ANL-7203 ADDENDUM
Reactor Technology

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ADDENDUM TO THE REPORT ON HIGH CONVERSION CRITICAL EXPERIMENTS

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ABSTRACT AND SUMMARY

This addendum presents revised results for the conversion-ratio measurements reported in ANL-7203. All the results have been revised downward, by net relative reductions of ~4-15%. The revision is necessary because of the application of a more appropriate method of data analysis than originally used. The improved analysis adjusts accurately for the epicadmium flux component in graphite thermal columns, which affected the reference irradiations used for normalization of these measurements.

The revised results for initial conversion ratios range from 0.44 ± 0.01 to 1.25 ± 0.04 for $H:U^{238}$ atom ratios of 4.16 down to 0.50 (which is a subcritical composition), using 3.04 w/o enrichment UO_2 fuel and H_2O moderator and reflector. the UO_2 pellet diameter was 0.935 cm, and an average of 853.3 g was contained as a 121.9-cm-length column in each fuel element. The cladding was 1.06-cm-OD stainless steel with 0.05-cm wall thickness.

INTRODUCTION

In ANL-7203,[1] pp. 50-51, it had been stated that the results given for conversion ratio in the last core of the series assembled for a series of experiments seemed to be too high and that the reason for this might have affected other results as well. A remeasurement had been made, but the additional data did not change the results. Since then the original data have been re-evaluated with appropriate corrections made for epithermal effects in the reference foils irradiated simultaneously in a predominantly thermal neutron flux.

ORIGINAL METHODS OF DATA TREATMENT

The modified conversion ratio (MCR) and initial conversion ratio (ICR) were measured according to Eqs. 1A and 2 below. The measured capture and fission rates in the cores were compared to those of similar materials irradiated simultaneously in the thermal, or reference, flux of

a graphite thermal column for which thermal cross sections apply.

$$MCR = \frac{N^{28}}{N^{25}} \frac{P}{R} \left[\frac{\sigma_c^{28}}{\sigma_{fiss}^{25}} \right]_{TC} ; \qquad (1A)$$

$$ICR = MCR \frac{1}{1+\alpha^{25}}. \qquad (2)$$

Here N^{28} and N^{25} are the atom number densities of U^{238} and U^{235}, respectively, in the core. P is the measured ratio of neutron capture by U^{238} in a thin slice of UO_2 across a fuel pellet, to capture by U^{238} in an identical UO_2 disc during a simultaneous irradiation in a thermal flux (i.e., a reference irradiation in a thermal column). R is the ratio of fissions in U^{235} in the same two samples. The cross sections (σ's) are for the thermal-column (reference) flux only, and it is necessary to adjust those values quite accurately for self-shielding, as an example, to avoid errors in the estimates of capture-to-fission ratios in the reference irradiations. The term α^{25} is the calculated capture-to-fission ratio for U^{235} in the core.

Difficulty was encountered because of a significant epithermal flux component present in the thermal-column regions used for reference irradiations, some of which were made fairly close to the source plane. Because U^{238} has a low energy resonance, epicadmium capture by U^{238} in the thermal columns varied significantly with surface area. For the first two reference irradiations, the 0.03-mm-thick gold cadmium ratios (CdR's) were 50 and 90, respectively. Following these, reference irradiations were made in a semipermanent graphite thermal column at locations where the gold CdR's ranged from 105 to 115. For the last two core loadings, the usual location was not available, and the reference measurements were made in thermal-column locations for which the CdR's were 32 and 35 for gold and 7.9 and 8.7 for U^{238}. From the observed ratio of $(CdR - 1)_{U238}/(CdR - 1)_{Au} = 0.22 \pm 0.01$, it was concluded that CdR's of U^{238} equal to ~12 and ~21 corresponded with gold CdR's of 50 and 90, respectively.

The thermal-column flux had been too low for accurate measurement of the U^{238} CdR initially, and it was decided to use the formula for Westcott's (two-group) cross sections[2] to account for the epicadmium flux component present, as in the following equation:

$$\sigma_{eff} = \sigma_{2200} \sqrt{\frac{\pi T_0}{4T_n}} \left[g(T) + rS(T) \right]. \qquad (3)$$

Substitution of these terms into Eq. 1 results in

$$MCR = \frac{N^{28}}{N^{25}} \frac{P}{R} \left[\frac{\sigma_c^{28}}{\sigma_{fiss}^{25}}\right]_{2200} \left[\frac{g_c^{28}(T) + rS_c^{28}(T)}{g_{fiss}^{25}(T) + rS_{fiss}^{25}(T)}\right]_{TC} . \qquad (1B)$$

Since the ratio $\sigma_c^{28}/\sigma_{fiss}^{25}$ is used, the explicit temperature terms of Eq. 3 cancel in Eq. 1B. (It can be shown that the implicit temperature dependence is small, by substitution of typical approximations for g(T) and S(T) for which the temperature dependence cancels.) The neutron temperature in a graphite thermal column is close to room temperature, and any reasonable estimate of neutron temperature should be adequate for use with Eq. 1B, since only the ratios of capture-to-fission in the thermal columns are obtained from calculated cross sections.

