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TUNGSTEN RESONANCE INTEGRALS AND DOPPLER COEFFICIENTS

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TUNGSTEN RESONANCE INTEGRALS AND DOPPLER COEFFICIENTS THIRD QUARTERLY PROGRESS REPORT (JANUARY – MARCH 1966)

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CONTENTS

TABLES

I. PROJECT OBJECTIVES

The objective of this project is to carry out an integrated experimental and analytical study of resonance integrals and Doppler coefficients of various samples of natural tungsten and separated tungsten isotopes.

Measurements of resonance integrals and Doppler coefficients are made in the 10.6-in. lattice of the Sodium Graphite Reactor Critical Assembly (SGR-CA), in which the flux spectrum in the resonance region is approximately 1/E. Both reactivity and activation methods are used; the reactivity oscillator is calibrated with the known resonance integral of gold; in addition, the resonance integral of W^{186} is being measured over a limited temperature range by the activation technique.

Resonance integrals and Doppler coefficients for each sample are calculated with TRIX-I, a fast-running resonance-integral code. Other methods, such as the RIFF-RAFF code and Monte Carlo calculations, are used as a check and to investigate some possible problem areas.

II. SUMMARY AND EVALUATION

The final 15-group calculation of parameters of the 10.6-in. SGR-CA lattice has been completed. Multigroup effective cross sections have been calculated for the actual isotopic compositions of the W^{182} and W^{186} samples. From these (and previous) cross sections and the multigroup real and adjoint fluxes, expected reactivities have been computed for samples of natural tungsten, W^{182}, W^{186}, and gold. Doppler coefficients have been calculated for the three tungsten samples. Agreement of these quantities with experiment is as good as could be expected for absolute reactivity calculations; however, measured reactivity ratios among the three samples are significantly different from calculated ratios.

An existing Monte Carlo resonance-escape code has been modified so that only those neutron histories resulting in passage through the sample are followed. This relatively minor modification has greatly increased the efficiency of the code.

Activation measurements of the W^{186} resonance integral have been completed at two temperatures, 293 and 473°K.

Reactivity worths and Doppler coefficients of samples enriched in W^{182} and in W^{186} have been measured in the cadmium sleeve at the center of the SGR-CA. Doppler measurements were made at several temperatures from room temperature to above 1000°K.

Reactivities of gold samples with widely varying surface-to-mass ratio have been measured to allow absolute calibration of the reactivity measurements.

Development of high-temperature ovens on a related AEC-sponsored project is being accelerated to allow completion of NASA high-temperature measurements within the contract period.

III. PROGRESS DURING REPORT PERIOD

A. THEORY

1. Neutron Spectrum

The final 15-group calculation of parameters of the 10.6-in. SGR-CA lattice has been completed. The most important results of this calculation are the real and adjoint neutron-flux spectra in the 1.33-in. -diameter central void with and without the surrounding 0.031-in. -thick cadmium sleeve (Table 1). The

TABLE 1
CALCULATED SGR-CA NEUTRON SPECTRA

Group No.	Upper Energy Limit	No Cadmium Sleeve		Cadmium Sleeve	
		Real Flux*†	Adjoint Flux*	Real Flux*†	Adjoint Flux*
1	10.0 Mev	0.0732	0.8453	0.0723	1.0850
2	3.0 Mev	0.6784	0.8335	0.6675	1.0528
3	1.4 Mev	0.8031	0.8494	0.7914	1.0453
4	0.9 Mev	1.1908	0.8672	1.1811	1.0513
5	0.4 Mev	0.7930	0.8942	0.7890	1.0568
6	0.1 Mev	0.8580	0.9138	0.8521	1.0538
7	17. kev	0.9241	0.9348	0.9140	1.0463
8	3.354 kev	0.9742	0.9539	0.9618	1.0377
9	454. ev	0.9964	0.9775	0.9866	1.0242
10	61.44 ev	1.0000	1.0000	1.0000	1.0000
11	22.6 ev	0.9977	1.0087	1.0094	0.9671
12	8.3 ev	0.9888	1.0146	1.0047	0.9005
13	3.06 ev	0.9767	1.0168	0.9702	0.7700
14	1.126 ev	0.9631	1.0152	0.4696	0.3568
15	0.414 ev	-	-	-	-

*Relative to Group 10
†Per unit lethargy

principal difference from the calculation reported in the previous quarterly report[1] was improved treatment of heterogeneity effects in fast fission. The loading required for criticality was calculated to be 20.4 fuel elements, compared with the actual value of 19.4 elements. Calculated values of delayed-neutron fraction and neutron lifetime were 0.00698 and 0.001240 sec.

