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Measurement of Epithermal Spectra in Fast Assemblies Using Resonance Sandwich Detectors*

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ABSTRACT

Resonance detectors, in general, are not suited for spectrum measurements in fast assemblies because of the contributions of $1/v$ captures and captures in resonances of higher energy. The sandwich-foil technique can be used to eliminate, for the most part, these interfering activities.

Sandwich-foil measurements were made in the fast core of the Mixed Spectrum Critical Assembly (MSCA) and in the ZPR-3 Assembly 48. The detector foil sandwiches had been previously calibrated in a known $1/E$ spectrum. The results reported in this paper are of a preliminary nature only. They show that the sandwich-foil technique does provide a measure of the fast-reactor spectra below 3 keV. However, the present state of the art only gives a qualitative picture of the spectrum.

1. Introduction

Resonance detectors, in general, have more than one resonance and a non-resonance $1/v$ cross section which cannot be neglected. In thermal-reactor spectra the contributions of the higher resonances and the $1/v$ cross sections are small compared with the main resonance, and may be minimized by using very dilute concentrations of resonance materials.⁽¹⁾ In a fast-reactor spectrum, however, the contributions of the $1/v$ captures and captures in resonances of higher energy are larger, and it is difficult to correct for these contributions to obtain the activity caused by the main resonance.

The sandwich method can be used to eliminate, for the most part, contributions to the activity from small cross sections, and the measured activity is a sum of contributions from large resonances only. The measurements reported in this paper are of a preliminary nature only. They show that the sandwich-foil technique does provide a measure of the fast-reactor spectra below 3 keV, but the present state of the art only gives a quali-

tative picture of the spectrum. Further improvements in both the experimental and analytical aspects of the problem should make the sandwich-foil measurements more quantitative. The method offers a low-energy complement to proton-recoil spectrometry which presently has a lower energy cutoff of about 1.5 keV.

2. Theory of the Measurement

The theory of the sandwich measurements has been given by Judd⁽²⁾ and Ehret.⁽³⁾ In essence, the reaction rate of a foil may be divided into two parts: one due to the strong resonances, and the other due to very weak resonances and the $1/v$ cross section. The foil thickness is chosen so that the self-shielding is small away from the strong resonances. Following Judd and assuming that the introduction of the foil does not perturb the spectrum, we may write for the foil reaction rate

$$R(t) = \int_{\text{non-res}} t \Sigma_a(u) \Phi(u) du + \sum_i R_{\text{res } i}, \quad (1)$$

where $\Phi(u)$ is the flux per unit lethargy, i denotes a particular resonance, t is the foil thickness, and R_{res} is the resonance reaction rate. The integral is taken over the lethargies where the flux depression is small, i.e., where the reaction rate is linear with t . The sum represents the strongly self-shielded resonance contributions. To calculate R_{res} for a single resonance, Judd used the Breit-Wigner line shape for the cross section. However, some detectors such as gold, indium, or tungsten have several higher-energy resonances which become important in a fast-reactor spectrum, and Doppler broadening of the line shape should be considered. Thus, assuming that a neutron is removed from a resonance by either absorption or scattering and that the fraction $\sigma_a/(\sigma_a + \sigma_s)$ results in the production of an active nuclide, one obtains for the activation rate

$$R_{\text{res } i} = \frac{1}{2} \Phi_i(u) \Gamma_i \int_0^\infty [1 - 2E_3(Z, \Psi(\theta, x))] dx, \quad (2)$$

where $\Phi_i(u)$ is the flux per unit lethargy at the reso-

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nance energy, Γ_γ is the gamma-radiation width, $E_3(x)$ is the exponential integral $\int_1^\infty (e^{-xt}/t^3) dt$, Z is the foil thickness in total mean free paths at the peak of the resonance, and $\Psi(\theta, x)$ is the usual Doppler-broadened Breit-Wigner line shape. The assumption that neutrons are removed from a resonance by one elastic scattering is valid for most foil materials under consideration since the total widths, Γ , are small relative to the average energy loss per collision. If a sandwich of three identical foils is irradiated, the reaction rates of the outer and inner foils of the sandwich are, respectively,

$$C_0 = \frac{1}{2}R(t) - \frac{1}{2}R(2t) + \frac{1}{2}R(3t); \quad (3)$$

$$C_i = R(2t) - R(t),$$

and therefore the difference in activity is

$$C_0 - C_i = \frac{1}{2}[3R(t) - 3R(2t) + R(3t)]. \quad (4)$$

Upon substituting Eq. (1) in Eq. (4) it is seen that the integral term vanishes because it is linear with t . The remaining difference activity, C , is due to the strong resonance contribution and may be expressed as

$$C = C_0 - C_i = \alpha \sum_{i=1}^n K_i \Phi_i(u), \quad (5)$$

where α is a proportionality constant typical of each material. It includes the detector efficiency, constant gamma width (Γ_γ), and the amount of material, as well as a flux-normalization factor. Also, $K_i = [3I(t) - 3I(2t) + I(3t)]$, where the I 's are the integrals in Eq. 2 taken for one, two, and three foil thicknesses.

