NONUNIFORM TEMPERATURE EFFECTS ON
THORIUM RESONANCE ABSORPTION

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by

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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

An experiment is being conducted by The Babcock & Wilcox Company to determine the effect of nonuniform temperature distributions on the neutron resonance capture integral of thorium metal and oxide rods. This experiment is a part of the Joint U.S. -Euratom Research and Development Program. The second quarter has been devoted primarily to the development of suitable thermal chambers and heating and cooling systems. The designed thermal chambers allow the experiment to be conducted under an inert atmosphere, and a helium-cooled connector has been developed to prevent burnout of the section of the heater exposed by thermal expansion.

Fabrication of the thorium oxide pellet samples has been completed, and activation measurements on these samples are under way. Resonance capture measurements have been made on the axially heated samples having radial temperature drops up to 300 C. Corresponding measurements at uniform temperatures up to 350 C have also been made. Data from these measurements will not be available for several weeks because the counting samples must be dissolved and chemically separated. Additional thorium oxide measurements are planned to give radial temperature drops up to 1000 C. In the measurements on thorium metal, the maximum radial temperature drop will be determined experimentally.

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1. INTRODUCTION

Described are the activities of the second quarter of an experimental program to study the effect of nonuniform temperature distributions on the neutron resonance capture integrals of thorium metal and oxide rods. This investigation is being carried out at the Critical Experiment Laboratory of The Babcock & Wilcox Company as a part of the Joint U.S. -Euratom Research and Development Program.

Previous resonance integral investigations at uniform temperature conditions have been conducted at this laboratory under the same sponsorship; the results are reported in References 1 and 2, and are summarized in a recent comprehensive review by Chernick and Levine³. The previous work included measurements on both uranium and thorium and their oxides in the form of rods of various diameters. In these experiments the effective resonance integral was measured as a function of surface-to-mass ratio, sample temperature, and lattice spacing.

The present experiment is the first known attempt to measure the nuclear Doppler effect under nonuniform temperature conditions. For this reason, and because of the lack of a rigorous analytical treatment, these measurements must be regarded as partially exploratory. In these measurements the desired temperature gradients are produced by an axial heater wire. The resonance capture is measured by episcadmium activation techniques under various temperature gradient conditions, and the results will be compared with those of corresponding uniform temperature measurements on similar samples.

2. SYNOPSIS OF THE PROBLEM

In the presently accepted method of computing the Doppler coefficient of a crystalline absorber the absorber atoms are treated as an ideal gas and the absorption cross section is convoluted with a Maxwellian velocity distribution, with the introduction of certain approximations, to obtain an effective cross section in which the broadening effect is symmetrical about the line center and the line center does not shift as a function of temperature⁴. Lamb⁵ has given a criterion for the validity of this model which requires that

$$\Gamma + \Delta \gg 2 \Theta \quad (1)$$

where Γ is the natural line width, Δ is the Doppler width, and Θ is the Debye temperature in energy units. In some cases, e.g., ThO_2 , this criterion is flagrantly violated, and therefore some refinement of the model would seem to be required. It is found, however, that the Doppler coefficients calculated by the simple model are in reasonably good agreement with the previously measured Doppler coefficients, even for thorium oxide rods.

Although the foregoing experience seems to indicate that the simple analytical treatment is adequate for the purposes of reactor physics, there is a possibly significant effect which has been overlooked. This effect is the enhancement of the resonance capture in a nonuniformly heated crystalline sample caused by the progressive shift in the resonance energy along the temperature gradient. The enhancement of the absorption is a result of the reduction of the shielding of the inner regions of the sample by the outer regions at energies near strong resonance peaks. Since all previous measurements of the temperature dependence of resonance integrals of lumped absorbers have been made under uniform temperature conditions, this effect has not been encountered, and therefore the theory has not been put to a satisfactory test.

The purpose of the present experiments is to determine the magnitude of this effect by a direct comparison of resonance integral measurements under uniform versus nonuniform temperature conditions.

