

Considering the uncertainties that have been mentioned, the results of these measurements substantiate the claims made for correlation methods of being able to extract time-correlated information in the presence of a large time-independent background. We feel that this experiment has demonstrated the feasibility and some of the advantages in using this technique for making partial cross section measurements which inherently have a low signal-to-noise ratio.

We are presently constructing a Fourier chopper which we can use to make similar type of measurements for a comparison with the other systems. We feel that this will give us an additional advantage in neutron intensity over the correlation chopper because we can use multiple slits.

- [1] F. Gompf, W. Reichardt, W. Glaser, K. H. Beckurts, "The Use of a Pseudo-Statistical Chopper for Time-of-Flight Measurements", Neutron Inelastic Scattering. Proceedings of a Symposium on Neutron Inelastic Scattering Held by the International Atomic Energy Agency in Copenhagen, 20-25 May, 1968. Volume II, Vienna: International Atomic Energy Agency, 1968, pp 417-428.

Fig. 1 Fission cross section of ^{239}Pu taken with a standard 2-slot chopper at a flight path of 2.7 m. Data are normalized to the fission cross section in BNL-325.

Fig. 2 Fission cross section of ^{239}Pu measured with a correlation chopper at a flight path of 2.7 m. Data are normalized to the fission cross section in BNL-325. Lower curve is a plot of the total cross section of Gd which was used as the absorbing material.

AUTOMATIC CROSS SECTION ANALYSIS PROGRAM

O. D. Simpson, N. H. Marshall^[1]

The Automatic Cross Section Analysis Program (ACSAP) was developed to adjust the resonance parameters using an "experimentalists approach". That is, ACSAP computes a theoretical cross section curve from starting parameters, compares this curve with specified input points, then adjusts the parameters (much as an experimentalist would) to bring the theoretical curve closer to the input data points. ACSAP adjusts the parameters by mathematical algorithms which are very similar to those an experimentalist might use to adjust his starting parameters after plotting his theoretical data.

ACSAP produces either single level or multilevel resonance parameters. It automatically adjusts initial guess values of E_0 , Γ_n° , and Γ_x (Γ_x represents either Γ_v or Γ_f) until the resolution and Doppler broadened theoretical data pass through three prescribed experimental data points, one at the peak and one on each side of the resonance. The program will also correct for any constant bias that might be present in the experimental data. This is done by requiring the theoretical data to pass through designated valley points. A valley point is by definition a point which lies in a region where Doppler and instrumental resolution broadening do not affect the cross section data. Using the three experimental points mentioned, ACSAP can determine E_0 , Γ_n° , and either Γ_v or Γ_f (but not both) for each resonance. This process is referred to as shape fitting. At energies where shape fitting is not desirable (eg, where the resolution width dominates the resonance), ACSAP can do a peak fit where only Γ_n° or Γ_x is determined. Only the peak point is needed for a peak fit. There may be instances where it is not desirable to use a peak point (eg, where the transmission goes to zero in the region of the resonance peak data). In this case a wing fit may be done using only points on the side of the resonance. A wing fit can yield an adjustment to E_0 and to either Γ_n° or Γ_x .

A basic flow chart of ACSAP is shown in Figure 1. The program permits a mixture of resonances from 25 different isotopes with a maximum of 250 resonances to be fit at any one time. Data can be automatically fit in the form of cross section versus energy or transmission verses delay.

To demonstrate that the program would work properly, ²³³Pa resonance parameters as quoted by Simpson^[2] were used to generate a set of resolution and Doppler broadened cross section data in the energy region from 1 to 18 eV. This set of ²³³Pa data was interpreted as representing a set of experimental data having perfect statistics. Initial guess resonance parameters were arbitrarily chosen and ACSAP was allowed to fit the data automatically. Figure 2 shows the differences between the "true" parameters and those obtained by ACSAP. Here the percent difference is plotted verses the resonance number for both Γ_n° and Γ_x . It should be noted that in all cases the agreement of the Γ_n° 's was within 1/2%. The Γ_v 's agreed to 1% or less except for the last resonance which is still less than 2%.

To see how well the program would describe a set of total cross section data from a fissile nucleus, ACSAP was used to fit the ²³²U data from 1 to 31 eV. The Reich-Moore multilevel equation was used. Figure 3 shows the

^{232}U cross section data. Superimposed on the data are the cross section curves obtained from the initial guess parameters and from the final resonance parameters determined by ACSAP. ACSAP cannot determine the type of interference (destructive or constructive between resonances) or the channel in which fission occurs; therefore, the channel and interference signs were assumed to be the same as those obtained by Simpson[3]. A constant value of $\Gamma_\gamma = 47$ meV was assumed for the capture widths, and the fission widths were allowed to vary until the desired fit was obtained. This required a total of about ten minutes of computer operating time (approximately \$150). Original analysis took three to four months.

Figure 4 compares the ^{232}U parameters determined by ACSAP with those found by the earlier analysis. The percent difference is plotted versus the resonance number. Note that the Γ_n 's are consistent within 5%, and the Γ_f values are within 10%. It is felt that the ACSAP analysis is an improvement over the earlier analysis.

Several avenues of development for ACSAP are anticipated in the future. Some are as follows:

- (1) Combining ACSAP with SCORE. The SCORE program was written principally by Charles Dunford of Atomics International and Bob Creasey of IBM. It allows display and manipulation of cross section data using interactive graphics. Since ACSAP still cannot determine all of the variables associated with multilevel analysis (eg, the type of interference between levels), and since it may be desirable to monitor the convergence of ACSAP, it is felt that the ACSAP-SCORE marriage will be a big boom to evaluators. Preliminary work has been done to perform this marriage and the combining of these two programs is well underway.
- (2) Fit total and fission data simultaneously. It is felt that an analysis of total and fission data simultaneously will lead to better and more consistent parameter values.
- (3) Fit total, fission, capture, and scattering data simultaneously. Once step two has been done the complete analysis of all the cross section data seems a logical step.
- (4) Least square fitting of data. Considerable work has already been done on the least square analysis of cross section data. This technique requires good starting values to ensure convergence. Often these are difficult to determine. It is felt that ACSAP may be used to provide starting values for the least square processes.

Work done on ACSAP to date has been most gratifying. Local physicists have been pleased with the results. It is felt that this is a good tool for the cross section evaluator.

[1] Member of Computer Science Center Branch.

Fig. 1 Basic flow chart for ACSAP.

Fig. 2 The percentage difference between the "true" ^{233}Pa resonance parameters and those obtained by ACSAP.

Fig. 3 ^{232}U total cross section data from 1 to 30 eV. The data were fit using the Reich-Moore multilevel equation. The solid lines compare the initial guess curve with the final output using ACSAP.

Fig. 4 The percentage difference between the ^{232}U resonance parameters obtained by ACSAP and those determined from an earlier analysis.

DEVELOPMENT OF LONG-TERM INTEGRATED FLUX MONITORS

J. A. Dunbar, R. P. Schuman

There is a need for flux monitors that can be used for long-term irradiations. Ideally, it would be best to have two monitors, one which is sensitive to resonance neutrons (i.e., the ratio of resonance integral to thermal cross section is high) and one which is sensitive to thermal neutrons so that both thermal and resonance integrated fluxes can be determined.

Most flux monitors depend upon the formation of an active nuclide which can be determined by absolute counting. For accurate flux monitoring, such monitors should be irradiated for less than one "in reactor" half life (half life for radioactive decay and neutron burnup) and the target nucleus should be burned up well under 50%. The activation of cobalt to ^{60}Co is one of the best long time flux monitors, but after several years (due mainly to ^{59}Co burnout) the activity of ^{60}Co will change very little with increasing nvt, and ultimately the ^{60}Co activity will actually decrease (due to ^{60}Co decay and burnout) with increasing nvt.

Manganese has been used as a flux monitor for a long time, but usually only for short irradiations due to the short half-life of ^{56}Mn (2.58 hrs). The thermal cross section and resonance integral of ^{55}Mn are 13.2 ± 0.5 barns and 14.0 ± 0.7 barns, respectively[1]. Manganese upon capturing neutrons gives iron, so it can be used as a long-term, high nvt, flux monitor by colorimetric analysis of the resulting iron and the residual manganese in the sample. Manganese can be precisely (+1%) and quickly determined on a microgram scale by oxidation to permanganate and determination of the absorption at 525 m μ [2]; the iron can equally precisely (+1%) and quickly be determined on a microgram scale by formation of the ferrous orthophenanthroline complex and determination of the absorption at 510 m μ [2]. The iron produced by manganese neutron capture is not appreciably burned up even after very long irradiations since the ^{56}Fe first formed captures neutrons ($\sigma_c = 2.7$ b) to give stable ^{57}Fe which in turn captures neutrons ($\sigma_c = 2.3$ b) to give stable ^{58}Fe [3]. It is only the capture by ^{58}Fe ($\sigma_c = 1.1$ b) to ^{59}Fe which decays to cobalt, which will result in the burnout of iron.

A sample of 9.58 mg specific pure manganese was irradiated for six years as a flux monitor for the ^{94}Nb production irradiation. The manganese was allowed to decay for about a month so there was little residual radioactivity, then dissolved in diluted HNO_3 and iron and manganese determined colorimetrically. The irradiated sample was found to contain 4.81 mg manganese and also 4.81 mg iron. The integrated pile flux (assume $\sigma = 14.0$ barns as for a thermal flux), as calculated from the iron to manganese molecular ratio is 4.9×10^{22} n/cm 2 . The estimated integrated flux from pile operation is 5.6×10^{22} n/cm 2 . The activities in the manganese after two months' decay are ^{54}Mn (due to fast n,2n reaction) and ^{59}Fe and ^{60}Co due either to the activation of trace impurities of iron and cobalt or perhaps to capture of 4 and 5 neutrons successively by the manganese. In either case the impurity iron or burnout of iron did not cause a significant error in the integrated flux. Manganese had been previously used as a flux monitor for irradiations of nvt = 1.8×10^{20} to 2.2×10^{22} n/cm 2 [4].

The niobium activation to 2.03×10^4 yr [5] ^{94}Nb can also be used as a high nvt flux monitor up to an nvt of around 4 to 5×10^{22} ; above this nvt the ^{94}Nb activity starts to level off and the monitor is no longer very sensitive to increased nvt. Due to second order capture to 35 d ^{95}Nb , the direct determination of ^{94}Nb is difficult until most of the ^{95}Nb has decayed. Figure 1 shows a Ge(Li) gamma spectrum of a sample of the niobium after three months decay. The amount of niobium capture is most accurately determined by measuring the ^{95}Nb second order product which is proportional to the ^{94}Nb concentration. The cross section of both ^{93}Nb and ^{94}Nb have been measured as:

$$^{93}\text{Nb}, \sigma_{\text{th}} = 1.0 \pm 0.1 \text{ b, R.I.} = 8.5 \pm 0.5 \text{ b}^{[1]} \text{ and}$$

$$\sigma_{\text{th}} = 1.1 \pm 0.1 \text{ b and R.I.} = 9.2 \pm 10 \text{ b.}^{[6]}$$

$$^{94}\text{Nb}, \sigma_{\text{th}} = 13.6 \pm 1.5 \text{ b, R.I.} = 125 \pm 8 \text{ b}^{[1]} \text{ and}$$

$$\sigma_{\text{th}} = 16.8 \pm 1.5 \text{ b and R.I.} = 122 \pm 10 \text{ b.}^{[6]}$$

Until better cross sections are determined for the two niobium isotopes, and the possibility of 35 day ^{95}Nb burnout checked, the chemical determination of iron buildup in manganese will be a more precise flux monitor. For the most accurate monitoring, manganese self-absorption of resonance neutrons will have to be corrected. Since niobium capture is more sensitive to resonance neutrons than manganese capture, manganese and niobium could be used as a pair of flux monitors so that both thermal and resonance nvt could be determined.

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 - [5] R. P. Schuman and P. Goris, "Half-Life and Decay of Niobium-94", J. Inorg. Nucl. Chem., 12 (1959) pp 1-5.
 - [6] G. E. Moore (ed.), Chemistry Division Annual Progress Report for Period Ending May 20, 1968, ORNL-4306 (September 1968).

Fig. 1. Ge(Li) gamma spectrum of highly irradiated niobium.

BURNOUT OF CADMIUM SHIELDS BY LONG REACTOR IRRADIATIONS

E. D. Gregory, R. P. Schuman, L. G. Miller

A number of radioactive nuclides that have high thermal neutron cross sections can be produced in considerably higher yields by resonance flux irradiation than by normal reactor flux irradiation. We have produced a number of chopper cross section samples by the ETR irradiation of the target material inside a cadmium shield. The cadmium shielding gives the desired resonance flux; however, an appreciable thickness of cadmium is burned out during the irradiation. Since sample production often requires very high integrated resonance fluxes, and since the desired product will be rapidly destroyed once the cadmium shield is burned out, the burnout of ^{113}Cd in a shield that had been irradiated for two cycles in the ETR was investigated.

The cadmium shield was in the form of a hollow cylinder, 60 mm long, 20mm OD, and 6 mm ID. A slice of cadmium about 2 mm thick was cut from the shield using the hack saw in the MTR hot alpha cave. The slice, which had decayed sufficiently so that it could be handled unshielded with tongs, was then neutron radiographed with the MTR neutron radiography facility in order to measure the thickness depleted in ^{113}Cd . A neutron radiograph taken on the slice by itself showed a ring of neutron absorbing material which at first glance indicated no burned out section; however, the absorbing section was about 2.5 mm smaller than the actual ring. A closer examination showed a barely visible ring of low neutron absorbing material about 1.3 mm thick outside of the unburned cadmium. The slice was then inserted into a 2.0-cm diameter hole in a 1-mm cadmium sheet, and a second neutron radiograph taken; this radiograph showed clearly the ≈ 1.3 mm thick section of burned out cadmium. The neutron radiograph is shown in Figure 1 and a photograph of the slice in the cadmium sheet is shown in Figure 2.

Fig. 1 Neutron radiograph of slice of highly irradiated cadmium shield inserted into hole in cadmium sheet -- actual size.

Fig. 2 Photograph of same slice of cadmium shield in cadmium sheet -- actual size.

The integrated epicalcium flux received by the shield was measured as 2.55×10^{20} n/cm² per 1n E interval. The integrated unperturbed thermal flux in the irradiation position was expected to be 2.67×10^{21} n/cm². The burnout of ¹¹³Cd in 1.3 mm of normal cadmium corresponded to the absorption of 0.74×10^{21} neutrons/cm², which is roughly what was expected from the unperturbed flux of the irradiation position. The burnout shows that cadmium shields could be safely irradiated to an integrated thermal flux of two to possibly three times the thickness in atoms ¹¹³Cd/cm² and so the 7 mm cadmium shield could be safely used in the same irradiation position for six ETR cycles. The complete burnout of the cadmium is expected to occur for an integrated thermal flux of about four times the thickness in atoms ¹¹³Cd/cm².

SAMPLE PREPARATION TECHNIQUES

J. R. Berreth, D. K. Oestreich

One of the prime duties of the Chemistry Group is to prepare samples for various research disciplines. This includes preparation of the materials for irradiation in the reactors, proper irradiation of the materials, determination of integral flux, and necessary handling after the irradiation.

In certain instances radioactive isotopic materials are received in the oxide form from off site and processed here by mixing the material with aluminum if necessary and compacting the material into proper size samples. Materials prepared in this fashion were four ¹⁵²Eu samples, three ²³⁶U samples, and one ²³⁰Th sample.

Samples irradiated in the reactor in the form of the metal and prepared for experiments were ²⁰⁴Tl, ⁹⁴Nb, and a ²³⁵U fuel mock-up sample.

Several fission foil samples on thin backings were prepared by electroplating. These included ^{236}U , ^{238}Pu , and ^{235}U .

During the course of the year we were asked to measure the total neutron cross section of the curium isotopes. Because of hot cell use timing it was necessary to extend our knowhow on oxide sample preparations to Savannah River laboratories by lending our specialized dies and consulting with them on the preparation of these curium samples. We have also undertaken at their request the preparation of an enriched ^{246}Cm sample for nuclear explosion experiments. This was done because of our capability to prepare wafer thin oxide samples, thus saving them the cost of tool up and development of the technology.

A RAPID SOURCE PREPARATION TECHNIQUE FOR HIGH-RESOLUTION ALPHA-PARTICLE SPECTROSCOPY [1]

R. A. Deal^[2], R. N. Chanda

Because of their high rate of ionization, alpha-particle ranges in matter are quite small. Consequently, high-resolution alpha-particle spectroscopy requires extremely thin and uniform sources so as not to degrade and spread the energy of the alpha particle leaving the source.

