NONUNIFORM TEMPERATURE EFFECTS ON
THORIUM RESONANCE ABSORPTION

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by

W. G. Pettus
M. N. Baldwin

Critical Experiment Laboratory

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FOREWORD

On May 29 and June 18, 1958, the United States and the European Atomic Energy Community (EURATOM) signed an agreement to cooperate in programs for the advancement of the peaceful applications of atomic energy. This agreement, in part, provides for the establishment of the Joint U.S. -Euratom Research and Development Program to promote the construction of reactors in Europe.

This report represents the U.S. -Euratom effort to share scientific and technical information. Their contributions to the Program minimize the duplication of effort by the limited pool of technical talent available in Western Europe and the United States.

ABSTRACT

Described are the third-quarter activities of an experimental investigation of the effects of nonuniform temperature distributions on the resonance capture integrals of thorium metal and oxide rods. This work, which is jointly sponsored by the USAEC and Euratom, is being performed at the Nuclear Development Center of The Babcock & Wilcox Company. The activation technique exposes samples having different radial temperature gradients in a cadmium thimble at the center of the Lynchburg Pool Reactor. The counting samples are dissolved, and the separated Pa-233 is gamma-counted to obtain the relative integral neutron capture rates. The activations of the oxide series are completed. These include a set of 13 nonuniform temperature measurements having radial temperature drops up to 1000 C and a comparable set of 8 uniform temperature measurements from room temperature to 350 C. In the metal series, 8 activations of the nonuniform temperature sequence are completed. Further activations of the metal series are in progress, as are chemical processing, counting, and data analysis of both the oxide and metal samples.

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1. INTRODUCTION

The third-quarter progress of an experimental investigation of the effects of nonuniform temperature distributions on the resonance capture of neutrons in thorium metal and oxide rods is described. These experiments are being carried out by The Babcock & Wilcox Company as a part of the Joint U.S.-Euratom Research & Development Program.

The temperature dependence of resonance capture integrals arises from the spread of the velocities of the target nuclei in thermal motion. Since the thermal energy of the target nuclei at ordinary temperatures is much less than the neutron energy in the resonance region above 1 ev, the effect on the reaction rate is important only where either the absorption cross section or the neutron flux is rapidly varying in energy. These conditions occur in lumped absorbers having strong resonance peaks. In these cases the calculation of the integral reaction rate becomes a very formidable problem. Although no rigorous and general treatment of this problem has appeared, reasonably good agreement with experimental results has been obtained in special cases of uniform sample temperature and weak binding of the target nuclei¹. On the basis of a more idealized model, the inference has been drawn that, in practical cases, the absorption of a lump depends only on the average temperature and not on whether the temperature distribution is uniform.² Since all previous direct resonance integral measurements have been made at uniform temperature, this conjecture has not been tested, and one purpose of the present experiment is to provide information for this purpose. These experiments also constitute a test of the weak binding assumption for a case (ThO_2) in which that assumption is not theoretically justified³. A discussion of the way in which the crystal binding and the temperature gradients can conceivably produce a significant effect has been given in a previous report⁴.

In the present experiment the relative epicadmium neutron capture of axially heated thorium and thorium oxide rods is measured by activation techniques in the Lynchburg Pool Reactor. Since the axial heat source produces the steepest temperature gradient near the axis of the sample, where the resonance capture rate is low, the effect of the temperature gradient is not expected to be as large as that found in the ideal case of a uniformly heated sample, which gives the steepest gradient near the surface, where the capture rate is high. The choice of the axial heater was dictated by experimental feasibility considerations, but the test should be sufficiently sensitive to disclose any gross deficiencies of the idealized theoretical treatment.

Activation measurements have been made on thorium oxide samples having radial temperature drops up to approximately 1000 C. Comparable measurements on identical samples held at uniform temperature were also made. Similar measurements have been made on thorium metal samples having radial temperature drops up to 200 C, and further measurements are in progress. All of the thorium oxide counting samples have been dissolved, and nearly all have been processed to separate the Pa-233 and have been subsequently gamma-counted. Data analysis is progressing concurrently with the experimental work, but final conclusions cannot be drawn until all the samples of a given type have been counted.

2. EXPERIMENTAL EQUIPMENT

The major facilities and apparatus were described in a previous report⁴. In the course of the experiment, however, it was found that several important modifications were required. The first apparatus deficiency to occur was an excessive temperature rise (above the boiling point of the coolant water) in the sample cooling jacket due to an inadequate flow rate in the highest temperature thorium oxide measurements. This condition was remedied by doubling the cooling coil on the jacket. In a subsequent attempt to achieve a thorium oxide activation run with a 1000 C ΔT , a sharp temperature excursion of undetermined maximum was observed in the sample. Subsequent examination revealed that a section of the tantalum heater and its aluminum oxide support tube were disintegrated and that a small cavity was formed in the thorium oxide sample. It was surmised that a hot spot had formed on the heater due to corrosion caused by gettering of traces of moisture or oxygen from the helium atmosphere and that this hot spot had terminated in an arc sufficiently hot to cause a reaction between the components in its vicinity. To prevent recurrence of this condition, a liquid nitrogen cold trap and a hot tantalum getter were installed in the helium line to remove the moisture and oxygen as shown in Figure 1. In addition, the aluminum oxide tube was eliminated, and a larger heater wire was installed. With this arrangement, the high-temperature thorium oxide activation was completed without further difficulty.