By irradiating the sandwich foils in a flux that is known to be $1/E$ -dependent, the constant α is determined from

$$\alpha = \frac{C(1/E)}{\Phi_{1/E} \sum_{i=1}^n K_i}, \quad (6)$$

where $C(1/E)$ is the $1/E$ difference activity. Thus an unknown flux at the lowest-energy resonance, $\Phi_{1X}(u)$, may be expressed as

$$\Phi_{1X}(u) = \frac{1}{K_1} \left\{ \frac{C(X)}{C(1/E)} \Phi_{1/E} \sum_{i=1}^n K_i - \sum_{i=2}^n K_i \Phi_{iX}(u) \right\}, \quad (7)$$

where $C(X)$ is the measured difference activity in the unknown flux. If there is only one resonance, as in La-139, Cd-114, and Na-23, the activity difference between the outer and inner foil is due only to this resonance and no corrections are necessary, i.e.,

$$\Phi_{1X}(u) = \frac{C(X)}{C(1/E)} \Phi_{1/E}. \quad (8)$$

The data from the single-resonance detectors are used as bases for computing the higher-resonance contributions for the other sandwich materials.

3. Description of the Measurements

3.1. MSCA Fast Core

Sandwich-foil measurements were made at Vallecitos Atomic Laboratory as part of the Mixed Spectrum Superheater Critical Experiments.⁽⁴⁾ The Mixed Spectrum Critical Assembly (MSCA) is a full-scale mockup of a 75- to 150-MWe power plant reactor core. The reactor contains four distinct fuel zones: a fast core, a fast buffer, a thermal buffer, and a thermal core. The fast core of approximately 407 liters contains about 2000 kg of UO_2 with an average enrichment of 23.1 % U-235, and 1100 kg of Inconel. The fast buffer zone contains 1.3 w/o enriched UO_2 fuel rods clad in stainless steel and effectively absorbs most of the thermal-neutron current emanating from the thermal region. This arrangement results in an asymptotic spectrum in the fast core which approximates that of a large, dilute, fast ceramic reactor.

Indium, gold, tungsten, lanthanum, cadmium, sodium, and manganese were found to be suitable for the sandwich measurements. The foil thicknesses were chosen so that the center foils were substantially self-shielded at the dominant resonance cross section but had much smaller or negligible self-shielding at the other resonance and non-resonance cross sections. The foil thicknesses and materials used are given in Table 1.

All foils were 0.5 in. in diameter and were enveloped by a ring foil to position the foils and to prevent edge effects. The ring foils were three times the thickness of the single detector foils and were 0.75 in. in diameter. Each sandwich was taped together with small pieces of Teflon tape. To prevent oxidation of the lanthanum foils during the experiment, a thin coating of silicon vacuum grease was used. All sandwiches, including those of cadmium, were irradiated in small boxes of 20-mil-thick cadmium. This prevented a thermal-flux depression in the sandwiches during the $1/E$ calibration. To verify the cadmium measurements, cadmium sandwiches were also irradiated in covers of enriched B-10.

The foils were first irradiated in the Nuclear Test Reactor at Vallecitos, which has a $1/E$ -dependent epithermal spectrum. The gamma activities were measured with a 3 x 3-in. NaI detector and a 256-channel analyzer. In this manner the foil response and detector efficiencies were calibrated for the known spectrum. In the $1/E$ spectrum, greater than 97 % of the difference activity measured for each material was due to its lowest resonance. The measured differences varied from

about 0.5 % of a single-foil activity for sodium to about 50 % for indium. Considerable care was exercised to obtain good statistics for the smaller difference activities.

Identical sandwiches were subsequently irradiated at the center of the fast core of the MSCA, and the foils were counted in the same manner as before. The relative fluxes at the lanthanum, cadmium, and sodium resonances were determined by means of Eq. (8). Based on these results, an iterative procedure was used to apportion the measured difference activities among the several contributing resonances for the tungsten, gold, and indium foils. For the iteration it was assumed that the flux $\Phi(u)$ had the form aE^b , i.e., a straight line on a log-log plot of $\Phi(u)$ vs E .