Several source preparation methods have been in use for some time including vacuum sublimation from a hot filament, electrospraying, and electrodeposition. However, these methods are all time consuming and involve rather elaborate apparatus. The method we developed is rapid and involves no apparatus other than standard laboratory equipment.

To prepare an alpha source, the alpha emitting nuclide is extracted into TTA (thenoyltrifluoroacetone) dissolved in benzene and a portion of the organic fraction transferred via a micropipette or spitzer onto a platinum disc. In order to confine the organic deposit to a small area a slight depression is made in the platinum disc. After drying under a heat lamp the plate is slowly flamed over a burner until no organic deposits remain. The entire procedure takes on the order of five minutes with 100% yields in all cases. An example of a spectrum taken with a TTA-evaporated plate is shown in Figure 1. Since the quality of the TTA evaporated plates is at least as good as plates prepared by other methods, we now use TTA plates almost exclusively.

[1] R. A. Deal and R. N. Chanda, "A Rapid Source Preparation Technique for High-Resolution Alpha-Particle Spectroscopy", Nucl. Instr. & Methods, 69 n 1 (March 1969) pp 89-92.

[2] Presently with Dow Chemical Company, Rocky Flats Division, Golden, Colorado.

Fig. 1. Alpha-particle spectrum of ^{244}Cm recorded on a 25 mm^2 area by 100 mm depletion depth silicon gold surface-barrier detector. The source was prepared by a TTA evaporation onto platinum.

CATALOGUE OF SEMICONDUCTOR
ALPHA-PARTICLE SPECTRA[1]

R. N. Chanda, R. A. Deal[2]

The use of semiconductor detectors for alpha-particle detection and spectroscopy has been gaining in popularity with the advent of improved detectors and the increased reliability of solid state electronics. However, outside of a few research labs there appears to be a general lack of information concerning not only the use of these semiconductor detectors but the improved alpha-particle resolutions that are now routinely available. A catalogue has been prepared to fill this information gap and stimulate the more widespread use of alpha spectroscopy.

The Catalogue contains alpha spectra of some 35 nuclides of both the natural decay chains and a number of the most frequently encountered artificially produced nuclides. In order that this data be useful for analytical purposes, all spectra were recorded using a high-resolution semiconductor detector (Figure 1) in a standard geometry with a standard gain scale. The spectra are presented in graphical (Figure 2) as well as digital form. In addition, the electronics, detectors, and source preparation techniques are described in detail. Also, to aid the user of the Catalogue, appendices are included listing nuclides according to Z, A, and alpha-particle energy, a table of specific activities, the natural radioactive series, and range energy curves.

[1] To be issued as USAEC Report IN-1261.

[2] Present Address: Dow Chemical Co., Rocky Flats Division, Boulder, Colorado.

Alpha-Particle Spectrometer

Fig. 1 Alpha-particle spectrometer used for recording the alpha spectra in the Catalogue.

Fig. 2 Example of an alpha-spectrum as found in the Catalogue.

NEUTRON RADIOGRAPHY AS AN ANALYTICAL TOOL

L. G. Miller

Neutron radiography as an analytical tool may be used for the determination of concentrations and uniformity of one material in another to a high degree of accuracy. As a test of this technique, gadolinium in aluminum has been tested from 0.012 to 1 weight percent Gd. Due to the nonuniformity of the gadolinium in the aluminum, the data deviate from a straight line for very low concentration (see Figure 1). With better samples and more accurate densitometer equipment this technique can be extended to even smaller concentrations. This technique can be useful for many material combinations such as Gd in Zr, Hf in Zr, Ag in Pb, Ir in Pt, etc. Accuracy and range will depend on neutron cross sections of material involved.

Fig. 1 A plot of the film density obtained from neutron radiographs of a number of 0.525 inch thick gadolinium-aluminum samples versus weight percent of gadolinium.

DEVELOPMENT OF RADIOACTIVE TAGGING OF BORON WITH ⁶⁰Co IN REACTOR FUEL ASSEMBLIES

E. L. Murri, J. E. Cline

A method is being developed which will permit accurate and simultaneous assay of reactor fuel plates for ²³⁵U and boron, the latter of which is a burnable poison. In the method boron is uniformly tagged with ⁶⁰Co and then is homogeneously dispersed with UAl_x powder.

The assay utilizes two gamma-ray detectors with single channel analyzers. The ²³⁵U content and uniformity is measured with the single channel analyzer of one detector covering the energy region of the 163 and 185 keV gamma rays from ²³⁵U. The boron assay is made with the window of the other analyzer covering only the energy region of the ⁶⁰Co gamma rays, well above those emitted from uranium.

Initial laboratory tests indicated that ≈ 0.4 μ curie of ^{60}Co per gram of boron glass would provide suitable sensitivity for the boron scan and would produce no detectable interference in the fuel scan due to Compton scattering. Ten test fuel plates were fabricated on this basis and were assayed by the above techniques. The boron determinations were within 10% and the uranium determinations were within 3% of the known values.

Since most field applications of this assay technique will be under less ideal background radiation conditions, the quantity of ^{60}Co should be increased. Fuel plates presently being fabricated for further study under field conditions will contain ≈ 2.0 μ curies of ^{60}Co per gram of boron glass. This will require a small, but constant, correction to the ^{235}U count rate from the ^{60}Co Compton distribution.

Very effective techniques for blending the ^{60}Co tagging radioisotope with the boron glass have been developed by the Reactor Engineering Branch. Uniformity of ^{60}Co in the glass has been achieved to within 1% among a large number of random samples.

DETERMINATION OF FUEL COMPOSITION USING THE MTR FAST CHOPPER AS AN ANALYTICAL TOOL

D. K. Oestreich, J. R. Berreth, F. B. Simpson

A nondestructive technique for determining fuel burnup is being developed using the MTR fast chopper as a "neutron spectrophotometer" by looking at the characteristic neutron resonances. With this technique one can determine the amounts of capture products and the amounts of poison-type fission products formed in a reactor fuel piece with a relatively high degree of accuracy.

Fuel burnup has been previously determined mainly by mass spectroscopy and by radiochemical analysis of fission products. These methods are destructive to the sample. Mass spectroscopy has the disadvantage that chemical separation must be done prior to analysis since isobars are not separated by the spectrometer. Radiochemistry is at a disadvantage with the neutron capture products of uranium in that the gamma-ray energies are quite close together for many of these products.

To test the new technique a fuel plate consisting of 25% uranium, enriched to 5% ^{235}U and 75% Al by weight, was irradiated in the MTR for two cycles to an nvt of 1.36×10^{21} neutrons/cm². The transmission of neutrons through the irradiated fuel plate as a function of time-of-flight was then compared to an unirradiated fuel plate using the fast chopper, Figure 1. Burnup was observed in the ^{235}U resonances. The production of new materials in the irradiated sample is indicated by the resonances marked in the figure. These data are presently being analyzed to determine the amount of burnup and the ratio of capture to fission cross sections. Preliminary results indicate a fuel burnup of 55.8% based on the 8.79 eV ^{235}U peak, and a burnup of 63.9% based on the 5.47 eV ^{236}U peak. Table I lists the composition of the sample before and after irradiation as determined from the neutron resonance parameters. In the future, mass spectrometry will be done on the sample to evaluate the consistency of the two methods.

TABLE I

COMPOSITION OF SAMPLE BEFORE AND AFTER IRRADIATION
AS DETERMINED FROM RESONANCE PARAMETERS

<u>Nuclide</u>	<u>Energy of Resonance</u>	<u>I/N Determined From Fit of Data</u>	<u>Atoms After Irradiation</u>	<u>Atoms Before Irradiation</u>
²³⁴ U	5.14 eV	312,350	9.24×10^{17}	9.24×10^{17}
²³⁵ U	8.77 eV	9,800	2.95×10^{19}	6.67×10^{19}
²³⁶ U	5.44 eV	46,000	6.27×10^{18}	-----
²³⁸ U	6.67 eV	237	1.22×10^{21}	1.23×10^{21}
²³⁹ Pu	0.296 eV	-----	-----	-----
	7.76 eV	22,334	1.29×10^{19}	-----
²⁴⁰ Pu	1.059 eV	142,610	2.02×10^{18}	-----
¹⁴⁷ Pm	5.35 eV	362,000	7.97×10^{17}	-----
⁹⁹ Tc	5.61 eV	141,000	2.05×10^{18}	-----
¹³³ Cs	5.83 eV	75,000	3.84×10^{18}	-----
¹⁵² Sm	7.98 eV	1.14×10^6	2.53×10^{17}	-----
¹³¹ Xe	14.3 eV	not yet determined	not yet determined	-----

Fig. 1 Transmission versus time-of-flight for unirradiated and irradiated fuel pieces.

PRESENT APPLICATIONS OF GERMANIUM GAMMA SPECTROSCOPY IN ANALYZING REACTOR COOLANT WATER

A. L. Erikson

Application of the Chemistry Section's lithium-drifted germanium detector has been made to the problem of identifying unusual radio-activities appearing in the gross gamma spectra of MTR and ETR reactor coolant waters or, occasionally, in radiochemically separated samples during routine radiochemical analysis of the reactor coolant water. The standard approach before the application of the Ge(Li) detector was to investigate the chemistry followed by an unknown activity and try to isolate it so that its chemistry, half life, or gamma spectrum (as determined with a NaI detector) would allow identification. Now merely counting the source containing the unknown activity on the Ge(Li) detector is usually sufficient. The superior resolution of the Ge(Li) detector usually enables one to determine photopeak energies of a unknown activity quite accurately and without interference from other gammas.

To calculate accurately the large number of photopeak energies and areas from Ge(Li) spectra with the long turn-around time previously necessary on the computers at Central Facilities, a Fortran IV program was written for the PDP-9 computer and used for this purpose until the GAP program[1] for gamma-ray analysis was developed by Wayne Black's group for the PDP-9. The Fortran program which was developed interpolated the channel locations of photopeaks and corrected channel locations for analyzer nonlinearity, summed the photopeak areas correcting for Compton and background contributions, and calculated photopeak energies from the energies of known peaks in the spectrum. Corrected photopeak areas and energies were output. Because of the improvements in the GAP program over the Fortran program the analysis of unknown nuclide gamma spectra is done now using the GAP program.

Quantitative analysis of radionuclides by germanium counting has been made possible by a determination of the efficiency of the Chemistry Section's Ge(Li) detector at three geometries. The first self-efficiency curve along with results comparing NaI and Ge(Li) analyses of some chemically separated reactor process water radionuclides and radionuclides in a gross Retention Basin Holding Tank sample have been previously reported[2].

The use of the Ge(Li) detector for the complete analysis of reactor water is hindered by the fact that the most abundant radioactivity in the reactor coolant water is 15 hour ^{24}Na which has several high energy gammas. The Compton contribution of these gammas to the reactor water gross gamma spectrum is sufficient to swamp out the photopeaks of some of the needed lower intensity activities.

If the large ^{24}Na activity could be removed there would be a better chance of analyzing the lower level activities. Retention of sodium activity on a hydrated antimony pentoxide column from a sample 6 M in hydrochloric acid has been presented by Girardi and Sabbioni of the Euratom Joint Nuclear Center in Italy. A batch equilibration for 15 minutes with ETR reactor water 6 M in HCl using hydrated antimony pentoxide resulted in a decontamination factor (DF) of 300. Column separation has not been tried, but using this technique DF's of 10^6 have been reported. Retention of sodium activity was also tried on a specially

prepared sodium chloride column by placing a small volume sample containing a drop of HCl on the column and eluting with acetone. This technique was developed by C. S. Maletskos of the Harvard Medical School and Massachusetts Deaconess Hospital. Evaporation of 200 mls of ETR reactor coolant water to a small volume and applying Maletskos' technique completely removed ^{24}Na activity with a DF of greater than 10^5 . Investigation of these methods is continuing. The possible retention of activities other than sodium must be evaluated.

- [1] R. C. Davis et al, "Gamma-Ray Analysis Package", this report.
- [2] H. S. Cole et al, "Use of Lithium-Drifted Germanium Detector for Radiochemical Analysis", in D. R. deBoisblanc, W. C. Francis and R. G. Fluharty, Nuclear Technology Branches Quarterly Report - January 1 - March 31, 1967, IN-1117 (September 1967), pp 39-42.

GAMMA-RAY ANALYSIS WITH ON-LINE COMPUTERS

R. C. Davies, G. C. Johnson^[1], E. W. Killian, C. W. Richardson, W. W. Black

A package of programs has been written to do gamma-ray spectrum analysis on a small computer system. The programs were written to greatly shorten the data reduction time. In the past the analysis of a gamma-ray spectrum has involved the following steps:

The spectral data are transferred to punched cards

This card deck is used to obtain a computer listing and plot of the data

By correlating the information contained on the listing and the plot, a set of control cards were prepared for an analysis program

Typically, at least one or more computer run is required either to change control parameters and/or to correct key punching errors.

By direct user interaction with a computer-driven oscilloscope, the present approach enables the researcher to complete the analysis of a spectrum in a few minutes. Experience to date has shown that the present approach allows data to be analyzed approximately a factor of 20 times faster. A detailed description of how to use the program is given in Reference 2.

The package of programs described here was written for a computer system containing the following components:

- (1) A Digital Equipment Corporation PDP-9 computer with 8192 words of memory and the extended arithmetic element
- (2) At least one DECtape transport
- (3) The type RM09 drum

- (4) The 339 display and display processor with the character and vector generator options added
- (5) A special ten key function box.

The following is a short functional description of each of the programs that are now in the package. It is not intended that this package of programs will satisfy all the needs for gamma-ray spectrum analysis. However, it is intended that those described here will form the basic nucleus from which any package can be derived. The system has been set up in such a way that users may add their own programs at will. The present package provides the basic functions of display, peak analysis, and energy determination. A short description of each program presently in the package follows:

DRMCOM. This program permits the user to take the gamma-ray spectral data and put it on the PDP-9 drum. At present, the data may be in the form of a paper tape from a multi-channel analyzer or may be on a PDP-8 DECTape. Eventually the program will allow data to also be read from an IBM compatible tape. Because there is presently no IBM compatible tape transport on the PDP-9, it is necessary to convert IBM compatible tapes to DECTape. DRMCON also allows the user to enter any kind of identifying commentary about the spectrum that is read in. Once a spectrum is placed upon the drum it may be saved for any future use by writing it back on DECTape with one computer command.

PKLOC. This program provides three things: (a) the display of the data, (b) recovery of raw data (channel number, counts, etc), and (c) performing Gaussian fits to individual peaks.

EDIT. It can be expected that errors will be made in the normal routine of obtaining Gaussian fits to the photopeaks. In case channels for the fits are deemed undesirable or peaks are classified in the wrong category, etc, the EDIT program provides the means for correcting these mistakes and entering any additional data that are desired. Three types of information can be operated on by this program, each type having two subgroupings. The first type of data is the list of centroids of peaks. The two subgroups within this type are the centroids of the calibration peaks and the centroids of the unknown peaks. The second type of data is the coefficients and correction data involved in analyzing gamma-ray spectra. The first subset is the coefficients that describe the non-linearity of the ADC systems in use, and also other parameters associated with the detector efficiency table. The second subgroup is the efficiency data (tables) for the detector being used. The third type of data is raw intensities associated with the photopeaks. The first subgroup is the intensities for the unknown peaks and the second subgroup is the intensities associated with the calibration peaks. The EDIT program allows all of these various data sets to be displayed on the 339 oscilloscope screen and also to be listed on the teleprinter.

ANAL. This program computes the energies and the intensities which are corrected for the efficiency of the detector. It, of course, receives its input data from previous programs described above with the exception of the energies associated with calibration peaks. These energies are entered at the time ANAL is executed.

It is important to note that the four programs that make up the program package may be used in any order desired. However, it is important to also remember that in some cases data that are required as input for one program

may be generated by another program. For example, the program ANAL requires as its input the centroids of the photopeaks and the coefficients that describe both the linearity and intensity of the analyzer system being used. In this example, these data may be provided in more than one way. For example, all the data could be provided by first calling the EDIT program and inserting the necessary values. More typically, the centroids would be provided by the program PKLOC.

