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FAIRCHILD

ENGINE AND AIRPLANE CORPORATION

RESTRICTED DATA

ATOMIC ENERGY ACT OF 1946

SPECIFIC RESTRICTED DATA CLEARANCE REQUIRED

THE HEAT CAPACITY OF U_3O_8 POWDER AS A
FUNCTION OF TEMPERATURE
(400-1800°F) PROJECT NC-Ph-90113
(METHOD NC-Ph-80720)
NEPA-934-SCR-34

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~~RESTRICTED DATA~~~~ATOMIC ENERGY ACT 1946~~~~SPECIFIC RESTRICTED DATA CLEARANCE REQUIRED~~ABSTRACT

The mean heat capacity of a sample of powdered U_3O_8 , a possible fuel material for a nuclear reactor, was determined as a function of temperature from measurements of heat content. A thin-walled nickel capsule containing the material was heated in an electric furnace and dropped into a water calorimeter. The mean heat capacity values calculated from the experimental data were nearly constant over the temperature range, room temperature to $250-975^{\circ}C$ ($482-1790^{\circ}F$). The average value was 0.074 ± 0.004 cal $gm^{-1}^{\circ}C^{-1}$ (0.074 ± 0.004 BTU $lb^{-1}^{\circ}F^{-1}$). From $975-1100^{\circ}C$ ($1790-2012^{\circ}F$) a small rise was observed.

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INTRODUCTION

This report is one of a series on the measurement of the heat capacity of materials of project importance. A certain amount of data of this type already exists in the literature, but such data are meager at high temperature. The accuracy of some published data does not seem to be very high. This is particularly true for U_3O_8 , for which heat contents have been calculated by Brewer, Bromley, Gilles and Lofgren¹ from the estimates of its heat capacities given by MacWood and Altman² and by Davidson and Seaborg³ based upon comparison with UO_2 and UO_3 .

It was desired to measure the heat capacity of fuel or moderator materials and mixtures of fuel and moderator materials as a function of temperature, regardless of chemical reactions or phase changes which might occur. Reactions or phase changes would be expected to cause discontinuities in the heat capacity-temperature curve if the new phase was characterized by a distinctly different heat capacity. No such discontinuities have been specifically reported in the literature, although some of the literature data, as well as the measurements presented in this and succeeding reports, definitely suggest the possibility of phase changes at the highest temperatures reached. It is proposed to investigate the heat capacity curves further at higher temperatures, but it was first necessary to standardize the method at lower temperatures, and to obtain results over the entire temperature range, so as to characterize definitely any points of discontinuity.

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1. Brewer, Bromley, Gilles and Lofgren CC-3234
  2. MacWood and Altman RL.4.7.600
  3. Davidson and Seaborg MUC-GTS-1012

The objective of this project was to measure the mean heat capacity of uranium oxide (U_3O_8) as a function of temperature from room temperature to 250-1050°C (482-1922°F) with an experimental error of 15% or less. The experimental procedure was almost the same as that reported in NEPA-932-SCR-32, and NEPA-937-SCR-37. Nickel capsules were employed, because nickel showed better oxidation resistance under the conditions used than either copper or #303 stainless steel.

Values of the enthalpy, the true heat capacity, $C_p = \left(\frac{\partial H}{\partial T} \right)_p$, and the entropy for U_3O_8 will be given in a later report which will summarize all the data obtained to date in this laboratory. Detailed calculations for the enthalpy, true-heat capacity, and entropy for Be_2C , UO_2 , and for mixtures of the uranium oxides with BeO will also be presented in this report.

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APPARATUS AND EXPERIMENTAL METHOD

The sample was heated in a furnace to a known temperature and dropped into a copper block in a water calorimeter; the heat evolved in cooling the sample to equilibrium temperature in the calorimeter was measured. The sample was contained in a thin-walled nickel capsule with a screw bottom. The mean heat capacity of the capsule and the heat loss during the drop were corrected for by "blank" experiments with an empty capsule. The temperature of the capsule was measured with a chromel-alumel thermocouple.

