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HIGH-TEMPERATURE THERMAL PROPERTIES
OF SNAP-10A FUEL MATERIAL

by

Sidney G. Nelson

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HIGH-TEMPERATURE THERMAL PROPERTIES OF SNAP-10A FUEL MATERIAL

Sidney G. Nelson

The enthalpy, specific heat, thermal diffusivity, thermal expansion, density, and thermal conductivity of as-received and of as-received and dehydrided SNAP-10A fuel material were measured or determined as a function of temperature. Also, hemispherical total-emittance data were obtained on dehydrided, dehydrided-oxidized, and dehydrided-nitrided fuel material as a function of temperature. Noteworthy in thermal-property measurements were the effects of homogenization of the dehydrided fuel on the thermal properties. Areas rich in uranium caused by segregation during the original hydriding of the fuel became less rich during exposure to elevated temperatures. Anisotropy in fuel rods was observed. Some significant differences between thermal conductivities determined in this study and thermal conductivities previously published were observed.

The solubilities of carbon, of nitrogen, and of oxygen in the molten (dehydrided) fuel alloys were determined up to about 2200 C. Data on carbon solubilities compared well with data previously reported on the carbon solubilities in unalloyed zirconium. The melting temperatures of fuel alloys containing 1 to 4 w/o nitrogen and of pure zirconium containing 4 w/o nitrogen were found to be as much as 300 C higher than those reported for alloys in the zirconium-nitrogen system. The melting points of alloys containing oxygen agreed with literature values for the zirconium-oxygen system except at 6 and 11 w/o oxygen, where the melting points of the fuel alloy were 100 C higher.

INTRODUCTION

The results of research studies conducted during the last several years have indicated the feasibility of constructing a mathematical model to evaluate realistically the effects of such variables as fuel-element design, reactor-system design, and orbit selection on the expected behavior of SNAP fuel elements during re-entry into earth's atmosphere. In the construction of a realistic mathematical model of the ablation process, it is necessary that the changes in physical properties of the fuel-element material with temperature be known. Recently, hyperthermal wind-tunnel experiments have been performed on fuel-element samples to observe the effects of various re-entry variables on the ablation process. Again, for the interpretation of data obtained in these tests, it is necessary that the effects of temperature on the physical properties of the fuel materials be known.

Various physical and mechanical properties of zirconium-uranium-hydride fuel materials have been evaluated in the past. (1-5)* The principal objective of the study described in this report has been to evaluate certain physical and chemical properties of a specific modified UZrH_x (SNAP-10A) fuel as a function of temperature.

Simulation of the precise conditions of re-entry burnup of nuclear fuels during property determinations is obviously very difficult, if not impossible, to achieve in the

*References are listed on page 114.

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laboratory. Perhaps the best approach to such determinations is to cover the required temperature ranges of interest, simulating as many of the conditions of re-entry as possible. Such property data then can be used, along with results from other tests such as hyperthermal wind-tunnel tests, to predict the behavior of the nuclear fuel material under more precise re-entry conditions.

GENERAL APPROACH

In this study, the thermal history of a re-entering fuel material was separated into three temperature intervals. In the first interval, approximately 0 to 650 C, it was assumed that little change in the composition and microstructure of the hydride fuel occurs during rapid heating. Thermal conductivity (diffusivity), enthalpy and specific-heat, and thermal-expansion measurements were made in this interval, while the hydrogen dissociation pressure of the material at temperature was balanced with a partial pressure of hydrogen. Measurements were conducted on the hydride fuel up to a temperature where the hydrogen dissociation pressure was 1 atm. Diffusivity and thermal-expansion measurements were also made between room temperature and 650 C in a dynamic vacuum; that is, by removing continuously all hydrogen leaving the specimens while heating specimens at a constant rate.

Notable changes in composition, principally loss of hydrogen, and in microstructure occur in the second temperature interval, from about 600 to 875 C. It is in this temperature range that difficulties in obtaining meaningful data on the properties of the fuel material are encountered. Both the microstructure and composition of the fuel material are dependent upon heating rate in addition to temperature. For this reason, it would be highly desirable if properties could be measured in this temperature interval utilizing heating rates comparable to those seen in re-entry. Unfortunately, equipment and techniques are not available to perform such measurements. If one considers only composition, however, and neglects microstructural changes, e.g., microcracking, phase separation, etc., an alternative approach is available which can yield valuable information. Because the composition of SNAP-10A fuel material has been determined for each given temperature when 1-atm back pressure is maintained, physical property measurements on specimens may be conducted within this temperature range with 1 atm back pressure and the respective compositions will be known. Employment of a dynamic vacuum will not yield as much useful data, since the compositions of the specimens before and after tests are the only ones generally determinable. Thermal-conductivity (diffusivity) and thermal-expansion-versus-time measurements were conducted from 650 C to well above 875 C on the hydride fuel in hydrogen at 1 atm with the specimens held at different constant temperatures spaced at discreet intervals.

At temperatures above 875 C, it was assumed that the fuel material would be completely dehydrided. Actually, it was found that 10 to 15 min was necessary at 875 C before the hydrogen composition of a full section of a fuel rod was down to 50 ppm, but it was also found that the length of time for essentially complete dehydriding decreased rapidly with increase in temperature above 875 C. Thermal-conductivity (diffusivity), enthalpy and specific-heat, and thermal-expansion measurements were made in vacuum on dehydrided fuel material from room temperature (from 150 C for thermal diffusivity)

to near the melting temperature of the material. Enthalpy and specific-heat measurements were also conducted on molten dehydrided material above the melting temperature.

ENTHALPY, SPECIFIC HEAT, HEAT OF FUSION, AND
HEAT OF PHASE CHANGE OF SNAP-10A FUEL MATERIAL

Experimental Procedures

A Bunsen ice calorimeter, similar to the one described by Ginnings and Corruccini,⁽⁶⁾ was utilized in the measurement of enthalpy, specific heat, heat of fusion, and heat of phase changes of SNAP-10A fuel material containing natural uranium. In an ice calorimeter, heat from the specimen melts ice that is in equilibrium with water in a closed system. The resulting volume change of the ice-water system is determined by weighing the mercury displaced as the system expands and contracts. The ratio of heat input to the mass of mercury replaced is a constant and is used as a calibration factor to obtain heat inputs to the calorimeter. There is no temperature change in the calorimeter since all heat transfer occurs at the ice point.

The particular Bunsen ice calorimeter used for measurements has been described by Deem and Lucks⁽⁷⁾. This apparatus is shown schematically in Figure 1. The calorimeter consists of a double-walled vessel with dry CO₂ gas between the walls. The outer vessel (A) contains a mercury reservoir chamber (B) which holds more mercury than is displaced during the freezing or melting of an ice mantle. The reservoir contains an adequate supply of mercury at the ice point to prevent the introduction of heat with reference to the ice-point temperature existing in the calorimeter. Encircling the outer vessel is a bath containing a mixture of ice and water. This bath maintains the entire calorimeter at the ice point. Mercury is introduced into the reservoir from the external mercury accounting system through 1/8-in. OD, 1/32-in. ID stainless steel tubing (C) passing down through the external ice and water baths. Similar tubing extends from the mercury reservoir to the calorimeter chamber.

The inner vessel (D) is the calorimeter chamber. This vessel is filled with outgassed distilled water and has about 1/4 in. of mercury at its bottom. The calorimeter is filled with water through a stainless steel tube (E) fitted with a special blunt-taper stainless steel needle valve. A low-wattage resistance heater wound around the inner vessel, is used to melt the outer part of the ice mantle should it touch the metal wall of the inner vessel.

An ice mantle is frozen on the copper fins with the aid of a copper tube containing crushed solid CO₂. The copper tube is placed inside the stainless steel dropping tube and is moved slowly downward to freeze an ice mantle evenly upon the copper fins. Earlier experience showed that mantles frozen in this manner in a transparent (Pyrex) calorimeter were very uniform and were not in contact with the inner vessel. Newly frozen mantles were often checked by dropping a specimen filled with Al₂O₃ and comparing the results obtained with reported literature values. The results obtained

FIGURE 1. STAINLESS STEEL BUNSEN ICE CALORIMETER

always compared very well with these values. Such successful tests, frequently repeated, have given confidence to this method.

A stainless steel specimen-dropping tube (F) extends down through the tops of the outer and inner vessels. This tube is machined to a 1/32-in.-thick wall and possesses two flanges by which it is welded to the tops of the two vessels.

Copper, because of its high thermal diffusivity, is used for the fins and tube around which the ice mantle is frozen. The fins and tube (G) are machined from one piece of copper bar and are tinned to prevent attack by water. It should be noted that the copper-fin assembly does not extend the full height of the calorimeter chamber. This procedure reduces the chance of the ice mantle freezing against the ends of the chamber. Purified argon enters the bottom of the dropping tube through tubing (H) and protects the specimen capsule (L) and the dropping wire from oxidation at high temperatures. A normal gas flow of approximately 30 bubbles per min through an oil bubble bottle is used during the measurements. The well and furnace are purged by increasing the flow of gas for at least 10 min.

A gate (I) is placed across the dropping tube to reduce heat transfer by radiation from the furnace. This gate is opened momentarily at a drop and then closed. A notch in the gate allows it to close with the dropping wire in place. Radiation losses from the capsule in the calorimeter in the direction of the gate are reduced by the use of a thin platinum radiation shield (M) attached to the supporting wire just above the capsule.

Figure 2 shows the calorimeter and furnace assembly. The specimens, encapsulated in Nichrome V alloy or tantalum, are heated to the desired temperature in a platinum resistance furnace (1). The temperature is regulated by controlling the voltage supplied and the specimens are dropped into a stainless steel ice calorimeter (2). A Pyrex jar (3) in which the calorimeter well is immersed contains a mixture of ice and water during the measurements. Mercury, which is moved during the melting or freezing of the ice mantle in the calorimeter well, is handled in the external accounting system (4) and is weighed on an analytical balance (not shown in photograph).

The atmosphere furnace shown in Figure 2 is used to 700 C. Above this temperature, a vacuum furnace is employed to prevent oxidation of the tantalum capsules which are used at high temperatures. Figure 3 is a sketch showing the vacuum furnace (1) in place over the calorimeter well (10). The capsule (6) is hung on a fusible link (7) and drops in free fall into the calorimeter well. A resilient pad of metal wool in the calorimeter tube cushions the impact. The capsule remains in the calorimeter tube in a vacuum until its temperature becomes so low that heat transfer by radiation is no longer effective. Gas at 0 C is introduced into the calorimeter well from the gas storage chamber (12) to aid in heat transfer. The vacuum gate (9) is closed after the drop to maintain a vacuum in the furnace. The capsule temperature is measured by an optical pyrometer through a sight port (5). A double-walled tantalum tube heater (2) is used to heat the capsule.

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FIGURE 2. CALORIMETER AND FURNACE ASSEMBLY

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FIGURE 3. HIGH-TEMPERATURE VACUUM FURNACE AND
BUNSEN ICE CALORIMETER

Experimental Results

The enthalpy of dehydrated SNAP-10A fuel material was measured in the Bunsen ice calorimeter. Specimens were encapsulated in tantalum capsules which contained ATJ-grade graphite liners. The liners were utilized to prevent reaction between the fuel and the tantalum capsules. Specimens evaluated in the molten state were not held in the graphite liners for longer than 3 min. A separate specimen was used for each data point in the liquid range, and a total of 45 specimen drops were made from 0 to 2157 C, encompassing a total of 10 different specimens.

The enthalpy data were fitted with equations in which the constants were obtained by least squares. Table 1 shows the observed enthalpy data, enthalpy data calculated using the equations, and the difference between the observed and equation data. The first derivatives of the enthalpy equations were used to calculate the instantaneous specific heats at interpolated temperatures. Table 2 shows the instantaneous specific heat values for the dehydrated material.

Temperatures used to calculate heats of transformation and of fusion were as follows:

595 ± 5 C, alpha + delta ⇌ alpha + beta transformation

840 ± 10 C, alpha + beta ⇌ beta transformation

1810 ± 25 C, solidus

1870 ± 25 C, liquidus

The enthalpy, H_T , and specific heat, C_P , equations with constants used to fit the enthalpy versus temperature data were:

$$H_T (\text{Solid}) = 0.066882 T + 10.606 \times 10^{-6} T^2 \quad (1)$$

(0-595 C)

$$C_P (\text{Solid}) = 0.066882 + 21.212 \times 10^{-6} T \quad (2)$$

(0-595 C)

$$H_T (\text{Solid}) = 28.447 - 0.017319 T + 81.239 \times 10^{-6} T^2 \quad (3)$$

(595-830 C)

$$C_P (\text{Solid}) = -0.017319 + 162.48 \times 10^{-6} T \quad (4)$$

(595-830 C)

$$H_T (\text{Solid}) = -123.46 + 0.39316 T - 247.583 \times 10^{-6} T^2 + 0.065349 \times 10^{-6} T^3 \quad (5)$$

(830-1810 C)

TABLE 1. ENTHALPY VALUES FOR DEHYDRIDED MATERIALS

Temperature, C	Enthalpy, H_o^T , cal per g		Equation	Specimen	Difference
	Observed	Equation			
0	0	0	1	--	--
134	8.8 ₁	9.15	1	9A	-0.34
137	9.0 ₃	9.36	1	9A	-0.33
302	21.9 ₁	21.17	1	9A	+0.74
303	21.0 ₀	21.24	1	9A	-0.24
473	33.9 ₆	34.01	1	9A	-0.05
474	34.0 ₆	34.08	1	9A	-0.02
588	43.0 ₀	42.99	1	9A	+0.01
589	43.0 ₃	43.07	1	9A	-0.04
595	--	43.69	1	--	--
595	--	46.83	3	--	--
632	49.6 ₈	49.95	3	9A	-0.27
636	50.1 ₀	50.29	3	9A	-0.19
691	55.6 ₉	55.27	3	9A	+0.42
693	55.9 ₂	55.46	3	9A	+0.46
733	60.1 ₁	59.40	3	9A	+0.71
735	59.8 ₂	59.60	3	9A	+0.22
775	61.4 ₃	63.82	3	9B	-2.39
806	67.5 ₅	67.26	3	9B	+0.29
821	69.7 ₅	68.99	3	9B	+0.76
830	--	70.04	3	--	--
830	--	69.67	5	--	--
866	74.7 ₉	73.78	5	9B	+1.01
905	76.9 ₅	78.01	5	9B	-1.06
943	81.3 ₉	81.93	5	9B	-0.54
963	85.6 ₅	83.91	5	9D	+1.74
994	85.9 ₆	86.90	5	9B	-0.96
1006	88.7 ₃	88.03	5	9D	+0.70
1028	88.7 ₈	90.06	5	9F	-1.28

TABLE 1. (Continued)

Temperature, C	Enthalpy, H_O^T , cal per g		Equation	Specimen	Difference
	Observed	Equation			
1049	92.2 ₃	91.96	5	9B	+0.27
1056	93.4 ₈	92.58	5	9D	+0.90
1083	93.9 ₂	94.95	5	9B	-1.03
1118	97.9 ₀	97.95	5	9B	-0.05
1181	101.7	103.2	5	9E	-1.5
1221	106.9	106.4	5	9B	+0.5
1300	112.5	112.8	5	9D	-0.3
1366	119.8	118.2	5	9B	+1.6
1464	128.1	126.5	5	9F	+1.6
1492	130.1	129.0	5	9B	+1.1
1581	137.2	137.5	5	9E	-0.3
1635	141.1	143.1	5	9B	-2.0
1650	144.1	144.8	5	9C	-0.7
1700	149.7	150.5	5	9E	-0.8
1764	158.5	156.4	5	9D	+1.9
1810	--	164.6	5	--	--
1818	166.9	--	--	9F	--
1835	--	168.2	5	--	--
1835	--	193.3	7	--	--
1858	186.5	--	--	9J	--
1860	--	201.3	7	--	--
1941	214.4	214.4	7	9H	0.0
1990	221.3	222.4	7	9C	-1.1
2061	238.3	233.9	7	9G	+4.4
2157	249.3	249.4	7	9K	-0.1

TABLE 2. SPECIFIC HEAT VALUES FOR DEHYDRIDED MATERIALS

Temperature, C	Specific Heat, C_p , cal/(g)(C)	Equation
0	0.0669	2
100	0.0690	2
200	0.0711	2
300	0.0732	2
400	0.0753	2
500	0.0774	2
595	0.0794	2
595	0.0791	4
650	0.0880	4
700	0.0960	4
750	0.1042	4
800	0.1123	4
830	0.1175	4
830	0.1172	6
900	0.1097	6
1000	0.0982	6
1100	0.0907	6
1200	0.0872	6
1300	0.0877	6
1400	0.0922	6
1500	0.1007	6
1600	0.1132	6
1700	0.1297	6
1800	0.1502	6
1810	0.152	6
1835	0.158	6
1835	0.162	8
1860	0.162	8
1900	0.162	8
2000	0.162	8
2100	0.162	8
2157	0.162	8

$$C_P (\text{Solid}) = 0.39316 - 495.166 \times 10^{-6} T + 0.19605 \times 10^{-6} T^2 \quad (6)$$

(830-1810 C)

$$H_T (\text{Liquid}) = -100 + 0.162 T \quad (7)$$

(1870-2150 C)

$$C_P (\text{Liquid}) = 0.162 \quad (8)$$

(1860-2150 C)

The heat of phase change at 595 C (alpha + delta $\rightleftharpoons$ alpha + beta transformation) was 3.1 cal per g. It was calculated by taking the difference between the solution of Equation (2) and the solution of Equation (1) at 595 C. There was no heat of phase change at 840 C (alpha + beta $\rightleftharpoons$ beta transformation). The phase change showed up as a change in slope in the enthalpy curve and the data indicated that this change was nearer 830 C. The slight difference in calculated enthalpies at 830 C was thought to be more a consequence of curve fitting than a real difference. The heat of fusion, calculated at 1835 C using the difference between Equation (7) and Equation (6), was 29.1 cal/g.

