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RADIATION EFFECTS ON THERMOELECTRIC MATERIALS

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May, 1961

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by

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ABSTRACT

To evaluate the effect of nuclear irradiation on typical thermoelectric compounds, two types of measurements were made of their thermoelectric properties: (1) during irradiation, and (2) pre- post-irradiation. The tests are described and data are presented.

The irradiations were performed in two reactors; the Brookhaven Graphite Moderated Reactor and the Westinghouse Testing Reactor. Total dosages of $\sim 10^{19}$ nvt thermal and $\sim 10^{18}$ nvt fast neutrons (thermal 0.2 ev) were accumulated in the Brookhaven reactor. Samples irradiated in the Westinghouse Testing Reactor experienced fluxes of $\sim 1.5 \times 10^{20}$ nvt fast flux and a like amount of thermal flux.

For the materials $\text{Li}_x\text{Ni}_{1-x}\text{O}$, GeTe , Bi_2Te_3 , PbTe , and $\text{Bi}_{.05}\text{Ge}_{.95}\text{Te}$, it was found that when the thermoelectric material was irradiated at reactor ambient temperatures and slightly above ($50^\circ\text{-}100^\circ\text{C}$), the usual result was an increase in electrical resistivity and Seebeck coefficient. In most cases, this resulted in a decrease in the term α^2/ρ (power number), which would be reflected in a decrease in the thermoelectric figure of merit, assuming no decrease in the thermal conductivity. Raising the average temperature of the sample to $150^\circ\text{-}200^\circ\text{C}$ tended to anneal out the incurred damage for most of the materials tested.

The material which best appeared to resist irradiation damage was $\text{Bi}_{.05}\text{Ge}_{.95}\text{Te}$. However, the results indicated that most thermoelectric materials have some temperature where the irradiation damage annealing equals the damage rate, so that no net change occurs in that material's figure of merit.

INTRODUCTION

The efficiency of conversion of heat into electrical energy is limited by two factors: (1) the Carnot efficiency ($\Delta T/T_{\text{max}}$), and (2) energy losses in the conversion device. The first of these factors is a consequence of thermodynamics and is an absolute limit to the efficiency of any conversion scheme. Although this limitation cannot be evaded, its effect can be reduced by increasing the temperature of the heat source while holding the temperature of the heat sink constant. The second factor reflects the energy losses that inevitably afflict any practical device. These losses may be due to mechanical friction (in bearings, liquids, etc.), heat leaks (by conduction or stack losses), or electrical losses (Joule heating).

In the past century the major heat source for electrical power production has been the combustion of fossil fuel such as coal, gas, and oil. During this period the overall efficiency of generating plants has increased from a few percent to a plateau of approximately 40 per cent.

The advent of nuclear energy as a supplement to (and eventual substitute for) the world's dwindling supply of fossil fuels has stimulated a re-evaluation of energy conversion systems which can utilize the unique advantages of a reactor as a compact, high-density energy source capable of operating at high temperatures.

Because of recent advances in materials technology, one of the most promising direct conversion systems appears to be thermoelectricity. In this concept, thermoelectric materials are placed between the reactor fuel and the coolant. With the coolant at a low temperature, many of the problems of corrosion and pressure are bypassed. Further, under this concept the temperature of the heat source (and thus the Carnot efficiency) is limited only by the fuel and thermoelectric materials.

With present-day materials such as lead telluride (PbTe), a thermoelectric generator operating in the temperature range from 650°C to room temperature has a conversion efficiency of about 10 per cent. While this is low, there is an extensive program in progress to develop better thermoelectric material and to develop materials that can be operated at higher temperatures. The efficiency of a thermoelectric generator, unlike that of a conventional conversion system, is independent of plant size. This makes a thermoelectric conversion system more nearly competitive in the range of 1 KW to 5 MW.

The use of thermoelectric materials in the core of a reactor hinges upon finding materials that do not absorb an excessive number of neutrons and that are not damaged by neutron irradiation. One task under the Thermoelectric Nuclear Fuel Element Program has been to determine the effects of irradiation on a number of thermoelectric materials. This report summarizes the results of this phase of the effort and covers the period from March, 1959 to May, 1960. Since no information was available in the literature about the effects of irradiation on such materials, preliminary experiments were necessary to provide information about the gross effects of damage to be expected in typical thermoelectric materials and to roughly establish the temperature range in which appreciable annealing of radiation damage occurs.