In order to accelerate the measurement of the heat capacity of several materials the single unit apparatus (described in NEPA-816-SCR-28 and NEPA-932-SCR-32) was replaced by a six-unit apparatus. This apparatus is illustrated in the drawing, Figure 1, and the photograph, Figure 2. The apparatus had the following features:

1. Six capsules could be heated simultaneously.
  2. The temperature of each furnace was regulated by a Bristol Pyrometer Controller, a Variac, and a relay connected for high-low control with about 15% power difference between the high and low setting. Each furnace was set at a different temperature. The range was from 250°C (482°F) to 1050°C (1922°F). Higher temperatures up to 1150°C (2102°F) have been used; but these temperatures curtailed the life of the furnace (wound with chromel-A wire).
  3. A special table to hold the controllers and furnaces was designed and constructed. This table was provided with six shelves, one to hold each calorimeter. The shelves helped in the alignment of furnace and calorimeter. (Previously, uncertainty of alignment sometimes prevented the capsule from dropping centrally into the vase-shaped copper block.) The hinged shelves were supported by chains provided with a pin-release mechanism which permitted the
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calorimeter to be removed easily and placed on a table for time-temperature readings after the drop. In Figure 2 a calorimeter is shown in place under the second furnace; an operator is shown making time-temperature readings on another calorimeter; and a capsule is shown suspended over furnace # 5.

4. The six furnace temperatures were recorded continuously by a multi-point Micromax recorder. The temperatures could also be read intermittently by a Leeds and Northrup thermocouple potentiometer and a multipoint switch.

5. The time-temperature curve could be determined by thermometer readings at specified intervals with a timer, as in previous experiments. A curve was plotted automatically by a Brown High Speed Elektronik Potentiometer with an iron-constantan thermocouple. The potentiometer had an expanded range of 0°C - 100°C (32°F - 212°F) and a chart speed of 90"/hr. The thermocouple was inserted into the calorimeter through a glass tube to minimize heat conduction, and soldered into a copper cylinder to provide some integration of the calorimeter temperature.

Not only was the experimental method improved by the use of the multiple unit apparatus, but also by modification of the capsule design. The capsules used in previous experiments were sealed by threaded top closures. The thickness necessary for the inside thread determined the amount of metal which could be cut away from the side walls of the capsule. In order to decrease the weight of the capsule and hence increase the ratio of weight of sample to weight of capsule, the capsules were made as follows:

1. The inside taper was drilled and bored to the required depth, and the roughed-out piece cut off.
2. A top was welded on the roughed-out piece.
3. A hole was bored and threaded in the bottom.
4. The outside taper of the capsule was turned.
5. A screw closure for the capsule bottom was made.

By this method capsules with wall thicknesses from 0.009" to 0.015" were fabricated. The weight was reduced from a range of 43-66 grams to a range of 18-23 grams.

EXPERIMENTAL DATA

Twenty determinations of the heat content of U_3O_8 powder in nickel capsules were made over the temperature range, room temperature to $250-1050^{\circ}C$ ($482-1922^{\circ}F$). From these, the mean heat capacity of U_3O_8 was calculated as a function of temperature, using the equation for the mean heat capacity of nickel derived in NEPA-937-SCR-37. Data for the mean heat capacity of U_3O_8 are shown in Table I, and the data are graphed in Figure 3. The value $C_p = 0.074 \pm 0.004 \text{ cal gm}^{-1}\text{ }^{\circ}C^{-1}$ ($0.074 \pm 0.004 \text{ BTU lb}^{-1}\text{ }^{\circ}F^{-1}$) expresses the heat capacity from room temperature to $975^{\circ}C$ ($1790^{\circ}F$) within the limits of experimental error.

The U_3O_8 was made by the Mallinckrodt Chemical Company, designated as Standard Sample MS-ST, and was nearly 100% U_3O_8 .

In making the first runs, time-temperature readings were compared with curves plotted on the Brown Electronik Potentiometer. A careful calibration of the Brown Potentiometer against a good mercury thermometer was made. The corrected readings were used in the calculations.

