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INTERIM REPORT

THERMAL CONDUCTIVITY STUDIES ON
VARIOUS CALCINED "COLD" WASTES

By

W. V. DeMier

Process Equipment Development
Chemical Development
Chemical Research and Development
HANFORD LABORATORIES OPERATION

March 11, 1960

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INTERIM REPORT - THERMAL CONDUCTIVITY STUDIES
ON VARIOUS CALCINED "COLD" WASTES

INTRODUCTION

Accelerated accumulation of high level radioactive wastes has given impetus to the development of safer and more economical waste storage methods. Several processes for converting these wastes to granular or porous-solid form are being investigated^(1,2). In any waste volume-reduction process, containment of the final material in a sealed metal shell is desirable as a precaution against accidental release of radioactive material. While the final waste forms are generally stable, they are, if uncontained, subject to some degree to leaching, volatilization, and dusting caused by fracturing due to thermal stresses.

An experimental program is now underway to develop an understanding of the variables in waste systems so that suitable storage containers may be designed. This report covers the first phase of the program, a study of heat transfer properties of self-heating granular solids.

SUMMARY AND CONCLUSIONS

Effective thermal conductivity (K^*) of granular alumina was found to increase with temperature from <0.2 Btu/(hr sq ft °F/ft) at 250 F to <0.4 Btu/(hr sq ft °F/ft) at 1000 F. The latter value is about one-tenth of the thermal conductivity of zero porosity Polycrystalline alumina as given in the literature⁽⁴⁾.

The effective thermal conductivity of finer materials, similar to those produced by spray and fluid bed calciners was found to increase from 0.1 Btu/(hr sq ft °F/ft) at 200 F to <0.25 Btu/(hr sq ft °F/ft) at 1000 F.

The rate of heat transfer through granular alumina was found to be dependent on particle size; the rate through 8 mesh alumina being 1.5 times the rate through 120 mesh material of the same bulk density.

Experimental equipment was built to permit simulation of uniform heat generation in granular materials. Temperature profiles and heat transfer rates were determined in these materials at various heat generation rates.

EQUIPMENT

The studies were conducted in three different units. Two of the units provide an approximation of uniform heat generation. Thermocouples in these units are placed at various points along two mutually perpendicular diameters in the middle of the cylinder length. The third unit provides a means for determining thermal conductivities of materials that sinter or fuse during tests, as such materials are difficult to remove from the uniform heat apparatus.

Uniformly Heated, Four-Inch Diameter Cylinder (Fig. 1)

A two foot length of 4-inch, schedule 40, 304 stainless steel pipe forms the shell for this unit. Heat is supplied by 60 Nichrome V wires of 0.028-inch diameter, connected in three parallel circuits of 20 wires each. The wires are enclosed by a 1/8-inch diameter stainless steel sheath, with magnesia

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insulation between the sheath and the wire. Heater wires are on 7/16-inch equilateral triangular spacing parallel to the cylinder axis. Heated length is 22 inches.

Several other heating methods were considered. Among them were induction heating, dielectric heating, and resistance heating using the material to be tested as the resistor. In the final analysis the resistance wires presented the most satisfactory method.

Temperatures are measured by means of 12 stainless steel-sheathed, 1/16-inch diameter, chromel-alumel thermocouples spaced at various known radii with hot junctions all in a plane perpendicular to and midway down the pipe axis.

Uniformly Heated, Two-Inch Diameter Cylinder (Fig. 2)

A one foot length of 2-inch schedule 40, 304 stainless steel pipe forms the shell for this unit. Heat is supplied by 19 sheathed Nichrome V wires of the same type as used in the 4-inch unit above, but on 3/8-inch triangular spacing. Thermocouple type and arrangement for this unit are similar to those in the four-inch unit, but only four thermocouples are used.

Axially Heated, Two-Inch Diameter Cylinder (Fig. 3)

This unit differs from the first two units described in that all the heat is applied along the axis, rather than uniformly throughout the sample. Two stainless steel-sheathed thermocouples are used to determine temperatures at two different radii within the sample. The heater is a helix about 8-1/2 inches long, made of 0.036-inch diameter Nichrome V, wound on a 1/8-inch diameter alumina tube.

DISCUSSION

Scope of Initial Studies

The work reported here represents one phase of a program to develop a container for storing high level radioactive solids such as calcined separations process wastes. A basic problem encountered in storing such material is the removal of the heat which accompanies radioactive decay. The removal rate must be great enough to prevent internal temperatures from reaching some maximum value. Generally this maximum is considered to be the temperature at which the waste was calcined or at which volatilization of the waste or waste fission products would occur. An indication of the heat generation rates involved is given in Tables 1 and 2.