The results indicate that a number of materials of interest can be operated in-pile without objectionable damage. Work now in progress will yield more precise information about radiation effects on PbTe and will more accurately define the limitations of its use in an in-core generator.

EXPERIMENTAL METHODS

Two types of irradiation experiments were performed on "representative" thermoelectric materials. These were termed (1) In-Pile Measurements, and (2) Post-Irradiation Measurements. The first type of experiment consisted of exposing thermoelectric materials to reactor irradiation and concurrently measuring the Seebeck coefficient (α) and electrical resistivity (ρ). The second type consisted of making measurements of α and ρ on pellets of thermoelectric materials, exposing the measured pellets to a reactor flux, and repeating the measurements after removing the irradiated pellets from the reactor.

Worth-while advantages accrue from both types of experiment; in the following discussion each will be considered separately.

A. In-Pile Instrumentation

1. Brookhaven In-Pile Experiments

The initial in-pile irradiation studies were performed in the Brookhaven Graphite-Moderated Test Reactor (BTR). This reactor is air cooled with an ambient temperature of approximately (170°F).

The neutron flux of the Brookhaven reactor at the location of interest (Hole E-53) is primarily thermal (<0.4 ev) with a value of $\sim 8.5 \times 10^{12}$ nv. Neutrons with energies greater than 0.4 ev were present at a level of $\sim 5 \times 10^{11}$ nv. The gamma flux was in the range of 60 milliwatts per gram.

The first thermoelectric material utilized for an in-pile experiment was $\text{Li}_{1-x}\text{Ni}_x\text{O}$. This required a neutral or oxidizing test atmosphere since NiO is an unstable oxide under even mildly reducing conditions. The first in-pile experiment was therefore not sealed to exclude the reactor coolant gas from the sample. For this first experiment samples of

$\text{Li}_{.013}\text{Ni}_{.097}\text{O}$ were placed in two aluminum cans and leads taken out through an aluminum pipe as shown in Figure 1.

The sample assembly can and internal arrangements are shown in Figure 2. The can is an aluminum pipe about 3-1/2 inches in diameter with aluminum flanges tie-bolted, welded, or soldered to the ends. The electrical leads from the heater, sample, etc., emerge through drilled holes in the bottom flange. The leads, individually insulated at the flange holes by aluminum oxide "spaghetti", extend in a bundle to the reactor face via the aluminum pipe as in Figure 1.

A sample was prepared by surface grinding the ends of a 1/2 inch diameter pellet to obtain parallel faces for bonding the pellet to thin Inconel disks. For $\text{Li}_x\text{Ni}_{1-x}\text{O}$ the braze material was silver. Each of the Inconel disks was drilled diametrically to permit insertion of a thermocouple. The thermocouple was connected to serve either as a probe for resistivity and Seebeck measurements or for measuring temperature.

A #1 headless screw threaded into a radial tap hole normal to the thermocouple was tightened against the couple to insure good thermal and electrical contact. The ends of the bonded sample assembly terminated in 1/4 inch diameter by one inch long rods, one of copper for the heater core and the other of Inconel for the cold sink.

The heater core was inserted into a platinum-wound heater. The cold sink terminated in the can, or, in later experiments, actually protruded through the flange into the reactor coolant flow. The 7052 glass was intended as a thermal conductivity standard. In later experiments at Brookhaven the glass was omitted because of difficulties in bonding the glass into sample assemblies. A later variation of sample assembly is shown in Figure 3. The essential difference here is the introduction

FIGURE 1 IN-PILE EXPERIMENT

FIGURE 2. IN-PILE EXPERIMENT 1, CAN #1

IN-PILE ASSEMBLY FOUR PROBE EXPERIMENT

FIGURE 3

of the four-probe method for measuring electrical resistivity, which is an improvement in that it enables elimination of the bond resistance from the resistivity measurements.