Since the manganese resonances are wide compared with the average energy loss per elastic collision, the broad higher resonances should not contribute to the measured difference activity. For this reason the manganese activity was assumed to be due only to the contribution of the lowest resonance. A more detailed treatment of the manganese data is required, however, to assure its validity.

3.2. ZPR-3 Assembly 48

A similar sandwich-foil measurement was made at the center of the ZPR-3 Assembly 48.⁽⁵⁾ Since the neutron spectrum was expected to be harder, the indium-foil measurements were omitted. Three sets of foils were irradiated at the core center and two sets in the blanket. The manganese foils were counted at ZPR-3 using a Co-58 standard to intercalibrate with the Vallecitos counting equipment. The other foils were counted at Vallecitos, and the data were treated in the above manner.

4. Results and Discussion

The results of the MSCA measurements are shown in Fig. 1, and compared with a calculated 24-group spectrum by a least-squares normalization. Several of the calculated higher-resonance points are included together

FIG. 1. Spectrum in Fast Core of MSCA.

with the percentage of the measured activity contributed by the resonance. It is seen that only 3.3 % of the gold activity is due to the 4.9-eV resonance, accounting for the large uncertainty in that point. The higher resonances, containing more of the activity, have smaller uncertainties and so may be used as flux indicators. The cadmium point is an average of the cadmium- and boron-covered sandwich measurements, which agreed to within 10 %. The large error bars on all of the data were primarily due to the fact that anisotropies in the flux often caused the two outer foils to have different activities. Theoretical considerations indicate that small flux gradients across the sandwiches may be neglected and the activities of two outer foils averaged. However, it was decided at this state of the measurement to treat the differences as errors. By neglecting this uncertainty the error bars would have been reduced to well under ± 20 % in most cases.

Except for the sodium point, which appears to be low, and the cadmium point, which appears to be a little high, the measured spectrum is in general agreement with the calculation.

The results of the measurements at the center of ZPR-3 Assembly 48 are shown in Fig. 2, and compared with two preliminary calculated spectra: a 24-group calculation by ANL and a 60-group calculation by General Electric. The blanket data did not yield significant results because of low count rates.

In Fig. 2 it is seen that the nuclides that have higher resonance corrections are not in agreement with the other nuclides. It is probable that two factors contribute to this. One is that there are very few neutrons

TABLE 1. SANDWICH-FOIL MATERIALS

Nuclide	Energy of Principal Resonance, eV	Material		Thickness (mils)
In-115	1.46	In-Pb	50% alloy	6
Au-197	4.9	Au	Metal	1.5
W-186	18.8	W	Metal	2
La-139	73.5	La	Metal	10
Cd-114	120	Cd	Metal	20
Mn-55	335	Mn-Cu	80%-20% alloy	2
Na-23	2950	NaCl	Crystal	20

FIG. 2. Spectrum in ZPR-3 Assembly 48.

t the energies of the lower gold and tungsten resonances, as predicted by the 24-group calculation. The other is that the contributions of the higher resonances are overestimated. This effect would become more sig-

nificant in a spectrum with a smaller low-energy tail such as is indicated for Assembly 48.

The method of correcting for the higher resonances was further investigated by irradiating 7-foil sandwiches of gold and tungsten in a $1/E$ spectrum. The normalized measured difference activities are shown in Fig. 3, together with values calculated in a manner analogous to Equations 2 through 5. The agreement appears to be quite good. However, the data were normalized at the outer foils and, in the $1/E$ spectrum, the difference activity of the outer foil is dominated by the lowest-energy resonance. This has the effect of masking the contributions of the higher-energy resonances. It is seen that the experimental points for foils (2 and 6) and (3 and 5) fall somewhat below the analytical values, particularly for the tungsten. Even a slight overestimation of the higher-energy resonance contributions would become quite significant in the fast spectra of the MSCA and Assembly 48. For this reason, additional effort, both analytical and experimental, is still required in order to obtain a more consistent picture of the resonance self-shielding in the sandwich foils. Measurements with 7-foil sandwiches in the fast spectra should be particularly useful.

It should be noted in both Figures 1 and 2 that the

FIG. 3. Flux Depression in Seven-foil Sandwiches in a $1/E$ Spectrum.

cadmium data points appear high in each case. A possible cause is that flux depressions due to the other cadmium isotopes cause measurable perturbation in the Cd-114 difference activities. This is not unreasonable since, while the $1/v$ capture cross section is low at the higher energies, the relative flux is much greater. This perturbation effect should be evaluated for all of the foil materials containing more than one nuclide.