The magnitude of the expected effect is exceedingly difficult to estimate beforehand, but some indication of its potential significance can be obtained from the following considerations. When a neutron is captured by a free atom which is initially at rest, the conservation of energy and momentum requires that the compound nucleus recoil with an energy

$$R = \frac{m}{m + M} E \quad (2)$$

where m is the neutron mass, M is the mass of the capturing nucleus, and E is the incident neutron energy. The energy available for excitation of the compound nucleus above the binding energy is then $E - R$. However, if the target nucleus is infinitely tightly bound in a crystal, the entire crystal recoils, and the recoil energy is

$$R_c = \frac{m}{m + M_c} E \quad (3)$$

where M_c is the mass of the crystal. Since in any practical case R_c is effectively zero, the entire energy of the incident neutron is available for excitation of the compound nucleus. Between the weak-binding and the strong-binding limits there is therefore a maximum possible shift of $mE_r/(m + M)$ in the apparent resonance energy. Even for the lowest positive energy resonance of thorium, for instance, this amounts to a shift of more than twice the natural width of the resonance. The relative effect is much greater for the higher energy resonances since the shift is proportional to the energy and the capture width is about constant. If an experiment could be performed on a sample with a temperature gradient from absolute zero to the melting point, one would expect to measure an effective resonance integral several times larger than would be obtained at uniform temperature. A much smaller effect is expected in the present experiments since the temperature range is more restricted, but even with the presently achievable temperature gradients a significant change in the Doppler coefficient would not be surprising.

3. OBJECTIVE AND SCOPE OF WORK

The objective of the present experiment is, broadly stated, to obtain an experimental comparison of temperature dependence of the resonance capture of nonuniformly heated solid samples with that of identical samples uniformly heated. This comparison will test the validity of the ideal theoretical model of the Doppler effect in which certain crystal binding effects have been overlooked and, more directly, will test the validity of the conjecture⁶ that the resonance integral depends only on the average sample temperature and not on the temperature distribution.

Thorium oxide has been chosen as the material of primary interest for these measurements. This material was chosen because it has nuclear and solid state properties which render the conventional theoretical model highly questionable and because it has thermal properties which are favorable for the establishment of the desired temperature gradients. Furthermore, the uniform temperature Doppler effect of thorium oxide has been previously measured² at this laboratory. Comparative measurements on thorium metal samples are also planned as a test of a material for which crystal binding effects are expected to be of much less importance due to its relatively low Debye temperature.

In the thorium oxide case a series of epicadmium activation measurements will be made in which the 3/4-inch-diameter samples are heated by an axial resistance wire to give approximate radial temperature drops of 0, 50, 100, 200, 300, 600, and 1000 C. The average sample temperature in these cases will range from room temperature to about 300 C. Corresponding measurements will be made on identical samples with surface heating to obtain uniform temperatures of approximately 20, 100, 200, and 350 C.

A similar set of measurements is planned for a thorium metal sample of the same diameter. In this case the range of radial temperature drops which can be achieved will be determined by auxiliary

out-of-pile measurements. On the basis of preliminary measurements on a stainless-steel mock-up a maximum radial ΔT of about 300 C is indicated. The limitation is caused by the relatively high thermal conductivity, which necessitates a much higher input power density than is required for the corresponding oxide case, and by the large temperature drop across the insulation surrounding the heater wire in this case.

The resonance capture in each case will be determined from the induced activity of a representative section of the sample after a monitored exposure in the cadmium thimble of the Lynchburg Pool Reactor. The counting sample will be dissolved, and the Pa-233 from the decay of the Th-233 will be chemically separated and gamma-counted.

4. EXPERIMENTAL FACILITIES AND APPARATUS

4.1. Reactor Facility

The present measurements are being carried out in the cadmium thimble of the Lynchburg Pool Reactor. The core configuration is shown in Figure 1. Each fuel element consists of 10 plates of uranium-aluminum alloy with about 19 grams of U-235 in each plate, and each control element contains 6 similar plates. The central element position is occupied by a void test thimble, which extends above the pool surface. The lower end of the thimble, which penetrates through the core, consists of a 1-5/8-inch-OD cadmium tube having a 0.0226-inch wall sandwiched between two thin-walled aluminum tubes. An adjustable aluminum spacer is used at the bottom of the tube to center the samples vertically in the core.

4.2. Thermal Chambers

As shown in Figure 2 the containment for the axially heated thorium oxide samples consists of a close-fitting copper tube onto which is brazed a copper coil for coolant flow. A 60-mil-diameter Nichrome V heater wire is brazed into a copper plug, which screws into the base of the sample tube. The base end of the heater wire is grounded through the coolant supply lines. The top end of the heater is connected through a flexible braided conductor to a 1/8-inch copper tube, which passes through a Fiberglas sleeve to the hot side of the power transformer. Helium flow is also supplied through the 1/8-inch tube to cool the section of heater wire exposed by thermal expansion during operation. The sample-cooling jacket is held in an outer container of stainless steel. A photograph of this apparatus partially disassembled is given in Figure 3.