The enthalpy of hydrided SNAP-10A fuel material was also measured in the Bunsen ice calorimeter. A single specimen, encapsulated in Nichrome V, was evaluated to a temperature of 660 C, where the partial pressure of hydrogen over the specimen was 1 atm absolute. A total of 14 drops were made on the hydrided fuel between 0 and 666 C. Enthalpy and specific heat equations were calculated from the observed data in the same manner as that for the dehydrided material. The enthalpy, H_T , and the specific heat, C_P , for the hydrided material were found to be represented by:

$$H_T (\text{Solid}) = 0.078094 T + 86.818 \times 10^{-6} T^2 \quad (9)$$

(0-670 C)

$$C_P = 0.078094 + 173.63 \times 10^{-6} T^3 \quad (10)$$

(0-670 C)

Table 3 shows the observed enthalpy values, enthalpy data calculated using the equation, and the difference between the observed and equation data. Table 4 shows the instantaneous specific heat values for the hydrided material. Figures 4 and 5 show the enthalpy and specific heat curves, respectively, for the dehydrided and hydrided materials.

The hydride specimen was encapsulated in a closed container having a free volume of approximately 2 cm³. Recombination of the free hydrogen surrounding the specimen upon cooling rapidly from a given temperature causes an exothermic reaction. Calculations of the probable heat of solution at 660 C, where the specimen is in equilibrium with hydrogen at 1 atm, show a reduction in enthalpy of less than 0.18 per cent, and therefore, an insignificant amount.

TABLE 3. ENTHALPY VALUES FOR HYDRIDED MATERIALS

Temperature, C	Enthalpy, $H_{O,T}^T$, cal per g		Equation	Specimen	Difference
	Observed	Equation			
0	0	0			
79	6.57	6.71	9	7B	-0.14
81	6.80	6.90	9	7B	-0.10
150	13.33	13.67	9	7B	-0.34
156	13.88	14.30	9	7B	-0.42
236	23.36	23.27	9	7B	+0.09
298	30.92	30.98	9	7B	-0.06
301	31.40	31.37	9	7B	+0.03
372	41.37	41.07	9	7B	+0.30
373	41.49	41.21	9	7B	+0.28
455	53.57	53.51	9	7B	+0.06
543	67.93	68.00	9	7B	-0.07
598	77.83	77.75	9	7B	+0.08
666	90.47	90.52	9	7B	-0.05
666	90.52	90.52	9	7B	0.00

TABLE 4. SPECIFIC HEAT VALUES FOR HYDRIDED MATERIAL

Temperature, C	Specific Heat, C_p , cal/(g)(C)	Equation
0	0.0781	10
100	0.0955	10
200	0.1128	10
300	0.1302	10
400	0.1475	10
500	0.1649	10
600	0.1823	10
666	0.1937	10

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FIGURE 4. ENTHALPY OF HYDRIDED AND DEHYDRIDED MATERIALS

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FIGURE 5. SPECIFIC HEAT OF HYDRIDED AND DEHYDRIDED MATERIALS

THERMAL DIFFUSIVITYExperimental Procedures

One of the most important thermal properties needed for the development of a re-entry model is thermal conductivity. Certain restrictions, however, make the measurement directly of thermal conductivity undesirable. Among these are: the inaccuracies introduced by the large-scale hydrogen diffusion which will occur during such measurements; the inability to measure conductivity in more than one direction (longitudinal); and the inability to measure conductivity at the very high temperatures because of lack of available equipment. Owing to these restrictions, it was decided that measurement of thermal diffusivities and calculation of conductivities from diffusivities would be more practical, since conductivity is equal to thermal diffusivity times density times specific heat. This method, of course, would depend upon the specific-heat and linear-thermal-expansion measurements which were planned.

Figure 6 illustrates the method employed for making thermal-diffusivity measurements by the single-flash method using a laser as the heat source. A small disk specimen (1), nominally 3/8-in. in diameter and about 0.040 in. thick, is in semi-blackbody condition in a double-wall tubular heater (2). Thermal-radiation shielding (3) in a vacuum envelope (4) permits specimens to attain temperatures near the melting temperature of the material being tested. A 3/8-in.-diameter laser (5) is directed onto one side of the specimen through a window in the vacuum envelope. A lead sulfide radiation receiver (6) and optical system (7) are used to measure the time-temperature history of a small area on the back face of the specimen as the laser is fired. An orifice (8) is used so that the radiation receiver does not see more of the back face of the specimen than it should and views only the central part where the heat flows are undisturbed by edge effects. The thermal diffusivity of the specimen is calculated from a photographed oscilloscope pattern as illustrated in Figure 6.

The times involved in the actual measurements are short. The laser flash lasts about $0.5 \mu\text{sec}$, and the back face of the specimen reaches one-half maximum amplitude in about 20 or 30 μsec . The temperature rise in the specimen is approximately 5 C and, therefore, little hydrogen migration or loss takes place from equilibrium conditions during the actual measurement. With the use of the present resistance-heated tube furnace, approximately 10 min are necessary to heat the specimen from room temperature to 900 C and to level off at that temperature for a measurement. This heating time is for vacuum conditions and is reduced when a gas atmosphere is used to aid heat transfer to the specimen at the lower temperatures.

Figure 7 is a photograph of the thermal-diffusivity apparatus. It shows the vacuum chamber and furnace (1); the vacuum system (2) capable of maintaining a vacuum of 2×10^{-5} torr or better in the chamber; the laser head (3); the nonparallax telescope system (4), which allows visual alignment of the laser on the specimen position; the laser power supply (5); the lead-sulfide radiation detector housing (6), which can be liquid-nitrogen filled to increase the detector sensitivity; and the atmosphere supply (7) and associated pressure gages (8), from which a vacuum or gas pressure up to 1 atm absolute may be controlled. Not shown in Figure 7 are the bridge balance circuit, the

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FIGURE 6. SCHEMATIC DIAGRAM OF THERMAL-DIFFUSIVITY APPARATUS

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FIGURE 7. THERMAL-DIFFUSIVITY APPARATUS, SINGLE-FLASH METHOD USING LASER

oscilloscope, and the camera with which the output signal from the detector is measured. Specimen temperatures are measured with a thermocouple placed in the specimen heater and previously calibrated against a thermocouple placed in a dummy specimen to obtain an observed-versus-true specimen temperature curve. Diffusivity values obtained with this apparatus are believed to be in error by no more than ± 5 per cent.

Experimental Results

Thermal Diffusivity of Dehydrated Material

Thermal-diffusivity measurements were made on two specimens of SNAP-10A fuel which had been dehydrated at a slow rate (approximately 4 C per min). These specimens possessed bulk densities of 6.86 and 6.92 g per cm³, respectively. Post-measurement analysis showed oxygen contents of 2700 ppm for the first specimen and 8800 ppm for the second specimen. Although there was appreciable difference in density and oxygen content between the two specimens, the experimental results as shown in Table 5 and Figure 8 indicated that no change in diffusivity between the two could be noted within experimental error. A preliminary test specimen of dehydrated SNAP-8 fuel was also examined and results for this material agreed well, within experimental error, with the results on the two SNAP-10A specimens.

Graphical presentation of the data for both the SNAP-8 and SNAP-10A materials showed definite changes in the slope of the curves at about 600 C, the $\alpha + \delta \rightleftharpoons \alpha + \beta$ transformation, and at about 1100 C, near the melting point of uranium. Furthermore, although within the normal scatter produced by this method of measurement, data plotted for both SNAP-8 and SNAP-10A specimens showed definite groupings of points, as shown by the dashed lines in Figure 8, indicating probable discontinuities at about 830 C, the $\alpha + \beta \rightleftharpoons \beta$ transformation, and at 1100 C, as well as the changes in slope mentioned above.

Table 6 and Figure 9 show the thermal-diffusivity data for the dehydrated fuel, dehydrated at a fast rate (approximately 30 C per min). Up to 600 C, these data agree well with those of material dehydrated at a slow rate. Above 600 C, however, the data of the specimen dehydrated at the fast rate diverges to a maximum difference of about 11 per cent at 1100 C and converges to within 3 per cent of the slow-rate data at 1300 C. Difficulties with the measuring system prohibited measurements on this specimen above 1300 C.

Thermal Diffusivity of Hydrided Material

Three separate groups of thermal-diffusivity measurements were made on the hydrided SNAP-10A fuel. Measurements were made under equilibrium hydrogen conditions by balancing hydrogen pressure to the hydrogen-vapor-pressure curve as the temperature was increased, in a dynamic vacuum at a heating rate of 4 C per min as the evolved hydrogen was pumped off, and in 1 atm of hydrogen with excess evolved hydrogen pumped off to maintain 1 atm as the temperature was raised.

TABLE 5. THERMAL DIFFUSIVITY OF DEHYDRIDED MATERIAL^(a)
(DEHYDRIDED AT A SLOW RATE) MEASURED IN A
DYNAMIC VACUUM

Specimens 8D and 8L

Temperature, C	Diffusivity, cm ² per sec	
	Experimental	Interpolated From Curve
250	--	0.070
262	(0.071)	
262	(0.067)	
267	0.070(a)	
270	(0.070)	
300	--	0.073
347	0.076(a)	
377	(0.076)(a)	
400	--	0.077
440	0.079	
440	0.079	
500	--	0.079
526	0.080(a)	
578	0.079	
578	0.082	
600	--	0.080
634	(0.090) and (0.087)	
689	0.095(a)	
700	--	0.093
736	0.097	
800	--	0.108
811	0.105	
811	0.108	
852	0.118	
852	0.114	
900	--	0.122
913	0.126(a)	
965	(0.131) and (0.126)(a)	
799	0.105(a)	
985	0.126	
985	0.135	
1000	--	0.134
1078	0.151	
1079	0.145	
1100	--	0.147
1110	(0.155) (0.136) (0.148)(b)	
1163	0.151	
1163	0.145	

TABLE 5. (Continued)

Temperature, C	Diffusivity, cm^2 per sec	
	Experimental	Interpolated From Curve
1200	--	0.148
1253	0.145	
1300	--	0.149
1366	0.151(a)	
1400	--	0.150
1468	0.154	
1500	--	0.151
1570	0.151	
1600	--	0.152(c)

(a) Two points of equal value.

(b) Data in parentheses indicate Specimen 8D - approximate density 6.86 g per cm^3 . All other points indicate Specimen 8L - approximate density 6.92 g per cm^3 .

(c) Extrapolated.

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FIGURE 8. THERMAL DIFFUSIVITY OF DEHYDRIDED MATERIAL (DEHYDRIDED AT A SLOW RATE) MEASURED IN VACUUM

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TABLE 6. THERMAL DIFFUSIVITY OF DEHYDRIDED MATERIAL
(DEHYDRIDED AT A FAST RATE) MEASURED IN A
DYNAMIC VACUUM

Specimen 8AA

Temperature, C	Diffusivity, cm^2 per sec	
	Experimental	Interpolated From Curve
193	0.068	
196	0.069	
200	--	0.069
280	0.072	
300	--	0.074
318	0.075	
319	0.072	
318	0.075	
400	--	0.078
413	0.077	
458	0.078	
458	0.080	
488	0.084	
487	0.080	
500	--	0.080
534	0.079	
535	0.077	
600	--	0.078
608	0.083	
493	0.082	
493	0.078	
537	0.078	
537	0.078	
585	0.079	
585	0.078	
635	0.079	
635	0.080	
685	0.089	
685	0.089	
700	--	0.088
738	0.096	
738	0.092	
780	0.094	
780	0.095	
800	--	0.099
833	0.096	
836	0.097	

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TABLE 6. (Continued)

Temperature, C	Diffusivity, cm ² per sec	
	Experimental	Interpolated From Curve
900	0.109	
900	--	0.111
901	0.117	
897	0.120	
898	0.120	
1000	--	0.121
1042	0.126	
1044	0.126	
1100	--	0.130
1112	0.129	
1113	0.127	
1200	--	0.139
1220	0.134	
1221	0.134	
1220	0.134	
1221	0.138	
1300	--	0.146
1326	0.144	
1331	0.173	
1331	0.146	

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FIGURE 9. THERMAL DIFFUSIVITY OF DEHYDRIDED MATERIAL (DEHYDRIDED AT A FAST RATE) MEASURED IN VACUUM

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Table 7 gives the experimental and interpolated data for a single specimen measured in a hydrogen pressure balanced to the vapor-pressure curve. A qualitative evaluation of the data from the second specimen was also made.

Postmeasurement analysis of the initial specimen indicated a reduction in hydrogen content from 1.82 w/o to 1.64 w/o. The oxygen content of the specimen after test was nearly the same as that of the material before test. The equipment was calibrated before the measurements on fuel samples by obtaining measurements on both Type 347 stainless steel and Armco iron, which are comparison standard materials used in the laboratory, and by comparing the results with the known properties. Very close agreement was noted in these calibration measurements. Postmeasurement calibration, however, showed diffusivity values obtained with the apparatus to be 30 to 40 per cent lower than known values at 100 C, 15 to 20 per cent lower than known values at 200 C, and within 5 per cent of the known values at 300 C and above. These differences are believed to be due to a change in the characteristics of the lead-sulfide radiation detector during the measurements made in a hydrogen atmosphere, since measurements made earlier on dehydrated materials showed good agreement with the initial measurements on the same material.

The diffusivity between 200 C and 350 C for the hydrided material measured in equilibrium hydrogen pressure and for the material measured in 1 atm of hydrogen have been corrected according to results obtained from the postmeasurement of Type 347 stainless steel calibration. The corrected data are shown in Figures 10 and 12, and tabulated in Tables 7 and 9.

Table 8 and Figure 11 show the results of measurements on as-received SNAP-10A fuel material in a dynamic vacuum at a constant heating rate of 4 C per min. Two specimens were used to obtain the data shown. Data from 240 to 635 C were obtained on the first specimen. At the higher temperatures in this range, the diffusivity dropped so rapidly that before data could be recorded, the diffusivity had again increased to the extent that balancing of the oscilloscope-bridge system could not be accomplished. A second specimen was therefore heated at the rate of 4 C per min to 650 C where measurements were then resumed and the diffusivity data shown in Figure 11 were obtained. Chemical analyses of a diffusivity specimen prior to testing showed an oxygen content of 914 ± 20 ppm and a hydrogen content of 1.82 w/o (18,200 ppm). Analyses of the first specimen after testing disclosed an oxygen content of 1123 ± 20 ppm and a hydrogen content of 15 ± 5 ppm. The second specimen showed an oxygen content of 1440 ± 15 ppm and a hydrogen level of less than 10 ppm. Since these measurements were made between measurements of the dehydrated materials and before the suspected change in the lead-sulfide detector, no low-temperature corrections were made to these data.