The results presented at this time are of a preliminary nature only. There is still a need for considerable further work, particularly in the analysis of the resonance self-shielding effects that give rise to the measured difference activities. However, it is believed that the experimental results to date, even using the simplest analytical approach, are encouraging. With additional effort the sandwich-foil technique should offer a quantitative measure of fast reactor spectra below 3 keV.

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Discussion

Mr. Weale (UK-Aldermaston): I would like to comment really to point out something you didn't mention but no doubt realize, that this is a potentially valuable technique as compared with the proportional counter and time-of-flight methods. It is an in-core technique and it can be put into a very small cavity in the core without such problems as disturbing the material around the sample. We have tried to use manganese. It seems that if one assumed that the flux was known, one could get the slope of the spectrum in the region from 330 eV to the next higher resonance in manganese at 1 keV. Alternatively, if you know the

slope of the spectrum you could get its absolute value, but in both cases the results don't seem very reliable. We found errors of about 30 % or so in this.

Mr. Weitzberg: An additional difficulty with manganese is that the high resonances are quite broad, and the assumption that you can remove a neutron from a resonance just by single scattering is not valid. Inherent in Judd's first formulation was the assumption that a neutron is removed either by absorption or scatter, and then one takes the ratio Σ_a/Σ_t . Now I think in broad resonances you get a contribution from multiple scattering and this should be taken into account. In fact, simple analysis obviously doesn't work, and it's a question of going perhaps to ELMOE-type calculations, not only for manganese, but for the others as well.

Mr. Weale: McCracken at Harwell did some interesting work on this recently, involving measurements in the ZEBRA blanket. This was reported at the International Conference on Radiation Measurements in Nuclear Power, held at Berkeley, England, in September 1966. It would be interesting to compare that work.

Mr. Pearlstein (BNL): One of the comprehensive experimental papers on the sandwich-foil technique is that by Ehret of Germany. In his paper he showed that he could not get agreement between calculated and experimental ratios of the outer foil to the inner foil. Now at Brookhaven, what we did was a calculation including Doppler broadening and scattering, and we produced agreement. Unless you calibrate your foils in a flux that you know to be $1/E$, you are in trouble if you rely on calculations that don't include these sophisticated corrections.

Mr. Weitzberg: I agree that $1/E$ calibration is the only absolute standardization that you can get, but unfortunately such a spectrum deviates drastically from a fast spectrum. You would be shifting emphasis from where your activity is perhaps 97 % in the lowest energy resonance to where it is less than 1 %. It would be nice if there were some higher-energy spectrum that could be used but this is not the case.

Mr. Weale: Could you comment on the possibility of calibrating the foil method using time-of-flight techniques?

Mr. Weitzberg: Yes, this would be valuable.

Mr. Kulian (AEG—Germany): At the 1965 Conference here (ANL-7120), some Doppler activation measurements in the MSCA were reported. The comparison with theory indicated that the experimental spectrum might be softer than calculated. Now, you show good agreement with the calculated spectrum within the limits of error. I would like to know if you checked against those old Doppler measurements?

Mr. Weitzberg: Yes. In the work which you men-

tioned the Doppler results were in fairly good agreement with this spectrum. This was the so-called soft spectrum. One of the possible reasons for the fact that the gold and indium sandwich foils gave quite high results was the possibility of streaming in the assembly. When these measurements were repeated, we took great care to do the measurements in a little central void. Now the sandwich-foil measurements are fairly consistent with the calculations, which are in turn consistent with the calculated Doppler effect and that measured by Pflasterer. The reason for the large error bars is because there is very small activity in the gold. The error bars are still quite large, and the reason for this is that in doing these measurements I have decided that, when anisotropies in the flux result in different outer foil activities, I would treat them as errors rather than averaging them. The theory indicates that for small flux gradients you could just average the outer

two foils, but I think that in the present state of the technique larger error bars are more honest.

Mr. Stegemann (Karlsruhe): I have a comment on the sodium-activation measurements. There are three points which have to be considered. The first one was mentioned by Weitzberg—the interference with chlorine. You can get rid of this if you use sodium fluoride instead of sodium chloride, which we did with measurements in STARK. The second thing is that for intensity reasons you have to use thicker foils of sodium fluoride. You have to take into account multiple scattering. This has been done, at our Laboratory, by a numerical procedure which will be published later. The third thing I would like to mention is that if you measure the sodium activity absolutely by gamma-gamma coincidence techniques, you have to be careful because there are angular correlation effects, requiring a correction factor of up to 10 %.