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MTR-ETR TECHNICAL BRANCHES
QUARTERLY REPORT
April 1-June 30, 1963

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NATIONAL REACTOR TESTING STATION
US ATOMIC ENERGY COMMISSION

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MATERIALS TESTING REACTOR-ENGINEERING TEST REACTOR TECHNICAL BRANCHES
QUARTERLY REPORT
APRIL 1 - JUNE 30, 1963

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MTR-ETR TECHNICAL BRANCHES QUARTERLY REPORT
1963 QUARTER 2
APRIL 1 - JUNE 30, 1963

SUMMARY

Distortion and cracking of portions of the MTR beryllium reflector are reported. Preliminary metallurgical and gas analyses on fracture chips indicate that some of the fractures may have occurred at exposures much lower than 10^{22} nvt (> 1 Mev).

The resonance integrals of mixtures of gold and indium oxide as calculated by a modified ZUT code were compared with the integrals calculated with the RBU Monte Carlo program. Although there were some discrepancies between the results of the two methods, both methods showed some interaction between the prominent gold and indium resonances.

The geometry routine, zone construction subroutine, and particle search subroutine have been programmed and checked for the Phillips general purpose Monte Carlo program. Flow charts are being written for the collision routine and the pre-Monte Carlo routine.

An unsuccessful attempt was made to observe gamma rays emitted in the decay of Pt-200. The energies and intensities of the gamma rays from Au-200 were remeasured.

Statistical techniques were employed to determine those angles and relative frequencies of measurements which will, for a given total number of observations, give the smallest variances for the directional-correlation coefficients in correlation functions which contain $P_6(\cos \theta)$ and $P_8(\cos \theta)$ terms.

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I. REACTOR PHYSICS AND ENGINEERING

1. REACTOR FUELS AND MATERIALS DEVELOPMENT

(W. C. Francis)

1.1 Irradiation Effects on Beryllium (J. M. Beeston, R. L. Tromp, W. F. Zelezny)

Recent observations of the MTR beryllium reflector showed considerable distortion and cracking in the regions of high radiation exposures. As a result a program was planned to study samples of the highly irradiated MTR pieces for the purpose of establishing the responsible mechanism and obtaining a quantitative measure of the damage as a function of irradiation history. A preliminary report of results is given here.

The irradiation effects on beryllium are being examined by means of several procedures. The Beryllium Gases Experiment, which has been underway for several years, has been discussed previously [1]. A high temperature capsule irradiation of beryllium is in preparation. Reported here are three procedures, along with preliminary results, used with samples of MTR reflector beryllium: (a) determination of bowing of lattice and fixed reflector pieces, (b) testing samples cut from the LB-15 lattice piece, and (c) examination of the fracture fragments from the fixed reflector.

The MTR lattice beryllium pieces have shown bowing over the length of the piece. The bowing of a number of these pieces was measured as was that between the MTR reflector faces as indicated in Table I-1. The relationship

TABLE I-1

VALUES OF STRAIN FOR VARIOUS
MTR BERYLLIUM LATTICE AND REFLECTOR PIECES

LB Piece	Bowing(a) (in.)	Exposure ($n/cm^2 > 1 \text{ MeV}$ $\times 10^{-21}$)	Strain (in./in.)	
			Measured	Calculated
LB-4	0.035	4.20	0.0005	0.0002
LB-9	0.035	4.28	0.0005	0.00021
LB-13	0.035	1.93	0.0005	0.0001
LB-4X-3	0.035	3.91	0.0005	0.00019
LB-14	0.045	7.10	0.0006	0.00035
LB-8	0.065	7.71	0.0013	0.00038
LB-16	0.065	7.37	0.0013	0.00036
LB-15	0.10	10.23	0.0021	0.00062
E-3	0.053	≈ 45	0.0005	0.0022
E-4	0.091	≈ 45	0.0009	0.0022

(a) The bowing on the LB pieces was measured over 24 in. length, while that on the reflector pieces was measured over 39.7 in.

An irradiated MTR lattice beryllium piece (LB-15) was used to produce a number of samples for measurements of strength changes and evidences of internal stresses. During the underwater cutting several longitudinal cracks developed, six to eight inches long, perpendicular to the saw cut. X-ray diffraction measurements on some of the samples indicate the possibility of the

presence of non-uniform strain in the irradiated beryllium. These measurements are continuing.

The results of compression tests on samples 1/2 in. square by 1-1/2 in. long cut from the LB piece are presented in Table I-2. The samples were cut from two sections of the piece, ie, the middle and the bottom. The fluxes at the samples on the horizontal mid-plane of the piece, as well as at the samples on the bottom where there is a considerable flux gradient were calculated using the measurements given in IDO-16678 [2]. A flux gradient, extending away from the core, of 60% in 1-1/2 in. was assumed linear to calculate the flux at the center of the 1/2 in. samples, ie, the flux 1/4 in. from the higher flux side and the lower side of the LB-15 piece. The flux gradient in the widest core direction is neglected, but most of the irradiation exposure resulted in a position toward the core center in this direction. The exposure values represent a summation at 30 and 40 MW from the various lattice positions from which LB-15 received its irradiation.

TABLE I-2
COMPRESSION STRENGTH OF IRRADIATED BERYLLIUM^(a)
(LB-15 Piece)

Sample Number	Exposure ^(b) ($n/cm^2 > 1 \text{ MeV}$ $\times 10^{-21}$)	Compression, Yield Strength (0.2% offset) (Psi $\times 10^{-3}$)	Compression, Ultimate Strength (Psi $\times 10^{-3}$)	Total Strain (% in 1.5 in.)	Plastic ^(c) Strain (% in 1.5 in.)
LL-6,7,8,9	0.6	79	153	14.4	12.9
LT-6,7,8,10	1.1	98	184	11.5	11.0
LL-1,2,3	1.6	117	194	13.9	12.4
LT-3,4,5	3.2	122	129	3.1	1.5
MT-6	5.0	163	163	2.8	0.7

(a) Values are average of number of samples indicated in first column.

(b) Calculated from flux profiles of MTR Cycle 146 for various positions in which LB-15 received irradiation (approximate values).

(c) Measured from yield point to point of fracture.

Samples at the middle section toward the core having exposures of $14 \times 10^{21} \text{ n/cm}^2$ ($> 1 \text{ MeV}$) cracked too badly to test, as did some of those on the side away from the core having exposures of $5 \times 10^{21} \text{ n/cm}^2$.

The compression yield strengths and ultimate strengths generally show the characteristic increase with increased irradiation exposure while the percent plastic strain decreases. The deviation in individual test values was not great and could be explained partly by specimen chipped corners, variation in strain rate (from 0.001 to 0.01 in./in./min), misalignments, and differing exposure from a flux gradient in a direction in the widest core dimension in the reactor core. These are preliminary values as the work is still in progress.

Examination of the beryllium reflector in the MTR, after the discovery of several pieces of beryllium in the flash evaporator of the process water system,

showed several fractures in the beryllium wall surrounding the HB-2 facility. Some of the pieces found in the flash evaporator appeared to match up with missing areas in the reflector. In an attempt to establish the cause of the beryllium fracturing, the maximum temperature produced in the beryllium, the neutron exposure prior to fracture, and the time elapsed since the fracture occurred, an extensive investigation was initiated which included analysis of fracture surface by fractography techniques, of density of the beryllium, of gas analysis, and of annealed swelling. Some of these results were reported previously [1].

Fractography results, gas analysis, density measurements, and analysis of strain measurements are as follows:

- (1) The electron microscopy of the corroded fracture surface of the small fragment (Figure I-2) indicates that fracture occurred a long time ago. The fresh fracture (Figure I-3) indicates no swelling and no increase in gas bubble voids as compared with the fracture surface of the original MTR beryllium, (Figure I-4).

The electron microscopy of the corroded fracture surface of the F-2 large fragment (Figure I-5) indicates that this fracture occurred some time ago. The fresh fracture of the F-2 large fragment (Figure I-6) shows an increase in gas bubble voids which can be due to higher exposure with increased partition of gas atoms to bubbles or higher temperature than a comparison fracture sample (Figure I-7).

- (2) Gas analysis results to date are given in Table I-3.

- (3) The density measurements give for both the F-2 large fragment (average of 2 samples) and for the small piece 1.83 g/cm^3 Be. The standard deviation of these measurements of ± 0.002 is expected to be as measured for other samples [3]. Since the densities for the original beryllium were given [4] as 1.85 g/cm^3 , this represents a one percent density change.

Samples from the F-2 large fragment were annealed for 20 hr at various temperatures with no appreciable swelling observable until a temperature of 600°C was reached. The temperatures and corresponding densities were as follows: 500°C - 1.82; 600°C - 1.75; and 700°C - 1.60.

- (4) Values of strain for various MTR beryllium lattice and reflector pieces are given in Table I-1. From the exposure values given in this table, it is possible to calculate values of strain based on the assumption that the gas atoms are contained in the crystal structure, an assumption that should be reasonably valid for irradiation temperature $< 350^\circ\text{C}$ and for exposures not too great. The question of how great an exposure remains to be answered. This assumption is a first approximation since the expansion produced by the presence of the irradiation-produced defects, other than helium atoms, is neglected as well as the possibility of the helium atoms being held in other trapping centers. In the

Fig. I-2 Corroded fracture surface of small fragments of beryllium found in flash evaporator.

