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July 1 - September 30, 1966

IDAHO NUCLEAR CORPORATION

NATIONAL REACTOR TESTING STATION

IDAHO FALLS, IDAHO

U. S. ATOMIC ENERGY COMMISSION

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SUMMARY

Values for β/Λ obtained recently in the ATRC using reactor noise techniques are reported, and the procedures for data analysis currently in use are described. Effects of flux trap experiments and outer shim positions on the ATRC power distribution are described.

The importance of motion of ATR fuel assemblies induced by the operating conditions was evaluated. Although significant motion was calculated, the geometrical changes that result do not appear detrimental.

Hydraulic testing of production ATR fuel elements was continued. Inadequate roll-swage-joint strength appears to be the cause of the premature failure of one fuel assembly.

From a study of the physical, chemical, and metallurgical changes which take place in various fuel systems during irradiation, some of the fuel stability variables which reduce uniform fission product swelling and lower the accelerated swelling threshold temperature have been identified. Large differences in uniform swelling rates and in accelerated swelling temperatures from one system to the next suggest that control of these variables could result in greater fuel stability. The incorporation of initial porosity to accommodate uniform swelling from fission improves the irradiation performance of aluminum plate-type elements.

Type 2219 aluminum alloy clad fuel plates have been corrosion tested in-pile under ATR conditions. Type 2219 aluminum in both the annealed (-0) and precipitation-hardened (T-62) condition appears superior to Type 6061-0 aluminum for application in the ATR.

UAl alloys and intermetallics appear to be compatible with liquid lead for short periods (≈ 72 hours) at 600°C . For longer periods, varying reaction and lead penetration occur.

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I. REACTOR ENGINEERING

1. CRITICAL FACILITIES (E. E. Burdick)

1.1 Measurements of β/Λ in the ATRC Using Reactor Noise Analysis (N. C. Kaufman, R. P. Morrell [a])

In January of this year, measurements of the ATRC power spectral density were performed by the Systems Analysis Group of Phillips Petroleum Company in order to obtain values for β/Λ , the ratio of the effective delayed neutron fraction to the effective neutron generation time. These measurements were performed in the 39-kg ATRC core with several different core conditions. The values obtained and the pertinent core conditions for each measurement are summarized in Table I-1. Also included in Table I-1 are values and pertinent core conditions for measurements performed previously [1] in the ATRC 35-kg core.

TABLE I-1

SUMMARY OF ATRC NOISE ANALYSES FOR β/Λ .

Run No.	Date	Analysis Technique	Chamber Type	Chamber Position	Core U-235 Load	Neck Shims Inserted	H-hole Hf Inserts	Outer Shim Position (degrees)	$\beta/\Lambda(\text{sec}^{-1})$ [a]	Other Core Conditions[b]
1	6-64	Auto-power	B-10 CIC	Center flux trap	35 kg	All No. 6	None	13.0	163 + 1.8	Pressure tube and flow tube out of center trap
2	6-64	Auto-power	B-10 CIC	Center	35 kg	All Nos. 5,6	None	47.2	151.3 $\pm$ 5.2	
3	6-64	Auto-power	B-10 CIC	Center	35 kg	All Nos. 2,3, 4,5,6	None	78.3	135.3 $\pm$ 6.0	
4	6-64	Auto-power	B-10 CIC	Center	35 kg	All Nos. 2,3, 4,5,6	None	107.5	117.6 $\pm$ 7.6	
5	6-64	Auto-power	B-10 CIC	Center	35 kg	All	None	137.0	106.9 $\pm$ 3.7	
6	1-66	Auto-power	He-3	East	39 kg	All except Nos. 2,4	6	77.0	150.7 $\pm$ 1.8	Instrument chimble in center and east flux traps
6	1-66	Cross-power	He-3	Center and east	39 kg	All except Nos. 2,4	6	77.0	142.9 $\pm$ 1.3	
6	1-66	Auto-power	He-3	Center	39 kg	All except Nos. 2,4	6	77.0	148.5 $\pm$ 2.8	
7	1-66	Auto-power	He-3	East	39 kg	All except Nos. 1,2,3	6	47.0	168.6 $\pm$ 2.1	
7	1-66	Cross-power	He-3	Center and east	39 kg	All except Nos. 1,2,3	6	47.0	161.0 $\pm$ 1.6	
7	1-66	Auto-power	He-3	Center	39 kg	All except Nos. 1,2,3	6	47.0	167.4 $\pm$ 1.8	
8	1-66	Auto-power	He-3	East	39 kg	All No. 1	6	0.0	192.3 $\pm$ 1.9	
8	1-66	Cross-power	He-3	Center and east	39 kg	All No. 1	6	0.0	183.5 $\pm$ 1.5	
8	1-66	Auto-power	He-3	Center	39 kg	All No. 1	6	0.0	190.0 $\pm$ 1.9	
9	1-66	Auto-power	He-3	East	39 kg	All except Nos. 2,4	None	64.0	153.1 $\pm$ 1.6	
9	1-66	Cross-power	He-3	Center and east	39 kg	All except Nos. 2,4	None	64.0	147.4 $\pm$ 0.9	
9	1-66	Auto-power	He-3	Center	39 kg	All except Nos. 2,4	None	64.0	155.8 $\pm$ 2.0	

[a] Quoted errors are one standard deviation

[b] Unless stated otherwise all other core conditions are the same for all runs.

[a] Present address: Hewlett-Packard, Colorado Springs, Colorado.

In the summary presented in Table I-1, there are basically three major distinctions in the core conditions and techniques used: (a) In early runs, a boron-lined gamma-compensated ion chamber (CIC) was used; whereas, in subsequent runs, a He-3 chamber was used. (b) In early runs, only the auto-power spectrum was measured; whereas, in the recent runs, both auto-power and cross-power spectra were measured. (c) Relative to the core conditions used in the early runs, the core contained an additional 4 kg of fuel in the recent runs, and sufficient neutron absorbers were added to compensate for the reactivity gain associated with this increase in fuel.

The compensated ion chamber was replaced with a He-3 chamber since the latter type is much more efficient than the former and is more convenient to use because of its smaller size. However, changing the detector material is not expected to influence the value obtained for β/Λ , especially since both He-3 and B-10 are near-1/v absorbers in the thermal energy region.

The values in Table I-1 indicate a small but definite difference in the measured β/Λ depending on whether an auto-power or cross-power measurement was made, the auto-power values being consistently larger than the cross-power values. The probable cause of this difference stems from two factors, both of which imply that the cross-power result is more nearly correct. First, the recording system noise can practically never be completely removed from the recorded reactor noise in an auto-power measurement; whereas, the system noise, being uncorrelated, is fully removed in a cross-power measurement. Second, as will be subsequently described, the functional model to which the auto-power data are fit in a least-squares fashion requires the determination of both a zero and a pole (the pole is equal to β/Λ); whereas, the model for the cross-power data requires only the determination of the pole. Consequently, in determining β/Λ from auto-power data, since the fitting procedure obtains a best value for both parameters at the same time, the value for any one may not be the best estimate. (In particular, the value of the pole may not be a best estimate for β/Λ .) Additionally, the variance quoted will not indicate how good the value obtained is since in the two-variable analysis the covariance of the parameters will also be important. Thus, since on both accounts the cross-power values are better, those values are most likely to be near the value characteristic of the reactor.

In order to evaluate the changes in β/Λ which occurred as the result of varying the fuel loading and the neck shim and outer shim positions, it will be assumed that any differences in the type of chamber used are unimportant, that values from auto-power measurements should be compared with values from auto-power measurements only, and that the best value for a particular core condition is given by a cross-power analysis. Under these assumptions, the two curves of β/Λ as a function of outer shim position (shown in Figure I-1) can be drawn, one for each fuel loading. The difference between the two is the combined change in generation time due to increasing the fuel loading and adding sufficient absorbers to compensate for the reactivity effect of the fuel. These two curves are the result of auto-power measurements; whereas, the curve shown in Figure I-2 is the companion curve for the 39-kg core taken from cross-power data and, thus, is the most nearly correct curve.

It must be emphasized that although the curves in Figures I-1 and -2 have been plotted as a function of outer shim position, there are a given number of poison rods (including both H-hole inserts and neck shims) corresponding

Fig. I-1 Values obtained for β/Λ in ATRC 35- and 39-kg cores with auto-power spectrum measurements.

Fig. I-2 Values obtained for β/Λ in ATRC 39-kg core with cross-power spectrum measurements.

to each outer shim position which are required to maintain criticality. (The number of such rods inserted appears in parentheses at each data point in Figures I-1 and -2.) Consequently, the shape of these curves cannot be interpreted as representing the change in β/Λ as the outer shims rotate. However, provided the assumption is justified that the effective absorption cross section for each neck shim and H-hole insert is the same, the curves can be interpreted as representing the change in β/Λ as the outer shim position at critical is varied in a cold, clean core with no experiments. This latter interpretation can be quite useful in accident studies.

A description of the data acquisition and analysis procedures for Runs 1 through 5 in Table I-1 may be found in Reference 2. For each of the Runs 6 through 9, both the auto-power and cross-power spectra were obtained from approximately 15-minute recordings, with the reactor critical at a constant power level of 1 watt. A description of the data acquisition and analysis procedures for these four runs is presented below.

Two Texas Nuclear He-3 chambers were used; one was located in the east flux trap, the other in the center flux trap. (See ATRC core diagram in Figure

I-3.) Their output currents were detected with high-frequency current amplifiers, filtered and amplified with Fairchild A006 amplifiers, and recorded simultaneously with an Ampex FR-1300 tape recorder using a tape speed of 3-3/4 inches

Fig. I-3 ATRC core diagram.

per second (ips). Sine calibration signals in the frequency range from 63.1 to 1000 cps also were recorded on the tape from a constant-current chamber-simulator through the identical instrumentation system used to record the data. This was done for the purpose of system frequency calibration. In addition, a channel of system noise was recorded with the chambers removed from the reactor for the purpose of determining any possible significant system noise corrections to the spectra.

The data were analyzed with the modified Boonshaft and Fuchs 711CL spectrum analyzer, and corrections were made to the resulting auto- and cross-spectra as follows:

- (1) The high-frequency (63.1 to 1000 cps) instrumentation system response for Channel X (east flux trap channel) and Channel Y (center flux trap channel) were obtained from the sine calibration signals on the tape channels used for Run 6. Two cases are of interest: (a) playback at a tape speed of 3-3/4 ips for analysis of the high-frequency portion of auto-power spectra from 125 to 1000 cps, and (b) playback at a tape speed of 30 ips for analysis of the low-frequency portion of all spectra from 63.1 to 125 cps. The response above 63.1 cps is essentially that of the tape recorder at 3-3/4 ips and is typical of all channels. All auto-power spectra were corrected for the appropriate 3-3/4-ips, high-frequency, instrumentation response. The 30-ips response for both channels was essentially flat in the region from 63.1 to 125 cps. Thus, no corrections were required to the auto-power or cross-power spectra in this frequency range.
- (2) The system low-frequency response in the range of 2.51 to 63.1 cps was measured to within about 0.05 dB with the modified Boonshaft and Fuchs analyzer. Both X and Y channels were essentially the same in this frequency range. Each of the cross-power spectra was analyzed with and without the low-frequency corrections, and in each case the low-frequency corrected β/Δ breakpoints were about 1.8 percent lower than the uncorrected breakpoints. The auto-spectra values have not, as yet, been corrected for the low-frequency response with the exception of the data for Run 6. As with the cross spectra, the corrected values of β/Δ for this run are about 1.8 percent lower than the uncorrected values.
- (3) The analyzer-frequency response was measured using a pseudo-random digital noise source whose spectrum was flat to within better than 1 percent in the frequency range of analysis. The necessary corrections are principally due to two uncorrelated effects: (a) the frequency response of a 3-kcps low-pass filter, which is part of the analyzer input amplifier and which contributes the bulk of the total correction--about 0.4 dB at 1000 cps, and (b) the small variations in the analyzer oscillator output voltage. The most recently measured analyzer response corrections were used to adjust all of the spectra, both auto and cross.
- (4) System noise corrections for the auto spectra were obtained by analyzing the noise channels which were recorded with the chambers removed from the reactor. The corrections were not very large and were used only with the data of Run 6. The effect of neglecting them for the other runs was to slightly increase the values of the data points over what they should have been. This resulted in a computed breakpoint for β/Δ which is slightly low. An estimate of the noise correction error, based on the observed

changes of the breakpoints for the data with and without the low-frequency system corrections, is about 1 percent. The cross spectra were not corrected for system noise because meaningful estimates of the noise cannot be obtained unless the chamber is in the same position in the reactor and biased to the same voltage used for obtaining the data. However, there was no apparent correlated noise problem judging from the lack of resonances in the cross spectra. (The uncorrelated noise in both channels does not contribute to the cross spectra.)

(5) The auto spectra were weighted according to the function,

$$W_i = \frac{bT}{y_i^2} \quad (1)$$

where

b = analysis bandwidth

T = analysis time

and

y_i^2 = value of analysis point at i^{th} frequency.

The cross spectra were weighted according to the function,

$$W_i = \frac{2bT}{\left(\frac{1}{\alpha}\right)^2 + 1} \cdot \frac{1}{y_i^2} \quad (2)$$

$$\alpha^2 = \frac{\phi_{xy}^2}{\phi_{xx} \phi_{yy}} \quad (3)$$

The corrected and weighted spectra were fitted with a nonlinear least-means-square curve fitting program on the IBM-7040 computer to the following functional forms:

(a) Auto Power:

$$\phi_{11}(\omega) = \frac{k(\omega^2 + Z_1^2)}{(\omega^2 + P_1^2)} \quad (4)$$

(b) Cross Power:

$$\left| \phi_{12}(\omega) \right| = \frac{k}{(\omega^2 + P_1^2)} \quad (5)$$

In each case the pole at P_1 is the β/Λ frequency cutoff of the reactor. The cross-power spectral analysis procedure determines the existence and magnitude of any possible phase difference between the two signals for each analysis frequency. In this case, there were no observable phase differences in the two chamber currents within the frequency range of the cross-spectra analysis.

The standard deviations quoted in Table I-1 are obtained from the curve fitting program used, and provide estimates of how well the experimental data actually fit the chosen function. In summary, all of the spectra for these four runs were corrected for analyzer response. All of the cross terms and the auto term for Run 6 (only) were corrected for low-frequency system response; all the auto-power terms were corrected for high-frequency system response. Only the auto terms for Run 6 were corrected for system noise.

1.2 Effects of Flux Trap Experiments and Outer Shim Positions on ATRC Power Distribution (J. B. Willis)

Power distribution effects of experiments in ATR flux traps have a direct bearing on the operation of the ATR. Problems of reactor operation that may result from experiment effects are

- (1) Possible power peaking in hot-spot fuel elements adjacent to the experiment
- (2) The selection of the shims to be used to compensate the reactivity effects of experiments such that the interaction of experiments is minimized.

The following power distribution characteristics, either explicitly or implicitly associated with experiment changes, have been observed in the ATRC:

Peaking factors in fuel elements surrounding interior flux traps are affected primarily by the metal-to-water ratio rather than the fuel or poison content of the flux trap. (An interior flux trap is a flux trap surrounded by six concave fuel elements as shown in Figure I-3.) Specifically, changes in fuel element fission rates as a result of changes in experiment U-235 content and shrouds are generally less than 10 percent in Fuel Plate 2 (ie, the second narrowest fuel plate of a fuel element). No meaningful changes in fission rate due to changes in experiment fuel content or shrouds are detectable beyond the eleventh fuel plate. The change in the radial fission rate profile in a fuel element due to an extreme change in the flux trap neutron poison (ie, shrouding the fueled experiment with 100 volume percent hafnium) is shown in Figure I-4.

Fig. I-4 Radial fission rate profiles in Fuel Element Position 6 showing the effect of a 100 volume percent hafnium shroud on the fueled experiment in the NE flux trap.