As an example of how the program package might be used in a typical situation, look at the following two cases: In the first case assume that an estimate of the gain and zero for the analyzer system is known. Then, the first program called would be DRMCON. With this program the data would be read into the computer either from PDP-8 DECTape or paper tape and stored on the drum. At this time any identifying information about the spectrum would be entered. Next, the program PKLOC would be called. With this program centroids would be obtained for all the photopeaks in the spectrum. Figure 1 shows a typical oscilloscope display using PKLOC. Note that one channel in the center of the screen is displayed with a brighter intensity. All the numeric information shown relates to the "bright dot" channel. The two numbers in the top line are the bright-dot channel number and the energy in keV of the bright-dot channel, respectively. The third number is the number of counts in the bright-dot channel. Once the photopeaks were obtained the program EDIT might be called to add any additional information or correct any errors that may have been made in the fitting of the photopeaks. Finally, the program ANAL would be called to obtain the energies corresponding to the photopeaks and the intensities corrected for the efficiency of the detector. Figure 2 shows a typical output obtained from ANAL.

In the second case assume that the user has no estimate of the gain and zero of the analyzer system for the data to be analyzed. In this case, the user would again read the data onto the computer drum using the program DRMCON. Then the program PKLOC would be called into core and as many of the calibration peaks as desired fit. After these few peaks have been fit then the program ANAL could be called into core and the gain and zero would be computed from the centroids of the calibration peaks. At this point the user would then return to the program PKLOC and fit all the peaks in the spectrum. As before, the procedure from this point would be to use the programs EDIT and ANAL in that order.

It should again be emphasized that the two sequences of usage described above are only typical situations and that the programs may be used in any order desired with the restraints of providing the proper input data.

To obtain intensities corrected for detector efficiencies and to obtain correct energies, it is necessary to have information on the linearity and the detector efficiencies of the analyzer system in the computer's memory. At present the linearity corrections and the efficiency corrections are being handled in two different ways. The linearity corrections are obtained from a set of coefficients. These coefficients are those obtained from a power series fit to the linearity data. Intensity corrections are computed from a look-up table in the same manner that has been done on the large computers in the past. The linearity coefficients and intensity table are read into the computer from a paper tape. The input dialogue for ANAL asks if new intensity and linearity data are to be read in. If the answer is yes, the appropriate paper tape is read

in at this time. The table is limited to a maximum of five sections with a maximum of 20 entries in each section.

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- [1] Now employed at UNIVAC Corp.
 - [2] R. C. Davies, G. C. Johnson, E. W. Killian, C. W. Richardson, W. W. Black,
A User's Manual for GAP, Idaho Nuclear Corp. Manual.

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Fig. 1 A typical oscilloscope display generated by the program PKLOC. A portion of a gamma-ray spectrum is shown. The alphanumeric information in the upper-left hand corner of the screen relates to the center data point which is displayed with a brighter intensity. The two numbers in the top line are the channel number of the "bright dot" and the bright-dot energy in keV, respectively. The third number is the number of counts in the bright-dot channel.

+C
DRUM MONITOR

*K

MONITOR V3C

\$E ANAL

INPUT NEW COEF (Y=YES,N=NO)

Y

TYPE IN THE ENERGY OF PEAK LOCATION 288.8791

278.83

TYPE IN THE ENERGY OF PEAK LOCATION 894.0164

892.01

TYPE IN THE ENERGY OF PEAK LOCATION 1094.0399

1090.035

FIRST SET OF LINEARITY COEF

-0.86644397E+01

0.95999040E-01

-0.42766429E-03

0.95727091E-06

-0.10696298E-08

0.47034578E-12

SECOND SET OF LINEARITY COEF

0.58035989E+00

-0.094000629E-03

-0.11869576E-05

0.86962973E-09

-0.17042402E-12

0.11326651E-16

ZERO SHIFT IS 12.223229 CHANNELS

GAIN IS 1.010005 KEV/CHANNEL

	LINEARITY CORRECTION	CALCULATED ENERGY	CORRECT ENERGY	EFF.	INTENSITY
326.2049	-0.0128	317.1102		0.118E-02	0.28194E+09
681.1505	-0.3880	675.2281		0.265E-03	0.26412E+08
979.3588	-0.8440	975.9594		0.140E-03	0.31458E+08
288.8791	-0.0722	279.3509	278.8300	0.158E-02	0.12382E+09
894.0164	-0.7198	889.8887	892.0100	0.163E-03	0.54089E+09
1094.0399	-0.9997	1091.6307	1090.0350	0.115E-03	0.29546E+09

STOP 000000

DRUM MONITOR

*

Fig. 2 A representative output from the program ANAL. The user has indicated that he wants to enter new coefficients which describe the linearity and efficiency characteristics of the detector system. Following this the energies corresponding to the calibration photopeaks were entered. This was followed by the print-out of the linearity coefficients and the calculated zero shift and gain for the detector system. Finally, the results of the energy analysis are printed in columnar fashion.

III. SOLID STATE DETECTOR DEVELOPMENT

GERMANIUM DETECTOR PULSE SIMULATOR

E. E. Owen

A multipurpose long-tail pulser has been developed for the Nuclear Technology Branch. It was designed primarily to test the linearity of germanium detector pulse counting systems. The instrument provides two independent channels of pulses and one channel that is the inverted sum of the other two.

A block diagram of the system is shown in Figure 1. The unique property of this design is its ability to combine the A and B output pulses in a summing amplifier producing a composite pulse similar to the pulse pile-up experienced in most pulse counting systems.

A waveform photograph is shown in Figure 2. Pulse channels A and B have independent and separate front panel controls to adjust the delay, rise time, fall time, polarity, and amplitude. The wide band DC amplifier output drivers load into 50 ohm continuously variable output attenuators that provide infinite resolution of the signal while maintaining a constant output impedance. Some other specifications are:

Rep. Rate: 100 Hz, 1000 Hz, or externally triggered

Rise Time: 30 nanoseconds to 1 microsecond

Fall Time: 10 microseconds to 1 microsecond

Delay: 0.25 microsecond to 70 microseconds

Amplitude: 0 to 1 volt (50 ohm load)

Polarity: Positive or Negative

DC offset available on all 3 channels.

Fig. 1 Germanium detector pulse simulator.

Fig. 2 Photograph of typical waveform.

HIGH SPEED AMPLIFIER

L. O. Johnson

A high speed feedback stabilized amplifier stage was designed and tested. Applications of the amplifier stage range from use as a current sensitive preamplifier in high count rate gross counting applications to use as a high speed, gain stable amplifier stage in short clip delay line amplifiers.

Referring to Figure 1, high speed is accomplished through use of an input common base transistor, Q-1, feeding the base of a common emitter gain stage, Q-2. The common base transistor serves as an impedance matching device to provide low input impedance plus a nearly ideal high impedance driver for the common emitter transistor, Q-2. This impedance conversion provides for maximum gain with minimum phase shift in the amplifier stage. System open loop gain is as:

$$\frac{R_{18}}{R_{12} + r_e(Q-1)} \times \beta(Q-2).$$

Q-3 and Q-4 serve as low impedance output drivers. Q-3 and Q-4 are included in the feedback loop to ensure gain stability.

The amplifier stage may be operated as a voltage amplifier with a gain of 10 and rise time of 15 ns or as a current-to-voltage converter with current-to-voltage conversion gain of 2000 with a rise time of 15 ns.

Fig. 1 High speed amplifier.

LOW NOISE FET PREAMPLIFIERS

L. O. Johnson

Special problems associated with coincidence systems, rise time studies, and high voltage detectors necessitate the design of new high speed preamplifiers with input DC coupling. A new FET was found to replace the 2N3823 for higher input capacity loadings.

It was found that with the high voltage detectors developed in this laboratory, excessive microphonic noise was introduced due to the presence of high voltage on the input leads of AC coupled detector-preamplifier systems. Microphonic noise was substantially reduced by direct coupling one side of the detector to the gate lead of the input FET.

Rise time studies were found to be limited by slow rise times in the preamplifiers. A new fully bipolar charge sensitive preamplifier exhibiting rise times of less than 75 ns with 50 pf external capacity was designed and built (see Figure 1). Its noise performance was identical to previously reported preamps. The open loop gain of the preamplifier is very high (100,000) to enhance gain stability and linearity over a wide dynamic range.

Two new T I FET's (a TIS-75 and SFB-8558) have been found to have noise characteristics superior to any combination of 2N3823 FET's for input capacity loadings in excess of 15 pf. Most recent FET noise data (Ge FWHM) is listed in Table I.

Fig. 1 Charge sensitive preamplifier.

TABLE I

FET NOISE DATA

FET	Zero External Capacity		50 pf External Capacity	
	Room Temperature (keV)	Cooled to Optimum (keV)	Room Temperature (keV)	Cooled to Optimum (keV)
2N3823	0.52	0.36	2.8	1.9
2N4416	0.415	0.37	2.55	1.85
TIS-75	0.74	0.46	1.75	1.05
SFB-8558	0.55	0.38	1.65	1.05

INSTRUMENTATION IMPROVEMENTS IN THE PROTON-RECOIL SPECTROMETER

J. W. Rogers, L. O. Johnson

Proton-recoil spectrometers have been used extensively for neutron spectrum measurements in the CFRMF and the filtered MTR neutron beams. The spectrometer has performed well with very few component failures and has demonstrated excellent stability. The spectrometer has been successfully interfaced and operated with an on-line digital data processing computer (PDP-9).

The addition of a live timer system has been made to the spectrometer instrumentation. The live timer was incorporated so that very accurate live times for the spectrum measurements could be measured. This is necessary for the proper interpretation of the proton spectrum data and for determining the absolute magnitude of the neutron flux. The live timer is composed of a clock time base and a dual OR gate.

The clock time base is a Nuclear Data #536CTD unit which can be gated on or off and will, after reaching a preset time, generate a stop signal. The dual OR gate was designed and built to gate off the clock time base while the ADC is busy or while an overload pulse is being rejected or both. Figure 1 is a circuit diagram of this unit. The location of this device is depicted in the proton-recoil spectrometer block diagram shown in Figure 2.

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Fig. 1. Circuit diagram of OR gate used with proton recoil spectrometer.

Fig. 2. Block diagram of the proton recoil spectrometer.

A DIRECT-COUPLED GAMMA-RAY SPECTROMETER FOR HIGH COUNTING RATES

L. O. Johnson

The full potential of high-resolution gamma-ray spectrometry with Ge(Li) detectors has been severely hampered by degradation in the quality of the pulse-height spectrum at high input data rates. This usually results from three generally recognized effects: (a) rate-dependent baseline shifts, (b) pulse peak pile-up, and (c) pulse tail pile-up. Conventionally, rate-dependent baseline shifts have been minimized by the use of a DC restorer, and much work has been done on its development. However, since the optimum baseline restorer for high-rate applications has not been the optimum restorer for low rate applications, some peak broadening has occurred with its use. Peak and tail pile-up rejection circuits have generally required delay of the primary pulse train, and rejection has been accomplished through a linear gate external to the ADC. This also may result in resolution degradation, even at low rates.

A new application of low drift circuitry has been explored to facilitate high counting-rate pulse-height analysis. A direct-coupled system featuring peak pile-up rejection, tail pile-up rejection, RC shaping and, uniquely, no DC voltage restoration or primary pulse train delay has been built.

The output charge from a high quality Ge(Li) detector is direct-coupled into a low noise, cooled, FET charge-sensitive preamplifier which is stabilized against DC drift. The long-tailed preamplifier output pulse is direct-coupled to an RC shaping amplifier composed of commercially available low-drift chopper stabilized amplifiers. The shaping amplifier output is direct-coupled to the linear gate of the ADC. No coupling capacitors are used anywhere in the primary pulse train path.

A system block diagram is shown in Figure 1. As shown, peak pile-up rejection is accomplished by clipping a preamplifier output at 50 ns and amplifying this signal in a fast amplifier. The amplifier output feeds a discriminator whose output is a constant amplitude, 50-nsec-wide pulse for each preamplifier pulse above noise level. The discriminator output and ADC busy signal feed an anti-coincidence gate such that every preamplifier pulse generates a 50-ns-wide gate output pulse whenever the ADC is not busy. The trailing edge of the gate output pulse triggers a 3- μ sec single-shot. The single-shot output and anti-coincidence gate output feed a coincidence gate. The 3- μ sec single-shot overlaps the ADC peak-voltage detection time, so that a reject pulse from the peak pile-up rejector coincidence gate is generated any time a second preamplifier pulse occurs in the time period from 50 ns after the first preamplifier pulse occurs until the ADC detects the peak value of that pulse. Pulse rejection is completed in the ADC reject circuitry. Thus, peak pile-up rejection is accomplished with no delay of the primary pulse train. Tail pile-up rejection is accomplished by examining the shaping amplifier output with a sensitive DC comparator (baseline discriminator). The baseline discriminator output and ADC busy signal feed a bistable flip-flop whose output controls the ADC linear gate. Conditions which must be satisfied to open the linear gate are (a) ADC not busy and (b) baseline returned to the noise level. The only condition that will close the linear gate is that the ADC be busy.

Some results are shown in Figures 2 and 3. Resolution degradation in the Co^{60} 1.33 MeV line is seen to be about 30%, from 1.92 keV at low rates to 2.50 keV at 75,000 cps. The effect of baseline inspection (tail pile-up rejection) is seen to be insignificant up to 50,000 cps. Above this rate, high-side tailing is significantly reduced through its use. No short-term rate-dependent peak shift is detectable with proper pole-zero adjustment. A long-term rate-dependent peak shift is thought to be due to component heating. These results were taken from a breadboard system which is presently being optimized in a printed circuit board layout.

Fig. 1 System block diagram.

1.33 MeV Co^{60}

Fig. 2 Sample spectra obtained with proton recoil spectrometer.

Fig. 3 Sample spectra obtained with proton recoil spectrometer.

ELECTRONIC CIRCUIT ANALYSIS PROGRAM

J. B. Thompson, E. V. Waite

A 360 version of the IBM 1620 Electronic Circuit Analysis Program (known as ECAP) has been obtained and is now operational. This is an integrated system of programs which can be used by the electrical engineer in the design and analysis of electronic circuits. This system of programs can produce DC, AC, and/or transient analysis of electrical networks from a description of the connections of the network (the circuit topology).

A second system of programs known as Sceptre is being obtained through the Air Force Weapons Laboratory, Kirtland Air Force Base. They have just received from IBM an up-dated program to which some revisions have been added, thereby increasing its versatility. When it is released by the Weapons Laboratory, we will receive a copy. Some of the Sceptre features are: stored models, automatic initial conditions, rerun, increase in the number of outputs, subprogram capabilities and economical operation, to name a few. It should be operational some time in September.

MEASUREMENT OF OXIDE COATING THICKNESS ON REACTOR FUEL ELEMENTS BY EDDY CURRENT SENSOR

T. J. Boland, E. G. Grafwallner, E. E. Owen

Aluminum oxides on the surface of reactor fuel plates impair heat transfer to the coolant in water-cooled reactors. This thermal barrier limits the in-service life of the fuel elements. When the oxide coating exceeds a particular thickness the element can no longer be cooled adequately and must be removed from service. Reliable nondestructive measurements to determine the oxide thickness permit extending the in-core life of fuel elements beyond that which is based on a computed average.

An instrument has been designed utilizing eddy currents which accurately measures the thickness of insulating coatings on aluminum and other metals. A 1 MHz magnetic field from a small probe coil placed on the surface of the fuel plate induces eddy currents in the base metal. The eddy currents, in turn, set up a second magnetic field which opposes the magnetic field of the probe coil. The strength of this opposing magnetic field is inversely proportional to the distance between the probe and the aluminum base. A second coil which is essentially identical to the probe coil is used as a reference in a null balance circuit. When the probe and the reference coil are both resting on the same metal surface, a null balance obtains. If the coil is moved away from the metal surface an unbalance results. Displacement of the reference coil the same distance as the probe again restores the null balance.

Oxide thickness measurements must be made while the fuel element is under eight or more feet of water. The probe sensor is placed on the surface of the outer fuel plate. The reference coil and null detector are located at the operating console. The reference coil is mounted in a holding fixture which can be moved up or down with respect to a reference metal surface. The displacement of the reference coil from the reference surface is measured

by a precision dial indicator which is calibrated in 50 microinch increments. Interpolation to 12.5 microinches is possible. The gain of the null detector is adjusted to provide a full scale range of 0.0015 inch.

The probe sensor is incased in stainless steel to make it waterproof. A thin (approximately 0.0015 inch) stainless steel window allows the 1 MHz magnetic field to penetrate through it with only a small loss.

Figure 1 is a simplified sketch of the null balance system.