*TABLE 1. HEAT GENERATION IN PUREX 1WW
FOR URANIUM IRRADIATED TO 800 MWD/T AT 7 MW/T

Solid Waste Volume → ↓Cooling Time	1 cu ft	0.6 cu ft	0.3 cu ft
	Ton U	Ton U	Ton U
90 days	8390 Btu/hr cu ft	13,980 Btu/hr cu ft	28,000 Btu/hr cu ft
1 year	1520	2,530	5,000
2 years	280	460	930
5 years	120	200	400

FIGURE 1
Uniformly Heated, 4-Inch Diameter Cylinder

FIGURE 2

Uniformly Heated, 2-Inch Diameter Unit

FIGURE 3
Axially Heated Unit

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*TABLE 2. HEAT GENERATION IN PUREX LWW
FOR URANIUM IRRADIATED TO 1500 MWD/T at 10 MW/T

Solid Waste Volume → ↓ Cooling Time	1 cu ft	0.6 cu ft	0.3 cu ft
	Ton U	Ton U	Ton U
90 days	13,700 Btu/hr cu ft	22,800 Btu/hr cu ft	45,700 Btu/hr cu ft
1 year	2,760	4,500	8,900
2 years	500	850	1,700
5 years	200	400	750

*These tables are based on calculations reported
in reference (3)

A waste volume reduction to 0.3 cubic foot per ton of uranium irradiated is well within the capabilities of the calciners now being developed. The optimum interim storage period cannot, of course, be determined at present. A heat generation rate of 45,700 Btu/hr cu ft was set as a nominal upper limit for experimental testing. As shown in Table 2, this corresponds to the specific heat generation rate of 90-day-cooled LWW from fuel irradiated to 1500 MWD/T at 10 MW/T, calcined to a solids volume of 0.3 cu ft/Ton U.

Previous Work on Subject

Little experimental work on temperatures in uniformly heated systems has been reported in the literature. The mathematical analysis for ideal systems is well known^(5,7), but is not applicable to practical systems without the use of correction factors to allow for deviations from perfectly spherical or cylindrical particle shapes.

To provide experimental data for estimating temperatures in granular solids at various rates of self-heating, the equipment described in the previous section was built.

Derivations of equations for representing heat flow in the two kinds of apparatus are available in any standard text on heat transfer by conduction⁽⁵⁾. The integrated forms, applicable at steady state are shown here for convenience. They are applicable only where all heat flow may be considered to be radial (i.e., cylinder may be considered infinite). Thermal conductivity is assumed constant over the range T_1 to T_2 .

For the axially heated unit:

$$q = \frac{2\pi K^* L (T_1 - T_2)}{\ln (r_2/r_1)}$$

For the uniformly heated units:

$$q = \frac{4K^* (T_1 - T_2) V}{(r_2^2 - r_1^2)}$$

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where:

q = heat generation rate, Btu/hr

r = volume of sample, cu ft

r_1, r_2 = radii at points 1 and 2, ft

T_1, T_2 = temperatures at points 1 and 2, °F

L = heated length, ft

K^* = "Effective" thermal conductivity in Btu/(hr sq ft °F/ft)

(Note: In this report the symbol K^* or the term "Effective thermal conductivity" will be used in referring to the experimentally determined transfer rate through granular material. The term will be understood to consolidate effects of particle contact films and transfer by radiation.)

The two-inch axially-heated unit and the two-inch uniformly-heated unit were checked for accuracy by using them to determine effective thermal conductivities of diatomaceous earth and comparing the results with those given in the International Critical Tables⁽⁶⁾. The four-inch unit was not checked because of the difficulty of obtaining uniform packing of the diatomaceous earth, which was considerably more difficult to pack than were the other materials tested. Results of the check (Figure 4) show a maximum disagreement of about 6% between values in the International Critical Tables and those determined by the equipment check.

After the apparatus was completed, and before simulated calcined waste was available, studies were made on heat flow through granular fused alumina. These were directed toward learning whether some relationship might exist between particle size and effective thermal conductivity. Studies reported in the literature generally have dealt with effects of temperature and bulk density, but not with the effect of particle size; although some reports do include particle "shape" corrections⁽⁷⁾.

Investigation of Particle Size Effects

Two or more granular systems of the same material (hence the same density at zero porosity) may have different particle sizes and yet have the same bulk density, or porosity, provided that the range of particle sizes within a given sample is small, and the particles have a nearly spherical shape.

On this basis four closely graded sizes of Alundum 38 alumina were obtained for the studies:

- A. 100% through 8 mesh, 90% on 10 US mesh Density = 1.78 gm/cc
- B. 100% through 18 US mesh, 90% on 25 US mesh Density = 1.84 gm/cc
- C. 100% through 40 US mesh, 90% on 50 US mesh Density = 1.77 gm/cc
- D. 100% through 120 US mesh, 90% on 170 US mesh Density = 1.87 gm/cc

The different densities may be attributed to (1) deviation of particle shapes from perfect spheres, and (2) the difference in particle sizes occurring in spite of efforts to minimize the difference. The density of a given sample may

also vary with the degree of tamping. The values given above were determined for the samples while they were in the test unit, with just enough tamping to assure no cavities were present in the sample.