2. Calculated Reactivities and Doppler Coefficients

Real and adjoint fluxes from the above SGR-CA calculation have been used with multigroup tungsten and gold cross sections to calculate Doppler coefficients of bare and cadmium-covered tungsten samples and reactivities of cadmium-covered samples of natural tungsten, W^{182}, W^{186}, and gold. Cross sections used for the two enriched samples corresponded to actual isotopic compositions of the samples; for previous calculations it had been assumed that each sample consisted of a single isotope. For the Doppler coefficient, the principal isotope contributes half or less of the total. These results, together with their experimental counterparts, are presented in Tables 2 and 3. The calculated W^{186} reactivity value may be too large, since the 2200-m/sec cross section calculated by TRIX-1 from the resonance parameters is 25% larger than a recently reported value.[2] Reduction of the W^{186} reactivity by this order of magnitude would result in approximate equality of the W^{182} and W^{186} calculated reactivities, in agreement with the measured reactivities of these samples. Since much of the W^{186} reactivity is due to the 18.8-ev resonance and the 1/v cross section, it is possible that adjusting the radiation width of this resonance to force agreement with the new measured value of σ_a may reduce the calculated reactivity by as much as 10 to 20%.

A number of approximations are implicit in the calculation of flux and importance used in the perturbation-theory computation of sample reactivities. Among these approximations are: homogenization of most of the reactor, use of only one spatial dimension, and use of only 15 energy groups. Although these approximations are perfectly adequate for calculating integral properties of the reactor (such as critical mass), where cancellation of errors tends to occur, they are less adequate for determining detailed properties such as flux or importance in a particular energy group. In view of these approximations, as well as uncertainties in the basic cross sections used in the reactor calculation,

TABLE 2

SAMPLE REACTIVITIES AT ROOM TEMPERATURE IN CADMIUM SLEEVE

Sample	Diameter (in.)	Length (in.)	Reactivity (¢)	
			Calculated	Measured
W^{182}	0.438	4.00	1.447 (79% 182)	1.770 ± 0.001
W^{186}	0.438	4.00	1.773 (91% 186)	1.744 ± 0.001
Natural W	0.438	4.00	1.826	1.94 ± 0.01
Gold	0.235	3.683	0.950	1.207 ± 0.001

TABLE 3

DOPPLER COEFFICIENTS OF STANDARD-SIZE TUNGSTEN SAMPLES AT 745°K

Sample	Doppler Coefficient (¢/1000°C)			
	Bare		Under Cadmium	
	Calculated	Measured*	Calculated	Measured
W^{182}	0.0859 (45% 182)	-	0.0448 (40% 182)	0.070 ± 0.006
W^{186}	0.0700 (43% 186)	0.124	0.0369 (36% 186)	0.0471 ± 0.0016
Natural W	0.151	0.157	0.0797	0.094 ± 0.005

*Uncorrected for thermal expansion

the overall agreement of calculated and measured reactivities (usually within 25%) is as good as can be expected. Reactivity ratios, whether calculated or measured, are of more interest, because they depend much more directly on the sample cross sections.

Doppler coefficients are presented in Table 3 as though they were independent of temperature, for the sake of representing each one by one easily understandable number. In fact, they decrease with temperature, presumably as $T^{-1/2}$ for a thermal reactor. This decrease is readily apparent for both measured and calculated values. Final results will be quoted in terms of least-squares power-law fits to the data.

3. Monte Carlo Calculations

Compilation, checkout, and debugging of the Monte Carlo program has been completed. This program is basically a modified version of an existing resonance-escape code; the efficiency of the original version has been greatly increased by following only those neutron histories resulting in passage through the sample. Cross sections for natural tungsten and gold were prepared with the aid of a new version of the UNICORN code (to be published), for calculating °K pointwise cross sections, and an existing Doppler-broadening code. A test case run for a 0.235-in.-diameter gold sample was converging well after only 1000 histories. Failure, for thick samples, of the flat-source approximation used in TRIX is evident in the following comparison (Table 4).