Fig. I-3 Fresh fracture surface of small fragments of beryllium found in flash evaporator.

Fig. I-4 Fresh fracture surface of original beryllium.

Fig. I-5 Corroded surface of large fragments of beryllium from F-2 reflector piece as found in flash evaporator.

table, a comparison is made between the values calculated in this manner and the measured values.

It is noteworthy that the calculated value of strain is lower than the measured value except in the case of the reflector pieces E-3 and E-4. There are four possibilities which might explain why the calculated strain on these reflector pieces is higher than the measured values:

Fig. I-6 Fresh fracture surface of large fragments of beryllium from F-2 fragment of beryllium from F-2 reflector piece found in flash evaporator.

Fig. I-7 Fresh fracture surface of LB-15 beryllium piece.

TABLE I-3

HELIUM CONTENT OF FRAGMENTS FROM FIXED REFLECTOR PIECES

<u>Origin of Fragment</u>	<u>Sample Number</u>	<u>Helium Content</u> ($\frac{\text{cc He}^4}{\text{cc Be}}$)
F-2	1	28.3
F-2	2	24.4
F-2	3	26.3
F-6 (possibly)	1	0.82

(1) If the flux values used were too high, the calculated values would be higher than the measured value. At 40 Mw the value used was 2×10^{14} n/cm² sec (> 1 MeV), and at 30 Mw the value used was 1.5×10^{14} n/cm² sec (> 1 MeV). These values were considered to be the best available.

(2) An increase in the fraction of the gaseous atoms increases the partition of the gaseous atoms into the original void space.

(3) An increase in the thickness of the reflector pieces E-3 and E-4 offers increased restraint.

(4) Loss of gas by diffusion out of the pieces.

From the above incomplete results, it appears that the temperature of the beryllium in the F-2 large fragment was between 150 and 650°F but probably closest to the lower value; that the main fracture occurred some time ago; and that the smaller flat piece was in the reactor a much shorter time. The final analysis of the problem awaits completion of the data.

2. REACTOR EXPERIMENTS

(E. Fast)

2.1 A Monte Carlo Study of Resonance Absorption in Gold and Indium Lumps (W. K. Foell, R. A. Grimesey, and S. Tong)

In an attempt to study in detail the phenomenon of resonance absorption in a lumped absorber, calculations are being carried out using the RBU general purpose Monte Carlo code [5]. This code provides the basis for a microscopic study of the spatial and energy dependence of the neutron flux in a heterogeneous system without resorting to any of the usual idealizations of current resonance integral theory.

The particular system being studied is a model of a cell in the Advanced Reactivity Measurement Facility (ARMF). The cell, located in a lattice position of the reactor, contained an absorbing cylinder of gold and lead oxide in one case, indium oxide and lead oxide in a second case, and a mixture of gold and indium oxide in the third case. In each instance the cylinder was enclosed by twenty mil cadmium and surrounded by a fuel composition characteristic of the ARMF. Because the geometry routine of RBU can track particles in three dimensional space, it was possible to treat the absorber as a finite cylinder instead of using the customary one dimensional approximation.

An extremely fine microscopic energy group structure, containing 134 energy groups between 0.55 and 7.9 eV, was chosen to permit high resolution in the vicinity of the resonance absorption peaks. At each particle energy, RBU calculates the exact cross section directly from the Doppler broadened single-level Breit-Wigner formula, so that no group averaging of cross sections was necessary. The group structure was used only as a means of tallying fluxes for output.

The results of this investigation were compared with a solution of the first flight coupled integral equations as used by Nordheim and co-workers in the ZUT code [6]. Several of the idealizations of his method are obviated in the full Monte Carlo treatment. In particular, no model of the slowing down kernel was necessary since, after a sufficient initial equilibrium time, the high energy neutrons acting as a slowing down source should be in a distribution characteristic of the particular system. Furthermore, no use was made of the flat flux approximation and the resulting first flight, one dimensional escape probabilities.

A comparison of the fluxes in the gold-indium mixture as calculated by RBU and ZUT-MOD-3 is shown in Figure I-8. ZUT-MOD-3 is a modification of the ZUT code, developed by Brown and Connolly at Stanford University [7], which makes possible the treatment of two resonance absorbers with overlapping cross sections.

The resonance integrals, covering the range from 0.55 to 7.9 eV, are given below as calculated by the two techniques.

<u>Material in Lump</u>	<u>Isotope</u>	<u>RBU</u>	<u>ZUT</u>
Indium Oxide-Lead Oxide	In-115	219.6 b	222.9 b
Indium Oxide-Gold	In-115	192.8	210.5
Indium Oxide-Gold	Au-197	101.8	115.4
Gold-Lead Oxide	Au-197	109.0	133.4

Fig. I-8 Comparison of the fluxes in the gold-indium mixture as calculated by RBU and ZUT-MON-3.

Both methods indicate some interaction between the resonances of gold and indium as described by the activation experiments of Drown, Connolly, and Foell [8]. There also are some discrepancies between some of the RBU and ZUT results. In part, this discrepancy points out the difficulty of attempting to use the idealized definition of the resonance integral in a case where the flux may not be in an asymptotic distribution. Because the Monte Carlo treatment generates its own slowing down source, as contrasted to the asymptotic $1/E$ flux of the ZUT code, it was necessary to normalize the RBU resonance integral by means of a calculated source function. This was accomplished by a least squares fit of the RBU flux to a C/E dependence in the 10 eV-100 eV region. The deviation of the Monte Carlo flux from an exact C/E behavior may account for some of the difference between results of the two methods.

3. REACTOR PHYSICS (D. R. Metcalf)

3.1 Phillips General Purpose Monte Carlo Program for the IBM-7040 (R. A. Grimesey and S. Tong)

Progress on the Phillips general purpose Monte Carlo program for the IBM-7040 computer proceeded to the point where the beginning of the flow charts for the collision routine using a monoenergetic thermal group option was started. The flow charts for the pre-Monte Carlo routine where the cross sections for the specific isotopes in each material are merged with an arbitrary group structure were essentially completed and the "Geometry Routine" of the Monte Carlo was programmed and checked. It locates particles, advances particles to collision, computes distance to boundary and advances particles across boundaries.

The types of surface allowed in the "Geometry Routine" are given by the equation $A(x-x_0)^n + B(y-y_0)^n + C(z-z_0)^n + D = 0$ where $n = 1$ or 2 . If $n = 2$, then $A = B = 1$ and $C = 1$ or 0 .

There are 12 types of regions allowed in the "Geometry Routine". These 12 types of regions are bounded by a combination of plane and curved surfaces and include most reactor geometries.

In order to reduce the machine time for locating a particle in a region the reactor is subdivided into zones. Each zone contains one or more regions. But no two zones contain the same region. In the "Particle Search" subroutine the particle is first located in a zone, then the particle is searched through the regions which are contained in that zone. There are 3 types of zone geometries used which depend on the reactor geometry; they are rectangular, cylindrical, and spherical. The "Zone Construction" and "Particles Region Search" subroutines are programmed and checked. It was found that more than 50% of machine time is saved in the "Particle Search" subroutine if the zone search method is used.

The actual programming of the pre-Monte Carlo cross section routine will follow later.

3.2 U-238 Resonance Integral Calculations (R. L. Curtis)

A study was undertaken and completed comparing the U-238 resonance integrals calculated by the GAM-I code [9] with those computed by the Nordheim ZUT code [10]. The purpose of this study was to verify the fast multigroup diffusion constants calculated by the GAM-I code for heterogeneous low-enriched fuel pins of the PL-2 type with a pitch of 0.585 in.

The model for the GAM-I equations is that of a homogeneous bare core reactor in one dimension. The NR and NRIA resonance approximations [11] are utilized for the heterogeneous resonance integral calculations.

A more accurate model of resonance absorption is incorporated in the Nordheim code. In this code, a Simpson's rule numerical integration of the resonances can be used as an option rather than the NR and NRIA approximation. In addition, the heterogeneous spatial dependence of the flux used in the Nordheim code is a more accurate model than the homogeneous solution obtained in the GAM-I code.

Table I-4 contains a comparison of the average resonance absorption cross sections from GAM-I and the Nordheim code over specified energy ranges corresponding to the GAM-I group structure in the resolved resonance range. The comparison indicates that the GAM-I results are quite accurate in view of the rather stringent approximations involved. Although the comparison of average cross sections over individual energy ranges differs by as much as + 22.62% and -26.4%, the total cross section over the resolved resonance range differs only by +3.34%. Since the neutron absorption in the fuel pins is proportional to the total absorption, it is felt that the GAM-I treatment of the resonance absorption is adequate for low enrichment oxide pin type cores of the pin size and pitch utilized in this study.