The removal of a flux trap filler [3] from an interior flux trap, thereby reducing the flux trap metal-to-water ratio, causes a localized increase in fission rates of the order 20 percent in Fuel Plate 2.

The rotational position of outer shim cylinders near fuel elements adjacent to interior flux traps has a decided effect on the fission rates in the exterior plates of these elements. Although more pronounced in the exterior (widest) plates, the effect extends more than halfway through the element. These effects are shown in Figure I-5. The increase in the fission rates near the flux trap

Fig. I-5 Radial fission rate profiles in a fuel element adjacent to an outer shim showing the effects of outer shim positioning.

baffle (narrow fuel plates) for outer shim positions 108 and 155 degrees, as compared to the less withdrawn positions, is principally attributed to the absence of a flux trap filler.

The discussion above shows that the rotational position of the outer shims strongly influences the fission rates in nearby fuel elements. Further, any change in the power generated in a fuel element will, in turn, cause a change in the radiation environment of nearby experiments. Many experiment irradiation programs are conducted simultaneously in a test reactor, and it is generally desired that each experiment be in a constant and known radiation environment independent of other experiments. Consequently, it is particularly important in ATR that the reactivity effect of experiment changes be compensated by moving shims adjacent to the experiment. This control philosophy is in keeping with the concept of independent irradiation programs and will tend to minimize secondary power distribution effects in the vicinity of other experiments.

2. ENGINEERING EXPERIMENTS (V. A. Walker)

2.1 ATR Fuel Element Development (M. L. Griebenow, G. N. Fillmore, J. A. Hendricks)

2.1.1 Thermal Bowing. The geometrical arrangement of the ATR fuel assemblies, within the core, produces a marked radial flux peaking across the assembly. The flux peaking and the subsequent radial temperature variation produces a thermal stress within the fuel assembly that tends to be relieved by bowing the assembly toward Channel 20. The bowing causes the flow areas in Channels 1 and 20 to vary with axial position thereby affecting the local coolant velocities, pressures, and the heat removal capability. An extensive analytical investigation was performed to determine the influence of the bowing on the fuel assembly performance capability.

The extent of the bowing, analyzed by assuming the fuel assembly was a simply supported beam, was found to be limited by side plate contact with the beryllium (or the neck shim housing -- depending on the fuel element position). The complex flow pattern through the bowed assembly was analyzed on the IBM-7040 using the MACABRE code⁴. The variable velocities affect the local static pressures and produce a pressure differential acting across the fuel assembly, tending to bend it as a beam, and acting across Plates 1 and 19, tending to bend these plates independently. The additional beam bending of the fuel assembly was computed assuming the assembly behaves as a beam simply supported at three points. The additional bending changes the lateral pressure differentials that produced the bending. The bending and flow rate solution requires an iteration between the MACABRE-computed pressure differentials and the bending. The iterative solutions converged prior to producing any mode of stress in excess of the yield strength of the fuel assembly. The local deflection of Plate 19, due to the pressure differential between Channel 19 and 20, does not exceed 0.004 inch.

The thermal hydraulic solutions for the bowed assembly show a small reduction in the hot-spot temperature. This results from the diversion of the

coolant from Channels 18 and 20 toward the internal channels in the region of the hot spot. The flow redistribution causes a more even division of the heat flowing from Plates 18 and 19 to the adjacent channels thereby slightly reducing the maximum heat flux. The thermal bowing will, therefore, not be pernicious to the operation of the fuel assembly, and no hot-spot hot-channel factors are required to allow for the bowing.

2.12 Hydraulic Testing. Eighty-seven, fully enriched, ATR fuel assemblies have been proof tested to date. The proof test consisted of flow testing at 120 percent of nominal flow (1050 to 1100 gpm) and 350°F for 6 to 8 hours. None of the assemblies have failed catastrophically; however, they are all permanently deflected, the assemblies bowing, as a beam, toward Channel 20. The extent of the bow is shown in Figure I-6.

Fig. I-6 ATR post-proof-test permanent set.

The bowing, in an assembly originally meeting design specifications, should not impair its operation since all of the internal coolant channels remain essentially unchanged.

Two depleted fuel assemblies recently were hydraulically tested to determine their mechanical integrity and their mode of failure. These tests were to be conducted in the same manner as the assemblies received previously. The flow rate through the assembly was to be increased, in 50-gpm increments, at 350°F to 120 percent of the nominal flow (1050 to 1100 gpm). This flow rate and temperature, maintained for 8 hours, constitutes a proof test, and it is believed that any assembly withstanding these conditions should perform satisfactorily in the reactor. Subsequent to the proof test, the assemblies were to be subjected to an increase in flow (50-gpm incremental steps) up to the flow limit of the hydraulic facility (1300 gpm) or failure if it occurs before the loop capacity is reached. Initial indications of incipient failure in the first assembly started at 600 gpm, and at 1000 gpm the swage joint at the top of Plate 17 failed. The failure of the second assembly was similar, but failure occurred at 1200 gpm and at 400°F.

The failures appear to be caused by inadequate swage joint strength. Figure I-7 shows a failed plate and one additional plate where the penetration into the

Fig. I-7 ATR fuel plate failure.

side plate is insufficient to provide good joint strength. This problem has been brought to the attention of the fabricator, and corrective action has been initiated.

3. METALLURGICAL DEVELOPMENT (G. W. Gibson)

3.1 Fuel Stability (M. J. Graber)

3.11 Uniform Swelling. Calculations have shown that in a fuel core composed of 30 volume percent fuel and an aluminum matrix material having a 60 percent reduction of area as determined by uniaxial tensile test, matrix cracking from fuel particle swelling is expected at about 14 percent core swelling provided the fuel particles are uniformly dispersed.

Experimental results on a nearly dense, 27 volume percent, niobium-coated UO_2 dispersion in Type X8001 aluminum fuel plates showed matrix cracking in a plate after 11.8 percent swelling and irradiation to 17×10^{20} fission/cc. The fuel dispersion in these plates was less than ideal. On the other hand, 95 percent dense powder metallurgy cores containing UAl_3 as the original dispersant have been irradiated to 16.3×10^{20} fission/cc with only 5.1 percent swelling.

Table I-2, which contains new swelling data, and Figure I-8, which is a combined plot of the previous and new data, show the swelling data for U_3Si_2 ,

TABLE I-2

SWELLING DATA FOR VARIOUS FUEL MATERIALS DISPERSED IN ALUMINUM
IRRADIATED IN THE G-12 PRESSURIZED WATER LOOP OF THE ETR

Plate Number	Composition	Fission/cc $\times 10^{-20}$	Expected $\Delta V/V$ (percent)	Observed $\Delta V/V$ (percent)	Swelling Accommodated (percent)	Fabrication Voids (percent)	Reaction Voids (percent)	Total Voids Available (percent)	Void Volume Remaining (percent)	Average Operating Temperature ($^{\circ}\text{C}$)
P-14-766	44 wt% UAl_2	10.9	7.0	3.1	3.9	3.9	0.5	4.4	0.5	111
P-14-769	44 wt% UAl_2	12.1	7.7	3.0	4.7	4.1	0.5	4.6	-0.1	126
P-12-727	50 wt% UAl_3	10.9	7.0	4.0	3.0	8.4	0.9	9.3	6.3	110
P-12-735	50 wt% UAl_3	13.5	8.6	1.8	6.8	8.1	0.9	9.0	2.2	129
P-12-736	50 wt% UAl_3	14.9	9.5	4.4	5.1	9.9	0.9	10.8	5.7	150
P-1-1003	51 wt% UAl_3	3.3	2.1	-1.1	3.7	11.7	0.9	12.6	9.4	232
P-1-1005	51 wt% UAl_3	6.2	4.0	-1.7	5.7	10.1	0.9	11.0	5.3	253
P-1-1007	51 wt% UAl_3	5.8	3.7	-2.5	6.2	11.3	0.9	12.2	6.0	238
P-1-1046	51 wt% UAl_3	3.0	1.9	-1.1	3.0	10.4	0.9	10.3	8.3	225
O-3-816	41 wt% U_3O_8	14.2	9.1	3.3	5.8	5.3	7.0	12.3	6.5	143
O-3-071	41 wt% U_3O_8	14.3	9.1	1.7	7.4	4.2	7.0	11.2	3.0	147
O-3-872	41 wt% U_3O_8	12.0	7.7	2.4	5.3	4.2	7.0	11.2	5.9	126
P-2-1022	42 wt% U_3O_8	3.3	2.1	-2.2	4.3	5.1	7.0	12.1	7.8	232
P-13-755	38 wt% U_3Si_2	11.4	7.3	4.4	2.9	0.9	-0.4	0.9	-2.4	113
P-13-761	38 wt% U_3Si_2	13.4	8.5	6.6	1.9	1.4	-0.4	1.4	-1.1	124
P-13-765	38 wt% U_3Si_2	12.5	8.0	4.7	3.3	3.4	-0.4	3.4	-0.3	127

UAl_2 , U_3O_8 , and the coated UO_2 as well as UAl_3 . In general, the cores with the lowest theoretical densities (largest amount of fabrication voids) show the lowest amount of swelling. The coated UO_2 system has the highest swelling rate. With this system, there was not sufficient provision for fission product accommodation within the particle because of high particle density, and the particles could not conform to any fabrication voids which might be present because of the rigidity of the hard spherical niobium shells around the particles. The U_3Si_2 cores had a relatively high starting density, and there was a slight volume increase when U_3Si_2 reacted with aluminum to form UAl_3 . This accounts for the high swelling rate of the U_3Si_2 plates. The U_3O_8 and UAl_2 cores in this series of tests had higher starting densities than the UAl_3 . A few of the test plates that were irradiated at the higher temperatures (230 to 250°C) and to very low radiation exposures exhibited negative swelling, indicating radiation-induced sintering.

The role of built-in porosity to accommodate fission products is not restricted to aluminum dispersion fuel systems. Two recent test series employing

Fig. I-8 Swelling of various fuel materials dispersed in aluminum (irradiated in G-12 pressurized water loop of the ETR).

low-density (82 and 86 percent particle density) UO_2 in stainless steel resulted in reduced swelling rates. The swelling data are shown in Table I-3 and Figure I-9. Although the initial porosity in the A-15 composition was 4.4 percent of the core compared to 6.4 percent for the A-16 composition, fission product accumulation at these low levels of exposure was insufficient to fill the voids, and the swelling performance of each amounted to approximately 1/3 that expected from dense fuel. The average swelling rate in terms of volume percent changed per 1 atom percent uranium fissioned was 0.60 and compares favorably with the 0.40 percent initial experimental swelling value reported by Bleiberg [5] for less than theoretically dense UO_2 . After densification as a result of fission

TABLE I-3
LOW-DENSITY UO_2 - STAINLESS STEEL SWELLING DATA

Plate Number	Average Fiss/cc. x 10^{-20}	U-235 Burnup (percent)	Expected Core Swelling ($\Delta V/V \times 100$)	Measured Core Swelling ($\Delta V/V \times 100$)	Swelling Accommodated by Voids ($\Delta V/V \times 100$)	Original Void. (volume percent)	Voids Remaining (volume percent)	Measured Swelling Rate [A]
A-15-923	3.68	6.2	2.35	1.64	0.71	4.4	3.7	1.10
A-15-925	8.74	14.7	5.57	2.71	2.86	4.4	1.5	0.75
A-15-926	8.74	14.7	5.57	2.62	2.95	4.4	1.0	0.75
A-15-933	4.31	6.9	2.75	0.47	2.28	4.4	1.7	0.27
A-16-943	11.13	21.1	7.10	1.94	5.16	6.4	1.2	0.43
A-16-945	11.13	21.1	7.10	3.34	3.76	6.4	2.6	0.74
A-16-947	5.10	9.5	3.25	-0.76	---	---	---	---
A-16-952	5.36	10.0	3.42	0.42	3.00	6.4	3.4	0.19
								7/4.23
								Average 0.60

[A] VOLUME PERCENT PER 2.46×10^{20} FISS/CC OR VOLUME PERCENT PER 1 ATOM PERCENT FISSIONED.

Fig. I-9 The swelling rate of low-density UO_2 -SS fuel cores compared to the swelling rate of dense UO_2 determined experimentally and theoretically.

product formation, the Bleiberg experimental value of 1.70 percent swelling per atom percent fissioned compares with that theoretically expected of dense materials or 1.57 percent. The swelling values (0.40, 0.60, 1.57, and 1.70) for 1 atom percent fissioned occur at 2.46×10^{20} fiss/cc.

3.12 Accelerated Swelling. The combined effect of the fuel stability variables, as they are manifested in the various individual fuel materials, each of which was fabricated by a similar technique, has been studied by post-irradiation anneal tests. These tests have shown, and continue to show, that the temperature at which accelerated (or breakaway) swelling occurs depends on what dispersant fuel material is used.

For instance, the accelerated swelling temperature of UO_2 dispersed in aluminum is about 250°C while that of UAl_3 is about 500°C at 10×10^{20} fiss/cc. Figure I-10

Fig. I-10 Failure/no-failure lines for various fuel materials dispersed in aluminum.

shows the failure/no-failure lines for various fuel materials dispersed in aluminum. These curves are similar to those shown previously [6], but have been confirmed and extended by additional data. Also, the effect of no boron in the UAl_3 composition is indicated.

Stainless steel plates containing dispersions of UO_2 and representing three different fabrication processes, the A-15, the A-16, and the 136 compositions, were also blister tested.

The objective in these fabrication processes, as well as to produce a low-density material for better fission product accommodation, was to promote uniform dispersions so that accelerated swelling of higher weight percent fuels (37 to 39 weight percent UO_2 in stainless steel) would be retarded. The fabrication of the A-16 composition employed special compacting and rolling techniques; the A-15 employed a UO_2 particle precoated with stainless steel; and the 136 composition utilized a binder to facilitate handling a powdered core and a powdered clad.

The blister anneal data points of the three compositions were plotted on the 18 to 30 weight percent fuel stability curves determined by Keller, Figure I-11.

Fig. I-11 Blister anneal points of high weight percent (37 to 39) UO_2 dispersed in stainless steel, spotted on low weight percent (18 to 30) curves.

It was found that the blister resistance of these higher weight percent (37 to 39) dispersions (A-15 and A-16 composition) is comparable to that which was achieved previously with low weight percent (18 to 30) fuels. However, the 136 composition prematurely developed one small blister on one of the plates.

3.13 Particle Mobility. As shown in a preceding section, the swelling rate of a powder metallurgy $\text{UAl}_3\text{-Al}$ plate that was irradiated to about 10×10^{20} fiss/cc

is about 1/6 that of dense material, and the swelling rate of a low-density UO₂ stainless steel dispersion may be 1/3 that of dense material. The difference in performance of these two systems is fuel particle mobility. Typically, the UO₂ particles in stainless steel have a recoil zone about 0.0001 inch (0.00025 cm) while the UAl₃ particles in aluminum undergo a significant diffusion reaction. Not only does enough aluminum diffuse into the particle to satisfy the formation of UAl₅ (crystallographically UAl₄), but, apparently, the aluminum also combines with fission products. This mobility allows the swelling fuel particle to conform to voids in the core rather than cause core swelling.

3.2 Corrosion Testing of Type 2219 Aluminum for High Flux Reactor Applications (M. Zukor, B. G. Carlson, G. W. Gibson)

In high-flux aqueous reactor systems, the use of aluminum cladding materials is limited by the buildup of an insulating oxide corrosion layer [7]. With heat fluxes of the order of 2×10^6 Btu/hr-ft², this oxide layer increases fuel plate cladding temperatures to such an extent that the mechanical stresses exceed the yield strength of the material, thereby leading to fuel element failure. To prevent this, the corrosion characteristics of aluminum alloys with good elevated temperature mechanical properties are being investigated.

Aluminum alloy Type 2219 is an aluminum-copper alloy developed by Alcoa for elevated temperature applications. The typical mechanical properties of Type 2219 aluminum in the range 260 to 316°C are superior to those of any other wrought aluminum alloy [8].

