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THORIUM RESONANCE INTEGRAL EXPERIMENT
JOINT U. S. -EURATOM RESEARCH AND
DEVELOPMENT PROGRAM
QUARTERLY TECHNICAL REPORT NUMBER 1
January-March 1963

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January-March 1963

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Submitted to
THE UNITED STATES ATOMIC ENERGY COMMISSION
By
THE BABCOCK & WILCOX COMPANY
Nuclear Development Center
Lynchburg, Virginia

FOREWORD

The United States and the European Atomic Energy Community (EURATOM), on May 29 and June 18, 1958, signed an agreement which provides a basis for cooperation in programs for the advancement of the peaceful applications of atomic energy. This agreement, in part, provides for the establishment of a Joint U. S. -Euratom research and development program which is aimed at reactors to be constructed in Europe under the Joint Program.

The work described in this report represents the Joint U. S. -Euratom effort which is in keeping with the spirit of cooperation in contributing to the common good by the sharing of scientific and technical information and minimizing the duplication of effort by the limited pool of technical talent available in Western Europe and the United States.

P. R. Augustine
Technical Advisor
U. S. Atomic Energy Commission

ACKNOWLEDGMENT

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ABSTRACT

The first-quarter activities are reported for the thorium resonance integral experiment, a Joint U. S. -Euratom research and development program authorized under AEC Contract AT(30-1)-3125. The objectives of the program are to measure (1) the effective neutron resonance-capture integral of thorium and ThO_2 as a function of rod diameter and temperature for isolated rods and (2) the mutual resonance shielding of parallel rods as a function of rod spacing. A technique is described for dissolving hard-fired ThO_2 pellets and separating out the Pa^{233} resulting from the decay of Th^{233} formed by the (n, γ) reaction in Th^{232} . The 310-Kev gamma from Pa^{233} is used as a measure of the integrated neutron capture. Details of the sample irradiation and counting procedures are also given, and plans for further experiments are outlined.

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1. INTRODUCTION

Described herein is the initial work done in the investigation of thorium resonance capture under the Joint U. S. -Euratom research and development program authorized by AEC Contract AT(30-1)-3125. This report covers the first calendar quarter of 1963 and the period from November 15 to December 31, 1962.

In over-all scope the program includes the following:

1. The measurement of the variation of the effective resonance integrals for Th and ThO₂ rods as a function of rod diameter in the range from 1/8 to 1-1/4 inches.
2. The measurement of the temperature dependence of the effective resonance integral for a ThO₂ rod of approximately 1/4-inch diameter in the temperature range from 20 to 800 C.
3. The measurement of the Dancoff Effect for ThO₂ and UO₂ rods of approximately 3/8-inch diameter separated by various intervals of D₂O moderator.
4. The auxiliary measurements and calculations necessary for correction and calibration factors plus exploratory measurements for the determination of the resonance integrals of thorium and uranium at infinite dilution.

Most of the present report period has been devoted to the development of chemical separation and counting procedures necessary for the activation measurements. Initial activation measurements have been made for all samples, and further measurements are in progress. The problem of spectrum measurement in the reactor test thimble has been considered.

2. REACTOR FACILITY

The experimental phases of this work are being carried out at the Lynchburg Pool Reactor (LPR). This reactor, shown in Figure 1, uses fuel elements of the MTR type; each fuel element consists of 10 plates of uranium-aluminum alloy with about 19 grams of U^{235} in each plate. Each control element contains six of the same plates. Figure 2 shows the core configuration used for the measurement of the geometric and temperature effects. The central fuel-element position is occupied by a cadmium-covered test thimble surrounded by a water gap. An extension on the test thimble rises above the pool surface to permit sample changing. The cadmium cover is sealed between two thin-walled aluminum tubes and extends about 6 inches above and below the fuel region. A plenum is provided for forced cooling in the water gap around the thimble during heated sample runs, and a thermocouple is fastened to the outside of the thimble for temperature monitoring.

