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Joint U.S.-Euratom

NON-UNIFORM TEMPERATURE RESONANCE ABSORPTION EXPERIMENT

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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

As a part of the Joint U.S.-Euratom Research and Development Program, an experimental investigation on the effect of non-uniform temperature distributions on the neutron resonance capture integral of thorium metal and oxide rods is in progress at the Critical Experiment Laboratory of The Babcock & Wilcox Company. The activities of the first quarter have been devoted primarily to sample procurement, thermal calculations, and apparatus design and development. A thorium metal sample has already been fabricated, and ThO_2 pellet samples are being fabricated. These samples, approximately 3/4 inch in diameter by 6 inches long, contain holes for an axial heater and a thermocouple.

A cooling jacket and an inert atmosphere containment are being constructed to allow inpile operation having radial temperature drops up to 1000 C. Under these conditions, the resonance capture will be measured by activation or reactivity techniques and will be compared with the results of similar measurements at uniform temperature. The goals of this investigation are to test the assumption that the effective resonance integral is dependent only on the average sample temperature and to observe, if possible, the effect of shifting a resonance peak along a thermal gradient.

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1. INTRODUCTION

As a part of the Joint U.S.-Euratom Research and Development Program, the Critical Experiment Laboratory (CEL) of The Babcock & Wilcox Company (B&W) is conducting an experimental investigation on the effect of non-uniform sample temperatures on the neutron resonance capture integrals of thorium metal and oxide rods. The primary objective of this work is to test the conventional assumption that the effective resonance integral of a fuel rod depends only on the average temperature of the rod and not on the temperature distribution.

Because of the experimental difficulties involved, the preceding assumption has not been tested previously. Also, a rigorous theoretical treatment of resonance capture in a rod having an arbitrary temperature distribution is probably out of the question at this time, but the possible existence of a significant effect due to temperature gradients in some cases may be inferred from the following considerations.

In the capture process[1], a part of the energy of the incident neutron goes into the recoil of the compound nucleus and subsequently into the lattice vibrations in the case of bound target nuclei. This recoil has the effect of a shift in the apparent resonance energy; the magnitude of the shift depends on the strength of the binding and, in turn, on the local temperature. Therefore, in the case of an absorbing rod having a radial temperature gradient, a varying shift is expected in the resonance energy as a function of radial position. For cases where there are strong temperature gradients in materials having a high Debye temperature (strong binding), this progressive shift in the resonance peak may reduce the sample self-shielding to a measurable extent.

In the present experiment, ThO_2 is the material of primary interest. This choice was based on the relatively high Debye temperature (782 K)[2]; the low thermal conductivity, which is favorable for producing large temperature gradients; and the uniform temperature resonance capture properties, which were determined from previous work at the CEL[3]. The ThO_2 measurements will be followed by similar measurements on thorium metal rods, in which case the Debye temperature is sufficiently low so that Lamb's weak binding criterion[1] is satisfied. Unfortunately, the necessity for electrical insulation of the heater and the higher thermal conductivity in the metal case limit the maximum achievable radial temperature drop to a value approximately one tenth of that attainable in the oxide case.

For the purposes of this work, the exact nature of the temperature distribution is not of primary concern and, therefore, no attempt is made to duplicate the temperature profile obtained in any particular type of fuel element. For experimental convenience, the desired temperature gradients will be established by axial heating and surface cooling, giving an approximately logarithmic radial temperature dependence.

The measurements will be made in the Lynchburg Pool Reactor (LPR), which is equipped with a central thimble containing a cadmium filter for resonance capture measurements. Depending on the results of preliminary measurements, either activation or reactivity techniques will be employed.

The work of the present report period has been devoted to thermal calculations, apparatus design and development, and material procurement. These activities and the plans for future work are described in the following sections.

