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Nuclear Coherent Scattering Amplitudes for Thorium, Uranium and Plutonium*

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Neutron diffraction transmission techniques were used to determine experimentally the nuclear coherent scattering amplitudes of thorium, uranium and plutonium. The thermal neutron wavelength employed was 1·391 Å, and experimental samples consisted of thin plates of ThO_2, UO_2, and PuO_2. Nickel, with a scattering amplitude of $1{\cdot}03 \times 10^{-12}$ cm., was used as the calibration standard. The experimental scattering amplitudes of thorium, uranium, plutonium and oxygen were determined to be 0·98, 0·84, 0·75, and $0{\cdot}58 \times 10^{-12}$ cm., respectively. These values are in good agreement with those reported by Bacon (1955) for thorium, uranium and oxygen, namely, 1·01, 0·85, and $0{\cdot}58 \times 10^{-12}$ cm., respectively.

Introduction

Determination of the nuclear coherent scattering amplitude of plutonium is required before a quantitative neutron diffraction investigation of plutonium, its alloys and compounds, can be accomplished. The techniques described here enabled that information for plutonium to be obtained. Preliminary work indicated that the neutron diffraction transmission technique used with very thin samples could be employed more successfully than either the reflection technique used with thick slabs of material or the transmission technique used with cylindrical samples, and was responsible for the choice of technique and sample geometry.

Theory

For a thin slab sample of powdered material having sufficiently large dimensions to intercept adequately an entire monochromatic beam of neutrons, the integrated intensity, I, from a set of hkl planes when the sample is placed in the symmetrical transmission position, is given by the following equation

$$I_{hkl} = K^2 t \frac{\varrho'}{\varrho} \frac{\exp\left[-\mu t \sec\theta\right]}{\sin^2 2\theta} j N_c^2 F_{hkl}^2 \times \exp\left[-2B \sin^2\theta/\lambda^2\right], \quad (1)$$

where K is a constant of the experiment and includes consideration of the geometry of the spectrometer and the intensity as well as the neutron wavelength of the incident beam. The quantity t is the thickness of the experimental sample. The quantity ϱ'/ϱ is the ratio of the measured density, ϱ', of the sample to its theoretical density, ϱ, which is calculated from the atomic weights of the elements composing the crystal and its known crystal structure. The quantity μ is the linear absorption coefficient of the sample and j is the multiplicity of the hkl planes. N_c is the number of unit cells per cm.^{3}, and B is the thermal parameter for the specimen. F_{hkl} is the structure amplitude factor per unit cell, which is a function of the nuclear scattering amplitude and number of each type of atom in the unit cell.

The transmission arrangement permits the correction for absorption in the sample to be calculated easily; this correction is represented by the term $\exp[-\mu t \sec\theta]$ in equation (1). The value of $\exp[-\mu t]$ is determined experimentally by direct measurement of the attenuation of the monochromatic beam by the sample in the 'straight through' or zero θ position.

Experimental results

The samples consisted of powder slabs of nickel, ThO_2, UO_2 + 15 vol./o. aluminum and PuO_2 + 15 vol./o. aluminum. The uranium and plutonium samples were encased in vanadium foil. The integrated intensities obtained for these samples are given in Table 1 and were obtained by summing neutron counts over a reflection and then subtracting back-ground.

Table 1. *Integrated intensities for experimental samples*

hkl	Ni	ThO_2	UO_2	PuO_2
111	448·1	126·3	14·4	7·1
200	246·1			
220	292·4	333·8	45·7	22·1
311		110·9	11·6	5·7
400		83·5	12·5	5·1
331		71·3	7·6	3·8
422		242·8	32·7	13·2

The information given in Table 1 was obtained on the neutron diffraction spectrometer at the Omega West Reactor at this Laboratory. The neutron wavelength was 1·391 Å, and was obtained by using the (111) planes of a lead crystal in transmission. The beam was collimated to give a width at half maximum of about 1·1° for a peak at a 40° scattering angle.

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The experimental results appropriate to each sample are derived and discussed below.

Nickel

The nickel sample was used to determine K^2 in equation (1). The nuclear scattering amplitude of nickel, b, is equal to $1{\cdot}03 \times 10^{-12}$ cm., (Bacon, 1955), and, since there are 4 atoms of nickel per unit cell, $F=4b$. The numerical values of the other constants used in equation (1) are as follows:

$$t=0{\cdot}179 \text{ cm.}, \quad \varrho'=6{\cdot}15 \text{ g.cm.}^{-3}, \quad \varrho=8{\cdot}90 \text{ g.cm.}^{-3},$$
$$a_0=3{\cdot}525 \text{ Å}, \text{ and } \exp[-\mu t]=0{\cdot}791 .$$