When measurements were made on materials other than $\text{Li}_{1-x}\text{Ni}_x\text{O}$, e.g. PbTe, it was necessary to fill the sample assembly can with argon and seal it by welding and soldering. In this case egress of the leads from the can was through Stupakoff seals. A quartz can, which was used for one experiment, involved such a multiplicity of glass-to-metal seals as to render the results unsatisfactory. The use of aluminum wherever possible in sample assemblies was requested by the Brookhaven staff in order to keep neutron activation problems to a minimum.

2. WTR In-Pile Experiments

The Westinghouse Testing Reactor (WTR) is light-water moderated and cooled. The coolant ambient temperature is about 100°F. Sample assemblies for this reactor were of necessity quite different from those used in BTR. The much greater neutron flux (fast $\sim 10^{14}$ nv, slow $\sim 10^{14}$ nv) as well as the much more intense gamma flux (5-10 watts/g) necessitated good sample cooling.

The experimental arrangement and sample assembly used in WTR are shown in Figures 4 and 5. Stainless steel outer tubes were used to provide protection and mechanical strength against coolant corrosion and turbulence. The swaged sample assembly was developed as part of the fuel element fabrication program of this contract. This type of assembly lends itself readily to the small spaces and rugged physical conditions which are encountered in the insertion. A further advantage of this geometry is that absolute thermal conductivity measurements can be made in-pile.

CAPSULE ARRANGEMENT

FIGURE 4

ED. SK. 288128 C

Figure 5

The swaged assemblies were made with stainless steel conductor and contact materials which, based on the information available, were thought to be compatible with the thermoelectric materials. More recent information shows that Armco iron contacts for PbTe and nickel-phosphorous coated iron contacts for GeTe are superior to stainless steel. However, considering the limited times and temperatures obtained in the experiments here reported, it is not likely that the stainless steel caused a serious reaction.

B. Post-Irradiation Capsule Design

All post-irradiation measurements were made on right cylinders of thermoelectric material approximately 1/2 inch in diameter and of various lengths from 1/2 to 1 inch. The pre-irradiation measurements were made under an appropriate atmosphere on the same apparatus used for post-irradiation measurements.

BTR capsules were merely aluminum cans of appropriate dimensions welded shut about the pellets. The pellets were in reasonably good thermal contact with the exterior surface of the capsule. Since the gamma and neutron flux was quite low, it was felt that the center-line temperature of most pellets did not greatly exceed the reactor ambient.

WTR post-irradiation capsules were somewhat different from those for the BTR. Since the samples in WTR experienced much greater gamma and neutron fluxes, good thermal contact with the capsule exterior had to be maintained to prevent excessive heating. Even if the heating did not cause melting or some deleterious reaction, it could anneal damage as it is incurred and prevent its detection.

The WTR capsule consisted of a thick-walled aluminum tube loaded with a series of aluminum-foil-wrapped thermoelectric pellets separated by Lavite insulating disks. The jacket tube was a 1 inch diameter by 12 inch long rod drilled axially 11-3/4 inches deep. The

1/2 inch diameter by 3/4 inch long wrapped pellets and the Lavite separating disks were forced in succession into the close-fitting jacket. After an aluminum cap was welded onto the loaded capsule, the sealed unit was leak checked.

After irradiation of the completed capsule, it was sectioned transversely through the Lavite spacers and the pellets were pressed out of the segments. Since this seamless tube construction has lead to severe sticking of some of the materials, later capsules have been built with a split tube liner to facilitate pellet removal after irradiation.

C. Thermoelectric Parameter Measurements

1. In-Pile Measurements

For the in-pile experiments, equipment was provided for both continuous recording and manual check reading of data to measure the thermoelectric parameters. For the manual check reading, a Leeds & Northrup K-3 potentiometer was used. A multi-point Minneapolis-Honeywell recorder supplied data as a function of reactor cycle time or irradiation flux. Data for the Seebeck voltage and for the temperatures at the ends of the sample were recorded continuously throughout the reactor cycle; resistance measurements were recorded at regular intervals when a timer-actuated switch pulsed a current from a six volt wet-cell battery through the sample.

The following data were recorded for a typical experiment:

- (1) The temperatures at the hot and cold ends of the sample pellet.
- (2) The Seebeck voltage across the pellet. This combined with the temperatures gives the Seebeck coefficient.