As a precaution against interaction between the thorium metal samples and the insulator surrounding the heater, the aluminum oxide insulators were replaced by beryllium oxide. This material also gives a smaller temperature drop across its wall and therefore allows the heater to operate at a lower temperature for a given input power. No difficulty was experienced with this apparatus in any of the 8 thorium metal activations already completed.

Figure 1. Power Supply and Coolant Flow Diagram

3. EXPERIMENTAL PROCEDURES

3.1. Sample Activation

Just prior to activation a disc of the sample material is carefully loaded between the upper and lower halves of the sample, and the heater and thermocouples are installed. The system is then placed in the outer container (see Reference 4), and the electrical connections, the coolant connections, and the helium flow connections are made. The coolant line is then pressure tested for leaks, and the system is lowered through the test thimble to the center of the reactor core, where it rests on a rigid stop. Two gold monitor foils are then installed at accurately reproducible locations near the core. The heater circuit and the thermocouple response are then checked. If all instruments are responding properly, the cold trap is filled, the tantalum getter is heated, and the coolant pump and the helium supply are turned on. The system is then slowly brought to the desired operating temperature and stabilized. The reactor is then brought to a power of 500 watts, and the sample is given an exposure of 5 kilowatt-minutes. During the activation the sample temperature, the coolant temperature, the heater voltage drop, and the heater current are continuously monitored. After reactor shutdown the sample is cooled slowly to room temperature, and after waiting several hours for decay of the short-lived radioactivity the system is removed from the reactor, and the sample disc and the monitor foils are unloaded.

3.2. Chemical Processing

The thorium oxide samples are dissolved in about 200 ml of hot refluxing aqua regia containing a pinch of sodium fluosilicate (Na_2SiF_6). The samples require from one to two weeks for dissolving, and during this time the aqua regia is sometimes replenished. When dissolution is complete, the solution is evaporated to near dryness and cooled. The

resulting hydrated crystals are then dissolved in cold concentrated hydrochloric acid (14 N) to form the feed solution.

The feed solution is passed slowly through a resin column of Dowex 21 K resin, which has been initially conditioned with concentrated hydrochloric acid. Protactinium is strongly absorbed by the resin, but thorium and its daughters are only weakly absorbed and appear in the feed effluent. After all the feed solution has passed through the column, the flask used for dissolving is washed with concentrated hydrochloric acid several times, and this wash is passed through the resin column to remove any remaining thorium from the column.

The elutriant for the resin column is 1.5 N hydrochloric acid having about 15 drops of concentrated hydrofluoric acid added to each 100 ml. The elutriant is mixed in small batches in the glassware used to dissolve the oxide sample and is then passed through the resin column and is caught in 100 ml volumetric flasks.

3.3. Sample Counting

The separated sample solutions are gamma-counted on a 3-inch sodium-iodide crystal in conjunction with a 400-channel analyzer system. A lucite ring over the crystal is used to center the 100 ml flasks for counting. Several 20-minute counts are taken on each sample with the analyzer gain adjusted so that the 0.31 Mev peak of protactinium falls roughly in Channel 170. Background is subtracted automatically by the analyzer. A least-squares Gaussian fit of the analyzer data determines the exact channel position X_0 of the 0.31 Mev peak, and the relative activity of the sample is taken to be the total counts recorded between $0.9 X_0$ and $1.1 X_0$. At least one week is allowed between separation and counting to enable Pb-212 to decay away. This isotope, which is present in the decay chain of Th-232, has a half-life of 10.6 hours and has been observed in the sample solution immediately after separation.

4. METHODS OF ANALYSIS

4.1. Data Reduction

For each sample the directly measured quantity is the gamma count in each of a number of channels, which together span the resolved portion of the Pa-233 photo-peak at about 310 kev. The gamma spectrum of a typical sample is shown in Figure 2. The vertical lines constitute the bounds of the integration band, and the total count within this band is taken as a relative measure of the sample activation. The contribution of the portions of the boundary channels to the integration band is obtained by interpolation. This integral count is corrected for differences in reactor exposure, flask factor, and wait time, and finally the specific activity of each sample is obtained. The specific activities are then proportional to the total epicadmium neutron capture rates of the respective samples.

4.2. Deduction of the Resonance Integral Variation

Since it is more instructive to examine the variation of the resonance integral than that of the total epicadmium activation, it is desirable to separate out the "1/v" absorption below the first resonance and to correct for any departure of the neutron flux from the 1/E shape. This is done as explained in a previous report⁶, and the variation in the resonance integral is given by

$$I(T) = I(T_0) \left[(1 + \epsilon) \frac{A(T)}{A(T_0)} - \epsilon \right] \quad (1)$$

where $I(T)$ is the resonance integral at effective temperature T , $A(T)$ is the sample specific activity at effective temperature T , T_0 is the reference or room temperature, and ϵ is a number on the order of 0.1 arising from the flux shape and the "1/v" absorption corrections.