Figure 1. Lynchburg Pool Reactor

Figure 2. Core Configuration

3. EXPERIMENTAL PROCEDURES

3.1. Sample Activation

The experimental effort during this report period has been confined to the investigation of the geometric dependence of the effective resonance integral by the activation method. In this measurement the sample rod of Th and ThO₂ is placed at the center of the reactor in the cadmium thimble and given an integrated exposure of about 10 kilowatt-minutes. The exposure is monitored by a precisely positioned gold foil located about 10 inches from the core. The holder assembly is shown in Figure 3 with a typical sample of stacked ThO₂ pellets. Pellet alignment is maintained by the close-fitting cladding tube. This tube is centered by a mechanical constraint in the outer holder, which is coupled by a short rod to a light chain for insertion and withdrawal of the assembly. All components of the holder assembly are made of aluminum except a small stainless-steel setscrew in the coupling rod.

The test rods are approximately 6 inches in length, and a small counting sample is located at the vertical center of each rod. The thorium-metal-rod diameters are 0.158, 0.397, and 1.155 inches, and the counting samples are thorium discs of approximately 56-mil thickness machined to precisely match the rod diameter. The thorium-oxide-rod diameters are 0.158, 0.266, and 0.936 inches. For the two smaller rods the center pellet of the stack is used as the counting sample. The largest oxide rod has pellets too large for convenient dissolving, and disc counting samples have been provided by cutting 100-mil slices from a pellet with a diamond saw.*

* - - - -
This work was done by the Geoscience Instrument Corporation, 110 Beekman Street, New York 38, N. Y.

3.2. Chemical Processing

In the present measurements the counting samples are of such varied sizes that direct counting is not feasible. The samples are therefore dissolved and counted in solution in volumetric flasks. In the process the protactinium that is counted is chemically separated from the thorium to minimize background correction. Since the development of a satisfactory technique for this task represents a major share of the work for this period, a description of the process is given in some detail.

The Th^{233} formed in the (n, γ) reaction in Th^{232} decays by the following scheme:

In this work a gamma line associated with the Pa^{233} decay is used as a measure of the neutron capture, and it is therefore necessary to wait several days after activation for the Th^{233} to decay before chemical separation is accomplished.

Dissolving thorium metal presents little difficulty, since it is vigorously attacked by hydrochloric acid, and the black residue produced will vanish upon the addition of a small amount of sodium fluosilicate. A small amount of white residue (probably ThO_2) sometimes remains but dissolves when nitric acid is added and the solution is boiled.

Dissolving of sintered thorium-oxide pellets is more difficult but has been accomplished successfully and completely by boiling for one to two weeks in aqua regia (3 parts concentrated hydrochloric acid and 1 part concentrated nitric acid) to which a pinch of sodium fluosilicate (Na_2SiF_6) has been added. The fluoride ion is necessary not only to help in dissolving the oxide but also to keep the protactinium in solution. Without the fluoride ion present the protactinium tends to adhere to the glassware. Care must be taken, however, to keep the amount of sodium fluosilicate to a minimum, since any excess will create an insoluble thorium-fluoride precipitate. For boiling pellets for long periods of time, as required in this investigation, it has been convenient to use the continuous refluxing apparatus shown in Figure 4. Occasional evaporation almost to dryness is desirable, however, so that fresh aqua regia can be added.

Thorium and its daughters can be separated from protactinium by utilizing the ion-exchange properties of Dowex 21 K resin. The feed solution is approximately 14 N HCl from which protactinium is strongly absorbed by the resin. Thorium and its daughters are only weakly absorbed and appear in the feed effluent. The elutriant is 1.5 N HCl with about 5 drops of concentrated HF added to each 100 ml.

The solution of dissolved Th or ThO₂ is brought to the proper normality in HCl by successively evaporating almost to dryness and rediluting with concentrated HCl. These successive operations of evaporation and dilution are necessary to eliminate most of the nitric acid, since thorium forms adsorbable nitrate complexes in a nitric acid solution. After the final dilution with concentrated HCl, the solution is not heated, since this will drive off HCl gas and thus reduce the acid concentration of the feed solution.