The quantity $rS_{fiss}^{25}(T)$ proved to be negligible in the reference irradiations, but $rS_c^{28}(T)$ varied significantly.

Equations 3 and 1B apply only to infinite dilution conditions, and adjustments must be made for both thermal and resonance self-shielding in the discs irradiated for reference use if these equations are used with accuracy.

Some preliminary results were obtained using Eq. 1B, effectively, but in subsequent reference irradiations the calculated cross sections for U^{238}, using Eq. 3 were not very consistent with the measured specific activities. A wide variety of compositions and dimensions of uranium-metal foils and UO_2 discs and pellets were irradiated in the graphite thermal column in testing the calculated cross sections.

The measured subcadmium U^{238} capture rates and U^{235} fission rates were quite consistent with the calculated self-shielded cross sections, and as a consequence the subcadmium U^{238} capture rates were used for all the MRC and ICR results given in ANL-7203. Effectively, each value of P used in Eq. 1A had been revised by multiplication by the quantity CdR/(CdR - 1) measured, or estimated, for U^{238} capture in the corresponding reference irradiation.

It is shown later, under the heading "Modified Equations for the Use of Thermal Activation Data," that the subtraction of epicadmium activities requires the removal of the rS(T) terms from Eq. 1B as in the revised Eq. 1D. A recent review of the Hi-C data showed that the results given in ANL-7203 had been obtained by substitution of terms resembling P* as defined for use with Eq. 1D, in place of P in Eq. 1B.

RELATION OF THERMAL ACTIVATION TO BARE FOIL AND NONRESONANCE ACTIVATIONS IN THE THERMAL COLUMNS

As an approximation, one usually estimates the ratio of thermal activation to total activation of a foil from the CdR, as in the following equation:

$$\frac{\text{Thermal activation}}{\text{Total activation}} \doteq \frac{\text{Subcadmium activation}}{\text{Bare-foil activation}} = \frac{\text{CdR} - 1}{\text{CdR}}. \qquad (4)$$

Westcott indicated the following expression for infinite dilution cadmium ratio,[2] shown as Eq. 5. (This expression is limited to values of $g \approx 1$.) The 2200-m/s cross section cancelled from the numerator and denominator of the following equation, and the term P_{cd} is seen to represent the epicadmium portion of the 1/v part of the cross section:

$$\text{CdR} = \frac{g(T) + rS(T)}{rS(T) + rP_{cd}(T)}. \qquad (5)$$

By use of Eq. 5, the approximation given as Eq. 4 can be compared to the following form, which is more precise:

$$\frac{\text{Nonresonance activation}}{\text{Total activation}} = \frac{g(T)}{g(T) + rS(T)} = \frac{\text{CdR} - 1}{\text{CdR}} + \frac{rP_{cd}(T)}{g(T) + rS(T)}. \qquad (6A)$$

The value of $rP_{cd}(T)$ is indicated by substitution of an infinite-dilution gold CdR into Eq. 5 and solving for $rP_{cd}(T)$. Since the thermal-column flux is very close to thermal-neutron temperature, the 25°C values for g(T) and S(T) were used. Reference 3 shows calculated and measured values of CdR - 1 for various thicknesses of gold in a graphite internal column. The factor ~0.39 is shown for correction of these 50-mg/cm^2 gold-foil (0.03-mm-thick) CdR's to infinite dilution. Thus the infinite-dilution CdR's are determined from the measured values of gold CdR by the equation $(\text{CdR})_{\text{inf dil}} = 1 + 0.39(\text{CdR} - 1)_{\text{meas}}$. The infinite-dilution gold CdR's of 13.1, 14.3, 20.1, 35.7, 41.6, and 45.5 correspond to measured gold CdR's of 32, 35, 50, 90, 105, and 115 respectively. Using the tabulated[2] values of ($g = 1.006$) and ($S \doteq 17.5$) for gold indicated that $0.0005 < rP_{cd}(T) < 0.0025$ for the various thermal column locations. (The value of $P_{cd}(T)$ is approximately[2] 0.5.)