2. THERMAL CALCULATIONS

The theoretical temperature profile through the samples has been calculated in the infinite cylinder approximation with various assumptions for the temperature dependence of the thermal conductivity. Calculations have also been started on the finite cylinder temperature distribution, which will be of interest if a reactivity technique is adopted. Additional calculations have been made to estimate input power requirements and heat removal rates for various coolants.

2.1. Radial Temperature Distribution in an Infinite Cylinder

The basis for these calculations is reviewed in a separate topical report, BAW-TM-395[4]. The report shows that the radial temperature equation to be satisfied in the sample region when the thermal conductivity is a function of temperature is

$$\frac{d^2T}{dr^2} + \frac{1}{r}\frac{dT}{dr} + \frac{1}{K}\frac{dK}{dT}\left(\frac{dT}{dr}\right)^2 = 0, \qquad (1)$$

where K is the thermal conductivity. The solutions of Equation 1 that satisfy the necessary boundary conditions for several useful cases are presented below.

2.1.1. Case 1: K = Constant

$$T = T_2 + \frac{P_\ell}{2\pi K} \log \frac{r_2}{r} \qquad (2)$$

where P_ℓ is the input power per unit length. In this case, the volume weighted average temperature is given by Equation 3:

$$\langle T \rangle = T_2 + \frac{P_\ell}{2\pi K}\left[\frac{1}{2} - \frac{r_1^2}{r_2^2 - r_1^2} \log \frac{r_2}{r_1}\right] \qquad (3)$$

It is interesting to note that the average temperature obtains when

$$r = \frac{r_2}{\sqrt{e}}\left(\frac{r_2}{r_1}\right)^{\left(\frac{r_1^2}{r_2^2 - r_1^2}\right)}, \tag{4}$$

and that this value is independent of the boundary temperature and the power level. Therefore, a thermocouple located at this position would always read the average sample temperature.

2.1.2. Case 2: $K = A + BT$

$$T = \frac{A}{B}\left\{\left[1 + \frac{2B}{A^2}\left(AT_2 + \frac{BT_2^2}{2} + \frac{P_\ell}{2\pi}\log\frac{r_2}{r}\right)\right]^{1/2} - 1\right\} \tag{5}$$

Here, the average temperature is determined as follows:

$$\langle T\rangle = \frac{A}{B}\left\{\left[\frac{r_2^2\sqrt{\alpha-\beta\log r_2} - r_1^2\sqrt{\alpha-\beta\log r_1}}{r_2^2 - r_1^2} - \frac{e^{2\alpha/\beta}}{r_2^2 - r_1^2}\sqrt{\frac{\pi\beta}{8}}\right.\right.$$
$$\left.\left.\left[\operatorname{erf}\sqrt{\frac{2}{\beta}(\alpha-\beta\log r_2)} - \operatorname{erf}\sqrt{\frac{2}{\beta}(\alpha-\beta\log r_1)}\right] - 1\right\} \tag{6}$$

where

$$\alpha = \left(1 + \frac{B}{A}T_2\right)^2 + \frac{BP_\ell}{\pi A^2}\log r_2$$

$$\beta = \frac{BP_\ell}{\pi A^2}$$

Using the asymptotic expansion of the error-function[5]

$$\operatorname{erf}(x) \approx 1 - \frac{e^{-x^2}}{x\sqrt{\pi}}\left(1 - \frac{1}{2x^2} + \frac{3}{4x^4} - \frac{15}{8x^6} + \cdots\right), \tag{7}$$

the following equation is obtained:

$$\langle T\rangle \approx \frac{A}{B}\left\{\left[\frac{r_2^2\sqrt{\alpha-\beta\log r_2} - r_1^2\sqrt{\alpha-\beta\log r_1}}{r_2^2 - r_1^2} + \sqrt{\frac{\beta}{8}}\frac{1}{r_2^2 - r_1^2}\right.\right.$$
$$\left.\left.\left[\frac{r_2^2}{x_2}\left(1 - \frac{1}{2x_2^2}\right) - \frac{r_1^2}{x_1}\left(1 - \frac{1}{2x_1^2}\right)\right] - 1\right\} \tag{8}$$