The containment used in the uniform temperature measurements is shown in Figure 4. Here the sample pellets are held in a copper jacket, which is wound with an asbestos-covered heater coil to provide

surface heating. The heater coil is overlaid with asbestos paper, and this assembly is surrounded by a water-cooled copper jacket to prevent radial heat flow to the cadmium sleeve or the reactor core. The outer containment is the same stainless-steel container used for the axially heated samples.

4.3. Sample Heating and Cooling System

A schematic drawing of the power supply is shown in Figure 5. The sample heater is fed directly by a four-section transformer, each section of which has a 115-volt primary and an 18-volt center-tapped secondary capable of continuous service at several hundred amperes. The primary is fed by a 6-kilowatt autotransformer. Forced cooling is provided by pumping water through the cooling jacket. Flow regulators of various capacities are used to adjust and stabilize the sample surface temperature. A water switch is used in the coolant line to trigger a relay to break the input power circuit in the event of loss of coolant.

4.4. Instrumentation

A Chromel-P Alumel thermocouple is imbedded at an accurately known position in the sample and a second thermocouple is brazed to the coolant jacket. The sample thermocouple is continuously monitored during operation by a Leeds and Northrup K-3 potentiometer with an electronic null detector. The jacket thermocouple is continuously monitored by a Rubicon portable potentiometer. The RMS voltage across the heater is measured by a Hewlett-Packard Model 403-A voltmeter, and the current through the heater is measured by a six-range Amprobe snap-around type ammeter reading from 0 to 600 amperes.

4.5. Chemical Processing and Counting Equipment

The process for dissolving the activated samples, separating out the Pa-233, and gamma-counting the separated sample is exactly the same as used in previous similar experiments at this laboratory. This process and the apparatus used are described in Reference 2.

Figure 1. Reactor Core Configuration

Figure 2. Thermal Chamber for the Axially Heated Samples

Figure 3. Thermal Chamber Partially Disassembled

Figure 4. Thermal Chamber for the Uniformly Heated Samples

Figure 5. Power Supply and Coolant Flow Diagram

5. EXPERIMENTAL SAMPLES

The thorium metal samples and the stainless-steel mock samples were machined at this laboratory and were described in the First Quarterly Report⁷. The oxide samples were fabricated by the Nuclear Materials and Equipment Corporation of Apollo, Pennsylvania. These samples consist of stacks of 10 pellets and a counting disc of 0.789-inch diameter. The total length of each sample is about 7-1/2 inches. The counting disc and the pellets below the counting disc have a single axial hole of about 1/16-inch diameter for the heater wire, and the pellets above the disc have an additional hole of about 0.050-inch diameter for a thermocouple. The density is about 85% of the theoretical value. A representative sample is shown in Figure 6 together with two individual pellets and a heating jacket used for the uniform temperature measurements. The chemical analysis supplied by the vendor is given in Table 1.

Table 1. Chemical Analysis of Thorium Oxide Samples

<u>Element</u>	<u>ppm</u>	<u>Element</u>	<u>ppm</u>	<u>Element</u>	<u>ppm</u>
Fe	220	Sn	<10	Mo	<10
B	<1	Cu	<10	V	<10
Co	<5	Pb	<10	Sb	<10
Cd	<1	Cr	95	Ca	<20
Mn	<5	Si	<50	--	--
Al	<20	Ti	<10	Th	87.88%
Mg	<10	Ni	150	--	--

Figure 6. Thorium Oxide Sample

6. SUMMARY OF EXPERIMENTAL WORK

Most of this quarter was devoted to several problems which arose in connection with the development of the thermal test chambers and to determining the limits of capability of the heating and cooling systems. The most severe problems were those related to the axial heater system for the thorium metal. The thermal conductivity of thorium (≈ 0.1 cal/cm-sec-C) is such that an input linear power density of about one watt per centimeter is required for each centigrade degree of radial temperature drop. This means that the present 6-kilowatt power supply is taxed to its limit to produce a modest 300 C radial temperature drop. This high power density requirement for the thorium metal also implies a rather large temperature drop across the electrical insulation separating the heater wire from the sample. At a power density of 300 watts/cm this ΔT is such that the heater must operate at a temperature above the melting point of the Nichrome V wire used in the initial tests. This problem was overcome by the use of tantalum wire, which added the complication that the thermal chamber had to be redesigned to permit operation under a helium atmosphere, since tantalum oxidizes very rapidly in air at these temperatures.