This system is being used to routinely measure the oxide coating thickness on ETR and MTR fuel elements. It was also used recently to measure a special ATR element and will be used to measure the oxide thickness on MTR Phoenix core elements. The Phoenix elements will be measured before use and three to five times during the Phoenix core operation.

Fig. 1 Eddy current null balance instrument.

AUTOMATIC CALIBRATION CORRECTION FOR AN ULTRASONIC LEVEL MONITOR

T. J. Boland

A monitor has been developed for the continuous measurement of liquid or solids materials stored in tanks, silos, bins, or other containments. A single transducer mounted near the top of the containment vessel is used to transmit an ultrasonic pulse and receive the echo from the surface of the liquid or solids material. Because of the low velocity of ultrasonic waves in air, the wavelengths are very short, even at relatively low frequencies. The resolution is therefore better than can be obtained with most other methods.

The accuracy of calibration of an ultrasonic level monitor is subject to errors caused by change of temperature and/or density of the air medium according to the relation:

$$c = \sqrt{\frac{\gamma P_o}{\rho_o}}$$

where

$$\gamma = \text{ratio of specific heats, } = \frac{C_P}{C_V}$$

C_P = specific heat at constant pressure

C_V = specific heat at constant volume

P_o = static pressure

ρ_o = density.

The velocity of sonic waves in gases increases with temperature approximately 0.6 meter/second/°C. This becomes a problem when a temperature or density gradient exists along the signal path between transducer and a monitored reflective surface.

Radioactive waste solids are stored in large underground tanks at the Idaho Chemical Processing Plant. These solids materials are thermally as well as radioactively "hot". An ultrasonic transducer mounted at the top of the waste storage tank operates in approximately a 100°F environment. The temperature of the solids waste material is upwards of 1000°F. There exists, therefore, a large temperature gradient and the ultrasonic pulse signal accelerates toward the surface of the solids and the reflected signal decelerates as it travels back toward the transducer.

Since the basis of the distance or level measurement is the time of travel of the signal, it is necessary to correct the calibration errors caused by the changing signal velocity. A system has been developed which accomplishes calibration error correction automatically. This is shown in block diagram form in Figure 1. Small marker reflective plates are installed along the signal path. These are uniformly spaced starting from the transducer face. The echo pulses from these markers are of smaller amplitude than the echo from the surface being monitored (level of the stored solids in this case). With reference to the block diagram, a three digit in-line decimal counter is used for level readout. The marker pulses are counted in the "tens" decade. Pulses from the clock-rate generator are counted in the "units" and "tenths" decades. The clock-rate generator nominally produces 100 pulses between marker pulses. The units and tenths counter is reset each time a marker pulse is received. If the time between marker pulses changes, the clock-rate is changed accordingly. This is accomplished by dividing the clock frequency by 100 and comparing this divided frequency with the marker pulse frequency. An error voltage proportional to the difference in rates is applied to the voltage controlled clock-rate generator to cause the divided clock-rate to coincide with the marker rate. The readout is stopped for display or recording by the level pulse.

It can be seen that the "tens" digit is always a true reading of the range or level. The units and tenths digits are subject to an error due to velocity changes which occur after the last marker pulse correction. This error follows the relations:

$$\text{Error} = \left(\frac{V_i - V_t}{V_t} \right) d$$

where

V_i = indicated velocity

V_t = true velocity

d = distance between last marker pulse and the level pulse.

Reducing the spacing between markers decreases the error to whatever value is acceptable. For long signal paths, the markers can be spaced farther apart than would be necessary for short signal paths.

The calibration correction system described was designed for use in a new waste storage facility in anticipation of the problems imposed by the known temperature gradient. Since the waste storage facility is still under construction, the calibration correction system has not yet been tested and proven.

Fig. 1 Automatic error correction system block diagram.

IMPROVEMENT IN THE CHART READOUT OF THE ETR SECONDARY WATER MONITOR

J. B. Thompson

The ETR secondary water monitors are installed in the vicinity of the heat exchangers. Their purpose is to detect the presence of radioactive particles in the secondary water if a leak develops between the primary and secondary systems. The monitor utilizes a multipoint strip chart recorder for information readout. The system as originally designed was not satisfactory because the desired average count rate was masked by the large fluctuations in the instantaneous counting rate. The counting rate is low so the statistical variation in the output voltage from one printout to the next produces a wide trace on the recorder chart. With four channels being printed on the same chart the points were so intermingled it became difficult to detect any deviation from the normal average output level of any one of the channels.

In low counting rate systems the output signal is a composite of the instantaneous pulses from the detector. The receiver requires a given bandwidth to pass this information, thus in the output there is the desired signal (the average counting rate) and the instantaneous variation about this signal which is considered noise. This noise ranges in amplitude from zero to some finite voltage around the signal thus masking the signal and making it difficult to determine the counting rate or detect when a change occurs. Applying the technique of post-detection filtering, the noise is reduced and the signal is enhanced.

The random noise energy in a given bandwidth of frequencies is given by the expression[1]:

$$\epsilon = \frac{1}{2\pi} \int_{\omega_1}^{\omega_2} E_n(\omega) d\omega \quad (1)$$

In the case where energy is uniformly distributed over the bandwidth of frequencies (Gaussian distribution) this equation reduces to

$$\epsilon = \frac{E_n}{2\pi} [(\omega_2 - \omega_1)] \quad \text{or} \quad E = \sqrt{\frac{E_n}{2\pi} Bw} \quad (2)$$

where

E_n = energy source

Bw = bandwidth ($\omega_2 - \omega_1$)

ϵ = (energy) = E^2 (voltage squared).

[1] Private communication from T. J. Boland.

For the low pass filter, $\omega_1 = 0$ and $\omega_2 = \omega_c = \text{cutoff frequency}$.

The Fourier transform pair

$$\left\{ \begin{array}{l} f(\tau) = \int_{-\infty}^{+\infty} g(\omega) e^{j\omega\tau} d\omega \end{array} \right. \quad (3)$$

$$\left\{ \begin{array}{l} g(\omega) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} f(\tau) e^{-j\omega\tau} d\tau \end{array} \right. \quad (4)$$

show the interchangeability of time and frequency.

The transfer function of a network is the ratio of its output to its input $\frac{E_o}{E_i}$. For a simple RC low pass filter

$$\frac{E_o(s)}{E_i(s)} = \frac{1}{\tau s + 1} = \frac{1}{\tau s + 1} \quad (5)$$

where

$$\tau = RC$$

and

$$\frac{1}{\tau s + 1} = \frac{\frac{1}{\tau}}{s + \frac{1}{\tau}} = \frac{j\omega}{s + j\omega} \quad (6)$$

which relates the cutoff frequency ω_c to a time τ where $\omega_c = \frac{1}{\tau} = \frac{1}{RC}$.

The strip chart recorder has a response time of one second which represents a bandwidth of 1 cps = $\frac{\omega_c}{2\pi}$. The output voltage due to noise is represented by

$$E_{o1} = \sqrt{\frac{E_n^{\omega} c_1}{2\pi}} \quad \text{or} \quad E_{o1} = \sqrt{\frac{E_n}{2\pi\tau_1}} \quad (7)$$

Now if an RC filter is added to the system, the output noise voltage becomes

$$E_{o2} = \sqrt{\frac{E_n^{\omega} c_2}{2\pi}} = \sqrt{\frac{E_n}{2\pi\tau_2}} \quad (8)$$

The noise will be reduced by the ratio

$$\frac{E_{o2}}{E_{o1}} = \frac{\sqrt{\frac{E_n^{\omega} c_2}{2\pi}}}{\sqrt{\frac{E_n^{\omega} c_1}{2\pi}}} = \sqrt{\frac{\omega_{c2}}{\omega_{c1}}} \quad (9)$$

or

$$E_2 = E_1 \sqrt{\frac{\tau_1}{\tau_2}} \quad (10)$$

which states the noise voltage will be reduced by the square root of the ratio of the time constants. It is evident that an improvement in the signal-to-noise voltage is purchased with time which is directly related to the change in the time constants. The limiting factor for signal-to-noise improvement occurs when the bandwidth is reduced beyond the point where the system will no longer respond to the desired signal change or when the time involved is too long to be practical.

This filter has been built and installed in the circuit. The results can be observed in Figure 1, which is a reproduction of a recorder chart before the addition of the filter, and Figure 2, which is a chart made after installation of the smoothing filter. For this comparison, sections of the two charts were selected where the average values of the signals are approximately equal. In the chart from which Figure 1 was reproduced, there were areas where the width of a channel was approximately two inches. A reduction of 20 would bring this width down to 0.1 inch. The theoretical reduction based on the components used is 19.2. A study of these charts indicates the noise voltage has been substantially reduced so that changes in the signal can be readily observed.

Fig. 1 Reproduction of recorder chart before addition of filter.

Fig. 2 Reproduction of recorder chart after addition of filter.

AUTOMATIC SAMPLE FUEL PLATE RADIOGRAPH DENSITOMETER

F. A. Meichle, R. L. Sumstine, E. G. Grafwallner, N. S. Graham

Evaluation of fuel element plate radiographs for the test reactors consists of a visual and densitometer inspection. Both inspections are manual operations. As the number of fuel plate inspections increased, manpower availability and the possibility of errors, due to the physiological fatigue of the inspectors, initiated the development of a prototype densitometer to inspect the fuel plate radiographs. The prototype was designed to scan automatically a radiograph, sense the radiograph density and density location (X-Y coordinates), digitize the three signals, and format and record the signals for data processing. To minimize development costs, the prototype was designed to inspect sample fuel element plate (5.75 x 1.25 inch) radiographs and to use in-house components which were available.

The prototype densitometer evaluation will investigate optical and density sensing techniques, fuel-to-film density correlation, data acquisition, and computer programs. A computer was not considered as part of the densitometer because of the large number of calculations, storage requirements, and plotting graphic which can be performed on the data. The data processing facilities are available in-house.

The prototype densitometer block diagram is shown in Figure 1. The major components are the film transport, light source, light sensor, amplifier, multiplexer, controller, analog-to-digital converter (DVM), format generator, and paper tape punch.

The transport incorporates the film holder, drive motors, position transducers, and limit switches. Starting from a specified corner the transport moves the film between the light source and sensor. The film is scanned back and forth along the X-axis while incrementing along the Y-axis.

The light source beam is collimated and the light sensor positioned so that a circular area of 0.080 inch on the film is inspected. The light sensor is an SGD-100[1] photo diode chosen for its low leakage current, linearity, and stability. The output of the photo diode is amplified and fed to the multiplexer. Two other signals (X and Y axis positions) are also fed to the multiplexer. The multiplexer, driven by the controller, scans the three signals, feeding them sequentially to the digital voltmeter (DVM).

The digital voltmeter, supplied with digital output, is used as the analog-to-digital converter. The information in the DVM is gated out in parallel to the format generator. The format generator provides the paper tape format, parallel to serial conversion, timing, and drive for the paper tape punch. Additional functions as paper tape leader and run identification are accomplished by the format generator.

The information is punched on the paper tape in the following form: X-axis position (3 digits), space, Y-axis position (3 digits), space, photo diode reading (3 digits), double space, X-axis position, etc.

[1] EG and G, Electronics Division.

Readout intervals during a scan are accomplished by monitoring the X-axis position signal. The position signal is analog and derived from a potentiometer. Applying the X-axis position digital signal to the controller, the controller logic senses the number "eight" in the least significant digit. Whenever the "eight" is sensed, the controller initiates a multiplexing operation. The Y-axis incrementing is accomplished in the same as the X-axis under controller control.

A computer program has been written to assemble the paper tape information into a X and Y map, plotting the densitometer readings. The program also computes the number of readings, sum, and average. The program was extended to map all values which exceed a selected percentage value above the average value. Percentage selection is under program control. Zeros replace all densitometer readings whose values are under the selected percentage.

The program is presently being extended to compute the areas and locations of the reading exceeding the selected percentages. The areas computed will then be compared with the fuel homogeneity specification for classification.

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Fig. 1 Automatic sample fuel plate densitometer.

NITROGEN LEVEL DETECTOR FOR USE WITH Ge(Li) DETECTORS

B. G. Nelson

Operation of Ge(Li) detectors requires the use of a cryostat and a dewar containing liquid nitrogen to maintain the detector at the required temperature of 77° K. To prevent damage to the solid state detectors, the nitrogen level in the dewar must be maintained at or above a minimum level. This is accomplished by refilling the dewars periodically. In an installation where a number of cryostats are in use, detectors can easily be ruined through inadvertent warming due to failure of the dewar, vacuum failure of the cryostat, or failure to fill the dewar, etc. To provide a warning when the nitrogen level is low, a device that senses the nitrogen level has been developed. It provides both audible and visual alarms when the nitrogen level drops below the sensor.

The sensitive unit in the instrument is a zener diode whose voltage characteristic changes approximately 25% when immersed in liquid nitrogen. The discrimination between the liquid and gas phases in the dewar is improved by operating the diode at a power level. In these units a 1N4747 (20 volt) diode is operated at one-half watt.

The schematic diagram of the instrument is shown in Figure 1. The operation is as follows: When the liquid level drops below the diode, the temperature of the diode increases, causing the voltage drop across it to increase. This decreases the voltage at the emitter of Q1 and it turns off. The collector voltage of Q1 rises turning on transistor Q2. Transistor Q2 applies voltage to transistors Q3 and Q4 which form a free running multivibrator. The visual alarm is produced by the flashing of lamp L1. The audible alarm is from a "sonalert" unit connected across lamp L1. The potentiometer R1 sets the alarm level.

For our "dip stick" cryostats the zener diode is mounted in a nylon ring which slips on the end of the cold finger. Power for the alarm unit is obtained from the preamp power supply.

The liquid nitrogen level detector has been installed on several solid state detector assemblies. These units have operated as designed; they alarmed when the nitrogen level dropped below the sensor. It is recommended that all cryostats be equipped with the nitrogen level detector.

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IV. EXPERIMENTAL APPARATUS AND ELECTRONICS

COMPUTER SYSTEM FOR GAMMA-RAY ANALYSIS OF LUNAR SAMPLES

W. W. Black, C. W. Richardson

A digital control panel has been added to the computer system for the low-level counting facility at the Lunar Receiving Laboratory in Houston, Texas. This facility is used to do gamma-ray spectroscopy analysis of lunar samples. The panel facilitates the operation of the computer display and other equipment. Figure 1 is a picture of the panel which shows the switch labels for the main program used on the system.

The panel is divided into three sections: (1) READ OUT, (2) EXPERIMENT CONTROL, and (3) DISPLAY CONTROL. The READ OUT section allows the operator to select the device as well as the information to be output. For example, to get a list of the scaler contents the device switch would be set to TELETYPE and the data switch would be set to SCALERS. The readout is initiated by pushing the START button.

The EXPERIMENT CONTROL section contains two switches for selecting timing constants and buttons for erasing data and turning the analyzer on and off. To protect against accidental loss of data, the erase button is not operable unless the erase enable button is also pushed.

The DISPLAY CONTROL section is used to specify the information and how it is to be presented on the scope. The positions labeled F, M, SUM, and PLANE select the four different two-dimensional arrays to be placed on the scope in the form of a point plot. The position F/M will select an overlapping display of both the F and M spectra. The spread button can be used to separate the spectra vertically. Other switches determine the number of channels in the display, the counts full scale for the linear plot, and the decades for the logarithmic display. A button is used to change from log to linear. If the number of channels selected is less than the length of the spectrum, the other channels can be rotated through the visible area. The direction and speed of rotation can also be controlled with the buttons. The position marked CONTOUR displays a three-dimensional array by intensifying points in a two-dimensional grid. The third dimension can be visualized since only those grid points in a narrow slice are intensified. Using the buttons the slice can be raised and lowered to study the array.

The values of such quantities as the percent live time, the scaler contents, and the number of error events can be displayed on the scope in the form of decimal digits. Two switches are used to select 15 different quantities.

A complete documentation of the computer programs and the digital control panel may be found in Reference 1.

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- [1] W. W. Black and C. W. Richardson, User's Manual for the Multiparameter Data Acquisition Program COINC Version 4 with Teletype and Panel Controls, Idaho Nuclear Corporation Manual.

Fig. 1 Control panel functions for the low level counting facility at the NASA Lunar Receiving Laboratory.