TABLE I-4

AVERAGE RESONANCES ABSORPTION CROSS-SECTIONS

Lower Energy (eV)	Values Calculated by GAM-I	Values Calculated by ZUT
748	0.3795	0.4296
583	0.4558	0.5151
454	0.4237	0.4779
354	0.3936	0.4193
275	0.3554	0.4291
215	0.4087	0.5282
167	0.8130	0.6432
130	0.3855	0.4049
101	1.4624	1.4023
78.9	0.5525	0.6053
61.4	1.2311	1.2245
47.9	0.0106	---
37.3	0.0125	---
29.0	2.6983	2.7844
22.6	0.0171	---
17.6	3.3657	3.6203
13.7	0.0233	---
10.68	0.0270	---
8.32	0.0313	---
6.48	7.2872	7.6450
5.04	0.0415	---
3.93	0.0478	---
Total	20.4235	21.1291

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II. NUCLEAR PHYSICS

1. CROSS SECTIONS PROGRAM

(M. S. Moore)

1.1 Fast Chopper

1.11 Ta-182 Cross Section (G. E. Stokes and T. E. Young). The occurrence of a large resonance at 0.147 eV in Ta-182 [1] suggests that this isotope may be useful in neutron temperature measurements. As a measure of this, Figure II-1 shows a plot of g vs neutron temperature. The quantity g defined by Westcott [2] gives the deviation from a $1/v$ cross section assuming a Maxwellian neutron distribution of temperature T . The slope of the curve is an indication of the temperature sensitivity. The steeper the slope the more sensitive the isotope is to temperature. The Lu-176 curve for the g factor is put in for

Fig. II-1 " g " vs neutron temperature for Ta-182.

Fig. II-2 Variation of the Ta-183/Ta-182 ratios for different irradiation times.

comparison. Schmid and Stinson [3] have demonstrated that Lu-176 is 7 times more sensitive to temperature change below 400°C than the commonly used Pu-239/U-235 fission activity ratio. The Ta-182 appears to have the same sensitivity as the Lu-176 for this temperature range.

The Ta-182 may have a place in temperature measurements where the Lu-176 is not practical. Ta-181 can be irradiated as a natural metal with a thermal absorption cross section of 21 ± 1 barn and a resonance integral of 590 barns. A fraction of the Ta-182 produced is converted to Ta-183 giving a Ta-183 to Ta-182 ratio which is a function of the effective cross section of Ta-182. Figure II-2 shows the variation of the Ta-183/Ta-182 ratios for different irradiation times as a function of neutron temperature. This ratio was calculated using a σ effective for Ta-182 that was the thermal value, 8200 barns, multiplied by the g factors for the various temperatures. The ratio remains good for long irradiations. R. P. Schuman [4] has irradiated Ta-181 foils in the MTR and the results show that the burnout rate is slow enough that Ta-182 may be useful where temperature measurements are desired for long irradiations.

2. NUCLEAR CHEMISTRY (W. H. Burgus)

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

Two small-scale Pa-233 processing runs were made in the hot alpha cave demonstrating the complete protactinium recovery scheme to be used in preparing an ≈ 1 gram sample for chopper cross section measurements. The first was a 1/50 scale test of chemical methods, performed in plastic and glass test tubes and using fritted glass filters. The second run was a 1/20 scale "pilot plant" experiment in which a stainless steel dissolver, glass fritted disc filters, and glass vessels were connected with plastic tubing to make a small-scale closed system in order that solution and precipitate handling techniques simulating those of the full-scale run could be tested. The chemical flowsheet consists of dissolution, two MnO_2 precipitations to carry protactinium, followed by two protactinium iodate precipitations, and calcination to Pa_2O_5 . The flowsheet was described in the previous quarterly report [5].

The first run produced 10.1 mg of nearly white Pa_2O_5 with a specific activity at 3 in. from the alpha cave monitor of ≈ 2500 r/hr/mg. This represented $\approx 85\%$ protactinium recovery in the final oxide product as measured by activity monitoring of feed and product.

A second run produced 9.7 mg of white Pa_2O_5 with a specific activity of ≈ 3000 r/hr/mg which was equivalent only to $\approx 40\%$ protactinium recovery as measured by activity monitoring. A more accurate method of determining the overall yield is based on aliquoting and counting a known fraction of the dissolver product and using this value along with the weight of the final Pa_2O_5 to compute overall recovery. On this basis, the overall recovery was only 34%. Up to the final protactinium iodate filtration the run had demonstrated $\approx 90\%$ Pa-233 recovery; at this point, however, over half of the protactinium iodate slurry was accidentally spilled. However, the final oxide was mixed with aluminum, pressed into a compact, and used for cross section measurements on the MTR fast chopper. Early inspection of the chopper data indicates that no significant quantities of materials other than Al and Pa_2O_5 are present in the sample.

The major difficulty shown by the 1/20 scale run (aside from the accidental spill) was failure of the Ascarite traps, located in vessel vent and vacuum lines, to absorb fission product iodine as it was released from solution during sparging or solution transfer operations. Installation of larger and more efficient iodine traps and a different vacuum setup is expected to solve this problem. The final slurry transfer which led to the spill was done in the 1/20 scale run into an open crucible filter as a matter of expediency. This will be a closed system in the full-scale work, thus avoiding the possibility of losing some of the product during transfer.

2.2 Repurification of U-232 and Isolation of Th-228 (R. P. Schuman, J. R. Berreth)

The U-232 sample was repurified by the procedure described in the last quarterly report [6]. The U_3O_8 -Al mixture was dissolved in HCl plus $\approx 1\%$ HNO_3 . The chloride solution was made ≈ 6 to 8M in HCl and fed to a Dowex

1-ion exchange column which absorbed the uranium and let aluminum and thorium pass through. The column was then washed with 8M HCl, and finally the uranium was eluted with H₂O. The uranium was recovered from the H₂O eluate by precipitation with NH₄OH, filtration into a ceramic filter crucible lined with ashless filter paper. It was then dried and ignited to U₃O₈ at 750 to 800°C. About 99% of the uranium was recovered. The purified U₃O₈ was pulverized, mixed with aluminum powder, and made into two ≈355 mg U-232 matrix element compacts for chopper cross section measurements (see section 2.3 below). Alpha pulse analysis of the purified uranium showed practically no Th-228 nor other daughter alpha activities.

About 60% of the Th-228 daughter activity originally present in the U-232 was isolated from the 6 - 8M HCl eluate from the anion column. This solution bubbled continuously due to radiolysis of the water. The solution was evaporated to about 1/4 its original volume, then excess 12N NaOH was added to precipitate Th(OH)₄ and to dissolve the aluminum as soluble aluminate. Attempts to filter the alkaline solution were unsuccessful, so the solution was centrifuged even though radiolytic decomposition of the water resulted in some stirring up of the Th(OH)₄ ppt. The precipitate was washed with NaOH solution and H₂O, and then dissolved in ≈ 3N HCl. The HCl solution was filtered, diluted to ≈ 1N HCl, and heated on a water bath. Saturated oxalic acid was added to precipitate Th-228 oxalate. The white thorium oxalate was filtered into a ceramic filter crucible lined with ashless filter paper, dried, and then ignited to ThO₂ at ≈ 800°C. A total of ≈ 2.9 mg of Th-228 O₂ was recovered and will be made into a pressed ThO₂-Al chopper sample. With the aid of this sample it should be possible to state conclusively which resonances observed in the U-232 samples are actually due to the Th-228 daughter which had grown into the U-232.

An alpha pulse spectrum of the isolated Th-228, taken after all the descendant activities had grown in, showed a clean separation from U-232 and the expected alpha peaks of Th-228, Ra-224, Rn-220, Po-216, Be-212, and Po-212. An alpha spectrum taken on an evaporated solution plate, is shown in Figure II-3.

The radiolysis of water due to the high alpha active solutions complicated the separations of U-232 and Th-228 by stirring up precipitates and causing columns to gas, but did not cause any serious trouble. However, U-232 and Th-228 represent very serious contamination potentials because of the Rn-220 gas and because the recoiling decay products have short half-lives. Chopper blocks, which have been only moderately contaminated, cannot be easily decontaminated apparently because the daughter recoil atoms are imbedded in the metal. Furthermore, Rn-220 is sufficiently soluble in organic material so that alpha activity will "diffuse" through rubber gloves and plastic sheets. The Rn-220 also passes through absolute filters, thus resulting in contamination (short-lived) of the cave exhaust system and control lines and syringes used for transferring solutions and for filtering precipitates in the cave. In order to prevent the escape of any of this activity into the room, all control lines were vented to the cave or to the hood exhaust system.

2.3 Total Neutron Cross Section of U-232 (J. R. Berreth, M. S. Moore, O. D. Simpson)

The total neutron cross section of U-232 was measured from 0.01 to 200 eV by use of the Materials Testing Reactor (MTR) fast chopper. Knowledge of the low-energy total cross section of U-232 is of importance to the Th-232/U-233 breeding cycle. It also is important to the study of the systematics of fission.

Fig. II-3 Alpha spectrum of Th-228 in equilibrium with decay products.

The U-232 was prepared by reactor irradiation of Pa-231 and subsequent chemical separation of the U-232 produced. The isotopic composition after separation of the uranium was as follows: U-232, 99.24%; U-233, 0.72%; traces of U-235 and U-238.

The nuclide U-232 has a half-life of 73.6 years and has as its daughter, 1.9-year Th-228. The latter nuclide is the head of the radioactive chain which extends to Tl-208; the longest-lived daughter in this chain is 3.6-day Ra-224. As can be readily deduced from these considerations, U-232 presents a difficult handling problem after a short time.

