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QUARTERLY REPORT

January 1 - March 31, 1962

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MTR ETR TECHNICAL BRANCHES QUARTERLY REPORT
1962 QUARTER 1
JANUARY 1 - MARCH 31, 1962

SUMMARY

ETRC measurements were made to confirm design parameters proposed for a new MTR shim rod. Increases of 13% and 26% were recorded for the thermal neutron flux peak, occurring in the water region between the fuel plates and the poison section of a partially withdrawn control rod, when the water gap was changed from 3/8 in. to heights of 1 in. and 1-3/4 in., respectively. It also was determined that boron-stainless steel flux-suppressor plates in ETR control rods were relatively more effective than nickel ones in reducing the pronounced thermal flux peaking. The maximum peak was reduced 29% by using boron-stainless steel plates as compared with a 17% reduction obtained with nickel suppressors.

Preliminary values have been obtained for the resonance absorption integral of natural dysprosium, natural tungsten, tungsten-184, and tungsten-186.

Measurements of activities of lutetium-175 and lutetium-176 in a natural lutetium-copper system were compared with those of a gold-copper system for neutron flux spectrum determinations. The results showed that natural lutetium is potentially useful for this purpose, but that presently available values of some of its nuclear constants are not sufficiently accurate.

A program (FLUSY) has been written for the IBM 650 to compute the necessary flux synthesis values of group-dependent buckling and flux-weighted reactor constants.

A sample design was finished for use in the long-term burnout study of the thorium-uranium-233 conversion chain. The design consists of a 1-in.-diameter x 4-in.-long 75-gram active core containing a maximum of 2.5 wt% of U-233, with the remainder Th-232, and clad with either zirconium or zircaloy-2.

Tensile testing was completed on 94 irradiated fuel-plate specimens with fuel cores of uranium-aluminum alloys, UO₂-aluminum cermet and U₃O₈-aluminum cermet clad with aluminum alloys. The most significant effect, resulting from fission damage, was the loss of ductility.

The fission cross section of plutonium-241 was measured from 0.025 to 100 ev. A value of 965 ± 40 barns was obtained for the 0.025 ev fission cross section based upon a normalization to the total cross section at the 6 ev level. A beryllium sample with a fast neutron dosage of 0.891×10^{21} n/cm² and a thermal neutron dosage of 1.001×10^{22} n/cm² was completely melted in the vacuum line apparatus for gas analysis. The total gas content determined by calculation agreed quite well with the gaseous products collected and analyzed. A series of swelling vs temperature experiments has commenced on beryllium samples with irradiation histories similar to the beryllium sample just described. In addition to swelling it has been determined that

evolution of reaction product gases at elevated but sub-melting temperatures also constitutes a potential hazard associated with beryllium components in reactors.

The specific 180-kev gamma emission rate for pure Ho_2O_3 was determined after six months irradiation in the MTR for a total nvt of $\sim 5.6 \times 10^{21} \text{ n/cm}^2$, followed by a four-month cooling period and purification of the weighable oxide sample. This permits determination of the half-life if the isotopic abundance of Ho-166 can be measured. An average value for the half-life of antimony-122m has been calculated as $4.21 \pm 0.02 \text{ min}$ by the IBM 650 computer using a weighted least-squares method. Yield ratios of indium-114 - indium-114m, produced by activation of 95%-enriched indium-113, were measured directly by following the β -decay through an absorber. The values obtained were $Y = \sim 2.5$ for thermal neutrons and $Y = \sim 1.7$ for epi-cadmium neutrons. These values differ considerably from values calculated from use of cross sections currently published in the literature.

The composition of irradiated thorium is currently being investigated by analyzing two thorium monitor samples in the hot alpha cave. These are in support of the fast chopper measurements of the total cross section of Pa-233 which have been completed but are not completely analyzed.

Data on the inelastic scattering of slow neutrons from sodium have been obtained at incident neutron beam energies of 0.026, 0.07, and 0.10 eV for sample temperatures of 23, 100, 110, and 150°C.

The study of five computational methods for calculating partial differential scattering cross sections was made and an evaluation of the methods prepared. The five methods were evaluated by comparing calculations on methane and propane with experimental results obtained from the MTR slow neutron velocity selector.

Examination of the full range of experimental results for the influence of interference scattering on the scattering of slow neutrons by methane is now possible because laboratory studies have shown that a correct quantum treatment of the rotational motion of methane is required to explain the scattering of low energy neutrons at forward angles. Also, computational procedures have been improved, and now large momentum transfers can be calculated.

A comprehensive review of the current status of conversion-electron spectroscopy used in decay scheme studies was compiled and is presented in this report.

Evidence for direct beta feeding of the 1.57-MeV state of molybdenum-94 from the decay of 6.6-min niobium-94m has been noted during coincidence experiments.

A programmer control was designed for the Nuclear Data ND-130 Analyzer which allows a restart after a prescribed time interval and also improves the maintenance schedule by turning off the tape perforator until required. Additional isolation was required in order to suppress noise in the ARMF regulating rod digital readout. This isolation includes output amplifiers at the angle encoder. A gated scaler and paper tape punch control unit was constructed for recording statistical data from neutron pulse counting systems. A magnetic tape pulse recorder was designed to record three channels of

individual neutron counts and a 100-kc timing reference channel to go with the gated scaler and data recording system. A time code generator was constructed to allow time marking on 16-mm motion picture film for use in transient information. A discrete time interval distribution of random pulses is derived from the binomial distribution that can be numerically evaluated from standard tables.

An overvoltage protector was designed which protects high-voltage systems when the pass tube in the high-voltage power supply shorts out. A circuit was designed that provides two timing pulses whose separation time is proportional to the ratio of one pulse to the sum of both pulses, where the pulses are the amplitudes of two 1-microsecond variable-height input pulses. An instrument was designed which measures the phasing of two-phased choppers in the MTR velocity selector to five parts in 10^5 corresponding to approximately two minutes of phase shift. A method was derived to obtain a rate signal from a mechanical milling machine without using a tachometer which utilizes the back emf of the motor but eliminates the effect of the armature current.

An improved air monitoring system was developed which will monitor the amount of alpha and beta activity in the air and give the ratio between the two. The operating noise of constant air monitors was reduced by the use of a simple noise filter attached to the muffler made of Tygon tubing with side branch resonators.

An IBM 650 program for the analysis of directional correlation of gamma ray data is being conducted to provide information concerning the nuclear levels and transitions involved. This program differs from similar ones in the past in that legendre polynomials are generated by the program, so that coincidence data may be taken at any desired set of angles between propagation vectors; whereas, investigators previously were limited to fixed location and numbers of angles.

The following new IBM 650 programs were completed and placed in service: an LSGF-1 program to perform a quantitative analysis, by the method of least squares, on gamma ray spectra obtained from the 256-channel analyzer, a MUFTR-E program using cross section and resonance decks from the MUFTR library to obtain averages of certain cross sections over the flux spectrum from a program problem, a REP1 program using the cross section and resonance decks of the MUFTR library to calculate resonance escape probabilities used in determining "L" factors for MUFTR, the BALD1 program to reduce and normalize data taken during testing of the chemical control out-of-pile loop, and a program called FLOOK which was written to compute the periodic input signal of a linear network when a periodic output signal and the transfer function are known.

The IBM 650 one-dimensional diffusion theory code (DDM) was again modified after it became apparent that the neutron balance used was not satisfactory. This modified program provides better approximations for derivatives at the boundaries so that the neutron balance will check out. It also includes certain absorption and source modifications.

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I. REACTOR PHYSICS AND ENGINEERING

1. REACTOR PHYSICS

1.1 Control Rod Water Gap Flux Peak (R. E. Oswald)

The thermal neutron flux peak which occurs in the water gap between the poison section and the fuel follower in an MTR shim rod becomes quite pronounced when the shim rod is partially withdrawn. A proposed design for a new MTR shim rod specifies a slightly greater water gap than is contained in the shim rods currently being used. Therefore, measurements were made to determine the effect that increasing the water gap might have on the flux peak.

The measurements were made in the ETRC in a control rod positioned at 18-1/2-in. withdrawal so that the top of the fuel region (3/8 in. + 1/8, -0 below the top edge of the fuel plate) was near the horizontal midplane of the core. This position was chosen to simulate the condition where the flux peak would be most pronounced. A standard ETR-type cadmium section was used as the poison piece and the fuel follower was a standard ETR fuel section (184 gram U-235 in sixteen 36-in. plates). The boron-stainless steel flux suppressor plates normally used in the ETR fuel sections were removed so the control rod was somewhat similar to an MTR shim rod. A 20-mil cadmium sleeve around the perimeter and extending below the poison section was used to vary the effective distance between the poison section and the fuel plates. This distance is 1-3/4 in. in the standard ETR control rod without the added cadmium sleeve.

Vertical thermal neutron flux profiles were measured with a water gap height of 5/8 in., 1 in., and 1-3/4 in., respectively. The resulting profiles are shown in Figure 1. As indicated in the graph the maximum thermal neutron flux peak increased 13% when the water gap was changed from 5/8 in. to 1 in. and increased 26% when the gap was changed from 5/8 in. to 1-3/4 in. The flux values were obtained from 1/4-in. bare and cadmium-covered gold wires. The data from all three flux runs were normalized to the same power level.

1.2 Control Rod Flux Suppressor Plates (R. E. Oswald)

The control rod fuel sections currently being used in the ETR contain a 1/2-in. flux suppressor plate at the top of each fuel plate. These suppressor plates are composed of stainless steel with 1 wt% boron enriched to at least 93% boron-10. The plates are employed in the control rod to suppress the pronounced thermal flux peaking that would otherwise occur in the water region between the fuel plates and the poison section of a partially withdrawn control rod.

Fig. 1 Vertical thermal neutron flux profiles.

It has been suggested that nickel plates, if effective enough as a flux suppressor, be used instead of the more expensive boron-stainless steel plates. Therefore, measurements were made in the ETRC to determine the relative effectiveness of nickel and boron-stainless steel flux suppressor plates. Three vertical thermal neutron flux profiles (Figure 2) were measured in the center region of a control rod positioned at 18-1/2-in. withdrawal to simulate the conditions where the peaking effect would be the most pronounced. A profile without suppressor plates was measured for a basis of comparison. The other two profiles were measured with type-A nickel and boron-stainless steel suppressors, respectively (the physical dimensions of both the nickel and the boron-stainless steel plates were identical). The flux measurements were obtained from 1/4-in. bare and cadmium-covered gold wires and the data from all three runs were normalized to the same power level.

Fig. 2 Vertical thermal neutron flux profiles.

As indicated in Figure 2, the nickel plates reduced the flux approximately 14% at the top of the control rod fuel region (3/8 in. + 1/8, -0 below the top edge of the fuel plate). The boron-stainless steel suppressors reduced the flux approximately 53%. The nickel plates reduced the maximum thermal flux peak 17%, and the boron-stainless steel plates reduced it 29%.

1.3 Resonance Absorption Integrals of Dysprosium and Tungsten (E. Fast, J. J. Scoville)

Preliminary values have been obtained for the resonance absorption integral of natural dysprosium, natural tungsten, tungsten-184, and tungsten-186. Reactivity measurements were made in the RMF inside of a 0.010-in.-thick walled cadmium tube, and compared to gold as a standard. The sample materials were dispersed in aluminum as a diluent in order to approach infinite dilution. A plot of the reactivity, ρ , as a function of the total sample atoms, N , was extrapolated to $N = 0$ to obtain $\left(\frac{d\rho}{dN}\right)_0$, the slope at infinite dilution. This quantity

is proportional to the resonance absorption integral.

The results which are relative to gold, and which assume a 0.30-ev cutoff energy are listed in Table I. The resonance integral for gold was assumed to be 1567 ± 40 barns per atom which includes the $1/v$ part and correction for the lower cutoff energy. In the case of gold and dysprosium, sufficient data were available to obtain an analytical fit to the equation:

$$\rho = A(1 - e^{-BN})$$

TABLE I
 RESONANCE ABSORPTION INTEGRAL MEASUREMENTS

Isotope or Element	Resonance Integral		
	Calculated	Literature [1]	RMF Measurements
Dy	1300	—	1270 ± 260
W (natural)	320	290 ± 35(a) 340(a) 353(b)	360 ± 70
W-186	460	355(c) 396(c) 481(b)	490 ± 100
W-184	6.34	8.6(b)	8.4 ± 2.0

(a) = absorption
 (b) = calculated
 (c) = activation

which has the slope

$$\left(\frac{dp}{dN} \right)_0 = AB$$

The uncertainties listed with the experimental values in Table I are estimated except for dysprosium in which case a calculated error in the slope was obtained, based on the fit to the above equation. The uncertainties given reflect the reproducibility in the experimental measurements and the published probable errors in the gold standard. Additional measurements, already in progress, are to be made in the ARMF both with and without cadmium shielding.

1.4 Lutetium as a Neutron Flux Spectrum Monitor (R. G. Nisle)

The nuclear properties of lutetium make it a unique substance for both the measurement of neutron temperature and for evaluating the flux spectrum. Several investigators [2, 3] have calibrated their foils in a heated thermal column where the neutron temperature was assumed equal to the thermal temperature. In another case [4] only relative values were used. The former procedure lumped the unknown terms into a calibration constant whereas these cancelled in the latter procedure. Such methods are not always useful in the general case, however, because they apply only to a particular setup, or because relative values are difficult to correlate when different experiments, performed under different conditions, are compared.

The general application of lutetium to the problems of flux spectrum and neutron temperature measurement is presently limited by a lack of precise values of its nuclear constants. For example the reported thermal cross

section varies from 13.3 to 35 barns for Lu-175. Furthermore, no information appears to be published on the branching ratio necessary to determine the saturated activity of Lu-176m which results in an 88-kev gamma of Hf-176.

In the work reported here, a procedure is described to measure the branching ratio, and the results are given both for this constant and the neutron temperature in two RMF locations.

The approach used here to evaluate the lutetium as a flux spectrum monitor is the inverse of the process that would be used if lutetium were being used to find F_1 , the parameter that characterizes the $1/E$ component of neutron flux [5]. The method consists of determining F_1 with the gold-copper monitor, and then calculating a value of the effective branching ratio in Lu-176m by use of this measured value of F_1 . Foils of Lu_2O_3 in copper, 0.005 in. thick, made by a powder metallurgy process in the MTR Cold Metallurgy Lab, were used for this purpose.

The procedure is outlined as follows:

Gold-copper alloy monitor wires and lutetium-copper foils were irradiated simultaneously in the RMF water hole and lattice positions. One wire was located close to a foil and three such wire-foil pairs were irradiated in each of the two RMF test positions. The gold-copper alloy was used to measure F_1 . This value of F_1 was used to find the ratio, R_p , of the effective cross section indices from Figure 3. The index ratio as defined in IDO-16612 [5] is

$$R_p (\text{Lu-175/Cu}) = \frac{[P(F_1, T)]_{\text{Lu-175}}}{[P(F_1, T)]_{\text{Cu}}} \quad (1)$$

and can be found experimentally from activation data by means of the equation

$$R_p (\text{Lu-175/Cu}) = \left[\frac{A}{\Sigma} \right]_{\text{Lu-175}} \left[\frac{A}{\Sigma} \right]_{\text{Cu}}^{-1} \quad (2)$$

where the A's are saturated activities, $\Sigma (\equiv N\sigma_0)$ is the 2200-m/s absorption cross section for the sample, N is the number of nuclei in the sample, and σ_0 is the 2200-m/s absorption cross section per atom. The saturated activities include a factor, b, that represents the fraction of the total number of disintegrations which result in a gamma ray of the energy that is being counted. This factor, b, is the effective branching ratio that is needed for Lu-

176m. A theoretical value of $\left[\frac{A}{\Sigma} \right]_{\text{Lu-175}}$

is found by use of R_p from Figure 3, Equation 2, and a measured value of

Fig. 3 Index ratio, R_p , vs F_1 for lutetium-175.

$\left[\frac{A}{\Sigma}\right]_{\text{Cu}}$. The saturated activity of Lu-176m is found, assuming $b = 1$. If this

last measured saturated activity is designated A' , then

$$\left[\frac{A'}{b\Sigma}\right]_{\text{Lu-175}} = \left[\frac{A}{\Sigma}\right]_{\text{Lu-175}} \quad (3)$$

Substitute Equation (3) in Equation (2) and solve for b

$$b = \frac{\left[\frac{A'}{\Sigma}\right]_{\text{Lu-175}}}{R(\text{Lu-175/Cu}) \left[\frac{A}{\Sigma}\right]_{\text{Cu}}} \quad (4)$$

Now, the value of b is independent of the place in the reactor at which the foil was activated. Hence, b , as measured from activation data, should be the same whether the irradiations are made in the RMF water hole or lattice positions. Table II shows the values resulting from the calculations just described.

TABLE II
DETERMINATION OF THE PARAMETER, b

RMF Position	F_1 Au-Cu	$R_p(\text{Lu-175/Cu})$	b Lu-176m
Water hole	0.0034	1.77	0.040
Lattice	0.0137	3.83	0.044

($\sigma_o = 35$ barns)
 $\sigma_o_{\text{Lu-175}}$

The agreement in b between the two RMF positions is seen to be fairly good and within the uncertainties inherent in the measurements of F_1 . The absolute value of b is dependent on the value of σ_o for which published values vary from 13.3 to 35 barns. However, the relative value is independent of σ_o .

