

account observations on polycrystalline graphite, it appears that the effect of irradiation in pyrolytic graphite is the intercalation of carbon complexes, probably larger than 3 atoms, between the layer planes.

These data also aid in understanding high-temperature radiation effects in polycrystalline graphite². The dimensional changes observed in pyrolytic graphite could be predicted qualitatively on the basis of the analysis of high-temperature neutron damage to polycrystalline graphite³, which predicts an expansion of the crystallites in the *c* direction and a contraction in the *a* direction. Because pyrolytic carbon is very nearly theoretical density and possesses a high degree of anisotropy (properties also common to crystallites) it was expected that radiation effects in pyrolytic carbon would exhibit the behavior inferred for the crystallites in polycrystalline graphite. These results for pyrolytic material thus imply that high-temperature radiation-induced contraction in polycrystalline reactor graphites must occur either by decreasing the porosity or by densifying any carbon that was initially less than theoretical density, but not by a volume contraction in the graphite crystallites.

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²R. E. NIGHTINGALE, Ed., *Nuclear Graphite*, Ch. 9, Academic Press, N. Y. (1962).

³D. R. de HALAS and H. H. YOSHIKAWA, *Proc. Fifth Carbon Conf.*, Pgs. 249-255, Pergamon Press, New York (1962).

Isotopic Hafnium Resonance-Absorption Data

The extensive use of hafnium as a control material in nuclear reactors has made necessary an accurate knowledge of its nuclear absorption properties. Although knowledge of the hafnium differential absorption cross section is fairly well detailed, available resonance data for the even-even isotopes is limited, and significant discrepancies still exist between calculated and experimental resonance integrals for natural hafnium. In this regard, I. Itkin¹ has pointed out that the upper and lower bounds on the calculated resonance integral are 1720 and 2150 barns while reported

experimental values range from 1300 to 2800 barns.

As part of the analytical and experimental investigations of the use of hafnium as a burnable poison in intermediate spectrum reactors², a sample of natural hafnium mixed with fuel was prepared for irradiation in such a system. The text specimen was a fuel element with 2 wt-% natural hafnium added to the fuel. After irradiation, radio-chemical and mass spectrometric analyses were performed at KAPL by F. M. Rourke and F. W. Walker to yield hafnium isotopic ratios and fuel-depletion data for several sections of the specimen.

This letter summarizes the results of the study of this isotopic-depletion data made to assess the present knowledge of the isotopic hafnium absorption cross sections in the resonance energy region. Isotopic-depletion data obtained in an intermediate-spectrum reactor emphasizes this energy region of interest.

For the system^a of Hf^{176,177,178,179,180} and U²³⁵, the experiment provides a value for the fuel depletion and for four ratios of the five isotopic number densities for each section of the specimen. Two interrelated procedures for comparison of the experimental data with analytical predictions are employed. The first procedure uses the solution to the system of sequentially coupled linear differential equations describing the time dependence of the isotopic hafnium number densities to calculate the isotopic ratios on the basis of the BNL-325, 2nd Ed. cross-section data and knowledge of the intermediate spectrum as obtained from experiment and analysis. Comparison of the calculated and measured isotopic ratios then provides a measure of the adequacy of the cross-section data. The second procedure consists of inverting these solutions, which describe the time dependence of the hafnium system, to obtain the ratios (α_i) of the effective absorption cross section of the *i*th hafnium isotope to the effective absorption cross section of fuel^b in terms of the experimental isotopic-

^aExamination of the isotopic depletion of natural hafnium shows that the contribution to the number density of Hf¹⁷⁶ by neutron captures in Hf¹⁷⁴ is negligible for the values of fuel depletion of interest.

$$\alpha_i = \frac{\int_R dr \int_0^\infty dE \sigma_i(E) \phi(r, E)}{\int_R dr \int_0^\infty dE \sigma_u(E) \phi(r, E)}$$

where the spatial integration is taken over the specimen, and σ_i and σ_u are respectively the microscopic absorption cross sections for the *i*th isotope of hafnium and for fuel.

²The Physics of Intermediate Spectrum Reactors, UC-81, (September 1958).

¹WAPD-TM-324, "Analysis of the Neutron Capture Cross Section and Resonance Integral of Hafnium," I. ITKIN, (August, 1962).

number density ratios. Comparison of these values with those calculated from BNL-325, 2nd Ed. data provides a more direct measurement of the adequacy of the hafnium cross-section data.

Table I summarizes the experimental and calculated isotopic ratios for two sections of the specimen that experienced significantly different fuel depletions.

The following should be noted from an examination of the results presented in Table I.