Due to the large quantities of alpha emitters as well as beta and gamma emitting nuclides associated with the U-232 and its decay products, it was necessary to prepare the sample for chopper measurements in the MTR hot alpha cave [7]. In order to prepare a uniform sample for cross section measurements, U₃O₈ powder was thoroughly mixed with aluminum powder and pressed at 70,000 psi to form a solid compact. This sample then was placed in a doubly-sealed sample holder for cross section measurements on the MTR fast chopper.

Figure II-4 is the total cross section as a function of energy from 0.01 to 200 eV. Strong resonances were observed below 100 eV in U-232 at 5.95, 12.68, 20.9, 23.8, 27.7, 43.2, and 75 eV. The resonance at 1.88 eV is due to a contaminant, probably Th-228, which had grown into the sample prior to measurement. The off-resonance cross section is unexpectedly large, ≈ 55 barns;

Fig. II-4 The U-232 total neutron cross section from 0.01 to 200 eV.

the reason for this large cross section has not yet been satisfactorily explained. A total cross section of 160 barns at 0.0253 eV is obtained if a cross section of 12 barns is assumed for the valleys.

Variation in the widths of the resonances suggests that fission is occurring. Figures II-5 and II-6 show a preliminary single level Breit-Wigner fit to the 5.95 eV and the 12.68 eV resonances. Comparison of these resonances indicates the presence of destructive interference between the two levels due to

Fig. II-5 Single level Breit-Wigner fit to the 5.95 eV resonance of U-232.

Fig. II-6 Single level Breit-Wigner fit to the 12.68 eV resonance of U-232.

fission. The resonance parameters are shown in Table II-1 for the first five resonances. A radiation width of 47 mV is assumed for these resonances based on a comparison of the data with that of James [8], who has been conducting fission cross section measurements on U-232. It is noteworthy that the average fission width is ≈ 6 times larger than the average fission width for U-235.

TABLE II-1
RESONANCE PARAMETERS FOR THE FIRST FIVE LEVELS OF U-232

E_0 (eV)	Γ_n^0 (mV)	Γ_γ^8 (mV)	Γ_f (mV)
5.945	0.61	47	23
12.656	1.75	47	253
20.95	0.55	47	753
23.71	1.20	47	93
27.65	0.75	47	453

Note: Assumed values

2.4 Beryllium Gases Experiment (R. L. Tromp)

Swelling tests were made at 400, 500, 600, and 700°C on separate samples from piece "A", the largest of the three beryllium fragments recovered from the MTR flash evaporator and believed to have broken off reflector piece F-2. (Previous fusion analyses indicated a reaction-gas content of approximately 30 cc/cc Be, which corresponds to a calculated fast nvt of $\approx 1.5 \times 10^{22}$ neutrons/cm².) The swelling results are plotted in Figure II-7 and tabulated in Table II-2.

Fig. II-7 Swelling behavior of sample "A", F-2 reflector.

These samples show no evidence of swelling at either 400 or 500°C. At 600°C a slow, fairly constant decrease in number density occurred from 1.85 to 1.75, with no significant evolution of reaction gas. At 700°C the swelling appears to have taken place entirely within the first two hours. Appreciable amounts of reaction gas were collected during all heating periods, although the evolution rate decreases with time.

Fusion-analysis of beryllium samples from the various segments of MTR lattice piece LB-15 also was carried out. From the middle (midplane) segment, sample M-12 yielded 36.5 cc total gas and sample M-14, 35.6 cc. Component analysis was completed only for M-12 showing:

He ⁴	91.3%	He ³	0.4%
H ¹ ₂	1.9%	H ³ ₂	6.0%

TABLE II-2

SWELLING OF IRRADIATED BERYLLIUM RECOVERED FROM MTR

Swelling Data			Evolved Gas Data				
Temp	Cumulative Heating Time (hr)	Be Density	Total Gas (cc/cc Be)	Volume Composition (%)			
				He ⁴	H ¹ ₂	H ³ ₂	He ³
400°C	0	1.8275	0.32	2.4	96.0	0.6	0.2
	6						
	12						
500°C	0	1.8345	0.27	0.6	97.2	1.9	0.3
	2	1.8293					
	6	1.8140					
	12	1.8238					
	20	1.8204					
600°C	0	1.8566	0.22	3.5	94.1	2.3	0.2
	2	1.8403					
	6	1.8122					
	12	1.7987					
	20	1.7473					
<u>Gas collected after each heat period</u>							
700°C	0	1.8458					
	2	1.5995	2.26	60.1	8.7	30.2	1.0
	6	1.6057	2.46	57.3	20.9	20.0	1.0
	12	1.5571	1.60	26.1	71.2	1.8	0.4
	20	1.6014	1.18	24.6	68.5	1.0	0.4
			7.50				

The high He⁴ content indicates that the gas is essentially all-reaction gas with no appreciable yield from extraneous sources. This would be expected since sample M-12 was cut from the interior of the middle segment; thus, there was no outer surface present to absorb larger quantities of H¹ from water recoil or other sources. The total gas content of roughly 35 cc/cc Be corresponds to a calculated fast nvt of 1.80×10^{22} n/cm² for this area of LB-15. Estimated nvt for the sample (from the irradiation history and MTR flux figures) was 1.02×10^{22} n/cm².

In connection with a proposal by ORNL to recover Be¹⁰ from highly irradiated MTR beryllium, irradiation histories of the various lattice "LB" pieces were again brought up to date, while thermal nvt totals were computed. Several pieces were found to have reached thermal nvt's (at midplane) equivalent to from 600 to 900 ppm of Be¹⁰ (assuming that the Be⁹ cross section for capture is 0.01 barn).

3. INELASTIC SCATTERING PROGRAM (R. E. Schmunk)

The velocity selector made the transition from operation at 7500 rpm with all rotors to operation at 12,000 rpm with choppers and 6000 rpm with rotating collimators. With the increased speed, an improvement of about 60% in time and energy resolution is realized.

4. LOW-ENERGY NUCLEAR PHYSICS-DECAY SCHEME STUDIES (R. L. Heath, C.W. Reich)

4.1 Level Structure of Odd-Odd Nuclei (R. G. Helmer)

A series of studies was undertaken to study the level structure of odd-odd nuclei by means of the radioactive decay of even-even nuclides. At various times in the future these studies will be reported. As an introduction to all of these studies in general and the decay of Pt-200 in particular, the following few paragraphs have been included.

One of the basic goals of nuclear physics is and always has been to determine the nature of elementary particles and the interactions between them. The problem of the interaction of elementary particles, and especially nucleons, has been studied extensively by means of scattering experiments. For the scattering of nucleons from very light nuclei, the angular distribution and cross sections should be related directly to the nucleon-nucleon interaction.

Another approach to the nucleon interactions is through the study of nuclear structure. However, in this case the difficulties are compounded by the inherent complexity of a many-body system. Because of insufficient knowledge to attack the problem directly, it has been expedient to formulate phenomenological theories of "models" of the nucleus. These models attempt to "explain" the experimental data in terms of a few ad hoc rules. Eventually these rules and models must be derived from physical laws; but in the meantime the models may provide some insight into the physical laws from which they arise.

Currently nuclei are discussed in terms of two complementary models, namely, the shell model or single-particle model and the collective model. In either case a nucleus is considered to be made up of a "core" with a small number of nucleons moving in orbits about the core. The properties of the nucleus are then deduced from those of the individual particles outside the core and the properties of the core. That is, the features of the individual nucleons in core are not considered, except that the properties of the core must result from those of the particles.

The shell model had its major success for nuclei which have proton and/or neutron numbers near 2, 8, 20, 50, 82, and 126. These numbers represent the number of particles for filled nuclear shells. For large numbers of nucleons outside these shells (eg, $150 < A < 190$ and $A > 225$), the collective model had a great deal of success in interpreting and predicting nuclear properties. In these regions the nucleus is considered to be deformed into a spheroidal or ellipsoidal shape from the spherical shape it has for the closed shell nuclei.

For a very simple form of collective model a deformed even-even nucleus can be considered as a core with no outside particles. Then the observed properties (energy levels, transition probabilities, etc) represent the various modes of excitation of this core. Similarly, odd-mass nuclei can be treated as consisting of an even-even core plus one particle outside the core. Then the nuclear properties are expected to result from excitation of the single particle, or the core, or a combination of the two. The remaining nuclei are the odd-odd nuclei which can be considered as an even-even core plus one neutron and one proton. The nuclear properties may then represent the excitation of the core or either of the two individual nucleons. A feature of this last case is that the observed properties may depend on the interaction of the two odd particles. If this is the case, the properties of odd-odd nuclei may be of interest in that they may yield some information about the neutron-proton interaction. The level structure of $^{67}\text{Ho}-166$, which was discussed by Helmer, Burson [9], and Newby [10], appears to be an example of this phenomenon.

For nuclei which can be described by the shell model the comments in the last paragraph are applicable. In other cases one may have to consider the coupling of several nucleons outside of the core and these simple arguments will not apply. At the ends of the regions of deformed nuclei these descriptions of nuclei do not suffice and these simple arguments may not hold.

Since it appeared from these considerations that there might be some special interest in the level structure of odd-odd nuclei, the author began a few years ago to study the level structure of these nuclei by observing the decay of radioactive even-even nuclei. Reports on the decay of $^{66}\text{Dy}-166$ and $^{68}\text{Er}-172$ have been published [9, 11]. Further experiments have been undertaken to determine the problems associated with the studies of the decay schemes of other even-even nuclei. The studies will be concerned with nuclei above mass 150.