Table 9 and Figure 12 show the thermal-diffusivity data from the hydrided fuel measured in 1 atm of hydrogen. Temperatures during these measurements were reached in steps. After each temperature change, measurements were made until no change in diffusivity with time could be noted. The "thermal equilibrium" data obtained in the first run are those shown in the table and as the solid line on the figure. Data are reported over the temperature range from 240 to 1000 C. The hydrogen and oxygen contents of this specimen after the measurements were 1.55 w/o and 5040 ppm, respectively.

TABLE 7. THERMAL DIFFUSIVITY OF HYDRIDED MATERIAL MEASURED
IN HYDROGEN PRESSURE BALANCED TO VAPOR-PRESSURE
CURVE

Specimen 4I

Temperature, C	Diffusivity, cm^2 per sec	
	Experimental	Interpolated From Curve
200	--	0.098
204	0.098	
244	0.091	
222	0.095	
250	--	0.095
280	0.096	
300	--	0.092
217	0.094	
345	0.092	
350	--	0.089
398	0.086	
400	--	0.087
421	0.088	
450	--	0.086
467	0.087	
500	--	0.085
506	0.084	
516	0.087	
537	0.083	
550	--	0.085
573	0.083	
590	0.087	
600	--	0.085
602	0.083	
609	0.083	
626	0.087	
633	0.088	
643	0.084	
645	0.084	
650	0.087	
650	0.090	
650	--	0.085
635	0.090	

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FIGURE 10. THERMAL DIFFUSIVITY OF HYDRIDED MATERIAL UNDER A BALANCED HYDROGEN PRESSURE

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TABLE 8. THERMAL DIFFUSIVITY OF HYDRIDED MATERIAL
MEASURED IN DYNAMIC VACUUM AT A HEATING
RATE OF 4 C PER MIN

Specimens 4B and 4C

Temperature, C	Diffusivity, cm ² per sec	
	Experimental	Interpolated From Curve
200	--	0.099
242	0.102	
247	0.096	
250	--	0.097
300	--	0.094
322	0.093	
326	0.091	
350	--	0.090
373	0.086	
386	0.089	
400	--	0.085
420	0.082	
424	0.082	
450	--	0.078
459	0.078	
460	0.080	
496	0.074	
499	0.076	
500	--	0.071
531	0.068	
534	0.067	
538	0.063	
550	--	0.062
598	0.051	
600	--	0.051
605	0.050	
625	--	0.045
638	0.043	
700	--	0.052(a)
720	0.061	
723	0.065	
735	0.078	
750	--	0.087
770	0.106	
780	0.121	
787	0.109	
800	--	0.115

TABLE 8. (Continued)

Temperature, C	Diffusivity, cm ² per sec	
	Experimental	Interpolated From Curve
808	0.108	
811	0.109	
842	0.126	
843	0.124	
850	--	0.137
865	0.150	
872	0.150	
877	0.153	
884	0.150	
900	--	0.156
901	0.150	
916	0.157	

(a) Interpolated.

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FIGURE 11. THERMAL DIFFUSIVITY OF HYDRIDED MATERIAL MEASURED IN DYNAMIC VACUUM AT A HEATING RATE OF 4 C PER MIN

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TABLE 9. THERMAL DIFFUSIVITY OF HYDRIDED MATERIAL
MEASURED IN 1 ATM OF HYDROGEN

Specimen 4K

Temperature, C	Diffusivity, cm^2 per sec	
	Experimental	Interpolated From Curve
240	0.086 ^(a)	
250	--	0.085
295	0.081	
300	0.081	
300	--	0.084
349	0.089	
350	--	0.083
355	0.086	
400	--	0.084
403	0.084	
408	0.084	
450	0.086	
450	--	0.087
451	0.086	
497	0.087	
500	--	0.089
501	0.087	
520	0.091	
523	0.091	
549	0.097	
550	--	0.091
553	0.092	
581	0.089	
583	0.091	
600	--	0.091
608	0.089 ^(a)	
625	0.091 ^(a)	
650	--	0.090
654	0.087	
655	0.087	
679	0.089	
678	0.079 ^(a)	
699	0.079	
700	--	0.081
703	0.077	
725	0.075	
726	0.073	
742	0.072	
745	0.071	

TABLE 9. (Continued)

Temperature, C	Diffusivity, cm^2 per sec	
	Experimental	Interpolated From Curve
750	--	0.068
762	0.061	
762	0.063	
792	0.061	
787	0.061	
800	--	0.061
828	0.061	
832	0.059	
840	0.057	
838	0.057	
910	0.052 ^(a)	
950	--	0.050
982	0.047	
984	0.047	

(a) Two points with the same value.

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FIGURE 12. THERMAL DIFFUSIVITY OF HYDRIDED MATERIAL MEASURED IN 1 ATM OF HYDROGEN

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The thermal-diffusivity values for the hydrided material measured under equilibrium hydrogen conditions (balanced against the dissociation pressure) were expected to correspond closely with those for the same material measured in 1 atm of hydrogen. The values for the first two specimens measured are shown as Run No. 1 in Figures 10 and 12. These values did correspond within ± 10 per cent, but the slopes of the curves for the two measurements were somewhat different. An appraisal of the method, data, and procedures was made in order to find the reason for these differences. A discussion of this evaluation is given in the section entitled Discussion of Results.

Difficulties in the temperature calibration of the specimen measured under a balanced hydrogen pressure resulted in specimen temperatures higher than expected or, conversely, hydrogen pressures lower than the actual dissociation pressures. The results of these measurements are shown by the dashed curve identified as Run No. 2 in Figure 10, and indicate, qualitatively, a loss of hydrogen from the specimen and a resulting decrease in thermal diffusivity above about 450 C.

The specimen in Run No. 2 was cooled (using the same pressure-temperature relationship) and was rerun under a constant hydrogen pressure of 1 atm. Qualitative results from this run are shown as Run No. 2 in Figure 12. Here it can be seen that 1 atm of hydrogen produced an overpressure of hydrogen up to the temperature of 550 C, thereby increasing the hydrogen content and the diffusivity values both on the heating portion and the cooling portion of the thermal cycle.

LINEAR-THERMAL-EXPANSION MEASUREMENTS

Experimental Procedures

Linear-thermal-expansion measurements were conducted in the recording dilatometer shown in Figure 13. A lever system, mounted in a pressure chamber (1), was used to magnify the total specimen expansion. Specimens were mounted in a furnace (2) which could be utilized to a temperature of 660 C. This furnace was replaced by a molybdenum-wound furnace to obtain temperatures up to 1650 C which was necessary for the measurement of dehydrided specimens. Total length changes were transmitted to an x-y recorder by means of a reversing motor, gear train, and Selsyn motor (3).

Expansion-versus-temperature curves were automatically plotted on 11-in.-strip chart paper on the x-y recorder (4). The x-axis movement designated temperature and the y-axis movement was specimen elongation. Continuous curves which were drawn were useful not only in showing the expansion and contraction of specimens but also in showing the temperatures at which transformation occurred. The dilatometer showed any permanent change in length at room temperature resulting from the experimental thermal cycle. The length change was also checked by making pre- and postlength measurements using a Pratt & Whitney measuring machine. Temperature rates were obtained with the aid of a programmer (5) and a proportional control (6). The hydrogen pressure was maintained by a supply system (7) with a precision pressure gage (8). A quartz structure was used with the hydrided specimens to 660 C.

FIGURE 13. RECORDING DILATOMETER

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The recording dilatometer in Figure 13 was modified to obtain temperatures to 1650 C for the dehydrided specimens. For these measurements, a graphite structure was substituted for the quartz structure in the dilatometer. The hydrogen-pressure-regulating apparatus (7 and 8) was removed, in addition, and the metal pressure chamber (1) was replaced by a conventional glass cylinder.

Experimental Results

Experimentally observed thermal-expansion values and data calculated from these values, which were used to generate the density data, are presented in Tables 10 through 14. Table 10 and Figure 14 show the expansion data for the hydrided material in both transverse and longitudinal directions under equilibrium hydrogen pressure. Tables 11, 12, and 13 give expansion data on dehydrided material. The expansion as a function of temperature for the dehydrided materials is plotted in Figures 15, 16, and 17. Table 14 and Figure 18 show expansion data on as-received fuel in the longitudinal direction under 1 atm of hydrogen.

The expansion coefficient, particularly for the first temperature cycle in the dilatometer, is very sensitive to quite minor specimen variations, such as differences in mechanical or thermal stresses in specimens, composition, impurity level, orientation, and, in the fuel materials, effects resulting from densification and hydrogen evolution. Many of these variations or effects are annealed or homogenized out during the first one or two cycles. In fact, the expansion coefficient is a sensitive way to show the results of these effects.

Measurements made in the quartz dilatometer are believed to be in error by no more than ± 2 per cent, and those made in the sapphire dilatometer by no more than ± 3 per cent.

DENSITY MEASUREMENTS AND CALCULATIONS

Room-Temperature Density

Room-temperature densities were calculated from measured volumes and weights of the specimens. Figures 19 and 20 and Table 15 give the variation of densities measured at 20 C by frequency of occurrence of the hydrided and the dehydrided material used in this program. Individual density measurements are given in Tables 16 and 17. Within experimental error, the hydrided specimens may be represented by their average density, 6.030 ± 0.018 g per cm^3 , which is 86.6 per cent of theoretical density. The standard deviation is based on measurements on 33 specimens and is taken as the standard deviation of the measuring method.

TABLE 10. THERMAL EXPANSION OF HYDRIDED MATERIAL UNDER
EQUILIBRIUM HYDROGEN PRESSURELongitudinal Specimen 1B
Transverse Specimen 2B

Temperature, C	Expansion, per cent		Temperature, C	Expansion Coefficient, 10 ⁻⁶ per C	
	Transverse	Longitudinal		Transverse	Longitudinal
20	0.000	0.000	35	7.7	5.3
50	0.023	0.016	75	8.0	7.0
100	0.064	0.051	125	8.8	8.2
150	0.108	0.092	175	9.6	9.2
200	0.156	0.138	225	10.2	10.0
250	0.207	0.188	275	11.0	10.8
300	0.262	0.242	325	11.4	11.6
350	0.319	0.300	375	12.0	12.2
400	0.379	0.366	425	12.2	12.6
450	0.440	0.424	475	13.2	12.8
500	0.506	0.488	525	13.6	13.2
550	0.574	0.554	575	14.8	13.4
600	0.648	0.621	625	15.8	13.8
650	0.727	0.690			

TABLE 11. THERMAL EXPANSION OF DEHYDRIDED MATERIALS - FIRST CYCLE TO
1000 C - USING A QUARTZ DILATOMETER STRUCTURE

Longitudinal Specimen 10A
Transverse Specimen 11A

Temperature, C	Expansion, per cent				Temperature, C	Expansion Coefficient, 10^{-6} per C			
	Longitudinal		Transverse			Longitudinal		Transverse	
	Heating	Cooling	Heating	Cooling		Heating	Cooling	Heating	Cooling
20	0.000	-0.164	0.000	-0.268	60	6.9	6.9	6.5	6.5
50	0.020	-0.144	0.018	-0.250	150	7.1	7.3	7.0	7.0
100	0.055	-0.109	0.052	-0.216	250	7.4	7.2	7.3	7.4
150	0.090	-0.074	0.087	-0.181	350	7.6	7.6	7.6	7.4
200	0.126	-0.038	0.122	-0.146	450	8.0	8.0	7.8	8.0
250	0.163	-0.001	0.158	-0.111	540	8.8	10.3	8.0	7.8
300	0.200	0.036	0.195	-0.072	625	6.4	6.4	2.8	6.6
350	0.238	0.074	0.232	-0.036	675	4.6	5.0	1.4	5.2
400	0.276	0.112	0.271	0.002	725	1.0	1.4	3.6	0.8
450	0.315	0.151	0.310	0.042	775	-4.0	0.0	-9.8	0.8
500	0.356	0.192	0.349	0.082	825	-3.4	7.8	-7.8	5.8
550	0.400	0.236	0.388	0.120	875	-4.6	10.8	0.0	7.2
595	0.446	0.301	0.425	0.176	925	+7.0	10.5	2.4	6.6
600	0.466	0.304	0.454	0.185	975	6.4	11.4	1.2	7.0
650	0.498	0.336	0.468	0.218					
700	0.521	0.361	0.475	0.244					
750	0.526	0.368	0.453	0.248					
800	0.506	0.368	0.404	0.252					
850	0.489	0.407	0.365	0.281					
900	0.506	0.461	0.365	0.315					
950	0.541	0.516	0.377	0.348					
1000	0.573	0.573	0.383	0.383					

TABLE 12. THERMAL EXPANSION OF DEHYDRIDED MATERIAL TO 1600 C -
USING A GRAPHITE DILATOMETER STRUCTURE (HEATING ONLY)Longitudinal Specimen 10C
Transverse Specimen 11C

Temperature, C	Expansion, per cent		Temperature, C	Expansion Coefficient, 10 ⁻⁶ per C	
	Longitudinal	Transverse		Longitudinal	Transverse
20	0.000	0.0000	60	6.06	5.89
100	0.0485	0.0354	150	6.85	5.69
200	0.117	0.0922	250	7.50	8.62
300	0.192	0.178	350	7.19	7.15
400	0.264	0.250	450	8.27	7.47
500	0.347	0.325	550	10.23	9.34
600	0.449	0.418	650	4.76	6.40
700	0.499	0.482	750	2.01	3.45
800	0.478	0.517	850	0.92	4.70
900	0.488	0.564	950	5.63	7.08
1000	0.544	0.634	1050	3.50	3.18
1100	0.579	0.666	1150	3.15	2.59
1200	0.610	0.640	1250	6.20	5.31
1300	0.672	0.587	1350	11.53	7.30
1400	0.788	0.660	1450	14.28	15.46
1500	0.931	0.815	1525	--	19.42
1550	--	0.912	1550	21.20	--
1600	1.14	--			

TABLE 13. EXPANSION DATA ON DEHYDRIDED MATERIAL TO
1575 C AND RETURN

Specimen 10D

Temperature, C	Thermal Expansion, per cent		Temperature, C	Expansion Coefficient, 10 ⁻⁶ per C	
	Heating	Cooling		Heating	Cooling
20	0.000	-0.941	60	6.9	6.5
100	0.055	-0.889	150	6.4	6.1
200	0.119	-0.828	250	6.5	6.7
300	0.183	-0.761	350	7.0	7.3
400	0.253	-0.688	450	6.9	7.2
500	0.322	-0.616	541	--	8.3
582	--	-0.548	550	7.7	--
591	--	-0.525	611	15.5	--
600	0.399	--	646	6.2	7.6
611	0.424	--	695	4.1	--
622	0.433	--	725	--	6.5
670	0.463	--	740	-0.2	--
700	--	-0.442	775	--	6.1
719	0.483	--	780	-3.6	--
750	--	-0.410	825	-1.2	13.8
760	0.482	--	875	5.1	8.9
800	0.468	-0.380	950	8.4	11.6
850	0.462	-0.311	1050	9.2	11.6
900	0.488	-0.268	1175	8.3	14.9
1000	0.572	-0.151	1225	5.8	--
1100	0.653	-0.034	1250	--	14.9
1116	--	-0.015	1275	7.7	--
1118	--	0.017	1350	9.2	19.1
1150	0.734	0.058	1450	12.9	32.6
1200	0.776	0.132	1538	17.8	53.3
1225	0.789	--			
1250	0.805	--			
1300	0.843	0.281			
1400	0.935	0.472			
1500	1.064	0.798			
1575	1.198	1.198			

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TABLE 14. THERMAL EXPANSION OF HYDRIDED MATERIAL
UNDER 1 ATM HYDROGEN

Specimen 1C

Temperature, C	Expansion, per cent	
	Heating(a)	Cooling
20	0.000	-0.149
100	0.047	-0.110
200	0.120	-0.028
300	0.215	0.094
400	0.331	0.243
500	0.455	0.355
595	0.597	0.421
608	0.617	
627	0.644	
635	0.644	
650		0.448
675	0.699	
695	0.714	
693	0.658	
700		0.465
730	0.701	
745	0.701	
740	0.620	
750		0.436
770	0.642	
800	0.647	0.274
795	0.568	
820	0.576	
840	0.559	
840	0.505	
850		0.123
870	0.502	
890	0.354	
890	0.227	
900		-0.017
920	0.101	
940	-0.007	
950	-0.071	
950	-0.178	0.178

(a) Heating curve built up of segments measured between hold temperatures.

