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TOPICAL REPORT: RESONANCE INTEGRALS MEASURED IN THE
ADVANCED REACTIVITY MEASUREMENT FACILITIES

MASTER

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In a program of this duration, the support and contributions of many individuals and organizations is necessary. We cannot acknowledge everyone, but must mention the technical aid of the Metallurgy and Analytical Chemistry groups of Idaho Nuclear Corporation, the staff of the ARMF's, and the continued support of the Division of Reactor Development and Technology of the U. S. Atomic Energy Commission.

SUMMARY

This report is intended to disclose the results of several years' effort of this laboratory in the measurement of resonance absorption integrals. Although the results included here have been previously reported in the open literature of the field, as has been the description of the various experimental techniques involved in these measurements, it was considered timely that all the results to date should be included in one document. References to the original publication of each resonance integral value are given.

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I. INTRODUCTION

The resonance integral of a material is defined to be its energy dependent microscopic neutron absorption cross section averaged over a $1/E$ dependent neutron flux spectrum,

$$R = \int_{E_c}^{\infty} \frac{\sigma(E)}{E} dE.$$

The lower limit of integration, though arbitrary in the definition, is chosen to include all energies not described by the thermal flux spectrum or to correspond to the cadmium cut off energy of an experiment. Recently, the acceptance of 0.5 eV has become fairly standard for E_c , and most of the results one finds in the literature adhere to this value.

Historically, reactors were designed via a two-energy group model, and the concept of resonance integral was used to calculate reaction rates in the resonance group. Now, however, with the advent of the many elegant multi-group models, reactor design proceeds by using differential cross section data, and averaging, group-wise, over a flux whose spatial and energy dependence applies to the particular reactor of interest. Hence the resonance integral plays a different role in reactor physics today. It is used primarily as a "handbook" number, which is more easily tabulated and discussed semi-quantitatively than the differential cross section data. An experimentally determined resonance integral also serves as a straightforward verification of

the differential cross section data, since the latter can be integrated over a $1/E$ flux distribution and compared directly to the measured resonance integral.

Resonance integrals have been determined experimentally several different ways. Activation measurements have contributed accurate measurements on isotopes where neutron capture leads to a radioactive nuclide that can be counted. Reactivity measurements, either static or oscillatory, can be used on any nuclide but require more sample material. Recently, the Moxon-Rae technique of counting "capture gamma rays" has been successfully applied to resonance integral measurements. No one method seems to be best, each has merit in particular instances.

This report is intended to summarize the procedures and results of resonance integral measurements made at the Advanced Reactivity Measurement Facilities (ARMF) in Idaho over the past several years. Most, if not all, the results listed have been previously reported in the open literature of the field. However, at this time, since a significant number of values have been measured, it seems pertinent to summarize all of them in one report. References to original publications will be given.

II. SAMPLES AND APPARATUS

A. Reactivity Measurement Facilities

Complete descriptions have been previously reported^(1,2) for both the ARMF-I and ARMF-II, so that a detailed discussion of the reactors is not needed here. They are designed for static reactivity measurements. The power level is maintained at a pre-set value by a servo-mechanical insertion or withdrawal of a regulating rod. The rod position is calibrated in terms of reactivity by means of period

measurements. The position during a measurement is determined to very high precision by a digital encoder and the position is averaged for a pre-set time. Several averages of this type are obtained for each reactivity measurement. The difference in the rod position with and without a sample inserted in the core is a measure of the sample's reactivity. Moderator temperature of the reactor is maintained at a constant value within a few thousandths of a degree centigrade. Resonance integral measurements are made in a cadmium lined dry tube inserted in the core (see Figure 1). As the capsule goes into the tube a cadmium lid goes on top of it, thereby completely enclosing the sample in a cadmium lined volume.

B. Hot Sample Changer

Because the cadmium tube described above is necessarily dry, it extends above the surface of the pool in which the reactors operate. Thus, samples are inserted into the tube by hand, precluding the use of the facility for making measurements on highly radioactive material. To allow remote handling and making "dry" measurements on radioactive, or "hot" samples, a special piece of equipment had to be designed and built. The hot sample changer, HSC, allows one to load up to eight samples into a round turret underwater, located just over the reactor core (see Figure 2). Once the samples are loaded, they can be dried by purging with either air or a dry gas, and remotely inserted and withdrawn from a cadmium tube extending into the reactor core.

C. Samples

Sample geometry considerations for the measurement of infinitely dilute resonance integrals must fulfill two requirements: (1) they must contain sufficient absorber to give rise to a reactivity response

CUTAWAY VIEW OF THE ARMF-II CORE

Figure 1

Figure 2 - Technician guiding hot sample changer into ARMF-II.

well above the reactor noise level, and (2) they should still be "thin" to neutron transmission. Several concentrations of absorber are required to extrapolate the results to the infinitely dilute situation. It is also desirable to make such samples economically and from a wide variety of materials.