A column of resin about six inches in height contained in a 50 ml buret is used in the present work. This apparatus is shown in Figure 5. A glass-wool plug at the bottom of the column supports the resin beads. The resin column is initially conditioned with concentrated HCl for several hours, and then the feed solution is poured in. The flow rate through the column is regulated to about one drop per second. After all the feed solution has passed through the column, it is washed several times with concentrated HCl to remove any remaining thorium from the resin.

About 75 ml of elutriant has been found sufficient to remove the last trace of protactinium from the column. Thus, the protactinium can be eluted directly into a 100-ml volumetric flask for counting.

3.3: Sample Counting

The flasks containing the protactinium solutions are positioned on a 3-inch sodium-iodide crystal by means of a lucite centering ring as shown in Figure 6, and the gamma spectra is analyzed through the 400-channel analyzer system shown in Figure 7. The protactinium gamma spectrum is shown in Figure 8. There are strong photo peaks at 105 Kev and 310 Kev. The latter peak is used for quantitative measurement, and the counts in the central 11 channels of this peak are summed. This operation is facilitated by automatic digital printout of the counts stored in each channel of the analyzer. Prior to sample counting, the flasks

are calibrated with a standard protactinium solution, and the analyzer response is checked with a Cs^{137} standard.

During this period three independent activation measurements were made on each of the three thorium-metal samples. No difficulty was experienced in separation or counting, and the results are internally consistent with a standard deviation of 1% or less. Several activation measurements on the oxide samples have also been made, but there is not yet enough data for a definitive check on the internal consistency. It may be expected, however, that the oxide results will be somewhat less reproducible due to possible trace losses of protactinium by adherence to the glassware in the long dissolving process. Since no spectrum corrections or calibration measurements have been made, the numerical results cannot yet be interpreted to obtain the resonance integral. The results will therefore be given in a later report after the analysis is complete.

Figure 3. Sample Holder Assembly

Figure 4. Refluxing Apparatus

Figure 5. Resin Column

Figure 6. Sample Solution in Counting Position

Figure 7. 400-Channel Analyzer and Data-Readout System

Figure 8. Protactinium Gamma Spectrum

4. FUTURE PLANS

4.1. Geometric Dependence Measurements

Activation measurements on the oxide samples will be continued until sufficient data are obtained to give satisfactory statistical significance. These data will then be calibrated in terms of the infinite-dilution resonance integral of U^{238} and Th^{232} by means of auxiliary measurements on these isotopes in various dilute mixtures with pure scattering materials.

The measurements of the geometric dependence by the reactivity method will probably not be made until the activation measurements are completed. The plans for this work are not yet complete in all details but will be given in a later report.

4.2. Doppler Coefficient Measurements

The temperature coefficient of the effective resonance integral for a ThO_2 sample of approximately 1/4-inch diameter will be measured by activation. Measurements will be made at sample temperatures of approximately 200, 500, and 800 degrees centigrade.

4.3. Measurement of the Dancoff Effect

The measurement of the Dancoff Effect will probably be postponed until after all other measurements have been completed, since this requires a modification of the core and test thimble. Detailed plans will be given in a later report.

4.4. Auxiliary Studies

Some consideration has been given to the possibility of spectrum measurement at various points in the resonance-energy region by means of resonance-foil activation. The relevant parameters of various isotopes (Rh^{103} , Au^{197} , W^{186} , Co^{59} , Se^{78} , Na^{23}) with isolated neutron-capture resonances are being reviewed. Consideration has also been given to the

problem of gamma spectra analysis to determine absolute disintegration rates, from which absolute neutron-capture rates may be deduced... If these contemplated measurements are successful, the results will be of significant value in confirming or correcting the calculated neutron-flux spectrum presently used in the interpretation of the data.

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