Supplying values of $g(T) \doteq 1.002$ and $S(T) \doteq 117$ for U^{238} indicated that the errors from setting $rP_{cd}(T) = 0$ in Eq. 6A range from 0.0005 to 0.002. Considering that the S(T) term would be substantially smaller if adjusted for self-shielding, the errors are bracketed by the values given here, as low estimates, and by $rP_{cd}(T)$, as high estimates.

Noting that the values of U^{238} (CdR - 1)/CdR are equal to or greater than ~(7.9 - 1)/7.9 = 0.8734 and that ~0.0025 error is additive to this permits an estimate of -0.0025/0.8734 = -0.3% worst error in setting $rP_{cd}(T) = 0$. The error ranges from -0.1 to -0.3%, depending on the CdR's measured in the thermal columns.

For consistency, equivalent treatment should be applied to the U^{235} fission rates. The CdR's of U^{235} fission, U^{238} capture, and gold activation were remeasured recently in the graphite thermal column. From this measurement it was estimated that the U^{235} fission cadmium ratio was about 300 for the thermal column irradiation giving the smallest gold CdR (32). This indicated that the term (CdR - 1)/CdR was about 0.9967 and the term $rP_{cd}(T)$ was about 0.0025. Summing the two as in Eq. 6A indicated that 0.9992 of the bare U^{235} fission rate was the thermal part. The results used in ANL-7203 for the bare U^{235} fission rates are slightly high (0.09%). This reduced the reported conversion ratios by the same percentage error, and this is cumulative with the correction for U^{238}.

The net effect of the foregoing treatment was to decrease the ratio of P*/R* by 0.1-0.4%. The distinction between P and R, and P* and R* is shown below. Most of this decrease is attributable to the $rP_{cd}(T)$ correction for U^{238} thermal capture, and only about a quarter of the decrease was attributable to the change from bare U^{235}-Al fission rates to thermal fissions.

MODIFIED EQUATIONS FOR THE USE OF THERMAL-ACTIVATION DATA

Thermal-activation cross sections having been chosen as being more accurate than bare-foil cross sections calculated for the thermal column, because of U^{238} resonance capture, Eq. 1A must be modified accordingly. Specifically, the bare activations must be eliminated from Eq. 1A by solving Eq. 6A for nonresonance (or alternatively subcadmium) activation as follows:

$$\text{Bare-foil activation} = (\text{Nonresonance activation})\left[\frac{g(T) + rS(T)}{g(T)}\right]; \qquad (6B)$$

$$P^* = \frac{\text{Bare } U^{238} \text{ capture rate in core}}{\text{Nonresonance capture rate in T.C.}} = P\left[\frac{g_c^{28}(T) + rS_c^{28}(T)}{g_c^{28}(T)}\right]; \qquad (7)$$

$$R^* = \frac{\text{Bare } U^{235} \text{ fission rate in core}}{\text{Nonresonance } U^{235} \text{ fission rate in T.C.}} = R\left[\frac{g_{fiss}^{25}(T) + rS_{fiss}^{25}(T)}{g_{fiss}^{25}(T)}\right]; \qquad (8)$$

$$MCR = \frac{N^{28}}{N^{25}} \frac{P^*}{R^*} \left[\frac{\sigma_c^{28}}{\sigma_{fiss}^{25}}\right]_{TC} \left[\frac{g_c^{28}(T)}{g_{fiss}^{25}(T)}\right]_{TC} \left[\frac{g_{fiss}^{25}(T) + rS_{fiss}^{25}(T)}{g_c^{28}(T) + rS_c^{28}(T)}\right]_{TC} . \quad (1C)$$

Substituting Eq. 3 into Eq. 1C cancels all the quantities involving S explicitly to give

$$MCR = \frac{N^{28}}{N^{25}} \frac{P^*}{R^*} \left[\frac{\sigma_c^{28}}{\sigma_{fiss}^{25}}\right]_{2200} \left[\frac{g_c^{28}(T)}{g_{fiss}^{25}(T)}\right]_{TC} . \quad (1D)$$

The revised results for conversion ratios given in Tables I and II were calculated according to Eq. 1D. The original results (in ANL-7203)