Radioactive even-even nuclides can be produced in a reactor only by double neutron capture in stable even-even nuclei. At the end of the irradiation the activity of the double-capture product will be small compared to that of almost any single-capture activity. Therefore, in most cases it is to be expected that the presence of other radioactive isotopes will prevent the observation of spectra of interest. In these cases the experiments carried out and the difficulties involved will be discussed.

The following report on the attempt to observe the gamma-ray spectrum of Pt-200 is the first of this series.

4.2 Attempt to Measure the Gamma-Ray Spectrum of Pt-200 (R. G. Helmer and J. A. Dunbar)

4.21 Introduction. The production of Pt-200 by double neutron capture in Pt-198 was reported by Roy et al [12]. The nuclides involved are tabulated on the following page. The properties of the Au-200 were reported earlier [13]. Roy et al [12] proved the presence of the Pt-200 activity by measuring the decay of the Au-200. At various times after the activation the gold fraction was extracted from the platinum and counted. The apparent half-life of the Au-200, ie, the half-life of Pt-200, was found to be (11.5 ± 0.5) hr. No information was reported on the radiations emitted in the decay of Pt-200.

The samples used in this study consisted of 0.2 to 2.3 mg of platinum metal enriched in Pt-198 (61% Pt-198, 26% Pt-196, 9% Pt-195, and 4% Pt-194). Each sample was sealed in a quartz ampoule and irradiated for about 12 hr in the VH-2 facility of the MTR. Chemical purifications were necessary in order to remove Au-198 and Au-199.

The following chemical procedure was used in the purification process. The quartz ampoule was opened and the Pt powder washed from the ampoule with distilled water. A mixture of three parts concentrated HCl and one part concentrated HNO₃ was then added to dissolve the Pt metal. The solution was evaporated to dryness, moistened with concentrated HCl to assure conversion to chloroplatinic acid, and again evaporated to dryness.

The residue was taken up in 1 N HCl, 10 mg of Au carrier added as chloroauric acid, and the gold extracted with an equal volume of ethyl acetate. An additional 10 mg of Au carrier was added and the extraction repeated. A total of four extractions to remove Au were made in this way.

Several drops of hydroxylamine hydrochloride solution were added to destroy any nitrates, and stannous chloride crystals were added to the Pt fractions to reduce the platinum. The red PtCl₄= complex was then extracted into 10 ml of ethyl acetate and washed twice with equal volumes of 6N HCl, after which, the wash was discarded.

Several ml of 1N HCl were added to the ethyl acetate phase and the ethyl acetate evaporated off by heating. The Pt fraction, now contained in 1N HCl, was diluted to $\approx$ 20 ml; 10 mg of La³⁺ were added and a La(OH)₃ scavenging precipitation was made by adding slight excess of 1 N NaOH solution. The Pt-containing supernate was re-acidified with HCl and a second La(OH)₃ scavenging carried out.

The supernate from the second La(OH)₃ scavenging was acidified with HCl and 1N NH₄NO₃ was added dropwise to precipitate (NH₄)₂(PtCl₆). The latter precipitate was filtered off, washed with alcohol, dried and mounted for counting.

Gamma-ray spectra were measured with a scintillation spectrometer consisting of a NaI(Tl) crystal 3 in. in diameter by 3 in. long, photomultiplier, preamplifier, A-8 amplifier, and a 512-channel pulse-height analyzer. The source was positioned along the axis of the crystal, with the source-detector distance determined by the source strength. A beryllium absorber (areal density = 1.18 g/cm²) was used to prevent electrons from reaching the crystal

(Figure II-8). In some cases a tantalum absorber (600 mg/cm²) also was used to reduce the relative intensity of the low-energy gamma rays and thereby permit the use of more intense sources.

4.22 Experimental Results. In order to verify the presence of Pt-200 and Au-200 in the purified platinum samples, gold was extracted from each of the platinum samples. The gamma-ray spectrum obtained from one such gold sample is shown in Figure II-8. The contributions from Au-198 and Au-199 are shown. The presence of these two isotopes indicates that the original purification of the platinum was not complete.

The Au-200 spectrum, after subtraction of the Au-198 and Au-199 spectra, is shown in Figure II-9. These data are consistent with the previous reports [11] that Au-200 decays primarily via coincident gamma rays of 370 and 1220 keV. The peak at 1600 keV results from coincident summing of these two radiations. This effect is especially pronounced because the source was only about 0.8 cm from the crystal. The solid lines in the figure represent the

Fig. II-8 Gamma-ray spectrum of gold fraction.

Fig. II-9 Gamma-ray spectrum of Au-200. Energies are in MeV.

calculated detector response [14] for gamma rays of 370 and 1220 keV and the corresponding calculated sum spectrum [14]. The gamma-ray relative intensities resulting from this analysis are shown in Table II-3 along with those of previous reports [13]. The gamma-ray energies were determined by comparison of pulse heights of the photopeaks with that of the 0.6616-MeV gamma ray from the decay of Cs-137. The calibration data in Figure 10 of reference 14, were used to obtain the energies.

Two of a set of gamma-ray spectra from a purified platinum source are shown in Figures II-10 and II-11. These two spectra were taken approximately 14 hr and 6 days after the irradiation. Therefore, the second spectrum shows only the longer lived platinum activities. The estimated contributions from the

TABLE II-3

GAMMA-RAY ENERGIES AND RELATIVE INTENSITY IN THE DECAY OF Au^{200}

Energy (MeV)	Intensity		
	Roy et al [13]	Girgis et al [13]	These data
0.368 ± 0.003	100	100	100
1.237 ± 0.010	100 ± 5	80 ± 8	80 ± 10
1.60	<5	2 ± 1	----

Fig. II-10 Gamma-ray spectrum of purified platinum sample about 14 hr after irradiation.

Fig. II-11 Gamma-ray spectrum of purified platinum sample 6 days after irradiation.

various platinum isotopes are shown in the figures. These activities (and half-lives) are Pt-191 (3.0 day), Pt-195m (4.1 day), Pt-197 (20 hr), and Pt-197m (1.3 hr). The only platinum isotopes that are not shown are Pt-199 (30 min) which is presumed to have decayed to a negligible amount, Pt-193 (<500 yr), which has no reported gamma rays, and Pt-193m (4.4 day), which has only a 136-keV gamma ray that would not be distinguished from the Pt-195m radiations.

In addition to these two spectra, others were obtained at about 16 hr, 23 hr, and 32 hr, after the irradiations. A comparison of these spectra did not reveal the presence of any radiations with a 10-hr half-life.

In Figure II-12 is shown the spectrum from another sample taken 12 hr after the irradiation. In this case the Au-200 contribution is discernible.

4.23 Discussion. From the data in Figure II-12 and the reported [13] decay scheme of Au-200, it is possible to estimate the decay rate for the Pt-200 present in the source. From this decay rate, the maximum intensity can be calculated as a function of energy of any gamma ray in the Pt-200 spectrum. This calculation leads to the conclusion that due to the presence of the intense Pt-197 spectrum it would not be possible to observe any gamma rays from the Pt-200 below about 300 keV. Between 300 and 600 keV, the gamma rays would have to represent at least 20% of the decays to be observed; and above 600 keV, 10% of the decays.

These data indicate that a meaningful study of the gamma-ray spectra of Pt-200 will not be possible until it is possible to produce a source in which the relative amount of the Pt-197 activity is reduced by a factor of the order of 100. In the future, this reduction might be possible by the isotopic separation of the sample after activation. The spectrum in Figure II-12 differs from that in Figure II-10 primarily in that it was taken with a source four times as intense. The cross-hatched region, which does not appear in Figure II-10, decays with a period of about 10 hr. However, these counts apparently result from random summing of two gamma rays ($191 + 191$ or $191 + 269$). Since the intensity of the random sum spectrum is proportional to the square of the counting rate, it has an apparent half-life (10 hr) one-half that of the half-life (20 hr) of the activity producing it.

4.24 Decay Scheme. From nuclear systematics and the decay scheme of Au-200, it is possible to determine a few features of the Pt-200 decay scheme. The data on the decay of Au-200 indicate that the spin and parity of the ground state of $^{79}\text{Au-200}$ are 0^- or 1^- . Since the $^{78}\text{Pt-200}$ is an even-even nucleus, its ground state is presumed to have $I^\pi = 0^+$.

The beta decay between these two states then would be an ordinary first-forbidden transition. The log ft value of this transition then would be expected to be between 6 and 8. From the systematics of beta-decay energies, the total decay energy for $\text{Pt-200} \rightarrow \text{Au-200}$ was estimated [15] to be 0.5 to 1.0 MeV. For any decay energy in this range, the log ft value will be between 6 and 8 if the branching to the ground state is more than about 5%. Therefore, it is

Fig. II-12 Gamma-ray spectrum of purified platinum sample 12 hr after irradiation. The cross-hatched area represents the contribution from accidental summing.

reasonable to expect some beta decay to excite states in Au-200 if low-energy states exist with spins of 0 or 1.