The 6.8-day Lu-177 activity of the same foils was used to determine the neutron temperature in the two RMF positions. The values of σ_o for Lu-176 [4] and a value of 0.18 for b [6] were used. Table III shows the results, using the F_1 from Table II and Figure 4.

Although a lutetium-copper system was used in these experiments, operating

TABLE III
NEUTRON TEMPERATURE IN THE RMF

Position	$R_p(\text{Lu-176/Cu})$	$T (^{\circ}\text{C})$
Water hole	1.38	25
Lattice	1.50	78

$$\sigma_o = 2049, b = 0.18$$

policy forbids the use of copper in the MTR or ETR. Cobalt is an acceptable substitute for copper for such measurements. However, cobalt in turn is unsuitable for RMF measurements because of the low flux density.

1.5 Flux Synthesis Program - FLUSY (A. W. Brown)

The flux synthesis technique is useful in obtaining approximate two-dimensional flux shapes from one-dimensional diffusion problems or approximate three-dimensional flux shapes from one- and two-dimensional diffusion problems. The method involves finding buckling and flux-weighted constants for several radial planes in the reactor and using these averaged constants for a calculation in the axial direction [7]. The solution involves an iterative procedure with the results from the axial solution being used on the next iteration of the radial solutions. The method will converge to a unique eigenvalue which is identical for both radial and axial solutions when the radial solutions are combined with only one axial solution [8]. The approximate group-dependent flux at any point in the reactor is then a product of the calculated radial flux and the axial flux at that point, divided by the integral of the radial flux. The radial flux integral is taken over the same volume as was used to determine the flux-weighted constants.

A program (FLUSY) has been written for the IBM 650 to compute the necessary flux synthesis values of group-dependent buckling and flux-weighted reactor constants. The results of an MDMM* problem are used directly as input to FLUSY.

The buckling as computed by FLUSY is as follows:

$$B_1^2 = \frac{1}{\int_V D_i \phi_i dv} \left\{ \frac{\chi_i}{K_{eff}} \sum_{j=1}^I \left[\int_V v_j \Sigma_{fj} \phi_j dv \right] + \sum_{j=1}^{i-1} \left[\int_V \Sigma_R^{j \rightarrow i} \phi_j dv \right] \right. \\ \left. - \int_V \Sigma_{ai} \phi_i dv - \sum_{j=i+1}^I \left[\int_V \Sigma_R^{i \rightarrow j} \phi_i dv \right] - \int_V D_i B_{1i}^2 \phi_i dv \right\}$$

where

i = the energy group index

*S. Tong, MDMM P. P. Co. Program numbers 00.094 and 00.095

Fig. 4 Index ratio, R_p , vs neutron temperature for lutetium-176.

I = the total number of energy groups

$\Sigma_R^{j \rightarrow i}$ = the downscatter cross section for scattering out of energy group j into energy group i

D = diffusion constant

Σ_f = macroscopic fission cross section

ν = neutrons produced per fission

Σ_a = macroscopic absorption cross section

χ_i = fraction of fission neutrons born in energy group i

B_1^2 = buckling perpendicular to direction of computation

ϕ = flux as computed by MDMM

K_{eff} = eigenvalue computed by MDMM

The trapezoidal rule is used to evaluate the integrations because the neutron balance equations, as expressed in difference form, are satisfied exactly by the converged MDMM solution when the integrals involved are computed by this rule.

The flux-weighted reactor constants are computed by

$$\bar{\Sigma}_i = \frac{\int \Sigma_i \phi_i dv}{\int \phi_i dv}$$

where Σ stands for $D, \Sigma_a, \nu\Sigma_f$, and Σ_R defined above.

All of the volume integrals are taken over the regions specified on input cards.

If desired FLUSY also will punch out for each region the following regional values:

$$A_i = \int \phi_i dv, \quad \bar{\phi}_i = \frac{\int \phi_i dv}{C(X_2^{\rho+1} - X_1^{\rho+1})}$$

$$\nu_1 \Sigma_{f1} A_1, \nu_1 \Sigma_{f1} \bar{\phi}_1, B_1^2, \sum_{i=1}^I \nu_1 \Sigma_{f1} A_i$$

$$\sum_{i=1}^I \nu_1 \Sigma_{f1} \bar{\phi}_i, \text{ and Area} = C(X_2^{\rho+1} - X_1^{\rho+1})$$

The following tabulation gives the values of ρ , dv , and C for the three geometries computed by MDMM:

	ρ	$\frac{dv}{dx}$	$\frac{C}{dx}$
Slab	0	dx	1
Cylinder	1	$2\pi r dr$	π
Sphere	2	$4\pi r^2 dr$	$4/3\pi$

The following inequalities must be satisfied for any problem run on FLUSY:

$$I \geq 2$$

$$4K + IN + 2I + 0.5IK(I + 9) < 1419$$

$$2K + IN + 7I + 5IK + 0.5I(I - 1)(K + 1) < 1419$$

If the values of B^2 put into the MDMM problem are group dependent then the following inequality must be satisfied:

$$3N + 2K + IN + 4I + 0.5IK(5 + I) + I < 1419$$

If the B^2 values are group and regional dependent this last inequality is changed to

$$3N + 2K + IN + 4I + 0.5IK(5 + I) + IK < 1419$$

where I = total energy groups

K = total number of regions

N = total number of mesh intervals

There are program stops built into FLUSY to check these inequalities. However, if the regional values are not wanted, the value of 1419 in the inequalities is increased to 1478 and the console switches set to minus.

2. THORIUM PROGRAM

2.1 Thorium-Uranium-233 Cycle Studies (R. G. Nisle, D. A. Millsap)

A sample design has been completed for a long-term burnout study of the thorium-uranium-233 conversion chain. Criteria for the design of such samples are mechanical ruggedness, capability of withstanding a high irradiating flux level, provisions for flux monitoring, a practical reactivity worth, and a convenient shape. These criteria together with the results of previous calculations suggested a thin-walled tubular sample of Th-232 containing a maximum of about 2.5 wt% of U-233. Hydraulic and heat transfer calculations were made to determine the best dimensions to give the lowest wall temperatures. The results of such calculations show that for any given weight and length of active core and cladding thickness there is some outside diameter for which the wall temperatures are a minimum. Other calculations showed that a 75-gram sample of the above composition would have a reactivity worth about equal to

that of the Advanced Reactivity Measurement Facility (ARMF) regulating rod. It was also shown that the outer cladding could safely be made somewhat thicker to allow for slots for the insertion of flux wires. The results of the heat transfer calculations for the 75-gram sample are shown in Figure 5.

The recommended design, therefore, consists of a 75-gram active core containing a maximum of 2.5 wt% of U-233 and the remainder Th-232. The outside diameter including cladding is 1.00 in. and the length of the active core is 4.00 in. The cladding material chosen is zirconium or zircaloy-2. This choice was based on the extrusion and alloying properties of zirconium as compared with those of aluminum.

Removable end fixtures will be used to center the sample in an X-basket and to hold flux wires in place. This design also permits measurements to be made in the ARMF with a minimum of extraneous structural parts. Figure 6 is a view of the assembled sample ready for irradiation.

Fig. 5 Calculated sample wall temperatures.

Fig. 6 Sample with centering fixture.

3. FUEL ELEMENT DEVELOPMENT

3.1 Sample Fuel Plate Irradiations (W. P. Scarrah, G. W. Gibson, M. J. Graber)

In addition to the fuel samples of uranium-aluminum and uranium oxide-aluminum cermet clad with 1100, APM, X8001, and 6061 aluminum alloys which were irradiated during the quarter, several unfueled samples of aluminum-alloy cladding materials were irradiated in the L-51 position of the MTR to determine mechanical property changes during irradiation. One sample of Dy_2O_3 stainless steel was added in collaboration with the HFBR (Brookhaven) control rod program.

As discussed in IDO-16574^[9], the initial experiments with burnable poison-fuel mixtures have emphasized that their combined effect on reactivity

is too complex to evaluate without more basic irradiation data on the burnable poisons alone. In order to secure these data, a program has been devised to study the two major variables affecting poison reactivity, ie, neutron flux spectra and self shielding.

During the quarter, the first set of samples required for this program were fabricated. These plates were the same size as the sample fuel plates, 3.213 x 2.875 in. overall, with a core thickness of 0.020 in. and a cladding thickness of 0.015 in. The cores each contained 136.8 mg of Dy_2O_3 dispersed in aluminum powder. The picture frame and cladding material were of 1100 aluminum.

3.2 Mechanical Property Measurements (G. W. Gibson, M. J. Graber, J. M. Beeston, E. H. Porter)

Microhardness tests were completed on several irradiated fuel samples. The results, although indicating the general increase in hardness expected with irradiation, gave a wide range of readings so that generalizations are difficult. As observed in earlier work [9, 10], the matrix material exhibited greater increases in hardness than the cladding.

Tensile testing was completed on 94 specimens of irradiated sample fuel plates. Fuel cores were composed of uranium-aluminum alloys, UO_2 -aluminum cermets and U_3O_8 -aluminum cermets; cladding materials were 1100, 6061, APM 257, X8001, X8003, and 5052 aluminum alloys. The most significant effect of the fission damage was the loss of ductility. At low levels of irradiation [about 2×10^{20} neutrons/cm² (thermal)] the maximum elongation retained by any one specimen was 4.5%; at 20×10^{20} , the maximum was 1%; and at 26×10^{20} , it was zero. In all these ranges of irradiation, some specimens showed zero ductility. Two specimens (see Figure 7) broke in three pieces while many others separated at the core-clad interface (see Figure 8). Tensile strengths increased from 3 to 125% with most increases falling between 30 and 80%. Two tests showed a decrease in tensile strength.

As part of a program to study radiation effects on reactor structural materials, preliminary conceptual designs and flow schematics have been initiated for an in-pile fatigue test loop. To be investigated are the fatigue life of structural steels as a function of temperature, irradiation exposure, and possibly dose rate, and the mechanism of defect generation and propagation of microcracks.

3.3 Uranium Oxide-Aluminum Reaction (J. M. Beeston, M. J. Graber)

A second sample of UO_2 dispersion in aluminum, thermally treated 65 hours at 600°C, was analyzed by electron beam microanalysis to determine the products of the UO_2 -Al reaction. A standard containing UAl_4 served to verify that UAl_4 was one of the products of the reaction. The intermetallic UAl_4 was also a product in the first thermally treated (40 hours at 600°C) UO_2 dispersion sample. As reported [10] for the first sample, the dispersion of Al_2O_3 in the reaction region must be extremely fine since it does not fluoresce under the electron beam. The diffusion of aluminum into the original UO_2 particle is evidently complete between 40 and 65 hours at 600°C as no solubility of aluminum was noted in the sample treated for 65 hours, while the one treated for 40 hours exhibited a gradient of aluminum over six microns of annular cross-sectional width of a 25-micron cross-sectional particle.

Fig. 7 Sample fuel plate tensile specimen which became embrittled due to fission damage.

Fig. 8 Sample fuel plate tensile specimen which became embrittled due to fission damage.

Since the production of UAl_4 was complete in these samples, with no plate warpage, this means that under certain conditions, the $UO_2 + Al$ reaction may take place without dimensional distortion, whereas under other conditions, it is well known that the production of UAl_4 produces dimensional instability with considerable warpage. These conditions have not, as yet, been completely defined.

3.4 EGCR Fuel Element Development (F. O. Haroldsen, E. H. Porter, M. W. Ellingford)

The irradiation of 13 EGCR capsules (UO_2 -SS) continued during the quarter with one of the capsules achieving a burnup of more than 7300 Mwd/MT. One capsule was discharged during the Cycle 45 shutdown with a calculated burnup of 6188 Mwd/MT.

Because the fuel burnup rates for several capsules during the past two or three cycles have exceeded the design value due to increases in neutron flux in the capsule positions, an absorber in the form of a bundle of nickel rods was placed in the reactor in two different positions to depress the neutron flux somewhat in these capsules. As a result, the high burnup rates were reduced 13 to 27% in the critical capsules and 5 to 10% in other capsules farther removed from the absorber.

A rather large variation in burnup values, as determined by calculations based on temperature measurements and as measured by radiochemical analysis of Co-60, Cs-137, and Ce-144, resulted in elimination of the 0.7

experience factor, which previously had been used in the burnup calculations (for 5-mil annulus capsules). The indicated burnup rate for these capsules, therefore, was increased by a factor of 1.43.

3.5 Pressure Deflection Testing of Fuel Elements (G. W. Gibson, M. L. Griebenow)

Three aluminum alloys have been static-pressure tested at room temperature to evaluate the strain predicted for fuel plates in a test reactor. The lateral pressure differential that produces the fuel plate loading and subsequent deflection is developed by adjacent channels of unequal spacing. Each of the three alloys (6061-0, 1100-0, and X8001-0) was used to fabricate a non-fueled plate 0.080 in. x 3.9 in. x 2 ft with a 5.4-in. radius of curvature. The 1100-0 and the X8001-0 plates were pinned, while the 6061-0 plates were roll-swaged into 2-1/2-in-wide side plates so that they formed the outside plates of a fuel section assembly. The side plates were supported at three axial positions (centered in the 2-1/2-in. dimension) by bolts. For 0.250-in. side plates, this should give a fairly close approximation of the side plate deflection that would be encountered in actual practice while in the case of 0.150-in. side plates, the side plate deflection would be higher than the deflections in actual elements due to the lack of interior support plates.

Figures 9 and 10 show the stress-strain relationships for the various alloys and side plate thicknesses at room temperature. It is apparent that at room temperature any of the alloys could tolerate at least 10 to 15 psi across the plate. The deflections are in the direction to reduce the ΔP ; ie, if at startup there was 10 psi across it, the plate would deflect up to 0.040 in. or 0.050 in. depending on the alloy, side plate thickness, etc, and stabilize with a pressure of less than 10 psi acting across it. It would deflect until the elastic force equaled to ΔP driving force remaining. The plastic flow yielding and elastic deformation both would occur as the reactor flow was brought up to design values (prior to irradiation which would tend to reduce the elongation) and only the elastic component would be required as the reactor went up and down in flow for refueling, shutdowns, scrams, etc. Additional work is planned using test sections with 19 plates to be tested at elevated temperatures.

Fig. 9 Total deflection of ATR plate 19.

Fig. 10 Plastic deflection of ATR plate 19.

3.6 Prototype Tests (M. L. Griebenow, O. L. Knighton, D. E. Williams, M. W. Ellingford)

Two 32-plate MTR-type (250-gram U-235) elements were hydraulically tested. The elements consisted entirely of 0.030-in.-thick, 6061-T6 clad fuel plates separated by nominal 0.068-in. channels. The outside dimensions were the same as existing MTR elements. In a partial core loading, these dimensions were necessary to prevent any increase in ΔP across adjacent standard MTR elements even though by so doing an unduly high ΔP results across the external plates of the test element. Any full core loading would, of course, have external channels equal to the internal channels. In the hydraulic test, a 0.065-in. channel was adjacent to an external channel of 0.120 in. Both elements withstood 140% of design flow at 100°F for 15 minutes*. A measurement of individual channel flows showed a maximum variation of 18.7% for the internal channels; the external channels, being larger, naturally had higher flow rates. This amounts to a range in channel velocity from -10.3 to +8.4%. Plans are made to insert the two elements in the MTR as soon as possible.

A "dummy" concentric-rounded square fuel element was fabricated for hydraulic testing. The fuel section was made of mild steel and the lower end box was made of aluminum. This fuel element was tested in the hydraulic facility. Although a detailed analysis has not been made, a preliminary investigation of the data indicates that for a given ΔP across the fuel element, the total flow for this element is higher than for a standard ETR fuel element. Conversely, the same ETR coolant flow would have resulted in a lower pressure drop and, thereby, less pumping power. In addition, the flow distribution across the fuel element appears to be relatively uniform.

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* This is a standard acceptance test for MTR fuel elements

II. NUCLEAR PHYSICS

1. CROSS SECTIONS PROGRAM (M. S. Moore)

1.1 Pu-241 Fission Cross Section (T. Watanabe, O. D. Simpson)

The Pu-241 fission cross section was measured from 0.025 to 100 ev. A flight path of 8.5 meters with an instrument resolution of 0.5 to 5.0 $\mu\text{sec}/\text{meter}$ was used. The data are shown in Figures 11 and 12. The data in Figure 11 represented by the open and closed circles were normalized one to another by requiring the integrals $\int \sigma dE$ from 2 to 100 ev to be equal. The data represented by the solid circles in Figure 11 are not presented above 3 ev because of the poor instrument resolution. The measured data represent a relative fission cross section, and absolute normalization was done at the peak of the 6-ev level by using the total cross section and the multilevel resonance parameters obtained from the total data [11], by the following procedure:

Most of the cross section of the 6-ev level is fission and was obtained from the following equation,

$$\sigma_{nf} = \sigma_{nT} - \sigma_{nn} - \sigma_{n\gamma}$$

where

σ_{nf} = fission cross section

σ_{nT} = total cross section

σ_{nn} = scattering cross section

$\sigma_{n\gamma}$ = capture cross section

Fig. 11 The fission cross section of Pu-241 from 0.025 to 100 ev. The data which are shown by a dashed curve from 2 to 20 ev are given in Fig. 12 on an expanded energy scale.

Fig. 12 The fission cross section of Pu-241 from 2 to 20 ev.