In order to ascertain the corrosion properties of Type 2219 Al alloy under high-power-density test reactor flow conditions, Type 2219 Al samples, 5.75 by 1.25 inches, were corrosion tested in the Reactor Engineering Corrosion Loop at 45 ft/sec and at 191°C for 1100 hours together with Types 6061 and X8001 Al alloy specimens for comparison. The nominal chemical compositions of Types 2219, 6061, and X8001 Al are shown in Table I-4.

TABLE I-4
NOMINAL CHEMICAL COMPOSITIONS OF TYPES 2219,
6061, AND X8001 ALUMINUM ALLOYS

	Cr (wt%)	Cu (wt%)	Fe (wt%)	Mg (wt%)	Ni (wt%)	Si (wt%)	Ti (wt%)	V (wt%)	Zr (wt%)	Al (wt%)
2219	----	6.3	----	0.3	----	----	0.6	0.10	0.18	Balance
6061	0.25	0.25	----	1.0	----	0.6	----	----	----	Balance
X8001	----	----	0.5	----	1.0	----	0.1	----	----	Balance

The loop was shut down, and the samples removed twice a week for weighing and corrosion-coating-thickness determination. The oxide coating buildup was measured by means of an eddy-current measuring instrument (Dermatron). Due to the sample holder design, there were small variations in flow velocities.

Type 2219 Al alloy in the heat-treated and aged (T-62) condition, was compared with Type 6061 Al in the fully annealed (-0), solution-heat-treated (T-4), and precipitation-hardened (T-6) conditions. The Type X8001 Al sample was in the fully annealed (-0) state. The oxide buildup is shown in Figure I-12. The flow past the two Type 2219 Al alloy specimens was 43 and 50 ft/sec, compared with 45 ft/sec for the Type 6061 Al. The Type 2219 sample, tested at 50 ft/sec, showed a considerably higher oxide buildup than the other samples tested. The Type 2219 Al alloy specimens also exhibited a variation in oxide thickness of about 1 mil (25 microns) along the sample. After about 800 hours, the oxide buildup on the 43 ft/sec Type 2219 Al sample and the Type X8001 Al sample leveled off at 1.75 and 1.50 mils, respectively. Figure I-13 shows the overall corrosion rates of the samples tested. The average corrosion rate of the Type 2219 Al, tested at 43 and 50 ft/sec, was about equal to that of the Type 6061 Al tested at 45 ft/sec and was slightly greater than that of Types X8001-0 and 6061 Al in the T-4 and T-6 conditions.

Fig. I-12 Oxide coating buildup on Type 2219 Al compared to Types 6061 and X8001 Al alloys.

Photomicrographs of the oxide coatings on the two Type 2219 Al alloy samples are shown in Figures I-14 and -15. The oxide coatings appear to show a laminar-type structure. For comparison, the Types 6061-0 and X8001 Al coatings are shown in Figures I-16 and -17.

Based upon the above findings, sample fuel plates clad with Type 2219 Al were irradiated in the G-12 pressurized water loop of the ETR at 46 ft/sec and at a pH of 5.0 ± 0.2 . Heat fluxes of up to 2.5×10^6 Btu/hr-ft² were obtained. The results of the post-irradiation hot cell examination of the Type 2219 Al plates together with Type 6061 Al companion plates are tabulated in Table I-5. The Type 2219 Al fuel plates tested were in the fully annealed (-0) and in the precipitation-hardened (T-62) conditions. The Type 2219 Al samples were compared against Type 6061-0 Al clad (the proposed ATR cladding material) fuel plates. In all cases, the Type 2219 plates exhibited lower final oxide coating buildups than the corresponding Type 6061-0 Al clad fuel plates. On all but one of the Type 2219 clad samples (P-19-1131), the oxide coating sloughed off over the core region. The average and maximum final oxide coating thicknesses for the Types 2219-0 and 6061-0 clad plates irradiated during Cycle 78 are shown in Figure I-18. This figure shows that Type 2219-0 Al offers an advantage over Type 6061-0 Al in terms of oxide coating buildup. Sufficient data are not available for a similar comparison between Type 2219 Al in the T-62 condition with Type

Fig. I-13 Comparison of corrosion rates of Types 2219, 6061, and X8001 aluminum alloys.

Fig. I-14 Photomicrograph of oxide corrosion coating formed on Type 2219 Al tested 50 ft/sec for 1123 hours at 190°C.

Fig. I-15 Photomicrograph of oxide corrosion coating formed on Type 2219 Al tested at 43 ft/sec for 1123 hours at 190°C.

Fig. I-16 Photomicrograph of oxide corrosion coating formed on Type 6061-0 Al alloy tested at 45 ft/sec for 954 hours at 190°C.

Fig. I-17 Photomicrograph of oxide corrosion coating formed on Type X8001 Al alloy tested at 46 ft/sec for 1123 hours at 190°C.

TABLE I-5

COMPARISON OF OXIDE BUILDUP ON TYPES 2219 AND 6061
ALUMINUM FUEL PLATES

Plate No. Cladding	Initial Plate Surface Temperature (degrees)	Avg. Heat Flux (Btu hr-ft ²)	Final Oxide Coating Thickness			
			Core		Edge	
			Avg (mils)	Max (mils)	Avg (mils)	Max (mils)
<u>Cycle 77 -- 359 hr</u>						
P-19-1074 2219-T62	199	2.07 x 10 ⁶	0.25	0.40	1.40	1.85
P-19-1077 2219-T62	197	1.92	0.40	1.45	1.40	1.70
P-19-1076 2219-T62	190	1.64	0.25	0.40	1.50	1.95
P-23-1101 6061-0	183	1.88	0.70	1.00	0.80	1.05
<u>Cycle 78 -- 631 hr</u>						
P-19-1131 2219-0	196	1.83 x 10 ⁶	1.55	2.20	1.50	1.70
P-14-775 6061-0	188	1.85	1.95	2.90	1.70	2.25
P-24-1142 6061-0	186	1.56	1.95	2.40	2.00	2.60
P-14-774 6061-0	182	1.83	1.05	1.60	1.25	2.00
P-19-1130 2219-0	176	1.68 x 10 ⁶	0.80	1.20	0.80	1.05
P-14-774 6061-0	182	1.83	1.05	1.60	1.25	2.00
P-24-1145 6061-0	180	1.68	1.45	2.00	1.40	2.00
P-20-1052 6061-0	170	1.28	1.80	2.20	1.85	2.10
P-20-1057 6061-0	168	1.08	1.90	2.25	1.90	2.20
P-19-1128 2219-0	139	0.81 x 10 ⁶	0.25	0.50	0.40	0.60
P-24-1143 6061-0	141	0.88	0.55	0.75	0.60	0.80
P-14-770 6061-0	139	0.81	0.55	0.60	0.65	0.90

Fig. I-18 Comparison of oxide coating buildup as function of initial plate surface temperature for Types 6061-0 and 2219-0 Al clad fuel plates.

6061-0 Al. However, the Cycle 77 Type 2219-T62 clad samples, although irradiated at a higher initial plate surface temperature, exhibit a lower final oxide coating than the Cycle 78 Type 2219-0 plates.

Figure I-19 compares the yield strength of Type 2219 Al in the -0 and -T62 conditions with Type 6061-0 Al. Based on a heat flux of 2×10^6 Btu/hr-ft², each mil of oxide increased the fuel plate cladding temperature by about 53°C. From Figure I-18, at the expected maximum nominal initial surface temperature for ATR fuel plates (177°C), the maximum oxide buildup is 1.2 mils for the Type 2219-0 Al clad plates and 1.6 mils on the Type 6061-0 plates. This corresponds to a final plate surface temperature of 241°C for the Type 2219-0 Al and 262°C for the Type 6061-0 Al. At these temperatures (see Figure I-19), Type 2219-0 has a 4.5-ksi yield strength advantage over Type 6061-0 Al. From the irradiation data available on the Type 2219-T62 samples irradiated during Cycle 77 at 241°C, Type 2219-T62 has a yield strength of 22.0 ksi, or 13.7 ksi better than Type 2219-0.

The Type 2219 Al alloy samples exhibited uniform corrosion with no pitting attack evident. The oxide corrosion coatings were stripped from the Type 2219 Al clad plates and most of the Type 6061 clad comparison plates with phosphoric-chromic acid. The overall corrosion rate in mg/dm²-day and the uniform cladding penetration after 17.0 days (1 ATR Cycle) is shown in Table I-6. The Type 2219-0 Al clad plates exhibited lower total corrosion rates than their corresponding Type 6061-0 plates. The uniform Al cladding penetration after 17 days was less than 0.6 mil on the Type 2219-0 clad samples and under 1.0 mils on the Type 6061 plates. The Type 2219-T62 clad samples showed overall corrosion

Fig. I-19 Comparison of yield strengths of Types 2219-0 and 2219-T62 with Type 6061-0 aluminum.

rates of 2 to 3 times that of the Types 2219 and 6061-0 samples, although the final oxide coating buildup was considerably less on the Type 2219-T62 samples. Even at these higher corrosion rates, the Al cladding penetration of the Type 2219-T62 samples amounted to less than 2.0 mils after 17.0 days. It should be noted that the flux and initial plate surface temperatures seen by the Type 2219-T62 samples were higher than those for the Type 2219-0 and the Type 6061 companion samples. Uniform cladding penetrations of up to 5 mils can be tolerated on ATR fuel plates.

Type 2219 Al alloy contains about 6.3 percent copper. Copper, traditionally, has not been used in reactor systems due to its tendency to plate out on other materials. Copper analysis on loop water samples, taken during reactor operation, turned up no copper present above background levels ordinarily found in demineralized water.

Thus, Type 2219 aluminum appears to be attractive for application as a cladding material in high-flux aqueous reactors. Five Type 2219 aluminum-clad ETR fuel elements are being procured for further testing.

TABLE I-6

COMPARISON OF IN-PILE CORROSION RATES
OF TYPES 2219 AND 6061 ALUMINUM

Plate No.	Cladding	Cycle	Avg Corc Final Oxide (mils)	Corrosion Rate (mdd)	Uniform Cladding Penetration (mils/17.0 days)
P-19-1074	2219-T62	77	0.25	52.1	1.23
P-19-1076	2219-T62	77	0.25	75.6	1.79
P-19-1077	2219-T62	77	0.40	85.0	2.01
P-23-1101	6061-0	77	0.70	14.2	0.37

TABLE I-6 (Contd.)

COMPARISON OF IN-PILE CORROSION RATES
OF TYPES 2219 AND 6061 ALUMINUM

Plate No.	Cladding	Cycle	Avg Core Final Oxide (mils)	Corrosion Rate (mdd)	Uniform Cladding Penetration (mils/17.0 days)
P-19-1131	2219-0	78	1.55	23.7	0.56
P-14-775	6061-0	78	1.95	39.1	0.93
P-24-1142	6061-0	78	1.95	37.1	0.88
P-14-774	6061-0	78	1.05	17.1	0.41
P-19-1130	2219-0	78	0.80	13.0	0.31
P-14-774	6061-0	78	1.05	17.1	0.41
P-20-1057	6061-0	78	1.90	25.4	0.60.
P-19-1128	2219-0	78	0.25	11.0	0.26
P-24-1143	6061-0	78	0.55	6.9	0.16

4. MATERIALS RESEARCH
(J. M. Beeston)4.1 Behavior of Uranium Intermetallic Compounds with Liquid Lead. (R. A. Moen)

Studies were initiated and reported last quarter [9] regarding the compatibility of liquid lead and uranium intermetallic compounds.

In the last quarterly report, only the visual examination of samples of UAl_3 , UAl_4 , and U-Al alloy heated with pure lead at temperatures ranging from 500 to 900°C for up to 72 hours was discussed. Since that time, an optical metallographic examination was completed. The examination showed that the first indication of attack of the UAl_3 surface by liquid lead came during the 700°C test. Subsequent tests at 800 and 900°C resulted in a penetration of up to 6 mils into the UAl_3 by the lead. UAl_3 heated with lead at 500 and 600°C was virtually in the as-fabricated condition. UAl_4 heated with lead at 700°C was attacked to the same extent as the UAl_3 at 700°C. The U-Al alloy surface was unattacked after heating at 700°C, but some basic microstructural change (homogenization) was noted as the result of the temperature alone.

The results of the above test showed that lead should be compatible with the uranium aluminide compounds for short periods of time (up to 72 hours) as long as the temperature remained in the 600°C range. To further evaluate

the UAl_3 and other uranium intermetallic compounds with pure lead for extended periods of time approaching those expected during future in-reactor testing, another test was performed. In this test, samples of UAl_3 , U_3Si , and UCu_5 were heated with lead at 600°C for 21 days. The resulting structures were compared metallographically with structures from samples heated at 600°C without lead and with nonheated control samples. The results show about a four mil penetration of the UAl_3 surface by the liquid lead, about an eight mil penetration of the U_3Si surface by the liquid lead and gross reaction between the UCu_5 and the liquid lead. Figure I-20 shows the microstructures of the three materials before and after heating with lead.

Fig. I-20 Microstructures of UAl_3 , U_3Si , and UCu_5 before and after heating with lead at 600°C for 21 days (250 X original magnification; reduced 60 percent).

5. REFERENCES

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II. NUCLEAR TECHNOLOGY

1. CROSS SECTIONS

(M. S. Moore)

1.1 Neutron Scattering by Detector Walls (T. E. Young, F. B. Simpson)

A layer of He^3 counters were installed in the fast chopper detector system to replace the 10 layers of BF_3^{10} counters previously used. It appears that high counting losses due to the increased efficiency of the He^3 detectors in the MeV energy region may be a problem. This is a result of the large number of fast neutrons in the initial neutron burst. Further investigation of the He^3 detectors will be carried out in the future.

Transmission measurements were taken on the Pu-238 sample to check on the neutron scattering effects in the detector system and to observe if there had been any increase in the cross section in the valleys. Previously, there had appeared, using ten layers of BF_3^{10} counters, a small transmission dip about 0.5 eV below the 18.56 eV resonance. Using a single layer of He^3 counters instead of ten layers of BF_3^{10} counters appears to eliminate this small transmission dip. This effect has also been observed for many other resonances. Figure II-1 shows the data taken under similar conditions with the exception of the counters. The solid circles are the data taken with the BF_3^{10} detector, and the open circles are the data observed with a single layer of He^3 as a detector. The explanation of these results is that the dip is caused by scattering and subsequent counting of about 3 percent of the neutrons by the BF_3 arrangement because of the massive amount of material in the beam.

Fig. II-1 Comparison of neutron transmission data taken with ten layers of BF_3 counters and with one layer of He^3 counters. The transmission dip of BF_3 data with respect to He^3 data at 18.1 eV can be explained by the detection of neutrons scattered within the detector system. A similar dip has been observed with various other samples.

1.2 Fission Fragments Range -- Energy Rotations (M. S. Moore, L. G. Miller)

An investigation into some of the phenomena associated with the slowing down of fission fragments in noble gases has been carried out on the crystal spectrometer. The experimental phase of the effort has been completed, and a paper is being prepared for publication. The data reveal significant departures from the theoretical predictions of Lindhard [1,2,3] when the velocity of the fission fragment approximates that of its most strongly bound electron. The Lindhard theory defines dimensionless energy and range parameters, ϵ and ρ , and relates them by the simple formula,

$$\frac{d\epsilon}{d\rho} = k^{1/2} \quad (1)$$

where k depends upon the charges and masses of the moving fragment and the atoms of the stopping medium. The integral of this formula relates the range loss to the energy loss:

$$\rho_0 - \rho = \frac{2}{k} (\epsilon_0^{1/2} - \epsilon^{1/2}) \quad (2)$$

In this experiment, solid-state detectors were used to measure the pulse height produced by the fission fragments after passage through varying amounts of stopping material. From these data were deduced the range loss versus energy loss curves shown in Figure II-2. On all curves the solid line connects the experimental points while the dashed line represents the predictions of the Lindhard theory. The departure from theory can be interpreted as representing a variation in the constant, k , of Equations (1) and (2). The variation in k required to describe the observed data is shown in Figure II-3 for the heavier absorbers investigated and in Figure II-4 for the lighter gases. In all cases the peak departure is for $\epsilon^{1/2} - \epsilon_0^{1/2} = 0$, which represents the condition that the moving fragment has a velocity close to that of an electron in a strongly bound shell.