TABLE I. Revised Conversion-ratio Data in Hi-C and BORAX-V Cores

Date	Run No.	Type of P Measurement, Foil Enrichment, etc.	Ratio[a] P*	Ratio[a] R*	Thermal Column Gold Cd Ratio	Modified Conversion Ratio (MCR)	Initial Conversion Ratio (ICR)
		1.27-cm Square BORAX-V, Core 1					
8/11/61	28-1	0.21% foil-count	2.65	0.691	90	0.354 (1)[b]	0.287 (1)[b]
10/4/61	38-7	0.21% foil-count	9.84	2.73	90	0.333 (1)	0.269 (1)
		1.27-cm Triangular BORAX-V, Core 2					
1/8/62	71-93	0.21% foil-count	0.504	0.127	50	0.397 (1)	0.317 (1)
		1.349-cm Square Hi-C, Core 4					
12/3/65	418-4	0.21% foil-count	2.35	0.643	35	0.560 (2)	0.457 (2)
12/7/65	418-5	0.21% foil-count	1.63	0.475	35	0.526 (2)	0.429 (2)
12/9/65	418-6	0.21% foil-count	1.78	0.506	35	0.539 (2)	0.440 (2)
		1.24-cm Square Hi-C, Cores 8 and 9					
12/13/62	183-6	0.21% foil-count	2.32	0.585	115	0.608 (1)	0.489 (1)
3/18/63	224-1	6 ppm foil-count	1.17	0.298	115	0.602 (1)	0.484 (1)
3/18/63	224-1	0.21% foil-count	1.21	0.298	115	0.622 (1)	0.500 (1)
5/6/64	383-1	0.21% foil-count	4.96	1.25	105	0.608 (1)	0.489 (1)
5/6/64	383-1	1.27-cm pellet-radiochem	5.11	1.25	105	0.627 (2)	0.504 (2)
6/3/64	383-10	0.21% foil-count	14.8	3.79[c]	105	0.599 (2)	0.481 (2)
6/3/64	383-10	1.27-cm pellet-radiochem	14.3	3.72	105	0.589 (2)	0.474 (2)
7/6/64	387-9	0.21% foil-count	11.4	3.04	105	0.575 (2)	0.462 (2)
7/6/64	387-9	0.21% foil-radiochem	11.9	3.04	105	0.600 (2)	0.482 (2)
7/6/64	387-9	1.27-cm pellet-radiochem	10.8	3.04	105	0.545 (2)	0.438 (2)
		1.27-cm Triangular Hi-C, Core 12					
7/17/63	283-7	0.21% foil-count	16.1	3.35	115	0.737 (2)	0.586 (2)
8/13/63	285-6	0.21% foil-count	1.18	0.271	115	0.667 (2)	0.531 (2)
		1.166-cm Triangular Hi-C, Core 14					
4/2/64	372-12	0.21% foil-count	2.65	0.399	105	1.018 (2)	0.786 (2)
4/3/64	372-13	0.21% foil-count	2.77	0.411	105	1.033 (2)	0.797 (2)
4/9/64	373-7	0.21% foil-count	2.49	0.364	105	1.049 (2)	0.809 (2)
		1.127-cm Triangular Hi-C, Core 19					
12/3/63	342-10	0.21% foil-count	4.61	0.598	115	1.182 (2)	0.897 (2)
12/3/63	342-10	d -radiochem	4.09	0.598	115	1.048 (2)	0.796 (2)
12/5/63	342-11	0.21% foil-count	4.74	0.582	115	1.249 (2)	0.948 (2)
12/23/63	342-22	0.21% foil-count	1.64	0.217	115	1.159 (1)	0.880 (1)
12/23/63	342-22	e -radiochem	1.61	0.217	115	1.137 (2)	0.864 (2)
		1.069-cm Triangular Hi-C, Core 21					
7/22/64	391-15	0.21% foil-count	2.21	0.192	32	1.765 (3)	1.296 (3)
7/23/64	391-16	0.21% foil-count	2.01	0.199	32	1.548 (3)	1.137 (3)
7/24/64	391-17	0.21% foil-count	2.22	0.189	32	1.801 (3)	1.322 (3)

[a]This ratio is defined in the text.
[b]Numbers in parentheses denote number of independent determinations.
[c]This value of R was determined by radiochemical techniques; all other values of R were obtained from foil-counting data.
[d]1.27-cm pellets used in the lattice; 0.254-cm pellets used in the ATSR.
[e]1.27-cm pellets used in the lattice; 0.127-cm pellets used in the ATSR.