4.25 Conclusions. No radiation from Pt-200 was observed in the platinum samples produced. It will be necessary to obtain sources in which the relative Pt-197 intensity is reduced by at least a factor of 100 before definitive data on the Pt-200 gamma-ray spectrum can be obtained. However, the information available on the decay of Pt-200 indicates that the decay scheme may involve some excited states and, therefore, may be of interest.

4.3 Extension of Variance Minimization Studies to Include $P_6(\cos \theta)$ and $P_8(\cos \theta)$ Terms in Angular Correlation Functions (J. A. Merrill and C. W. Reich)

The problem of determining the uncertainties to be associated with estimated coefficients in directional-correlation functions is important in the studies of properties of low-lying nuclear states. Certain problems concerning the estimation of the variances of directional-correlation coefficients have been studied at this laboratory and the results have been reported [16, 17, 18]. Since the interest was directed toward standard gamma-gamma directional correlation measurements, these previous studies were limited to functions in which the highest order Legendre polynomial included was $P_4(\cos \theta)$.

Some interest in correlational functions which might possibly contain P_6 and P_8 terms has recently arisen and, therefore, it seemed appropriate to extend the studies to include those terms. In these extended studies, we have considered both the proper choice of angles as well as the appropriate relative frequencies of observations at those angles to minimize the sum of variances associated with the estimated coefficients (or the trace of the inverse of the normal equation matrix). This report summarizes the results thus far obtained.

Since the method employed in the original study to determine the best relative frequencies of observations would have led to prohibitively unwieldy algebraic relationships, even in the first extension to include a P_6 term, it was necessary to adopt a modified approach. This modified approach consisted of treating the k angles plus $k-1$ of the relative frequencies of observations as the variables in a statistically designed experiment. The k^{th} relative frequency was always defined by the relationship $f_k = 1 - \dots - f_{k-1}$. The particular statistical design chosen for this work was the orthogonal composite design. The basic portion of such a design is a 2^{2k-1} -factorial design (or a half replicate of the full factorial) which provides the necessary information to determine the linear effects of the experimental factors (angles and relative frequencies) as well as all of the two-factor interactions ($\theta_i f_j$). In the composite design, sufficient additional experimental runs are required to provide the necessary information so that quadratic effects for the factors can be estimated. As in the original study, the statistical weights for observations at the various angles were all assumed to be unity. For this modified approach, the calculational procedure was as follows:

- (1) A fixed total number, N (100 was used), of observations was distributed uniformly among k equally spaced angles ($k = 4$ when only the P_6 term was added and $k = 5$ when both P_6 and P_8 terms were added).

(2) The k angles plus $k-1$ of the relative frequencies of observations at the angles were treated as the variables in an orthogonal composite design.

(3) The response R (defined as the sum of the variances of the expansion coefficients) was calculated for all of the factor level combinations required in the composite design.

(4) Since a composite design does provide sufficient information, a quadratic polynomial approximation (including all two factor interactions) was fitted to the response data.

(5) The minimum of this polynomial approximation was found employing the usual methods of the calculus.

(6) A comparison of the position of the center of the composite design and the point corresponding to the minimum of the quadratic response function provided information about both "direction" and "distance" to reach a point of minimum response.

(7) Points along the path joining the two above points were examined and the point having the smallest response was chosen as the center for a new composite design.

This procedure was repeated until significant reductions in the response could no longer be realized.

The results obtained for the extension to include $P_6(\cos \theta)$ and $P_8(\cos \theta)$ terms in the angular correlation model are shown in Table II-4. For comparison purposes, the cases of ten equally spaced angles having ten observations at each angle are included.

TABLE II-4
RESULTS OF VARIANCE MINIMIZATION STUDIES

Frequency	Angle	C ₁₁	C ₂₂	C ₃₃	C ₄₄	C ₅₅	Response ($=\sum_{\lambda} C_{\lambda\lambda}$)
<u>Including $P_6(\cos \theta)$ terms</u>							
0.1	90°						
0.1	100°						
-	-						
-	-						
-	-	0.01255	0.04660	0.08063	0.09238	---	0.23216
0.1	180°						
0.2633	90°						
0.3187	121.86°	0.01122	0.04356	0.06060	0.05360	---	0.16898
0.2773	148.52°						
0.1407	180°						
<u>Including $P_6(\cos \theta)$ and $P_8(\cos \theta)$ terms</u>							
0.1	90°						
0.1	100°						
-	-						
-	-	0.01270	0.04662	0.08661	0.11900	0.12349	0.38842
-	-						
0.1	180°						
0.2026	90°						
0.2552	114.75°						
0.2509	134.95°	0.01112	0.04532	0.07467	0.08859	0.07250	0.29220
0.1930	155.49°						
0.0983	180°						

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III. INSTRUMENT DEVELOPMENT

1. DETECTOR DEVELOPMENT

(S. D. Anderson)

1.1 Dynamic Pressure Generator (R. L. Kindred)

A pulse circuit was developed for the purpose of generating underwater explosions for dynamic testing of pressure transducers. In operation the energy of a spark gap controlled capacitor discharge is used to explode a small diameter wire in a water tank.

Figure III-1 is a photograph of the pulse circuit with the housing cover open. The capacitor serves as a mounting for all of the components in the housing and is located in the housing by two angle brackets. The capacitor is rated at 10 kV with a measured energy capacity of 1675 joules, an equivalent series inductance of 0.040 microhenrys, and a ringing frequency of 150 kc. The triggered spark gap consists of two 1.25-in. diam brass hemispheres and a stainless steel trigger needle which are adjustable to accommodate a voltage range of 2 to 10 kV. One hemisphere is mounted on the capacitor stud. The other hemisphere is mounted on a polystyrene bracket and is connected to the inner braid of a triaxial cable which is the current path to the exploding wire. The other braid is used for the return path. This cable arrangement is used to reduce the cable inductance. A current measuring resistor of 0.00243 ohm is connected between the outer braid and the capacitor ground. An oscilloscope sweep trigger signal is provided to facilitate the measurement of circuit and pressure parameters.

Safety features include a manual capacitor shorting switch, a manual interlock on the housing cover, an audible alarm during the capacitor charge time, a capacitor voltage monitoring meter, and programmed relays to shut off the high voltage and to shunt the power supply and capacitor to ground after firing is complete.

A separate control unit, not shown, furnishes charge, fire, programmed safety relay, and misfire signals to the pulser circuit. Indicators show charge and fire conditions and also the location of the manual capacitor shorting switch.

Operation of the circuit is as follows. A conventional 10 kV power supply is used to charge the capacitor through a charging resistor. When the capacitor is charged, the trigger needle voltage is 1/2 of the capacitor voltage due to a voltage divider across the gap. Since the needle is positioned approximately in the center of the gap, the gap voltage field is not disturbed. When the trigger firing switch is closed, a parallel capacitor and resistor circuit connected to the needle is grounded. The needle is momentarily at ground potential and the full gap voltage exists between the high side of the gap and the needle. Voltage breakdown and ionization occur in this region. The combination of ionization and a rapid rise in needle voltage creates a breakdown to the low side of the gap. In actual practice the needle is positioned somewhat closer to the high side hemisphere.

Peak current for a 10 kV capacitor discharge is 42,400 amp with a 1/2 cycle duration of the discharge of 18.6 μ sec, and a rise time of 3.3 μ sec for a 17.9 mil

Fig. III-1 Photograph of pulse circuit with housing cover open.

diam, 0.75 in. long, copper wire. Peak voltage across the wire for the same conditions is 12 kV with a rise time of 2.2 μ sec.

The 10 kV capacitor discharge through this wire produces a peak pressure of approximately 2200 psi with a rise time of 0.32 μ sec at an axial location six in. from the end of the wire. For the same discharge, the peak pressure six inch from the wire in a plane normal to the wire is about 4600 psi with a rise time of 0.36 μ sec.

2. ELECTRONIC SYSTEMS INSTRUMENTATION (T. J. Boland)

2.1 Fail-Safe Level Trip Circuit for ETR Differential Pressure Monitor (K. F. Smith and J. B. Colson)

An investigation was made to determine the safety of the ETR differential pressure monitor, where transistor circuits have been in operation for over two years. The level trip circuit used is a Schmidt trigger which has proven very reliable. However, if one of the transistors was to fail the circuit could become inoperative and would not produce a scram signal when required. Therefore, two units were operated in parallel to increase the probability that at least one circuit will be in operation at all times.

Several dc level trip circuits were studied to determine their fail-safe properties. The circuit described in Section 2.2, "High Impedance dc Level Trip Circuit", has proven to be fail-safe. Referring to the simplified diagram shown in Figure III-2, the active element is a solid state device called a silicon controlled rectifier (SCR). Under normal operating conditions the SCR is gated on and the circuit becomes a simple half-wave rectifier power supply which holds the relay in. If the gate voltage is removed the relay will drop out producing an alarm or scram. If the SCR fails open, there will be no voltage across the relay, or if the SCR fails short, an ac voltage will be applied to the dc relay. Either condition will drop out the relay and hence produce an alarm or scram. Failures of the capacitor or power supply which makes the circuit inoperative also will drop out the relay.

Fig. III-2 A simple dc level trip circuit.

This circuit showed good stability characteristics and was recommended for replacing the Schmidt trigger circuits in the ETR differential pressure monitor.