Rearranging equation (1) and substituting in it these values and the data of Table 1 yields an equation which has the general form

$$y=a \exp[cx], \qquad (2)$$

where $a=K^2$, $c=-2B$ and $x=\sin^2\theta/\lambda^2$. Equation (2) was programmed for an IBM 704 computer so that, for given values of x and y, a least-squares analysis could be performed to find a and c and their standard deviations. For nickel,

$$K^2=27298 \pm 1523 \quad \text{and} \quad 2B=0{\cdot}505 \pm 0{\cdot}518 .$$

Thorium dioxide

The ThO_2 sample was used as an independent check on the validity of K^2 as determined for the nickel sample. Values of the experimental constants for use in equation (1) are as follows:

$$t=0{\cdot}318 \text{ cm.}, \quad \varrho'=7{\cdot}29 \text{ g.cm.}^{-3}, \quad \varrho=10{\cdot}07 \text{ g.cm.}^{-3},$$
$$a_0=5{\cdot}584 \text{ Å} \text{ and } \exp[-\mu t]=0{\cdot}890 .$$

Substituting these values, the data from Table 1, and the value of K^2 determined for the nickel sample into equation (1) yields an equation similar to equation (2) except that $a=F^2$. Since ThO_2 has the CaF_2 structure, three different sets of F_{hkl} are available. One set, for which $h+k+l=2n+1$, consists of thorium atoms alone. The second set, for which $h+k+l=4n$, consists of thorium + 2 oxygen atoms. The third set, for which $h+k+l=4n+2$, consists of thorium − 2 oxygen atoms, but since the nuclear coherent scattering factor of thorium is approximately twice that of oxygen the intensities of these peaks are nearly equal to zero. Least squaring the data according to equation (2) yields values of $15{\cdot}47 \pm 0{\cdot}09$ for $|4b_{Th}|^2$ and $73{\cdot}13 \pm 2{\cdot}54$ for $|4(b_{Th}+2b_O)|^2$. From these values, $b_{Th}=(0{\cdot}98 \pm 0{\cdot}01) \times 10^{-12}$ cm. and $b_O=(0{\cdot}58 \pm 0{\cdot}02) \times 10^{-12}$ cm. are derived. These results are in good agreement with those reported by Bacon (1955), $b_{Th}=1{\cdot}01$ and $b_O=0{\cdot}58$. Experimental values of 2 B are $0{\cdot}83 \pm 0{\cdot}06$ for the thorium atom set and $1{\cdot}76 \pm 0{\cdot}27$ for the thorium + 2 oxygen atom set.

Uranium dioxide

The UO_2 + 15 vol./o. aluminum sample was designed as a mock-up for the plutonium sample. The 15 vol./o. aluminum was added to increase the strength and to decrease the brittleness of the sample. The sample was very thin,

$$t=0{\cdot}038 \text{ cm.}, \quad \varrho'=7{\cdot}67 \text{ g.cm.}^{-3}, \quad \varrho=9{\cdot}72 \text{ g.cm.}^{-3},$$
$$a_0=5{\cdot}470 \text{ Å} \text{ and } \exp[-\mu t]=0{\cdot}976 .$$

Since UO_2 and ThO_2 have the same structure the data for these two compounds were processed in a similar manner. The data for UO_2 yield a value of $11{\cdot}39 \pm 0{\cdot}54$ for $|4b_U|^2$ and $66{\cdot}61 \pm 5{\cdot}46$ for $|4(b_U+2b_O)|^2$. From these values, $b_U=(0{\cdot}84 \pm 0{\cdot}02) \times 10^{-12}$ cm. and $b_O=(0{\cdot}60 \pm 0{\cdot}05) \times 10^{-12}$ cm. are derived. These results are also in good agreement with those reported by Bacon (1955), $b_U=0{\cdot}85$ and $b_O=0{\cdot}58$. Experimental values for 2 B are $1{\cdot}59 \pm 0{\cdot}48$ for the uranium atom set and $1{\cdot}97 \pm 0{\cdot}61$ for the uranium + 2 oxygen atom set.

Plutonium dioxide

For the PuO_2 + 15 vol./o. aluminum sample,

$$t=0{\cdot}038 \text{ cm.}, \quad \varrho'=7{\cdot}10 \text{ g.cm.}^{-3}, \quad \varrho=10{\cdot}25 \text{ g.cm.}^{-3},$$
$$a_0=5{\cdot}397 \text{ Å} \text{ and } \exp[-\mu t]=0{\cdot}660 .$$

Processing the data in a manner similar to that used for UO_2 yields a value of $8{\cdot}99 \pm 0{\cdot}56$ for $|4b_{Pu}|^2$ and $55{\cdot}70 \pm 1{\cdot}00$ for $|4(b_{Pu}+2b_O)|^2$. From these values, $b_{Pu}=(0{\cdot}75 \pm 0{\cdot}03) \times 10^{-12}$ cm. and $b_O=(0{\cdot}56 \pm 0{\cdot}02) \times 10^{-12}$ cm. are derived. The result for oxygen is in good agreement with that reported by Bacon (1955), $b_O=0{\cdot}58$, whereas, to our knowledge, the result for plutonium is entirely new. Experimental values of 2 B are $1{\cdot}00 \pm 0{\cdot}60$ for the plutonium atom set and $2{\cdot}83 \pm 0{\cdot}14$ for the plutonium + 2 oxygen atom set.

Reference

BACON, G. E. (1955). *Neutron Diffraction*. Oxford: University Press.