The geometry used for samples in all our resonance integral measurements is that of a hollow cylinder whose overall diameter is 0.869 in. and is 4-1/2 in. high with 0.030 in. wall thickness (see Figure 3). This kind of sample is easily made by rolling the material of interest into plates 0.030 in. thick and then forming the plates into the cylinders of the desired dimensions.

Samples have been made in the above geometry from both alloys of metal samples with aluminum and from rolled out powder compacts of oxide samples with pure aluminum powder. Recently most of our samples have been of the powder compact type since better control is achieved in dispersion and particle size of the material to be measured. Sample controls consist of radiographs of the plates to measure homogeneity, photomicrographs of metallurgically mounted specimens to look at particle size distributions, and analytical chemistry specimens taken from areas of the rolled out plate adjacent to where cylindrical pieces are cut out. The latter analyses are used to check both homogeneity and concentration of the sample material. Since a final sample is made up of two sleeves of the type described each of which is 2-1/4 in. long, in order to make up the 4-1/2 in. height, we essentially have analyses from material at each end of the sample and also from the middle. These controls have become fairly routine, for all samples, with the exception of radioactive samples and in cases where we do not

Fabrication Of Resonance Integral Measurement Samples

Figure 3

have sufficient material to employ all the checks, and have alleviated many of the difficulties that are common to sample preparation for measurements of this type.

III. MEASUREMENT METHODS AND DATA ANALYSES

A. Reactivity Measurements

Reactivity measurements are made on the empty capsules that are used to contain the sample sleeves, and then repeated with the samples inside. In addition to the samples of varying concentration of the material of interest, measurements are made on sleeves of identical geometry made of the same pure aluminum that is used in the sample fabrication process. The measured reactivity of a loaded capsule is assumed to be the sum of the empty capsule reactivity and the reactivity of the sample inside. Thus the net reactivity of the sample itself is readily found. The reactivity of the pure aluminum sample is used to determine the reactivity worth per unit mass of aluminum, and hence allows one to subtract out the effect of the aluminum diluent in any other sample. The measurement procedure, therefore, provides one finally with a reactivity worth for varying masses of the absorber being investigated, distributed over a constant volume.

The assumption of additivity with regard to the absorber and diluent is not strictly true, and can lead to errors in interpretation. If the absorber is present in sufficient quantity to perturb the flux in the region of the sample significantly, then the reactivity response of the diluent will not be the same as in the unperturbed measurement of the pure diluent. We have investigated this effect by making measurements on gold, with three different diluents. Samples were made in several different concentrations, including zero, of gold in

aluminum, lead, and copper. Significant differences in the gold response were noted for the three different diluents at relatively large concentrations, but the result of extrapolating the results back to zero so as to get an infinitely dilute response to the gold were the same for all samples. Thus the effect is not a significant problem to this kind of measurement.

B. Comparison with Standards

In order to obtain consistency in interpreting the infinitely dilute reactivity response per atom of sample, the data are fit to an analytical expression. The expression used is a second order polynomial of the form $\rho = A + BN + CN^2$, when ρ is the reactivity, N is the number of atoms in the sample and the constants are determined in the fitting procedure. The constant term A should be very nearly zero, if the effects of the empty capsule and the aluminum diluent have been accurately subtracted out of the sample reactivity. The constant B , is the slope at the limit as N approaches zero, $B = \lim_{N \rightarrow 0} \left(\frac{\Delta \rho}{\Delta N} \right)$ and is therefore the infinitely dilute reactivity response of the material. The same kind of fitting is done to a set of standard samples, usually gold, and finally a resonance integral for the unknown material is derived by the comparison

$$R^{(X)} = \frac{B^{(X)}}{B^{std}} (R)^{std}$$

where the superscripts "std" and "(X)" refer to the standard and unknown respectively. Because the fitting is done by the method of least squares, a variance can also be calculated for B , and hence a fairly accurate method of determining the uncertainty of $R^{(X)}$ is available

which includes uncertainties in the reactivity measurements through the B's and also in the resonance integral of the standard.

C. Mixtures of Isotopes

The method described above applies to samples containing only the isotope being measured and the diluent. Often, one wants to measure the resonance integrals of the individual stable isotopes making up an element. Since one can never obtain 100% enrichment in isotopic separation, the samples will have some of each isotope present. The infinitely dilute sample reactivity will then be made up of the contributions from each isotope present, weighted by the appropriate atom density of that isotope. Thus the expression becomes a linear combination of the form

$$\left(\frac{\Delta\rho}{\Delta N}\right)_j = \sum_i \left(\frac{\Delta\rho}{\Delta N}\right)_i C_{ij}$$

where C_{ij} is the atom fraction of isotope i in a sample enriched in the isotope j ; $(\Delta\rho/\Delta N)_j$ is the reactivity per "average" atom in sample j (using an atomic weight adjusted for the amount of the various isotopes present); and $(\Delta\rho/\Delta N)_i$ is the reactivity contribution per atom of isotope i , whose atom fraction is C_{ij} . Thus if one has enriched samples of each of the isotopes, he can arrive at a system of simultaneous equations from which the unknown $(\Delta\rho/\Delta N)_i$'s can be obtained. It is apparent that measurements of this type might lead to larger uncertainties on the measured resonance integrals than for single isotope measurements. It becomes unreasonable to try to affect a measurement of a very low resonance integral isotope when other isotopes whose integrals might be much larger, are present in significant amounts. It can be seen from the table of results to

follow, that where isotopic measurements appear, any low cross section member will have a considerably higher percentage error than the high cross section isotopes of that same element. Of course the error will also depend on the isotopic enrichment as well as the magnitude of the resonance integral.