where

$$x_1 = \sqrt{\frac{2}{\beta}(\alpha - \beta \log r_1)}$$

$$x_2 = \sqrt{\frac{2}{\beta}(\alpha - \beta \log r_2)}$$

The thermal conductivity of thorium metal is such that Equation 5 holds to a good approximation with A = 0.0865 cal/sec-cm-C and B = 3.28×10^{-5} cal/sec-cm-C^2. This relation is plotted in Figure 1 for r_1 = 0.08 cm, r_2 = 1.00 cm, T_2 = 50 C, and P_ℓ = 100 watts/cm = 23.87 cal/cm-sec. The average temperature in this case is found to be about 71 C, and the radius at which this temperature occurs is very nearly that given by Equation 4.

2.1.3. Case 3: $K = \frac{A}{T + \tau}$

$$T = (T_2 + \tau)\left(\frac{r_2}{r}\right)^{\frac{P_\ell}{2\pi A}} - \tau \tag{9}$$

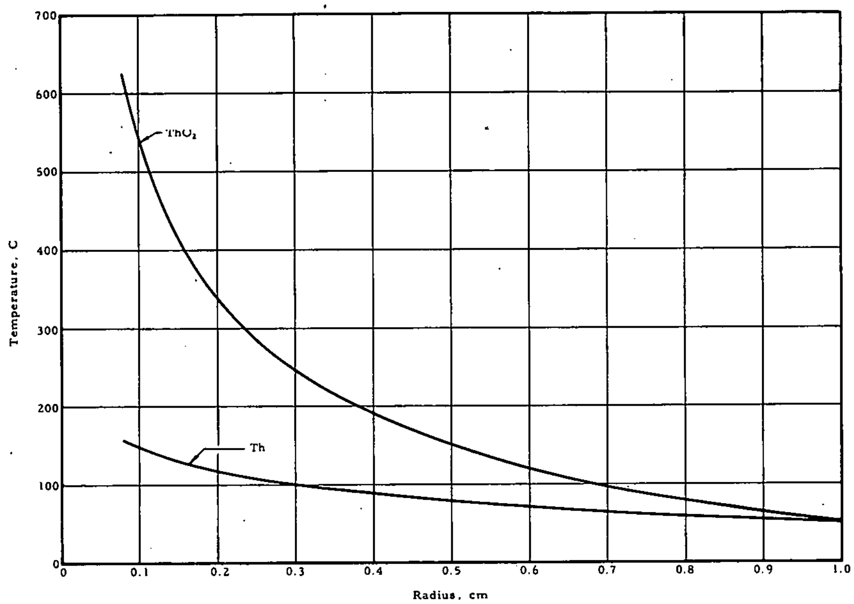
$$\langle T \rangle = \frac{2}{2 - \frac{P_\ell}{2\pi A}} \frac{T_2 + \tau}{r_2^2 - r_1^2}\left[r_2^2 - r_1^2\left(\frac{r_2}{r_1}\right)^{\frac{P_\ell}{2\pi A}}\right] - \tau \tag{10}$$

Equation 9 is a good approximation for ThO_2 where, at theoretical density, A = 8.71 cal/sec-cm and τ = 235 C. With the other parameters the same as in Case 2, we obtain the distribution shown in Figure 1, and we find that the average temperature is approximately 125 C. This temperature occurs where the following relation is satisfied:

$$r = r_2\left\{\frac{2}{2 - \frac{P_\ell}{2\pi A}} \frac{1}{r_2^2 - r_1^2}\left[r_2^2 - r_1^2\left(\frac{r_2}{r_1}\right)^{\frac{P_\ell}{2\pi A}}\right]\right\}^{-\frac{2\pi A}{P_\ell}} \tag{11}$$

Again, this value is within a few tenths of a millimeter of the position obtained from Equation 4.