LOW FREQUENCY DIGITAL FILTERING IN THE TIME DOMAIN

A. L. Connelly

A digital computer will be used in performing noise analyses on the Advanced Test Reactor. The essential computational tool for such work is the Fast Fourier Transform for transforming time domain data to the frequency domain. For reasons explained below it is necessary to filter the time domain data before transformation in order to increase the resolution of the frequency domain data and/or filter out certain frequency components. Present plans call for performing such filtering by digital means. To this end, digital filter subroutines are under development for future inclusion in a program package to be used on the ATR computer system in the computation of power spectra and related computations.

A subroutine implementing a hanning filter has been developed and tested in FORTRAN on the IBM 360. This filter is applied to data in the time domain before transformation into the frequency domain. The object of such a filter is to reduce the interdependence of frequency points in the power spectrum derived from the Fourier transform of the time domain data. This interdependence arises from the fact that only finite data records are transformed, and it results in a smearing of the frequency domain data, ie, lower resolution. It is intended to develop a similar subroutine in MACRO-9 for the PDP-9. Figure 1 shows raw frequency domain data before the application of a hanning filter routine; the result of such an application is shown in Figure 2.

A low-pass digital filter has been developed and tested on the IBM 360 in FORTRAN. The method of filter design suggested by Ormsby[1] has been followed in the development of the filter. The purpose of this filter is to obtain the low frequency components ($f \leq 1$ Hz) of the ATR power spectrum with a minimal data storage requirement and the least possible computation while minimizing Nyquist folding. An implementation of the filter on the PDP-9 in MACRO-9 is in progress. In this latter development only integer arithmetic is used in the first stages of computation. The filtered data are then stored in floating point. Figure 3 shows the result of applying the Ormsby type low-pass filter to the data of Figure 1.

Study is being carried on into recursive digital filtering techniques in hope of gaining further reduction of storage and computational requirements in filtering.

[1] J. F. A Ormsby, "Design of Numerical Filters with Applications to Missile Data Processing", J. Assoc. Compt. Mach., 8 (1961) pp 440-466.

Fig. 1 The ordinate is in terms of arbitrary intensity units, and the abscissa is in terms of arbitrary frequency units. The time domain data that are the source of this spectrum is also used in Figures 2 and 3.

Fig. 2 The axes are as in Figure 1. The unfiltered spectrum is the same as Figure 1. The hanning filter was used to obtain the filtered power spectrum.

Fig. 3 The axes are as in Figure 1. The longer effective data length that goes into the computation of this spectrum is responsible for the higher resolution. The small perturbations of the power spectrum are due to leakage of higher frequency components of the data through the filter and their folding over into the lower frequency portion of the spectrum by the Nyquist folding phenomenon.

SPC-12 -- PDP-9 BI-DIRECTIONAL DATA TRANSFER INTERFACE

R. A. Coates, G. O. English, E. E. Owens

A General Automation model SPC-12 control processor, which is a small 8-bit, 4 K word memory computer, has been interfaced with a PDP-9 general purpose computer. The interface allows either computer to stand alone or to communicate with the other through program control. The obvious advantage of this system is the use of many peripheral devices by the PDP-9 through the SPC-12. The SPC-12 can be treated as a relatively inexpensive common interface to these peripheral devices[1] with its own memory and control. It also relieves the use of memory in the PDP-9. Figure 1 is a block diagram of the system.

The SPC-12 -- PDP-9 Bi-Directional Data Transfer Interface actually consists of two parts: the program controlled transfers, which would normally be used for bi-directional instructions, and the DMT (direct memory transfer) option of the SPC-12 via the DCH (data channel) of the PDP-9. This would be used for the transfer of blocks of data between the two computers. All signals are routed through the PDP-9 Common Logic for compatibility between I. C. logic and the "R" series logic of the PDP-9. Figure 2 is a block diagram of the bi-directional interface.

- [1] E. W. Killian, G. B. Morgan, R. E. Brown, "Magnetic Tape Controller", this report.

Fig. 1 A block diagram of the system.

Fig. 2 A block diagram of the bi-directional interface.

ASSEMBLERS FOR ON-LINE COMPUTERS

C. W. Richardson, E. W. Killian, G. B. Morgan

The experiments conducted in this laboratory involve the use of several small digital computer systems. Each system has a unique set of peripherals which are designed for the needs of the experiment. Even though a considerable amount of time must be spent developing the computer programs for each system, peripherals to aid program development are usually not included.

It would be economically prohibitive to purchase the necessary peripherals for each system, and time on the small system is usually not available for program development due to the requirements of the experiment.

The program development for small systems can just as well be done on a large batch processing computer. Symbolic instructions for the small system are punched on cards and assembled to produce a paper tape and a listing. The paper tape contains the numbers to be loaded into the memory of the small computer. Each of our small systems have a paper tape input device. The listing contains a list of the instructions together with the memory map defined by the instructions. The memory map greatly facilitates check-out and development of the small computer program.

The term "assembler" is generally used for the program which processes the symbolic instructions to give the memory numbers. We have written an assembler in FORTRAN IV language which will process instructions for the PDP-8, -8/S, -7, and -9 computers.

Table I is a comparison of the Fortran assembler and the 360/75 machine language assembler. Due to the increased capabilities and efficiencies of the 360/75 assembler we are experimenting with the possibility of using it to assemble programs for small computers. The small computer's instructions are defined using the conditional macro generation statements of the 360/75 assembler. This is done by dividing the small computer instruction into three main parts: the tag, the operation, and the arguments (usually the address). The operation is used for the name of the macro and the arguments are used for the macro operands. Any desired manipulation of the arguments can be written into the macro definition to produce the object code for the small computer. The output from the 360 assembler is input to a PL/1 program to convert to the small computers format.

We are now using this general automation procedure to assemble programs for a General Automation model SPC-12 computer and plan to do the same for other computers as the need arises.

TABLE I

COMPARISON OF FORTRAN AND MACHINE ASSEMBLERS

<u>Fortran IV Assembler For Small Computers</u>	<u>360/75 Assembler for the 360/75</u>
No MACRO handling	MACRO's with conditional generation capabilities
FORTTRAN logic for tape searching	Very efficient machine logic for optimized tape searching
FORTTRAN logic for card scanning	Efficient machine logic for card scanning
Easily modified to handle other computers	Very difficult to modify

COMPUTER CONTROLLED DATA ACQUISITION FOR NEUTRON SCATTERING INSTRUMENTS

W. R. Myers, P. D. Randolph, G. B. Morgan

Three experimental facilities of the Solid State Physics Section have made use of the PDP-9 computer for data acquisition. A burst-mode time-of-flight interface[1] was used both at the High Pressure Neutron Diffraction facility and by the MTR phased chopper velocity selector. The operation of the triple-axis crystal spectrometer was automated using a second interface. An interface for statistical method (correlation or Fourier choppers) neutron time-of-flight experiments is being designed. These experiments were controlled by a Data Monitor Program[2] which has the capacity to handle data acquisition from one of the time-of-flight experiments and from the triple-axis spectrometer simultaneously. Increased efficiency in data acquisition was due to greater component reliability over the conventional analyzers and experimental control instruments, to the greater versatility in executing the experiments, and primarily for the time-of-flight experiments, due to the live display of data.

[1] G. B. Morgan, Time of Flight Interface, IN-1294 (June 1969).

[2] W. R. Myers, DATMON -- A PDP-9 Computer Program for the Simultaneous Acquisition of Data from One Neutron Time-of-Flight Spectrometer and One Neutron Triple-Axis Crystal Spectrometer -- A User's Manual, IN-1287 (July 1969).

MAGNETIC TAPE CONTROLLER FOR PDP-9 SYSTEM

E. W. Killian, G. B. Morgan, R. E. Brown

It has become apparent, as large amounts of data are being analyzed on the PDP-9B computer system, that a method is needed to permit transmission of data between IBM compatible magnetic tape and the PDP-9. With this problem in mind a General Automation Model SPC-12 computer is presently being interfaced to the PDP-9. The SPC-12 will itself be interfaced to an incremental magnetic tape transport. This will provide an indirect path to and from the PDP-9 and tape transport. The SPC-12 will be used as a magnetic tape unit controller for data transmission between the PDP-9 and IBM 7-track magnetic tape. The SPC-12 will control all functions of the tape unit, freeing the PDP-9 for other tasks. It is also envisioned that the SPC-12 will be able to perform some data modification (if desired) before transmission to the PDP-9.

The logic required to interface the SPC-12 to the magnetic tape transport was minimal (under \$200), since the control functions are to be implemented in the SPC-12 software. The single instruction functions that the SPC-12 can perform are listed below:

Command Functions

Write beginning of tape
Rewind tape
Stop capstan motor
Write end of file mark
Write end of record mark
Stop capstan
Stop command to magnetic tape transport
Ready magnetic tape for operation
Advance to load point
Initialize magnetic tape transport
Load command (write, speed, direct)

Test Functions

Test ready status of magnetic tape
Test busy status of magnetic tape
Test for end of record gap flag
Test for longitudinal parity error
Test for vertical parity error
Test rewind busy flag
Test write enabled ring
Check for load point marker

Data Functions

Write 6 bits on tape
Read 6 bits on tape.

The control and timing to allow the primary commands above to read and write records, search, space records, etc, are done in the SPC-12 software package.

AN AUXILIARY COMPUTER SYSTEM FOR THE ATR REACTOR

W. W. Black, G. B. Morgan, N. C. Kaufman, J. E. Anderson, D. D. Metcalf

A second computer system which will serve two primary purposes is to be installed at the Advanced Test Reactor. The first purpose is to provide the sponsors on the reactor loops with added output capabilities at the loop experiments and the second purpose is to provide a means of giving the reactor operators operating information and suggestions.

The loop sponsors had requested additional output capabilities at the loop experiments in the form of five typers and two paper tape punches. Cost studies showed that the most economical and most flexible way of providing this additional output was to install a second small computer system at the ATR. This equipment will also be equipped with request consoles to allow loop operators to request the type of information that they want output on the typers and punches.

This particular computer system will also be supplied with magnetic disk storage and display oscilloscopes. A display oscilloscope will be installed at the reactor console for use by the reactor operators. The first computer

system at the ATR already accumulates a running history of the reactor's operating conditions. At the present time the only means of obtaining this information is via printing devices. The second computer system will supply the capability to get this information on an oscilloscope screen not only in alphanumeric form but in the form of plots. This will permit the operators to ask for plotted histories of various parameters, and even overlapping plots of such histories for a time comparison. Such information and capability will be extremely valuable in analyzing any reactor scrams that might occur. Programs are already under development which exploit an algorithm that has been developed to compute the needed shim settings for the ATR reactor. Therefore, the capability will be provided to give the operator suggestions as to what the shim settings may be at any time via the computer-driven oscilloscope display.

Considerable research has been done on the possibility of digitally acquiring power spectral densities for the reactor. It has been conclusively shown that these power spectra densities may be acquired in a digital manner and that computers presently on the market are fast enough to allow real time acquisition of such data[1]. Therefore, the second computer system will be used to develop the means of supplying power spectra densities and other similar information on a real time basis.

Because of the stringent requirements for up-time on this computer system and the need to make the system operational as quickly as possible, it has been decided to take a newly developed approach to interfacing the various peripherals to the computer. The majority of the peripherals will be interfaced using the CAMAC system. This is a completely modular approach to interfacing computers and is designed to give the maximum independence of interfaces from the computer manufacturer. In this case a CAMAC bin will be acquired which will have slots for each of the typers, punches, and other peripheral interfaces. Only one slot in the bin will be computer-dependent and that will have a controller module in it. In the case of the typers and punches this will provide a very clean approach to the maintenance of these devices since they may be unplugged at any time and taken from the system without any interference to the operation of the reactor. The fact that the module interfaces themselves will be computer independent will mean that the same interfaces may be used on the ATR for any other application at a later time.

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- [1] B. J. Kramer, W. W. Black, A. L. Connelly, "Digital Neutron Analyses," this report.

COMPUTER INSTRUMENTATION SYSTEM FOR THE LMFBR LOOP EXPERIMENT ON THE ETR REACTOR

W. W. Black, G. B. Morgan, F. A. Meichle, R. C. Davies, C. W. Richardson

A sodium cooled loop is to be installed in the Engineering Test Reactor in support of the Liquid Metal Fast Breeder Reactor Program. Computations based on present theoretical models indicate that the failure of a fuel pin in a liquid metal fast breeder reactor might set off a chain of fuel pin failures

throughout the reactor core. The purpose of this experiment is to determine if a fuel pin failure can result in the propagation of such a transient through the reactor core and to find what the possible destructive effects might be. If such propagations can occur, the goal is to collect data which will show how to prevent the propagation of such transients. Present computations show that the total time elapsed in the propagation of a transient will be on the order of 100 microseconds.

Equipment configurations are now being considered and some initial equipment, including one computer system, is now on order. The signals that the computer system will have to handle may be broken into two categories, slow and fast. It is felt that slow signals which are approximately 50 in number will present no unusual problems. To accomplish the digitalization of these slow signals, both binary approximation, analog-to-digital converters, and voltage-to-frequency converters are being evaluated. It is assumed at this time that these signals will be stored on a bulk storage device, like magnetic disks, or magnetic tape, and will be available to the operators and experimenters.

The fast signals may require technical development; several methods are presently being investigated for handling them. Because the total time lapse for a transient is estimated to be of the order of 100 microseconds, extremely large amounts of data may have to be accumulated in this short time. Currently two possible ways of accumulating this data in real time are being investigated.

The computer system that has been selected will have an asynchronous memory bus system. This will allow more than one memory bank of different speeds to be attached to the computer system. Therefore, one of the methods of storing the data is to store it directly in magnetic core memory on a cycle stealing basis. If the normal cycle stealing mode is not sufficient to handle the data rates the asynchronous bus will permit interleaving of computer memory banks. For example, if four memory banks were interleaved, data could be stored at four times the rate of one normal bank.

The second possibility that is being investigated for the possible acquisition of the fast signals is an analog disk. Because the data need be stored only for a short period of time, it should be possible to write the data in real time on an analog disk. Once a transient had occurred the data would be immediately read off the analog disk and digitized. Therefore, the presentation of the digitized data could still be in real time on human reaction time scales.

For monitoring the experiment, a departure is going to be made from the more conventional methods of monitoring reactor loop experiments. Also because the principal investigators of this experiment are normally located at Argonne Laboratory in Lemont, Illinois, there is a requirement for the ability to monitor the experiment both locally at the ETR and at Lemont, Illinois.

Experiment monitoring will be carried out at the ETR by the use of computer-driven oscilloscope displays and digital control panels. It is anticipated that there will be at least one display and control panel which will be available for the reactor and loop operators and one display and control panel that will be available to the experimenters. Both the operations personnel and the experimenters may request via the control panels the information they desire displayed on the oscilloscope screens. Both alphanumeric and point plot data may be requested to be displayed and combinations thereof. This will allow the demarcation of axes with the appropriate engineering units and the plotting of data as it is accumulated. With the computational power of the computer available, plots

of data may be extrapolated ahead of current data taking on the basis of theoretical models to predict the future course of the experiment. Figure 1 shows simulated data for a transient displayed on a computer-driven display. Figure 2 shows a photograph of a typical digital control panel.

A study has been completed which considered the feasibility of also placing a computer-driven display at Lemont, Illinois; this would provide the results of experiments very soon after the experimental results have become available at the ETR. Based on the results of this study it would appear that this would be entirely feasible and that personnel at Lemont would be able to receive essentially the same information on their monitor that would be output by the monitors at the ETR. The remote monitor at present is envisioned with a communication link by ordinary telephones. Because the system would be compact enough to be mobile, it could be wheeled to the location of any ordinary telephone at Lemont and a connection made to the ETR loop experiment by simply dialing the appropriate phone number. It is also possible to provide the remote monitor at Lemont with the computational capability to allow the investigators there to do some model computations as the experimental results come from the ETR.

Fig. 1 A photograph of a computer-driven oscilloscope display showing a simulated reactor transient.

Fig. 2 A typical digital control panel used in connection with the control and acquisition of experimental data.

NEW DEVELOPMENTS IN COMPUTER INTERFACES

G. B. Morgan, C. W. Richardson, W. W. Black

In the last year we have developed several new computer interfaces which are of general interest. A very brief description of each of these is given below.