Estimates of σ_{nn} and $\sigma_{n\gamma}$ using the multilevel analysis of the total data are as follows: $\sigma_{nn} = 14 \pm 4$, and $\sigma_{n\gamma} = 14 \pm 7$. Combining these values with the measured $\sigma_{nT} = 434 \pm 6$ barns and solving for σ_{nf} , we obtain the value of 406 ± 10 barns for the fission cross section at the 6-ev level. Normalizing the fission data to this value we obtain a 2200-meter/sec fission cross section of 965 ± 40 barns.

Table IV compares this 2200-meter/sec value with other experimental measurements of the fission cross section of Pu-241. It is of interest to note that the integral measurements are, on the average, significantly higher than the (monoenergetic) spectrometer measurements. This is very suggestive of distortions introduced by the reactor spectra in which the integral measurements were made. Although at 0.0253 ev the cross section is very nearly $1/v$, a significant contribution to the integral cross section can be introduced if the reactor spectrum contains appreciable neutrons with energies from 0.1 to 0.3 ev. These effects are being investigated.

TABLE IV

2200-METER/SEC PU-241 FISSION CROSS SECTION

σ_{nf} (barns)	Experiment	Reference
1100 ± 30	Integral	ANL—1955 ^[12]
858	Integral	KAPL—1955 ^[13]
1060 ± 210	Integral	ANL—1956 ^[14]
1013 ± 15	Integral	Chalk River—1958 ^[15]
935 ± 40	Time-of-flight spectrometer	Harwell—1955 ^[16]
956 ± 43	Crystal spectrometer	Hanford—1959 ^[17]
965 ± 40	Time-of-flight spectrometer	MTR—1962 ^[18]

The solid line in Figure 13 shows the multilevel fit to the fission data as predicted by the resonance parameters obtained from the total cross sections [19]. There are several places where the fit does not describe the fission data and modifications of the parameters are needed. The data are presently being reanalyzed and it is expected that allowing variations in the radiative capture widths will permit a fit to both the total and fission data, within the accuracy of the data.

Fig. 13 The fission cross section of Pu-241, multiplied by the square root of the neutron energy, as a function of neutron energy below 12 eV. The solid curve shows the fission cross section predicted from the multilevel fit to the total cross section of Pu-241.

1.2 Fast Backgrounds (O. D. Simpson, N. H. Marshall, G. E. Stokes)

A technique has been developed whereby the fast neutron leakage through the chopper rotor (fast background) can be determined by using a $B_4^{10}C$ filter. A program has been written for the IBM 650 computer where the fast background is obtained from the incident and transmitted neutrons of a $B_4^{10}C$ filter. A water sample having a transmission of ~5% was found to yield the correct cross section in the region of 500 eV when the fast background constitutes 80% of the transmitted beam. Additional data are being taken and an IDO report is being prepared describing the technique in detail.

2. NUCLEAR CHEMISTRY (W. H. Burgus)

2.1 Beryllium Gases Experiment (R. L. Tromp)

Difficulties in the melting of irradiated beryllium for gas analysis mentioned in a previous quarterly report [20] have been rectified. One beryllium sample with a reported fast neutron dosage of 0.891×10^{21} n/cm² and thermal neutron dosage of 1.001×10^{22} n/cm² was quickly and completely melted in the vacuum line gas analysis apparatus and the gaseous products collected and analyzed. Detailed calculations were made of the quantities of gases expected, based upon the reported neutron dosages. These are compared with the measured quantities in Table V.

Agreement in total gas content is quite good but considerable variation exists in individual contributions such as $(H^1)_2$ and He^4 . The nvt values for the measured sample may be of limited accuracy and a detailed compilation based on further flux measurements and reactor operating data will be made to provide a check on these values. As has been observed with other samples previously analyzed, the measured $(H^1)_2$ content is a factor of 10 greater than the calculated content. In an attempt to explain this, a piece of unirradiated

TABLE V

CALCULATED VS MEASURED QUANTITIES OF GASES
IN NEUTRON-IRRADIATED BERYLLIUM

Calculated(a) (cc gas/cc Be)		Measured Sample D-6(b) (cc gas/cc Be)	
Gas	Quantity	Gas	Quantity
He ⁴	2.043	He ⁴	1.52
(H ³) ₂	0.113	(H ³) ₂	0.09
(H ¹) ₂	0.025	(H ¹) ₂	0.29
He ³	0.0002	He ³	0.01
Total	2.181	Total	1.91

(a) Fast nvt = 1.00×10^{21} n/cm²; thermal nvt = 1.1×10^{22} n/cm²; irradiation time = 3.96×10^7 sec.

(b) Fast nvt = 0.891×10^{21} ; thermal nvt = 1.001×10^{22} ; irradiation time = 3.96×10^7 sec.

beryllium of similar fabrication history but from a different source was melted for blank gas determination. Only (H¹)₂ was detected and amounted to 0.12 cc/cc beryllium. Both additional irradiated and unirradiated beryllium pieces are available for further analysis.

Using other samples of beryllium with irradiation histories similar to the one melted for determination of gas content, a series of swelling vs temperature experiments has been started. The beryllium irradiations were carried out in the ETR where the temperature during irradiation was < 100°C. Each beryllium sample, under high vacuum, was held at a fixed temperature for various lengths of time. After cooling to room temperature, the density was determined by a water pycnometer method, which for the size samples used, had a standard deviation of less than 0.2%. Within experimental error, none of the samples exhibited a change in density between the beginning and end of the irradiations. Upon heating at 600°C, no swelling (decrease in density) was observed within experimental error. Heating at 700°C for 16 hours resulted in only 1.26% swelling, but at 800°C, heating for 30 hours produced 15.01% swelling. The swelling vs temperature results are presented in Table VI and plotted graphically in Figure 14.

Micrometer measurement of the 800°C piece after 20-hours heating showed the swelling to be non-uniform and more pronounced in the direction of the shorter dimension. The length of the originally cylindrical piece had increased by 2.65% and the diameter increased variably between 4.36% and 6.96%.

In addition to swelling, the evolution of reaction product gases at elevated but sub-melting temperatures constitutes another potential hazard associated

TABLE VI

SWELLING OF IRRADIATED BERYLLIUM AS A FUNCTION OF TEMPERATURE

Beryllium irradiated to fast (> 1 Mev) neutron dosage of $\sim 1.0 \times 10^{21}$ n/cm², thermal neutron dosage of $\sim 1.0 \times 10^{22}$ n/cm².

600°C		700°C		800°C	
Total Heating Time (hr)	Density	Total Heating Time (hr)	Density	Total Heating Time (hr)	Density
0(a)	1.8471	0(a)	1.8488	0(a)	1.8482
2	1.8446	1	1.8346	1	1.7823
8	1.8419	2	1.8295	2	1.7302
16	1.8446	4	1.8288	4	1.6658
32	1.8473	8	1.8284	6	1.6482
		16	1.8258	10	1.6361
				20	1.6168
				30	1.6070

(a) As removed from reactor, before annealing experiments were started.

with reactor beryllium components (because of the tritium released). So far in the swelling measurements, no significant gas evolution was observed at either 600°C or 700°C but at 800°C a small amount of non-condensable gas (hydrogen and helium) was collected during each heating interval. After 30 hours at 800°C an accumulation of gas equal to approximately one-half the total gas content was released. Mass spectrographic analyses have shown the earliest evolution to be nearly all hydrogen, while tritium and helium evolution begins at a later time. Early evolution of hydrogen may be due to a combination of adsorption on the surface of the unirradiated metal, surface capture of recoil protons from the reactor cooling water, or higher mobility of hydrogen.

Fig. 14 Density change at constant temperature for beryllium previously irradiated to 1×10^{21} fast n/cm² and 1×10^{22} thermal n/cm².

2.2 Fission Mass Yields of U-235 as a Function of Neutron Energy (K. T. Faler, R. L. Tromp)

An additional neutron crystal spectrometer irradiation was carried out at 0.06 ev in an attempt to decrease the uncertainty of the "R" value (peak-to-valley ratio/peak-to-valley ratio at thermal energy) previously reported [21] at this energy. The new data permitted rejection of two previous values by the "Studentized" deviation test and the new value at 0.06 ev is 0.783 ± 0.021 . This work continues intermittently depending upon the availability of "cold neutron" and spectrometer time when these facilities are not in use for other experimental work.

2.3 Ho-166m Half-Life Determination (B. Keisch, K. T. Faler)

After a six-month irradiation in the MTR for a total nvt of $\sim 5.6 \times 10^{21}$ n/cm² followed by a four-month cooling period, a sample of Ho₂O₃ was purified three times by ion exchange and prepared in a weighable oxide form for γ counting. The specific 180-kev gamma emission rate for the pure Ho₂O₃ was determined. Mass analysis of the sample (still pending) is being carried out in order to determine, if possible, the fraction of holmium which is Ho-166. This along with the absolute disintegration rate will permit accurate calculation of the half-life which is of the order of ~ 30 years.

2.4 Sb-122m Half-Life Determination (B. Keisch)

In the course of yield ratio measurements for the Sb-122 - 122m isomer pair, it was discovered that the half-life for Sb-122m listed in the literature was in error. Accordingly, new measurements were made as previously reported [22]. The half-lives have now been computed by the IBM 650 computer using a weighted least squares method. The results are: sample 1, 4.21 minutes; sample 2, 4.23 minutes; sample 3, 4.19 minutes; average $4.21 \pm .02$ minutes. The long-lived tail which was subtracted amounted to 5% or less of the initial total activity in each case.

2.5 In-114 - In-114m (B. Keisch)

Yield ratios were measured directly by following the β decay (through an absorber) of In-114 and In-114m produced by activation of 95%-enriched In-113. Experiments were performed using thermal neutrons, epi-cadmium neutrons, and a mixture of the two. The values obtained range from $Y = \sim 2.5$ for thermal neutrons to $Y = \sim 1.7$ for epi-cadmium neutrons where the yield ratio $Y = \frac{\sigma_{\text{metastable}}}{\sigma_{\text{ground state}}}$. These values are considerably different from

the one calculable from use of the cross sections currently in the literature [23] ($Y = 23$), and so direct measurements of the formation cross sections are being made. Thus far, a value for the thermal neutron cross section for the formation of 72-sec In-114 has been obtained as 2.9 barns (literature value = 2 barns) and a tentative value for the formation of 50-day In-114m has been obtained which indicates that the literature value may be too high by a factor of 10. Measurements are being continued in order to confirm these results.

2.6 Ir-192 - Ir-192m Yield Ratio (B. Keisch)

The previously reported yield ratio [24] for the Ir-192 - Ir-192m isomeric pair indicates that the current value in the literature [25] is too low (literature values = 0.84, the previous results = 1.6). Determinations are now being

made using $4\pi\beta\text{-}\gamma$ coincidence-counting techniques, as before, but with an arrangement in which counting efficiency for the weak conversion electrons in the decay of Ir-192m can be measured. Efficiencies of $\sim 70\%$ have been obtained as opposed to a 90 to 100% efficiency previously assumed. Thus the value of the yield ratio is considerably higher (~ 2.0).

2.7 Preparation of U-236 Sample (J. W. Coddington)

Uranium from the MTR-30-6 irradiation of U-236 has been recovered and purified by a method outlined previously^[26]. The capsule had been irradiated in the MTR to about 1.5×10^{22} n/cm² in order to preferentially burn out the 4% U-235 initially present. Gross separation of the uranium from neptunium, plutonium and most fission products was accomplished by anion exchange; solvent extraction provided a final purification step.

The uranium product has been analyzed in several ways: a mass analysis was performed by the analytical group at the Chemical Processing Plant, a spectrophotometric analysis was done to measure total uranium recovered, and both alpha pulse-height and gamma scintillation spectra were taken to determine product purity. Table VII shows the uranium isotopic composition before and after irradiation. The final U-235 content seems satisfactorily low for the purpose of measuring the fission cross section of U-237 by in-pile fission counting of the re-irradiated material.

TABLE VII
ISOTOPIC COMPOSITION OF
U-236 SAMPLE MTR-30-6

Nuclide	Before Irradiation (wt%)	After Irradiation (wt%)
U-234	—	0.049
U-235	4.39	~ 0.005
U-236	93.60	97.556
U-238	2.02	2.389

Based on the optical density of the product solution at 415 m μ , the final product contained 62.1 mg of total uranium. Alpha pulse-height analysis showed 60.8 mg of U-236 in the final product; this is equivalent to about 62.3 mg of total uranium, based on the mass analysis.

Figure 15 shows the alpha spectrum. The amounts by weight of the various alpha active constituents have been calculated from the alpha spectrum and are given in Table VIII.

A gamma scintillation spectrum of the product, shown in Figure 16, indicates that the major fission product contaminant is Ru-106. The low levels of fission products present will present no difficulties when the fission cross section of U-237 is determined after formation of the U-237 by re-irradiation of the U-236.

2.8 Total Cross Section of Au-198 (F. B. Simpson, R. P. Schuman)

An additional sample of gold^[27] for MTR chopper cross section measurements of Au-198 was irradiated for two days in the hydraulic rabbit facility of the MTR. The sample was then taken into the chemistry alpha cave, where the welded aluminum rabbit was cut open and the gold sample removed and loaded into a chopper sample block. The gold sample was the first high-level gamma ray-emitting sample handled in the alpha cave, and allowed a check

Fig. 15 Alpha pulse-height analysis of electroplated U-236 product from MTR 30-6.

Fig. 16 Gamma ray spectrum of U-236 product from MTR 30-6, showing the major fission product contaminant, Ru-106.

TABLE VIII

U-236 PRODUCT COMPOSITION BASED ON ALPHA PULSE-HEIGHT ANALYSIS

Nuclide	Weight in Total Product (mg)
U-236	60.8
U-234	$2.6 \times 10^{-3}(a)$
Pu-238	2.0×10^{-5}
Cm-244	3.0×10^{-7}

(a) Corresponds to 0.043 wt%; cf 0.049% from mass analysis.

to be made of the shielding. Radiation leaks were discovered through the large plugs in the front face of the cave; these were stopped by adding more lead shot to fill the plugs (which were incompletely filled by the construction contractor) and by placing lead bricks in front of the remaining small leaks.

Gamma scintillation spectra have been taken of the irradiated gold after about 40 days decay. The gold shows 270-day Ag-110 and 74-day Ir-191, as well as Au-198 and Au-199. The decay of the gold activities is being followed, and an attempt will be made to calculate the initial concentration of Au-198 and Au-199 from the gamma spectra.

2.9 Total Cross Section of Pa-233 (F. B. Simpson, R. P. Schuman, J. W. Coddling, H. G. Miller, J. R. Berreth)

A new sample of thorium metal was irradiated in the ETR in order to produce a sample of thorium containing Pa-233 for total cross section measurements [28] of the latter isotope. The irradiation capsule contained two rectangular blocks of thorium metal (1.526 cm in the path of the chopper neutron beam, 2.862 cm long, and 0.140 cm wide), sealed inside aluminum blocks. In addition, two small thorium

monitors sealed inside aluminum and two cobalt-aluminum wire flux monitors were included in the irradiation capsule. The aluminum-clad thorium pieces were surrounded by close fitting heat sinks so that the heat could be conducted to the wall of the capsule.

The capsule was irradiated for one cycle in the G-12 NW position of the ETR. After the irradiation, the capsule was brought to the chemistry alpha cave. The capsule read $\sim 40,000$ r/m at 4 in., about nine hours after the end of the irradiation. The alpha cave was adequate to handle this level of activity, and the radiation level at the working face was less than 8 mr/hr.

The thorium pieces were removed by first sawing the ends off the capsule and cutting the capsule in two (between the two thorium samples). The shell of the capsule was then slit on both sides by sawing, and the thorium samples pried out from the slit sides of the capsule and the heat sinks. The two thorium samples were then placed side by side in a special chopper block (giving a neutron path in the irradiated thorium of 3.05 cm) and the block was taken to the chopper for cross section measurements.

The composition of the irradiated thorium was calculated from the expected flux of 7×10^{14} n/cm² sec and from literature values of the cross sections listed in Table IX. The calculated composition of the thorium at the end of

TABLE IX
CROSS SECTIONS AND HALF-LIVES USED TO
CALCULATE COMPOSITION OF IRRADIATED THORIUM^[29]

<u>Nuclide</u>	<u>Half-Life</u>	<u>Thermal Cross Section (barns)</u>	<u>Resonance Integral (barns)</u>
Th-232	Assume stable	7.4	21.0 ^[30]
Pa-233	27.4 days	56	670
U-233	Assume stable	591	927

irradiation corresponds to a total thickness for the neutrons of 8.769×10^{22} atoms Th/cm², 8.414×10^{20} atoms Pa-233/cm², and 2.362×10^{20} atoms U-233/cm².

A more accurate composition of the irradiated thorium is being obtained by analyzing the two thorium monitor samples in the hot alpha cave. The aluminum cladding was first dissolved from the samples with NaOH + NaNO₃ solution. Then the thorium plus insoluble residue was dissolved in HNO₃ + HF and a known U-236 spike added. Uranium has been separated from a portion of each solution by hexone extraction, and will be mass analyzed at CPP in order to determine the U-233 and U-234 content of the samples at the time of separation. After about six half-lives of Pa-233 decay, uranium will again be separated from the thorium, and the uranium mass analyzed to obtain the new U-233 content. The amount of increase of U-233 will correspond to the Pa-233 decay, and from the decay, the initial Pa-233 content can be determined.