- (3) A second reading of the Seebeck voltage with a direct current flowing through the sample. The algebraic difference between (2) and (3) is the I^2R drop of the sample. This combined with I gives the resistance of the sample.
- (4) The voltage drop across a known resistance in series with the sample when current is flowing. This permits calculation of the value for I to be used in (3).

From the data thus obtained, a timed record of the Seebeck coefficient and the resistivity could be constructed for the entire experiment.

Regular records were also made of the power input to the sample heater and the ambient reactor coolant temperature. (The Brookhaven Reactor had the restriction that the maximum allowable surface temperature of a capsule should not exceed 200°C , which limited the temperature and power applied to the sample.) All thermocouple cold junctions were maintained at 0°C .

2. Post-Irradiation Measurements

Samples of thermoelectric material were prepared in the form of right cylinders approximately $1/2$ inch in diameter and $1/2$ to 1 inch long. All specimens were carefully graded by visual observation and density measurements in an attempt to reject those with detectable imperfections. Room temperature resistivity measurements were then made on the better samples with a K-3 potentiometer to determine if the material had normal thermoelectric properties for its particular composition. Representative samples could then be selected for pre-irradiation measurements over the appropriate temperature range.

The apparatus used to measure the thermoelectric properties for the pre- post-irradiation studies is shown in Figure 6. The cylinders which hold the current terminals and make contact with the sample are of Monel, as is the core for the upper heater. The three supporting legs, the radiation shield, and the disk to which the latter is attached, are stainless steel. The core for the lower heater is copper. The base plate and the upper disk assembly are brass. Electrical connections are made through the base plate by means of Stupakoff seals and Conax connectors. The cover for the apparatus is 3/8 inch thick aluminum.

The lower contact cylinder rests on a ball bearing, correcting for non-parallelism of the sample faces and insuring uniform contact. The thermocouples are set into wells in the contact cylinders and held in position by set screws.

To measure Seebeck coefficient, the EMF at the current terminals is read. To measure resistance, the EMF across the potential terminals is read. The potential terminals consist of small (0-80) screws held by metal rings cemented to a ceramic cylinder which surrounds the sample. All potentials are measured on a Leeds & Northrup K-3 potentiometer.

The chamber was evacuated and back-filled with helium or argon before the heaters were turned on. The only exception to this procedure was that $\text{Li}_x\text{Ni}_{1-x}\text{O}$ was run in air.

D. Neutron Flux Measurements

Approximate neutron flux measurements were available at the reactor involved as part of the reactor service. A flux measurement was made of Hole E-53 at Brookhaven for the first pre- post- experiment. Al-Co standards were irradiated and then the induced activity was read with a gamma ray spectrometer. Since these measurements confirmed values obtained by the Brookhaven Reactor Staff, no further flux measurements were made during the remainder of the program at Brookhaven.

FIGURE 6 - Measuring Apparatus

EXPERIMENTAL RESULTS

A. In-Pile

Table I lists the thermoelectric materials which were instrumented and irradiated as in-pile studies.

TABLE I
IN-PILE IRRADIATION EXPERIMENTS

<u>Exp.</u>	<u>Material</u>	<u>Reactor</u>	<u>Total nvt (t)</u>	<u>Remarks</u>
#1	Li _{0.013} Ni _{0.987} Li _{0.013} Ni _{0.987} Li _{0.035} Ni _{0.965} ⁰	BTR	1.7 x 10 ¹⁹	
#2	Li _{0.05} Ni _{0.95} ⁰ GeTe Bi ₂ Te ₃	BTR	1.3 x 10 ¹⁹	
#3	Li _{0.05} Ni _{0.95} ⁰ Li _{0.05} Ni _{0.95} ⁰	BTR		lead failed
#4	Li _{0.05} Ni _{0.95} ⁰ Li _{0.05} Ni _{0.95} ⁰	BTR	8 x 10 ¹⁸	
#5	N-type PbTe P-type Bi _{0.1} Ge _{0.9} Te	BTR	~ 5 x 10 ¹⁹	
#6	P-N PbTe	WTR	1.5 x 10 ²⁰ (fast)	heater failed

The data obtained from the experiments listed in Table I are presented in Figures 7 to 14. Data on Li_xNi_{1-x}⁰ are illustrated in Figures 7-11. In-pile data on Bi₂Te₃ and GeTe are illustrated in Figures 12-13. P-type PbTe (0.3 a/o Na) in-pile results are depicted by Figure 14.