Fig. II-2 The slowing of median light and heavy fragments from U-235 fission in various gases and in nickel foil. The ordinate is proportional to the fragment range expended in the absorber, and the abscissa is proportional to the velocity loss of the fragment in the absorber. The suppressed origin should be noted for the slowing in nickel foil. In all cases, the linear relationship between the range and velocity parameters expected from the Lindhard treatment^[1,2,3] is given by the dashed line.

Fig. II-3 The ratio of the measured proportionality factor, k_{exp} , to the constant, k_{th} , as given by the Lindhard treatment[1,2,3] as a function of the difference from the critical velocity as described in the text. This ratio appears to be independent of both the fragment mass and the atomic mass of the absorbing gas for the heavier gases.

Fig. II-4 The ratio of the measured proportionality factor, k_{exp} , to the constant, k_{th} , as given by the Lindhard treatment[1,2,3] as a function of the difference from the critical velocity as described in the text. This ratio appears to be independent of the fragment mass but not of the atomic mass of the light absorbing gases.

2. NUCLEAR CHEMISTRY (R. P. Schuman)

2.1 Chopper Cross-Section Measurements (J. W. Coddington, J. R. Berreth, R. I. Tromp, D. K. Oestreich, R. P. Schuman)

A gram of Pm-147 has been purified, and several chopper samples have been fabricated. Cross-section measurements have been made with the fast chopper, and the material appears to be quite pure. Area analysis, COMBQ fitting, and resonance parameter calculations are being made on the cross-section data. The growth of Sm-147 daughter into the samples is being followed, and additional cross-section measurements will be made for this purpose.

A sample of thallium, which has been irradiated to an nvt of $\approx 1.6 \times 10^{22}$ n/cm², has been loaded into a chopper block, and Tl 204 cross-section measurements have started on the MTR fast chopper.

Some Am-243 has been purified and made into chopper samples. The Am-243 has been analyzed by alpha spectroscopy and by exchange column chromatography and has been found to contain 99.82 percent Am-243, 0.15 percent Am-241, and 0.03 percent Cm-244.

The Cm-244 used for the chopper cross-section measurements has been analyzed and found to contain about 1 percent Am-243 and 5.6 percent Pu-240. The purified material has been submitted for mass analysis.

2.2 Analysis of Reactor Coolant Water (E. H. Turk, S. D. Reeder)

A new procedure for the radiochemical determination of potassium in reactor primary coolant systems has been finalized. The distinguishing features of the method are (a) concentration of the activity from a large sample on a cation

exchange resin, (b) elution with hydrochloric acid, (c) scavenging for the removal of cesium and insoluble hydroxides, and (d) precipitation of potassium with sodium tetraphenyl boron. Using HCl and hexone, the potassium tetraphenyl boron is disproportionated making possible reprecipitations of potassium with complete removal of sodium and other radioactive nuclides.

The possibility of using high-resolution germanium gamma detectors for the analysis of reactor coolant water without radiochemical separations has prompted a study of activity collection on ion-exchange resins. Preliminary experiments indicate better than 98 percent collection of the activity in process water with only three rapid passes of the water through small tubes containing either mixed or separated Dowex-50 and Dowex-1 cation-anion resins. Comparison of results between standard radiochemical procedures and germanium gamma counting is in process.

2.3 Decay of Heavy Nuclides (R. N. Chanda, R. A. Deal, R. P. Schuman)

In conjunction with the Am-243 cross-section measurements, the decay properties of the Am-243 nucleus are being investigated utilizing Li-drifted germanium γ -detectors and surface barrier α -detectors. Prior to this study, only one γ -transition had been reported -- 74.6 keV. In addition, transitions of ≈ 30 , 43.5, 86.7, 117.4, and 142 keV have been identified. Alpha-gamma coincidence studies were made using a 2-mm by 1.0-cm² Ge detector and a 25-mm² α -detector. In this way, all 6 γ -transitions tentatively have been placed in a level scheme for Np-239. Due to the availability of two large-volume Ge detectors (4.0 cm³ and ≈ 13 cm³), an attempt was made to measure γ - γ coincidences. The "dead layers", however, on the larger detectors were sufficiently thick (ie $> 400\mu$) to absorb a large percentage of the low-energy γ -rays, thus rendering the experiment unfeasible. Analysis of the alpha spectrum taken on a surface barrier detector, while in substantial agreement with previous data taken on a high-resolution α -spectrometer by a Russian group, is not yet complete.

Studies have been made of the extraction of tracer Pa-233 from aqueous solutions into solutions of Aliquat 336 (tricaprylyl monomethyl ammonium chloride) in benzene. The extraction appears to be highly dependent upon the history of the tracer. For one batch of tracer, the extraction (Pa-233 in organic/ Pa-233 in aqueous) fell from 82 percent from 1M HNO₃ to 18 percent from 8M HNO₃ and 14.5 percent from 10M HNO₃. For another batch, the extraction increased from 39 percent from 1M HNO₃ to 97 percent from 4M HNO₃. Adsorption on walls was found to be quite small. The gross difference in extractability of the two batches of tracer has not been explained.

The Cm-246 and -248 decay products have been separated from californium, and half-lives of 4.3 and 10⁵ yr (partial alpha half-life) and 5.8 x 10³ yr have been obtained for Cm-248 and -246, respectively.

3. DECAY SCHEMES PROGRAM (R. L. Heath, C. W. Reich)

3.1 Gamma-Ray Transitions in the Decay of Nd-149 (R. G. Helmer, L. D. McIsaac)

A decay scheme of Nd-149 has been presented in previous reports[4,5,6]. Subsequently, the gamma-ray spectrum has been remeasured using a

lithium-drifted germanium detector of higher resolution with a 4096-channel pulse-height analyzer. With these improvements, many additional gamma rays have been observed, and better intensity and energy values have been obtained.

In this study, a 0.8-cm by 2.5-cm² Ge(Li) detector was used. The photopeak efficiency was determined at various energies by means of sources whose gamma-ray emission rates were determined with a NaI(Tl) scintillation spectrometer. The resulting efficiency curve is shown in Reference 6. For energy determinations, it is desirable to know the nonlinearity of the electronic system. These corrections are also shown in Reference 6.

The results of the two energy calibration runs are given in Table II-1. Only the strong, well-resolved peaks are analyzed here. For most of these peaks, the positions can be determined to ± 0.05 channels. Thus, for gain scales of 0.09 to 0.50 keV per channel as used here, the errors in the energies should be less than 0.05 keV plus the errors in the energies and positions of the calibration lines. Above 700 keV, the energies were calibrated by assuming that the energy of the 1234-keV transition was equal to the sums 579.25 + 654.82 and 696.29 + 537.87. This assumption was consistent with calibration measurements to within about 0.15 keV at 1234 keV.

The energies of the weaker lines from Nd-149 was determined by interpolation between the stronger lines. The spectra used are shown in Figures II-5 and -6. These spectra were obtained with sources purified by ion-exchange column separations. The latter spectrum involved six separations and about 36 hours of counting time. The resulting energies are given in Table II-2.

For the low-energy transitions, the energies from the previous conversion electron studies [4] have been considered in determining the final energies. The relative photon intensities obtained are also given in Table II-2. The intensities from the data in Figure II-6 include a correction for the attenuation of the photons in the absorber used. The intensities are normalized to percent of decays, assuming 8.4 percent for the 654-keV transition [4]. The uncertainties quoted are intended to be estimated standard deviations.

Currently it is not possible to carry out coincidence experiments that would provide information as to positions of the weaker transitions in the Nd-149 decay scheme; however, the precision of the energy measurements allows some hope for assembling a part of the scheme from the energy values. The coincidence results reported previously were used to aid in this analysis. The analysis was carried by the use of computer programs which used all the established level energies and all the unplaced gamma-ray energies to look for new levels. Possible levels that involved transitions to and from several other levels were tabulated. Those levels that (a) were consistent with the errors assigned to the levels and transitions, (b) were not in conflict with the coincidence information, and (c) involved several transitions were considered. A new proposed decay scheme which includes 100 transitions is shown in Figures II-7 and -8.

Figure II-7 shows the portion of the scheme which is considered to be well established. Recent studies of the decay of Nd-149 have included gamma-gamma angular correlation studies [7,8] and extensive internal-conversion coefficient and lifetime studies [9]. (The latter report uses the gamma-ray intensities from this study.) The multipolarities and spin and parity assignments from Reference 9

TABLE II-1

CALIBRATION OF Nd-149 GAMMA-RAY ENERGIES

Isotope	Peak Position (channels)	Energies (keV)		
		Calibration Value	Computed Value	Difference
Run 1				
Gd-153	1088.39 ± 0.03	97.43	97.41	+ 0.02
Gd-153	1153.00 ± 0.02	103.18	103.20	- 0.02
Lu-177	1262.17 ± 0.02	112.955	112.96	0.00
Nd	1277.48 ± 0.02	---	114.33	---
Nd	1741.98 ± 0.04	---	155.89	---
Lu-177	2328.26 ± 0.02	208.365	208.35	+ 0.01
Nd	2361.42 ± 0.03	---	211.32	---
Nd	2991.52 ± 0.09	---	267.69	---
Nd	3019.04 ± 0.07	---	270.15	---
Hg-203	3119.69 ± 0.07	279.15	279.16	- 0.01
Run 2				
Nd	1197.39 ± 0.02	211.32	211.30	+ 0.02
Nd	1361.16 ± 0.02	---	240.21	---
Nd	1516.93 ± 0.02	---	267.71	---
Nd	1530.93 ± 0.02	---	270.18	---
Hg-203	1582.03 ± 0.02	279.15	279.20	- 0.05
Nd	1850.46 ± 0.03	---	326.58	---
Au-198	2333.24 ± 0.01	411.795	411.80	0.00
Nd	2399.94 ± 0.04	---	423.58	---
Nd	2513.23 ± 0.05	---	443.57	---
Na-22	2894.86 ± 0.05	511.006	510.94	+ 0.07
Nd	3062.52 ± 0.02	---	540.54	---
Nd	3152.16 ± 0.012	---	556.36	---
Bi-207	3227.53 ± 0.03	569.62	569.66	- 0.04
Nd	3710.00 ± 0.04	---	654.83	---
Cs-137	3748.34 ± 0.03	661.595	661.60	0.00

Fig. II-5 Gamma-ray spectrum of Nd-149 below 0.7 MeV measured with an 8-mm by 2.5-cm² Ge(Li) detector. The energies are in keV. The (FWHM) is 0.9 keV at 100 keV and 1.5 keV at 650 keV.

TABLE II-2

GAMMA-RAY ENERGIES AND INTENSITIES

<u>Energy (keV)</u>	<u>Error</u>	<u>Photon Intensity (percent of decays)</u>	<u>Percentage Error (10 percent unless noted)</u>
30.00	0.10	0.15	---
58.90	0.03	1.3	20
65.37	0.05	0.041	20
74.03	0.02	2.2	---
74.07	0.02	2.2	20
75.55	0.02	2.2	---
91.12	0.05	0.063	20
97.00	0.03	1.3	---
107.76	0.10	0.055	20
114.31	0.03	16.1	---
116.09	0.02	0.10	20
122.41	0.05	0.20	---
126.62	0.05	0.10	---
137.02	0.08	0.040	20
139.21	0.05	0.41	---
155.87	0.03	5.2	---
177.82	0.05	0.14	---
185.50	0.08	0.079	20
188.64	0.03	1.7	---
192.03	0.05	0.51	---
198.93	0.03	1.25	---
208.15	0.05	2.5	---
211.32	0.03	23.4	---
213.95	0.15	0.35	---
226.85	0.05	0.14	20
229.57	0.05	0.43	---
240.23	0.03	3.4	---
245.71	0.05	0.89	---
250.84	0.10	0.032	20
254.20	0.07	0.075	20
258.07	0.05	0.33	---

TABLE II-2 (Contd.)

GAMMA-RAY ENERGIES AND INTENSITIES

Energy (keV)	Error	Photon Intensity (percent of decays)	Percentage Error (10 percent unless noted)
267.70	0.03	5.2	---
270.18	0.03	9.2	---
273.28	0.08	0.20	20
275.96	0.05	0.51	20
276.96	0.10	0.28	20
282.47	0.05	0.52	---
288.21	0.05	0.58	---
294.82	0.05	0.50	---
301.14	0.05	0.33	---
310.99	0.05	0.44	---
326.57	0.03	4.0	---
342.85	0.10	0.029	20
347.85	0.02	0.16	50
349.23	0.03	1.27	---
352.75	0.10	0.056	20
357.05	0.10	0.047	20
360.05	0.05	0.14	---
366.64	0.05	0.57	---
380.94	0.10	0.058	20
384.69	0.05	0.29	---
396.77	0.10	0.085	20
423.54	0.03	8.1	---
425.20	0.15	0.40	---
443.53	0.03	1.29	---
480.21	0.15	0.051	20
483.53	0.15	0.073	20
493.86	0.15	0.044	20
497.81	0.15	0.047	20
510.29	0.15	0.051	20
515.69	0.20	0.028	30
533.17	0.15	0.074	20

TABLE II-2 (Contd.)

GAMMA-RAY ENERGIES AND INTENSITIES

<u>Energy (keV)</u>	<u>Error</u>	<u>Photon Intensity (percent of decays)</u>	<u>Percentage Error (10 percent unless noted)</u>
538.01	0.03	0.093	30
540.49	0.03	6.6	---
556.40	0.10	1.00	20
579.25	0.07	0.077	---
583.07	0.07	0.072	---
594.48	0.10	0.028	20
598.06	0.15	0.033	20
630.24	0.07	0.19	---
635.47	0.07	0.096	---
654.82	0.03	8.4	---
661.08	0.03	0.016	25
671.06	1.0	0.012	100
673.06	0.05	0.014	100
675.08	0.02	0.028	20
681.29	0.02	0.011	20
686.96	0.07	0.092	---
696.29	0.05	0.17	---
712.62	0.07	0.072	---
718.40	0.10	0.026	---
727.90	0.15	0.019	---
740.61	0.10	0.017	20
749.68	0.15	0.012	---
754.33	0.10	0.030	---
758.06	0.03	0.014	50
761.45	0.15	0.026	20
768.17	0.10	0.059	---
781.42	0.20	0.006	30
786.74	0.10	0.011	20
793.43	0.10	0.022	20
795.84	0.20	0.006	50
808.80	0.10	0.146	---

TABLE II-2 (Contd.)

GAMMA-RAY ENERGIES AND INTENSITIES

<u>Energy (keV)</u>	<u>Error</u>	<u>Photon Intensity (percent of decays)</u>	<u>Percentage Error (10 percent unless noted)</u>
809.08	0.05	0.025	100
813.02	0.05	0.008	30
829.04	0.04	0.006	50
832.20	0.10	0.019	---
837.41	0.10	0.030	---
842.86	0.10	0.048	---
849.93	0.10	0.022	---
859.43	0.20	0.013	20
861.54	0.10	0.019	20
865.03	0.15	0.018	---
871.40	0.10	0.032	---
873.08	0.02	0.0052	50
886.59	0.03	0.0073	20
907.70	0.02	0.0063	50
915.03	0.03	0.0045	50
923.88	0.08	0.099	---
929.44	0.15	0.012	---
935.94	0.02	0.0056	20
938.77	0.02	0.0060	20
945.82	0.10	0.021	---
951.96	0.15	0.0086	---
964.00	0.15	0.018	---
967.44	0.15	0.0094	20
971.07	0.03	0.0033	20
979.03	0.08	0.096	---
986.07	0.03	0.0033	20
992.09	0.02	0.013	---
1022.79	0.08	0.105	---
1027.19	0.10	0.0098	20
1031.08	0.02	0.0041	20
1041.97	0.10	0.023	---

TABLE II-2 (Contd.)