TABLE II. Revised Conversion Ratios in Hi-C and BORAX-V Cores

Lattice	Core No.	H-to-U^{238} Atom Ratio	Modified Conversion Ratio (MCR)	α^{25}	Initial Conversion Ratio (ICR)
1.27-cm square BORAX-V	1	4.65	0.344	0.236	0.278 ± 0.02 (2)[a]
1.27-cm triangular BORAX-V	2	3.53	0.397	0.252	0.317 ± 0.02 (1)
1.349-cm square Hi-C	4	4.16	0.542	0.225	0.442 ± 0.01 (6)
1.24-cm square Hi-C	8 & 9	2.92	0.594	0.244	0.478 ± 0.01 (16)
1.27-cm triangular Hi-C	12	2.29	0.702	0.257	0.558 ± 0.02 (4)
1.166-cm triangular Hi-C	14	1.33	1.033	0.296	0.797 ± 0.01 (6)
1.127-cm triangular Hi-C	19	0.98	1.155	0.317	0.877 ± 0.02 (9)
1.069-cm triangular Hi-C	21	0.50	1.705	0.362	1.252 ± 0.04 (9)

[a]Numbers in parentheses denote number of independent determinations.

had been calculated much as if P* could be substituted directly into Eq. 1B for P. The foregoing discussion showed that the only way in which r and S enter into Eq. 1D is through very minor adjustments to P* and R* that are no greater in magnitude than 0.4% of the ratio P*/R*.

The ICR is plotted against H:U^{238} atom ratio in Fig. 1 along with calculated ICR's from Ref. 4. A slightly different set of cross sections and α^{25} values were used in the calculations. The use of those cross sections in the experimental data analysis would reduce the measured MCR's and ICR's by about 1.6%, and thus would increase the differences significantly. The use of the same values of α^{25} would slightly increase most of the experimental points, but no more than ±0.5% change would result. The net effect would be to slightly decrease the average difference.

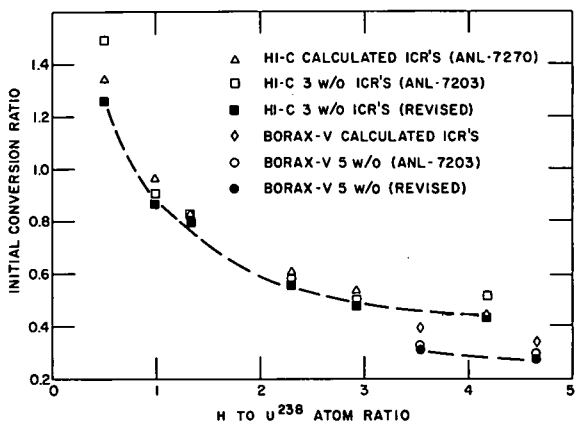

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Fig. 1. Hi-C Conversion Ratios

Alternatively to the preceding method, equations may be written in terms of the subcadmium activation rather than nonresonance activation. This is an equally direct algebraic derivation, and the resulting equations might be more useful in some instances than the preceding ones. The values of $rP_{cd}(T)$ must be subtracted from g(T) rather than being combined with other measured terms, and P** and R** are not quite identical with P* and R*. The resulting equations are

$$P^{**} = \frac{\text{Bare } U^{238}\text{capture rate in core}}{\text{Subcadmium capture rate in T.C.}} = P\left[\frac{g_c^{28}(T) + rS_c^{28}(T)}{g_c^{28}(T) - rP_{cd}(T)}\right]_{TC}; \quad (9)$$

$$R^{**} = \frac{\text{Bare } U^{235}\text{ fission rate in core}}{\text{Subcadmium } U^{235}\text{ fission rate in T.C.}} = R\left[\frac{g_{fiss}^{25}(T) + rS_{fiss}^{25}(T)}{g_{fiss}^{25}(T) - rP_{cd}(T)}\right]_{TC}; \quad (10)$$

$$MCR = \frac{N^{28}}{N^{25}} \frac{P^{**}}{R^{**}} \left[\frac{\sigma_c^{28}}{\sigma_{fiss}^{25}}\right]_{2200} \left[\frac{g_c^{28}(Th) - rP_{cd}(T)}{g_{fiss}^{25}(Th) - rP_{cd}(T)}\right]_{TC} \quad (1E)$$

As a final comment, Eqs. 1D and 1E should give results that are consistent with Eq. 1A. The advantage of Eqs. 1D and 1E is the use of sub-cadmium capture and fission rates, which avoids the difficulty of calculating the effective resonance cross section of U^{238} in the thermal columns.

EFFECT OF USE OF CURRENTLY RECOMMENDED CROSS SECTIONS

The "Recommended" cross sections stated by BNL-325, Second Edition, Supplement No. 2, Neutron Cross Sections, were used for the revised conversion ratios reported here. These include $\sigma_{fiss}^{25} = 577.1$ and $\sigma_c^{28} = 2.73$ barns for 0.0253-eV energy neutrons. The values used for the results given in ANL-7203 were listed as $\sigma_{fiss}^{25} = 581.95$ and $\sigma_c^{28} = 2.72$ barns. Use of the currently recommended cross sections increased the results by 1.2% relatively, compared to the use of the cross sections involved in ANL-7203.

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