Figure 1. Temperature Distributions in Thorium Metal and Oxide Rods (P_ℓ = 100 watts/cm)



2.2. Input Power Requirements

From Equation 5, the required input power per unit length for the thorium metal samples is given by

$$P_\ell = \frac{(A + BT_1)^2 - (A + BT_2)^2}{\frac{B}{\pi} \log \frac{r_2}{r_1}}, \qquad (12)$$

where the subscripts 1 and 2 refer to the inside and outside radii, respectively. Using the parameters of Case 2, we find that the input power for a 100 C radial temperature drop through the sample is 22.37 cal/cm-sec ≈ 93.7 watts/cm.

In the ThO_2 case, the input power requirement is given by

$$P_\ell = 2\pi A \frac{\log \frac{T_1 + \tau}{T_2 + \tau}}{\log \frac{r_2}{r_1}}$$

$$= 136 \text{ watts/cm} \qquad (13)$$

for a 1000 C radial temperature drop in a sample of theoretical density. By using powder samples, the input power requirement can be reduced to approximately 30 watts/cm.

2.3. Heat Removal Rates

The rate of heat removal per degree temperature rise is given by

$$R = 4.18 \rho c F \text{ watts/C}, \tag{14}$$

where ρ is the density (gm/cm^3), c is the specific heat capacity (cal/gm C), and F is the flow rate (cm^3/sec). For air, we have R = 0.13 watts/C when ρ = 0.0013 gm/cm^3, c = 0.24 cal/gm C, and F = 100 cm^3/sec. For water, we have R = 41.8 watts/C when F = 10 cm^3/sec.

3. APPARATUS DEVELOPMENT

The major item of experimental equipment to be perfected is a thermal environment chamber in which a desired radial temperature distribution in the sample can be established, maintained, and monitored. The greatest problem lies in obtaining an axial heater wire capable of sustained operation at temperatures above 1000 C or linear power densities greater than 100 watts/cm. A number of different heater materials have been considered and subjected to various tests to determine their suitability from the standpoint of high-temperature strength, corrosion resistance, thermal stability, and brazing characteristics. Some of the pertinent characteristics of these materials are listed in Table 1. Most of the parameters listed are temperature dependent, and the numerical values quoted should therefore be regarded as only rough order-of-magnitude approximations for semi-quantitative use.

Nichrome V and Kanthal A-1 have exceptionally low temperature coefficients of resistance and are less susceptible to hotspot formation than the other materials. Of these materials, Kanthal A-1 appears to have better high-temperature corrosion resistance but poorer ductility and high-temperature tensile strength. Platinum has excellent oxidation resistance but very little strength at high temperature. In air, stainless steel and nickel seem to corrode more rapidly than Nichrome and Kanthal. Tungsten, tantalum, molybdenum, and niobium must also be provided with protective atmospheres. The final selection of heater wire for the different cases to be investigated has not yet been made, but the initial tests indicate that either Nichrome V or Kanthal A-1 will be satisfactory for the ThO_2 powder samples and possibly for the ThO_2 pellet samples also. In the latter case, platinum or tungsten may be employed at the highest temperatures. In the thorium metal case, the heater must be electrically insulated from the sample. Because of the temperature drop across this insulator, the heater must operate at a much higher temperature than in the oxide case to maintain comparable temperatures at the inside boundary of the sample.