General Control Panel Interface. The control panel concept was developed to help make the digital computer more convenient for the layman. Most operators or experimenters are familiar with the front panel of a particular piece of equipment (such as nuclear ADC's or reactor control equipment). These people should not be forced to learn a completely new procedure in order to use a piece of equipment which is supposed to be more advanced. The control panel interface enables the computer to service the panel in an efficient manner. Whenever a switch position is changed the panel interface causes a computer interrupt and the computer then reads the new panel status in one simple instruction. The panel status is read into the accumulator in seven bits. Three bits indicate the new switch position (0 through 7) and the

other four bits indicate the switch that was changed. The panel consists of 12 eight-position switches and three sets of eight push buttons. Figure 1 is a photograph of a typical panel. The software to service such as input device is much simpler than most input devices such as light pens, keyboards, etc.

A small program package has been developed for decoding information from the panel. When a switch or button is changed the computer can read two binary numbers. The two numbers specify a switch and a position or a button. The two numbers are generally used to either select a constant or select a set of instructions to be executed. Figure 2 is a diagram showing the programming logic which is used. In this example switch 1 is used to select a constant. A different constant is allowed for each position. Switch 2 is used to execute the group of instructions corresponding to the position of the switch.

The coding for Figure 2 has actually been written and implemented for both PDP-8 and -9 computers.

Memory-to-Memory Interface for Input/Output Processor. A memory-to-memory interface has been designed which will enable communication and data transfer between the PDP-9 and the General Automation SPC-12[1]. The SPC-12 will be used as an input/output processor to the PDP-9. It is expected that the SPC-12 used as an I/O processor will relieve the PDP-9 from many of the current device handler functions. Peripherals which will be attached to the SPC-12 include an IBM compatible magnetic tape transport, a data phone, an 1130 transmission link, and a card reader.

CAMAC Crate Interfacing System. Currently the Scientific Computer Systems Group is evaluating the feasibility of using the new CAMAC crate system for doing the majority of interfacing jobs on small computers. The CAMAC crate accepts plug-in modules and provides access to the data way. The data way is carried on a multilayer printed circuit board which is assembled at the rear of the crate. The CAMAC module is one-half the standard U.S.AEC NIM module width and plugs into a data way connector which is a double-sided 43-way connector. The CAMAC system provides a method for computer-independent interfacing on an interchangeable basis on all computer systems. A CAMAC crate system with a PDP-8 controller will be evaluated.

[1] R. A. Coates, G. O. English, E. E. Owens, "SPC-12-PDP-9 Bi-Directional Data Transfer Interface", this report.

Fig. 1 Photograph of a typical digital control panel.

Fig. 2 A flow diagram of the programming logic used to service a digital control panel. In the example switch #1 selects different constants. Switch #2 selects different blocks of instructions to be executed.

SOFTWARE DEVELOPMENTS FOR A 50/50 PULSE-HEIGHT ANALYZER SYSTEM

E. W. Killian, C. W. Richardson

A data acquisition and analysis package is being written for a field computer analyzer system which is a modified Nuclear Data 50/50 analyzer. The package will allow the user to monitor the data as it is being acquired. This will eliminate time losses due to counting samples too long and rejection of samples. The time saved in this case is very valuable since expensive drilling equipment must wait while the counting is done.

The primary purpose of the data analysis is to determine the ratio of the areas of two photopeaks in the gamma-ray spectra acquired. The operator indicates which two peaks are to be used at the beginning of a series of experiments.

The operator has rather flexible capabilities for displaying any desired data by the use of a function box and cathode-ray-tube display that have been added to the 50/50 system. Both the function box and the display have been added directly to the PDP-8I computer built into the 50/50 system.

FOURIER TRANSFORMS IN REAL TIME

R. C. Davies

A fast Fourier transform subroutine package which is FORTRAN compatible has been developed for a PDP-9 computer to transform any type of discrete data. A 128K RM09 serial drum is used to readily access and store the data during computation of the fast Fourier transform. A 339 display unit is used to display the original data and the transformed data separately or simultaneously for comparison. A paper tape punch option supplies the user with permanent copies of portions or of all the data.

The above method proved to be too slow for the real time applications for which it was intended. The time required by this method to transform 1024 points was 2-1/2 minutes. Although the drum furnished the capability of transforming up to 8192 points, the amount of drum transfers required plus the actual Fourier transform time made transforming anything above 1024 points unfeasible. Therefore, different approaches have been taken. A brief discussion of the methods studied follows.

In order to do a transform in the shortest possible time, integer arithmetic was used. Integer arithmetic was chosen because it is hardware supported on the PDP-9 where floating point arithmetic is only software supported. Also integers use only one memory word; floating point uses two memory words.

Some difficulties arose in this method, however. It is evident, in using integer arithmetic, that the sines and cosines which are needed must also be integer numbers. This means all sine and cosine values must be multiplied by a constant. Because of this distortion of the sines and cosines, the results

of the Fourier transform became multiplicatively greater than before the sines and cosines were changed. Unless some scaling method could be found this method was not considered satisfactory. It should also be noted that in many cases accuracy was lost by using integer arithmetic. This method was abandoned for the following program.

A program was written which uses the "chaining" capabilities of the PDP-9. Chaining allows FORTRAN programs to be logically divided into segments. These segments can then be called into the computer memory one at a time and executed. This allows very large programs to be run in a small computer. The Fourier transform routine has also been written in Macro-9 language to give greater speed. A Fourier transform can be performed on 1024 floating point numbers with all 1024 points in the memory of the computer in about 34 seconds.

This method has been recently modified. Since most of the time is spent in the floating point package, the floating point package supplied by Digital Equipment Corporation has been replaced with one written by W. R. Meyers^[1]. This modification decreased the time to transform 1024 points to 19 seconds. This method is fairly fast and has an accuracy of six decimal digits. The data can have a magnitude of $10^{\pm 75}$.

Table I gives a comparison of the various methods described above. Figures 1 and 2 show sample data and the corresponding Fourier transforms, respectively.

- [1] W. R. Myers, FP9 -- A Floating Point Arithmetic Package for a PDP-9 Computer, CI-1151 (July 1969).

TABLE I

A COMPARISON OF DIFFERENT METHODS THAT HAVE BEEN DEVELOPED TO
COMPUTE FOURIER TRANSFORMS USING THE FAST FOURIER TRANSFORM ALGORITHM

<u>Method</u>	<u>Execution Time (sec)</u>	<u>Accuracy Decimal Digits</u>	<u>Maximum Number of Points</u>
Using the RM09 Drum	150	6	at least 8196
Using Integer Arithmetic	not known	very poor	2048
Using chaining feature and DEC's floating point package	34	6	1024
Using chaining feature and W. R. Myers floating point package	19	6	1024

Fig. 1 A gamma-ray spectrum as it appears on the screen of the oscilloscope prior to the Fourier transformation.

Fig. 2 The Fourier transformation of the gamma-ray spectrum shown in Figure 1 as it appears on the oscilloscope screen.

COMPUTER AIDED LOGIC DESIGN

G. B. Morgan

Some progress has been made in the last year in developing a Computer Aided Design (CAD) programming system which may be used in the layout, drafting, and wiring phases of digital logic systems. This is a description of the capabilities incorporated in the present CAD programs and a proposal for future capabilities to be developed.

In the design and fabrication of modern digital systems (computer interfaces, etc) the designer often finds that the actual creative design phase is very short compared to all the bookkeeping, routing, and layout planning functions which must be done. Once this creative design phase is finished the problem of converting this sketch to a finished (fabricated) system looms ahead.

The task of logic drafting, layout, and wire list generation is one which many times occupies more of the designer's time than the actual creative design. The modern digital computer is best suited to carry out this task. For this reason the CAD program which is now running has the following capabilities:

- (1) The system accepts a coded description of the logic sketch.
- (2) It then generates a finished logic schematic with pin numbers, modules numbers, etc.
- (3) It generates a complete module list and parts list.
- (4) It generates a sorted wire list which can be used by an automatic wiring machine.

The areas of continued CAD system development are chiefly associated with making the total system user-oriented. The first and most vital area is the development of an interactive oscilloscope graphics package as input to the existing CAD package. This would allow the designer the convenience and freedom of using his own familiar language (logic symbols) rather than the machine's language (coding statements). Until this is done the system will not be used by the engineering people. The second area of development is in the output of the CAD package. The current CAD system plots the logic schematic on a Calcomp plotter. For storage and handling convenience as well as output speed it is proposed that the output be done on the microfilm plotter system.

DIGITAL NEUTRON NOISE ANALYSES

B. J. Kramer^[1], W. W. Black, A. L. Connelly

The results summarized here are primarily those resulting from a thesis project completed by B. J. Kramer and given in Reference 2. There were two primary goals set and attained in this study. The first was to develop the techniques that would allow power spectral densities from neutron noise studies to be computed in a digital fashion rather than the analog fashion that had been used in this laboratory in the past. It is necessary to develop this digital capability for the future when digital computers will be used to control reactors and to run incipient failure tests on reactors. The second

goal of the study is a natural result of the first, namely to perform experiments which will provide the specifications for a small on-line computer that can perform power spectral density computations and other similar computations in real time. Theoretical calculations show that a frequency span for thermal test reactors up to approximately 100 cycles per second for the power spectral density is sufficient. And taking into consideration the Nyquist folding frequency this means a sampling capability must be developed which will provide a frequency spectrum out to 200 cycles per second.

The first task to be accomplished was the development of computer programs which would permit Fourier transforms of the data, auto- and cross-correlation computational abilities, and averaging techniques for the power spectral densities. The fast Fourier transform algorithm was used to compute the Fourier transforms. The Fourier transform of the data points themselves plus one additional operation gives the power spectral density. However, there are many more points in the data array than one would normally desire in the power spectral density. Therefore, averaging techniques were developed to give power spectral densities with reasonable number of points and also to improve statistics. These programs were all written in FORTRAN and developed on the large computer at the Central Computing Facility. All of the programs were tested with known functions such as periodic functions and exponential decays.

Once the programs had been written and tested on functional forms, analog tapes were obtained which contained data from neutron noise measurements at the Advanced Test Reactor Critical Facility. These tapes had been analyzed in an analog fashion at a spectrum analyzer in the laboratory. These analog tapes were digitized on a computer system which was made available at the TAN area. The resulting digital magnetic tapes were then used as input for the FORTRAN program package. As expected, the digital analysis gave results identical with those obtained in an analog fashion. This is demonstrated in Figure 1. Figure 1 shows an overlapping plot of the same data which has been analyzed by both digital and analog methods. Those data points with error bars are the results of the analog analysis and those points with no error bars are the results of the digital analysis. It can be seen easily that comparable results have been obtained.

After the ability to analyze the data in a digital fashion had been demonstrated with the FORTRAN program package, a small on-line computer system was used to determine what specifications would be needed for a machine installed at a reactor facility. The computer system used consisted of a computer with a 8192-word 18-bit memory, a 128K word drum, and a display oscilloscope. The memory cycle time of the computer was one microsecond. Several different programming approaches were tried to obtain a program package which would run fast enough to handle the data in real time. The first approach was to write a program in FORTRAN again for the small computer. As expected, these required too much memory space and did not run with the desired speeds. The final package to be developed is written almost entirely in the computer's assembly language and is within a factor of two of the desired running time. We are confident that the desired brief running time requirement can be met. Therefore, it has been concluded that a small computer system with approximately a one microsecond cycle time will be able to calculate the power spectral densities in real time. It has also been determined that a minimum of 8192 words of magnetic core memory are required and a reasonable sized magnetic drum or disk must be used for auxiliary storage.

- [1] Now employed at Douglas United Nuclear, Hanford Project.
- [2] B. J. Kramer, "Computation of Power Spectral Density Curves from Reactor Noise Data On a Digital Computer", masters thesis, University of Idaho.

Fig. 1 A comparison of a power spectral density that has been determined by both analog and digital methods. The points with error bars are those obtained by analog methods, and the remaining points were obtained by digital methods.

PSEUDORANDOM NUMBER GENERATOR

A. L. Connelly

Pseudorandom sequences of the numbers zero and one have wide application as sources of random noise, useful in system analysis. In addition certain properties of their autocorrelation function permit effective increases in data acquisition rates. For example, pseudorandom sequences have been used in the design of correlation choppers used in time-of-flight spectrometers[1,2]. This usage has provided improved data where the signal-to-background ratio is low. Pseudorandom digital signals have been applied to reactors in order to measure their frequency response functions[3], and for obtaining Rossi-alpha and pulse-propagation measurements in subcritical assemblies[4]. Thus considering the wide use of such sequences, a general way of obtaining such sequences with equipment at hand was sought.

A subroutine is currently under development on the PDP-9 for the generation of pseudorandom sequences of length 2^m-1 , $7 \leq m \leq 18$. Each call of the subroutine will leave an element of the sequence in the accumulator of the PDP-9 to be used by the caller of the subroutine in any way he desires. Each successive call will produce a result dependent on the previous call. The user of the subroutine must supply m to the subroutine each time it is called. The subroutine may be called recursively in order to obtain a complete maximum length sequence. Typically such a sequence would be stored in the computer or punched on a paper tape for future use.

A lower limit of $m = 7$ was chosen because shorter low-pass filtered sequences do not possess a Gaussian distribution[5]. The upper limit of $m = 18$ was chosen because the accumulator of the PDP-9 is 18 bits long, and the subroutine relies heavily on shifting the accumulator. It would be possible to set an upper limit of 36 if the accumulator was used in conjunction with the multiplier quotient register. This is not being implemented at this time because a very different type of program would be required.

The subroutine currently under development is a generalization of a completed program for generating sequences 511 bits long ($m = 9$).

-
- [1] K. Sköld, "A Mechanical Correlation Chopper for Thermal Neutron Spectroscopy", Nucl. Instr. & Methods, 63 (1968) pp 114-116.
 - [2] K. Sköld, "On a Special Application of Correlation Methods in Neutron Spectroscopy", Nucl. Instr. & Methods, 63 (1968) pp 347-348.
 - [3] M. Hara and N. Suda, "Some Investigations into the Application of Pseudorandom Binary Signals to Reactor-Dynamics Measurements", Neutron Noise, Waves and Pulse Propagations, AEC Symposium, Series 9, pp 247-270 (May 1967).
 - [4] R. E. Uhrig and M. J. Ohanian, "Pseudorandom Pulsing of Subcritical Systems", Neutron Noise, Waves, and Pulse Propagation, AEC Symposium Series 9, pp 315-332 (May 1967).
 - [5] K. R. Godfrey, "Theory and Application of Pseudo-Random Sequences", Control, 10 pt 1 (1966) pp 305-308.

BETA-RAY SPECTROMETER CONTROLLER SOFTWARE

E. W. Killian, C. W. Richardson

The air-cove $\pi/2$ β -ray spectrometer became operational during the past year. The program presently being used within the computer operated controller of this instrument has provided the developers of the software system with a method to control and test the spectrometer. The present software package will provide the basis for an extended data acquisition and control program which will be developed after operating experience has been gained with the instrument. This basic package is general in nature and modular in design and will serve as the foundation for a more sophisticated program as more peripherals and consequently more tasks are required to be handled by the controller.

The peripherals presently interfaced to the controller are:

- (1) A ASR-33 teletype
- (2) A storage oscilloscope
- (3) A 200 amp power supply via a 17 bit digital-to-analog divider
- (4) A 24 binary bit, 1.5 MHz counter
- (5) A 12 bit 60 Hz clock.

The computer uses these peripherals to perform three main tasks:

- (1) Determines from the operator (via the teletype) what the input parameters are for a particular run on the spectrometer. At present the only controller-determined variables allowed in the operation of the spectrometer are variations in the current supplied to the magnetic coils. The program asks for a starting current, a final current, a current increment size, and the length of time for the coils to be energized at each current level. The computer determines from these parameters what the initial setting of the power supply register should be, sets the register, and increments it until the final current is reached. The 12 bit clock serves to determine how long the power is supplied between increments.
- (2) During each step (ie, while current is supplied to the coils) electron counts are detected and recorded in the 24 bit counter. At the end of each step the counter is read, converted to a decimal number, and printed on the teletype together with the power level of the spectrometer. The counter is then cleared, the current incremented, and a new step begun.
- (3) At the end of each run the computer also records on the storage scope the number of counts for that step. To accomplish some degree of sensitivity in the plotting of these numbers, the operator is questioned as to the range he expects the counts

to fall within during a run. The range determines the number of decades which will be plotted on the scope. This method of plotting allows the operator to obtain plots with varying degrees of sensitivity.

The program operates under the control of a monitor. All signals to the computer from the peripherals are in the form of interrupts. The monitor determines which device caused the interrupt and initiates action, as required, to service the interrupt. All commands to control the spectrometer are given to the controller via the teletype. The monitor decodes these commands and initiates the required action. The design of the monitor is such that it will allow the addition of several more commands very easily.