TABLE 4

EFFECTIVE RESONANCE INTEGRAL OF THICK GOLD SAMPLE

Method Used	Effective Resonance Integral (barns) Above 0.55 ev
TRIX (corrected for 1/v self-shielding)	82
Monte Carlo (1000 histories)	69 ± 4
Experiment (Advanced Doppler Project)	64 ± 2

B. EXPERIMENT

1. Activation Measurements

Bare exposures of natural-tungsten foils at several S/M values have been completed at 293 and 473°K. In addition to measurements on a foil sandwiched between two halves of a standard sample, measurements on several thinner foils were performed, to obtain an experimental normalization of the resonance integral and its temperature change. This normalization is made necessary by the fact that a Cd^{113} resonance at 18.5 ev partially overlaps the first W^{186} resonance at 18.8 ev. Measurements by Baumann[3] indicate a significant effect of this overlap on the resonance integral; its effect on the Doppler coefficient can be of either sign. This possible difficulty has been avoided by making the Doppler activation measurements without cadmium. If cadmium were used, the

- ratio of the change in activation between two temperatures to the activation at one temperature would be directly proportional to the Doppler coefficient $\Delta I/I\Delta T$, without any normalization. Without cadmium, the change in activation is still proportional to ΔI but the total activation is not proportional to the resonance integral, I. Thus, the experimental normalization is required.

Data from runs at these two temperatures are being reduced and analyzed. Because of funding limitations, activation measurements will not be extended to higher temperatures.

2. Reactivity Measurements

Total reactivity worths of standard-size samples of W^{182} and W^{186} have been measured with and without the cadmium sleeve. Bare results were $20.764 \pm 0.014¢$ for W^{182} and $26.085 \pm 0.023¢$ for W^{186}; results for samples under cadmium were presented in Table 2, preceding.

The reactivity change with temperature has been measured for W^{186}, bare and under cadmium, and for W^{182} under cadmium. Large calculated values for the thermal-expansion reactivity coefficient[1] and large measured differences between bare and cadmium Doppler results have led to the conclusion that Doppler measurements on samples under cadmium are more reliable and require less correction than measurements on bare samples; therefore, no additional bare measurements will be done. This problem is much less severe in the activation measurements, for the following reasons. The foil being activated is sandwiched between two halves of the 4-in.-long tungsten slug; it is far enough from the ends of the slug that essentially none of its activation is due to neutrons entering the ends. The thermal-expansion coefficient of activation is then smaller by a factor of two than the corresponding coefficient for the reactivity measurements, because axial expansion has no effect on the activation of the foil.

Reactivity-vs-temperature measurements of W^{182}, W^{186}, and an empty oven, all under cadmium, were repeated one or more times. From the results of repeated runs, it now appears that the reactivity change with temperature of an empty oven is essentially zero; therefore, no oven subtraction was performed for the W^{182} and W^{186} data, and previous natural-tungsten data were reanalyzed. Results of these measurements appeared in Table 3, preceding, and are

also presented in Figure 1. Each curve is a composite of two or more reactor runs.

A tentative conversion of reactivity change (¢) to resonance-integral change (barns) has been included in the Figure. The scale factor for this conversion is uncertain by about 20%.

Reactivity measurements on a number of new gold samples under cadmium have been performed, and previous gold-sample measurements have been repeated (See Table 5). After conversion of thin-foil measurements to equivalent cylindrical geometry with TRIX-1, these data are expected to provide adequate coverage in S/M for epithermal calibration of the oscillator.

TABLE 5

REACTIVITY WORTHS OF GOLD SAMPLES IN CADMIUM SLEEVE

Length (in.)	Width* or Diameter (in.)	Thickness (in.)	Weight (gm)	Reactivity (¢)
4.0	1.045*	0.002	2.683	0.4046 ± 0.0006
4.0	0.990*	0.031	38.088	1.6511 ± 0.0010
4.0	0.030	(cyl)	0.876	0.0555 ± 0.0007
3.683	0.235	(cyl)	49.871	1.2073 ± 0.0009
0.67	0.67	(cyl)	74.4	1.1304 ± 0.0012
1.34	0.67	(cyl)	148.8	1.9818 ± 0.0010
2.01	0.67	(cyl)	223.2	2.8143 ± 0.0014
2.68	0.67	(cyl)	297.4	3.6310 ± 0.0023