Table 1. Properties of Various Heater Materials

Material	Melting point, C	Resistivity $\times 10^{-6}$, ohm-cm	Temperature coefficient $\times 10^{-3}$	Thermal conductivity, watts/cm-C	Expansion coefficient $\times 10^{-6}$	Maximum operating temperature in air, C	Resonance integral, barns
Platinum	1773	10	4	0.70	9	1600	114
Kanthal A-1	1510	148	0.03	0.10	19	1350	4
Nichrome V	1400	108	0.1	0.11	17	1175	4
Nickel	1455	6.8	4	0.53	15	1000	4
Stainless Steel 304	1400	73	1	0.13	10	1000	3
Tungsten	3370	5.5	5	1.6	4	500	350
Tantalum	3027	15	4	0.55	7	500	750
Molybdenum	2620	5.7	4	1.5	5	500	32
Niobium	1950	21	4	0.52	10	500	10

Furthermore, the higher thermal conductivity of the metal samples requires an increased input power density for a given radial temperature drop in the sample. These conditions of high temperature and high power density create some problems in regard to the chemical compatibility of the heater and insulator materials with thorium. It is especially desirable to avoid hotspot formation on the heater wire, because the inherent positive feedback associated with this phenomena tends to cause the temperature of the hotspot to increase rapidly to burnout. At the melting point, the wire is suddenly reduced in diameter causing a large increase in the local resistance and hence a sudden large local increase in temperature. The process terminates with a very hot arc and very likely fracture or melting of the insulation around the wire. There is some concern about the possibility of triggering an exothermic reaction with the thorium in this hot region, causing the cadmium in the test thimble to melt or causing the thimble to rupture. As a precaution, the inpile experiments will be preceeded by an outpile run under operating conditions.

The cooling jacket must be capable of dissipating the input power developed in the heater wire. Equation 14 gives the required flow rates for a given input power and coolant temperature rise. If we limit the coolant temperature rise to 50 C, then we can only remove about 30 watts by air cooling at the maximum flow rate of 200 cm^3/sec. Since this is sufficient to give a temperature drop of only about 200 C in a loose powder sample, it appears that gas cooling is not practical. By water cooling at 10 cm^3/sec, a temperature drop in excess of 1000 C can be attained in both the oxide powder and pellet samples without exceeding the 50 C rise in coolant temperature. For the metal samples, the maximum flow rate of 10 cm^3/sec for the present apparatus design limits the radial temperature drop to approximately 400 C. A bench-top test rig for outpile heat transfer measurements and heater tests is shown in Figure 2; two newer models of the cooling jacket are shown in Figure 3.