Another problem encountered at the higher power levels was the burnout of the heaters in the region between the connector and the sample exposed by thermal expansion. After several unsuccessful attempts to devise a sliding connector the problem was solved by the use of a special collet as a connector and by providing a helium flow through the collet to cool the exposed heater section.

Other problems related to the pyrophoric and chemical compatibility properties of thorium have been encountered. Since these problems could conceivably constitute a reactor operation hazard, they were reviewed with our internal Safety Review Board. Their recommendations, which included power interlocks, pressure relief devices, and inert atmosphere operation, have been adopted.

A number of preliminary tasks were performed before beginning the primary experimental operations. These included establishing a reactor core loading with suitable excess reactivity, installing the power system and cooling equipment, mapping the neutron flux through the thermal test chamber, and measuring the influence of the water in the cooling coils on the resonance flux.

The beginning of the primary activation measurements was delayed until late in the present reporting period because of the late delivery of the thorium oxide samples. The first measurements were made on September 15. Since then a complete set of measurements (room temperature to 350 C) has been made on the oxide samples at uniform temperature, and nonuniform temperature measurements have been made on the same samples with radial temperature drops up to 300 C. The data from these runs is not yet available, since several weeks are required for dissolving and chemically processing the counting samples. Similar activation runs are in progress, and no unusual operational difficulties have been encountered except thermal cracking of several of the thorium oxide pellets during the nonuniform temperature runs.

Fortran programs have been written to aid in the data analysis and to compute the effect of the finite sample length on the temperature distribution. An attempt is also being made to formulate a more rigorous model of the Doppler effect in the weak binding case.

7. FUTURE PLANS

The current series of thorium oxide measurements will continue through the first half of the next quarterly reporting period. This series will cover the entire range of radial temperature drops from 0 to 1000 C in addition to the uniform temperature measurements. It is anticipated that powder samples will have to be used at the higher temperatures because of thermal cracking of the pellet samples. The change in the form of the samples will cause no delay since the material is already on hand and the techniques have been developed in preliminary out-of-pile runs. When this series is completed, the available data will be analyzed, and the results will be used in determining the course of the remainder of the experiment.

If the crystalline binding effect under investigation is large and well-defined in the initial thorium oxide measurements, than a detailed set of corresponding measurements for thorium metal will begin. This series will probably run to the end of the quarter. If the effect in the initial thorium oxide measurements is not clearly significant, it may be desirable to repeat some of these measurements to improve the statistics before proceeding to the metal case. Since the effect is theoretically expected to be smaller for the metal than for the oxide, the oxide measurements will be given priority until the magnitude of the effect has been established.

In the absence of unforeseen difficulties it is expected that the experimental work will be essentially complete by the end of January 1965 and that all data reduction, analysis, and interpretation will be complete by the scheduled contract termination date of March 15, 1965.

REFERENCES

- ¹ Pettus, W. G., Baldwin, M. N., and Samuel, C., Resonance Absorption in U-238 Metal and Oxide Rods, The Babcock & Wilcox Company, BAW-1244, Lynchburg, Virginia, April 1962.
- ² Pettus, W. G., Baldwin, M. N., and Samuel, C., Resonance Absorption in Thorium Metal and Oxide Rods, The Babcock & Wilcox Company, BAW-1286, Lynchburg, Virginia, November 1963.
- ³ Chernick, J. and Levine, M. M., Developments in Resonance Absorption, Proceedings of the Third Geneva Conference, A/CONF. 28/P/262.
- ⁴ Dresner, L., Resonance Absorption in Nuclear Reactors, Pergamon Press, Inc., New York, 1960.
- ⁵ Lamb, W. E., "Capture of Neutrons by Atoms in a Crystal", Phys. Rev. 55, p 190 (1939).
- ⁶ Dresner, L., "Some Remarks on the Effect of a Nonuniform Temperature Distribution on the Temperature Dependence of Resonance Absorption", Nuclear Sci. and Eng. 11, p 39 (1961).
- ⁷ Pettus, W. G. and Baldwin, M. N., Non-Uniform Temperature Resonance Absorption Experiment, Quarterly Progress Report No. 1, The Babcock & Wilcox Company, BAW-3125-1, Lynchburg, Virginia, July 1964.
- ⁸ Baldwin, M. N., A Fortran Program for Calculating the Temperature Distribution Through a Set of Finite Axially Heated Cylindrical Shells, The Babcock & Wilcox Company, BAW-TM-401, Lynchburg, Virginia, August 1964.

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