In all previous heat capacity determinations the water equivalent was calculated from the mass and heat capacity of each constituent of the calorimeter. As an example, for a combination consisting of 300 cc of water, copper block #2, aluminum calorimeter cup #2, immersed portion of stirrer #8 and the immersed portion of the thermometer, the water equivalent was calculated to be $338.6 \text{ cal } ^{\circ}C^{-1}$. To check this calculated value the water equivalent for the same combination was determined by electrical heating. A heating coil was wound with asbestos-insulated chromel-A wire and placed in position in the calorimeter around the base of the copper vase. Potential and current leads were connected to the heating coil. The potential drop was measured by a L. & N. Type K-2 potentiometer, and the current by the voltage drop across a standard 0.1 ohm resistor. Current was supplied by two 6-volt storage batteries connected in series. Time-temperature readings were taken

over a temperature interval corresponding to the value of ΔT observed during a heat content measurement. Values of 338 and 334 cal $^{\circ}\text{C}^{-1}$ were obtained from two consecutive runs. Since these values agreed well with the calculated value given above, it is believed that the calculated values of the water equivalent are correct within one or two per cent.

DISCUSSION

The mean heat capacity of U_3O_8 calculated from the determinations made over the temperature range, room temperature to $250-975^\circ C$ ($482-1790^\circ F$) was constant within experimental error, and equal to $0.074 \text{ cal gm}^{-1}^\circ C^{-1}$ ($0.074 \text{ BTU lb}^{-1}^\circ F^{-1}$). Practically all of the points fell within $0.004 \text{ cal gm}^{-1}^\circ C^{-1}$ of this value. At the highest temperatures, in the neighborhood of $1100^\circ C$ ($2012^\circ F$), the mean heat capacity appeared to rise. Further experiments at higher temperatures will be carried out in the near future.

The measured values of the heat capacity of U_3O_8 agree with figures found in the literature. It was calculated from Report CC-3234 that the mean heat capacity of U_3O_8 from $25-227^\circ C$ ($77-441^\circ F$) was $0.070 \text{ cal gm}^{-1}^\circ C^{-1}$ ($0.070 \text{ BTU lb}^{-1}^\circ F^{-1}$). From $25-727^\circ C$ ($77-1341^\circ F$) it was $0.071 \text{ cal gm}^{-1}^\circ C^{-1}$ ($0.071 \text{ BTU lb}^{-1}^\circ F^{-1}$). From $25-1227^\circ C$ ($77-2241^\circ F$) it was $0.080 \text{ cal gm}^{-1}^\circ C^{-1}$ ($0.080 \text{ BTU lb}^{-1}^\circ F^{-1}$).

The total error of the determinations was estimated to be 15% or less. Factors contributing to this error were the following:

- 1) Errors in the thermocouples and thermometers.
- 2) Errors arising from impurities in the sample. As material of the highest purity was used, this error should be negligible.
- 3) Non-reproducible heat losses by radiation and convection during the time of fall of the capsule.
- 4) Errors in the extrapolated maximum temperatures.
- 5) Errors arising from a change of mean heat capacity of the capsule because of oxidation during successive heatings.
- 6) Errors arising from a change of chemical and physical composition of the sample during periods of heating and cooling.
- 7) Errors arising from differences between the true and calculated values of the water equivalent of the calorimeter. (The electrically measured

values of the water equivalent obtained as described in this report agree with calculated values.) The error from this source is less than 2%.

- 8) Errors caused by the heat of stirring. These are probably small in comparison with other errors.

Figure 1
MULTIPLE UNIT HEAT CAPACITY APPARATUS

TABLE I

TEMPERATURE AND HEAT CAPACITY DATA FOR U_3O_8 IN NICKEL CAPSULES *

<u>Run Number</u>	<u>Furnace Temp., $T, ^\circ C$</u>	<u>Mean Heat Capacity (C_p)_s, cal gm⁻¹°C⁻¹</u>
1	253	.068
2	255	.072
3	260	.075
4	452	.079
5	453	.080
6	466	.072
7	678	.072
8	688	.076
9	813	.072
10	822	.075
11	824	.075
12	901	.076
13	969	.072
14	969	.074
15	974	.075
16	1077	.070
17	1084	.079
18	1091	.085
19	1092	.086

*The weight of the sample was 59.80 grams throughout the series of runs.

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