Figure 2. Outpile Test Setup

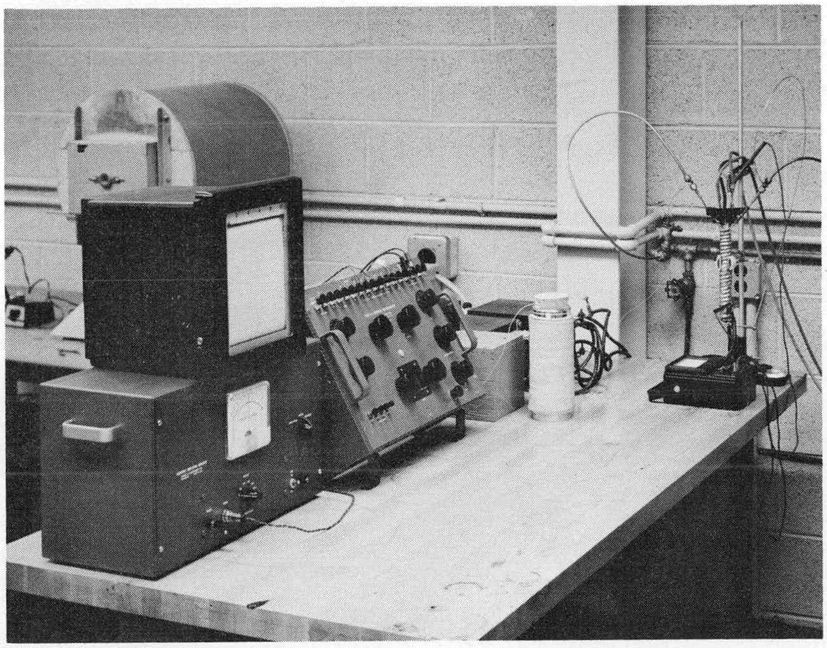
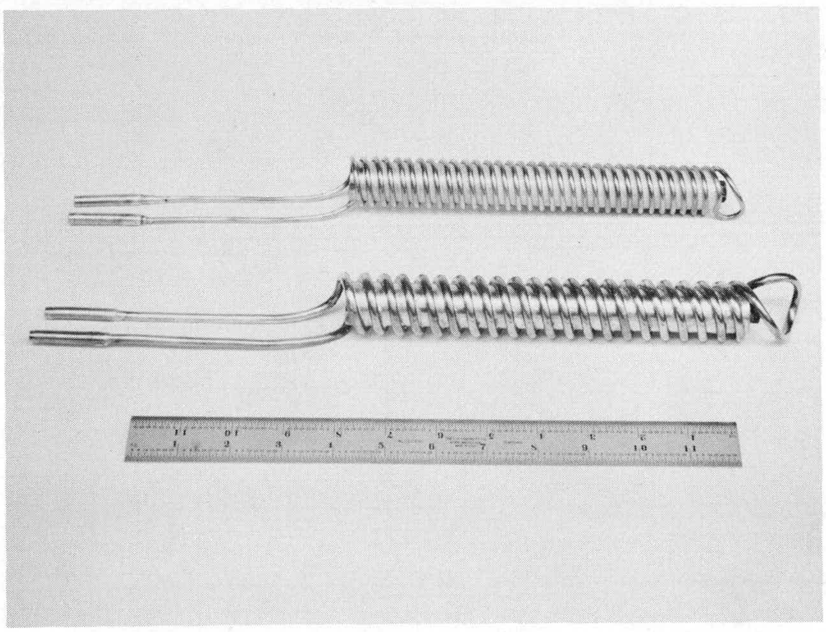


Figure 3. Sample Cooling Jackets



4. EXPERIMENTAL SAMPLES

The experimental samples are in the shape of right circular cylinders approximately 3/4 inch in diameter and 7 inches long including an end buffer zone. A thorium metal sample has been machined at the laboratory; the sample consists of a set of six matching cylinders, each approximately 3/4 inch long and having an axial hole 1/16 inch in diameter for the heater wire and insulator. Three of these pellets also have a 43-mil-diameter hole parallel to the axis and approximately 1/4 inch off center for placement of a thermocouple. As shown in Section 2, a thermocouple at this location measures very nearly the average sample temperature in all cases. A number of precisely matching discs about 1/16 inch thich have also been machined to serve as counting samples in the activation measurements. These samples are shown in Figures 4 and 5. A complete set of stainless-steel cylinders having dimensions identical to those of thorium have been machined for use in preliminary tests. Two of these cylinders will also be used as buffers at the ends of the thorium metal samples.

A set of fired ThO_2 pellets and discs having the same dimensions as the thorium samples has also been ordered from an independent supplier. These are to be ground to match and will have precisely located holes for the heater and thermocouple wires. ThO_2 powder samples are also being used in some of the preliminary tests and may be employed in the final measurements if cracking of the pellet samples becomes a serious problem.