A preliminary value of the Pa-233 content of the thorium has been obtained by gamma scintillation counting of very small aliquot from one of the thorium monitors. The calculation assumed that 95% of the decay of 27.4-day Pa-233 proceeds through the 313-kev, 301-kev, and 342-kev gamma rays, that the ~ 80% abundant 313-kev gamma had $ek/\gamma = 0.74$ and $K/L = 4.0$, and that the other two gamma rays were converted to about the same extent as the 313-kev gamma ray [31]. Based on the above assumptions, the gamma activity corresponds to a Pa-233 concentration at the end of irradiation of $0.0090 \times$ initial thorium so that the Pa-233 thickness in the neutron beam is 8.0×10^{20} atoms/cm². The calculated amount of Pa-233 agrees with the above measured amount well within the errors of the two values.

2.10 Preparation of Separated Pa-233 for Cross Section Measurements (J. W. Coddington)

Considerable information about the total cross section of Pa-233 has been obtained by measuring the neutron transmission of samples of irradiated thorium with the MTR fast chopper. However, because large corrections must be made for the absorption due to the thorium, the U-233, and other unwanted products of irradiation, and further because the composition of the sample is somewhat uncertain, the accuracy of the cross section determinations of the Pa-233 is not as great as desired, especially in the thermal energy region. Consequently, plans are being made to produce chemically separated samples of Pa-233 for MTR fast chopper cross sections.

Several hundred milligrams of Pa-233 will be produced by irradiating thorium metal in a high flux position of the MTR or ETR. After irradiation, the Pa-233 will be chemically isolated from the thorium, purified from U-233, U-234, fission products, and other impurities, converted to pure oxide, and then made into a suitable chopper sample by compacting the oxide with aluminum metal powder. After an appreciable amount of U-233 has grown into a sample, it will be necessary to dissolve the sample, repurify the Pa-233, add additional pure Pa-233 to make up for the decay, and make a new Pa-233 oxide - aluminum compact.

A solvent extraction scheme currently is being scoped for the separation and purification of 100-milligram quantities of Pa-233. The separation is complicated by the very high level of beta activity, by the relatively limited time available for processing, and by the complex and difficultly reproducible chemical behavior shown by protactinium in even quite dilute aqueous solutions. Accordingly, some preliminary work on the extraction of protactinium and uranium is being done using Pa-231 and U-233 as tracers.

Initial extraction measurements were made at the following aqueous conditions: 0.4 M Th⁴⁺, 0.2 M Al³⁺, 0.1 M F⁻, and about 6 M HCl. Such an aqueous phase can easily be obtained by dissolving the irradiated thorium in 8M HCl with some HF added, then adjusting the resulting dissolver solution with AlCl₃. Under these conditions 85% of the Pa-231, 24% of the U-233, and 0.2% of the thorium were extracted into methylisobutylketone.

Data collection for separation studies is considerably simplified if both components can be analyzed in a single measurement. Since the energies of the major alpha groups differ by about 200 kev, the amounts of Pa-231 and U-233 in any sample can be determined by careful alpha pulse-height analysis. Figure 17 shows the superimposed alpha spectra of Pa-231 and U-233, taken with a silicon surface barrier counter. The relative position of

the peaks shows that good analyses will require high resolution. The counting plates must be thin, even layers of active material, in order to obtain good alpha energy resolution. Sublimation is a technique for producing thin uniform counting samples and the spectra in Figure 17 were taken from sublimed plates. The sublimer, designed by J. R. Berreth, consists of a tungsten filament suspended between electrodes and located about 1 in. beneath a revolving sample holder. The sample holder can position any one of six counting plates above the filament. The sublimer is enclosed in a removable bell jar during evacuation. After the tungsten filament has been degassed by heating at a reduced pressure, a sample is placed on it and dried (if in solution) by slow warming. The system is then evacuated to $>10^{-3}$ mm Hg, a counting plate positioned over the sample, and the filament brought to a suitable temperature (usually around 1500°C for oxides) for sublimation of the sample. From 10 to 15% of the sample can be transferred to the plate, and deposited in a thin, uniform layer. The counting plates exhibit excellent alpha energy resolution.

Fig. 17 Alpha spectra of U-233 and Pa-231 taken from sublimed samples.

An alpha spectrum showing Pa-231 and its daughters is shown in Figure 18. The Pa-231 used had been stored as Pa_2O_5 for several years, and most of the alpha-active daughters can be readily detected in the spectrum.

2.11 Effective Resonance Activation Integral of Ta-181 (R. P. Schuman)

Plans are being made to irradiate tantalum metal in order to produce 115-day Ta-182 for total cross section measurements with the MTR chopper. Since 115-day Ta-182 has a very high thermal capture cross section, the amount of Ta-182 that can be produced by thermal neutron capture is seriously limited by burnout. Therefore, it may be possible to produce a higher concentration of Ta-182 by irradiating the tantalum metal inside a cadmium shield.

Ta-181, like Au-197, has strong neutron capture resonances in the epithermal energy region, and therefore the effective resonance integral will depend strongly on the thickness of the samples. In order to see the effect of self-shielding on the resonance activation of tantalum metal, six rectangular pieces of tantalum foil and two gold-aluminum flux monitors were irradiated inside a 0.8-mm-thick cadmium shield in the VG-7 facility of the MTR. The activation of the tantalum was determined by absolute gamma counting. The number of counts in the broad peak due to the 1.122-, 1.189-, 1.222-, and 1.231-Mev gamma rays (plus other very low intensity gamma rays) was determined, and from it the tantalum activity was determined assuming that 100% of the Ta-182 decays proceed through these gammas, and that the conversion of these gamma rays is insignificant [32]. The Au-198 in the gold-

Fig. 18 Alpha spectrum of sublimed Pa-231 showing daughter activities.

aluminum monitors was also determined assuming a resonance activation integral of 1558 barns [33] (including $1/v$ contribution from thermal) for Au-197.

The effective resonance activation integrals for the different thicknesses of tantalum are given in Table X. The specific surface area of each tantalum piece is the total surface area of the rectangular prism in square centimeters divided by the mass in grams. For the thinnest pieces, the specific surface area is two times the reciprocal of the thickness expressed in g/cm^2 ; for the thicker foils, the area of the sides is an appreciable fraction of the total area. The effective resonance activation integral as a function of $\sqrt{S/M}$ is shown in Figure 19.

2.12 Gold as a Long Term Monitor (J. R. Berreth)

Previous experiments with mercury-amalgamated foil have shown that it is possible to remove all of the mercury from the gold by heating [34]. The amount of mercury formed after complete decay of the Au-198 and Au-199 in an irradiated gold sample is an indication of the total neutron dosage received. The integrated neutron flux (nvt) received by gold in a long term irradiation can therefore be determined from the amount of mercury present.

TABLE X

EFFECTIVE RESONANCE INTEGRAL OF TANTALUM FOR VARIOUS THICKNESSES

Foil Thickness (mm)	Weight (mg)	Total Surface Area		Effective Resonance Integral (barns) (including 1/v)
		Weight (cm ² /g)	$\sqrt{S/M_1}$ (cm/g ^{1/2})	
0.0084	1.63	167	12.9	573
0.0124	2.26	104	10.2	517
0.0406	9.06	30.3	5.50	384
0.132	23.79	9.85	3.14	232
0.257	35.6	5.59	2.36	185
0.777	112.0	2.38	1.54	132

In order to test this procedure, a series of irradiations has been made in the MTR. One sample containing 82.6 mg of Au was irradiated for approximately two days, and one sample containing 33.4 mg of Au was irradiated for approximately four days in the VH-2 facility. A third sample containing 10.7 mg of 1-mil Au and 9.7 mg of 5-mil Au was irradiated for one cycle in the L-55 position of the MTR. A fourth sample containing 5.1 mg of 1-mil Au and 6.5 mg of 5-mil Au was irradiated for two cycles in L-55. The samples also contained manganese and/or cobalt flux monitors. These samples have now been out of the reactor for a sufficient length of time (> 30 days) so that essentially all the Au-198 and Au-199 have decayed and the amount of mercury formed can be determined.

Fig. 19 Effective resonance integral of tantalum.

The resonance flux the gold received is important and must be considered in this case because the mercury formed is due to both resonance and thermal neutrons. The resonance self-shielding of gold is considerable for 1- and 5-mil-thick foils. It was necessary therefore to determine an "effective" resonance integral for these thicknesses. To accomplish this a cadmium-shielded irradiation containing rectangular gold foils of various thicknesses was performed in the MTR facility, VG-7. Table XI lists the results of this irradiation. The 0.5% Au + Al foil is sufficiently thin so there is negligible self-shielding of resonance neutrons. A resonance integral of 1558 barns including the 1/v component was assumed for this foil [23]. The resonance integrals were determined by gamma ray analysis of the 412-keV gamma ray with a 256-channel analyzer. Figure 20 is a plot of effective resonance integral vs $\sqrt{S/M}$ in cm/gm^{1/2} of gold.

TABLE XI

EFFECTIVE RESONANCE INTEGRAL OF GOLD FOR VARIOUS THICKNESSES

Au Thickness (mm)	Mass Au (mg)	S/M (cm ² /gm)	$\sqrt{S/M_1}$ (cm/gm ^{1/2})	Effective Resonance Integral Including 1/v (barns)
0.1252	2.089	9.74	5.12	327
0.0277	1.042	37.4	6.12	540
0.0109	1.285	95.1	9.76	787
1% Au + Cu 40-mil wire	0.1536	521	22.8	1291

Fig. 20 Effective resonance integral of gold.

The results of this experiment will be used to calculate the effective resonance integrals of the gold foils in the long term irradiation, so that the resonance contribution to the mercury production can be determined (see Table XI).

3. INELASTIC SCATTERING OF SLOW NEUTRONS PROGRAM (R. M. Brugger)

3.1 Scattering of Slow Neutrons by Sodium (P. D. Randolph, R. E. West)

Data on the inelastic scattering of slow neutrons from sodium have been obtained at incident neutron beam energies of 0.025, 0.07, and 0.10 ev for sample temperatures of 23, 100, 110, and 150°C. These data have been converted to scattering cross section and to scattering law.

The data at 23°C do not show any strong indication of coherent diffraction effects, indicating that the microcrystals of the sample were randomly oriented. The data for the liquid runs, however, show strong diffraction effects. The scattering law has a sharp diffraction peak for energy transfers out to approximately 0.1 kT. Beyond 0.15 kT this peak disappears.

Only the 100°C data have been analyzed further. For this data the angular distribution and the total cross sections have been obtained. Also the two

lowest energy moments of the scattering law have been obtained for several values of the momentum transfer.

The angular distribution was obtained by integrating the measured partial differential cross sections over the scattered neutron wavelength. This has been compared with the neutron diffraction results of Gingrich and Heaton [35]. The position and amplitude of the principal maximum agree; however, the first minimum is not as pronounced in this data as it is in the results of Gingrich and Heaton.

Integration of the angular distribution over scattering angle was done to obtain the total scattering cross sections at 0.025 ev. The result agrees with the published value within 10%.

A check of the lowest energy moments of the scattering law has been made and compared with the so-called moment theorems [36]. These may be written

$$\begin{aligned} \text{zero}^{\text{th}} \text{ moment: } & \int_{-\infty}^{\infty} e^{-\beta/2} S(\alpha, \beta) d\beta = F(\alpha) \\ \text{1st moment: } & \int_{-\infty}^{\infty} e^{-\beta/2} \beta S(\alpha, \beta) d\beta = \alpha \end{aligned}$$

where $S(\alpha, \beta)$ is the scattering law, β is the energy transfer, and α is the square of the momentum transfer. The function, $F(\alpha)$, is known from diffraction experiments.

These integrals were done numerically for several values of α , using a best fit curve to the data for the scattering law, S . The results show that the experimental zeroth moment is in reasonable agreement with the theory and the first moment is a factor 2 to 3 higher than predicted. This disagreement, however, may not be entirely real. The reason is that the major contributions to the integral come from the larger values of the energy transfer, β . This is just the region where the scattering cross section becomes small, and small errors in making background correction have a large effect on the cross section. Such errors would have little effect on the zeroth moment since the major contribution to it comes from the smaller values of β where the cross section is high.

3.2 New Rotating Collimators (R. M. Brugger, D. E. Jackson)

Construction, balancing and installation of the new rotating collimators were completed. Compared to the old collimators, which are constructed of nickel and phenolic with a 2-in. chopping radius, the new collimators are made from steel and nylon with a 6-in. chopping radius. The housings for the new collimators were inserted in the beam near the first chopper in place of the old collimator housings. The four Ford motors which have been used so successfully to drive the rotors for the past four years were replaced with Elinco 6000-rpm, 8-pole hysteresis synchronous motors. The new crystal-controlled

oscillator replaced the temporary oscillator. After several days of adjustment, the velocity selector was put into operation at 6000 rpm with these new collimators.

Operating conditions are (a) the bearings and stator of the motors are at 30 to 40°C, (b) the voltages to each winding are 80 to 120 volts, (c) the currents to each leg are 3 to 6 amps, (d) the voltages to the two legs of a motor are not balanced, (e) adjustment of current and voltage are made for what appears to be minimum phase jitter, (f) the phase jitter between choppers A and D is about ± 2 μ sec, (g) the phase jitter between chopper A and collimators B or C is about ± 7 μ sec, and (h) after the motors have run for several days the choppers accelerate to speed in less than 1.5 min and the collimators in less than 8 min.

The phase jitter of the new collimators is not unsatisfactory. Measurements of the backgrounds caused by fast neutrons show that the new collimators have reduced these backgrounds by a factor of 10 to 20. Some shielding was removed and counters at angles from 90 to 140° were installed.

3.3 MTR Cold Neutron Facility (K. A. Strong)

At this facility, a pair of choppers has been installed and successfully phased when the choppers are driven by fractional horse power synchronous motors with power supplied by a motor generator source. A simple and accurate method of varying the phase is achieved with autotransformers. The phase holds to ± 3 μ sec over long periods. These experiments indicate that this relatively inexpensive chopper drive system is as satisfactory for certain neutron time-of-flight experiments as a system containing an oscillator and power amplifiers. A paper on this work entitled "Phasing Two Choppers from a Motor Generator Source" has been completed and was submitted for patent clearance prior to submission for publication.

Two fission chambers were built for use as beam monitors, and one was installed at the cold neutron facility and tested. A single-channel analyzer for use with the beam monitor has been designed and is 90% complete. The signal to background in the scattering detectors was studied using vanadium as a sample. Two 30-in. radius-of-curvature chopper inserts are being built and will replace the present 42-in. radius-of-curvature inserts in the choppers. These 30-in. radius-of-curvature inserts will substantially increase the neutron intensity. A motor and gear box to be used as a beam door drive has been received. A method of improving the resolution of the instrument by spinning choppers at higher speed in evacuated housings is in the planning stages. Most of the material for such a changeover is on hand.

4. THEORETICAL STUDIES (H. L. McMurry)

4.1 Techniques for Calculating Partial Differential Scattering Cross Sections (H. L. McMurry)

In the last quarterly report [37] the need for methods of calculating partial differential scattering cross sections of low energy neutrons was discussed

and results obtained from two approximate techniques were outlined. The study of computational techniques has been carried farther and two papers which describe and evaluate five methods have been prepared. All of these methods are approximations to the rigorous formalism of Zemach and Glauber [38] and are applied to the specific case of scattering by gases composed of semi-rigid molecules. However, one of them can be adapted to treating simple models of liquids and amorphous solids and appears to have promise as a practical computational tool. It is being applied to calculate the scattering by water.

The five methods were evaluated by comparing calculations on methane and propane with experimental results obtained from the MTR slow neutron velocity selector [39, 40] and by comparing the most approximate methods with the rigorous ones by means of calculations on a hypothetical oxygen-hydrogen molecule with several values assumed for the oxygen-hydrogen vibration energy. The methods and chief results are presented here in order of decreasing rigor (and increasing range of practical applicability).

Method 1

This method is quite rigorous and treats rotations exactly by quantum mechanics. It is limited to neutron energies and thermal energies kT well below the characteristic energies of the molecular vibrations, and is of such complexity that practical calculations are limited to highly symmetrical molecules. The only application has been to methane where it gives excellent agreement with data from the MTR slow neutron velocity selector [41]. In particular, this degree of rigor is necessary to explain the scattering of low energy neutrons (0.025 eV and 0.015 eV) scattered at angles less than 25° .

Method 2

This method treats rotations classically, includes the contributions arising from vibrational excitation and de-excitation, and averages over molecular orientations exactly by numerical integration. Vibrations are assumed to be harmonic and rotation-vibration coupling is neglected. Within these limitations the method is exact. The classical treatment of rotations simplifies it so that its range of application is wider than method 1. However, it is still a cumbersome computational procedure for molecules of complex structure and is useful as a means for evaluating more approximate, but more easily used, techniques. It has been evaluated by calculations on CH_4 . In the calculations on the hypothetical oxygen-hydrogen molecule it is the standard for evaluating more approximate techniques.

The calculations on CH_4 agree well with the experimental results, except for scattering of low energy neutrons at forward angles where the classical assumption of continuous rotational energy levels is not valid.

Method 3

This method is like method 2 except that averages over orientation are approximated in the manner of Krieger and Nelkin [42] by inserting average values of functions of the Eulerian angles wherever they appear. This simplification greatly increases the range of practical applicability.