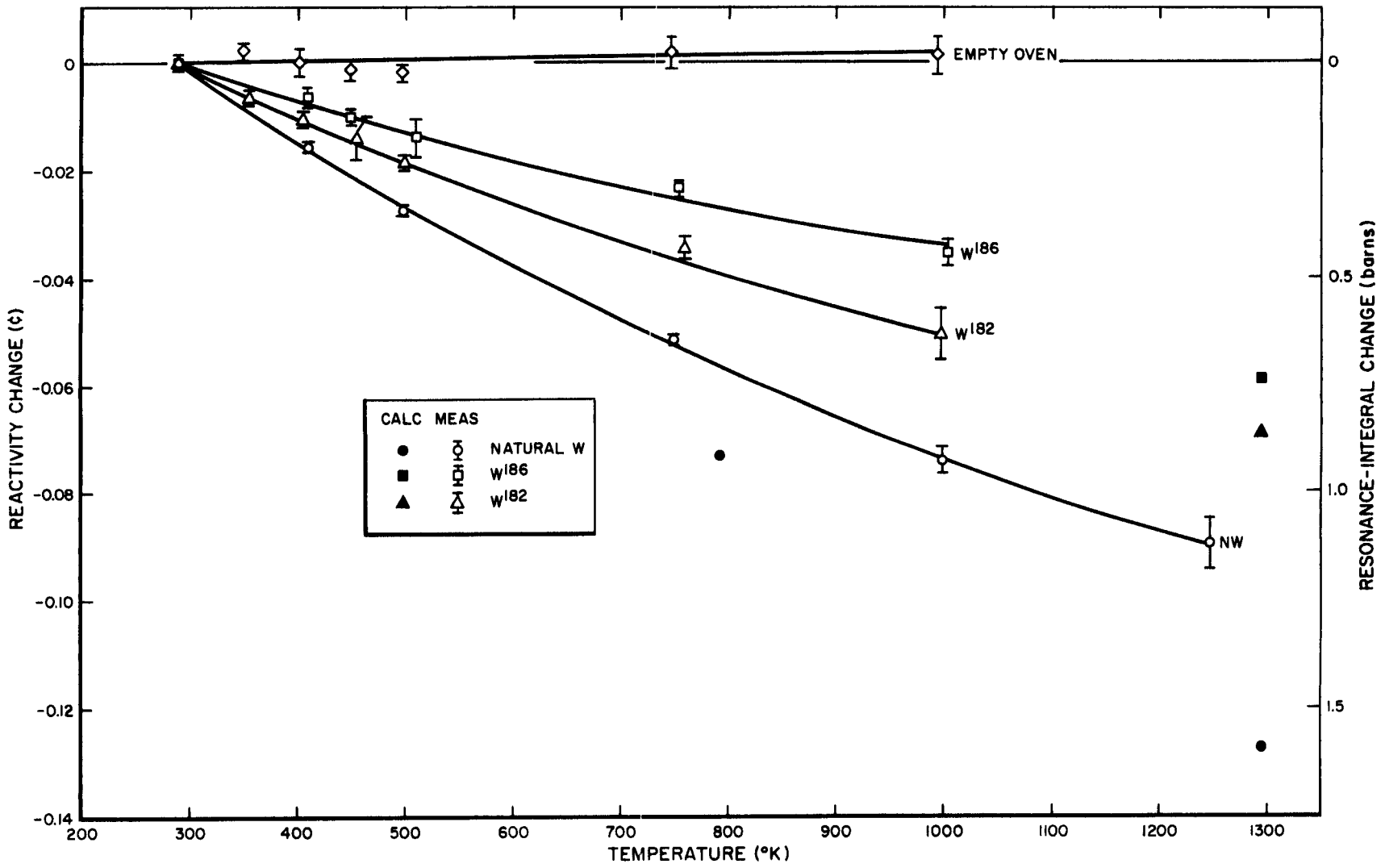
*Rectangular sample

C. RELATED PROJECTS*

1. Advanced Doppler Coefficients

Analysis of the gold data is nearly complete. An auxiliary experiment has demonstrated that interactions between foils in the new exposure wheel have been eliminated. Cadmium-covered exposures of natural-tungsten foils at 293 and 473°K have been completed.

*AEC-sponsored Doppler projects



Change in Reactivity (or resonance integral) vs Absolute Temperature.
Resonance-integral scale is tentative; 20% uncertainty.

2. Fast-Spectrum Doppler Measurements

The first oven utilizing the design proposed for reaching 2300°K has been assembled and is being tested. The only difference between this oven and the 2300° oven is the use of high-density alumina as insulating material rather than thoria; it is expected to reach a temperature of the order of 1800 to 2200°K. Work on two alternate designs has started. One design makes use of insulated heater wire rather than an insulating sleeve; the other uses a sleeve made of thoria rather than alumina.

IV. ACTIVITIES DURING NEXT REPORT PERIOD

Further investigation of problem areas will be carried out with the new Monte Carlo code. Reactivity and Doppler coefficient will be calculated for the W^{184} sample.

Reactivity and Doppler-coefficient measurements will be performed with the W^{184} sample. Doppler measurements on W^{182}, W^{186}, and natural tungsten will be extended to the operating limits of the present ovens, expected to be in the neighborhood of 1500°K. This will conclude Phase I of the project.

Upon successful operation of high-temperature ovens being developed on the Fast Doppler project, Doppler measurements on all tungsten samples will be extended to the neighborhood of 2000°K or higher.

Data analysis will continue and work will start on portions of the final report.

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