GAMMA-RAY ENERGIES AND INTENSITIES

Energy (keV)	Error	Photon Intensity (percent of decays)	Percentage Error (10 percent unless noted)
1051.09	0.03	0.0037	20
1076.05	0.15	0.021	20
1078.78	0.10	0.058	---
1100.78	0.10	0.052	---
1123.06	0.04	0.014	50
1125.41	0.15	0.025	---
1128.05	0.03	0.0034	20
1135.09	0.02	0.0021	20
1141.08	0.02	0.0034	20
1150.01	0.02	0.0030	20
1172.01	0.03	0.0044	20
1175.07	0.02	0.0044	20
1190.04	0.02	0.0029	20
1197.08	0.02	0.0066	20
1202.03	0.02	0.0019	30
1225.07	0.03	0.0014	20
1234.11	0.10	0.250	---
1239.00	0.06	0.0014	50
1259.06	0.02	0.0033	20
1264.00	0.02	0.0060	20
1280.03	0.03	0.0010	20
1284.03	0.03	0.0007	20
129.10	0.15	0.0039	20
1298.04	0.03	0.0015	20
1312.13	0.15	0.0068	---
1357.02	0.40	0.0021	20
1381.05	0.03	0.0021	20
1448.01	0.05	0.0004	30
1454.03	0.04	0.0011	20
1473.09	0.05	0.0003	50
1495.8	0.03	0.0008	30
1568.04	0.04	0.0005	30

Fig. II-6 Gamma-ray spectrum of Nd-149 measured with an 8-mm by 2.5-cm² Ge(Li) detector. The low-energy portion of the spectrum was attenuated with an absorber consisting of 6 g/cm² of Pb, 0.6 g/cm² of Ta, and 0.7 g/cm² of Cd.

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Fig. II-7 The well-determined portion of the proposed decay scheme of Nd-149. The energies are in keV, and the transition intensities are in percent of decays. The intensities of the beta-ray branches are deduced from the gamma-ray intensities. The ground-state spins have been measured. The other spins, parities, and multipolarities are taken from Reference 9 and are consistent with those in Reference 4.

Fig. II-8 The complete proposed decay scheme (see caption of Figure II-7).

are included in the decay scheme. These multipolarities assignments are consistent with those previously reported [4], but are more extensive and more definitive. The transition intensities shown in the decay scheme are based on the photon intensities in Table II-2 and the conversion coefficients in Reference 9.

A complete proposed decay scheme is shown in Figure II-8. The well-determined section is shown on the left in abbreviated form. The center section includes several cascades which have been observed to be in coincidence. Also several levels are included where the transition energies seem to fit together very well. The right-hand section includes several possible levels which are determined only by energy combinations. In this case, it is possible that some levels are accidental energy combinations.

The conversion-electron data of Reference 9 determine the multipolarities of several transitions shown in the center section. From these multipolarities, several possible spin and parity assignments are deduced and are shown at the right of the center section of Figure II-8.

The internal consistency of the gamma-ray energies is indicated by the agreement of the various energy sums which add up to the same level. Such comparisons are given in Table II-3. It can be seen that the differences in

TABLE II-3

COMPARISON OF CASCADE ENERGY SUMS AND CROSSOVER ENERGIES
(All cascade and crossover energies are transition energies from Table II-2 unless otherwise noted.)

Cascade Energies (keV)		Energy Sum (keV)	Crossover Energy (keV)	Sum-Crossover Difference (keV)	Total Spread in Level Energy (keV)
114.31	± 74.32 ^[a]	188.63	188.64	- 0.01	0.01
114.31	± 97.00	211.31	211.32	- 0.01	0.01
240.23	± 30.00 ^[a]	270.23	----	+ 0.05	----
211.32	± 58.90	270.22	270.18	+ 0.04	0.05
114.31	± 155.89	270.20	----	+ 0.02	----
114.31	± 245.71	360.02	360.05	- 0.03	0.03
188.64	± 198.93	387.57	----	----	0.02
114.31	± 273.28	387.59	----	----	0.02
270.18	± 126.62	396.80	396.77	+ 0.03	0.05
211.32	± 185.50	396.82	396.77	+ 0.05	0.05
188.64	± 208.15	396.79	396.77	+ 0.02	0.05
114.31	± 282.47	396.78	396.77	+ 0.01	0.05
288.21	± 137.02	425.23	425.06 ^[b]	----	0.11
211.32	± 214.02	425.34	425.06 ^[b]	----	0.11
114.31	± 310.99	425.30	425.06 ^[b]	----	0.11
387.57 ^[c]	± 74.66 ^[a]	462.23	----	----	----
114.31	± 347.00	462.01	----	----	----
270.18	± 192.03	462.21	----	----	----

TABLE II-3 (Contd.)

COMPARISON OF CASCADE ENERGY SUMS AND CROSSOVER ENERGIES
(All cascade and crossover energies are transition
energies from Table II-2 unless otherwise noted.)

Cascade Energies (keV)	Energy Sum (keV)	Crossover Energy (keV)	Sum-Crossover Difference (keV)	Total Spread in Level Energy (keV)
114.31 ± 423.54	537.85	538.00 ^[b]	----	0.04
188.64 ± 349.23	537.87	538.00 ^[b]	----	0.04
211.32 ± 326.57	537.89	538.00 ^[b]	----	0.04
270.18 ± 267.70	537.88	538.00 ^[b]	----	0.04
360.05 ± 177.82	537.87	538.00 ^[b]	----	0.04
114.31 ± 540.49	654.80	654.82	- 0.02	0.10
270.18 ± 384.69	654.87	654.82	+ 0.05	0.10
211.32 ± 443.53	654.85	654.82	+ 0.03	0.10
288.21 ± 366.64	654.85	654.82	+ 0.03	0.10
360.05 ^[c] ± 294.82	654.87	654.82	+ 0.05	0.10
537.87 ^[c] ± 117.03	654.90	654.82	+ 0.08	0.10
396.77 ± 258.07	654.84	654.82	+ 0.02	0.10
211.32 ± 1022.81	1234.13	1234.11	+ 0.02	0.21
270.18 ± 964.04	1234.22	1234.11	+ 0.11	0.21
288.21 ± 954.84	1234.05	1234.11	- 0.06	0.21
360.05 ± 874.03	1234.08	1234.11	- 0.03	0.21
396.77 ± 837.46	1234.23	1234.11	+ 0.12	0.21
537.87 ^[c] ± 696.29	1234.16	1234.11	+ 0.05	0.21
654.82 ± 579.20	1234.02	1234.11	- 0.09	0.21
114.31 ± 1197.84	1312.15 ^[d]	1312.08	+ 0.07	0.12
188.64 ± 1123.84	1312.48 ^[d]	1312.08	+ 0.40	0.12
211.32 ± 1100.78	1312.10	1312.08	+ 0.02	0.12
270.18 ± 1041.97	1312.15	1312.08	+ 0.07	0.12
360.05 ± 951.99	1312.04	1312.08	- 0.04	0.12
396.77 ± 915.37	1312.14	1312.08	+ 0.06	0.12
188.64 ± 842.90	1031.54	1031.78	- 0.24	0.24
270.18 ± 761.52	1031.70	1031.78	- 0.08	0.24
537.87 ^[c] ± 493.86	1031.73	1031.78	- 0.05	0.24
360.05 ^[c] ± 675.78	1035.83	----	----	0.32
537.87 ^[c] ± 497.64	1035.51	----	----	0.32
654.85 ± 380.94	1035.79	----	----	0.32
114.31 ± 1027.21	1141.52	1141.77	- 0.25	0.25
270.18 ± 871.44	1141.62	1141.77	- 0.15	0.25
360.05 ± 781.45	1141.50	1141.77	- 0.27	0.25
114.31 ± 1075.99	1190.30	1190.33	+ 0.03	0.07
211.32 ± 979.05	1190.37	1190.33	- 0.04	0.07
114.31 ± 1175.69	1290.00	1290.02	- 0.02	0.34
211.32 ± 1078.79	1290.11	1290.02	+ 0.09	0.34
654.82 ± 635.52	1290.34	1290.02	+ 0.32	0.34
188.64 ± 1259.58	1448.22	1447.97 ^[b]	----	0.46
396.77 ± 1051.91	1448.68	1447.97 ^[b]	----	0.46
654.82 ± 793.49	1448.31	1447.97 ^[b]	----	0.46

TABLE II-3 (Contd.)

COMPARISON OF CASCADE ENERGY SUMS AND CROSSOVER ENERGIES
(All cascade and crossover energies are transition energies from Table II-2 unless otherwise noted.)

Cascade Energies (keV)	Energy Sum (keV)	Crossover Energy (keV)	Sum-Crossover Difference (keV)	Total Spread in Level Energy (keV)
114.31 ± 1381.36	1495.67	1495.69	- 0.02	0.43
211.32 ± 1284.25	1495.57	1495.69	- 0.12	0.43
270.18 ± 1225.64	1495.82	1495.69	+ 0.13	0.43
360.05 ± 1135.95	1496.00	1495.69	+ 0.31	0.43
114.31 ± 1454.20	1568.51	1568.20 ^[b]	----	0.34
211.32 ± 1357.12	1568.44	1568.20 ^[b]	----	0.34
270.18 ± 1298.48	1568.66	1568.20 ^[b]	----	0.34
288.21 ± 1280.25	1568.46	1568.20 ^[b]	----	0.34
396.77 ± 1172.01	1568.78	1568.20 ^[b]	----	0.34

[a] Energy taken from Reference 9.

[b] The accuracy of the crossover transition energy is much less than that of the energy sums; therefore, the energy differences in Column 5 are not significant and those in Column 6 are computed without considering the crossover energy.

[c] This is a level energy.

[d] The 188 and 1123 sum is not considered in the value computed in Column 6.

Columns 5 and 6 are generally consistent with the errors quoted in Table II-2. The absolute accuracy of the energies is indicated by a comparison in Table II-4 with the transition energies from conversion-electron studies of Reference 9. Again the differences are consistent with the quoted uncertainties.

TABLE II-4

COMPARISON OF TRANSITION ENERGIES WITH THOSE OF REFERENCE 9

Transition Energies		Difference	
Table II-2	Backlin et al ^[a]	ΔE (keV)	$\Delta E/s$
97.00 ± 0.03 ^[b]	97.03 ± 0.03 ^[c]	- 0.03	- 1.0
114.31 ± 0.03	114.32 ± 0.03	- 0.01	- 0.3
117.03 ± 0.20	117.06 ± 0.05	- 0.03	- 0.6
122.41 ± 0.05	122.34 ± 0.10	+ 0.07	+ 0.7
126.62 ± 0.05	126.64 ± 0.20	- 0.02	- 0.1
139.21 ± 0.05	139.02 ± 0.10	0	0
155.87 ± 0.03	155.87 ± 0.06	0	0

TABLE II-1 (Contd.)

COMPARISON OF TRANSITION ENERGIES WITH THOSE OF REFERENCE 9

Transition Energies		Difference	
Table II 2	Backlin et al ^[a]	ΔE (keV)	$\Delta E/s$
188.64 $\pm$ 0.03	188.62 $\pm$ 0.07	+ 0.02	+ 0.2
192.03 $\pm$ 0.05	191.96 $\pm$ 0.07	+ 0.07	+ 1.0
198.93 $\pm$ 0.03	198.93 $\pm$ 0.07	0	0
208.15 $\pm$ 0.05	208.09 $\pm$ 0.07	+ 0.06	+ 0.9
211.32 $\pm$ 0.03	211.28 $\pm$ 0.05	+ 0.04	+ 0.8
214.02 $\pm$ 0.07	213.85 $\pm$ 0.15	+ 0.17	+ 1.1
240.23 $\pm$ 0.03	240.20 $\pm$ 0.06	+ 0.03	+ 0.5
245.71 $\pm$ 0.05	245.76 $\pm$ 0.10	- 0.05	- 0.5
267.70 $\pm$ 0.03	267.65 $\pm$ 0.06	+ 0.05	+ 0.8
270.18 $\pm$ 0.03	270.12 $\pm$ 0.10	+ 0.06	+ 0.6
273.28 $\pm$ 0.05	273.01 $\pm$ 0.03	+ 0.02	+ 0.6
275.46 $\pm$ 0.05	275.25 $\pm$ 0.30	+ 0.21	+ 0.7
282.47 $\pm$ 0.05	282.39 $\pm$ 0.20	+ 0.08	+ 0.4
288.21 $\pm$ 0.05	288.12 $\pm$ 0.15	+ 0.09	+ 0.6
301.14 $\pm$ 0.05	301.04 $\pm$ 0.04	- 0.03	- 0.8
310.99 $\pm$ 0.05	311.11 $\pm$ 0.20	- 0.12	- 0.6
326.57 $\pm$ 0.03	326.45 $\pm$ 0.11	+ 0.12	+ 1.1
349.23 $\pm$ 0.03	349.26 $\pm$ 0.25	- 0.03	- 0.1
423.54 $\pm$ 0.03	423.54 $\pm$ 0.20	0	0
540.49 $\pm$ 0.03	540.06 $\pm$ 0.03	- 0.01	- 0.3
654.82 $\pm$ 0.03	654.04 $\pm$ 0.05	+ 0.04	+ 0.8

[a] Reference 9.

[b] Estimated standard deviation ($\approx \sigma$).[c] Estimated error limit ($s \approx 2$ standard deviations).

3.2 Decay of Hf-183

The identification of the radioactive nuclide Hf-183 was first reported by Gatti and Flegenhimer [10]. The activity, which was produced by the $W(n,\alpha)$ reaction, had a half-life of (64 ± 3) min. The same reaction was used by Poularikas et al [11] to produce a (66 ± 4) min activity. Recently, a decay scheme has been proposed by Bakru and Mukerjee [12]. Blachot et al [13] report the results of gamma-gamma coincidence measurements and an analysis of the beta-ray spectrum obtained with a scintillation detector. The reported beta-ray energies (and intensities) are 1570 ± 100 keV (25 percent), 1200 ± 100 keV (50 percent), and $650 \pm$ keV (25 percent). The latter two studies also produced sources by the (n,α) reaction.

For this study, a source of Hf-182 was produced by double-neutron capture in Hf-180. A sample of about 100 mg of normal hafnium oxide was irradiated for 1 year in the Materials Testing Reactor in a flux of about 5×10^{14} neutrons/cm²-sec⁻¹. This sample was then mass separated in the Argonne Isotope Separator. It is estimated that the mass-182 part of the separated sample contained about 40 micrograms of hafnium.

Since this small amount of material was irradiated several times to produce Hf-183, the chemical processing of it was of special importance. The initial processing involved, in part, the removal of aluminum. After that the following procedure was used: The Hf activity was purified by selective elution with HCl from an anion resin column; the Hf fraction was made 8N in HCl then extracted into 100 percent tributyl phosphate; after washing three times with 8N HCl, the Hf was back-extracted into water and then carried on Fe (OH)₃ precipitated with ammonia; and the dried Fe (OH)₃ carrier was subsequently packaged for re-irradiation.

Gamma-ray spectra were obtained with a 3- by 3-inch NaI(Tl) scintillation system and a 0.8-cm by 2.5-cm² Ge(Li) detector system. The latter employed a 4096-channel analyzer. The linearity of the complete Ge(Li) detector system has been determined by the method described in Reference 14. The correction to the observed pulse height is shown in Figure II-9. For this detector, the photopack

Fig. II-9 Linearity correction to observed pulse height for 4096-channel analyzer with Ge(Li) detector (data from W. W. Black).

and double-escape peak efficiencies have been determined by comparison of photopeak areas with those obtained with a NaI(Tl) detector. The resulting efficiency curve is shown in Figure II-10. Although these absolute efficiencies apply to a source-detector distance of 3.0 cm, the relative efficiencies are expected to be quite accurate for other distances.