The method gives results for CH_4 almost as good as method 2. However, it becomes cumbersome when the neutron energy is much larger than the

characteristic vibrational energies of a molecule, because the number of ways open to vibrational excitation by neutron impact then become very large and each of these requires a separate computation. Furthermore, calculations on the hypothetical oxygen-hydrogen molecules show that when the characteristic vibrational energy is large (>0.2 eV), but much smaller than the incident neutron energy, the Krieger-Nelkin averaging gives poor results. However, the method is very good for certain ranges of neutron energies and vibrational energies and should be a practical computational tool.

Method 4

This method, called the short collision time (SCT) method because of the assumptions in the derivation of the equations, treats vibrations with characteristic energies much lower than the neutron energy in an approximate way [43]. The partial differential scattering cross section is expressed as a series. The first three or four terms are readily expressed but to go beyond this introduces very complicated expressions which cannot be used in a practical computational procedure. This severely limits what can be done using it. The SCT method was compared with the more rigorous method 2 in calculations on the hypothetical oxygen-hydrogen molecule. It gave fair results at low neutron energies (0.01 eV), but failed badly at higher energies (0.2 eV and above). It is not a practical computational method in its present form.

Method 5

This method treats vibrational motions classically when the vibrational energy is sufficiently below the neutron energy. It gives simple equations and greatly extends the range of practical calculations. Most of the evaluation has been by comparison against method 2 by calculations on the oxygen-hydrogen molecule, although some results have been obtained for CH_4 and propane. The results are very good provided the incident neutron energy exceeds the characteristic vibrational energy by a sufficient margin. The results indicate that the criteria in Table XII may serve for deciding when to treat a vibration classically, and when the Krieger-Nelkin (K-N) method should be used.

Method 5 has the correct asymptotic form in that if the neutron energy is much larger than all the characteristic vibrational energies so that a classical treatment of all vibrations is justified, the equation for the scattering reduces to that for unbound particles.

The criteria in Table XII greatly reduce the number of terms needed for calculating the scattering by a molecule and open the way to making practical calculations on complicated systems. If the behavior of a molecule in the liquid phase is described by assuming it to be free as far as translational motions go, but to be restricted to making torsional oscillations instead of rotations, the scattering from the liquid phase can be calculated by method 5 using the criteria in Table XII.

4.2 Influence of Interference Scattering on the Scattering of Slow Neutrons by Methane (G. W. Griffing)

Studies in this laboratory have shown that a correct quantum treatment of the rotational motion of methane is required in order to explain the scattering of low energy neutrons at forward angles [44]. The computational procedures have been improved so that cases involving large momentum transfers can be calculated, and the contributions arising from coherent scattering by atom

TABLE XII

CRITERIA FOR TREATING VIBRATIONS CLASSICALLY
AND BY THE KRIEGER-NELKIN METHOD

$\beta_\tau = E_o/kT(a)$	$\beta_o = E_o/kT$	Method
$0 < \beta_\tau \leq 1$	$0 < \beta_o < \infty$	C. A. ^(b)
$1 < \beta_\tau \leq 2$	$0 < \beta_o \leq \beta_\tau$	K-N ^(c)
	$\beta_\tau < \beta_o < \infty$	C. A.
$2 < \beta_\tau \leq 4$	$0 < \beta_o \leq 2\beta_\tau$	K-N
	$2\beta_\tau < \beta_o < \infty$	C. A.
$4 < \beta_\tau \leq 8$	$0 < \beta_o \leq 3\beta_\tau$	K-N
	$3\beta_\tau < \beta_o < \infty$	C. A.
$8 < \beta_\tau < 20$	$0 < \beta_o < 4\beta_\tau$	K-N
	$4\beta_\tau < \beta_o < \infty$	C. A.

(a) β_τ = energy of the characteristic vibration

E_o = energy of incident neutron

(b) C. A. denotes that a classical treatment of the vibration is permissible.

(c) K-N denotes that the Krieger-Nelkin method should be used.

pairs are included. Now the full range of the experimental results can be examined.

The most striking new result is that the interference scattering has an observable effect. It is revealed most clearly in plots of the Egelstaff [45] scattering function $S(\kappa^2, \beta)$. This function arises by expressing the partial differential scattering cross section as a product of factors - $\epsilon/\underline{E}b\tau$

in the form

$$\sigma(E_o, \epsilon, \mu) = \left(1 + \frac{\epsilon}{E_o}\right)^{1/2} e^{-\beta/2} S(\kappa^2, \beta)$$

where σ is the partial differential scattering cross section for scattering of a neutron with incident energy, E_o , through an angle, $\theta = \cos^{-1}\mu$, with an

energy change, ϵ . The $\beta = \epsilon/kT$ and $\underline{\kappa}$ is the momentum change of the neutron. Except for inclusion of a factor, $\frac{1}{kT}$, the function, S , is the Egelstaff scattering function. Convenient units for σ and S are barns/ev steradian molecule.

Figure 21 shows plots of $S(\kappa^2, \beta)$ vs $(\kappa b)^2$ for methane. Several values of $\epsilon = \beta kT$ are used and the experimental data are included for the case $\epsilon = 0$. A striking characteristic of the curves in Figure 21 is the inflections which are most noticeable for the curves with small ϵ . These have been shown to arise from the interference contributions which are negative in some $(\kappa b)^2$ ranges, positive in others and unimportant elsewhere. From curves like those in Figure 21 it is easy to tell at what neutron energies and scattering angles the interference effects are largest. By comparing with calculations which omit the interference effect, the magnitudes are easily obtained. In methane they can contribute up to 10% of the total.

The experimental data for the case $\epsilon = 0$ are consistent with the presence of a dip in the curve between $(\kappa b)^2$ values of 5 and 15, and a hump in the range 15 to 30. Examination of the published data for propane [40] indicates that interference contributions are seen in these experiments also.

Fig. 21 $S(\kappa^2, \epsilon)$ shown as a function of $(\kappa b_p)^2$ for selected energy transfers. The experimental points are for $\epsilon = 0.0$. The temperature of the gas is 21°C.

5. DECAY SCHEME STUDIES (C. W. Reich, R. L. Heath)

5.1 Current Status of Conversion-Electron Spectroscopy at the MTR-ETR Site (R. G. Helmer)

The decay schemes group has had for several years a set of four 180° uniform-field permanent-magnet spectrographs for the detection of conversion electrons from radioactive sources. These instruments have been used primarily to obtain the energies of internal-conversion electrons, and thereby, the energies of the associated gamma rays. The high resolution of this type of spectrograph often enables one to obtain gamma ray energies with an accuracy about one order of magnitude better than one can obtain by scintillation spectrometry. The higher resolution also may allow one to distinguish individual transitions whose photopeaks form one complex peak in the scintillation spectrum. Finally, the relative electron intensities give information concerning the multipolarity of the gamma ray transitions. By use of photographic detection one is able to record data over a large range of energies with no correction for source decay.

The features of the spectrographs on which the electron measurements rely are shown in Figure 22. Here w is the source width, a is the slit width, s is the source position, l is the line position, and x and y represent the dimensions of the camera. The camera is placed in a uniform magnetic field which is perpendicular to the plane of the paper. In this magnetic field the electron trajectory is a helix whose axis is parallel to the field lines. That is, the projection of the orbit in the plane of Figure 22 is a circle. Although the paths of monoenergetic electrons diverge as they pass through the slit, they converge again after traversing an angle of 180° (See reference 46 for a detailed description of this type of spectrometer).

Fig. 22 Camera for conversion-electron spectrograph.

Fig. 23 Calculated conversion-line intensity distribution for a source of infinitesimal height and width, w . Here $\phi \approx \tan \phi = \frac{a}{2x}$.

The profile of the calculated line shape for a source of width, w , and infinitesimal length (in the direction of the field) is given in reference 46 and shown in Figure 23. Since the high-energy edge of this line is quite sharp, its position is usually used as the line position. This edge is produced by electrons leaving the edge of the source nearer the photographic plate and traveling in a plane perpendicular to the magnetic field lines. Since they traverse an angle of 180° , the distance between this edge of the line and the nearer edge of the source is the diameter of a circle. If ρ is the radius of the circle, then from Figure 22

$$(2\rho)^2 = x^2 + (s + y + l)^2, \text{ or} \quad (1)$$

$$\rho = \frac{1}{2} \sqrt{x^2 + (s + y + l)^2}$$

Then, if B is the magnetic field strength in gauss and ρ is in centimeters,

$$\text{electron momentum} = B\rho \text{ gauss-cm}$$

$$\text{electron energy} = 510.98 \left[\sqrt{\left(\frac{B\rho}{1704.45} \right)^2 + 1} - 1 \right] \text{ kev}$$

The source position, s , must be measured for each source and the l for each line. B , x , and y are, in principle, constant and can be determined once for any spectrograph. They provide the calibration.

The four existing spectrographs have magnetic fields of 54, 136, 213, and 424 gauss and they span the energy ranges 2 to 95, 12 to 450, 22 to 850, and 50 to 1800 kev, respectively.

5.11 Energy Calibration - Computer Program. A computer program has been written [47] for the purpose of calculating the "best" values of the calibration parameters B, x, and y. For use with this program a series of exposures is made with sources containing lines whose energies (or momenta) are known quite accurately (to five parts in 10^4 or better) from independent measurements. A set of up to 50 lines is chosen from these data. From the corresponding s_i and ℓ_i (averaged over several measurements on each of several plates) and the known energies, the calibration parameters are calculated as follows:

Let

p_i = momentum of the i th line as calculated from the known energy,

and $\bar{p}_i(\ell_i, s_i, B, x, y)$ = momentum calculated from the parameters listed.

The program calculates the values of B, x, and y which minimize the function

$$\phi = \sum_{i=1}^{n \leq 50} (p_i^2 - \bar{p}_i^2)^2 \quad (2)$$

Each point is weighted equally because of the difficulty in choosing a more correct weighting function*. One effect of using equal weighting should be noted. On the average, one expects the difference, $p_i^2 - \bar{p}_i^2$, to increase proportionally to p_i^2 . So a line with a larger p_i will generally make a larger contribution to the function, ϕ . Thus, if the calibration lines were uniformly distributed along the plate, the high-energy region would dominate the fit. However, in practice the density of lines is usually much greater at the low-energy end of the plate. This distribution tends to make the contribution to ϕ from equal energy (or momentum) intervals more uniform.

5.12 Energy Calibration - Temperature Dependence of Magnetic Field. By May 1961, the four spectrographs had been calibrated using data for Ta-183, Bi-205, and Bi-206. (The Bi-205 and Bi-206 data were accumulated before July 1958, while the Ta-183 data were taken in February 1960. The Ta-183 data were used only in the two lower field magnets.) In 1961 several sources of Ir-192 and W-187 were run to check these calibrations. The energies of the gamma rays computed using the previous calibration were in serious disagreement with the values reported from measurements with a bent-crystal spectrometer [48, 49].

After several other possible sources of error were checked and ruled out, it was decided to measure the dependence of the line positions on the temperature of the whole spectrometer. Three plates were exposed using the same source

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* In the usual least-squares fitting problem one minimizes $\sum_i w_i (y_i - \bar{y}_i)^2$ where $w_i = \frac{1}{\sigma_{y_i}^2}$ and $\sigma_{y_i}^2$ = variance associated with y_i . In the justification of the least-squares method it is assumed that $\bar{y}_i$ has a zero variance (ie, it depends only on variables that are known exactly). This is not true here since $\bar{p}_i$ depends on the ℓ_i and s_i which are measured quantities. In fact, in the problem $\text{var } \bar{p}_i$ may be of the same order of magnitude as $\text{var } p_i$.

(μ constant except for negligible thermal expansion) with the spectrograph (136 gauss) temperature at 20, 24-1/2, and 34-1/2°C. A large shift in line position was observed. Analysis of the results indicates that all the data are consistent with the interpretation that the shifts are due to a change in the magnetic field with temperature, where

$$\frac{\Delta B}{B} = (2.0 \pm 0.5) \times 10^{-4} \Delta T \quad (3)$$

where ΔT is in °C. This value is much larger than one might expect from the thermal expansion coefficients of the materials in the spectrographs. (For example, the volume expansion coefficient of iron is about $3 \times 10^{-5}/^\circ\text{C}$).

The discrepancy between the new and old calibration data can be resolved if one assumes a decrease in the temperature of about 4°C from 1958 to 1961. (There are no temperature measurements to support this argument; however, the removal from the room of certain electronic equipment probably has lowered the temperature.) Using this value of ΔT , the results of the calibration of one magnet (216 gauss) are shown in Figure 24. The difference, ΔE , between "known" energy, E , and the calculated energy, $\bar{E}(B, x, y)$, is plotted against the energy. (The slight systematic difference between the B_i and W data may be due to any of several causes including the small number of readings on the W plates.) The solid lines indicate a deviation of one part in 10^3 . From this plot it would appear that if the assumed temperature decrease is correct, conversion lines can be measured with an accuracy of about one part in 10^3 .

The calibration program makes the assumption that the magnetic field is uniform. The fact that these data points scatter quite randomly around $\Delta E = 0$ over the entire range of the plate indicates that this assumption is justified (to an accuracy of about one part in 10^3).

Fig. 24 Data from calibration of 316-gauss spectrograph.

5.13 Relative Internal-Conversion-Line Intensities. In many cases it is desirable to know the relative intensities of the internal-conversion-electron lines (ie, the relative number of electrons emitted by the source). This information can be obtained from microphotometer traces of a photographic plate in the following manner:

The microphotometer trace consists of a plot of the intensity, L , of light transmitted through the emulsion as a function of the distance along the plate. The density, D , of the emulsion is defined as

$$D \equiv \log_{10} \frac{L_0}{L} \quad (4)$$

where L_0 is the light intensity through an unexposed portion of the plate.

The exposure, E , that a portion of a plate receives can be defined as the number of electrons per cm^2 which strike the plate. Since this quantity cannot be measured easily, it is often necessary to express the exposure in terms of a quantity (such as "hours" for a source with a constant decay rate) which is proportional to the electrons per cm^2 . For monoenergetic electrons the density will be some function of the exposure. Once this functional dependence, $E(D)$, is known, the microphotometer trace can be converted to a plot of exposure vs position.

The total exposure from the conversion electrons is given by the area, A , under the line profile. That is,

$$A = \int E \, d\ell - \int E_B \, d\ell = \int (E - E_B) \, d\ell$$

where

ℓ is the distance along the plate

$E = E(\ell)$ is the total exposure at the position ℓ

$E_B = E_B(\ell)$ is the exposure due to the spectral background

To obtain the relative number of electrons emitted from the source, one must correct the total exposure, or area, A , for (a) the sensitivity ϵ of the emulsion and (b) the variation in solid angle with position along the plate. The sensitivity (ie, the density per unit exposure as a function of the electron energy) must be measured, but the solid angle can be calculated. The change in solid angle is due to the motion of the electrons in the direction of the field. The fraction of the electrons emitted that are lost will increase proportionally to the distance they travel before striking the plate. This distance is $\pi\rho$. Therefore, the solid angle is $\frac{\text{constant}}{\rho}$.

Finally, the relative intensity, I , of any internal-conversion line is

$$I = \frac{A\rho}{\epsilon} \quad (5)$$

The area, A , is often very difficult to obtain due to the problem of estimating the background exposure under the line, especially the low-energy tail thereof. To circumvent this problem, Slätis [50] has reported a method in which he approximates the profile by a rectangle. The final expression for A depends on the explicit form of $E(D)$. If

$$E = \alpha D \quad (6)$$

where α is a constant,* one obtains

$$A = w \log \frac{L_B}{L_M} \quad (7)$$

---

* Slätis uses $D = C \log (\alpha E \pm 1)$ for a different type of photographic emulsion.

where

$$w = \text{line width at } L = L_{1/2} = \sqrt{L_B L_M}$$

L_B = light intensity corresponding to the background under the peak

L_M = light intensity at the peak of the profile

Experimental Data. As noted above there are two relationships that must be determined before relative internal-conversion-line intensities can be calculated. They are the density vs exposure, and the emulsion sensitivity as a function of energy. The former can be measured quite simply. A source of Pm-147, which has only a continuous beta-ray spectrum, was used for the data below. The plates were exposed to a narrow band of energies by covering the plate with an aluminum plate having a slit in it. Seven exposures were made on each plate by displacing the plate in the plate holder. Since all the exposures were at the same value of ρ and all on the same plate (same developing, washing, and fixing times) no known systematic errors were introduced. The results of these preliminary measurements are shown in Figure 25. The difference in slope for the two measurements with No-Screen plates may be due to different developing times. These data indicate that the relationship between D and E can be approximated quite well by a straight line for $D < 1$ for No-Screen plates and $D < 1.5$ for type A plates.

The measurement of emulsion sensitivity is not easy. A direct measurement requires a source of electrons of variable energy and known intensity. However, such measurements have been carried out for No-Screen plates by other experiments. Figure 26 shows such a curve [51]. The author does not know of any corresponding data for type A plates.

Two measurements were made in order to illustrate this method of measuring intensities. The first trace was of a type A plate exposed with Ir-192. It was found that the widths of single lines vary randomly between 2.2 and 2.7 mm (on the original chart); therefore all the widths were considered the same in Equation (7). Since the emulsion sensitivity of the plate was not known,

Fig. 25 Density vs exposure relationships for photographic plates used.

it was taken to be the same as the No-Screen type. (This will not be true, but it was hoped that the gross shape would be the same between 100 and 400 kev.) Also the three most intense

lines have $D = \frac{L_o}{L} > 1.5$, so that

Equation (6), and therefore Equation (7), may not be correct. The calculations and a comparison with the values of Baggerly et al [48] are given in Table XIII.