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FIGURE 14. THERMAL EXPANSION OF HYDRIDED MATERIAL UNDER EQUILIBRIUM HYDROGEN PRESSURE

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FIGURE 15. THERMAL EXPANSION OF DEHYDRIDED MATERIAL USING A QUARTZ DILATOMETER STRUCTURE

FIGURE 16. THERMAL EXPANSION OF DEHYDRIDED MATERIAL IN THE LONGITUDINAL DIRECTION USING A GRAPHITE DILATOMETER STRUCTURE

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FIGURE 17. THERMAL EXPANSION OF DEHYDRIDED MATERIAL USING A GRAPHITE DILATOMETER STRUCTURE

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FIGURE 18. THERMAL EXPANSION OF HYDRIDED MATERIAL IN THE LONGITUDINAL DIRECTION UNDER 1 ATM OF HYDROGEN

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FIGURE 19. HYDRIDED-MATERIAL DENSITY HISTOGRAM (AT 20 C)

FIGURE 20. DEHYDRIDED-MATERIAL DENSITY HISTOGRAM (AT 20 C)

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TABLE 15. MATERIAL DENSITY AT 20 C

Density, g per cm ³	Frequency	Density, per cent of theoretical
<u>Hydrided Material</u>		
5.970	1	85.7
5.980	0	85.9
5.990	1	86.0
6.000	0	86.2
6.010	1	86.3
6.020	8	86.4
6.030	6	86.6
6.040	14	86.7
6.050	1	86.9
6.060	1	87.0
<u>Dehydrided Material</u>		
6.64	1	95.3
--	--	
6.70	1	96.2
--	--	
6.74	2	96.8
6.76	1	97.1
6.78	1	97.4
6.80	0	97.6
6.82	1	97.9
6.84	2	98.2
6.86	1	98.5
6.88	4	98.8
6.90	13	99.1
6.92	3	99.4
6.94	5	99.7
6.96	2	100.0

TABLE 16. HYDRIDED-SPECIMEN DENSITY BY INDIVIDUAL SPECIMENS

Specimen	Density		Specimen	Density	
	G per Cm ³	Per Cent of Theoretical		G per Cm ³	Per Cent of Theoretical
4A	6.022	86.5	1A	6.046	86.8
B	6.017	86.4	1C	6.043	86.8
C	6.024	86.5	1D	6.039	86.7
D	6.026	86.5			
E	6.024	86.5	3A	6.040	86.7
F	6.022	86.5	3B	6.044	86.8
G	6.024	86.5			
H	6.031	86.6	5A	6.028	86.6
I	6.023	86.5	5B	6.010	86.3
J	6.064	87.1			
K	6.026	86.5	6A	5.986	86.0
L	6.023	86.5	6B	6.021	86.5
M	5.967	85.7			
N	6.027	86.5			
7C	6.043	86.8			
D	6.036	86.7			
E	6.042	86.8			
F	6.029	86.8			
G	6.043	86.8			
H	6.043	86.8			
I	6.040	86.7			
J	6.041	86.7			
K	6.041	86.7			
L	6.040	86.7			

Note: $x \approx 6.030 \pm 0.018$ g per cm³.
 ± 0.3 per cent.

TABLE 17. DEHYDRIDED SPECIMEN DENSITY BY INDIVIDUAL SPECIMENS

Specimen	Density		Specimen	Density	
	G per Cm ³	Per Cent of Theoretical		G per Cm ³	Per Cent of Theoretical
8A	6.906	99.2	10B	6.697	96.2
B	6.892	99.0	C	6.754	97.0
C	6.734	96.7	D	6.746	96.9
D	6.864	98.6	11C	6.643	95.4
E	6.934	99.6	8CC	6.904	99.1
F	6.812	97.8	DD	6.900	99.1
G	6.886	98.9	EE	6.965	100.0
J	6.908	99.2	FF	6.878	98.8
K	6.947	99.8	GG	6.830	98.1
L	6.923	99.4	HH	6.922	99.4
M	6.895	99.0	II	6.895	99.0
N	6.953	99.8	JJ	6.902	99.1
O	6.939	99.6			
P	6.902	99.1			
Q	6.888	98.9	Large specimen	6.773	97.3
S	6.927	99.5			
T	6.891	99.0			
9H	6.939	99.6			
I	6.908	99.2			
J	6.909	99.2			
K	6.938	99.6			
L	6.884	98.9			
M	6.838	98.2			
N	6.900	99.1			

Note: Average, 6.8736 g per cm³ 98.7 % theoretical.

The density of the dehydrated material falls into two sets. The smaller specimens average 6.897 g per cm³, which is 99.0 per cent of theoretical density, and the larger specimens, which were used in the thermal-expansion work, average 6.710 g per cm³, which is only 96.4 per cent of theoretical. The difference between the two sets is highly significant. The densities of the dehydrated material also show much more significant variation than do the densities of the hydrated material.

These observed densities represent the materials at the start of the physical-property measurements.

Table 18 gives the observed density of the arc-melted material, which is taken as the theoretical-density standard, and of the arc-melted material with oxygen and nitrogen additions. The data are plotted in Figures 21 and 22.

TABLE 18. MATERIAL DENSITY

Density, g per cm ³	Specimen Description
6.964	Arc-melted biscuit, 100 per cent dense
	<u>Per Cent Oxygen Additions(a)</u>
6.992	2
7.067	4
7.125	6
7.091	11
6.976	13
	<u>Per Cent Nitrogen Additions(a)</u>
6.964	1
7.001	2
7.025	3
7.030	4

(a) Additions were made to the biscuit during the arc-melting process.

Elevated-Temperature Density

Density data at elevated temperatures were calculated from the room-temperature densities and experimentally observed thermal-expansion data (given in Appendix).

Specifically, the observed measure of the density is the specific volume. To take into account the anisotropic behavior of the material, the specific volume is given by

$$V(T) = L \times L_T^2 ,$$

FIGURE 21. DENSITY OF ARC-MELTED MATERIAL WITH OXYGEN ADDITIONS

FIGURE 22. DENSITY OF ARC-MELTED MATERIAL WITH NITROGEN ADDITIONS

where L = relative length in the longitudinal direction and L_T = relative length in the transverse direction. Where the mass of the specimen does not change, as in the determination under equilibrium hydrogen pressure and, within limits, in the determination made on the dehydrided materials, the reciprocal specific volume is directly proportional to the density. In the tables, it is tabulated as relative density. The theoretical-density values given in the tables are based on the 100 per cent dense, arc-melted material.

Table 19 gives the density, over the temperature range 20 to 650 C, for the hydrided material under equilibrium hydrogen pressure. The density varies smoothly from 86.6 to 84.8 per cent of theoretical. Figures 23 and 24 give relative density and the density based on the measured average, 6.030 g per cm³.

Tables 20, 21, and 22 give density data for the dehydrided material calculated from linear expansion measured to 1000 and 1550 C in a vacuum. These data constitute four independent determinations. Longitudinal and transverse specimens were involved in each determination. To provide maximum information, both the heating and cooling data are presented since there was considerable difference in the data as a result of the densification mechanism. A quartz structure was used in the expansion measurements to 1000 C, and a graphite structure was used in the measurements to 1550 C. The density data are plotted in Figure 25.

Figure 26 gives the volumetric coefficient of expansion, calculated by differences from the tabulated data, for the hydrided and the dehydrided material. The data are in complete accord with the phase diagram. For the dehydrided material, the evidence of hysteresis or slight displacement of transition boundaries is considered characteristic of this particular alloy. Of most importance is the clearly defined evidence of free uranium in the alloy as shown by the break in the expansion curve at 1120 C.

Table 23 gives the relative volume and density data for the hydrided material under 1 atm hydrogen pressure over the temperature range 20 to 1000 C. These data are based only on the longitudinal direction, and the heating curve is built up of segments between temperatures at which the specimen was held for various times in order to find the length change caused by the temperature arrests. The relative volume is the experimentally observed quantity. Its reciprocal is accurately proportional to the density. The specimen, however, loses hydrogen under these conditions and the density changes accordingly. The density data above 650 C are estimated on the basis of a 1 per cent loss of hydrogen which is, at this time, simply a guess for the actual hydrogen loss. The relative densities are plotted in Figure 27.

Table 24 and Figure 28 give the observed change in length as a function of time when the material was held at various temperatures. The data are for a longitudinal hydrided specimen under 1 atm of hydrogen. The recording dilatometer with a quartz structure was used to generate these data. No kinetic analysis was made at this time.

The reported density values are believed to lie within a relative error band of a few per cent. The absolute values are believed to be in error by no more than ± 5 per cent.

TABLE 19. DENSITY OF THE HYDRIDED MATERIAL UNDER
EQUILIBRIUM HYDROGEN PRESSURE^(a)

Temperature, C	Relative Volume	Density		
		Relative	Per Cent of Theoretical	G per Cm ³
20	1.0000	1.000	86.6	6.030
50	1.0006	0.9994	86.5	6.026
100	1.0018	0.9982	86.4	6.019
150	1.0031	0.9969	86.3	6.011
200	1.0045	0.9955	86.2	6.003
250	1.0060	0.9940	86.1	5.994
300	1.0077	0.9924	85.9	5.984
350	1.0094	0.9907	85.8	5.974
400	1.0112	0.9889	85.6	5.963
450	1.0131	0.9871	85.5	5.952
500	1.0151	0.9851	85.3	5.940
550	1.0171	0.9832	85.1	5.929
600	1.0193	0.9811	85.0	5.916
650	1.0216	0.9789	84.8	5.903

(a) Equilibrium hydrogen pressure data supplied by Sandia.

(b) Density values based on the average density of the hydrided material, 6.030 g per cm³.

FIGURE 23. RELATIVE DENSITY OF HYDRIDED MATERIAL

FIGURE 24. DENSITY OF THE HYDRIDED MATERIAL UNDER EQUILIBRIUM HYDROGEN PRESSURE

TABLE 20. DENSITY OF DEHYDRIDED MATERIAL, FIRST CYCLE TO 1000 C AND RETURN

Temperature, C	Relative Volume		Relative Density		Density, per cent of theoretical	
	Heating	Cooling	Heating	Cooling	Heating	Cooling
20	1.00000	0.99302	1.00000	1.00703	96.40	97.08
100	1.00159	0.99459	0.99841	1.00544	96.25	96.92
200	1.00270	0.99670	0.99731	1.00331	96.14	96.72
300	1.00591	0.99892	0.99412	1.00108	95.83	96.50
400	1.00820	1.00116	0.99187	0.99884	95.62	96.29
500	1.01057	1.00356	0.98954	0.99645	95.39	96.07
550	1.01181	1.00477	0.98833	0.99525	95.28	95.94
600	1.01380	1.00675	0.98639	0.99330	95.09	95.75
650	1.01441	1.00773	0.98579	0.99233	95.03	95.66
700	1.01478	1.00852	0.98544	0.99155	95.00	95.59
750	1.01439	1.00867	0.98581	0.99140	95.03	95.57
800	1.01320	1.00875	0.98697	0.99133	95.14	95.56
850	1.01224	1.00972	0.98791	0.99037	95.23	95.47
900	1.01241	1.01095	0.98774	0.98917	95.22	95.36
950	1.01300	1.01216	0.98717	0.98799	95.16	95.24
1000	1.01344	1.01344	0.98674	0.98674	95.12	95.12

TABLE 21. DENSITY OF DEHYDRIDED MATERIAL, FIRST CYCLE TO 1550 C

Temperature, C	Relative Volume	Relative Density	Density, per cent of theoretical
20	1.00000	1.00000	96.4
100	1.00121	0.99879	96.3
200	1.00301	0.99699	96.1
300	1.00549	0.99454	95.9
400	1.00764	0.99241	95.7
500	1.00999	0.99010	95.4
550	1.01093	0.98919	95.4
600	1.01290	0.98726	95.2
650	1.01384	0.98634	95.1
700	1.01469	0.98552	95.0
800	1.01519	0.98503	95.0
850	1.01528	0.98495	94.9
900	1.01624	0.98402	94.9
950	1.01716	0.98312	94.8
1000	1.01823	0.98209	94.7
1050	1.01866	0.98167	94.6
1100	1.01923	0.98113	94.6
1150	1.01909	0.98127	94.6
1200	1.01902	0.98133	94.6
1300	1.01857	0.98176	94.6
1400	1.02122	0.97921	94.4
1500	1.02582	0.97482	94.0
1550	1.02887	0.97194	93.7

TABLE 22. DENSITY OF DEHYDRIDED MATERIAL TO 1575 C AND RETURN

Temperature, C	Relative Volume		Relative Density		Density, per cent of theoretical	
	Heating	Cooling	Heating	Cooling	Heating	Cooling
20	1.00000	0.972007	1.00000	1.028796	96.9	99.7
100	1.001651	0.973538	0.99835	1.027179	96.7	99.5
200	1.003573	0.975337	0.99643	1.025283	96.5	99.3
300	1.005527	0.977345	0.99450	1.023174	96.3	99.1
400	1.007633	0.979475	0.99242	1.020949	96.1	98.9
500	1.009721	0.981596	0.99037	1.018755	95.9	98.7
582	--	0.983620	--	1.016652	--	98.5
591	--	0.984305	--	1.015940	--	98.4
600	1.012012	--	0.98813	--	95.7	--
611	1.012806	--	0.98735	--	95.6	--
622	1.013068	--	0.98710	--	95.6	--
670	1.013953	--	0.98623	--	95.5	--
700	--	0.986769	--	1.013407	--	98.2
719	1.014557	--	0.98565	--	95.5	--
750	--	0.987721	--	1.012432	--	98.1
760	1.014557	--	0.98565	--	95.5	--
800	1.014134	0.988643	0.98606	1.011490	95.5	98.0
850	1.013953	0.990698	0.98623	1.009397	95.5	97.8
900	1.014708	0.992011	0.98550	1.008054	95.5	97.6
1000	1.017256	0.995474	0.98303	1.004550	95.2	97.3
1100	1.019746	0.998950	0.98063	1.001051	95.0	97.0
1116	--	0.999550	--	1.000450	--	97.0
1118	--	1.000540	--	0.999460	--	96.9
1150	1.022208	1.001741	0.97827	0.998263	94.8	96.7
1200	1.023491	1.003994	0.97704	0.996025	94.6	96.5
1225	1.023888	--	0.97667	--	94.6	--
1250	1.024340	--	0.97623	--	94.6	--
1300	1.025533	1.008486	0.97510	0.991591	94.5	96.0
1400	1.028346	1.014254	0.97243	0.985950	94.2	95.5
1500	1.031277	1.024127	0.96967	0.976438	93.9	94.6
1575	1.036369	1.036368	0.96490	0.964906	93.5	93.5

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FIGURE 25. RELATIVE DENSITY OF DEHYDRIDED MATERIAL

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FIGURE 26. INSTANTANEOUS VOLUMETRIC COEFFICIENT OF EXPANSION FOR THE DEHYDRIDED MATERIAL

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TABLE 23. RELATIVE VOLUME AND RECIPROCAL RELATIVE VOLUME OF
HYDRIDED MATERIAL UNDER 1 ATM HYDROGEN

Temperature, C	Relative Volume		Reciprocal Relative Volume		Density(a)			
					G per Cm ³		Per Cent of Theoretical	
	Heating	Cooling	Heating	Cooling	Heating	Cooling	Heating	Cooling
20	1.00000	0.99553	1.00000	1.00449	6.030	6.057	86.59	86.98
100	1.00141	0.99670	0.99859	1.00331	6.021	6.050	86.46	86.88
200	1.00360	0.99916	0.99641	1.00084	6.008	6.035	86.27	86.66
300	1.00646	1.00282	0.99358	0.99719	5.991	6.013	86.03	86.35
400	1.00996	1.00731	0.99014	0.99274	5.971	5.986	85.74	85.96
500	1.01371	1.01069	0.98648	0.98942	5.948	5.966	85.41	85.67
600	1.01817	1.01269	0.98215	0.98747	5.922	5.954	85.04	85.50
650	1.02030	1.01350	0.98010	0.98668	5.910	5.950	84.87	85.44
700	1.02160	1.01401	0.97886	0.98618	5.894	5.938	84.64	85.27
750	1.02115	1.01314	0.97929	0.98703	5.887	5.934	84.54	85.21
800	1.01956	1.00824	0.98082	0.99183	5.888	5.955	84.55	85.51
850	1.01653	1.00369	0.98374	0.99632	5.898	5.973	84.70	85.77
900	1.00903	0.99949	0.99105	1.00051	5.932	5.989	85.18	86.00
950	0.99775	0.99467	1.00226	1.00536	5.991	6.009	86.03	86.29
1000	0.98507	0.98507	1.01516	1.01516	6.060	6.060	87.02	87.02

(a) All density data above 650 C are estimated on the basis of a 1 per cent loss of hydrogen.