Fig. II-10 Photopeak and double-escape peak efficiency for 0.8-cm by 2.5-cm² Ge(Li) detector with 3.0-cm source-detector distance.

Gamma-gamma coincidence measurements were made with two 30 by 3-inch NaI(Tl) detectors and a 256- by 256-channel, two-parameter analyzer.

3.3 Experimental Results

A gamma-ray spectrum measured with the Ge(Li) detector is shown in Figure II-11. The presence of peaks from Hf-180m and Hf-181 result from incomplete isotopic separation. The peaks of the daughter Ta-183 grow, as expected for a sample which was pure Hf-183, about 1 hour before the start of the count. The Mn-56 and As-76 lines result from irradiation of the iron carrier used in the chemical processing and the arsenic impurity in the iron. The origin of the

Fig. II-11 Gamma-ray spectrum of Hf-183 measured with Ge(Li) detector. The energies are in keV.

Zr-97 - Nb-97 is not known, but zirconium is a common impurity in hafnium. Other spectra were obtained in which the Mn-56 and As-76 peaks were not observable.

The relative gamma-ray intensities were determined from the data in Figure II-11. The energies were determined for the strong transitions from similar spectra of mixed sources of Hf-183, Am-241 (59.568), Gd-153 (97.43 and 103.18), Lu-177 (112.955 and 208.365), Hg-203 (279.15), and Au-198 (411.795 keV) and Hf-183, Hg-203, Au-198, Na-22 (511.006), Cs-137 (661.60) Mn-54 (834.84), and Co-60 (1173.23 and 1332.48 keV. The energies of the weaker transitions were obtained from the Hf-183 spectrum by calibrating with the strong Hf-183 and Ta-183 lines.

The absolute gamma-ray intensities were determined by comparing the intensities of the Hf-183 and Ta-183 lines in three successive spectra obtained with a single source. The growth of the Ta-183 lines was used to determine the time at which the source would have been pure Hf-183. The Hf-183 intensities were compared with that of the 246-keV gamma ray from Ta-183 which was taken to represent 26.2 percent of the Ta-183 decays [15]. From these relative intensities and the appropriate growth and decay equations, the absolute intensities were determined. These intensities and energies are given in Table II-5.

TABLE II-5

GAMMA-RAY ENERGIES AND INTENSITIES

(The transition intensities are for the multipolarities E1 for 73 and M1+E2 for 113 and 143 keV.)

<u>Transition Energy</u> (keV)	<u>Photon Intensities</u> (percent of decays)	<u>Transition Intensity</u> (percent of decays)
73.08 $\pm$ 0.05	38.00 $\pm$ 5.00	67.00 $\pm$ 9.00
113.75 $\pm$ 0.01	0.16 $\pm$ 0.02	0.46 $\pm$ 0.75
143.01 $\pm$ 0.02	0.50 $\pm$ 0.15	0.07 $\pm$ 1.07
160.07 $\pm$ 0.02	0.05 $\pm$ 0.02	
165.00 $\pm$ 0.02	0.06 $\pm$ 0.03	
223.05 $\pm$ 0.02	0.09 $\pm$ 0.05	
224.99 $\pm$ 0.01	0.17 $\pm$ 0.04	
284.01 $\pm$ 0.01	0.38 $\pm$ 0.06	
315.86 $\pm$ 0.05	1.23 $\pm$ 0.15	
397.86 $\pm$ 0.05	3.04 $\pm$ 0.04	
459.08 $\pm$ 0.05	30.00 $\pm$ 2.00	
476.93 $\pm$ 0.01	0.77 $\pm$ 0.01	
481.04 $\pm$ 0.02	0.60 $\pm$ 0.15	
686.47 $\pm$ 0.01	0.30 $\pm$ 0.06	
691.69 $\pm$ 0.01	0.30 $\pm$ 0.06	
716.09 $\pm$ 0.02	0.10 $\pm$ 0.03	

TABLE II-5 (Contd.)

GAMMA-RAY ENERGIES AND INTENSITIES
(The transition intensities are for the
multipolarities E1 for 73 and M1+E2 for 113 and 143 keV.)

Transition Energy (keV)	Photon Intensities (percent of decays)	Transition Intensity (percent of decays)
735.14 $\pm$ 0.08	0.93 $\pm$ 0.10	
783.81 $\pm$ 0.05	65.00	
806.57 $\pm$ 0.15	0.17 $\pm$ 0.04	
841.02 $\pm$ 0.15	0.18 $\pm$ 0.04	
856.81 $\pm$ 0.08	0.07 $\pm$ 0.02	
1091.49 $\pm$ 0.15	0.12 $\pm$ 0.12	
1470.38 $\pm$ 0.08	2.03 $\pm$ 0.03	
1784.03 $\pm$ 0.02	0.06 $\pm$ 0.03	

Only the relative errors are given for the intensities. A 15 percent normalization error should be included to get the total uncertainty. The transition intensities were computed on the assumption that the 73-keV transition is E1 and that those at 113 and 143 keV are M1 + E2.

Some gamma-gamma coincidence relationships were determined from the NaI(Tl) singles spectra shown in Figure II-12. It is clear from the coincidence sum peaks that the 73-keV transition is in coincidence with those at 783 and 1470 keV and is not in coincidence with the one at 459 keV.

These coincidence relationships were verified, and others were established by a gamma-gamma coincidence measurement. The analyses of two of the coincidence spectra are shown in Figure II-13. The coincidence relations determined from this experiment are listed in Table II-6.

If the K X-ray peak in Figure II-11 is assumed to result completely from conversion of the 73-keV transition, the K conversion of this transition can be computed. The resulting value is $\alpha_K = 0.58$. The theoretical conversion coefficients [16,17] for various multipolarities are shown in Figure 14. These data indicate this transition has E1 character. This conclusion also follows from the fact that any other multipolarity would yield a transition intensity of > 400 percent of the decays.

A tentative proposed decay scheme is shown in Figure II-15. The order of the transitions in the 315-143-keV and the 284-113-keV cascades is not uniquely determined from the experiment. The intensities of the beta-ray branches are computed from the gamma-ray intensities. There are eleven gamma-ray transitions in Table II-5 which are not included in this scheme.

Fig. II-12 Gamma-ray spectra of Hf-183 measured with a NaI(Tl) scintillation spectrometer. The source-detector distances are both 1 cm. The lower curve was obtained with an absorber including 0.3 g/cm² Pb and 0.6 g/cm² Ta to eliminate the 73-keV gamma rays.

The 856-keV level is presumed to be a gamma-vibrational state with $K' = K-2$, based on the 9/2⁻[514] level at 73 keV. The large ratio of the intensities of the 783- and 856-keV transitions supports this assumption. Part of the particle configuration making up this state may be the three-particle configuration [21],

There is one apparent coincidence relation that is not in agreement with this scheme. The 735-keV transition appears to be in coincidence with the 284-keV transition, but not with the one at 397 keV. Interchanging the 113- and 284-keV gamma rays does not solve the problem since the 735-keV transition is more intense than the 284-keV transition.

Although no spins have been determined experimentally, a set of reasonable assignments can be made. The ground-state spin and parity of Hf-183 are expected to be 3/2⁻ or 1/2⁻, as is observed [18] for W-185 or Os-187, respectively. The observed beta-decay intensities along with the spins in Ta-183 discussed below suggest 3/2⁻. Assuming the Nilsson model [19] applies this should be the 3/2⁻[512] neutron state.

The ground-state spin of Ta-183 has been measured [18] to be 7/2. The data compiled by Bunker and Reich [20] suggest that this should be the 7/2⁺[404] proton state and that the 9/2⁻[514] level should be a low-energy excited state. The latter is taken to be the 73-keV level. This is in agreement with the multipolarity of the 73-keV transition. The 143-keV level is interpreted as the 9/2⁺ rotational state based on the ground state. The value of $\frac{\hbar^2}{2I} = 15.9$ keV is consistent with the observed values for bands based on this state [20]. (Since the order of the 315- and 143-keV transitions is not known, the second excited state could occur at 315 keV.

The 459-keV level is assumed to be the 5/2⁺[402] level. The energy of this state is in agreement with that expected in Reference 20. Also the log ft value of 7.0 is reasonable for the unhindered, first-forbidden beta decay to this state.

Fig. II-13 Gamma-gamma coincidence spectra obtained with two NaI(Tl) detectors. The energies of the gating channel are shown, and the portion of the decay scheme assumed to be producing the coincidences is shown.

TABLE II-6

GAMMA-GAMMA COINCIDENCE RESULTS

(An X represents an observed coincidence

relationship and an O one which is not observed.

A blank indicates a lack of definitive information.)

E_{γ} (keV)								
E_{γ} (keV)	73	113	143	223 225	284	315	397	459
284		X				X		X
315			X				X	
397	O					X		X
459	O	X	O	X	X	O	X	O
686 and 691								X
735				X(?)	X	O	O	O
783	X				O	O	O	O
1480	X					O	O	O

$$9/2^- [514]_p + \left\{ \begin{array}{l} 5/2^+ [402]_p \\ + 9/2^+ [404]_p \end{array} \right\} 2^+ .$$

A small amount of this configuration would allow the transition to the $5/2^+ [502]_p$ state at 459 keV. Another part [21] (perhaps ≈ 25 percent) of the 856-keV state might be

$$9/2^- [514]_p + \left\{ \begin{array}{l} 3/2^- [512]_n \\ + 7/2^- [514]_n \end{array} \right\} 2^+ .$$

The beta decay from Hf-183 would then proceed by the

$$7/2^- [514]_n \rightarrow 9/2^- [514]_p$$

transition which is an allowed unhindered decay. This would explain qualitatively the log ft value to this level.

The log ft value of 6.0 to the 1543-keV level requires that this state has $I^\pi = 1/2^-, 3/2^-,$ or $5/2^-$. The strong transition to the $9/2^-$ state then suggests an assignment of $5/2^-$.

Fig. II-14 Plot of theoretical K-shell internal conversion coefficients [15,16] for various multipolarities. The threshold values [16] have been interpolated to $Z = 73$.

herein.

(E. Fast)

4.1 AIM-6 Diffusion Code Calculations (D. A. Millsap, E. Fast)

a volume of 46 ml.

TABLE II-7

COMPARISON OF
AIM-6 CALCULATED WITH
MEASURED REACTIVITIES

Material	Reactivity in μK for 46-ml Sample		
	Calculated 4-Group	Calculated 18-Group	Measured
ARMF-I			
H ₂ O	- 232.4	- 226.2	- 194.7
D ₂ O	- 43.0	- 46.4	- 50.4
Be	- 13.4	- 15.4	- 27.5
C	- 12.7	- 13.9	- 15.8
Mg	- 21.8	- 22.4	- 15.8
Al	- 58.1	- 58.3	- 50.0
Zr	- 48.2	- 40.3	- 38.7
Pb	- 24.5	- 25.4	- 26.0
Bi	- 8.3	- 8.8	- 9.3
ARMF-II			
H ₂ O	244.4	222.5	76.1
D ₂ O	400.0	431.7	437.7
Be	410.6	451.2	449.4
C	89.3	115.6	123.5
Mg	- 161.1	- 148.1	- 2.9
Al	- 292.0	- 266.2	- 237.8
Zr	- 377.8	- 221.1	- 93.7
Pb	- 88.7	- 56.2	- 59.9
Bi	27.4	57.1	59.9

4.2 Eta of Plutonium Fissile Isotopes (E. Fast, E. F. Aber)

The neutron reproduction values, eta, for the plutonium fissile isotopes Pu-239 and Pu-241, relative to U-235, have been measured in the Advanced Reactivity Measurement Facilities, ARMF-I and -II. Samples were made up as 4N H₂SO₄ solutions in a series of concentrations approaching infinite dilution. Analogous series of samples of pure absorbers gave information on the energy dependence of the absorption statistical weights necessary for correcting the reactivity measurements of the fissile isotopes. A detailed analysis of all sources of errors was made to obtain the quoted uncertainty. The reactivity measurement itself contributed only a minor part of this value.

In the tabulation below are summarized both the effective eta ratios obtained in the two facilities and also the overall weighted averages for the 2200-m/sec and Maxwellian results.

4.3 Relative Yields of I-135 in Fission (R. G. Nisle, I. E. Stepan)

The measurement of the cumulative yields of the mass -135 fission product chain to I-135 for U-233, Pu-239, and Pu-241 relative to that for U-235 is a part of a general study of fission products

	$\eta(\text{Pu } 239)/\eta(\text{U } 235)$	
	ARMF-I	ARMF-II
Effective	1.0013 $\pm$ 0.0095	0.9965 $\pm$ 0.0260
2200 m/sec	1.0191 $\pm$ 0.0087	
Maxwellian	1.0077 $\pm$ 0.0086	
	$\eta(\text{Pu-241})/\eta(\text{U-235})$	
Effective	1.0546 $\pm$ 0.0175	1.0584 $\pm$ 0.0130
2200 m/sec	1.0450 $\pm$ 0.0103	
Maxwellian	1.0542 $\pm$ 0.0103	

currently underway at the Advanced Reactivity Measurement Facilities (ARMF-I and -II). The method may be divided into four phases: (a) irradiation of the sample and subsequent measurement of its transient reactivity in the ARMF reactors; (b) fuel and poison calibration of the ARMF reactors; (c) reduction of the raw reactivity data; (d) analysis of reduced data to obtain the respective yield ratios and to determine uncertainties.

The sample is tubular in shape to permit cooling on both inner and outer surfaces. The active isotopes are alloyed with, and clad in, high-purity aluminum. Irradiations were made in a Materials Testing Reactor rabbit facility for about 72 hours at a flux level of 1.6×10^{14} nv. Subsequent to irradiation, reactivity measurements were made in both ARMF-I and -II. Both reactors were calibrated by measuring fuel and poison standard samples containing known amounts of the various fissile isotopes and using Li-6 as a $1/v$ neutron absorber.

The reactivity data were converted to fuel and poison cross sections by use of the calibration factors in a computer code. The yield ratios were then calculated from the following equations:

$$\Sigma_p(t_i) = C_1 + C_2 \exp(-\lambda_I t_i) + C_3 \exp(-\lambda_{Xe} t_i) + C_4 \exp(-\lambda_{Pm} t_i) + C_5 \exp(-\lambda_{Rh} t_i) + C_6 \exp(-\lambda_{Ru} t_i) \quad (3)$$

$$C_2 = \sigma_{Xe} F_o \sigma_f \phi \exp(-\sigma_a \phi T) \frac{\gamma_I}{\lambda_I - \lambda_{Xe}} \quad (4)$$

and

$$\frac{[\gamma_I]_x}{[\gamma_I]_U} = \frac{[C_2]_x}{[C_2]_U} \frac{[F_o \sigma_f \phi \exp(-\sigma_a \phi T)]_U}{[F_o \sigma_f \phi \exp(-\sigma_a \phi T)]_x} \quad (5)$$

where

$\Sigma_p(t_i)$ is the poison cross section of the irradiated sample derived from reactivity measurements.

$C_1, C_2, \dots$ etc., are functions of parameters such as fission yields, decay constants, sample content, and fission cross section. C_2 is the only one of interest here and is given by Equation (2).

λ 's are the decay constants of the isotopes I-135, Xe-135, Pm-149, Rh-105, and Ru-105 indicated by the subscripts.

t_i is the sequence of measurement times subsequent to removal of the sample from the irradiating flux.

σ_{Xe} is the 2200 m/sec cross section of Xe-135.

F_0 is the number of fissile atoms initially present.

σ_f is the effective fission cross section in the irradiating flux, ϕ .

σ_a is the effective absorption cross section of the fissile isotope in the irradiating flux, ϕ .

T is the duration of the irradiation at the constant flux level, ϕ .