The good agreement over the whole range of energies is probably fortuitous. (Only the values at 284 kev differ by more than 16%). However, over small energy ranges where ϵ is about the same for all the lines, the agreement indicates the analysis is at least approximately correct.

The second example consists of traces of two No-Screen plates exposed with a source of Bi-205 and Bi-206. A photograph of the plate and a portion of the trace are shown in Figure 27. In this case the principal uncertainties are [46]

Fig. 26 Emulsion sensitivity for No-Screen type plates (From reference 55).

TABLE XIII

RELATIVE INTENSITIES FOR Ir-192

Electron Line Energy (kev)	Line Interpretation	$A = \log \frac{I_B}{I_M}$	ρ_4	Emulsion Sensitivity (ϵ^+)(a)	Relative Intensity(b)	Reported Relative Intensity(c)
132	K-206	0.754	12.2	0.90	0.6	0.6
217	K-296	1.36	16.2	0.50	2.6	2.3
230	K-308	1.20	16.7	0.48	2.5	2.4
238	K-316	2.18	17.1	0.46	4.8	5.4
284	L _{III} -296	0.117	18.9	0.42	0.32	0.22
296	L _{III} -308	0.093	19.4	0.40	0.27	0.28
304	L _{III} -316	0.246	19.8	0.40	0.7	0.6
389	K-468	0.334	23.1	0.35	1.3	1.4

(a) Values are taken from Fig. 26.

(b) Even if the assumptions concerning ϵ and D vs E are correct, the uncertainties are expected to be about 20%.

(c) These values are calculated from the gamma ray intensities and conversion coefficients[48]. The corresponding errors are not known.

Fig. 27 Photograph of plate Bi-205, -206 plate III-19 and a copy of a portion of the microphotometer trace of the same plate. The numbers associated with the lines are the same as those in Table XV.

the deviations from Equation (6) for the intense lines [47], the possible fading of the plate in the four years since its exposure, and applicability of the emulsion sensitivity curve in Figure 26. The resulting data are shown in Table XIV.

Considering the estimated average uncertainties of 15 to 25% in each set of data, the agreement seems satisfactory. This may indicate the absence of any large systematic errors in the analysis.

No attempt has been made to analyze the random errors. This is because there is a need for further evidence that the sensitivity, solid angle, and density-exposure relationships are correct before any such analysis is meaningful.

5.14 Method of Line Position Measurements. All previous line position measurements have been made with a linear comparator. The lines are viewed directly and a cross hair on the comparator is moved along until it is lined up with the high-energy edge of the line. The line positions are all measured with respect to the image of one of the notches in the plate holder (see Figure 27). The procedure has been to have three or more persons make readings on each plate. These readings are then averaged. The following are a few of the systematic errors that may appear:

TABLE XIV
RELATIVE INTENSITIES FOR Bi

Isotope	Line Energy (kev)	Relative Line Intensities			Reported Value ^(a)
		Plate III-19	Plate II-10	Average	
Bi-205	615	92		92	100
Bi-205	492	21		21	21
Bi-205	483	76		76	74
Bi-205	462	9		9	6
Bi-205	405	10	14	12	11
Bi-205	262	31	40	35	28
Bi-205	196	149(b)	127(b)	138(b)	160
Bi-205	194	45	38	41	41
Bi-205	172	112(b)	90(b)	101(b)	110
Bi-205	148	7	5.5(b)	7	8
Bi-206	449	2.4	2.4	2.4	2.1
Bi-206	428	1.9	2.2	2.0	1.9
Bi-206	409	1.4	1.6	1.5	1.4
Bi-206	310	1.8	1.8	1.8	1.8
Bi-206	298	—	0.14	0.14	0.13
Bi-206	255	6.0(b)	4.0(b)	5.0(b)	6.6
Bi-206	224	—	0.10	0.10	0.14
Bi-206	175	2.9(b)	1.3(b)	2.1(b)	2.1
Bi-206	146	0.10	0.10	0.10	0.24
Bi-206	115	0.06	—	0.06	0.02

(a) Values for Bi-205 are from ref. 52 and for Bi-206 from ref. 53.
Both sets have quoted errors of 15%.

(b) These lines have $D = \frac{L_o}{L} > 1$. Therefore, the corresponding intensities are expected to be low.

(1) Differences between persons in method of estimating the "edge" of the lines.

(2) Differences between persons in estimating the "edge" of a notch.

(3) For one person, variation in estimating edges of faint and dark lines.

Because of the above errors and also because of the large amount of tedious labor that must go into plate reading, it would be desirable to find an alternate method of measuring the line positions. A test run indicates that it may be feasible to use the microphotometer traces mentioned before. In this case the systematic errors are replaced by the problems of the dimensional stability of the chart paper, the uniformity of the chart drive speed, and the "noise" due to the finite size of the emulsion grain. However, it is hoped that it may be possible to make these errors at least as small as the direct-reading errors by measuring the line positions relative to the notches (which are two cm apart) on the plate. As a test case the chart shown in Figure 27 was used. In Table XV, the resulting line positions (in units of the centimeters on the plate) are compared with the corresponding values from a set of three linear comparator measurements. In the conversion of distances measured on the chart to those measured on the plate, there are two independent parameters. The first is the zero position and the second is the chart-distance to plate-distance conversion. The latter was computed from the distances between the 4th and 10th notches. These distances were 11.997 cm on the plate and 455.65 cm on the chart. On the scale in Table XV they occur at 18.223 and 30.220 cm, respectively. The two scales were then matched at the 6th notch (22.227 cm).

A more accurate method of correcting for long-term variations in the chart drive system would be to calculate the chart-distance to plate-distance conversion factor between each pair (or set of 3 or 4) of notches. Such an analysis should eliminate the systematic negative differences for lines 1 to 10 (Table XV). The systematic trend to positive differences for lines

TABLE XV
COMPARISON OF LINE POSITION MEASUREMENTS ON B1-205, -206 PLATE III-19

Line	Chart (cm)	Comparator (cm)	Difference (cm)
1	17.792	17.781	-0.011
2	17.911	17.908	-0.003
3	18.612	18.612	0
4	18.782	18.780	-0.002
5	18.978	18.978	0
6	19.804	19.807	+0.003
7	20.497	20.488	-0.009
8	20.583	20.579	-0.004
9	21.589	21.577	-0.012
10	21.648	21.639	-0.009
11	21.797	21.792	-0.005
12	21.906	21.904	-0.002
13	22.164	22.165	+0.001
14	22.304	22.307	+0.003
15	22.824	22.819	-0.005
16	22.912	22.908	-0.004
17	24.083	24.091	+0.008
18	25.531	25.531	0
19	25.579	25.576	-0.003
20	25.786	25.788	+0.002
21	25.992	25.993	+0.001
22	26.071	26.074	+0.003
23	26.575	26.575	0
24	27.799	27.790	-0.009
25	28.505	28.502	-0.003
26	28.747	28.745	-0.002
27	28.981	28.985	+0.004
28	30.636	30.638	+0.002
29	31.532	31.528	-0.004
30	31.661	31.662	+0.001
31	32.204	32.212	+0.008
32	32.381	32.379	-0.002
33	33.177	33.173	-0.004
34	33.633	33.623	-0.010
35	34.392	34.393	+0.001
36	34.504	34.525	+0.021
37	34.725	34.734	+0.009
38	35.056	35.065	+0.009
39	35.796	35.810	+0.014
40	39.134	39.155	+0.021

36 to 40 is due to extrapolation of this conversion factor beyond the last visible notch.

The accuracy of this method can be estimated crudely by computing the rms deviation from the mean of the comparator readings. Using only the lines between the 4th and 10th notches the rms deviation is 0.005 cm. This is about the same as the average rms deviation from the mean for any one set of readings on the comparator. Therefore, it appears that this method gives accuracy comparable with the comparator in those regions in which the notches are visible.

5.15 Source Making. The ideal source for this type of spectrometer is a line of active material about 2 cm long, and of infinitesimal width and thickness. In practice, the width and thickness are both governed by the specific activity of the source material.

Two satisfactory methods of source making are electroplating and vacuum evaporation. The former has not been used here extensively and will not be discussed. At present there are no facilities for evaporating radioactive materials. For evaporating stable materials, the apparatus used is shown in Figure 28. (This arrangement was discussed previously [47].) After reducing the pressure in the system to about 0.1 micron, the graphite filament is heated to the desired temperature by running an electric current through it. The maximum current the system allows is about 200 amps at 10 volts. For the filaments used currently, tungsten oxide (melting point of 1450°C at atmospheric pressure) can be evaporated at about 150 amps and 6 volts. Therefore, evaporated deposits of most elements can be obtained if the proper compound is chosen.

The material is collected on a blank (1/16 x 3/16 x 1-1/8 in.) which is usually made of high-purity graphite. The source region is defined on the sides by two slit jaws and on the ends by two shielding clips. The slit jaws can be

Fig. 28 Apparatus for vacuum evaporation of sources.

moved to give source widths of 0.0 to 0.1 in. although the range actually used is from 0.002 to 0.015 in.

For a given slit width and filament-to-blank distance, the efficiency of the evaporation process will depend primarily on the shape of the filament. Three filament shapes have been used recently. The cross sections are shown in Figure 28. The second and third models are preferred because they give much more uniform heating of the source material. For an evaporation of tellurium metal from the second model, a yield of about 1% was obtained, or about 40 micrograms (by weighing) from 4.3 milligrams of material. In this case the slit was 0.012 in. wide by 0.5 in. long and the blank was $7/32$ in. from the top of the filament. One attempt has been made to measure the yield from the third model using activation analysis. The results placed an upper limit of 1% on the yield for a slit 0.016 in. by 0.5 in. at $1/8$ in.

5.16 External Conversion. Experiments were undertaken to ascertain whether it is feasible to do external-conversion electron studies in these spectrographs. Since such experiments generally require sources whose decay rates are at least two orders of magnitude higher than those used in internal-conversion-electron studies, it was not obvious that one could satisfactorily shield the photographic plate from the direct gamma ray radiation.

The source arrangement used in these experiments is shown in Figure 29. The aluminum is placed over the source to stop the electrons emitted from the source. Some of the gamma rays leaving the source will be absorbed in the gold foil and will produce photoelectrons. These photoelectrons are monoenergetic and will produce conversion lines on the plate. After correcting for the photoelectric cross section, the intensities of these lines are proportional to the intensities of the original gamma rays. Also the ratio of the intensities of the internal- and external-conversion lines give the internal-conversion coefficients.

Using a $1\frac{1}{2}$ -millicurie source of Ir-192, several external-conversion lines were observed above the background caused by Compton scattering in the material around the source and by direct gamma ray radiation. The quality of the data could be improved by using a uranium or thorium converter since

for a higher Z the photoelectric cross section is larger relative to the Compton scattering cross section. Also for energies below about 500 kev the results will be better for a thinner foil (foils of 2 mg/cm² are commonly used).

Fig. 29 Source arrangement for external conversion.

5.2 Evidence for Direct Beta Feeding of the 1.57-Mev State of Mo-94 from the Decay of 6.6-minute Nb-94m (C. W. Reich and R. P. Schuman)

The decay scheme previously proposed [54, 55] for the two Nb-94 activities is shown in Figure 30.

It has been previously suspected [54] that the decay of 6.6-minute Nb-94m contains a weak beta

branch to the 1.57-Mev state in Mo-94. This was postulated because of the apparent "filling-in" of the valley and Compton edge of the spectrum of the 0.874-Mev gamma ray. The gamma ray spectrum from the decay of 6.6-minute Nb-94m is shown in Figure 31. In order to establish the existence of the 0.703-Mev gamma ray, a coincidence experiment was performed. The spectrum of gamma radiation in coincidence with the 0.874-Mev gamma ray was recorded using a 20-channel discriminator-type pulse-height analyzer of conventional design. Because of the short half-life of the sample and the low coincidence counting rates involved, no attempt to obtain the complete spectrum was made. The analyzer was set up to span the region between 560 and 940 kev. The Nb-94m sources were prepared by irradiation of samples of niobium metal for period of ~ five minutes in the VG-7 facility of the MTR. Four 400-second measurements of the coincidence counting rate from each of three sources were made. The composite spectrum obtained by combining these measurements is shown in Figure 32. The presence of the 0.874-Mev gamma ray in the spectrum is due to random coincidences. The presence of the 0.703-Mev gamma ray photopeak in the spectrum is clearly indicated. A gamma ray spectrum of one of the sources, taken several days after the coincidence experiment, gave no evidence of any 0.87- or 0.70-Mev gamma rays.

Fig. 30 Decay scheme for Nb-94 and Nb-94m.

Fig. 31 Gamma ray singles spectrum from 6.6-min Nb-94m.

Fig. 32 Spectrum of gamma rays in coincidence with the 0.874-Mev gamma ray.

Thus, the 0.703-Mev gamma ray in the coincidence spectrum cannot arise from a 20,000-year Nb-94 in the source.

The intensity of this gamma ray relative to the 0.874-Mev gamma ray was found from an analysis of the single spectrum of Figure 31 to be $\sim 1.3\%$. This result, however, is highly sensitive to details of the assumed shape of the 0.874-Mev gamma ray and must thus be considered as an order-of-magnitude estimate only.

III. INSTRUMENT DEVELOPMENT

1. DIGITAL SYSTEMS INSTRUMENTATION

1.1 Programmer Control for Nuclear Data ND-130 Analyzer (R. I. Little)

While the Nuclear Data ND-130 Analyzer is a versatile piece of equipment, it contains no features for restarting after prescribed time intervals and even in the direct analyze-readout-analyze-readout mode its tape perforator must run continually. This increases its preventative maintenance schedule to once each 200 hours. This programmer control overcomes both deficiencies. It employs a precise external clock for determining restart times and when the analyzer attempts to enter a readout mode the programmer starts the punch motor and places the analyzer in a hold status until the punch motor has reached operating speed.

The programmer (Figure 33) employs all solid state logic. When the START button is depressed, it sets the run binary coincident circuit to a kcps clock pulse. This in turn fires a monostable multivibrator which starts the analyzer and resets the timer. The next clock pulse starts counting. Three separate decades divide the clock to a one-second rate; the five succeeding decades present a visible display of time and feed the preset circuit. The preset circuit allows the operator to select time intervals in the sequence 1, 2, 4, 8, 10, 20, 40, 80, etc, up to 80,000 seconds. The analyzer run time must be adjusted appropriately to accomodate a readout sequence before restart.

When the ND-130 has finished the analyze mode (due to its own preset), it sets a punch binary circuit which starts the punch motor through a relay.

Fig. 33 Block diagram programmer control for ND-130 analyzer.

Also, it rapidly triggers a half-second, monostable multivibrator which inhibits all operation in the analyzer until termination of the half-second period. The programmer places the analyzer in the readout mode. After readout is complete, the analyzer returns to hold and, in doing so, resets the punch binary circuit, stopping the punch motor. The punch motor ac power has been routed externally to reduce transient effects.

The analyzer now sits idle until the programmer's timer reaches the preset count. At this time the preset circuit triggers the start-analyze monostable multivibrator which also resets the timer.

This procedure repeats itself until the operator interrupts the sequence with the STOP button.

1.2 Noise Suppression in the ARMF Regulating Rod Digital Readout (R. I. Little)

During the past quarter, the ARMF personnel replaced the shaft angle encoders on the shim and regulating rods. The new encoders contained improved strobe lamp electronics, but the one on the regulating rod was found to be load sensitive.

When the output amplifiers in the encoder on the regulating rod produced pulses, the trailing edge of these pulses falsely triggered other amplifiers. The effect was intermittent and somewhat position sensitive. The problem had arisen during initial construction of the ARMF with the original encoders and had been resolved by isolating the output amplifiers from the shielded drive lines with series 3 k Ω resistors at encoders. Any further load reduction would have degraded the outputs to the point of unreliability.

Three possible remedies presented themselves:

- (1) Remove the encoder and try to modify it to correct the problem. This meant loss of rod calibration and considerable reactor down time,
- (2) Isolate the encoder with active elements in such a way as to present a high impedance load with low impedance subsequent output,
- (3) Strobe for the output pulses. This seemed attractive because spurious pulses always appeared delayed in more time than valid pulses, as shown in Figure 34.

The solution employed both remedies (2) and (3). Figure 35 shows the basic circuit and Figure 34 shows the resultant waveforms. The entire assembly including its own power supply occupies $\sim 1/2$ ft³ on the bridge close to the encoder. Each encoder output, capacitively coupled in the encoder, drives a standard two-input "nor" element whose alternate input is grounded to establish proper bias. The input impedance of 33 k Ω substantially decreases encoder loading. A second "nor" element "ands" the output pulse, now positive going, with the positive interrogation pulse to yield a negative pulse with substantially the same characteristics as the original signals, but minus any spurious pulses. The interrogation pulse had to be adjusted to properly envelop the output pulses.

Fig. 34 Timing in shaft angle encoder noise suppressor.

Fig. 35 Typical noise suppression circuit.

The success of this small logic package demonstrates the advantages of maintaining a small stock of logic modules and associated hardware on hand, because from the time the problem was isolated, only nine hours passed before the package was installed and operating. As a result of its success, the package will be incorporated on both shim and regulating rod encoders in ARMF II presently under construction.