2.2 High Impedance Differential DC Level Trip Circuit (J. B. Colson and K. F. Smith)

A simple high impedance dc level trip circuit was developed. A schematic drawing of the simplified circuit is shown in Figure III-2.

This circuit takes advantage of the fact that the silicon controlled rectifier (SCR) acts as a controlled halfwave rectifier of the ac power supply. It will block both the positive and the negative half cycle until the minimum gate firing voltage has been exceeded. When this occurs, the SCR will conduct during the positive half cycle and block during the negative half cycle. During the positive half cycle, the capacitor C will charge and the relay will be energized. During the negative half cycle when the SCR is blocking, the capacitor will discharge through the relay coil. By proper choice of the RC time constant of the relay and capacitor, the relay will be held in during the negative half cycle. Thus the relay will be held energized as long as the minimum gate firing voltage has been exceeded. Once this control signal is removed, the SCR will block both half cycles again since it automatically turns off at the end of each positive

half cycle. The relay will now drop out. Because the power supply to the SCR is floating, either A or B may be used as either the reference or the dc input depending on the polarity of trip required. If the relay is required to energize when the dc input voltage is increasing in the positive direction, the reference should be tied to point B and the dc input to point A. Alternately if the relay is required to energize when the dc input voltage is increased in the negative direction, the reference should be tied to point A and the dc input to point B. These two different types of connections are shown in Figure III-3. Note that in both of these cases, the only current which passes between the dc input and the reference is the current required to fire the SCR. Through the use of high gain SCR's this current can be held to less than one μa which represents a very high input impedance device.

(a) Relay energizes in positive direction.

(b) Relay energizes in negative direction.

Fig. III-3 Different methods for energizing relay.

The dc voltage required to trip the circuit will vary with temperature because the firing voltage of the SCR has a negative temperature coefficient of approximately three $\text{mV}/^\circ\text{C}$. If a uniform firing voltage over a wide range of temperature is required, it will be necessary to use some type of compensation. A very excellent method of compensation involves the use of a PNP (or an NPN for the alternate method) transistor connected as an emitter follower as shown in Figure III-4. Compensation is achieved because the change in V_{be} of the transistor will null out the change in V_{gc} of the SCR. The transistor also offers an additional advantage of isolation between the dc input and the SCR.

Fig. III-4 A temperature compensated dc level trip circuit.

Other methods for applying the reference signal are shown in Figure III-5. These methods employ a current source which develops a reference voltage across resistor R_1 . Note that the method shown in Figure III-5 does not require a floating ac supply; however, the alternate circuit does.

Fig. III-5 Methods for using a current source as a reference.

A practical circuit shown in Figure III-6 was built and tested. The floating ac power supply is a 10 kc oscillator whose output is a 40 volt peak to peak square wave. The maximum time before the relay could start to energize is 0.05 msec, half the period of the oscillator frequency. Therefore, the speed of response is limited by the relay, which will energize in approximately 5 msec. Where the fast response is not required, a stepdown transformer from the power line could be used. With a line frequency of 60 cps, the maximum delay before the relay could start to energize would then be increased to 8.3 msec. An additional disadvantage of operating at 60 cps is that the filter capacitor has to be increased from 0.1 μ f at 10 kc to 15 μ f at 60 cps.

Fig. III-6 A practical differential dc level trip circuit.

2.3 Thickness Indicator Modification (J. F. Swartz)

It was desired to use an available commercial coating thickness indicator in the hot cells to measure an anodized coat on aluminum. This application requires operation with the sensing probe located at least 15 ft from the instrument whereas the manufacturer recommends a maximum cable length of 32 in.

The indicator uses four frequencies, 0.1, 0.5, 2, and 6 megacycles (mc) with each frequency requiring a separate probe. Due to the inverse variation of depth of penetration with frequency, thicker coatings require the use of lower frequencies. Since the immediate application is the measurement of an anodized coating on aluminum over a thickness range of 1 to 2 thousandths on an inch (mil), the use of 6 Mc and its associated probe is required.

A simplified schematic diagram of the instrument, showing only one probe, is seen in Figure III-7. This shows that the probe coil L_p is part of a parallel tuned circuit.

Fig. III-7 Simplified schematic of the coating thickness indicator.

As the cable is lengthened beyond a few inches the impedance it presents to the unit varies drastically, changing from inductive to capacitive in the region of a cable length of one quarter wavelength (27 ft for 6 Mc in RG-22). The solution to the problem is to go beyond this region and use a half-wavelength cable. The unit then "sees" the same impedance that is presented to the cable by the probe.

A total cable length (RG-22) of approximately 54 ft was used in this application and, with slight adjustment of C_2 , readings within ± 0.2 mil were obtained on coating samples. It is believed that better accuracy than this can be obtained but the non-uniformity of available samples made a closer check impossible.

A method of applying this cable lengthening scheme to the lower frequency probes may be to use an artificial or lumped line to make up a portion of the half-wavelength, thereby eliminating the use of excessive amounts of cable.

2.4 ETR Level Preamplifiers (J. B. Colson, J. B. Thompson)

At the request of ETR Operations, the ETR level preamplifiers were tested and evaluated to determine their continued ability to perform satisfactorily. It was determined that, after approximately four years of operation, the end of their reliable service life had essentially been reached. Their service life had been shortened by operation at high ambient temperature.

New preamplifiers which will operate satisfactorily up to -85°C have been built to replace the old ones. A new high performance type operational amplifier designed specifically for military applications was used and a power supply with reactor grade components has been designed for this application. A completely new mechanical layout was used to facilitate cooling of the critical components within the preamplifiers.

These preamplifiers have now been operating for several weeks with no indication of trouble. However, final changes in the power supplies are to be made pending procurement of parts. It has been recommended that cooling fans be placed in the cabinets containing these preamplifiers to maintain a lower ambient temperature.

IV. APPLIED MATHEMATICS AND MACHINE COMPUTATIONS

1. MODIFICATION OF CURIE

(Dean Ritchie)

CURIE is a FORTRAN program written by Atomic International to compute a library of fission product isotopes during and after reactor operation. In checking the results of test runs made against the equations supplied with the code, discrepancies were encountered, which led to the following modifications.

Equations were derived which lend themselves more readily to digital methods. These equations are designed for the decay of a chain, given no external source of any isotope in the chain, but an initial number of atoms of each isotope, and the decay of a chain, given no initial atoms in the chain, but a constant source supplying the first isotope.

Generalizing to any number of isotopes, we may write

$$N_i(t) = S_1 \lambda_1 \lambda_2 \dots \lambda_{i-1} \sum_{j=1}^i \frac{1 - e^{-\lambda_j t}}{\lambda_j (\lambda_1 - \lambda_j) (\lambda_2 - \lambda_j) \dots (\lambda_{j-1} - \lambda_j) (\lambda_{j+1} - \lambda_j) \dots (\lambda_i - \lambda_j)}$$

where $N_i(t)$ is the number of atoms of isotope i at time t (assuming $N_i(0) = 0$) and S_1 is the number of atoms of isotope 1 supplied per unit time.

If more than one isotope is supplied, this equation must be generalized in the following way. Taking isotope k as the source isotope, for $i \geq k$, we have:

$$N_i(t) = S_k \lambda_k \lambda_{k+1} \dots \lambda_{i-1} \sum_{j=k}^i \frac{1 - e^{-\lambda_j t}}{\lambda_j (\lambda_k - \lambda_j) \dots (\lambda_{j-1} - \lambda_j) (\lambda_{j+1} - \lambda_j) \dots (\lambda_i - \lambda_j)}$$

The total number of atoms of isotope i is given by the number of atoms formed by decay from isotope 1, plus the number supplied by isotope 2, etc, ie

$$N_i(t) = \sum_{k=1}^p S_k \lambda_k \lambda_{k+1} \dots \lambda_{i-1} \sum_{j=k}^i \frac{1 - e^{-\lambda_j t}}{\lambda_j (\lambda_k - \lambda_j) \dots (\lambda_{j-1} - \lambda_j) (\lambda_{j+1} - \lambda_j) \dots (\lambda_i - \lambda_j)} \quad (1)$$

Where isotope p is the last isotope in the chain such that $p \leq i$ or $S_p > 0$.

In practice, it is assumed that $p=i$. This is done with no error, since $S_k = 0$ implies no contribution by decay from isotope k as a source. (Isotope k may be an intermediate step of decay, however).

Equation (1) is the equation used to calculate fission product inventory at shutdown time in the modified CURIE code.

In the original CURIE code, matching limitations forced the use of double-precision, whenever the reactor operation time was small. In the modification, the machine limitation encountered is as follows:

In the computation of

$$1 - e^{-\lambda_j t}, \text{ if } \lambda_j t \text{ were small, say } 10^{-8}, e^{-\lambda_j t} = 1 - 10^{-8} = 0.99999999$$

When this is subtracted from 1.0, 10^{-8} is the result, which appears as 0.00000001 in the machine. This result is then normalized to 1.0×10^{-8} , which is the desired result. However, had $\lambda_j t$ been 1.9×10^{-8} , the machine limitation would have returned an answer of 1.0×10^{-8} (the 0.9 would be truncated off).