Examination of the data obtained on Li_xNi_{1-x}⁰ compositions leads to the following conclusions:

FIGURE 7 RESISTANCE OF Li_2SiO_3 AS A FUNCTION OF TEMPERATURE FOR PRE-IRRADIATION AND IN-PILE EXPERIMENTS

FIGURE 8. RESISTANCE OF $\text{Li}_{0.913}\text{Ni}_{0.987}\text{O}$ AND $\text{Li}_{0.935}\text{Ni}_{0.985}\text{O}$ AS A FUNCTION OF INTEGRATED THERMAL NEUTRON FLUX

FIGURE 9 - SEEBECK COEFFICIENT OF $\text{Li}_{0.98}\text{Ni}_{0.98}\text{O}$ AS A FUNCTION OF INTEGRATED THERMAL NEUTRON FLUX AT 464°C

FIGURE 10 - ELECTRICAL RESISTIVITY OF $\text{Li}_{0.95}\text{Ni}_{0.05}\text{O}$ AS A FUNCTION OF INTEGRATED THERMAL NEUTRON FLUX AT 464°C

FIGURE 11

TE PROPERTIES OF $\text{Li}_{.06} \text{Ni}_{.94} \text{O}$ VS. THERMAL FLUX

FIGURE 11 ELECTRICAL RESISTIVITY OF Bi_2Te_3 AS
A FUNCTION OF INTEGRATED NEUTRON FLUX

FIGURE 13 ELECTRICAL RESISTIVITY OF GeTe AS
A FUNCTION OF INTEGRATED NEUTRON FLUX

FIGURE 14

TE PROPERTIES OF P-TYPE PbTe VS THERMAL FLUX

1. For these compositions radiation damage more readily affects the electrical resistivity than the Seebeck coefficient.
2. Measurements made by the four-probe method indicate that the electrical resistivity decreased with thermal neutron flux, while measurements made through bond areas indicated an increase in resistivity. The Seebeck values, however, displayed no significant differences with respect to the two modes of measurement.
3. When the in-pile results are interpreted in the light of pre-post- data on $\text{Li}_x\text{Ni}_{1-x}\text{O}$ (Figures 17-18, p. 31-32), it appears that 400°C represents the lower limit of a safe operating temperature for $\text{Li}_x\text{Ni}_{1-x}\text{O}$ in a reactor environment. Sufficient annealing takes place above 400°C to essentially maintain the thermoelectric properties of Seebeck and resistivity at their original values.

In-pile data on Bi_2Te_3 and GeTe were obtained before the four-probe technique was adopted. Thus, the resistivity curves shown in Figures 12 and 13 represent both bond resistance and material resistance, so that they do not necessarily indicate the true resistivity of the samples. However, the minimum temperatures for annealing irradiation damage in these materials have been estimated from pre-post-irradiation studies (at KAPL¹ and herein reported) to be in the vicinity of 200°C .

The in-pile data on P-type PbTe (0.03% Na) were obtained by the four probe method; this eliminated the spurious influence of bond resistance on the values from which the thermoelectric properties were derived. The resistance of the PbTe at an average temperature of 375°C increased almost linearly with neutron flux while the Seebeck coefficient remained virtually unchanged. However, other

¹ Knolls Atomic Power Laboratory Report No. 2150, "Changes in the Thermoelectric Properties of PbTe , Bi_2Te_3 , and ZnSb During Reactor Irradiation," J. Corelli, R. Frost, ² Bi_2Te_3 & M. Balicki, February 17, 1961.

experience with P-type PbTe suggests that the resistance increase was due to other than irradiation effects. Resistance measurements made on unencapsulated P-type PbTe showed a similar behavior which was attributed to loss of sodium doping agent. Also, pre- post-irradiation data on P- and N-type PbTe at KAPL and WAPD show an increased Seebeck coefficient rather than the observed static Seebeck coefficient when samples accrue irradiation damage. It does not seem unreasonable, then, to attribute the resistance increase to loss of sodium doping agent rather than to irradiation damage.