1.3 Gated Scaler and Data Recording System (N. J. Veach)

A gated scaler and paper tape punch control unit have been constructed for recording statistical data from neutron pulse counting systems as shown in Figure 36. The gated scaler counts pulses for a preset time, transfers the accumulated count into the punch control unit, resets, and starts the next count. The cycling is automatic.

The gate time is selectable by front-panel switch settings and can be varied from 0.001 to 9.999 seconds in 0.0001-second increments. The basic time source is a crystal-controlled 100-kc oscillator. A front-panel switch prevents reset when accumulating counts over longer gate times. Manual timing control is used for the longer periods. The scaler has a two-pulse resolution of five microseconds and accepts positive four-volt pulses. The punch control unit has an internal 10-decimal-digit memory which holds an accumulated count for punching on paper tape while the next count is being accumulated. The number of digits per word can be varied 1 to 10 digits by means of a front-panel rotary switch. Maximum punching rate is 60 decimal digits per second. An identification punch (EL punch) is added to the last digit of each word. The gated scaler and punch control unit control each other so that the faster of the two units waits for the slower unit. A Tally model 420 paper tape punch is used in this system.

Fig. 36 Block diagram of gated scaler and data recording system.

The code used on the paper is 1-2-4-8 as used on IBM tape-to-card converter 47. The scaler drives an 8-line output (1-2-4-8 with complement) where zero is represented by zero volts and 1 (or 2 or 4 or 8 or a combination) by -12 volts.

1.4 Magnetic Tape Pulse Recorder (N. J. Veach)

A system has been constructed for recording on 1/2-in. magnetic tape, three channels of neutron counts and a 100-kc timing reference channel. The NRZ technique is used in which the tape flux is saturated continuously by current, thereby changing the direction of flux. Playback at reduced speeds is used to drive gated scaler and paper tape punch, or other recorders. Oscillographic records of the playback have been made for visual inspection of the data. Additional application is variable-rate recording of BCD digital data in study of variable-velocity transport problems. A diagram of the recorder is shown in Figure 37.

Fig. 37 Magnetic tape pulse recorder.

1.5 Time Code Generator (N. J. Veach)

A laboratory model of a time code generator has been constructed for recording time signals on 16-millimeter motion picture film. Pulse trains of seven different pulse sizes and repetition rates (0.1 to 1000 cps) are generated starting at the time of an external switch closure. Two of the pulse trains are selected by a front-panel switch and mixed for driving a single neon bulb in a remotely located camera. The light pulses from the neon bulb appear on the film sprocket track as small exposed areas. Known starting time of the pulse train and known pulse repetition rates enable the determination of the elapsed time. Figure 38 is a diagram of the time code generator.

Fig. 38 Time code generator.

1.6 The Discrete Binomial Time Interval Distribution (N. Wilde)

The time interval distribution as derived from the Poisson distribution is a useful relation when dealing with random pulses. In some pulse systems, the time interval is restricted to multiples of a finite resolving time. Hence, it would be desirable to have a discrete time interval distribution describing this condition. A discrete distribution is derived from the binomial distribution that can be numerically evaluated from standard tables.

For Poisson-distributed pulses, the probability of x pulses in time, t , is

$$P_x(t) = \frac{(at)^x}{x!} e^{-at} \quad (1)$$

where a is the average pulse rate. The probability of no pulses ($x = 0$) in time, t , is

$$P_0(t) = e^{-at} \quad (2)$$

If the period, t , is to be a time interval between successive pulses, the interval must terminate with the next pulse. The probability of a pulse in dt at t is ($a dt$). The combined probability of no pulse in time, t , followed by a pulse in dt at t is the product of these two probabilities.

$$dP(t) = a e^{-at} dt \quad (3)$$

The probability of a time interval less than t is obtained by integrating Equation (3)

$$P(t) = \int_0^t dP(t) = (1 - e^{-at}) \quad (4)$$

Note that Equations (3) and (4) are continuous functions.

A discrete relation analogous to Equations (3) and (4) can be derived from the binomial distribution. The probability of x pulses in r time increments of length, τ , is

$$B_x(r) = \binom{r}{x} (a\tau)^x (1 - a\tau)^{r-x} \quad (5)$$

where $\binom{r}{x}$ is the binomial coefficient and $r = t/\tau$ is an integer. The probability of no pulses ($x = 0$) in r increments is

$$B_0(r) = (1 - a\tau)^r \quad (6)$$

The probability of a pulse in the $(r+1)$ increment is $a\tau$. The combined probability of no pulses in r increments followed by a pulse in the $(r+1)$ increment, ie, the probability of a time interval r increments long, is

$$B(r) = a\tau (1 - a\tau)^r \quad (7)$$

The probability of a time interval less than n increments long is obtained by summing Equation (7)

$$B(n) = \sum_{r=0}^n B(r) = 1 - (1 - a\tau)^{n+1} \quad (8)$$

Equations (7) and (8), the desired discrete relations, are analogous to Equations (3) and (4), respectively.

It is readily shown that Equation (4) is the limiting expression for Equation (8) just as the Poisson distribution can be considered the limiting distribution for the binomial distribution. Writing Equation (8) in terms of t and τ

$$B(n) = 1 - (1 - a\tau)^{\frac{t}{\tau} + 1} \quad (9)$$

If $a\tau \ll 1$, $(1 - a\tau)$ is approximately $e^{-a\tau}$ and

$$B(n) \sim 1 - e^{-a(t + \tau)} \quad (10)$$

Letting the increment τ approach zero and n approach infinity such that $t = n\tau$ is finite yields

$$B(n) \sim 1 - e^{-at}$$

$$B(n) \sim P(t) \quad (11)$$

Numerical values of $B(n)$ are given in a National Bureau of Standards publication [56]. For table use, $B(n)$ is rewritten in terms of the binomial expansion as

$$B(n) = \sum_{r=1}^{n+1} \binom{n+1}{r} (a\tau)^r (1 - a\tau)^{n+1-r} \quad (12)$$

This form is tabulated in the tables [56].

The preceding relations have proven useful in evaluating sampling rates for digital data systems handling random Poisson-distributed pulses.

2. SPECIAL ELECTRONIC INSTRUMENTATION

2.1 Overvoltage Protector for High Voltage Power Supplies (K. F. Smith)

When the pass tube in a high voltage power supply shorts out, the high voltage output of the supply will go to a voltage which is much higher than the normal operating voltage. In certain applications, this voltage excursion can cause severe damage to equipment which is being supplied from these high voltage power supplies.

A simple circuit has been developed which will offer protection to equipment when this type of failure occurs. A schematic of the protector is shown in Figure 39. The protector senses the overvoltage condition through the breakdown of the NE 2 (neon lamp) chain. The magnetic latching relay will then energize and disconnect the power supply from the ac supply voltage. The relay in the circuit energizes in approximately one to two msec after the overvoltage occurs. After the cause of the failure has been corrected, the protector may be reset by depressing the push-button switch.

2.2 Pulse Height Ratio to Time Converter for Fission Studies (J. B. Colson)

A circuit is being designed for use with the Crystal Spectrometer in the peak-to-valley fission fragment measurements. This circuit is required to provide two timing pulses whose separation time is proportional to the ratio $A/(A + B)$, where A and B are the amplitudes of two 1- μ sec, variable-height

input pulses. These timing pulses are supposed to occur only if pulses A and B are in coincidence and exceed five volts in amplitude.

These requirements have been accomplished, except for the degree of precision, by discharging a capacitor with two current sources. The capacitor voltage is then proportional to the height of pulses A and B, providing a ramp voltage whose slope is proportional to $A + B$. When the voltage decreases an amount proportional to pulse height A, an output pulse is generated. The amount of delay is proportional to $A/(A + B)$.

The block diagram of the system is shown in Figure 40. The pulses A and B are amplified through two A-8 linear amplifiers. The discriminator level is set at five volts and the pulse height output is fed into a coincidence analyzer which starts a timing circuit. The amplifier outputs are gated by the timing circuit and stretched to about 250 μ -sec. The stretched pulses are then used to set the current level of two constant-current sources which discharge a capacitor. The pulse A is inverted and compared to the capacitor ramp voltage giving an output stop pulse when the ramp reaches the level of

Fig. 39 Block diagram overvoltage protector for high voltage power supplies.

Fig. 40 Pulse height ratio to time converter system block diagram.

pulse A inverted. The output start pulse occurs at the beginning of the cycle. The difference in time between the start and stop pulses is then proportional to $A/(A + B)$. The complete circuit is reset by the timing circuit at the end of each cycle.

The dead time in the circuit is about 350μ sec which produces little counting error when operated at the expected rate of 100-counts per minute. The accuracy desired is about $\pm 0.5\%$ and the present accuracy obtained is about $\pm 1.5\%$. The limitation in accuracy is due to the nonlinearities in the constant-current generators. Additional work is proceeding to improve these generators.

2.3 Instrument for the Analog Display of Time Delay or Frequency Drift (T. J. Boland)

To facilitate measurement of a long-term stability in the phasing of the two-phased choppers in the MTR velocity selector it became desirable to derive an analog voltage proportional to phase drift for display on a strip chart recorder.

The instrument shown in block diagram form in Figure 41 is useful for many measurements in addition to the one for which it was designed, Stability of pulse delay lines as a function of time or environment, and the frequency stability of a secondary standard generator compared with a primary standard, are examples. In the velocity selector time phase, stability of one chopper with respect to the other was recorded to within one microsecond.

Basically the instrument consists of two identical channels, each made up of a blocking oscillator driving a flip-flop. Two flip-flops provide square waves to a phase comparator whose output is a voltage proportional to phase or time difference between a reference pulse and a delayed pulse. In the case where the zero crossing of two sine waves is compared, the sine waves are first amplified and clipped and then differentiated before going to the blocking oscillators.

The instrument will readily detect a shift of five parts in 10^5 corresponding to approximately two minutes of phase shift. Indications are that filtering of the phase comparator output to reduce high frequency noise will improve this sensitivity considerably, but would slow the response correspondingly.

Fig. 41 Pulse-delay to analog converter and frequency comparator.

2.4 Method for Obtaining a Rate Signal Without Using a Tachometer (H. R. Hansen)

For the remotely controlled milling machine it was necessary to obtain a tachometer signal to stabilize the servos. But because of physical limitations, a tachometer could not be mounted on the mill. Hence, in place of using a tachometer the back emf, which is proportional to the speed of the motor, was used for a tachometer signal.

To obtain a signal proportional to the back emf, $V_{\text{-emf}}$, of the motor but independent of the armature current, a bridge circuit was used. This bridge circuit is shown in Figure 42. When the bridge is balanced

$$\left(\frac{R_1}{R_2} = \frac{R_a}{R_3} \text{ and } C_1 = \frac{L_a}{R_1 R_3}\right)$$

the output from the bridge is independent of the armature current and proportional to the back emf. At low frequencies, the output equals

$$V_o = \frac{R_2}{R_1 + R_2} V_{\text{-emf}}$$

This assumes that R_1 and R_2 are chosen to be much larger than R_a and R_3 respectively.

Fig. 42 Bridge circuit for obtaining speed-sensitive voltage.

3. HEALTH PHYSICS INSTRUMENTATION

3.1 Alpha-Beta Air Monitor (J. B. Thompson and K. F. Smith)

An improved air monitoring system has been developed which will monitor the amount of alpha and beta activity in the air and give the ratio between the two with much greater accuracy than in commercially available instruments tested here.

The system uses a new end-window proportional counter [57] to detect the alpha and beta activity that is present on a filter. The filter is used to collect particulate matter in a constant air flow system. The alpha and beta pulses are amplified in a common amplifier to increase their amplitude to a level where separation can be made using a simple diode discriminator. The alpha pulses which exceed the discriminator threshold are amplified to a level where they will trigger a univibrator. The output of the univibrator feeds the alpha log count ratio channel and an anticoincidence circuit.

The "beta-plus-alpha" channel amplifies the beta and alpha pulses simultaneously to increase the amplitude of the smallest beta pulses to a level that will trigger a univibrator. The output of this univibrator is fed into the anti-coincidence circuit where the alpha pulses are subtracted from the "alpha-plus-beta" pulses. The beta pulses are then fed into the beta log count-rate channel. The two log count-rate channels are identical and linear with respect to each other over count rates from 50 to 50,000 cpm. The outputs of the two log count-rate circuits are used to drive meters and recorders that indicate the count rate in each channel. These two outputs are also impressed on the count rate in each channel, and on the grids of a difference amplifier which indicates

the ratio between the two counts. This ratio output is displayed on a meter and a recorder.

The breadboard model has been tested and found to operate satisfactorily over a temperature range of from 0 to 150°F. The alpha and beta channels up to the input of the log count-rate circuits were designed using transistor circuitry. Relatively inexpensive transistors were used throughout.

The log count-rate channels and the ratio amplifiers were designed using vacuum tubes. A block diagram of the system is shown in Figure 43.

Fig. 43 α, β air monitor.

3.2 Constant Air Monitor Noise Reduction (G. F. Brockett, W. L. Fouts)

The operating noise of Constant Air Monitors (CAM) is a familiar and often unpleasant working condition in many locations. A CAM was obtained for experiment and its noise viewed by the method of narrow-band analysis. The generated noise was considered to exceed that described by acoustic authorities as "office environment unsatisfactory".

During subsequent experimental work an exhaust muffler was evolved which reduced the operating noise as much as 34 db in one portion of the noise spectrum. The inexpensive device consists of a Tygon-tubing tail pipe muffler with three side branch resonators. The mechanical arrangement is shown in schematic form in Figure 44. The improved noise conditions for personnel with this arrangement is dramatic, yet the cost of making the improvement is small.

Fig. 44 Constant air monitor muffler.

IV. APPLIED MATHEMATICS

1. IBM 650 PROGRAM FOR THE ANALYSIS OF DIRECTIONAL CORRELATION OF GAMMA RAYS DATA (Darwin D. Metcalf)

This program carries out a weighted least-squares analysis of directional correlation data. These data consist of measurements of the coincidence-counting rate of two cascade nuclear radiations as a function of the angle between their propagation vectors. From the results of this analysis, one may obtain some information concerning the nuclear levels and the nuclear transitions involved in the decay.

The program input data consist of several coincidence-counting rates for each of a number of angles, and the standard deviation of the rates. The random coincidence-counting rate, its standard deviation, and the average of all counting rates for all angles are also input.

The program determines the least-squares coincidence-counting rate (gross minus random), and its statistical weight, which is the reciprocal of the corrected squared deviation. With these data, the program calculates the coefficients A_i , $i = 0, 2, 4$, of the expression for the directional correlation of two cascade radiations

$$A_0 + A_2 P_2(\cos\theta) + A_4 P_4(\cos\theta)$$

where θ is the angle between the propagation vectors of the two radiations, and $P_i(\cos\theta)$ is the i th order legendre polynomial. The values of the polynomials are generated by the program. The values of A_i are found by matrix methods. The A_i obtained give the best fit in the least-squares sense to the previously determined coincidence-counting rates. The standard deviations of the coefficients also are obtained.

The ratios $\frac{A_2}{A_0}, \frac{A_4}{A_0}, \frac{A_2 + A_4}{A_0}$

and their standard deviations are calculated. These quantities are useful in comparing the experimental results with theoretical calculations.

The program obtains a "figure of merit" for each run. This is a value that, depending upon its deviation from unity, indicates the importance of non-statistical errors.

In addition to the quantities mentioned above, the program also punches out the elements of the symmetric matrix used in obtaining the A_i , the corrected counting rates, their standard deviation, the angles at which data were taken, the random-counting rate, its standard deviation, and the average coincidence-counting rate.

The program differs from similar previous programs in the respect that the legendre polynomials are generated by the program, so that the coincidence

data may be taken at any desired set of angles, in contrast to fixed locations and numbers of angles in the previous programs.

2. LSGF-1: QUANTITATIVE ANALYSIS OF GAMMA RAY SPECTRA
BY THE METHOD OF LEAST SQUARES
(W. A. Hestir, R. L. Heath, R. D. Helmer)

LSGF-1 is an IBM program to perform a quantitative analysis of gamma ray spectra obtained from the 256-channel analyzer of the physics section.

Part I of the program accepts card data converted from the paper tape punch on the 256-channel analyzer. This data is then fit with known standard gamma ray spectra using the method of least squares to obtain a quantitative analysis. Double precision arithmetic is used to insure that the matrix inversion involved in the least-squares analysis will not fail for slightly ill-conditioned matrices.

Part II of the program computes the error analysis terms associated with the computations performed in Part I of the program. These terms are punched out in fixed point format to facilitate plotting for comparative purposes. An option to punch the computed value of the curve that has been fit in the standard curve format for later use as a standard curve is provided.

The program is currently being used on a routine basis.

3. MUFTR-E: FLUX WEIGHTING EDIT FOR MUFTR
(W. A. Hestir, A. W. Brown)

MUFTR-E is an IBM 650 program using the cross section and resonance decks from the MUFTR library to obtain averages of the cross sections σ_s , σ_a , $\bar{\mu}\sigma_s$, ν , σ_f , $\nu\sigma_f$, and σ_{in} over the flux spectrum from a MUFTR problem.

The program is designed to be run in conjunction with MUFTR in that it is assumed that the flux spectrum and slowing-down density tables computed in MUFTR are available in the computer at the time this program is run.