Figure 2. Protactinium Gamma Spectrum

4.3. Average Temperature Calculations

In attempting to understand the meaning and implications of the experimental results, it is desirable to compare the resonance absorption of a sample activated under nonuniform temperature conditions with that of a sample activated at a corresponding uniform temperature, which is called the effective temperature. If the conjecture of Reference 2 is correct, then the proper effective temperature which yields the same resonance integral is simply the average sample temperature or

$$\langle T \rangle = \frac{2}{r_2^2 - r_1^2} \int_{r_1}^{r_2} T(r) r dr \quad (2)$$

The evaluation of this integral has been carried out for both Th and ThO₂ with the effect of the temperature dependence of the thermal conductivity included, and these results have been given in closed form in a previous quarterly report⁵.

It is also interesting to consider a comparison based on a slightly different effective or equivalent temperature as follows. It is well established that the resonance integral at uniform temperature is linearly dependent on the square root of the absolute sample temperature. One may suspect then that a more realistic effective temperature could be obtained by averaging the square root of the absolute temperature over the volume of the sample and then squaring the result, or

$$\langle \sqrt{T_K} \rangle^2 = \left[\frac{2}{r_2^2 - r_1^2} \int_{r_1}^{r_2} \sqrt{T_K(r)} r dr \right]^2 \quad (3)$$

Using the radial temperature dependence previously obtained for the oxide case⁵, we find

$$\langle \sqrt{T_K} \rangle = \frac{2}{p(r_2^2 - r_1^2)} \int_{r_1^p}^{r_2^p} \sqrt{a + \beta u} u^{(4-3p)/2p} du \quad (4)$$

where

$$p = P_\ell / 2\pi A$$

$$d = (T_2 + \tau) r_2^p$$

$$\beta = 273 - \tau$$

T_2 = centigrade temperature at r_2

Equation 4 can be evaluated for values of p when $(4 - 3p)/2p$ equals an integer m , and interpolation will yield intermediate values. For a given m the result is

$$\langle \sqrt{T_K} \rangle = \frac{(-1)^m}{r_2^2 - r_1^2} \frac{(2^m \times m!)^2}{(2m + 1)!} \left(\frac{a}{\beta}\right)^{m+1} \sqrt{a}$$

$$\left[\left(1 + \frac{\beta u}{a}\right)^{3/2} \sum_{n=0}^m (-1)^n \frac{(2n+1)!}{(2^n \times n!)^2} \left(\frac{\beta u}{a}\right)^n \right]_{r_1^p}^{r_2^p} \quad (5)$$

In the present experiments this relation gives an effective temperature only 4% less than that given by Equation 2 for the most severe gradient condition, and the difference is much smaller in the other cases. It is concluded that a comparison based on a mean-root-square effective temperature is essentially equivalent to that based on a simple volume average of the temperature.

The question of what is the proper effective temperature for comparing samples under different temperature conditions is not completely resolved, and further consideration of this point will be deferred to a later report.

5. CURRENT STATUS

5.1. Thorium Oxide Measurements

As of the end of the third quarter the thorium oxide activations are considered complete and sufficient for the goals of the experiment provided no significant discrepancies appear in the analysis of the results. These activations include 8 uniform temperature measurements up to 300 C and 13 nonuniform temperature measurements with radial temperature drops up to about 1000 C. Also, all of the oxide samples have been dissolved and chemically processed to separate out the Pa-233, and most of the samples have been gamma-counted. Data reduction and analysis is in progress, but is deferred when necessary to complete the experimental work on other samples. Only preliminary results without final corrections are available for those samples which have been counted, and therefore no conclusions can be deduced at this time.

5.2. Thorium Metal Measurements

Eight thorium metal runs have been made in the nonuniform temperature series, giving radial temperature drops up to about 200 C. In these cases much higher input power is required to achieve a given temperature drop than in the corresponding oxide cases. This is due to the higher thermal conductivity of the thorium metal. The power requirement for the 200 C ΔT case is about 3.7 kilowatts, which must be developed in a 7-inch length of 30-mil wire. Under these conditions imperfections in the heater wire may lead to a hot-spot formation and subsequent heater burnout. Attempts will be made to achieve still higher radial temperature drops, but the uniform temperature measurements will be made first before the samples and apparatus are subjected to possible damage.

6. FUTURE PLANS

The current series of thorium metal activations will be continued until about the end of January 1965. Six activations at uniform temperature and at least two more activations at nonuniform temperature are planned. The uniform temperature activations will cover the range from room temperature to about 350 C, and in the nonuniform temperature measurements a maximum radial temperature drop of about 300 C with the average sample temperature at about 350 C will be attempted.

By the end of January the data analysis on the thorium oxide cases should be sufficiently advanced to indicate whether or not any repeat measurements are desirable. If repeat measurements are indicated, these will be made immediately, and the remainder of the contract period, through March 15, will be devoted to sample processing and counting along with data analysis and interpretation. It is expected that all work will be complete by the specified termination date.

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