γ_I is the cumulative fission yield of the mass-135 chain to I-135.

The results are shown in Table II-8 together with results published by other investigators. Each of the yield ratios reported in this paper is an average of

TABLE II-8

YIELD RATIOS OF I-131 IN THE
FISSION OF VARIOUS ISOTOPES RELATIVE TO U-235

	This Paper	Nisile and Stepan ^[22]	Okazaki and Walker ^[23]	Weaver et al ^[24]
U-233	$0.768 \pm 0.160^{[a]}$	$0.700 \pm 0.156^{[b]}$	$0.764 \pm 0.011^{[c]}$	0.749
Pu-239	$1.006 \pm 0.067^{[b]}$	$1.204 \pm 0.214^{[b]}$	$0.955 \pm 0.014^{[c]}$	1.024
Pu-241	$1.221 \pm 0.089^{[b]}$	----	$1.205 \pm 0.030^{[c]}$	----

[a] Average of three values -- confidence interval at the 90 percent level.

[b] Average of two values -- confidence interval at the 90 percent level.

[c] Uncertainties quoted as rms deviations.

two or more determinations based on separate complete sets of transient reactivity measurements made on different samples. For any one fissile isotope, the different samples were of very nearly the same composition. The uncertainties quoted are confidence intervals at the 90 percent confidence level, based on standard deviations from the average of the separate determinations.

4.4 Resonance Integrals (J. W. Rogers, J. J. Scoville)

Measurements of the infinite dilution resonance integrals of several elements and some of their isotopes have been made in the ARMF-II cadmium tube and are given in Table II-9. Since some of these materials have resonances between cadmium cutoff and the 4.91-eV resonance of the gold standard where the neutron importance varies appreciably, it was necessary to make some corrections to the experimental values. These corrections were made using the interpretation and calculations of Foell [25] and of Kaufman [26], and the computer code COMBCO [27]. They are subject to rather high uncertainties due to the uncertainties in the resonance parameters. The infinite dilution resonance integrals of these materials were calculated using parameters for only the resolved resonances. These calculated values are also recorded in Table II-9.

TABLE II-9

INFINITE DILUTION RESONANCE INTEGRALS

Material	Measured Value	Correction	Corrected Value	Calculation
As	81 ± 3	0	81 ± 3	54
Ru	82 ± 3	0	82 ± 3	43
Pd	121 ± 5	0	121 ± 5	105
La	19 ± 1	0	19 ± 1	10
Eu	3037 ± 92	1899	4936 ± 850	5308
Eu-151	4050 ± 129	3850	7900 ± 2000	9777
Eu-153	1780 ± 55	112	1890 ± 110	1212
Lu	830 ± 25	14 ^[a]	843 ± 35	542 ^[a]
Lu-175	874 ± 28	12	886 ± 35	536
Lu-176	None	80 ^[a]	-----	758 ^[a]
Ta	715 ± 22	19	734 ± 35	688
Os	209 ± 8	0	209 ± 8	181
Ir	1774 ± 54	558	2330 ± 325	1974
Ir-191	None	1206	-----	3116
Ir-193	None	173	-----	1294
Np-237	863 ± 28	213	1075 ± 130	649
Hf-176	400 ± 21	0	400 ± 21	-----
Hf-177	7670 ± 230	1015	8685 ± 760	7340
Hf-178	1330 ± 40	0	1330 ± 40	1810
Hf-179	640 ± 20	0	640 ± 20	397
Hf-180	11 ± 6	0	11 ± 6	28
Hf	2130 ± 60	187	2315 ± 150	2050

[a] The 0.142-eV resonance is not included.

4.5 Hafnium Irradiation Studies (J. W. Rogers, J. J. Scoville)

Hafnium-aluminum alloy samples of 0.0, 1.0, 2.5, and 5.0 wt % hafnium have been irradiated in the MTR up to about 6×10^{21} n/cm² (thermal). The samples were of hollow cylindrical geometry, 0.86 inch diameter and 4 inches long, with a 0.030-inch wall thickness, weighing a total of about 15 grams. Reactivity measurements of these samples were made in the ARMF-I water hole at various intervals of irradiation to measure the changes in reactivity. Figure II-16 shows

Fig. II-16 Reactivity response as a function of irradiation for hafnium-aluminum alloy samples.

the reactivity response as a function of irradiation for these samples. Approximately 65 percent reduction in the reactivity effect of the hafnium was observed in all samples, indicating little self-shielding. No reactivity change was observed in the blank aluminum sample. The deviation of the 1-percent sample data from the expected curve is attributed to a faulty capsule. Neutron exposure uncertainties are about ± 6 percent.

Solid hafnium slugs of 0.282 inch diameter of 0.564 inch diameter have been irradiated to 1.4×10^{21} n/cm² (thermal) the reactivity of both samples was reduced by only 5 μ k indicating that the irradiation is affecting both samples about the same. The flux monitors indicated strong perturbation due to the samples which is to be expected. These samples are receiving further irradiation in the MTR.

5. THEORETICAL PHYSICS (H. L. McMurry)

5.1 Scattering Kernels for Water at Elevated Temperatures (H. L. McMurry)

The McMurry-Russel (MR) model for water gives calculated scattering in agreement with Brugger's room-temperature data [28]. Simple modifications in the parameters have been made to optimize agreement with Haywood's data [29] for water at 150°C and Page's data [30] for water at 277°C.

The MR model treats water as a mixture of 10 percent free H₂O molecules and 45 percent for each of two aggregates in which the effective scattering masses for H atoms are 75 and 150. Each of the larger aggregates exhibits internal vibrational modes which help to determine the scattering.

To optimize agreement with the high-temperature data, the fraction of free H₂O molecules was adjusted upward to 25 percent for the 150°C water and 28 percent for the 277°C water. Equal parts of the large aggregates were assumed. This work has been reported in detail [31].

5.2 Scattering Kernels for D₂O

The MR model for water at 20°C has been modified to fit D₂O by adjusting some of the internal frequencies and vibration amplitudes in the aggregates in a reasonable way and by altering the fraction of free D₂O molecules to optimize agreement with Haywood's data [32]. Further alteration of the fraction of free D₂O molecules has been used to optimize agreement with Haywood's 150°C data. Interference scattering is neglected. The results have been reported in detail [33].

5.3 Preparation of Water Kernels for the ENDF File

Two problems are associated with preparing scattering kernels for water based on the MR models for the ENDF file at BNL [34]. One is simply that of translating the results from the computational program used here so they are in the conventional ENDF format. There are no real problems here. The second arises from the fact that the MR model gives calculated $S(\kappa^2, \beta)$ versus β curves which exhibit sharp resonances at low κ^2 values. The resonances arise whenever $\beta \approx E_\tau/kT$ where E_τ is a characteristic vibration in the model. It may be undesirable to submit results showing such sharp structure to ENDF. Besides giving users a possible erroneous picture of the structure of water, it appears that to obtaining good interpolation more β points might be needed than are desirable.

To deal with the problem, the computational routines has been modified so that the $S(\kappa^2, \beta)$ data computed for the ENDF file are "smoothed" in the region of κ^2, β space where the resonances are sharp. The smoothing function has the form,

$$S_s(\kappa^2, \beta) = S_I(\kappa^2) e^{-C_I(\kappa^2) (\beta - \beta_{I-1/2})}; \beta_{I-1/2} \leq \beta \leq \beta_{I+1/2}.$$

In Equation (6), $S_s(\kappa^2, \beta)$ refers to the "smoothed" value to be submitted to ENDF. The variable, β , runs from $\beta_{I-1/2}$ (a value of β below the resonance which is at β_I) up to a value, $\beta_{I+1/2}$, which is above the resonance. The coefficient, $S_I(\kappa^2)$, is chosen, if possible, so that the smoothed function is continuous at the points, $\beta_{I\pm 1/2}$. The $C_I(\kappa^2)$ is chosen so that the area under $S_s(\kappa^2, \beta)$ $\beta_{I-1/2}$ and $\beta_{I+1/2}$ is the same as is obtained from the actual MR model.

The details of how this is accomplished are being reported in an IN report which is being prepared.

5.4 Slow Neutron Scattering by Crystals (A. W. Solbrig, INC; C. Noble, PPCo)

Calculations of the scattering of slow neutrons from atomic systems requires knowledge of the eigenstates and dynamical behaviors of the scattering atoms. One method for calculating molecular dynamics involves general quadratic valence force fields and uses symmetry coordinates to factor the secular equation. McMurry's version of this method for semi-rigid molecules [35] has been adapted to lattice dynamics calculations. This lattice dynamics calculation has been programmed in FORTRAN for the IBM-7040. The program has been checked by several methods.

The program is being used to calculate dispersion curves for BeO. It is hoped that a set of valence for constants can be found which will yield dispersion curves in agreement with the experimental curves of Brugger and Carpenter for BeO. The program is being extended to include calculation of structure factors for inelastic scattering of neutrons.

5.5 Pulsed Neutron Research (G. W. Griffing)

At the beginning of the reporting period, theoretical studies were started on pulsed neutron research. Primarily, this effort is intended to give theoretical support to an experimental program of pulsed neutron research. Hopefully, this program will lead to the development of a pulsed reactor. Experimentally, the program consists of pulsing a moderator with neutrons and observing the subsequent temporal and energetic behavior of the neutrons. The theoretical support consists of attempting to predict the observed neutron spectra or the life history of the neutrons. Fundamentally, it is believed that such phenomena are described by the Boltzmann equation. Significant theoretical and experimental research in this area has been going only for about 10 years; consequently, it is a relatively new field.

Much of the time during the reporting period was devoted to becoming familiar with the published experimental and theoretical researches. A computational program called TIDES was obtained from Rensselaer Polytechnic Institute, and a few explanatory calculations for water and polyethylene have been undertaken.

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ERRATA FOR IN-1048

NUCLEAR TECHNOLOGY BRANCHES QUARTERLY REPORT July 1 - September 30, 1966

Attached are substitute pages correcting the following tables:

Table II-2	-	Pages 35, 36, 37, 38, 39
Table II-3	-	Pages 43, 44, 45
Table II-4	-	Pages 45, 46
Table II-5	-	Pages 50, 51

TABLE II-2

GAMMA-RAY ENERGIES AND INTENSITIES

<u>Energy (keV)</u>	<u>Error</u>	<u>Photon Intensity (percent of decays)</u>	<u>Percentage Error (10 percent unless noted)</u>
30.00	0.10	0.15	---
58.90	0.03	1.3	20
65.37	0.05	0.041	20
74.3	0.2	2.2	---
74.7	0.2	2.2	20
75.55	0.2	2.2	---
91.12	0.05	0.063	20
97.00	0.03	1.3	---
107.76	0.10	0.055	20
114.31	0.03	16.1	---
116.9	0.2	0.10	20
122.41	0.05	0.20	---
126.62	0.05	0.10	---
137.02	0.08	0.040	20
139.21	0.05	0.41	---
155.87	0.03	5.2	---
177.82	0.05	0.14	---
185.50	0.08	0.079	20
188.64	0.03	1.7	---
192.03	0.05	0.51	---
198.93	0.03	1.25	---
208.15	0.05	2.5	---
211.32	0.03	23.4	---
213.95	0.15	0.35	---
226.85	0.05	0.14	20
229.57	0.05	0.43	---
240.23	0.03	3.4	---
245.71	0.05	0.89	---
250.84	0.10	0.032	20
254.20	0.07	0.075	20
258.07	0.05	0.33	---

TABLE II-2 (Contd.)

GAMMA-RAY ENERGIES AND INTENSITIES

Energy (keV)	Error	Photon Intensity (percent of decays)	Percentage Error (10 percent unless noted)
267.70	0.03	5.2	---
270.18	0.03	9.2	---
273.28	0.08	0.20	20
275.96	0.05	0.51	20
276.96	0.10	0.28	20
282.47	0.05	0.52	---
288.21	0.05	0.58	---
294.82	0.05	0.50	---
301.14	0.05	0.33	---
310.99	0.05	0.44	---
326.57	0.03	4.0	---
342.85	0.10	0.029	20
347.85	0.2	0.16	50
349.23	0.03	1.27	---
352.75	0.10	0.056	20
357.05	0.10	0.047	20
360.05	0.05	0.14	---
366.64	0.05	0.57	---
380.94	0.10	0.058	20
384.69	0.05	0.29	---
396.77	0.10	0.085	20
423.54	0.03	8.1	---
425.20	0.15	0.40	---
443.53	0.03	1.29	---
480.21	0.15	0.051	20
483.53	0.15	0.073	20
493.86	0.15	0.044	20
497.81	0.15	0.047	20
510.29	0.15	0.051	20
515.69	0.20	0.028	30
533.17	0.15	0.074	20

TABLE II-2 (Contd.)

GAMMA-RAY ENERGIES AND INTENSITIES

<u>Energy (keV)</u>	<u>Error</u>	<u>Photon Intensity (percent of decays)</u>	<u>Percentage Error (10 percent unless noted)</u>
538.1	0.3	0.093	30
540.49	0.03	6.6	---
556.40	0.10	1.00	20
579.25	0.07	0.077	---
583.07	0.07	0.072	---
594.48	0.10	0.028	20
598.06	0.15	0.033	20
630.24	0.07	0.19	---
635.47	0.07	0.096	---
654.82	0.03	8.4	---
661.8	0.3	0.016	25
671.6	1.0	0.012	100
673.6	0.5	0.014	100
675.8	0.2	0.028	20
681.29	0.2	0.011	20
686.96	0.07	0.092	---
696.29	0.05	0.17	---
712.62	0.07	0.072	---
718.40	0.10	0.026	---
727.90	0.15	0.019	---
740.61	0.10	0.017	20
749.68	0.15	0.012	---
754.33	0.10	0.030	---
758.6	0.3	0.014	50
761.45	0.15	0.026	20
768.17	0.10	0.059	---
781.42	0.20	0.006	30
786.74	0.10	0.011	20
793.43	0.10	0.022	20
795.84	0.20	0.006	50
808.80	0.10	0.146	---

TABLE II-2 (Contd.)

GAMMA-RAY ENERGIES AND INTENSITIES

<u>Energy (keV)</u>	<u>Error</u>	<u>Photon Intensity (percent of decays)</u>	<u>Percentage Error (10 percent unless noted)</u>
809.8	0.5	0.025	100
813.2	0.5	0.008	30
829.4	0.4	0.006	50
832.20	0.10	0.019	---
837.41	0.10	0.030	---
842.86	0.10	0.048	---
849.93	0.10	0.022	---
859.43	0.20	0.013	20
861.54	0.10	0.019	20
865.03	0.15	0.018	---
871.40	0.10	0.032	---
873.8	0.2	0.0052	50
886.59	0.3	0.0073	20
907.70	0.2	0.0063	50
915.3	0.3	0.0045	50
923.88	0.08	0.099	---
929.44	0.15	0.012	---
935.94	0.2	0.0056	20
938.77	0.2	0.0060	20
945.82	0.10	0.021	---
951.96	0.15	0.0086	---
964.00	0.15	0.018	---
967.44	0.15	0.0094	20
971.7	0.3	0.0033	20
979.03	0.08	0.096	---
986.7	0.3	0.0033	20
992.9	0.2	0.013	---
1022.79	0.08	0.105	---
1027.19	0.10	0.0098	20
1031.8	0.2	0.0041	20
1041.97	0.10	0.023	---

TABLE II-2 (Contd.)