4. REP1: CALCULATION OF RESONANCE ESCAPE PROBABILITIES
(W. A. Hestir, D. R. Metcalf)

REP1 is an IBM 650 program using the cross section and resonance decks of the MUFTR library to calculate resonance escape probabilities used in determining the "L" factors for MUFTR. The MUFTR "L" factors are used to correct for the effect of self-shielding and Doppler broadening of U-238 in the MUFTR program.

5. BALD1: DATA REDUCTION OF THE CHEMICAL CONTROL OUT-OF-PILE LOOP

(W. A. Hestir, F. H. Just, J. M. Waage)

BALD1 is an IBM 650 program to reduce and normalize data taken in the testing of the chemical control out-of-pile loop.

Data for the ink detectors, armature voltages, and pneumatic-to-current convertors are processed and punched out in a form suitable for plotting.

6. HED1F: FORTRAN-SOAP OUTPUT SUBROUTINE

(W. A. Hestir, L. J. Gannon, R. J. Wagner)

HED1F is an IBM 650 Fortran-Soap subroutine for obtaining alphabetic or numeric output.

This is a modified version of the older HEADF subroutine using the special utility boards for the 533 and 407 to get up to 60 columns of alphabetic headings or eight 10-digit words of numeric output.

7. COMPUTATION OF A PERIODIC INPUT SIGNAL

(L. A. Schmittroth, D. H. Gipson)

An IBM 650 program called FLOOK has been written to compute the periodic input signal of a linear network when a periodic output signal and the transfer function are known. The method is general but the program, FLOOK, currently is tailored to the analysis of pressure transducers.

If T represents the period of both the known output signal, e_o , and the unknown input signal, e_I , and $\omega = 2\pi/T$, each may be expanded into a Fourier series:

$$e_I(t) = A_o^I + \sum_{n=1}^{\infty} (A_n^I \cos n\omega t + B_n^I \sin n\omega t)$$

$$e_o(t) = A_o^O + \sum_{n=1}^{\infty} (A_n^O \cos n\omega t + B_n^O \sin n\omega t)$$

The assumption of linearity implies that if $f_1(t)$ and $f_2(t)$ are inputs and $g_1(t)$ and $g_2(t)$ are outputs then the output with $\alpha_1 f_1 + \alpha_2 f_2$ as input is just $\alpha_1 g_1 + \alpha_2 g_2$. Further if the input is $e^{i\omega t}$ (ie, a purely sinusoidal complex signal) the output is $T(i\omega) e^{i\omega t}$.

The input and output as sums of components $e^{in\omega t}$ are written, therefore, as:

$$e_I(t) = \sum_{n=-\infty}^{\infty} C_n^I e^{in\omega t}$$

with $A_0^I = C_0^I$, $A_n^I = 2 \operatorname{Re} \left\{ C_n^I \right\}$ $B_n^I = -2 \operatorname{Im} \left\{ C_n^I \right\}$

and $C_n^I = \bar{C}_n^I$, where the bar denotes the complex conjugate. Similarly,

$$e_0(t) = \sum_{n=-\infty}^{\infty} C_n^O e^{in\omega t}$$

Now, because of linearity

$$C_n^O e^{in\omega t} = T(in\omega) C_n^I e^{in\omega t}$$

and with the exponential cancelled $C_n^I = C_n^O / T(in\omega)$. The relations between the C's and A's and B's may be written

$$C_n^I = \frac{A_n^I - i B_n^I}{2}$$

$$C_n^O = \frac{A_n^O - i B_n^O}{2}$$

If $T(in\omega) = T_1(n\omega) + iT_2(n\omega)$, then

$$\begin{aligned} A_n^I - i B_n^I &= \frac{A_n^O - i B_n^O}{T_1(n\omega) + iT_2(n\omega)} \\ &= \frac{A_n^O T_1(n\omega) - B_n^O T_2(n\omega) - i B_n^O T_1(n\omega) + A_n^O T_2(n\omega)}{T_1^2(n\omega) + T_2^2(n\omega)} \end{aligned}$$

Separating real and imaginary parts

$$A_n^I = \frac{A_n^O T_1(n\omega) - B_n^O T_2(n\omega)}{T_1^2(n\omega) + T_2^2(n\omega)}$$

$$B_n^I = \frac{B_n^O T_1(n\omega) + A_n^O T_2(n\omega)}{T_1^2(n\omega) + T_2^2(n\omega)}$$

$$T_1(n\omega) = \frac{\omega_n^2 T_0 (\omega_n^2 - n^2 \omega^2)}{(\omega_n^2 - n^2 \omega^2)^2 + (\xi \omega_n n\omega)^2}$$

$$T_2(n\omega) = - \frac{\omega_n^2 T_0 \xi \omega_n n\omega}{(\omega_n^2 - n^2 \omega^2)^2 + (\xi \omega_n n\omega)^2}$$

ω_n = natural frequency of the transducer and is determined experimentally

ξ = damping of the transducer and is determined experimentally

T_0 = value of the transfer function at $\omega = 0$

$\omega = 2\pi/T$

T = fundamental period of the output signal as read from the trace of the output signal.

The program computes the input signal, $e_I(t)$, at equal intervals of time for J intervals from 0 to T .

$$t_j = j (T/J), \quad j = 0, 1, 2, \dots, J$$

From the computed signal it computes the flow in gpm by

$$GPM_j = \sqrt{C e_I(t_j)}$$

where C = multiplier which converts $e_I(t_j)$ to $(GPM_j)^2$, and is an experimentally determined calibration factor of the particular transducer.

Then the program computes

$$(GPM_j)_N = \frac{GPM_j}{(GPM)_{Max}}$$

where $(GPM)_{Max}$ is a predetermined maximum flow.

The program allows for two sets, ($k = 1, 2$), of values ($\xi_k, \omega_{n_k}, T_{0_k}$) of the transfer function, $T(in\omega)_k$, and four values of C . The transducers, FE-11 and FE-12, use the transfer function, $T(in\omega)_1$, and the C multipliers, C_1 and C_2 , respectively. The transducers, FE-101 and FE-102 use transfer function, $T(in\omega)_2$, and the C multipliers, C_3 and C_4 , respectively. The identification numbers (FE-11, FE-12, FE-101, and FE-102) refer to four different transducers in the system.

8. MODIFICATION TO DMM-II (MDMM-II) (Steven Tong)

It was found that the neutron balance of the one-dimensional diffusion code, DMM, is not satisfactory. It was also found that the neutron balance of MDMM-I, although far better than that of DMM, is still not highly satisfactory in some cases. Investigations showed that the discrepancy is caused by the approximation of the derivatives at the boundaries (inner, outer, or an interface). This program provides better approximations for the derivatives at the boundaries so that the neutron balance will check out. It also includes the absorption and source modifications made in MDMM-I.

8.1 Derivative at Outer Boundary

Let N be the mesh point which lies on the boundary. Let N-1 be the adjacent mesh point to the left of N. Let m be the mid-point between N-1 and N. Let h be the mesh width (see diagram).

Then the following equation holds:

$$\iiint_V -D \nabla^2 \phi dV = \iiint_V H dV - \iiint_V T \phi dV \quad (1)$$

Where V is the small volume of length $\overline{X_m X_N}$

$\iiint_V -D \nabla^2 \phi dV$ is the leakage term

$\iiint_V H dV$ is the source term

$\iiint_V T \phi dV$ is the absorption term

$$\text{now, } \iiint_V -D \nabla^2 \phi dV = \alpha \left[-DX^0 \frac{d\phi}{dx} \right]_{x=x_m}^{x=x_N}$$

$$= \alpha \left[-DX_N^\rho \left(\frac{d\phi}{dx} \right)_{x=x_N} + D x_m^\rho \left(\frac{d\phi}{dx} \right)_{x=x_m} \right]$$

$$\approx \alpha \left[-DX_N^\rho \left(\frac{d\phi}{dx} \right)_{x=x_N} + D \left(x_N - \frac{h}{2} \right)^\rho \left(\frac{\phi_N - \phi_{N-1}}{h} \right) \right]$$

where $\alpha = \begin{cases} 1 & \text{if } \rho = 0 \\ 2\pi & \text{if } \rho = 1 \\ 4\pi & \text{if } \rho = 2 \end{cases}$

and $\rho = 0, 1, 2$ refers to slab, cylindrical and spherical geometries, respectively.

But, $\iiint_V H dV - \iiint_V T \phi dV = \iiint_V (H - T\phi) dV$

$$= \alpha \left[\int_{x_m}^{x_N} x^\rho (H - T\phi) dx \right]$$

$$\approx \alpha \left[x_N^\rho (H_N - T_N \phi_N) \frac{h}{2} \right]$$

Hence, Equation (1) becomes

$$\alpha \left[-D x_N^\rho \left(\frac{d\phi}{dx} \right)_{x=x_N} + D \left(x_N - \frac{h}{2} \right)^\rho \left(\frac{\phi_N - \phi_{N-1}}{h} \right) \right] = \alpha \left[x_N^\rho (H_N - T_N \phi_N) \frac{h}{2} \right]$$

or

$$-DX_N^\rho \left(\frac{d\phi}{dx} \right)_{x=x_N} = -D \left(x_N - \frac{h}{2} \right)^\rho \left(\frac{\phi_N - \phi_{N-1}}{h} \right) + x_N^\rho (H_N - T_N \phi_N) \frac{h}{2} \quad (2)$$

Let $J^-(x_N) = \lim_{x \rightarrow x_N^-} \left[-D \frac{d\phi}{dx} \right]$

Then $J^-(x_N) = \frac{-D}{h} \left(1 - \frac{h}{2x_N} \right)^\rho (\phi_N - \phi_{N-1}) + \frac{h}{2} (H_N - T_N \phi_N) \quad (3)$

8.2 Derivative at Inner Boundary

Let N be the mesh point which lies on the boundary. Let $N + 1$ be the adjacent mesh to the right of N . Let w be the mid-point between N and $N + 1$ and let k be the mesh width (see diagram).

Then the following equation holds:

$$\iiint_V -D \nabla^2 \phi dV = \iiint_V H dV - \iiint_V T \phi dV \quad (4)$$

Where V is the small volume of length $\overline{X_N X_w}$

$\iiint_V -D \nabla^2 \phi dV$ is the leakage term

$\iiint_V H dV$ is the source term

$\iiint_V T \phi dV$ is the absorption term

$$\begin{aligned} \text{now, } \iiint_V -D \nabla^2 \phi dV &= -\alpha \left[-DX^\rho \frac{d\phi}{dx} \right]_{x=x_N}^{x=x_w} \\ &= -\alpha \left[-DX_N^\rho \left(\frac{d\phi}{dx} \right)_{x=x_N} + DX_w^\rho \left(\frac{d\phi}{dx} \right)_{x=x_w} \right] \\ &\approx -\alpha \left[-DX_N^\rho \left(\frac{d\phi}{dx} \right)_{x=x_N} - D \left(X_N + \frac{k}{2} \right)^\rho \frac{\phi_N - \phi_{N+1}}{k} \right] \end{aligned}$$

where α, ρ are as defined before.

$$\text{But, } \iiint_V H dV - \iiint_V T \phi dV = \iiint_V (H - T\phi) dV$$

$$= \alpha \left[\int_{X_N}^{X_w} X^\rho (H - T\phi) dX \right]$$

$$\approx \alpha \left[X_N^\rho (H_N - T_N \phi_N) \frac{k}{2} \right]$$

Hence Equation (4) becomes

$$-DX_N^\rho \left(\frac{d\phi}{dx} \right)_{x=x_N} = \frac{D}{k} \left(X_N + \frac{k}{2} \right)^\rho (\phi_N - \phi_{N+1}) - X_N^\rho (H_N - T_N \phi_N) \frac{k}{2} \quad (5)$$

If $\rho = 0$, Equation (5) becomes

$$-D \left(\frac{d\phi}{dx} \right)_{x=x_N} = \frac{D}{k} (\phi_N - \phi_{N+1}) - (H_N - T_N \phi_N) \frac{k}{2} \quad (6)$$

Let $J^+(X_N) = \lim_{X \rightarrow X_N^+} \left[-D \frac{d\phi}{dx} \right]$

Then $J^+(X_N) = \frac{D}{k} (\phi_N - \phi_{N+1}) - (H_N - T_N \phi_N) \frac{k}{2} \quad (7)$

If $\rho \neq 0$

$$X_N \neq 0$$

and $J^+(X_N) = \frac{D}{k} \left(1 + \frac{k}{2X_N} \right)^\rho (\phi_N - \phi_{N+1}) - (H_N - T_N \phi_N) \frac{k}{2} \quad (8)$

$$X_N = 0$$

Then, by symmetry, $J^+(X_N) = 0 \quad (9)$

8.3 Derivative at an Interface

Clearly, Equations (3) and (8) are $J^-(X_N)$ and $J^+(X_N)$, respectively, for an interface.

8.4 The Neutron Balance

Let X_1 and X_N be the lower bound and upper bound of a region, respectively.

$$\begin{aligned} \text{Then, } \iiint_V -D \nabla^2 \phi dV &= \alpha \left[-DX^\rho \frac{d\phi}{dx} \right]_{x=x_1}^{x=x_N} \\ &= \alpha \left[X_N^\rho J^-(X_N) - X_1^\rho J^+(X_1) \right] \end{aligned} \quad (10)$$

where V is the volume of that region and α is as defined before.

Now, $\iiint_V (H - T\phi) dV = \alpha \int_{X_1}^{X_N} X^\rho (H - T\phi) dx$

If the length is subdivided into $N-1$ equal subintervals and the trapezoidal rule is used

$$\alpha \int_{X_1}^{X_N} X^\rho (H - T\phi) dx = \frac{\alpha h}{2} \left[X_1^\rho (H_1 - T_1 \phi_1) + 2X_2^\rho (H_2 - T_2 \phi_2) + \dots + 2X_N^\rho (H_N - T_N \phi_N) \right] \quad (11)$$

At a point X_n of the region, the following difference equation holds:

$$-D \left[\frac{\phi_{n-1} - 2\phi_n + \phi_{n+1}}{h^2} + \frac{\rho}{X_n} \frac{\phi_{n+1} - \phi_{n-1}}{2h} \right] = H_n - T \phi_n$$

Knowing the fact that within a region $T_1 = T_2 = \dots = T_N = T$, Equation (11) becomes

$$\alpha \int_{X_1}^{X_N} X^\rho (H - T\phi) dx = \frac{\alpha h}{2} \left[X_1^\rho (H_1 - T \phi_1) + 2X_2^\rho S_2 + \dots + 2X_N^\rho S_N + X_N^\rho (H_N - T \phi_N) \right]$$

where $S_n = -D \left[\frac{\phi_{n-1} - 2\phi_n + \phi_{n+1}}{h^2} + \frac{\rho}{X_n} \frac{\phi_{n+1} - \phi_{n-1}}{2h} \right]$

Therefore,

$$\int_{X_1}^{X_N} X^\rho (H - T\phi) dx = X_1^\rho \left[\frac{h}{2} (H_1 - T\phi_1) - \frac{D}{h} \left(1 + \frac{\rho h}{2X_1} \right) (\phi_1 - \phi_2) \right] + X_N^\rho \left[\frac{h}{2} (H_N - T\phi_N) - \frac{D}{h} \left(1 - \frac{\rho h}{2X_N} \right) (\phi_N - \phi_{N-1}) \right] \quad (12)$$

Now, for neutron conservation the following equation must hold:

$$\iiint_V -D \nabla^2 \phi dV = \iiint_V (H - T\phi) dV$$

From Equation (10).

$$\begin{aligned} \iiint_V -D \nabla^2 \phi dV &= \alpha \left[X_N^\rho J^-(X_N) - X_1^\rho J^+(X_1) \right] \\ &= \alpha \left\{ X_N^\rho \left[\frac{-D}{h} \left(1 - \frac{h}{2X_N} \right)^\rho (\phi_N - \phi_{N-1}) + \frac{h}{2} (H_N - T\phi_N) \right] \right. \\ &\quad \left. - X_1^\rho \left[\frac{D}{h} \left(1 + \frac{h}{2X_1} \right)^\rho (\phi_1 - \phi_2) - \frac{h}{2} (H_1 - T\phi_1) \right] \right\} \quad (13) \end{aligned}$$

by Equations (3) and (8).

From Equation (12)

$$\begin{aligned} \iiint_V (H - T\phi) dV &= \alpha \int_{X_1}^{X_N} X^\rho (H - T\phi) dx \\ &= \alpha \left\{ X_N^\rho \left[\frac{-D}{h} \left(1 - \frac{\rho h}{2X_N} \right) (\phi_N - \phi_{N-1}) + \frac{h}{2} (H_N - T\phi_N) \right] \right. \\ &\quad \left. - X_1^\rho \left[\frac{D}{h} \left(1 + \frac{\rho h}{2X_1} \right) (\phi_1 - \phi_2) - \frac{h}{2} (H_1 - T\phi_1) \right] \right\} \quad (14) \end{aligned}$$

For $\rho = 0$ and 1, Equations (13) and (14) are identical.

Hence the neutron balance is exact.

For $\rho = 2$, assuming h is small, the equations are

$$\left(1 + \frac{h}{2X} \right)^\rho \approx 1 + \frac{\rho h}{2X}$$

and

$$\left(1 - \frac{h}{2X} \right)^\rho \approx 1 - \frac{\rho h}{2X}$$

Hence, Equations (13) and (14) are equal and the neutrons are conserved.

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