FIGURE 27. RELATIVE DENSITY OF HYDRIDED MATERIAL UNDER 1 ATM OF HYDROGEN

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TABLE 24. LENGTH CHANGE OF HYDRIDED MATERIAL HELD UNDER
1 ATM HYDROGEN AT VARIOUS TEMPERATURES

Time, min	Length Change, $\Delta l/l \times 10^{-4}$, at Indicated Arresting Temperature, C							
	595	627	695	745	800	840	890	950
0	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
1	0.77	0.00	0.00	-0.15	-0.15	-0.29	-0.58	-0.88
2	1.05	0.00	-0.29	-0.29	-0.73	-1.36	-2.49	-2.05
3	1.08	0.00	-0.58	-0.58	-1.61	-2.34	-4.24	-2.63
4	1.22	0.00	-0.88	-1.02	-2.49	-2.92	-5.70	-2.92
5	1.24	0.00	-1.16	-1.61	-3.07	-3.22	-7.02	-3.80
6	1.24	0.00	-1.75	-1.90	-3.65	-3.80	-7.60	-4.53
7	1.24	0.00	-2.19	-2.92	-4.09	-3.95	-8.33	-5.12
8	1.25	0.00	-2.48	-3.22	-4.39	-4.09	-8.92	-5.55
9	1.10	0.00	-2.63	-3.51	-4.68	-4.24	-9.50	-5.85
10	1.25	0.00	-2.78	-3.80	-4.97	-4.24	-10.09	-5.99
11	1.11	0.00	-2.92	-4.09	-5.12	-4.24	-10.52	-6.29
12	1.11	0.00	-3.22	-4.39	-5.26	-4.24	-10.82	-6.43
14	1.11	0.00	-3.51	-4.53	-5.70	--	-11.84	-6.87
16	1.11	0.00	-3.36	-4.97	-5.99	--	-12.86	-7.45
18	0.97	--	-3.36	-5.12	-6.14	--	-13.02	-7.89
20	--	--	-3.51	-5.70	-6.43	--	-13.59	-8.48
25	--	--	-3.73	-6.10	-6.90	--	-14.25	-9.65
30	--	--	-4.09	-6.58	-7.31	--	-15.06	-10.52
35	--	--	-4.82	-6.70	-7.31	--	--	-10.67
40	--	--	-4.82	-6.87	-7.31	--	--	-10.67
50	--	--	--	-7.00	--	--	--	--
60	--	--	--	--	--	--	--	--

FIGURE 28. LENGTH CHANGE IN HYDRIDED MATERIAL HELD AT VARIOUS TEMPERATURES UNDER 1 ATM HYDROGEN

THERMAL CONDUCTIVITY

Thermal conductivities were calculated by multiplying density times specific heat times diffusivity, where each factor was measured under, as nearly as possible, similar conditions. Values used in the computation were taken from the interpolated data shown in the individual sections of this report.

Thermal-conductivity values for the dehydrated SNAP-10A fuel are given in Table 25 with the density, specific heat, and diffusivity values used in the computations. A plot of the conductivity data from Table 25 is given in Figure 29.

Table 26 gives the calculated thermal conductivity for the hydrided fuel measured under equilibrium hydrogen pressure. Data from this calculation are shown plotted in Figure 30.

Calculations of the thermal conductivity of the hydrided fuel, measured in 1 atm of hydrogen, are given in Table 27 and the results are shown graphically in Figure 31. The conductivity values above 660 C are somewhat in doubt since the specific-heat data were measured to only 660 C and were extrapolated above that temperature.

Since thermal conductivity values were calculated from measured thermal-diffusivity and specific-heat data and from calculated density data, the cross product of these three values enlarges any errors which might lie in the original data. Any error in analysis of the thermal conductivity data must, therefore, be referred to the probable errors in original data, which have been discussed at the end of each section.

HEMISPHERICAL TOTAL EMITTANCE

Experimental Procedures

Hemispherical total-emittance measurements were made by the hole-in-tube method. Specimens of the dehydrated SNAP-10A fuel were machined into 6-in.-long tubes with approximately 1/4-in. ID and with 0.020-in. wall thickness. Measurements were made in the apparatus shown partially disassembled in Figure 32. The specimen (1) was mounted vertically between two water-cooled current clamps (2) so as to be at least one specimen diameter from the axis of the blackened, water-cooled vacuum chamber (3) when assembled. This eliminated first reflections from the chamber walls which could cause error in the readings. The lower current clamp is moveable and was adjusted vertically to allow for specimen expansion during measurement. A small sight hole approximately 1/64 in. in diameter was drilled through the specimen wall midway between the specimen ends. Two 15-mil tungsten potential leads were pinned into holes 3/4 in. on either side of the sight hole. The assembly, suspended from the vacuum chamber top plate as shown, was then lowered into the chamber and positioned so that the sight hole could be viewed through the shuttered window (4). The vacuum system (5) was turned on and allowed to pump out the chamber over night,

TABLE 25. THERMAL CONDUCTIVITY CALCULATIONS FOR DEHYDRIDED MATERIALS

Temperature, C	Density, g per cm ³	Specific Heat, cal/(g) (C)	Diffusivity, cm ² per sec	Conductivity, cal/(sec)(cm)(C)
200	6.902	0.0711	0.068	0.0333
300	6.885	0.0732	0.073	0.0368
400	6.870	0.0753	0.077	0.0398
500	6.854	0.0774	0.079	0.0419
600	6.835	0.0795	0.080	0.0435
650	6.828	0.0880	0.085	0.0511
700	6.823	0.0960	0.093	0.0609
800	6.819	0.1123	0.108	0.0827
830	6.817	0.1173	0.112	0.0896
900	6.812	0.1097	0.122	0.0911
1000	6.799	0.0982	0.134	0.0895
1100	6.792	0.0907	0.147	0.0906
1200	6.794	0.0872	0.148	0.0877
1300	6.797	0.0877	0.149	0.0888
1400	6.779	0.0922	0.150	0.0938
1500	6.749	0.1007	0.151	0.103
1600	6.709(a)	0.1132	0.152(a)	0.115
1700	6.649(a)	0.1297	0.153(a)	0.132

(a) Extrapolated.

TABLE 26. THERMAL CONDUCTIVITY CALCULATIONS FOR HYDRIDED MATERIALS (EQUILIBRIUM HYDROGEN PRESSURE)

Temperature, C	Density, g per cm ³	Specific Heat, cal/(g) (C)	Diffusivity, cm ² per sec	Conductivity, cal/(sec)(cm)(C)
200	6.003	0.1128	0.098	0.0663
250	5.994	0.1215	0.095	0.0690
300	5.984	0.1302	0.092	0.0715
350	5.974	0.1389	0.089	0.0737
400	5.963	0.1475	0.087	0.0765
450	5.952	0.1562	0.086	0.0800
500	5.940	0.1649	0.085	0.0831
550	5.929	0.1736	0.085	0.0874
600	5.916	0.1823	0.085	0.0915
650	5.903	0.1910	0.085	0.0958

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FIGURE 29. THERMAL CONDUCTIVITY OF DEHYDRIDED MATERIAL

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FIGURE 30. THERMAL CONDUCTIVITY OF HYDRIDED MATERIAL MEASURED
IN BALANCED HYDROGEN PRESSURE

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TABLE 27. THERMAL CONDUCTIVITY CALCULATIONS FOR HYDRIDED MATERIALS (MEASURED IN 1 ATM OF HYDROGEN)

Temperature, C	Density, g per cm ³	Specific Heat, cal/(g)(C)	Diffusivity, cm ² per sec	Conductivity, cal/(sec)(cm)(C)
200	6.008	0.1128	0.086	0.0583
250	5.999	0.1215	0.085	0.0620
300	5.991	0.1302	0.084	0.0655
350	5.981	0.1389	0.083	0.0690
400	5.971	0.1475	0.084	0.0740
450	5.960	0.1562	0.087	0.0810
500	5.948	0.1649	0.089	0.0873
550	5.935	0.1736	0.091	0.0938
600	5.922	0.1823	0.091	0.0982
650	5.910	0.1910	0.090	0.1016
700	5.894	0.1997 ^(a)	0.081	0.0953
750	5.887	0.2084 ^(a)	0.068	0.0834
800	5.888	0.2171 ^(a)	0.061	0.0780
850	5.898	0.2258 ^(a)	0.057	0.0759
900	5.932	0.2345 ^(a)	0.053	0.0737
950	5.991	0.2432 ^(a)	0.050	0.0728
1000	6.060	0.2519 ^(a)	0.046	0.0702

(a) Extrapolated from 660 C.

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FIGURE 31. THERMAL CONDUCTIVITY OF HYDRIDED MATERIAL MEASURED
IN 1 ATM OF HYDROGEN

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FIGURE 32. HEMISPHERICAL TOTAL EMITTANCE APPARATUS

attaining a vacuum of approximately 1×10^{-6} torr. Power was introduced to the specimen assembly and was measured over the measured section between the potential leads by the calibrated meter (6) and the Ballentine electronic voltmeter (7). Temperature of the specimen was measured by sighting, through the window (4), into the center (black-body) cavity of the specimen with an optical pyrometer (8). Since the specimens were thin walled, it was assumed that the internal blackbody temperature was also the external surface temperature of the specimen within experimental error. The hemispherical total emittance (ϵ_{ht}) was then calculated at each equilibrium temperature from the known surface area between the potential leads, the specimen temperature, and the power dissipated from the measured section using the following relationship:

$$\epsilon_{ht} = \frac{IV}{A\sigma(T_s^4 - T_w^4)},$$

where

I = current through specimen, amp

V = potential drop between potential leads, v

A = surface area of specimen between potential leads

σ = Boltzmann's radiation constant

T_s = specimen temperature, K

T_w = chamber wall temperature, K.

The chamber wall temperature was considerably colder than the specimen and was neglected in the calculations since it was found to change the emittance by less than 1 per cent, even at the lowest temperatures measured.

Experimental Results

Dehydrided Fuel

Table 28 shows the experimental data, calculated from the relationship given above, for the dehydrided SNAP-10A fuel having a surface roughness between 6 and 7 μ in. rms. Vacuum readings are given as a control check on the system during measurement. Figure 33 shows a plot of the experimental data. It can be seen that some surface contamination was present initially on the specimen, as indicated by the high values obtained during the first heating. The lower and reproducible data obtained during subsequent cooling and heating cycles indicate that this contamination was removed from the surface, giving consistent data for the bare material. This specimen was later used for initial oxygen-addition tests and, therefore, could not be analyzed after the run to check contamination from the vacuum system. The data themselves, however, and the visual appearance of the specimen, indicate little or no contamination of the specimen during the run. Interpolated data are given in Table 29.

TABLE 28. HEMISPHERICAL TOTAL EMITTANCE OF DEHYDRIDED MATERIAL, SPECIMEN 290-6-F (EXPERIMENTAL DATA)

Temperature, C	Emittance	Vacuum Reading, 10 ⁻⁶ torr	Temperature, C	Emittance	Vacuum Reading, 10 ⁻⁶ torr
<u>First Heating</u>			<u>Second Heating</u>		
800	0.234	2.95	905	0.229	1.70
899	0.270	3.80	996	0.237	1.70
1035	0.287	3.00	1068	0.241	1.80
1139	0.293	2.40	1134	0.251	1.85
1235	0.286	2.55	1211	0.258	1.90
1303	0.287	2.41	1287	0.257	1.95
1379	0.294	2.34	1367	0.263	2.00
1474	0.289	2.40	1446	0.268	2.03
1554	0.291	2.38	1520	0.271	2.10
1603	0.297	2.23	1595	0.276	2.10
1704	0.283	2.21	1623	0.287	2.15
			1655	0.289	2.15
<u>Second Cooling</u>			<u>Second Cooling</u>		
1644	0.279	1.90	1465	0.273	2.01
1582	0.271	1.80	1294	0.256	2.00
1508	0.274	1.75	1109	0.252	1.94
1450	0.269	1.70	933	0.240	1.90
1370	0.266	1.70	795	0.229	1.90
1292	0.256	1.70			
1215	0.251	1.65			
1118	0.247	1.63			
1120	0.245	1.63			
1034	0.237	1.70			
945	0.236	1.70			
850	0.225	1.70			
786	0.225	1.72			

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FIGURE 33. HEMISPHERICAL TOTAL EMITTANCE OF DEHYDRIDED MATERIAL

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TABLE 29. HEMISPHERICAL TOTAL EMITTANCE OF
DEHYDRIDED MATERIAL, SPECIMEN
296-6-F (INTERPOLATED DATA)

Temperature, C	Emittance
800	0.225
900	0.230
1000	0.237
1100	0.245
1200	0.251
1300	0.258
1400	0.265
1500	0.272
1600	0.279
1700	0.286

Dehydrated and Oxidized Fuel

Oxygen additions were made to a second specimen. A valving system, shown behind and below the optical pyrometer in Figure 32, allowed discrete and reproducible quantities of gas to be introduced into the vacuum chamber. All oxygen additions were made at approximately 1500 C where preliminary measurements had indicated a very rapid absorption of oxygen by the material. After oxygen additions were made with the vacuum chamber isolated from the pumping system, the oxygen was allowed to remain in the chamber until thermocouple-gage pressure readings showed zero pressure in the system. The chamber was then opened to the pumping system.

Measurements were made at equilibrium temperatures on cooling from the temperature of oxygen addition to as low as allowed by the pyrometer and then on heating to about 1650 C and back to about 1500 C before the next oxygen addition. **Experimental** data are given in Table 30 and are shown graphically in Figure 34. **Data interpolated** from the curve are given in Table 31.

Postmeasurement analysis of the specimen showed an oxygen content of 3.48 w/o. Since the 3.48 w/o is well below the total oxygen solubility for the material, and since similar conditions prevailed during each oxygen addition, it was assumed that one-sixth of the total oxygen in the specimen was absorbed during each of the six additions.

Dehydrated and Nitrided Fuel

Nitrogen additions and subsequent emittance measurements were made in the same manner as were the oxygen additions described above.