D. Corrections

Since the infinitely dilute resonance integral is defined in terms of a "slowing down" spectrum whose energy dependence is proportional to $1/E$, then any measurement of the quantity should be made in just such a spectrum. The spectrum present inside the cadmium facility of the ARMF-II has been calculated via S_{11} transport theory⁽³⁾ and found to be $1/E$ within the accuracy that such a calculation can be made, down to energies at which the cadmium begins to have an appreciable cross section. The spectrum does not deviate appreciably from a $1/E$ shape at or above the 4.9 eV resonance in gold. Below that, corrections to the measured resonance integrals must be made if the cross section is appreciable. When corrections are necessary, they are made assuming that the differential data available is accurate and this is numerically integrated over the neutron statistical weight (flux times adjoint flux) to yield a reactivity correction. The correction procedure also allows the integral to be cut off sharply at 0.5 eV by merely terminating the numerical integration. Hence, results are reported for a low energy limit of integration of 0.5 eV, which has become a fairly standard cut-off point for the purpose of reporting numbers in the literature.

E. Error Analysis

The least-squares fitting technique employed in the data analysis yields not only the values of the constants A, B, and C, but also a variance for each of these. Thus, if one includes all the reactivity measurements made on a set of samples, including each repeat measurement on each sample, as opposed to perhaps including an average of the repeated measurements for a given sample, the variance then calculated for the parameter B, is a realistic estimate of the error on the slope at infinite dilution for the sample set. This error can then be propagated in the usual fashion, together with that for the slope of the standard, and the error of the resonance integral of the standard, to determine an error for the unknown resonance integral measurement.

The error analysis affected for sets of isotopically enriched samples is slightly more involved. First one must determine the errors on the slopes at infinite dilution for each set of enriched samples [the quantities $(\frac{\Delta\rho}{\Delta N})_j$]. These errors are then propagated through the system of simultaneous equations. The errors that may be present in the C_{ij} 's are assumed to be negligible compared to those on the slopes. The error propagation involves the usual techniques, and is readily apparent after writing the determinant solution to the simultaneous equations. Finally then, the errors on the slopes of the individual isotopes $(\frac{\Delta\rho}{\Delta N})_i$ are treated as before with those of the standards to obtain final errors on each isotopic resonance integral.

IV. RESULTS

Table I lists measured as well as calculated values of resonance integrals of some 36 different elements or isotopes. The calculated values are made with the best parameters available at the time, and

include both the $1/v$ and an unresolved region contribution. The calculation is a fairly standard one, but the results are included in the Table for the purposes of comparison and to allow one to judge the adequacy of the existing parameters and/or differential data for these elements.

V. FUTURE WORK

Results reported here are the culmination of many years effort in measuring resonance integrals at the ARMF's. The program will continue along the same lines, with emphasis given to the measurement of priority items listed by the Cross Sections Advisory Group and the Advisory Committee on Reactor Physics of the AEC. We have also begun work on measuring flux averaged cross sections of interest to the fast reactor physics programs.

TABLE I

ELEMENT	REFERENCE	MEASURED RI	CALCULATED RI
As	4	81 ± 3 b.	57 b.
Ru	4	82 ± 3	59
Rh	9	1200 ± 100	1100
Pd	4	121 ± 5	126
La	4	19 ± 1	14
Nd	7	45 ± 5	30 (incomplete parameters)
Eu	4	2870 ± 85	
Eu-151	4	3740 ± 120	3208
Eu-153	4	1830 ± 55	1723
Tb	7	630 ± 25	356
Dy	6	1840 ± 180	1240
Dy-160	6	1160 ± 130	(no parameters)
Dy-161	6	1670 ± 165	
Dy-162	6	3325 ± 400	2570
Dy-163	6	1960 ± 175	1630
Dy-164	6	375 ± 35	360
Ho	7	860 ± 25	504
Er	7	1370 ± 40	920
Tm	7	1735 ± 55	1580
Yb	7	195 ± 10	129
Lu	4	840 ± 25	712
Lu-175	4	885 ± 30	707
Lu-176	4	1160 ± 35	901
Hf	8	2300 ± 60	1919
Hf-176	8	880 ± 40	112
Hf-177	8	7000 ± 240	7200
Hf-178	8	1710 ± 50	1877
Hf-179	8	595 ± 55	446
Hf-180	8	52 ± 7	31
Ta	4	735 ± 25	720
W	5	370 ± 60	320
W-184	5	7.5 ± 1.0	6.3
W-186	5	490 ± 80	460
Os	4	210 ± 10	271
Ir	4	2290 ± 70	1991
Np	4	900 ± 30	519

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