A summary of the commands which can be given to the controller are:

(1) SETUP

Upon receiving a SETUP command the program will ask for the eight input parameters. The storage scope is erased and the axes drawn.

(2) RESET

The RESET command uses the input numbers from the last SETUP to re-initialize the system for a new run.

(3) GO

After all input parameters have been entered, the GO command is used to start the series of runs.

(4) HALT

The HALT command stops the spectrometer interface.

(5) CONTINUE

By setting the left most switch (bit zero) on the switch register of the computer, the operator signals the controller to stop the interface at the end of each time interval. To continue to the next step the operator types the CONTINUE command. Similarly, if the operator sets the second left most switch, the computer will stop the interface at the end of each run. Again, the CONTINUE command is given to proceed to the next run.

(6) REPEAT

As an alternative to the CONTINUE command the REPEAT command is used at the end of each step or run to repeat the last step or run.

The controller requires that eight input parameters be supplied to initiate a series of runs. The eight input parameters are:

(1) Range

The number supplied here must correspond to the manual switch on the power supply. It is used by the program to scale all current level parameters.

(2) Number of Runs

The integer supplied for this parameter determines the number of runs the operators wish to run consecutively without further intervention. This option is supplied in order to eliminate the necessity for supplying the remaining input parameters at the beginning of each run. The value for this parameter must be less than six.

(3) Initial Current

The initial current is used as the initial current setting for the start of the run.

(4) Delta Current

Delta current is used to increment the initial current to subsequent power settings.

(5) Final Current

The initial current will be incremented by delta current until the accumulated current is greater than or equal to this parameter.

(6) Delta Time

The integer given here determines how long the time interval will be between the initiation of a new power level and the conclusion of that step. Time settings are multiples of one minute.

(7) Maximum number of counts (MAXC)

(8) Minimum number of counts (MINC)

The values for these two parameters are used to scale the storage scope. The controller program divides the scope into decades as determined by the difference between MAXC and MINC.

Figure 1 is a sample of the teletype printout which is produced when a run is initiated. All underlined text was typed by the operator; all other was supplied by the controller.

```

*SETUP
*NUMBER OF RUNS = 3
*RANGE = 50
*MIN. NUMBER OF COUNTS = 2.3
*MAX. NUMBER OF COUNTS = 3.2
*
*RUN # 1
*INITIAL I = 33.90
*FINAL I = 33.94
*DELTA I = .01
*DELTA T = 4
*
*RUN # 2
*INITIAL I = 33.95
*FINAL I = 34.03
*DELTA I = .001526
*DELTA T = 4
*
*RUN # 3
*INITIAL I = 34.04
*FINAL I = 34.08
*DELTA I = .01
*DELTA T = 4
*
*GO
*
*PERIOD   COUNTS   CURRENT   TIME   COUNTS/DELTA TIME
*  1      175 +    33.8999    4      +  43.7500
*  2      170 +    33.9098    8      +  42.5000
*  3      182 +    33.9197   12      +  45.5000
*  4      207 +    33.9297   16      +  51.7500
*  5      167 +    33.9396   20      +  41.7500
*
*END OF RUN # 1
*
*TYPE R OR C CONTINUE
*
*PERIOD   COUNTS   CURRENT   TIME   COUNTS/DELTA TIME
*  1      234 +    33.9499    4      +  58.5000
*  2      180 +    33.9514    8      +  44.9999
*  3      176 +    33.9529   12      +  43.9999
*  4      210 +    33.9545   16      +  52.4999
*  5      222 +    33.9560   20      +  55.5000
*
*
*
*  50     143 +    34.0246   200     +  35.7500
*  51     167 +    34.0262   204     +  41.7500
*  52     167 +    34.0277   208     +  41.7500
*  53     167 +    34.0292   212     +  41.7500
*
*END OF RUN # 2
*
*PERIOD   COUNTS   CURRENT   TIME   COUNTS/DELTA TIME
*  1      156 +    34.0399    4      +  38.9999
*  2      179 +    34.0498    8      +  44.7500
*  3      159 +    34.0597   12      +  39.7499
*  4      173 +    34.0697   16      +  43.2500
*  5      184 +    34.0796   20      +  46.0000
*
*END OF RUN # 3
*
*ALL RUNS COMPETED
*

```

Fig. 1 A sample printout from a series of runs on the beta-ray spectrometer. In the initial dialogue the number of runs and the parameters for each run have been established.

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G. COOPERATIVE PROGRAMS WITH EDUCATIONAL INSTITUTIONS

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COOPERATIVE PROGRAMS WITH EDUCATIONAL INSTITUTIONS

As the major operating contractor at the NRTS, Idaho Nuclear Corporation continues to stress the importance that the U. S. Atomic Energy Commission places on the stimulation and support of education. President Nixon's statement[a] clearly indicates the importance that the administration places on this area. Thus, the program of the Nuclear Technology Branch continues to emphasize activities to strengthen ties between the research programs at the National Reactor Testing Station and those presently being conducted by regional universities. This effort, which has met with an increasingly enthusiastic response over the past several years, is implemented in a variety of ways. Examples of activities in this area during the period covered in this report are briefly described below.

INC Educational Fund

The Idaho Nuclear Corporation contributes about \$100,000/year to educational activities with a substantial portion of this contribution related to programs within the Nuclear Technology Branch. Such connected funding during FY-1969 for support of university faculty members was:

<u>Faculty Member</u>	<u>University</u>	<u>Project Area</u>
T. J. Connolly	Stanford	Radiation Induced Nucleation of Bubbles
E. Fast	Idaho Nuclear Corporation	Teacher at Idaho State University
D. M. Grant	University of Utah	Neutron Molecular Spectroscopy
R. J. Pugmire	University of Utah	Neutron Molecular Spectroscopy
V. Ryan	University of Wyoming	Activation Analysis
T. E. Young	Idaho Nuclear Corporation	Teacher at Idaho State University

[a] "The equipment of many federal laboratories is superb and often unique. This investment should be viewed as a national resource and not one for the exclusive use of the laboratory staff members. While many scientists and engineers from outside now frequently use federal research facilities, an even closer and more extensive cooperative relation will be productive. I am therefore approving a policy designed to bring this about. Dr. DuBridge will be able to implement the policy with the help of the Federal Council for Science and Technology, which has recommended its adoption."

In addition to the above mentioned faculty participation, other activities involving the Nuclear Technology Branch which received support from the Idaho Nuclear Corporation Educational Funds are as follows:

- (1) Learning Systems Inc. was commissioned to produce a 35-minute video tape (or film) on the uses of neutrons in research. This tape was then presented to universities and research foundations throughout the country. The emphasis of this presentation was on the use of high flux reactor neutron sources such as the MTR, in research disciplines such as biology, agriculture, geology, etc.
- (2) A graduate student, R. Martin, of the University of Chicago was provided a dislocation allowance so that he could work with Dr. H. L. McMurry of the Nuclear Technology Branch.
- (3) R. J. Pugmire of the University of Utah was provided with travel expenses so that he could present a seminar on neutron molecular spectroscopy at Montana State University.
- (4) Development of a technical display for the Idaho State Fair was provided to present a demonstration of advanced computer technology. This was partially planned and manned by members of the Nuclear Technology Branch.

Association of Western Universities Program

Under the auspices of the Association of Western Universities (AWU), scientists are able to visit and use the facilities of the National Reactor Testing Station to conduct research projects, participate in existing programs, or carry out independent research leading to advanced degrees. During the 1969 fiscal year, 20 faculty members from 9 different colleges and universities worked in the Nuclear Technology Branch under the AWU program. These activities are listed below.

<u>Faculty Member</u>	<u>University</u>	<u>Project Area</u>
J. J. H. Berlijn	Valdosta State College	Radioactive Decay
M. K. Corbett	Idaho State University	Trace Elements in Geological Formations
G. P. DeVault	Montana Technical	Shock Effects and Phonon Viscosity
L. H. Johnston	University of Idaho	Radioactive Decay
R. J. Kearney	University of Idaho	Solid State Physics
J. K. Kliwer	University of Nevada	Radioactive Decay
R. E. McAdams	Utah State University	Radioactive Decay

<u>Faculty Member</u>	<u>University</u>	<u>Project Area</u>
B. H. Muehler	University of Wyoming	Dynamics of Liquids
O. H. Otteson	Utah State University	Radioactive Decay
R. A. Porter	University of Idaho	Radioactive Decay
J. E. Price	Idaho State University	Radioactive Decay
R. J. Pugmire	University of Utah	Neutron Molecular Spectroscopy
R. Ragsdale	Utah State University	Radioactive Decay
V. A. Ryan	University of Wyoming	Activation Analysis
G. M. Sandquist	Utah State University	Pollution Detection by Activation
D. A. Tripp	Weber State College	Radioactive Decay
S. H. Vegors	Idaho State University	Radioactive Decay
H. Willmes	University of Idaho	Radioactive Decay
A. E. Wilson	Idaho State University	Nuclear Engineering
J. K. Woods	Utah State University	β -ray Spectrometry

During this period 13 graduate students also carried out research projects under the joint direction of their on-campus major professor and under a Nuclear Technology Branch staff member. They were:

<u>Student</u>	<u>University</u>	<u>Idaho Nuclear Staff Director</u>	<u>Project Area</u>
W. R. Bohl	University of Wisconsin	O. D. Simpson	Neutron Adsorption Cross Sections
T. A. Dardis	University of Nevada	R. J. Gehrke	Radioactive Decay
David Fleischer	University of Wyoming	R. N. Chanda	Gamma Detection
B. J. Kramer	University of Idaho	W. W. Black	Reactor Noise
Larry Kroger	University of Wyoming	R. R. Spencer	Fission Process

<u>Student</u>	<u>University</u>	<u>Idaho Nuclear Staff Director</u>	<u>Project Area</u>
R. C. Livingston	University of Utah	R. J. Pugmire	Neutron Molecular Spectroscopy
R. A. Lokken	Utah State University	R. G. Helmer	Nuclear Decay Schemes
E. L. Mark	University of Idaho	R. P. Schuman	Surface Absorption
D. J. McMillan	Vanderbilt	C. W. Reich	Nuclear Level Structure
T. D. Rupp	Idaho State University	C. W. Reich	Radioactive Decay
M. R. Serpa	Idaho State University	T. E. Young	Cross Sections
R. W. Tarr	University of Idaho	C. W. Reich	Nuclear Structure
A. G. Taylor	Utah State University	R. L. Heath	Low Energy Nuclear Spectroscopy

Summer Employees

In addition to the AWU-supported program, faculty members and graduate students are brought into the company as regular employees for short periods during academic vacations. This allows the university people to become acquainted with facilities and programs at the NRTS Site. During FY-1969 those employed under this program included two graduate students, James K. Boyter of the Idaho State University and Kelby J. Anderson of the University of Colorado, Thayne M. Thompson, a student in Physics at the Illinois Institute of Technology and Glenn F. Goff, an instructor in electronics at Idaho State University.

AEC Post-Doctoral Fellowships

David McMillan of Vanderbilt University won an AEC post-doctoral fellowship and chose the MTR as his location. He is assisting in nuclear structure studies.

Faculty and Staff Exchange Program

Idaho Nuclear Corporation has established a program by which staff members of the company can be assigned to teach at a university. There are basically two kinds of arrangements that can be made:

- (1) For a period of a semester or a year, a staff member and a faculty member can exchange positions.

- (2) Where the school is within commuting distance of the NRTS, a staff member may be assigned to teach on a half-time basis, ie, either two or three days a week, while continuing to work the balance of his time on his regular job.

Under the first program, no exchanges were arranged this year.

In the second program, K. T. Faler was assigned to teach half time in the Chemistry Department of Idaho State University while E. Fast and T. E. Young were assigned to the Physics Department at Idaho State University.

Adjoint Professorships

In several of the situations where a graduate student at a regional university has completed his course work toward an advanced degree and plans to carry out thesis research at the NRTS, arrangements are made whereby the staff member of the Nuclear Technology Branch responsible for supervising the on-Site research is appointed to the university staff as an Adjoint Professor. This arrangement has in the past proved to be a highly beneficial one, greatly improving the communication between the university and the NRTS. During this fiscal year, four members of the Branch have served in this capacity with Utah State University. They are J. E. Cline, R. L. Heath, R. G. Helmer, and C. W. Reich. In addition because of his association, R. J. Pugmire at the NRTS, of the University of Utah staff was appointed an adjoint professor in the Chemistry Department at Idaho State University.

NRTS Educational Program

Idaho Nuclear Corporation, in cooperation with the universities of Idaho, administers a continuing educational program at the NRTS. This program leads to the degree of Master of Science in various technical fields. The following members of the Nuclear Technology Branch taught courses.

Faculty

R. P. Schuman

J. J. Scoville

O. D. Simpson

T. G. Worlton

The following members of the Branch earned their master's degree through this program:

Student

Gordon E. Stokes

Barry W. Howes

Thesis Title

"The Total Neutron Cross Section of Thulium-170 in the Resonance Region"

"The Materials Testing Reactor 24 keV Neutron Beam"

University Seminars and Colloquium Presented

The staff of Nuclear Technology Branch is frequently called upon to provide seminars and colloquia at universities. In FY-1969 these were:

<u>Staff</u>	<u>University</u>	<u>Topic</u>
R. B. Bennion	Montana State University	High Pressure Neutron Diffraction
W. W. Black	Texas Technological College	Application of On-line Computers to Nuclear Spectroscopy
R. M. Brugger	Idaho Falls Rotary	Need for Intense Neutron Sources
R. M. Brugger	Eastern Idaho ANS	Beam Research at MTR
R. M. Brugger	Idaho State University Student Chapter of ANS	Beam Research at MTR
R. M. Brugger	University of Michigan	
E. Fast	Denver University New Mexico State University Brigham Young University University of Idaho Montana State University	Physics Measurements with a Low Power Nuclear Reactor
R. L. Heath	Oak Ridge National Laboratories	Series of lectures on nuclear spectroscopy
B. W. Howes	University of Idaho	The MTR 25-keV Neutron Beam
H. L. McMurry	University of Maryland National Bureau of Standards	The Use of Valence Force Potentials for Calculating Crystal Vibrations
P. D. Randolph	University of Wyoming	Neutron Inelastic Scattering from Liquids
C. W. Reich	University of Idaho	Nuclear Level Structure
C. W. Reich	University of Nevada	Nuclear Level Structure
R. E. Schmunk	Marquette University	High Pressure Neutron Diffraction
R. E. Schmunk	Idaho State University	Four lectures on neutron diffraction
R. P. Schuman	Idaho State University	Chemistry of Actinides

H. PUBLICATIONS

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NUCLEAR TECHNOLOGY BRANCH
PAPERS PUBLISHED IN OPEN LITERATURE

AUTHOR(S)	TITLE	PUBLICATION
O. D. Simpson L. G. Miller	A Technique to Measure Neutron Cross Sections in the Low keV Energy Region	<u>Nuclear Instruments and Methods</u> <u>61</u> , 245-250 (1968).
G. E. Stokes R. P. Schuman O. D. Simpson	The Total Neutron Cross Section of 115-Day Tantalum-182 from 0.01 to 1000 eV	<u>Nuclear Science and Engineering</u> <u>33</u> , 16-23 (July 1968)
T. G. Worlton D. L. Decker	Neutron Diffraction Study of the Magnetic Structure of Hematite to 41 kbar	<u>The Physical Review</u> <u>171</u> , 596-599 (July 10, 1968)
R. G. Helmer C. W. Reich	Decay of an Isomeric State in ^{178}Hf with $K \geq 16$	<u>Nuclear Physics</u> <u>A114</u> , 649-662 (1968)
W. R. Myers P. D. Randolph	Low Frequency Vibrations in Polyethylene at 300°, 77°, and 4.2°K	<u>Journal of Chemical Physics</u> <u>49</u> , 1043-1054 (August 1, 1968)
R. C. Greenwood	Observation of a 9.15 MeV Doublet Level in ^{15}N from a Study of the $^{14}\text{N}(n,\gamma)^{15}\text{N}$ Reaction	<u>Physics Letters</u> <u>27B</u> , 274-276 (August 5, 1968)
L. D. McIsaac	Levels in ^{133}I from the Decay of $^{133\text{m}}\text{Te}$ and $^{133\text{g}}\text{Te}$	<u>The Physical Review</u> <u>172</u> , 1253-1261 (August 20, 1968)
R. M. Brugger	Slow Neutron Scattering from Water	<u>Nuclear Science and Engineering</u> <u>33</u> , 187-194 (August 1968)
J. W. Rogers J. J. Scoville	Infinite Dilution Resonance Absorption Integrals of Hafnium	<u>Nuclear Science and Engineering</u> <u>33</u> , 350-352 (September 1968)
K. T. Faler R. R. Spencer R. A. Harlan	A $K^\pi = 0^+$ Vibrational Band in ^{184}W	<u>The Physical Review</u> <u>175</u> , 1495-1497 (November 20, 1968)