In-pile irradiation data must be obtained for higher levels of integrated neutron flux to verify the present tentative conclusion that thermoelectric materials operating at some minimum temperature just above the critical annealing temperature can withstand neutron fluxes of at least 10^{20} to 10^{21} fast nvt. Although each thermoelectric material has its own annealing temperature, a general annealing level can be named for the materials previously studied (Bi_2Te_3 , GeTe , $\text{Bi}_x\text{Ge}_{1-x}\text{Te}$, PbTe , and $\text{Li}_x\text{Ni}_{1-x}\text{O}$); it appears to be between 150 and 400°C. Thus, with the possible exception of Bi_2Te_3 , an in-pile application exploits most of the efficient range of working temperature of a given thermoelectric material.

B. Post-Irradiation Studies

The pre- post-irradiation experiments were performed in two reactors, the Brookhaven Graphite Moderated Test Reactor and the Westinghouse Testing Reactor (WTR). These are basically different reactors in the quality and intensity of their neutron spectrum energies. WTR has a fast neutron flux >0.3 ev, about equal to the thermal neutron flux of <0.3 ev. The Brookhaven reactor has a ratio of thermal-to-fast neutrons of about 10:1. Thus the Brookhaven reactor is basically a thermal neutron source while the WTR reactor is a fast neutron source.

Pre- post-irradiation data are presented in Figures 15 to 28. The flux levels are indicated on the figures. Qualitatively, the data on $\text{Bi}_{.05}\text{Ge}_{.95}\text{Te}$, GeTe , and N-type PbTe showed similar behavior after irradiation. That is, for each of these the Seebeck coefficient and resistivity were both higher after irradiation than before, and post-irradiation heat treatments at $200\text{-}500^{\circ}\text{C}$ partially restored their pre-irradiation values. Although for these particular samples no definite "annealing" temperature was identifiable, it appeared that at $200\text{-}300^{\circ}\text{C}$ average temperature almost complete annealing would take place. The significance of the annealing behavior observed is that a given one of these thermoelectric materials can probably be used in a reactor environment when the lowest operating temperature of the material is above its particular annealing temperature. The ideal case in this respect would be a material with a broad range of operating temperatures and with the annealing temperature below the bottom of the range.

A plot of α^2/ρ is also included on the figures showing the pre- post- data. The change in this term depends on the relative change between the resistivity and the Seebeck coefficient. Since in most cases both terms tend to increase, large changes in S^2/ρ were not observed, although N-type PbTe , ZnSb , $\text{CeS}_{1.37}$, and GeTe as represented in Figures 17, 18, 19, 20, 21, and 22 did show appreciable decreases. Annealing the irradiated samples tended to restore the pre-irradiation values for S^2/ρ , as would be expected.

Since the pre- post-irradiation experiments were originally designed to provide rapid information on gross effects of reactor irradiation, the type of data obtained did not yield sharply-defined annealing temperatures. This was primarily due to the rather large temperature gradient used to obtain the Seebeck voltages, which resulted in a poorly-defined temperature dependence of the sample properties.

PRE-POST IRRADIATION THERMOELECTRIC PROPERTIES AS A FUNCTION OF TEMPERATURE FOR $\text{Li}_{0.613}\text{Ni}_{0.987}\text{O}$ AFTER 1.4×10^{19} NVT THERMAL

FIGURE 15

ED. SK. 288028 C

PRE-POST IRRADIATION THERMOELECTRIC PROPERTIES AS A FUNCTION OF TEMPERATURE FOR $\text{Li}_{0.613}\text{Ni}_{0.987}\text{O}$ AFTER 1.39×10^{19} NVT THERMAL

FIGURE 16

ED. SK. 288028 C

PRE-POST IRRADIATION THERMOELECTRIC PROPERTIES AS A FUNCTION OF TEMPERATURE FOR N-TYPE PbTe AFTER 1.4×10^{19} NVT THERMAL

FIGURE 17

ED. SK. 288025 C

PRE-POST IRRADIATION THERMOELECTRIC PROPERTIES AS A FUNCTION OF TEMPERATURE FOR N-TYPE PbTe AFTER 1.5×10^{20} NVT THERMAL