The results obtained are given in Tables 32 and 33 and are shown graphically in Figure 35. Less emittance change was noted with nitrogen additions than with the

TABLE 30. HEMISPHERICAL TOTAL EMITTANCE OF DEHYDRIDED MATERIAL
WITH OXYGEN ADDED, SPECIMEN 290-4-12C
(EXPERIMENTAL DATA)

Temperature, C	Emittance	Vacuum Reading, torr	Temperature, C	Emittance	Vacuum Reading, torr
0.58 w/o Oxygen Added			1.16 w/o Oxygen Added		
1462	0.294	2.4×10^{-6}	1447	0.343	1.9×10^{-6}
1345	0.283	1.9×10^{-6}	1320	0.333	1.8×10^{-6}
1206	0.280	1.8×10^{-6}	1173	0.318	1.8×10^{-6}
1063	0.271	1.8×10^{-6}	1047	0.303	1.8×10^{-6}
880	0.254	1.8×10^{-6}	947	0.298	1.8×10^{-6}
825	0.251	1.8×10^{-6}	826	0.289	1.8×10^{-6}
825	0.251	1.8×10^{-6}	971	0.291	1.8×10^{-6}
892	0.262	1.8×10^{-6}	1105	0.313	1.8×10^{-6}
1006	0.274	1.8×10^{-6}	1237	0.317	1.8×10^{-6}
1170	0.292	1.8×10^{-6}	1390	0.334	1.9×10^{-6}
1249	0.289	1.8×10^{-6}	1529	0.345	1.9×10^{-6}
1347	0.287	1.8×10^{-6}	1676	0.312	2.0×10^{-6}
1551	0.301	1.9×10^{-6}	1501	0.320	1.9×10^{-6}
1672	0.308	1.9×10^{-6}			
1518	0.299	1.8×10^{-6}			
1.74 w/o Oxygen Added			2.32 w/o Oxygen Added		
1401	0.380	1.9×10^{-6}	1420	0.413	1.5×10^{-6}
1254	0.376	1.8×10^{-6}	1243	0.398	1.5×10^{-6}
1107	0.363	1.8×10^{-6}	1096	0.395	1.5×10^{-6}
970	0.349	1.8×10^{-6}	970	0.385	1.4×10^{-6}
823	0.337	1.8×10^{-6}	828	0.359	1.4×10^{-6}
986	0.350	1.8×10^{-6}	965	0.381	1.4×10^{-6}
1113	0.359	1.8×10^{-6}	1105	0.389	1.5×10^{-6}
1241	0.352	1.8×10^{-6}	1239	0.398	1.5×10^{-6}
1360	0.376	1.8×10^{-6}	1413	0.408	1.6×10^{-6}
1521	0.374	1.9×10^{-6}	1518	0.389	1.6×10^{-6}
1666	0.359	2.0×10^{-6}	1676	0.408	2.2×10^{-6}
			1503	0.395	1.6×10^{-6}

TABLE 30. (Continued)

Temperature, C	Emittance	Vacuum Reading, torr	Temperature, C	Emittance	Vacuum Reading, torr
2.90 w/o Oxygen Added			3.48 w/o Oxygen Added		
1399	0.462	1.7×10^{-6}	1337	0.582	1.9×10^{-6}
1244	0.475	1.6×10^{-6}	1221	0.583	1.8×10^{-6}
112	0.467	1.6×10^{-6}	1111	0.582	1.8×10^{-6}
967	0.462	1.6×10^{-6}	948	0.586	1.8×10^{-6}
824	0.435	1.6×10^{-6}	817	0.552	1.8×10^{-6}
968	0.453	1.6×10^{-6}	960	0.574	1.8×10^{-6}
1107	0.462	1.7×10^{-6}	1105	0.586	1.8×10^{-6}
1243	0.463	1.7×10^{-6}	1239	0.580	1.8×10^{-6}
1399	0.458	1.7×10^{-6}	1385	0.571	1.9×10^{-6}
1518	0.457	1.8×10^{-6}	1534	0.532	2.1×10^{-6}
1682	0.449	1.9×10^{-6}	1674	0.524	3.6×10^{-6}
1502	0.442	1.8×10^{-6}	1736	0.519	4.5×10^{-6}
1501	0.434	1.8×10^{-6}	1782	0.529	6.0×10^{-6}
			1841	0.529	1.1×10^{-5}
			1897	0.521	1.35×10^{-5}

FIGURE 34. EMITTANCE VERSUS TEMPERATURE WITH O₂ ADDITIONS TO A DEHYDRATED SNAP-10A FUEL SPECIMEN

Specimen separated at about 1927 C. Final O₂ analysis: 3.45 ± 0.05 w/o.

TABLE 31. HEMISPHERICAL TOTAL EMITTANCE OF DEHYDRIDED MATERIAL WITH OXYGEN ADDED, SPECIMEN 290-4-12C (INTERPOLATED DATA)

Temperature, C	Emittance With Indicated Oxygen Addition(a), w/o					
	0.58	1.16	1.74	2.32	2.90	3.48
825	0.252	0.283	0.338	0.360	0.437	0.555
900	0.260	0.296	0.396	0.370	0.447	0.568
1000	0.268	0.304	0.356	0.380	0.450	0.581
1100	0.276	0.313	0.365	0.390	0.462	0.590
1200	0.285	0.322	0.372	0.400	0.465	0.590
1300	0.293	0.330	0.376	0.405	0.465	0.584
1400	0.301	0.339	0.377	0.406	0.463	0.569
1500	0.310	0.345	0.375	0.406	0.460	0.544
1600	0.318	0.335	0.368	0.404	0.453	0.530
1675	0.325	0.315	0.360	0.400	0.450	--
1700	--	--	--	--	--	0.529
1800	--	--	--	--	--	0.525
1900	--	--	--	--	--	0.523

(a) Each of the six runs is assumed to have taken up one-sixth of the total oxygen present in the specimen after the final run, or one-sixth of 3.48 w/o.

TABLE 32. HEMISPHERICAL TOTAL EMITTANCE OF DEHYDRIDED MATERIAL
WITH NITROGEN ADDED, SPECIMEN 290-4-12B
(EXPERIMENTAL DATA)

Temperature, C	Emittance	Vacuum Reading, torr	Temperature, C	Emittance	Vacuum Reading, torr
Bare Specimen			0.545 w/o Nitrogen Added		
794	0.243	3.2×10^{-6}	1546	0.448	2.1×10^{-6}
897	0.248	3.8×10^{-6}	1434	0.466	1.9×10^{-6}
1048	0.243	3.6×10^{-6}	1318	0.453	1.8×10^{-6}
1171	0.296	3.9×10^{-6}	1215	0.440	1.8×10^{-6}
1266	0.320	3.4×10^{-6}	1091	0.426	1.8×10^{-6}
1381	0.337	3.2×10^{-6}	955	0.397	1.8×10^{-6}
1482	0.371	3.6×10^{-6}	815	0.363	1.7×10^{-6}
1574	0.407	4.7×10^{-6}	962	0.410	1.8×10^{-6}
1686	0.418	1.4×10^{-5}	1113	0.412	1.8×10^{-6}
1563	0.516	2.6×10^{-6}	1225	0.436	1.9×10^{-6}
			1379	0.452	2.0×10^{-6}
			1519	0.457	2.1×10^{-6}
			1679	0.478	4.3×10^{-6}
			1536	0.481	2.1×10^{-6}
0.273 w/o Nitrogen Added			0.818 w/o Nitrogen Added		
1400	0.464	2.0×10^{-6}	1557	0.456	1.9×10^{-6}
1271	0.448	1.8×10^{-6}	1431	0.463	1.7×10^{-6}
1170	0.431	1.8×10^{-6}	1317	0.456	1.6×10^{-6}
1019	0.423	1.8×10^{-6}	1177	0.430	1.6×10^{-6}
913	0.388	1.7×10^{-6}	1058	0.419	1.6×10^{-6}
817	0.361	1.6×10^{-6}	939	0.394	1.6×10^{-6}
965	0.396	1.7×10^{-6}	811	0.364	1.5×10^{-6}
1000		1.8×10^{-6}	953	0.397	1.6×10^{-6}
1240	0.432	1.9×10^{-6}	1100	0.423	1.6×10^{-6}
1373	0.466	2.0×10^{-6}	1245	0.431	1.6×10^{-6}
1528	0.454	2.3×10^{-6}	1395	0.464	1.7×10^{-6}
1686	0.464	5.6×10^{-6}	1524	0.4693	1.8×10^{-6}
1533	0.459	2.3×10^{-6}	1672	0.483	3.2×10^{-6}
			1538	0.481	1.9×10^{-6}

TABLE 32. (Continued)

Temperature, C	Emittance	Vacuum Reading, torr	Temperature, C	Emittance	Vacuum Reading, torr
1.36 w/o Nitrogen Added			2.73 w/o Nitrogen Added		
1564	0.449	1.9×10^{-6}	1522	0.457	1.8×10^{-6}
1420	0.454	1.6×10^{-6}	1398	0.448	1.5×10^{-6}
1280	0.424	1.6×10^{-6}	1247	0.423	1.4×10^{-6}
1108	0.405	1.5×10^{-6}	1107	0.405	1.4×10^{-6}
967	0.379	1.5×10^{-6}	952	0.383	1.35×10^{-6}
836	0.362	1.5×10^{-6}	812	0.338	1.3×10^{-6}
957	0.386	1.5×10^{-6}	964	0.388	1.4×10^{-6}
1103	0.403	1.6×10^{-6}	1097	0.418	1.4×10^{-6}
1228	0.476	1.6×10^{-6}	1246	0.432	1.5×10^{-6}
1378	0.459	1.6×10^{-6}	1378	0.447	1.5×10^{-6}
1523	0.453	1.8×10^{-6}	1511	0.473	
1678	0.475	3.3×10^{-6}	1679	0.499	3.3×10^{-6}
1521	0.474	1.8×10^{-6}	1547	0.477	1.65×10^{-6}
2.38 w/o Nitrogen Added			3.27 w/o Nitrogen Added		
1571	0.435	2.0×10^{-6}	1546	0.462	1.95×10^{-6}
1384	0.434	1.8×10^{-6}	1420	0.442	1.6×10^{-6}
1240	0.403	1.7×10^{-6}	1314	0.435	1.5×10^{-6}
1112	0.388	1.6×10^{-6}	1192	0.452	1.5×10^{-6}
956	0.357	1.6×10^{-6}	1098	0.477	1.5×10^{-6}
809	0.332	1.5×10^{-6}	932	0.394	1.4×10^{-6}
816	0.333	1.4×10^{-6}	870	0.381	1.4×10^{-6}
968	0.363	1.5×10^{-6}	1012	0.414	1.4×10^{-6}
1098	0.382	1.6×10^{-6}	1095	0.436	1.5×10^{-6}
1233	0.390	1.6×10^{-6}	1236	0.460	1.6×10^{-6}
1388	0.428	1.8×10^{-6}	1374		1.6×10^{-6}
1510	0.444	2.0×10^{-6}	1496	0.501	1.6×10^{-6}
1680	0.471	3.0×10^{-6}	1612	0.513	1.6×10^{-6}
1531	0.457	1.6×10^{-6}	1674	0.512	3.4×10^{-6}
			1546	0.488	1.7×10^{-6}
			1708	0.511	4.2×10^{-6}
			1767	0.510	1.0×10^{-5}
			1802	0.525	2.1×10^{-5}
			1848	0.537	4.0×10^{-5}
			1905	0.541	6.5×10^{-5}

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FIGURE 35. EMITTANCE VERSUS TEMPERATURE WITH N₂ ADDITIONS TO A DEHYDRATED SNAP-10A FUEL SPECIMEN

Surface roughness: 10 μ in. rms. Final N₂ analysis: 3.27 w/o.

oxygen additions. The total postanalysis nitrogen content of 3.27 w/o was again divided equally among the individual additions as was done for the oxygen additions. It was apparent from the data that reversals in the emittance characteristics occurred. The emittance was seen to increase initially with small additions of nitrogen (0.27 w/o), to remain essentially constant with further additions up to 1.36 w/o, to drop then slightly with further additions up to about 2.38 w/o, and, finally, to increase again with more additions. The reversals observed in the emittance data appear to be related to changes occurring in the material, i. e., diffusion of nitrogen into the specimen, buildup of a zirconium nitride layer on the surface of the specimen, etc.

TABLE 33. HEMISPHERICAL TOTAL EMITTANCE OF DEHYDRIDED MATERIAL WITH NITROGEN ADDED, SPECIMEN 290-4-12B (INTERPOLATED DATA)

Temperature, C	Emittance With Indicated Nitrogen Addition, w/o			
	None	1.36	2.38	3.27
800	0.235	0.360	0.325	0.367
900	0.255	0.380	0.345	0.375
1000	0.275	0.400	0.360	0.402
1100	0.295	0.415	0.377	0.428
1200	0.314	0.450	0.395	0.450
1300	0.332	0.465	0.410	0.466
1400	0.351	0.470	0.425	0.482
1500	0.370	0.470	0.490	0.492
1600	0.391	0.470	0.504	0.508
1700	0.412	0.470	0.516	0.517
1800	--	--	--	0.528
1900	--	--	--	0.536

Note: Surface roughness - 10 μ in. rms.

The major probable error in the emittance measurements is the uncertainty in reading the temperatures with the optical pyrometer. Because all temperature data were taken by the same person, it is believed that the maximum relative error for these measurements is ± 3 per cent. The data are believed in absolute error by less than ± 8 per cent. It must be remembered, however, that the measurements are reproducible only under similar conditions of surface roughness, surface contamination, oxide or nitride coating thickness, and tenacity of the coatings.

SOLUBILITY OF CARBON, NITROGEN, AND
OXYGEN IN MOLTEN SNAP-10A FUEL

To define what maximum levels of impurities might be encountered during the high-temperature thermal-property studies and to aid in the interpretation of phenomena which occur during re-entry, the solubility limits of carbon, nitrogen, and oxygen in the molten fuel alloy were determined. In the case of carbon, the molten fuel was directly saturated with the element at various fixed temperatures. The composition of the fuel alloy was then determined. For nitrogen and oxygen, it proved more expedient to prepare fuel alloys with known additions of these elements and then to determine the melting points (liquidus temperatures) of the resulting alloys. Descriptions of initial experiments and of improved techniques that were developed in the course of the study follow.

Carbon Solubility

The solubilities of carbon in a molten zirconium-10 w/o uranium alloy at various temperatures were determined by heating the alloys to various temperatures in graphite crucibles, holding them at those temperatures until equilibrium amounts of carbon were dissolved, and quenching them rapidly to room temperature. The resulting alloys were then examined metallographically and analyzed chemically to determine the solubility limits.

Five 25-g melts of the zirconium-10 w/o uranium alloy (degassed) were induction melted in 3/4-in.-ID halogen-purified graphite crucibles. The metal was contained in the crucibles during all heat-ups. Melt 1 was held at temperature for 60 min, after which the power was turned off and the sample solidified immediately with the temperature dropping approximately 100 C per min. A similar procedure was conducted for Melt 2 (only in this case the alloy was held at a higher temperature for 30 min). The resulting ingots were then sectioned; Figure 36 shows the microstructure of the Melt 2 ingot. Two distinct regions of carbide precipitation can be seen in this microstructure. The lower region shows zirconium solid solution with carbides precipitated during cooling; the upper region contains a larger amount of carbides, many of which apparently were caused by slight fluctuations of temperature or a small temperature gradient at the melt surface during holding. Samples taken from the tops and bottoms of Melts 1, 4, and 5 and the bottoms only of Melts 2 and 3 were analyzed. The results are listed in Table 34. Values listed for bottom samples are considered to be saturation values for the temperatures of the melts. To avoid the excess of carbides in later tests, certain precautions were taken, i. e., the holding time for a melt was reduced to 15 min. Microstructures of the resulting ingots from Melts 3, 4, and 5 displayed no distinct regions. Figure 37 shows the ingot from Melt 3 for which the growth of carbides in the melt was reduced to about a minimum for this technique.

Approx. 5X

RM29768

FIGURE 36. MICROSTRUCTURE OF MELT 2 INGOT IN
CARBON-SOLUBILITY STUDY

Approx. 4X

RM30447

FIGURE 37. UNIFORM MICROSTRUCTURE OF MELT 3 INGOT IN
CARBON-SOLUBILITY STUDY

TABLE 34. SUMMARY OF CARBON SOLUBILITIES IN MOLTEN ZIRCONIUM-10 w/o URANIUM MELTS

Carbon Melt	Temperature, C	Time at Temperature, min	Carbon, w/o	
			Top	Bottom
1	1850	60	1.41 ^(a)	0.15
5	1925	15	0.58	0.29
2	2000	30	--	0.49
4	2100	15	0.89	0.63
3	2200	15	--	0.74

(a) Large carbide particles present.

The solubility values are plotted in Figure 38. The results are compared with results of a similar study of unalloyed zirconium made in 1950 by the Bureau of Mines⁽⁸⁾. The latter's time at temperature was 25 min. It is seen that the values are of the same order of magnitude. In the practical application of the solubility data, it should be kept in mind that unless a melt is entirely isothermal, a two-phase system will be produced with reduction of fluidity and eventual solidification of the melt. For this reason, the lower-carbon-content values are considered most accurate and these were the values plotted in Figure 38. The distribution of uranium in the experimental ingots was not determined.

Nitrogen Solubility

Attempts were made to saturate alloy melts with nitrogen by dissolving ZrN from crucibles of this material. Because commercial ZrN crucibles were not available, ZrN powder (-80 mesh with 50 per cent -200 mesh) was purchased from Hummel Chemical Company and was tamped into crucible form in molybdenum-lined graphite heaters and fired therein. The initially prepared crucibles showed over 20 per cent shrinkage upon heating to 1700 C. To overcome this, ZrN powder was pressed and prefired in vacuum in the heater assembly. This material was then crushed, sieved, and mixed with some unfired ZrN, and the mixture was hand-tamped into crucible form as before. A binder of Parawax in carbon tetrachloride was used for green strength. This was baked out at 300 C in vacuum before final firing. Although shrinkage of about 10 per cent occurred on firing to 1700 C, the resulting crucibles were uncracked and usable.