GAMMA-RAY ENERGIES AND INTENSITIES

Energy (keV)	Error	Photon Intensity (percent of decays)	Percentage Error (10 percent unless noted)
1051.9	0.3	0.0037	20
1076.05	0.15	0.021	20
1078.78	0.10	0.058	---
1100.78	0.10	0.052	---
1123.6	0.4	0.014	50
1125.41	0.15	0.025	---
1128.5	0.3	0.0034	20
1135.9	0.2	0.0021	20
1141.8	0.2	0.0034	20
1150.1	0.2	0.0030	20
1172.1	0.3	0.0044	20
1175.7	0.2	0.0044	20
1190.4	0.2	0.0029	20
1197.8	0.2	0.0066	20
1202.3	0.2	0.0019	30
1225.7	0.3	0.0014	20
1234.11	0.10	0.250	---
1239.0	0.16	0.0014	50
1259.6	0.2	0.0033	20
1264.0	0.2	0.0060	20
1280.3	0.3	0.0010	20
1284.3	0.3	0.0007	20
1290.10	0.15	0.0039	20
1298.4	0.3	0.0015	20
1312.13	0.15	0.0068	---
1357.2	0.4	0.0021	20
1381.5	0.3	0.0021	20
1448.1	0.5	0.0004	30
1454.3	0.4	0.0011	20
1473.9	0.5	0.0003	50
1495.8	0.3	0.0008	30
1568.4	0.4	0.0005	30

are included in the decay scheme. These multipolarities assignments are consistent with those previously reported [4], but are more extensive and more definitive. The transition intensities shown in the decay scheme are based on the photon intensities in Table II-2 and the conversion coefficients in Reference 9.

A complete proposed decay scheme is shown in Figure II-8. The well-determined section is shown on the left in abbreviated form. The center section includes several cascades which have been observed to be in coincidence. Also several levels are included where the transition energies seem to fit together very well. The right-hand section includes several possible levels which are determined only by energy combinations. In this case, it is possible that some levels are accidental energy combinations.

The conversion-electron data of Reference 9 determine the multipolarities of several transitions shown in the center section. From these multipolarities, several possible spin and parity assignments are deduced and are shown at the right of the center section of Figure II-8.

The internal consistency of the gamma-ray energies is indicated by the agreement of the various energy sums which add up to the same level. Such comparisons are given in Table II-3. It can be seen that the differences in

TABLE II-3

COMPARISON OF CASCADE ENERGY SUMS AND CROSSOVER ENERGIES
(All cascade and crossover energies are transition energies from Table II-2 unless otherwise noted.)

Cascade Energies (keV)	Energy Sum (keV)	Crossover Energy (keV)	Sum-Crossover Difference (keV)	Total Spread in Level Energy (keV)
114.31 ± 74.32 ^[a]	188.63	188.64	- 0.01	0.01
114.31 ± 97.00	211.31	211.32	- 0.01	0.01
240.23 ± 30.00 ^[a]	270.23		+ 0.05	
211.32 ± 58.90	270.22	270.18	+ 0.04	0.05
114.31 ± 155.89	270.20		+ 0.02	
114.31 ± 245.71	360.02	360.05	- 0.03	0.03
188.64 ± 198.93	387.57		----	
114.31 ± 273.28	387.59		----	0.02
270.18 ± 126.62	396.80		+ 0.03	
211.32 ± 185.50	396.82	396.77	+ 0.05	0.05
188.64 ± 208.15	396.79		+ 0.02	
114.31 ± 282.47	396.78		+ 0.01	
288.21 ± 137.02	425.23		----	
211.32 ± 214.02	425.34	425.6 ^[b]	----	0.11
114.31 ± 310.99	425.30		----	
387.57 ^[c] ± 74.66 ^[a]	462.23		----	
114.31 ± 347.8	462.1		----	
270.18 ± 192.03	462.21		----	

TABLE II-3 (Contd.)

COMPARISON OF CASCADE ENERGY SUMS AND CROSSOVER ENERGIES
(All cascade and crossover energies are transition
energies from Table II-2 unless otherwise noted.)

Cascade Energies (keV)	Energy Sum (keV)	Crossover Energy (keV)	Sum-Crossover Difference (keV)	Total Spread in Level Energy (keV)
114.31 ± 423.54	537.85	538.00 ^[b]	----	0.04
188.64 ± 349.23	537.87		----	
211.32 ± 326.57	537.89		----	
270.18 ± 267.70	537.88		----	
360.05 ± 177.82	537.87		----	
114.31 ± 540.49	654.80	654.82	- 0.02	0.10
270.18 ± 384.69	654.87		+ 0.05	
211.32 ± 443.53	654.85		+ 0.03	
288.21 ± 366.64	654.85		+ 0.03	
360.05 ^[c] ± 294.82	654.87		+ 0.05	
537.87 ^[c] ± 117.03	654.90		+ 0.08	
396.77 ± 258.07	654.84		+ 0.02	
211.32 ± 1022.81	1234.13	1234.11	+ 0.02	0.21
270.18 ± 964.04	1234.22		+ 0.11	
288.21 ± 954.84	1234.05		- 0.06	
360.05 ± 874.03	1234.08		- 0.03	
396.77 ^[c] ± 837.46	1234.23		+ 0.12	
537.87 ^[c] ± 696.29	1234.16		+ 0.05	
654.82 ± 579.20	1234.02		- 0.09	
114.31 ± 1197.84	1312.15	1312.08	+ 0.07	0.12
188.64 ± 1123.84	1312.48 ^[d]		+ 0.40	
211.32 ± 1100.78	1312.10		+ 0.02	
270.18 ± 1041.97	1312.15		+ 0.07	
360.05 ± 951.99	1312.04		- 0.04	
396.77 ± 915.37	1312.14		+ 0.06	
188.64 ± 842.90	1031.54	1031.78	- 0.24	0.24
270.18 ± 761.52	1031.70		- 0.08	
537.87 ^[c] ± 493.86	1031.73		- 0.05	
360.05 ± 675.78	1035.83		----	0.32
537.87 ^[c] ± 497.64	1035.51		----	
654.85 ± 380.94	1035.79		----	
114.31 ± 1027.21	1141.52	1141.77	- 0.25	0.25
270.18 ± 871.44	1141.62		- 0.15	
360.05 ± 781.45	1141.50		- 0.27	
114.31 ± 1075.99	1190.30	1190.33	+ 0.03	0.07
211.32 ± 979.05	1190.37		- 0.04	
114.31 ± 1175.69	1290.00	1290.02	- 0.02	0.34
211.32 ± 1078.79	1290.11		+ 0.09	
654.82 ± 635.52	1290.34		+ 0.32	
188.64 ± 1259.58	1448.22	1447.97 ^[b]	----	0.46
396.77 ± 1051.91	1448.68		----	
654.82 ± 793.49	1448.31		----	

TABLE II-3 (Contd.)

COMPARISON OF CASCADE ENERGY SUMS AND CROSSOVER ENERGIES
(All cascade and crossover energies are transition
energies from Table II-2 unless otherwise noted.)

Cascade Energies (keV)		Energy Sum (keV)	Crossover Energy (keV)	Sum-Crossover Difference (keV)	Total Spread in Level Energy (keV)
114.31	± 1381.36	1495.67		- 0.02	
211.32	± 1284.25	1495.57		- 0.12	
270.18	± 1225.64	1495.82	1495.69	+ 0.13	0.43
360.05	± 1135.95	1496.00		+ 0.31	
114.31	± 1454.20	1568.51		----	
211.32	± 1357.12	1568.44		----	
270.18	± 1298.48	1568.66	1568.20 ^[b]	----	0.34
288.21	± 1280.25	1568.46		----	
396.77	± 1172.01	1568.78		----	

[a] Energy taken from Reference 9.

[b] The accuracy of the crossover transition energy is much less than that of the energy sums; therefore, the energy differences in Column 5 are not significant and those in Column 6 are computed without considering the crossover energy.

[c] This is a level energy.

[d] The 188 and 1123 sum is not considered in the value computed in Column 6.

Columns 5 and 6 are generally consistent with the errors quoted in Table II-2. The absolute accuracy of the energies is indicated by a comparison in Table II-4 with the transition energies from conversion-electron studies of Reference 9. Again the differences are consistent with the quoted uncertainties.

TABLE II-4

COMPARISON OF TRANSITION ENERGIES WITH THOSE OF REFERENCE 9

Transition Energies		Difference	
Table II-2	Backlin et al ^[a]	ΔE (keV)	$\Delta E/s$
97.00 ± 0.03 ^[b]	97.03 ± 0.03 ^[c]	- 0.03	- 1.0
114.31 ± 0.03	114.32 ± 0.03	- 0.01	- 0.3
117.03 ± 0.20	117.06 ± 0.05	- 0.03	- 0.6
122.41 ± 0.05	122.34 ± 0.10	+ 0.07	+ 0.7
126.62 ± 0.05	126.64 ± 0.20	- 0.02	- 0.1
139.21 ± 0.05	139.2 ± 0.10	0.0	0
155.87 ± 0.03	155.87 ± 0.06	0.00	0

TABLE II-4 (Contd.)

COMPARISON OF TRANSITION ENERGIES WITH THOSE OF REFERENCE 9

Transition Energies		Difference	
Table II-2	Backlin et al ^[a]	ΔE (keV)	$\Delta E/s$
188.64 $\pm$ 0.03	188.62 $\pm$ 0.07	+ 0.02	+ 0.2
192.03 $\pm$ 0.05	191.96 $\pm$ 0.07	+ 0.07	+ 1.0
198.93 $\pm$ 0.03	198.93 $\pm$ 0.07	0.00	0
208.15 $\pm$ 0.05	208.09 $\pm$ 0.07	+ 0.06	+ 0.9
211.32 $\pm$ 0.03	211.28 $\pm$ 0.05	+ 0.04	+ 0.8
214.02 $\pm$ 0.07	213.85 $\pm$ 0.15	+ 0.17	+ 1.1
240.23 $\pm$ 0.03	240.20 $\pm$ 0.06	+ 0.03	+ 0.5
245.71 $\pm$ 0.05	245.76 $\pm$ 0.10	- 0.05	- 0.5
267.70 $\pm$ 0.03	267.65 $\pm$ 0.06	+ 0.05	+ 0.8
270.18 $\pm$ 0.03	270.12 $\pm$ 0.10	+ 0.06	+ 0.6
273.28 $\pm$ 0.05	273.1 $\pm$ 0.3	+ 0.2	+ 0.6
275.46 $\pm$ 0.05	275.25 $\pm$ 0.30	+ 0.21	+ 0.7
282.47 $\pm$ 0.05	282.39 $\pm$ 0.20	+ 0.08	+ 0.4
288.21 $\pm$ 0.05	288.12 $\pm$ 0.15	+ 0.09	+ 0.6
301.14 $\pm$ 0.05	301.4 $\pm$ 0.4	- 0.3	- 0.8
310.99 $\pm$ 0.05	311.11 $\pm$ 0.20	- 0.12	- 0.6
326.57 $\pm$ 0.03	326.45 $\pm$ 0.11	+ 0.12	+ 1.1
349.23 $\pm$ 0.03	349.26 $\pm$ 0.25	- 0.03	- 0.1
423.54 $\pm$ 0.03	423.54 $\pm$ 0.20	0.00	0
540.49 $\pm$ 0.03	540.6 $\pm$ 0.3	- 0.1	- 0.3
654.82 $\pm$ 0.03	654.4 $\pm$ 0.5	+ 0.4	+ 0.8

[a] Reference 9.

[b] Estimated standard deviation ($\approx \sigma$).[c] Estimated error limit ($s \approx 2$ standard deviations).

Zr-97 - Nb-97 is not known, but zirconium is a common impurity in hafnium. Other spectra were obtained in which the Mn-56 and As-76 peaks were not observable.

The relative gamma-ray intensities were determined from the data in Figure II-11. The energies were determined for the strong transitions from similar spectra of mixed sources of Hf-183, Am-241 (59.568), Gd-153 (97.43 and 103.18), Lu-177 (112.955 and 208.365), Hg-203 (279.15), and Au-198 (411.795 keV) and Hf-183, Hg-203, Au-198, Na-22 (511.006), Cs-137 (661.60) Mn-54 (834.84), and Co-60 (1173.23 and 1332.48 keV. The energies of the weaker transitions were obtained from the Hf-183 spectrum by calibrating with the strong Hf-183 and Ta-183 lines.

The absolute gamma-ray intensities were determined by comparing the intensities of the Hf-183 and Ta-183 lines in three successive spectra obtained with a single source. The growth of the Ta-183 lines was used to determine the time at which the source would have been pure Hf-183. The Hf-183 intensities were compared with that of the 246-keV gamma ray from Ta-183 which was taken to represent 26.2 percent of the Ta-183 decays [15]. From these relative intensities and the appropriate growth and decay equations, the absolute intensities were determined. These intensities and energies are given in Table II-5.

TABLE II-5

GAMMA-RAY ENERGIES AND INTENSITIES
(The transition intensities are for the
multipolarities E1 for 73 and M1+E2 for 113 and 143 keV.)

<u>Transition Energy</u> <u>(keV)</u>	<u>Photon Intensities</u> <u>(percent of decays)</u>	<u>Transition Intensity</u> <u>(percent of decays)</u>
73.08 ± 0.05	38 ± 5	67 ± 9
113.75 ± 0.1	0.16 ± 0.02	0.46 - 0.75
143.1 ± 0.2	0.50 ± 0.15	0.7 - 1.07
160.7 ± 0.2	0.5 ± 0.2	
165.0 ± 0.2	0.06 ± 0.03	
223.5 ± 0.2	0.09 ± 0.05	
224.99 ± 0.1	0.17 ± 0.04	
284.01 ± 0.1	0.38 ± 0.06	
315.86 ± 0.05	1.23 ± 0.15	
397.86 ± 0.05	3.4 ± 0.4	
459.08 ± 0.05	30.0 ± 2.0	
476.93 ± 0.1	0.77 ± 0.1	
481.4 ± 0.2	0.60 ± 0.15	
686.47 ± 0.1	0.30 ± 0.06	
691.69 ± 0.1	0.30 ± 0.06	
716.9 ± 0.2	0.10 ± 0.03	

TABLE II-5 (Contd.)

GAMMA-RAY ENERGIES AND INTENSITIES
(The transition intensities are for the
multipolarities E1 for 73 and M1+E2 for 113 and 143 keV.)

Transition Energy (keV)	Photon Intensities (percent of decays)	Transition Intensity (percent of decays)
735.14 $\pm$ 0.08	0.93 $\pm$ 0.10	
783.81 $\pm$ 0.05	65	
806.57 $\pm$ 0.15	0.17 $\pm$ 0.04	
841.02 $\pm$ 0.15	0.18 $\pm$ 0.04	
856.81 $\pm$ 0.08	0.7 $\pm$ 0.2	
1091.49 $\pm$ 0.15	0.12 $\pm$ 0.12	
1470.38 $\pm$ 0.08	2.3 $\pm$ 0.3	
1784.3 $\pm$ 0.2	0.06 $\pm$ 0.03	

Only the relative errors are given for the intensities. A 15 percent normalization error should be included to get the total uncertainty. The transition intensities were computed on the assumption that the 73-keV transition is E1 and that those at 113 and 143 keV are M1 + E2.

Some gamma-gamma coincidence relationships were determined from the NaI(Tl) singles spectra shown in Figure II-12. It is clear from the coincidence sum peaks that the 73-keV transition is in coincidence with those at 783 and 1470 keV and is not in coincidence with the one at 459 keV.

These coincidence relationships were verified, and others were established by a gamma-gamma coincidence measurement. The analyses of two of the coincidence spectra are shown in Figure II-13. The coincidence relations determined from this experiment are listed in Table II-6.

If the K X-ray peak in Figure II-11 is assumed to result completely from conversion of the 73-keV transition, the K conversion of this transition can be computed. The resulting value is $\alpha_K = 0.58$. The theoretical conversion coefficients [16,17] for various multipolarities are shown in Figure 14. These data indicate this transition has E1 character. This conclusion also follows from the fact that any other multipolarity would yield a transition intensity of > 400 percent of the decays.

A tentative proposed decay scheme is shown in Figure II-15. The order of the transitions in the 315-143-keV and the 284-113-keV cascades is not uniquely determined from the experiment. The intensities of the beta-ray branches are computed from the gamma-ray intensities. There are eleven gamma-ray transitions in Table II-5 which are not included in this scheme.