Figure 5 shows that no significant difference occurs between K^* values (at any given temperature) for the three materials composed of less than 18-mesh particles. The effective conductivity of the 10-mesh material is, however, higher than the others by a factor of nearly two although the densities are essentially equal. While no statistically valid conclusion can be reached from the amount of data taken, the results do show that methods for predicting effective thermal conductivities on the basis of void fraction (or bulk density) and the true thermal conductivities of the gaseous and the solid phases probably should not be applied to systems composed of large particles. That is, the heat transfer rate through given granular materials is affected by the size of the particles, once the size reaches a certain value.

Heat Transfer Rates in Simulated Calcined Wastes

The effective thermal conductivities of three fluid-bed calcined simulated wastes are shown in Figure 5.

Waste 1 is a fluid-bed-produced, simulated waste from Idaho Chemical Processing Plant. Composition of the material resembles that of Hanford's Redox waste. Screen analysis for the material is:

+20 Tyler mesh	5.8% by wt
+50	90.0
+100	3.3
+200	0.02

Tapped density is 0.77 gm/cc

Wastes 26Pa and 28Pa were produced in the Hanford Chemical Engineering Development Operation fluid bed unit⁽²⁾. The density of the material as tested was 1.3 gm/cc. Screen analysis is as follows:

<u>Waste 26Pa</u>		<u>Waste 28Pa</u>	
+14 Tyler mesh	0.9% by wt	+14 Tyler mesh	3.1% by wt
+20	0.3	+20	0.4
+35	11.7	+35	37.7
+48	48.0	+48	46.8
+100	39.0	+100	1.6
+200	0.1	+200	Trace
+325	0		

Wastes 26Pa and 28Pa were calcined from formaldehyde-killed, 3-fold concentrated, simulated Purex LWW waste at bed temperatures of 500 C and 575 C respectively. No feed additives or post calcination treatment for improving thermal conductivity were used in preparing the wastes.

On the basis of K^* values shown in Figure 5, the permissible radius of a cylinder of calcined waste may be estimated. If one assumes a maximum permissible center temperature of 600 C (1112 F), a heat generation rate

Figure 5
EFFECTIVE THERMAL CONDUCTIVITY OF GRANULAR MATERIALS

- Alumina "A", density 1.78 gm/cc, 100% thru 8 mesh, 90% on 10 mesh
- ◇ Alumina "B", density 1.84 gm/cc, 100% thru 18 mesh, 90% on 25 mesh
- Alumina "C", density 1.77 gm/cc, 100% thru 40 mesh, 90% on 50 mesh
- △ Alumina "D", density 1.87 gm/cc, 100% thru 120 mesh, 90% on 170 mesh
- + Waste 28Pa, density 1.3 gm/cc (See Discussion for screen analysis)
- Waste 26Pa, density 1.3 gm/cc (" " " " ")
- ▽ Waste 1, density 0.77 gm/cc (" " " " ")

of 28,000 Btu/hr cu ft, and a temperature of 200 C (392 F) at the surface of the material (not the container) the maximum permissible radius is

$$r = 2\sqrt{\frac{K^* (T_1 - T_2)}{q/v}}$$

From figure 5, K^* is approximately 0.12 at a mean temperature of 752 F. Hence:

$$r = 2\sqrt{\frac{(0.12)(1112-392)}{28000}}$$

$$= 0.11 \text{ ft or } 1.3 \text{ inches.}$$

Maximum permissible radii at other heat generation rates, using the basic assumptions above, are shown in Figure 6. The assumption of a temperature of 200 C at the surface of the material is arbitrary. A more realistic idea of the temperatures involved is shown in Figure 7, where temperature profiles are shown for a cross section of a uniformly heated cylinder of waste. For three cases the temperatures at the outer surface of the two-inch, schedule 40 pipe containing the material are shown. Cooling was by natural convection only.

Direction of Future Work

In general, the low conductivities observed in the materials tested to date indicate a need for study in the field of improving heat transfer rates through solid wastes. Some ways that offer promise of accomplishing this improvement are: (1) pre-calcination treatment, such as addition of melt forming constituents to calciner feed; (2) post-calcination treatment such as compaction, sintering, or dense melt formation; and (3) calcination by a process which forms a solid continuous product instead of a granular product.

Studies in heat transfer characteristics of simulated wastes incorporating these features are planned as a part of the continuing program.

Figure 6
RADIUS LIMITATIONS

assumes 200°C surface temp.
600°C max internal temp

Figure 7
TEMPERATURE PROFILES

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