The initial melt of the fuel alloy in the nitride crucible at 2000 C is shown on the right in Figure 39. The majority of fuel alloy disappeared from inside the crucible during test and the alloy appeared to coat the entire crucible. A duplicate melt was made in the crucible shown on the left in Figure 39 with a similar result. In both cases, a large amount of the alloy was found outside the crucible, but no evidence of a

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FIGURE 38. CARBON SOLUBILITY IN LIQUID ZIRCONIUM-10 w/o URANIUM ALLOY

CONFIDENTIAL

FIGURE 39. ZIRCONIUM NITRIDE
CRUCIBLES RUN IN
NITROGEN-SOLUBILITY
STUDIES

RM31116

750X

RM31312

a. Before Test

750X

RM31120

b. After Test

FIGURE 40. MICROSTRUCTURE OF
ZIRCONIUM NITRIDE
CRUCIBLE

hole or crack in the crucible which would permit leakage was noted. Figures 40a and 40b show the microstructure of the second ZrN crucible before and after test. It can be seen that the crucible initially consisted solely of ZrN and contained a considerable amount of porosity. In Figure 40a penetration of metal into the crucible can be seen; the white areas in the metal between the ZrN grains appear to be α -zirconium formed upon initial cooling, while the surrounding gray metal probably is $\alpha + \beta$ zirconium formed upon complete solidification of the permeating metal.

It appears that the alloy must have had remarkable fluidity to have permeated the crucible walls. Also superior wetting power and surface diffusion of the metal in the nitride appears to be demonstrated. In view of this interesting but bothersome phenomenon and of the fact that solution of zirconium from the ZrN crucibles reduces the overall uranium content of the alloy, a different technique for the solubility study was sought.

A second approach was taken in which a series of fuel alloys containing 1 to 4 w/o additions of nitrogen was prepared by arc melting. The gas was measured manometrically and was added directly to the arc chamber during melting. Alloys were exposed to pressures up to 400 mm of nitrogen in a 2600-cc chamber. About 95 per cent of the gas additions were dissolved in a single melting. After the final gas additions were made, remelting permitted all residual gas to be dissolved. Alloy buttons were turned over and remelted four times after the final gas addition. Attempts were then made to determine the melting points of the fuel alloys with the known gas contents. Figure 41 shows the microstructures of these alloys.

Various-melting-point determination techniques were considered. That of Deardorff and Hayes⁽⁹⁾ was chosen as most promising. Specimen pedestals 1/4-in. square and 1 in. long with a cap piece in the form of a 1/4-in. cube were machined from the button ingots. A sight hole 0.035-in. in diameter and 3/16 in. deep was drilled into the cap. The specimen and cap with the sight hole on top was supported vertically on a molybdenum wire and heated by a tantalum susceptor in an induction furnace. The specimen was placed low in the susceptor so that the bottom of the pedestal was cooler than the cap. Various arrangements were tried for thermally insulating the susceptor and for supporting the pedestal. Eutectic formation between the alloy and the molybdenum support always occurred allowing contact between the specimen and the susceptor. The tantalum susceptor was suspended by molybdenum wires inside a cylindrical Vycor radiation shield. A calibration run was made with a zirconium-metal specimen. The appearance of liquid viewed in the sight hole of the cap gave a melting point in excellent agreement with the literature value. A specimen of the fuel alloy containing 3 per cent nitrogen gave the first appearance of liquid at 1865 C. This temperature apparently is that of the solidus isotherm for the region corresponding to liquid + α -zirconium in the zirconium-nitrogen phase diagram. The temperature was raised further to determine the liquidus temperature as evidenced by a rounding of specimen edges and corners. The liquidus temperature was not clearly observed because the softened sample repeatedly came in contact with the susceptor. Use of this technique was suspended while a third, simpler approach was evaluated and finally was adopted.

In this third approach, a cube-shaped alloy sample was dropped into a graphite crucible preheated to a known constant temperature. Specimens reached constant temperature in about 10 sec. They were held at this temperature for 2 min and

750X

RM36857

Dehydrated SNAP-10A Fuel
with 1 w/o Nitrogen

750X

RM26854

Dehydrated SNAP-10A Fuel
with 2 w/o Nitrogen

750X

RM36677

Dehydrated SNAP-10A Fuel
with 3 w/o Nitrogen

750X

RM36858

Dehydrated SNAP-10A Fuel
with 4 w/o Nitrogen

FIGURE 41. MICROSTRUCTURES OF AS-CAST, DEHYDRATED SNAP-10A FUEL ALLOYS WITH ADDITIONS OF NITROGEN

then were rapidly cooled to room temperature. Little or no reaction between unmelted specimens and graphite was seen and only very small reaction zones were noted between the molten alloys and graphite. Apparently a thin layer of ZrC was formed between the liquid alloys and graphite which prevented excessive carbon contamination of the molten alloys. A typical interface between an alloy molten during test and graphite is shown in Figure 42. In the initial trial, three specimens of the fuel alloy containing 3 per cent nitrogen were examined utilizing this technique. At 2265 C an initial specimen retained sharp corners; at 2315 C the second specimen displayed slight distortion; at 2365 C the third specimen melted down completely leaving only the impression of a slight residue on the melt surface.

The latter temperature was selected as the "best" estimate of the melting point. Melting points were determined for dehydrided fuel alloy arc melted with additions of 0.0, 1.0, 2.0, 3.0, and 4.0 w/o nitrogen. A complete record of the experimental heatings of these alloys is given in Table 35. The nitrogen solubilities and corresponding melting points are plotted in Figure 43.

TABLE 35. MELTING POINT DETERMINATIONS OF FUEL ALLOYS
CONTAINING NITROGEN ADDITIONS

Nitrogen Content, w/o	Alloy was Solid at:	Alloy was Molten at:	Best Estimate of Melting Point
0.0		1850 C	1850 C
1.0	2115 C	2255 C (Complete) 2175 C (Slight residue)	2175 C
2.0		2235 C (Slight residue)	2235 C
3.0	2265 C 2315 C	2365 C (Slight residue)	2365 C
4.0	2455 C (Surface appeared molten)		2455 C

150X

RM36862

FIGURE 42. FORMATION OF A THIN ZIRCONIUM CARBIDE LAYER
BETWEEN THE MOLTEN FUEL ALLOY AND THE
GRAPHITE CRUCIBLE DURING A TYPICAL TEST

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

FIGURE 43. NITROGEN SOLUBILITY IN LIQUID ZIRCONIUM-10 w/o URANIUM ALLOY

It was noted that the experimental melting points of the fuel alloy-nitrogen alloys are considerably higher than the estimated literature values for the unalloyed zirconium-nitrogen system. For example, the melting point of the 3 w/o nitrogen alloy was 2365 C compared to 2075 C as indicated in the literature.⁽¹⁰⁾ The presence of 10 w/o uranium in the fuel alloy is not expected to be responsible for this difference since uranium would probably lower the melting point somewhat. The apparently low estimated liquidus temperature by Domagala, et al., may have been influenced by a eutectic formation between the specimen and the tungsten wire suspension used in their experiments. Such an action was postulated by Deardorff and Hayes for McPherson's melting-point determination for hafnium.

The melting points of the present study were obtained with a Pyro optical pyrometer sighted on the inside bottom of the empty graphite crucible. This brightness temperature was corrected to true temperature by adding 65 C, the correction determined by duplicate measurements of the melting point (1845 C) of pure zirconium. As a check at higher temperatures, comparison readings were taken with the Pyro and a Leeds and Northrup optical pyrometer. The latter is a reference pyrometer calibrated by the National Bureau of Standards. The pyrometers agreed within ± 10 C at 2050 C and ± 20 C at 2450 C.

As a further check on the questioned literature melting-point values, a 4 w/o addition of nitrogen was made to pure zirconium. The melting point of this material by Battelle's experimental techniques was 2475 C compared to the estimated literature value of 2200 C.

Oxygen Solubility

In early experiments the oxygen solubility in a zirconium-10 w/o uranium alloy (degassed) was determined by equilibrating melts of the alloy held in coarse-grain stabilized-zirconia crucibles. The crucibles were 1 by 1 in. with 1/8-in. walls. The crucible containing about 25 g of alloy was induction heated by a sheet molybdenum susceptor nested in ZrO_2 granular packing in a fused silica thimble. A melt made at 1879 C was held 5 min while dissolving oxide from the crucible. The ingot appeared uniform except for a 1/32-in. layer on the side and bottom which appeared to be uranium rejected from the melt as oxygen was absorbed. However, analyses of this region and of the body of the ingot showed no marked change from the original 10 w/o uranium. Duplicate samples of the 1870 C melt averaged 1.11 ± 0.03 w/o oxygen by the vacuum-fusion platinum-bath technique. For unalloyed zirconium this oxygen solubility would correspond to that for about 1925 C according to the zirconium-oxygen phase diagram.

Another oxygen solubility melt attempted at 2000 C spattered and then formed a bridge about 3/8-in. up from the (empty) bottom of the crucible. The metal was hard and brittle and two samples analyzed 3.56 and 5.64 w/o oxygen. Such results for the unalloyed zirconium-oxygen system would correspond to a temperature slightly under 1975 C. An attempt to remelt partially oxygen-saturated alloy in a previously vacuum-fired crucible failed when the crucible cracked. It appears that some of the stabilizing oxide, e.g., CaO , was lost from the refractory during the prior vacuum heating.

Because the technique for determining melting points of arc-melted alloys with known nitrogen additions was so successful and because in this technique the problem of refractories was avoided, this same technique was applied to the oxygen alloys. Arc melts of degassed fuel alloy with oxygen additions of 1.5, 4.0, 6.0, 11.0, 13.0, and 17.0 w/o oxygen were prepared. The microstructures of these alloys in the as-cast condition are shown in Figure 44. **The results of the melting-point determinations on these alloys are summarized in Table 36 and the melting points are plotted against reciprocal temperature in Figure 45.**

The melting points agree with literature values for the zirconium-oxygen system⁽¹¹⁾, except at 6.0 and 11.0 w/o oxygen where the melting points of the fuel alloy appear to be about 100 C higher. No line was drawn through the solubility values for oxygen since they do not represent a single phase relationship such as that of 2.0 to 4.0 w/o nitrogen solutions.

DISCUSSION OF RESULTS

Pre- and Postmeasurement Examinations

Particular emphasis in the thermal-properties studies was placed upon the quality control of measurements and materials. Detailed examination of test specimens before and after all thermal-property measurements were made to ensure that the properties measured were those of materials with known compositions and structures.

Specimens before and after measurements were examined macroscopically for cracks, surface imperfections, discoloration, etc.; metallographically for microstructural variations and changes; and chemically for impurity content. In general, the results of these examinations showed that the specimens measured were uniform in composition and structure and that little contamination of the specimens occurred during measurements.

The results of chemical analyses of specimens before and after measurements are listed in Tables 37 and 38. **From these results, it can be seen that all hydride specimens, with the exception of diffusivity specimens, retained their hydrogen fairly adequately. The hydride diffusivity specimens tested in balanced hydrogen pressures lost approximately 0.18 to 0.27 w/o hydrogen during measurements.**

Several hydride and dehydrided specimens showed increases in oxygen content during tests. **Surface discoloration was also noted on specimens showing large increases in oxygen content. In most cases, specimens showing oxygen contamination were set aside and new specimens were run.**

Little change in the microstructure of hydride fuel specimens occurred during measurements (see Figures 47 through 49). **The microstructures of all specimens examined showed the presence of the epsilon-hydride phase. Somewhat more notable changes were apparent in the microstructures of dehydrided materials after**

150X

RM36916

Dehydrided SNAP-10A Fuel
with 2 w/o Oxygen

150X

RM36917

Dehydrided SNAP-10A Fuel
with 4 w/o Oxygen

150X

RM36921

Dehydrided SNAP-10A Fuel
with 6 w/o Oxygen

150X

RM36919

Dehydrided SNAP-10A Fuel
with 11 w/o Oxygen

FIGURE 44. MICROSTRUCTURES OF AS-CAST, DEHYDRIDED SNAP-10A

150X

RM36920

Dehydried SNAP-10A Fuel
with 13 w/o Oxygen

150X

Dehydried SNAP-10A Fuel
with 17 w/o Oxygen

FIGURE 44. (CONTINUED)

TABLE 36. MELTING POINT DETERMINATIONS OF FUEL ALLOYS
CONTAINING OXYGEN ADDITIONS

Oxygen Content, w/o	Alloy was Solid at:	Alloy was Molten at:	Best Estimate of Melting Point
0.0		1850 C	1850 C
1.5		2025 C (Complete after 23 seconds)	1925 C
		1965 C (Complete after 22 seconds)	
		1925 C (Complete after 140 seconds)	
4.0	1925 C	2055 C (Complete after 25 seconds)	1990 C
	1965 C	2015 C (Complete after 55 seconds)	
6.0	2035 C	2165 C (Complete)	2060 C
		2115 C (Complete after 18 seconds)	
		2080 C (Complete after 45 seconds)	
11.0	1965 C	2085 C	2085 C
	2015 C	(Complete after 90 seconds)	
13.0		2075 C (Complete after 60 seconds)	2075 C
		2075 C (Complete after 90 seconds)	
17.0	2220 C	2255 C (Complete after 70 seconds)	2255 C

NOTE: Specimens prepared from arc-melted and alloyed, dehydrided SNAP-10A fuel material.

FIGURE 45. OXYGEN SOLUBILITY IN LIQUID ZIRCONIUM-10 w/o URANIUM ALLOY

TABLE 37. CHEMICAL ANALYSES OF FUEL SPECIMENS BEFORE AND AFTER MEASUREMENTS

Thermal Diffusivity

Before (Specimen 4T):

Hydrogen - 1.82 w/o
Oxygen - 914 ppm $\pm$ 20 ppm

After (Specimen 4I - Balanced H₂ Pressure):

Hydrogen - 1.64 and 1.54 w/o $\pm$ 0.02 w/o
Oxygen - 1360 ppm $\pm$ 15 ppm

(Specimen 4K - Balanced H₂ Pressure-Leakage):

Hydrogen - 1.55 w/o $\pm$ 0.02 w/o
Oxygen - 5040 ppm $\pm$ 15 ppm

(Specimen 4B - Dynamic Vacuum):

Hydrogen - 15 ppm $\pm$ 5 ppm
Oxygen - 1123 ppm $\pm$ 20 ppm

(Specimen 4C - Dynamic Vacuum):

Hydrogen - <10 ppm
Oxygen - 1440 ppm $\pm$ 15 ppm

Specific HeatBefore (Specimen 7L):

Hydrogen - 1.82 w/o
Oxygen - 861 ppm $\pm$ 20 ppm

After (Specimen 7B):

Hydrogen - 1.54 w/o and 1.78 w/o $\pm$ 0.2 w/o
Oxygen - 654 ppm $\pm$ 15 ppm

Thermal ExpansionBefore (Specimen 1E):

Hydrogen - 1.78 w/o $\pm$ 0.04 w/o
Oxygen - 955 ppm $\pm$ 30 ppm

After (Specimen 1B - Longitudinal):

Hydrogen - 1.83 w/o $\pm$ 0.01 w/o
Oxygen - 996 ppm $\pm$ 15 ppm
Nitrogen - 6 ppm $\pm$ 3 ppm

(Specimen 2B - Transverse):

Hydrogen - 1.75 w/o $\pm$ 0.01 w/o
Oxygen - 920 ppm $\pm$ 15 ppm
Nitrogen - 20 ppm $\pm$ 3 ppm

TABLE 38. CHEMICAL ANALYSES OF DEHYDRIDED FUEL SPECIMENS
BEFORE AND AFTER MEASUREMENTS

Thermal DiffusivityBefore (Specimen 8Y):Hydrogen - 48 ppm $\pm$ 2 ppm
Oxygen - 809 ppm $\pm$ 15 ppmAfter (Specimen 8D):Oxygen - 2700 ppm $\pm$ 35 ppm
Nitrogen - 216 ppm $\pm$ 5 ppm

(Specimen 8L - Leakage):

Oxygen - 8760 ppm

Specific HeatBefore (Specimen 9N):Hydrogen - 65 ppm $\pm$ 2 ppm
Oxygen - 914 ppm $\pm$ 20 ppmAfter (Specimen 9A - Heated in Graphite and Evaluated in Solid Region):Oxygen - 865 ppm $\pm$ 20 ppm
Carbon - 0.14 w/o

(Specimen 9C - Heated in Graphite and Evaluated in Liquid Region):