NUCLEAR TECHNOLOGY BRANCH
PAPERS PUBLISHED IN OPEN LITERATURE

AUTHOR(S)	TITLE	PUBLICATION
R. M. Brugger	We Need More Intense Thermal-Neutron Beams	<u>Physics Today</u> <u>21</u> , 23-30, (December 1968)
R. G. Helmer A. Backlin	Numerical Construction of Level Schemes - ^{149}Pm	<u>Nuclear Instruments and Methods</u> <u>65</u> , 31-44 (1968)
R. G. Fluharty F. B. Simpson G. J. Russell J. H. Menzel	Moderator Studies for a Repetitively Pulsed Test Facility (RPTF)	<u>Nuclear Science and Engineering</u> <u>35</u> , 45-69 (January 1969)
R. M. Brugger K. A. Strong R. J. Pugmire D. M. Grant	Observation of the Least-Energetic E_u Vibration of Cyclohexane	<u>The Journal of Chemical Physics</u> <u>50</u> , 1029-1031 (January 15, 1969)
R. J. Gehrke R. G. Helmer	Decay of ^{102}Rh and ^{102m}Rh	<u>The Physical Review</u> <u>177</u> , 1792-1797 (January 20, 1969)
K. T. Faler R. R. Spencer R. A. Harlan	Excited States in ^{178}Hf from Resonance Neutron-Capture Gamma-Ray Studies	<u>Nuclear Physics</u> <u>A123</u> , 616-626 (January 27, 1969)
L. G. Miller H. G. Miller A. N. Ellis	Hot Sample Changer and Transfer Cask for the Nuclear Explosion Neutron Source Experiment	<u>Nuclear Instruments and Methods</u> <u>67</u> , 121-124 (1969)
R. G. Nisle	The Decomposition of Empirical Functions as Applied to Recoil-Proton Spectrometer Data	<u>Nuclear Instruments and Methods</u> <u>70</u> , 169-172 (1969)

NUCLEAR TECHNOLOGY BRANCH
PAPERS PUBLISHED IN OPEN LITERATURE

AUTHOR(S)	TITLE	PUBLICATION
J. E. Cline	Energy Dependence of Germanium (Lithium) Detector Efficiency	<u>Semi-Conductor Nuclear-Particle Detectors and Circuits</u> , National Academy of Sciences, Publication 1593, Washington, D. C. (1969) pp. 241-247.
R. L. Heath	Energy Dependence of Gamma-Ray Line-Widths in Germanium Detectors	Ibid, pp. 247-252
R. L. Heath	Detector Efficiencies: A Clarification of Terms	Ibid, pp. 252-258
R. L. Heath	Problems in the Calibration and Use of High-Resolution Germanium (Lithium) Spectrometer Systems	Ibid, pp. 642-660
H. F. Bezdek R. E. Schmunk L. X. Finegold	An Oven for a Neutron Spectrometer to 1000°K	The Review of Scientific Instruments, in press
R. E. Schmunk D. R. Winder	Lattice Dynamics of Sodium Chloride at Room Temperature	J. Phys. Chem. Solids, in press

NUCLEAR TECHNOLOGY BRANCH
PRESENTATION OF TECHNICAL PAPERS

AUTHOR(S)	SUBJECT	GROUP	PLACE, DATE
C. W. Reich M. E. Bunker	Systematic Features of Non-Rotational States in the Odd-A Deformed Nuclei	International Symposium on Nuclear Structure	Dubna, USSR July 1968
R. L. Heath	Application of High-Resolution Gamma-Ray Spectrometry to Non-destructive Testing	Symposium on Physics and Nondestructive Testing	Argonne National Lab. Chicago, Illinois Sept. 24-26, 1968
R. L. Heath	Gamma-ray Spectrometry and Automated Data Systems for Activation Analysis	IAEA-NBS International Symposium on Activation Analysis	Greenbelt, Maryland October 7, 1968
J. J. H. Berlijn R. C. Greenwood C. W. Reich	Levels in ^{174}Yb from the $^{173}\text{Yb}(n,\gamma)$ ^{174}Yb Reaction	American Physical Society Fall Meeting	Miami Beach, Florida November 1968
R. C. Greenwood C. W. Reich	Levels in ^{158}Gd from the $^{157}\text{Gd}(n,\gamma)$ ^{158}Gd Reaction	American Physical Society Fall Meeting	Miami Beach, Florida November 1968
C. W. Reich J. E. Cline	Experimental Investigation of the Gamma-Vibrational Band of ^{166}Er	American Physical Society Fall Meeting	Miami Beach, Florida November 1968
G. B. Morgan	Recent Experiences in Designing Module Interfaces Using M-series Logic	Digital Equipment Computer User's Society Fall Symposium	San Francisco, Calif. December 1968
G. E. Stokes D. R. Staples	A Flexible Data Acquisition and Control System Utilizing a PDP 8/S	Digital Equipment Computer User's Society Fall Symposium	San Francisco, Calif. December 1968
R. M. Brugger	Neutron Diffraction Using Pulsed Sources	American Nuclear Society Winter Meeting	Albuquerque, New Mexico January 28-30, 1969

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AUTHOR(S)	SUBJECT	GROUP	PLACE, DATE
R. L. Heath	The Techniques of High-Resolution Gamma-Ray Spectrometry	Symposium on Applications of Nuclear Engineering in Biology and Medicine	Argonne National Lab. Chicago, Illinois February 4, 1969
R. L. Heath L. O. Johnson	System Requirements for High-Resolution Gamma-ray Spectrometry at High Counting Rates	IEEE-Euratom Symposium on Nuclear Electronics	Ispra, Italy May 6-9, 1969
R. L. Heath W. W. Black	Development of a Laboratory Computer System Concept for Physics Research	Conference on Computer Systems in Experimental Nuclear Physics	Skytop, Pennsylvania March 1969
W. W. Black	Utility of Correlation Methods for Interactive Data Reduction and Acquisition	Conference on Computer Systems in Experimental Nuclear Physics	Skytop, Pennsylvania March 1969
O. D. Simpson N. H. Marshall	Automatic Analysis of Neutron Cross Section Data Using Interactive Graphics as an Aid	Conference on Computer Systems in Experimental Nuclear Physics	Skytop, Pennsylvania March 1969
R. M. Brugger R. B. Bennion T. G. Worlton W. R. Myers	Neutron Diffraction at High Pressures	American Crystallographic Association Meeting	Seattle, Washington March 1969
R. C. Greenwood C. W. Reich	Levels in ^{156}Gd from the $^{155}\text{Gd}(n,\gamma)$ ^{156}Gd Reaction	American Physical Society Spring Meeting	Washington, D. C. April 1969
R. P. Schuman	Resonance Integral Measurements	American Physical Society Spring Meeting	Washington, D. C. April 1969
R. G. Nisle	Analysis Methods for Recoil-Proton Spectrometer Data	Idaho Academy of Sciences	Nampa, Idaho April 25-26, 1969

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<u>AUTHOR(S)</u>	<u>SUBJECT</u>	<u>GROUP</u>	<u>PLACE, DATE</u>
J. W. Rogers	Proton Recoil Spectrometry	Idaho Academy of Sciences	Nampa, Idaho April 25-26, 1969
R. G. Nisle	The Beta-Decay Half-Life of Pu ²⁴¹ Recoil-Proton Spectrometry	Idaho Academy of Sciences	Nampa, Idaho April 25-26, 1969
A. L. Erikson R. P. Schuman R. J. Gehrke	The Use of Germanium Gamma Spectroscopy in Analyzing Reactor Coolant Water	American Chemical Society Northwest Regional Meeting	Salt Lake City, Utah June 1969
E. L. Mark R. A. Porter R. P. Schuman	The Direct Counting of ²³³ U Ions Adsorbed on Glass Scintillators	American Chemical Society Northwest Regional Meeting	Salt Lake City, Utah June 1969
R. M. Brugger	The WBRR's Role in Nuclear Education	American Nuclear Society Annual Meeting	Seattle, Washington June 1969
G. J. Russell R. M. Brugger	Very Intense Neutron Source (VINS)	American Nuclear Society Annual Meeting	Seattle, Washington June 1969
J. R. Berreth F. B. Simpson B. C. Rusche R. A. Gregg	Total Neutron Cross-Section Measurements of ²⁴³ , ²⁴⁵ , ²⁴⁶ Cm	American Nuclear Society Annual Meeting	Seattle, Washington June 1969
O. D. Simpson N. H. Marshall	An Automatic Fitting Technique for Obtaining Neutron Cross Section Resonance Parameters	American Nuclear Society Annual Meeting	Seattle, Washington June 1969
B. W. Howes R. M. Brugger J. W. Rogers R. P. Schuman O. D. Simpson	The Materials Testing Reactor 25 ± 1 keV Neutron Beam	American Nuclear Society Annual Meeting	Seattle, Washington June 1969

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AUTHOR(S)	SUBJECT	GROUP	PLACE, DATE
J. J. Scoville E. Fast D. A. Millsap R. G. Nisle J. W. Rogers	A Coupled Fast Reactivity Measure- ment Facility	American Nuclear Society Annual Meeting (Informal Session)	Seattle, Washington June 1969
R. C. Greenwood R. A. Harlan R. G. Helmer C. W. Reich	Neutron Capture Gamma-Ray Measure- ments with a Reactor-Produced 2-keV Neutron Beam	International Symposium on Neutron Capture Gamma- ray Spectroscopy	Studsvik, Sweden August 11-15, 1969
R. G. Helmer R. C. Greenwood C. W. Reich	Levels in ^{182}Ta	International Symposium on Neutron Capture Gamma- ray Spectroscopy	Studsvik, Sweden August 11-15, 1969
R. B. Bennion H. G. Miller W. R. Myers	100 kbar Press for Time-of-Flight Neutron Diffraction	VIII International Congress of Crystallography	Stony Brook, New York August 13-23, 1969
W. R. Myers R. B. Bennion R. M. Brugger	Pressure Dependence of the Debye Temperature of NaCl to 30 kbar	International Congress of Crystallography	Stony Brook, New York August 13-23, 1969

REPORTS ISSUED DURING FY 1969

- IN-1067 Analysis of ARMF Measurements by One-Dimensional Multigroup Diffusion Calculations, by D. A. Millsap, July 1968
- IN-1199 Scattering Law for Liquid Sodium at Small Momentum Transfers, by P. D. Randolph, July 1968
- IN-1214 Introductory Computer Programs for Neutron Diffraction, by J. K. Boyter and R. M. Brugger, August 1968
- IN-1226 DIPOLE SUM -- A Computer Program for Calculating Dipole Sums for an Arbitrary Crystal, by J. K. Boyter and T. G. Worlton, November 1968
- IN-1222 Low Energy Neutron Spectroscopy, Measurements on Fissile Nuclides, by M. S. Moore, January 1969
- IN-1280 MACS C -- A Computing Program for Crystal Vibrations Which Includes Long-Range Coulomb Interactions, by J. K. Boyter, H. L. McMurry and A. W. Solbrig, Jr., and Richard M. Martin (University of Chicago)
- IN-1282 MVFC -- A Computer Program for Constructing Force Constant Matrices for Use in Valence Force Potential Functions in Molecules and Crystals, by J. K. Boyter and H. L. McMurry, April 1969
- IN-1182 An Evaluation and Compilation of Neptunium-237 Cross Section Data for the ENDF/B File, by J. R. Smith and R. A. Grimesey, May 1969
- IN-1296 Resonance Integral Measurements, by R. P. Schuman and J. R. Berreth, May 1969
- IN-1130 Annual Progress Report Gamma-Ray Spectrometry of Neutron-Deficient Isotopes, by J. E. Cline and R. L. Heath, May 1969
- IN-1287 DATMON -- A PDP 9 Computer Program for the Simultaneous Acquisition of Data from One Neutron Time-of-flight Spectrometer and One Neutron Triple-Axis Crystal Spectrometer -- A User's Manual -- by W. R. Myers, July 1969
- IN-1294 Time-of-Flight Interface, by G. B. Morgan, June 1969
- IN-1308 The MTR 25-keV Neutron Beam by B. W. Howes, R. M. Brugger, J. W. Rogers, R. P. Schuman, F. B. Simpson and O. D. Simpson, July 1969

"Status of Nuclear Chemistry and Radiochemistry", National Academy of Sciences, Prepared in Cooperation with the Subcommittee on Radiochemistry, one of the members being R. P. Schuman, Washington, D. C. (1969).

I. RELATED ACTIVITIES OF BRANCH PERSONNEL

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RELATED ACTIVITIES OF BRANCH PERSONNEL

Besides their direct research accomplishments and their activities in support of education, Branch personnel are often called upon to assume other professional responsibilities and duties. During the past year the following staff members have been so engaged:

J. R. Berreth: Member, Research Materials Committee, AEC.

W. W. Black: Member, Chapter Executive Council, Association for Computing Machinery.

R. M. Brugger: Program Committee of Reactor Physics Division of American Nuclear Society, Nuclear Cross Sections Advisory Committee, Subcommittee on Evaluation of Thermal Cross Sections of NCSAC.

J. E. Cline: Reviewer for Phys. Rev. and J. Nucl. Application. Alternate, Nuclear Detector Subcommittee, National Academy of Science.

K. T. Faler: Reviewer for Phys. Rev., Member, Reactor Safety Committee for Idaho State University.

E. Fast: Reviewer for Nuclear Applications, Chairman, Reactor Safety Committee for Idaho State University.

R. L. Heath: Member, Board of Editors, Nuclear Instruments and Methods; Contributing Editor, Handbook of Chemistry and Physics; Member, National Technical Program Committee, American Nuclear Society; Member, Executive Committee, I and R Division, American Nuclear Society; Reviewer for the Physical Review, Analytical Chemistry, Nuclear Applications and Nuclear Science and Engineering; Chairman, Nuclear Detector Subcommittee, National Academy of Sciences.

R. G. Helmer: Reviewer for the Physical Review.

G. W. Griffing: Reviewer for Nuclear Science and Engineering.

C. H. Hogg: ASTM Committee, E-10, Subcommittee on Neutron Dosimetry; Irradiation Standards and Procedures Committee of the AEC Irradiation Effects on Reactor Structure Materials Program.

H. G. McMurry: Reviewer for Nuclear Science and Engineering; Mathematics Committee, School District #91.

F. A. Meichle: Chairman of Educational Committee of Idaho Falls Section of Instrument Society of America.

G. B. Morgan: Teacher, Introduction to Computer Systems, Glenn E. Clark Business College, Idaho Falls.

D. A. Pearson: Alternate, ASTM Committee, E-10, Subcommittee on Neutron Dosimetry.

R. P. Schuman: Subcommittee on Radiochemistry of National Science Foundation-National Research Council, Chairman of Idaho Section of American Chemical Society.

J. J. Scoville: Reviewer for Nuclear Science and Engineering; Reviewer for ANS-AEC monograph series.

F. B. Simpson: Exofficio member of Transplutonium Committee, AEC.

D. R. Staples: Secretary for Idaho Falls Section of Instrument Society of America.

G. E. Stokes: Chairman, Eastern Idaho Chapter of the Association for Computing Machinery. Chairman, Food Irradiation Committee of Eastern Idaho Nuclear Industrial Council; Chairman, Nuclear Committee of Blackfoot Chamber of Commerce.

K. A. Strong: Reviewer for J. Chem. Phys.

E. H. Turk: Representative of Idaho Section of American Chemical Society to Idaho Falls Technical Council.

D. E. Jackson: Member, Board of Directors, Idaho Falls Section of the Instrument Society of America.

R. E. Schmunk: Reviewer for J. Phys. Chem. Solids.

J. R. Smith: Member, Cross Sections Evaluation Working Group.

Support of Secondary Schools

The staff of the Nuclear Technology Branch has been engaged in a number of cooperative programs with the local school districts. During the reporting period members of the staff were instrumental in encouraging and assisting in the acquisition of a small computer for Idaho School District #91. They are assisting the school in implementing the machine into programs for computer assisted instruction (CAI) and computer techniques in problem solving.