1. Using the values for the α_i 's based on available cross section information, the measured isotopic ratios can be predicted to within about 15% for a fuel depletion of about 7%.
2. Since all calculated ratios relative to Hf^{176} are low in comparison to experiment, it is suggested that the value for α_{176} is low.

Table II summarizes the values for α_i calculated from BNL-325, 2nd Ed. cross-section data and those obtained by inversion of the depletion equations. The uncertainties shown for the α 's obtained by inversion are based on a conventional linear error analysis taking into account the 1% precision of the ratios and the 5% precision in the fuel-depletion determination. Table II also lists an average α_i selected to minimize the error in predicted isotopic ratios for both data points. When the depletion calculation is repeated using the average α_i 's, the maximum error as presented in Table I is reduced from 15% to 5%, and the errors at both fuel depletions are of opposite sign and comparable magnitude indicating a best fit to α_i satisfying both sets of data.

It is seen that the calculated value for α_{176} is low by a factor of about sixty, the values calculated for α_{177} and α_{178} are in agreement with experiment to within about 10%, and those calculated for Hf^{179} and Hf^{180} seem to be low notwithstanding the large errors assigned to the α 's obtained by inversion. The results shown in Table II are consistent with those obtained by D. J. Anthony and R. T. Frost³

³*Ibid*, Chapter 17, *Critical Assembly Investigation of Burnable Poisons*, by D. J. ANTHONY and R. T. FROST, Table 17.1.

TABLE I
Isotopic Ratios, N_i/N_j

| i/j | Fractional Fuel Depletion (U^{235}) 0.052 | | | Fractional Fuel Depletion (U^{235}) 0.074 | | |
|---------|--|-------|---------|--|-------|---------|
| | Exp. | Calc. | % Diff. | Exp. | Calc. | % Diff. |
| 180/179 | 2.12 | 2.06 | -2.8 | 1.98 | 1.89 | - 4.5 |
| 180/178 | 1.17 | 1.19 | +1.7 | 1.18 | 1.19 | + 0.8 |
| 180/177 | 3.19 | 3.15 | -1.3 | 3.57 | 3.94 | +10.0 |
| 180/176 | 7.28 | 6.87 | -5.6 | 7.43 | 6.92 | - 6.9 |
| 180/176 | 7.28 | 6.87 | -5.6 | 7.43 | 6.92 | - 6.9 |
| 179/176 | 3.43 | 3.34 | -2.6 | 3.75 | 3.66 | - 2.4 |
| 178/176 | 6.22 | 5.79 | -6.9 | 6.28 | 5.84 | - 7.0 |
| 177/176 | 2.28 | 2.18 | -4.4 | 2.08 | 1.76 | -15.0 |

from reactivity-coefficient measurements employing isotopically enriched samples.

Since no resonance parameters are listed for Hf^{176} in BNL-325, 2nd Ed., the calculated value for α_{176} is based on an assumed $1/v$ energy dependence of the absorption cross section. The discrepancy between the calculated and experimental value of α_{176} is sufficiently large that the difference may be attributed to undetected resonances. Preliminary results from experiments performed by T. Fuketa and J. E. Russell⁴ of Rensselaer Polytechnic Institute using the RPI Linac confirm this hypothesis.

Using isotopically enriched hafnium samples, Fuketa and Russell performed capture and transmission measurements at RPI and Oak Ridge. Even though the analysis of these experiments required to obtain the resonance parameters is incomplete, the resonance energies are available. For the energy region above 1.7eV the ten lowest lying resonances for Hf^{176} were found at 48.3, 53.3, 67.1, 122.4, 123.9, 133.9, 177., 201.5, 243., and 255eV. An examination of the transmission data indicates that the lowest lying resonance at 48.3eV is a strong resonance. However, any quantitative statements about its strength must await the completion of their analysis.

⁴T. FUKETA and J. E. RUSSELL, Private Communication, (March 1964).

TABLE II

| | α_i Calculated from BNL-325 2nd Ed. Data | α_i Obtained from Inversion of Depletion eqns. for fuel depletion of 0.052 | α_i Obtained from Inversion of Depletion eqns. for fuel depletion of 0.074 | Average α_i Obtained by Inversion |
|----------------|---|---|---|--|
| α_{176} | 0.016 | 1.02 ± 0.36 | 0.95 ± 0.25 | 0.98 ± 0.35 |
| α_{177} | 9.2 | 9.74 ± 0.63 | 8.25 ± 0.51 | 8.85 ± 0.59 |
| α_{178} | 2.7 | 2.62 ± 0.59 | 2.48 ± 0.41 | 2.55 ± 0.57 |
| α_{179} | 0.72 | 1.11 ± 1.4 | 0.97 ± 0.95 | 1.04 ± 1.4 |
| α_{180} | 0.069 | 0.16 ± 0.97 | 0.18 ± 0.67 | 0.17 ± 0.93 |

Finally, it should be noted that the data employed for Hf^{177} and Hf^{178} , which yield good agreement with the intermediate-spectrum experiment, also represent the major contribution ($\sim 95\%$) to the conventional natural-hafnium resonance integral of about 2000 barns. Thus the results of the present study are consistent with the work of F. Feiner⁵ and I. Itkin¹ where it is concluded that a resonance integral closer to 2000 rather than 2800 barns is appropriate for natural hafnium.