Oxygen - 865 ppm $\pm$ 15 ppm
Carbon - 0.37 w/oThermal ExpansionBefore (Specimen 10B):Hydrogen - 40 ppm $\pm$ 0.5 ppm
Oxygen - 988 ppm $\pm$ 20 ppm
Nitrogen - 75 ppm $\pm$ 3 ppmAfter (Specimen 10A - Longitudinal):Hydrogen - 11 ppm $\pm$ 1 ppm
Oxygen - 1450 ppm $\pm$ 25 ppm
Nitrogen - 38 ppm $\pm$ 5 ppm

(Specimen 11A - Transverse):

Hydrogen - 13 ppm $\pm$ 1 ppm
Oxygen - 769 ppm $\pm$ 10 ppm
Nitrogen - 6 ppm $\pm$ 2 ppm

TABLE 38. (Continued)

Total Hemispherical EmittanceBefore (Specimen 12AA):Hydrogen - 40 ppm $\pm$ 0.5 ppmOxygen - 859 ppm $\pm$ 15 ppmNitrogen - 65 ppm $\pm$ 3 ppmAfter (Specimen 12 B - Run in Nitrogen):

Nitrogen - 3.27 w/o

(Specimen 12 C - Run in Oxygen):

Oxygen - 3.48 w/o $\pm$ 0.05 w/o

(Specimen 12D - Run in Vacuum after Contamination):

Oxygen - 2085 ppm $\pm$ 20 ppmNitrogen - 16 ppm $\pm$ 1 ppm

FIGURE 46. TYPICAL MICROSTRUCTURE OF AS-RECEIVED SNAP-10A FUEL MATERIAL

FIGURE 47. MICROSTRUCTURE OF SNAP-10A DIFFUSIVITY SPECIMEN 4K AFTER MEASUREMENTS

250X

RM32605

FIGURE 48. MICROSTRUCTURE OF SNAP-10A TRANSVERSE THERMAL EXPANSION SPECIMEN 2B AFTER MEASUREMENTS

250X

RM38503

FIGURE 49. MICROSTRUCTURE OF SNAP-10A SPECIFIC HEAT SPECIMEN 7B AFTER MEASUREMENTS

measurements (Figures 50 through 54). Among these were: (1) a coalescence and spherodization of void spaces within specimens which were exposed to high temperatures; (2) a change in the etching behavior of specific heat specimens which had dissolved small amounts of carbon from the graphite liners used during measurements; and (3) a reduction in the grain size and a slight change in the nature of the alpha phase in the diffusivity specimens. The increase in oxygen content occurring in the latter material may have been responsible for some of the microstructural changes noted. Figure 55 shows the microstructures of dehydrided emittance specimens after tests. The effects of nitrogen and oxygen additions on the microstructure of the fuel are readily apparent in these photomicrographs.

Dehydriding of SNAP-10A Fuels

After a somewhat detailed investigation of the variables involved in dehydriding SNAP-10A fuel rods, a procedure consisting of placing sections of as-received fuel into a furnace at room temperature, heating the rods with the furnace to a temperature of 930 C, holding the rods at 930 C for 5 to 10 hr, and furnace cooling the rods to room temperature was selected and was used for the preparation of the dehydrided fuel specimens utilized in thermal-property measurements. Normally, a time of 3 to 4 hr was necessary to heat rods in the furnace from room temperature to 930 C. Hydrogen was pumped continuously from the furnace chamber during heating and holding of the fuel at 930 C. At temperatures approaching 930 C, audible snapping occurred in the fuel when essentially all hydrogen given off was pumped from the chamber. The snapping sounds were found to be directly associated with large-scale internal cracking of the fuel. By maintaining a hydrogen back pressure of about 1/2 atm, however, no snapping was noted and the resulting material contained a uniform distribution of small, irregularly shaped voids no more than 2 or 3 mils long instead of large radial cracks which were seen in specimens dehydrided with no back pressure. A back pressure therefore was used in the dehydriding of all material for thermal-property measurements with the exception of several samples which were placed directly into a hot furnace for a more rapid dehydriding. Diffusivity specimens were taken from this latter material and measurements conducted on these specimens were compared with measurements on the more slowly dehydrided materials.

Results of Thermal-Property Measurements

Enthalpy, Specific Heat, Heat of Fusion, and Heat of Phase Change

Enthalpy measurements made in the Bunsen ice calorimeter are believed to be in error by less than 0.5 per cent for measurements below about 700 C and by less than 1 per cent for measurements above 700 C. Specific heats, calculated from the enthalpy data, are believed to be in error by less than 2 per cent for values below about 700 C and by less than 5 per cent for values above 700 C.

150X

RM31250

FIGURE 50. TYPICAL MICROSTRUCTURE OF DEHYDRIDED SNAP-10A FUEL MATERIAL BEFORE MEASUREMENTS

150X

RM31796

FIGURE 51. MICROSTRUCTURE OF DEHYDRIDED SNAP-10A DIFFUSIVITY SPECIMEN 8L AFTER MEASUREMENTS

150X

RM32609

FIGURE 52. MICROSTRUCTURE OF DEHYDRIDED SNAP-10A LONGITUDINAL THERMAL EXPANSION SPECIMEN 10A AFTER MEASUREMENT

150X

FIGURE 53. MICROSTRUCTURE OF DEHYDRIDED SNAP-10A SPECIFIC HEAT SPECIMEN 9A AFTER DROPS FROM TEMPERATURES UP TO 730 C

150X

FIGURE 54. MICROSTRUCTURE OF DEHYDRIDED SNAP-10A SPECIFIC HEAT SPECIMEN 9C AFTER A DROP FROM 2000 C (MOLTEN REGION)

150X

RM36915

a. Run in Nitrogen Atmosphere

150X

RM32608

b. Run in Oxygen Atmosphere

FIGURE 55. MICROSTRUCTURES OF DEHYDRIDED SNAP-10A EMITTANCE SPECIMENS

The heat of phase change reported for the $\alpha + \delta \rightleftharpoons \alpha + \beta$ transformation as 3.1 cal per g at 595 C is believed to be in error by less than ± 5 per cent, although the temperature of transformation might lie anywhere between 590 and 625 C, as indicated by the measured data. The heat of fusion reported as 29.1 cal per g is believed to be in error by less than ± 10 per cent. Both the solidus and liquidus temperatures, 1810 ± 25 C and 1860 ± 25 C, respectively, are believed to be valued somewhat toward the low side, on the basis of experimental data, and probably lie within the ± 25 C error band given.

Thermal Diffusivity

The thermal-diffusivity values for the hydrided material measured in a hydrogen atmosphere balanced to the dissociation pressure curve for the material should correspond closely with values for the same material measured in 1 atm of hydrogen up to near a dissociation pressure of 1 atm, or about 650 C. Although the values did correspond within ± 10 per cent, the slopes for the curves for the two measurements were different. This difference in the slopes has led to a further evaluation of the data and to additional measurements in order to remove possible uncertainties.

Evaluations of the method, data, and procedures used pointed to the following three areas of possible variation in the final data:

- (1) Possible variations due to the method and the apparatus
- (2) Changes in a specimen during measurement
- (3) Differences between starting specimens.

These three possibilities are discussed in detail below.

Evaluation of the Method and Apparatus Used. The following points were considered:

- (1) A check of the apparatus was made before measurements on the Sandia materials were started. A specimen of Type 347 stainless steel was measured at temperatures from 300 to 1400 C. This specimen was fabricated from material used as a secondary thermal conductivity standard. The measurement agreed within 5 per cent over the above temperature range both with data calculated from known thermal conductivity, specific heat, and density values and with diffusivity values from the literature.
- (2) Measurements of three specimens of the dehydrided material (one SNAP-8 specimen and two SNAP-10A specimens) produced data within ± 5 per cent of each other.

The excellent sensitivity of the measurements was indicated by the abrupt changes in slope or apparent discontinuities of the plotted data

at temperatures where phase diagrams indicated changes of phase in the uranium-zirconium system.

- (3) During measurements on the dehydrided specimens and the hydrided specimen under dynamic vacuum, all systems appeared to be working well and the data were believed to be in error by less than ± 5 per cent.
- (4) Prior to measurements of the hydrided material in a hydrogen atmosphere, in balanced hydrogen pressure atmosphere and under 1 atm of hydrogen, the specimen furnace was changed from a tantalum tube furnace to a molybdenum-wound ceramic tube furnace, and a graphite specimen holder was used in place of the normally used boron nitride specimen holder.
- (5) Measurement on a Type 347 stainless steel specimen was made after the hydrogen atmosphere runs. This measurement indicated values in agreement with the comparison curve above 350 C but showed a linear negative deviation to about 25 per cent at 200 C. Subsequent measurements with the apparatus showed that the room-temperature resistance of the lead-sulfide detector had changed from its original value of approximately 0.4 megohms to about 1.2 megohm. It is possible that this may have affected the time response of the detector and, therefore, the low-temperature results for the balanced hydrogen and the 1-atm hydrogen measurements. It is believed, however, that both measurements would have been changed similarly (which did not occur) should the resistance change have been the sole problem.
- (6) Laser-specimen-detector alignment may have caused difficulties during one or both of the hydrogen runs. Alignment was checked carefully, however, when specimens were changed and, therefore, was not considered as a probable difficulty.
- (7) Distribution of energy over the laser beam may have changed during one or several measurements. Such changes have been noted. Measurements in the past have indicated, however, that these changes produce little, if any, effect in resulting data.

Evaluation of Changes Which Might Occur in a Specimen During Measurement.

Recent measurements have indicated that the thermocouples may not have been functioning properly during measurements in hydrogen. Incorrect temperatures would have resulted in improper balancing of hydrogen pressure, and data obtained would not have been on material of known composition but on material of some other composition. A difference of 10 C, for example, would have made a difference of about 12 cm of mercury pressure near 650 C.

In recent measurements, it was shown that a 10 per cent change in diffusivity occurs when a fuel specimen is held at approximately 500 C and the hydrogen pressure is changed from 29 to 10 cm of mercury pressure. In all cases, changes in diffusivity occurred very rapidly and reached a new equilibrium value within 3 min after a pressure change.

Tenacious oxide layers appearing on the surfaces of specimens may have been responsible for differences noted in results. It is postulated that, in some cases, the laser beam may have removed the oxide layer from one side of the specimen allowing the hydrogen pressure to change rapidly. Some specimens may have possessed heavier or more tenacious layers than others thereby suppressing the hydrogen pressure change and possibly creating a measurable difference in diffusivity.

Evaluation of Possible Variations Between Specimens. Although specimens were well characterized before and after machining and after measurements, specimen histories during storage were uncertain. It is possible that changes may have occurred in diffusivity specimens during storage prior to measurements.

It is believed that the relative differences shown between specimens measured in 1 atm of hydrogen and a balanced hydrogen atmosphere are actual measured differences rather than measurement errors.

Slopes of the curves below 350 C for the hydrided materials run in hydrogen have been changed, using the Type 347 stainless steel calibration curve to obtain the corrected diffusivity curve.

Linear-Thermal-Expansion

The linear-thermal-expansion data are believed to be in error by less than ± 2 per cent for the measurements made in the quartz-structure dilatometer, and by less than ± 3 per cent for measurements made in the sapphire-structure dilatometer.

Only slight coalescence or densification of hydrided specimens was noted after thermal cycling. Coalescence of the dehydrided material was quite pronounced and quite dependent upon maximum temperature of the run. The specimens measured to 1000 C and cooled showed a decrease in length of 0.16 per cent for the longitudinal direction and 0.26 per cent for the transverse direction, as shown in Figure 15. The longitudinal specimen run to 1680 C, however, as shown in Figure 16, coalesced 0.94 per cent upon cooling to room temperature.

Density

The bulk densities measured at room temperature showed variations between specimens as pointed out earlier in this report. The hydrided specimens varied statistically only ± 0.3 per cent from an average value of 6.030 g per cm³ or 86.6 per cent of theoretical density. The dehydrided specimens, however, varied from an average of 6.897 g per cm³ for the smaller specimens (diffusivity) to 6.710 g per cm³ for the larger specimens (expansion).

Elevated-temperature densities were calculated from the room-temperature density, and the volume expansion calculated for the particular material. Densities were calculated in per cent density change in order to be useful in calculating thermal conductivities for a thermal-diffusivity specimen of measured room-temperature density.

Thermal Conductivity

Thermal-conductivity values at a given temperature were calculated by multiplying the specific heat times the density times the thermal diffusivity at that temperature

The resulting thermal conductivity values are believed to be in error by no more than ± 10 per cent, assuming errors of each measurement to be additive.

Hemispherical Total Emittance

The emittance data are believed to be in error by less than ± 8 per cent, absolute, and by less than ± 3 per cent, relative. **However, the measurements are reproducible** only under similar conditions of surface roughness, surface contamination, coating thickness, and tenacity of the coatings.

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APPENDIX

DESCRIPTION OF STARTING MATERIALS AND
SPECIMENS USED IN MEASUREMENTS

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APPENDIX

DESCRIPTION OF STARTING MATERIALS AND SPECIMENS USED IN MEASUREMENTS

Approximately 300 in. of 5/8-in.-diameter SNAP-8 fuel material (two rods) and 78 in. of 1-1/4-in.-diameter SNAP-10A fuel material (six rods) prepared at Atomics International were received from the Sandia Corporation. The SNAP-8 fuel was used in preliminary studies, e.g., machining studies, investigation of dehydriding techniques and rates, etc. Thermal-property determinations and solubility studies were conducted on the SNAP-10A fuel material. Test and inspection data which were received with the fuel are given in Table A-1.

Specimens for the various thermal-property measurements and studies were prepared from five of the six SNAP-10A fuel rods received. The sixth fuel rod will be returned to the Sandia Corporation. Figure A-1 shows the regions of the fuel rods from which specimens were prepared. Table A-2 gives the dimensions of thermal-property specimens that were utilized.

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TABLE A-1. SNAP-10A-TYPE FUEL ROD TEST AND INSPECTION DATA

Fuel Rod No.:	290-4	290-5	290-6	291-1	291-2	838-1
SNAP Fuel Type:	10A	10A	10A	10A	10A	10A
<u>Dimensions, In.</u>						
Length	13.236	13.249	13.273	13.191	13.246	--
Diameter (max)	1.302	1.300	1.287	1.282	1.305	--
Diameter (min)	1.300	1.297	1.294	1.277	1.302	--
Out of straight as TIR	0.014	0.007	0.018	0.031	0.031	--
<u>Fuel Alloy Chemistry</u>						
Uranium(a), w/o						
Average	10.29	10.29	10.29	10.27	10.27	10.08
Maximum	10.34	10.34	10.34	10.31	10.31	--
Minimum	10.25	10.25	10.25	10.21	10.21	--
Uranium, g	175.1	174.4	171.4	168.0	175.7	--
Carbon(a), w/o	0.18	0.18	0.18	0.19	0.19	0.21
Hydrogen, w/o	1.828	1.828	1.828	1.824	1.815	1.728
<u>Fuel Rod Physical Data:</u>						
Density, g per cm ³	6.056	6.055	6.053	6.0555	6.058	6.064
Weight, g	1733.347	1726.822	1696.720	1666.615	1742.372	1672.080
N _H	6.61	6.61	6.61	6.60	6.57	6.26

(a) For melt from which these rods were produced.

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TABLE A-2. DIMENSIONS OF SPECIMENS UTILIZED IN
THERMAL-PROPERTY MEASUREMENTS

Specimen	Configuration	Dimensions
Thermal expansion (longitudinal)	Right cylinders	2-1/2 in. long with 3/8-in. diameter
Thermal expansion (transverse)	Right cylinders constructed of two joined parts	2-1/2 in. long with 1/4-in. diameter
Diffusivity	Disks	50 mils thick with 3/8-in. diameter
Specific heat (hydride material)	Right cylinders	2 in. long with 7/16-in. diameter
Specific heat (dehydrided material)	Right cylinders	2 in. long with 3/8-in. diameter
Emissivity	Tube	6 in. long with 1/4-in. ID ($<1/32$ -in. wall)

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ROD NO. 291-1

ROD NO. 290-6

ROD NO. 290-4

ROD NO. 290-5

ROD NO. 291-2

-KEY-

TE(L) - Thermal expansion
(longitudinal) specimen

TE(T) - Thermal expansion
(transverse) specimen

D - Diffusivity specimen

S.Ht - Specific heat specimen

E - Emittance specimen

DS - Material used in dehydrating
studies

SS - Material used in solubility
studies

- Metallographic and/or
chemical samples

-H - As-received hydride fuel

-D - Dehydrated prior to
specimen preparation

FIGURE A-1. SPECIMEN LOCATION WITHIN FUEL RODS

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