This limitation was eliminated by a simple expansion of $e^{-\lambda_j t}$

$$e^{-\lambda_j t} = 1 - \frac{\lambda_j t}{1} + \frac{(\lambda_j t)^2}{2!} - \frac{(\lambda_j t)^3}{3!} + \dots$$

$$1 - e^{-\lambda_j t} = \lambda_j t - \frac{(\lambda_j t)^2}{2!} + \dots$$

Then, $\lambda_j t = 1.9 \times 10^{-8}$ leads to

$$1 - e^{-\lambda_j t} = 1.9 \times 10^{-8} - 3.61 \times 10^{-16}$$

which in the machine, is calculated as

$$\frac{1.9000000 \times 10^{-8} - 0.0000000 \times 10^{-8}}{1.9000000 \times 10^{-8}}$$

Therefore, double precision is not necessary. Whenever $\lambda_i t < 0.1$ the above expansion is used.

After the reactor is shut down, equation (2) is used for an initial number of atoms in each isotope, and the numbers given by equation (1) are used as the initial number of atoms. Before calculation, these numbers may be modified, isotope by isotope, by adding a fission product inventory from a previous period of reactor operation, and/or by multiplying by a fraction of 1 to simulate fractional release. After these modifications are made, the code considers the numbers in the library of isotopes to be $N_i(0)$ for the strict decay calculation.

$$N_i(t) = N_i(0) e^{-\lambda_i t} + \lambda_{i-1} N_{i-1}(0) \left(\frac{e^{-\lambda_i t}}{\lambda_{i-1} - \lambda_i} + \frac{e^{-\lambda_{i-1} t}}{\lambda_i - \lambda_{i-1}} \right) + \dots$$

$$+N_1(0) \lambda_1 \lambda_2 \dots \lambda_{i-1} \sum_{j=1}^1 \frac{e^{-\lambda_j t}}{(\lambda_1 - \lambda_j)(\lambda_2 - \lambda_j) \dots (\lambda_{j-1} - \lambda_j) \dots (\lambda_i - \lambda_j)} \quad (2)$$

Since the calculation of $e^{-\lambda_j t}$ is accomplished satisfactorily, no modification to this formula is necessary for small λt .

2. ANGULAR CORRELATION OF TRIPLE GAMMA-RAY CASCADE (G. A. Cazier, N. D. Dye)

A complex quadratic expression was programmed for the IBM 7090 which describes an angular correlation of triple gamma-ray cascades. The roots of the equations expressing the counting rate of the apparatus were obtained initially by using the Newton Raphson method of iteration. However it was found that for certain cases the first derivative of the function in the neighborhood of the root was very close to zero. A new approach to the problem showed that with a sizeable amount of algebraic manipulation the equations could be solved with a direct application of the quadratic formulas, thus yielding both roots simultaneously.

3. IBM 7090 FAP PROGRAM FOR PROCESSING BINARY MAGNETIC TAPE OUTPUT FROM A MULTICHANNEL ANALYZER (D. D. Metcalf, D. E. Shippen)

The Multichannel Analyzer recently installed at the MTR in the Physics Section writes binary records of 100 computer words each, 36 binary bits per word. Each computer word consists of two pairs of x,y coordinates which are the locations in a 256 by 256 grid. Each pair (x,y) gives the location of an event. The program tallies the counts for all 65,536 locations in the grid, in effect giving a three dimensional histogram. This requires "packing" in the core of the 7090 because the number of grid locations is double the number of core location in the 7090. The tallies are packed three to a computer word, and 8000 core locations are reserved for overflows. Each 1/3 of a computer word will accommodate 4096 counts, and when this value is exceeded, count for that grid element is reset to zero in the word segment. An overflow location then tallies the overflows for this particular grid element. Without such packing, three machine passes would be required to process the tapes.

Output from the core is a binary tape that is processed by a FORTRAN subroutine which prints out the spectra according to x indices, and then y indices. The format for the print-out is identical to the format for all gamma spectrum currently being processed.

The output record is specified as binary in order that tapes may be added together, element for element, over a portion of two output tapes, or the whole of two output tapes that are of identical length. This is necessary for instances of accumulations where the number of overflow locations exceeds 8000. When this occurs, the counts in core are dumped out on tape, the core area cleared,

and accumulation of counts resumed. Forms other than binary cannot reenter the computer for further computer operations.

4. GAMMA-RAY PHOTOPEAK PARAMETERS

(G. A. Jayne)

For use in the generation of spectra of monoenergetic gamma rays, it is necessary to interpolate the values of α_1 and α_2 for the desired gamma ray energy or value of x_0 . This can be done most efficiently by representing α_1 and α_2 as functions of x_0 . Helmer (IDO-16857) found that a polynomial of the form

$$\text{Log } \alpha = a_0 + a_1 (\log x_0) + a_2 (\log x_0)^2 + a_3 (\log x_0)^3 + \dots$$

suited the need very well. An IBM 7090 program was written to evaluate these polynomials for α_1 and α_2 as well as calculating the area under the photo peak.

5. CYLINDRICAL RADIATION DETECTOR EFFICIENCY

(G. A. Jayne)

A FORTRAN program was written for the IBM 7090 to calculate the efficiency of right, circular, cylindrical NaI(T) crystals in detecting gamma rays emitted from a source centered on the extended crystal axis. The sources considered were a point source, a line source perpendicular to the end of the crystal, and a thin disc source whose plane is perpendicular to the end of the crystal and whose radius is less than the radius of the crystal. Input parameters for any given problem are the radius and height of the crystal, type and size of the source, distance between the crystal and the source, and the absorption cross section associated with the energy level of the gamma rays.

6. GAUSS-HERMITE NUMERICAL INTEGRATION

(Dean Ritchie)

An IBM 7090/7040 subroutine for the evaluation of integrals of the form

$$\int_{-\infty}^{\infty} f(x) e^{-a^2 x^2} dx$$

was written and checked out. The Gauss-Hermite method of numerical integration is used with up to 20 mesh points available. A significant saving of computer time was achieved in the evaluation of the integrals which appear in the CLOUD program.

7. GENERATION AND TESTING OF RANDOM NUMBERS

(L. J. Gannon and L. A. Schmittroth)

In connection with the general purpose Monte Carlo program, PMC, being written here for the IBM 7040, an extensive test of random number generators was conducted. The field was narrowed to two methods. The multiplicative congruential

$$x_{n+1} \equiv \lambda x_n \pmod{P} \quad (1)$$

where $P = 2^b$, and b is the length of the computer word in binary bits (in this case $b = 35$). The second method tested was the mixed congruential method,

$$x_{n+1} \equiv \lambda x_n + \mu \pmod{P} \quad (2)$$

In order to determine the most satisfactory parameters λ , or λ and μ , each method was subjected to the following statistical tests:

(1) Frequency Test. This tests the uniformity of the distribution on $0 \leq x \leq 1$.

(2) Matrix Test. This tests for undue correlation between elements of the sequence.

(3) Auto correlation. This also is a test of the independence of the elements.

(4) Frequency of Length of Runs. An occurrence of k consecutive numbers forming a monotonic increasing (or decreasing) sequence which is broken by the $(k+1)$ st number is called a run of length k . The test compares observed frequencies of length of runs with the theoretical expected values.

(5) Frequencies in Subsequences. In the auto correlation test the main concern was the correlation between every j^{th} number. Below is a discussion of the subject and another test for randomness. In the following example, suppose that the coordinates (x_i, y_i, z_i) of a large number of points uniformly distributed throughout the unit cube are desired. Then it is necessary to use one of the random number generators to generate a sequence of random numbers r_i ($0 < r_i < 1$) and assign the desired coordinates these values, ie $x_1 = r_1$, $y_1 = r_2$, $z_1 = r_3$ and $x_2 = r_4$, $y_2 = r_5$, $z_2 = r_6$, etc. This is the type of application mentioned earlier where every j^{th} number (in the example $j = 3$) is associated with a given variable. Thus it is visible that difficulties are encountered if the subsequences of a given sequence are not random. In this example the points would not be uniformly distributed.

To test for this randomness of subsequences, it was necessary to set up j tables, each with K subintervals, and tally every j^{th} number of the sequence in the j^{th} table as was done in the frequency test.

It was found that the length of run test is the most powerful of the foregoing. It rejected more of the generators than any other test, and rejected all generators that were rejected by any of the other tests.

One striking result of these tests is that for all cases tried, the straight multiplicative method is superior to the mixed method. Of those generators tested, the multiplicative method with $\lambda = 2^{18} + 3$ was the most satisfactory.

V. PUBLICATIONS OF MTR-ETR TECHNICAL BRANCHES

PAPERS PRESENTED AT MEETINGS

F. D. Terry, S. D. Anderson, Effects of Transient Nuclear Radiation on Transducers and Electrical Cable, ANS Summer Meeting, Salt Lake City, Utah, June 18, 1963.

N. Wilde, Generation of Poisson Time Distributed Random Pulses, IEEE Electro Nuclear Conference, Richland, Washington, April 30, 1963.

IDO REPORTS ISSUED

S. R. Gossman, A Transfer Function for D. C. Operational Amplifiers with Generalized External Networks, IDO-16809, (February 25, 1963).

G. E. Putman, D. M. Shapiro, (Internuclear Co., Inc., Clayton, Mo.), MIST (Multigroup Internuclear Slab Transport), for Phillips Petroleum Co., U. S. A. E. C. Division at Idaho Falls, Idaho, IDO-16859 (May 10, 1963).

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