FIGURE 18

ED. SK. 288025 C

PRE-POST IRRADIATION THERMOELECTRIC PROPERTIES AS A FUNCTION OF TEMPERATURE FOR N-TYPE Pb Te AFTER 1.5×10^{20} NVT THERMAL

FIGURE 19

ED. SK. 288024 C

PRE-POST IRRADIATION THERMOELECTRIC PROPERTIES AS A FUNCTION OF TEMPERATURE FOR Zn Sb AFTER 5.6×10^{18} NVT THERMAL

FIGURE 20

PRE-POST IRRADIATION THERMOELECTRIC PROPERTIES AS A FUNCTION OF TEMPERATURE FOR $\text{CeS}_{1.37}$ AFTER 5.0×10^{18} NVT THERMAL

FIGURE 21

ED. SK. 288022 C

PRE-POST IRRADIATION THERMOELECTRIC PROPERTIES AS A FUNCTION OF TEMPERATURE FOR GeTe AFTER 1.5×10^{-6} NVT THERMAL

FIGURE 22

ED. SK. 288027 C

PRE-POST IRRADIATION THERMOELECTRIC PROPERTIES AS A FUNCTION OF TEMPERATURE FOR Ge Te AFTER 4.5×10^{18} NVT THERMAL

FIGURE 23

ED. SK. 283022 C

PRE-POST IRRADIATION THERMOELECTRIC PROPERTIES AS A FUNCTION OF TEMPERATURE FOR $\text{Bi}_{0.05}\text{Ge}_{0.95}\text{Te}$ AFTER 9.43×10^{18} NVT THERMAL

FIGURE 24

ED. SK. 288023 C

PRE-POST IRRADIATION THERMOELECTRIC PROPERTIES AS A FUNCTION OF TEMPERATURE FOR $\text{Bi}_{0.05}\text{Ge}_{0.95}\text{Te}$ AFTER 2.75×10^{19} NVT THERMAL

FIGURE 25

ED. SK. 288024 C

PRE-POST IRRADIATION THERMOELECTRIC PROPERTIES AS A FUNCTION OF TEMPERATURE FOR $\text{Bi}_{0.05}\text{Ge}_{0.95}\text{Te}$ AFTER 1.5×10^{20} NVT THERMAL

FIGURE 26

ED. SK. 288027 C

PRE-POST IRRADIATION THERMOELECTRIC PROPERTIES AS A FUNCTION OF TEMPERATURE FOR $\text{Bi}_{0.05}\text{Ge}_{0.95}\text{Te}$ AFTER 9.4×10^{18} NVT THERMAL

FIGURE 27

ED. SK. 288023 C

PRE-POST IRRADIATION THERMOELECTRIC PROPERTIES AS A FUNCTION OF TEMPERATURE FOR Mn Te AFTER 4.5×10^{18} NVT THERMAL

FIGURE 28

ED. SK. 288026 C

Since the cooler portion of the sample was not exposed to a high temperature, annealing would not be expected to occur there as it did in the hotter end. In future work a modified apparatus will be available which will permit maintenance of very small-to no temperature differences across pre- post- samples.

CONCLUSIONS AND RECOMMENDATIONS

This phase of the study of irradiation damage has indicated that when a typical thermoelectric material is irradiated at a low temperature, its electrical resistivity and Seebeck coefficient are increased and its power number (α^2/ρ) is reduced. The figure of merit ($\alpha^2/\rho k$) may also be reduced, but not to as great an extent since the thermal conductivity k should be reduced due to both the disordering of the lattice and the trapping of carriers. As the temperature of the sample is raised, one finds that radiation damage previously incurred anneals out at a relatively low temperature. For each material there appears to be some temperature above which the material would not incur a net effect on the $\alpha^2/\rho k$ term. For in-pile use, the lowest temperature that a given thermoelectric material will see should be above the annealing temperature for that material.

The best thermoelectric material now available appears to be P and N-type PbTe. Its annealing temperature is about 150°C and the material has useful thermoelectric properties up to 700-750°C. Within this temperature range the radiation damage would be negligible and the Carnot efficiency would still be in the neighborhood of 50-60 per cent.

Although the number of irradiation experiments performed and their duration is not sufficient to fully guarantee long life operation in-core, the results are encouraging and justify a more definite investigation of the irradiation effects on the better thermoelectric materials.

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