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⁵KAPL-2000-16, Reactor Technology Report No. 19 -
Physics, December 1961, "Resonance Integrals of Manganese, Hafnium and Niobium," by F. FEINER.

In^{115} Thermal Cross Section Ratios*

In a recent paper by K. H. Beckurts *et al.*¹, data are given for the ratio of thermal activation cross sections for the 14-sec and 54-min activities of In^{116} produced by capture of thermal neutrons in In^{115} which is in "striking disagreement" with that obtained by the Greenfield-Koontz's² re-evaluation of Domanic and Sailor's³ experiment.

*This work was done under the auspices of the USAEC.

¹K. H. BECKURTS, M. BROSE, M. KNOCH, G. KRÜGER, W. PÖNTIZ and H. SCHMIDT, *Nucl. Sci. Eng.* **17**, No. 3, 329 (1963).

²M. A. GREENFIELD and R. L. KOONTZ, *Phys. Rev.* **123**, 197 (1961).

³F. DOMANIC and V. L. SAILOR, *Phys. Rev.* **119**, 208 (1960).

Domanic and Sailor had published data on the dependence of the In^{116} activation ratio on neutron energy. They found that foils of different thicknesses (26.8 and 96 mg/cm²) gave quite different values for the activation ratios for pile neutrons; their values are listed as $A_{14 \text{ sec}}/A_{54 \text{ min}}$ in column 2 of Table I. In a subsequent publication² we suggested the need to take account of the rather different values for self-absorption and self-scattering for these foil thicknesses for the beta spectra corresponding to the 54-min activity. In Reference 2 application of the correction factor for an end-window G.M. detector with narrow geometry (7%) did remove the anomaly observed by Domanic and Sailor³. The correction factors were used for the 54-min activity only.

The choice of the narrow geometry correction factors has now been reviewed, and it has become apparent that the self-absorption and self-scattering factors appropriate for 2π or 4π geometry should have been used. This becomes clear when one examines the geometry employed by Domanic and Sailor³. Values for the correction factors, $f_{s,54 \text{ min}}$, for 2π geometry^{4,5} are given in column 3 of Table I for the foil thicknesses of interest. It also appears from the work of Beckurts *et al.*¹, that a correction factor for the short-lived activity, $f_{s,14 \text{ sec}}$, should be used. Table I summarizes the original data of Domanic and Sailor as well as the various correction factors just described, and also presents corrected ratios of the activities; i.e. the ratio of thermal activation cross sections.

Table I indicates that the anomalous results for differing foil thicknesses are removed by including the self-absorption and self-scattering correction factors. Further, the application of the correction factors of Table I to other data reported by Domanic and Sailor³ indicates no dependence of the ratio of the 14-sec to 54-min activities on the neutron energy up to 2.66 eV (as had been pre-

⁴M. A. GREENFIELD, R. L. KOONTZ, A. A. JARRETT, *et al.*, *Nucleonics* **15**, No. 3, 57 (1957).

⁵M. A. GREENFIELD, R. L. KOONTZ and A. A. JARRETT, "Absolute Beta Counting of Indium Foils," Part II, NAA-SR-1137, Atomics International, (1955) (unpublished).

TABLE I
Ratio of 14-sec to 54-min Activities of In^{116} Corrected for Self-Absorption and Self-Scattering based on 2π or 4π Geometry.

| Foil thickness mg/cm ² | Ratio of activities ³ $A_{14 \text{ sec}}/A_{54 \text{ min}}$ | $f_{s,54 \text{ min}}$ ^{4,5} (for 2π or 4π geometry) | $f_{s,14 \text{ sec}}$ ¹ | Corrected ratio of activities $\frac{A_{14 \text{ sec}}/f_{s,14 \text{ sec}}}{A_{54 \text{ min}}/f_{s,54 \text{ min}}}$ |
|--------------------------------------|---|--|-------------------------------------|--|
| 96 | 0.586 | 0.339 | 0.86 | 0.231 |
| 26.8 | 0.331 | 0.632 | 0.94 | 0.223 |