Figure 4. Thorium Metal Sample and Activation Discs

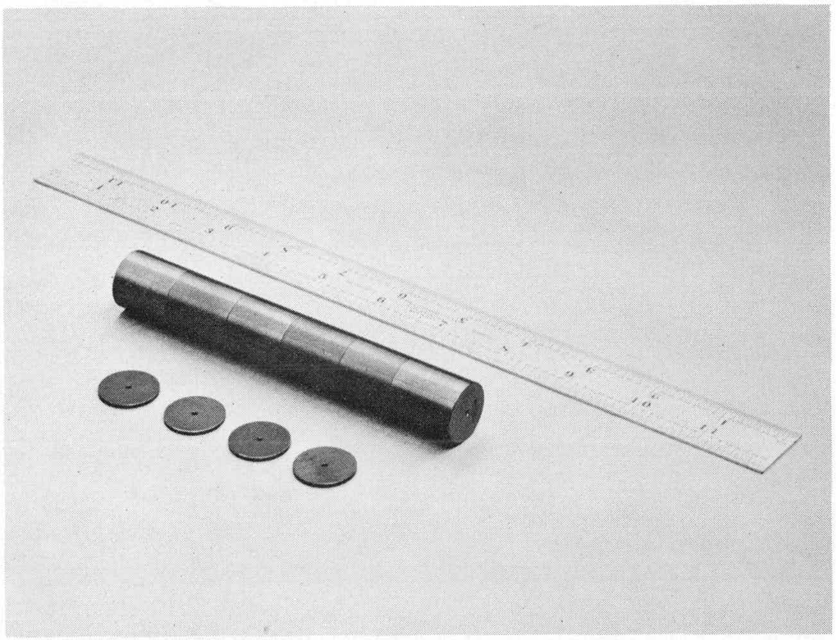
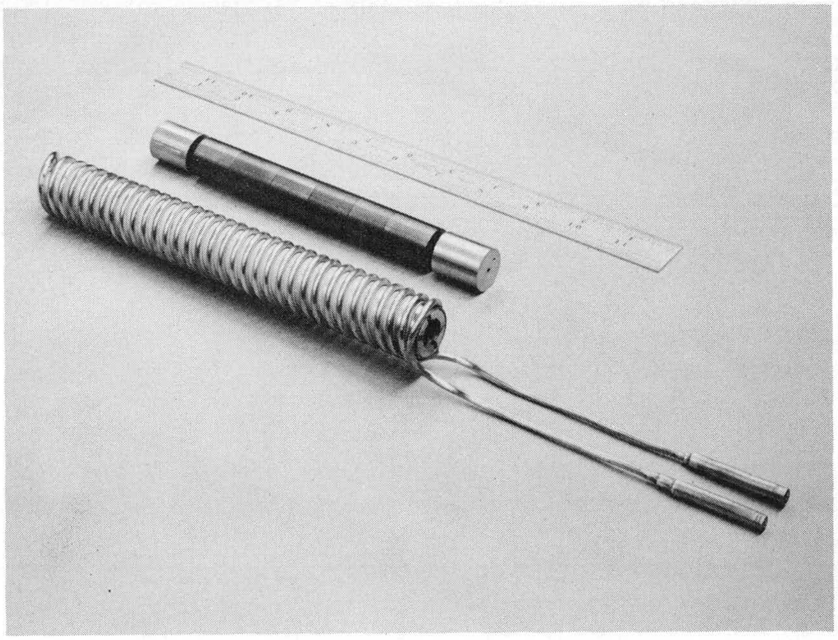


Figure 5. Thorium Metal Sample Including Buffer Pieces and Cooling Jacket



5. FUTURE PLANS

A number of problems related to the thermal test chamber are presently under an investigation that will continue through most of the next quarter. These problems include the following:

1. Selection of a heater material and electrical insulator compatible with thorium metal and useful for extended periods at temperatures above 1000 C.

2. Development of a fluid-cooled electrical connector capable of sustained operation at 200 amps while allowing free thermal expansion of the heater up to 0.2 inch.

3. Construction of a thermal test chamber containment to allow inpile operation in an inert atmosphere.

4. Installation and calibration of instrumentation for measurement of sample temperature, coolant temperature, and coolant flow rate.

The ThO_2 pellet samples are scheduled for delivery around July 1, 1964. These pellets will be sorted into matching sets and ground or lapped to assure accurate alignment. Outpile thermal tests will be conducted using both metal and oxide samples to determine operational limits and compatibility of materials.

Preliminary inpile tests should begin around August 15, 1964. Systematic data accumulation should begin near the end of the second quarter. The third and fourth quarters will be devoted primarily to data accumulation, data reduction, and analysis.

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