The Use of a Thermal Probe to Determine the Effective Thermal Conductivity of Packaging Contents*

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INTRODUCTION

Contact-handled transuranic (CH-TRU) waste materials are nonradioactive items contaminated with alpha-emitting transuranium radionuclides. The most common transuranium radionuclides in the waste are plutonium and associated daughter products. These contaminants are usually in the form of oxides and are embedded, trapped, or otherwise attached to a variety of inert host or parent materials. Due to the high efficiencies of the recovery process, the actual contaminants are normally small in size and the contaminants remaining in the waste are usually well attached to their host.

The TRU waste is typically sealed in plastic bags which, in turn, are placed within cans or other containers and are subsequently placed in 55-gallon steel drums with polyethylene liners. These procedures prepare the waste for transport and storage and reduce the possibility of contaminant release.

Department of Transportation Regulation 49CFR173.24, as described in Code of Federal Regulations, Title 49, Part 173, "Standard Requirements for All Packages," 1985, requires that "there will be no mixture of gases and vapors in the package which could, through any credible spontaneous increase of heat or pressure or through an explosion, significantly reduce the effectiveness of the packaging." Since many CH-TRU wastes produce gases, the designers of radioactive transport containers must consider the consequences of gas generation to ensure the safe operation of transport containers.

During transport, gases are primarily generated in CH-TRU waste through three mechanisms: radiolysis, bacteriological decay, and thermal degradation. Radiolysis of organic materials, such as cellulosics, plastic, and oil, generates hydrogen, carbon dioxide, and carbon monoxide and depletes oxygen. Bacteriological decay of organic materials produces carbon dioxide or methane.

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To calculate the amount of gas generated during normal transport, the designers of the transport container require the waste centerline temperature as a function of the total decay heat load. The waste centerline temperatures thus far have been calculated using a conservative value for the thermal conductivity of the actual waste. Using a low value for the thermal conductivity can limit the payload of the transport container by overpredicting the waste centerline temperature. In addition, since the amount of gas generated is strongly dependent on the waste centerline temperature, the amount of gas generation and pressure buildup during normal transport could also be overpredicted. Therefore, an investigation was conducted to determine the feasibility of a method for estimating the effective thermal conductivity of CH-TRU waste. The method ultimately chosen should be reliable, nondestructive, and easy to implement; the data obtained must be defensible and must augment the thermal analysis of the transport container.

SIMULATED WASTE FORMS AND REFERENCE MATERIAL

CH-TRU waste can be generally classified into five categories for transportation purposes: (1) soft waste, (2) hard waste, (3) processed/treated waste, (4) large equipment waste, and (5) special waste, which is considered to be more mobile than the other four types. The predominant characteristic of soft waste, the waste form of interest for this investigation due to its inherently low thermal conductivity, is the flexible nature of the host. These CH-TRU contaminated wastes consist of radioactive elements embedded or trapped in the fibers of paper, rags, protective clothing, plastic bags, liners, or other similar host materials. Consequently, to prove the method, three simulated waste forms were chosen: paper, disposable clothing, and plastic.

The simulated disposable clothing waste was obtained from the clean rooms of Sandia's Radiation Hardened Integrated Circuit Laboratory and consisted of paper coveralls, caps and booties, plastic gloves, and the plastic envelopes which contained the coveralls. The simulated paper waste form consisted of the Marathon Paper Company's multipurpose wipes, 860W "Maratuff." The wipes are 29 by 37 cm when folded in quarters. Each wipe was unfolded, crumpled, and placed in a 55-gallon steel drum and liner. The simulated plastic waste consisted of plastic sheeting 0.2 mm thick and 6 m wide. The plastic sheets were cut into smaller sections and densely packed into the drum.

In addition, to add confidence to the data collected, the thermal conductivity of silica microspheres (a reference material) was determined and compared to values reported in the literature. The microspheres ranged from 0.2 to 0.9 mm in diameter; the predominant size was approximately 0.7 mm. The article "Induced Convection During Cylindrical Probe Conductivity Measurements on Permeable Media" featured in Thermal Conductivity 17, Fodemesi and Beck, 1983, reports the thermal conductivity of similar size silica microspheres when saturated with air to be approximately 0.19 W/m-K.

Packaging densities for the three simulated waste forms and the reference material are given in Table I.

Table I. Packaging Densities

Waste Form	Density* (kg/m^3)
Clothing	100.9
Wipes	77.7
Plastic	107.8
Silica Microspheres	1464

*Drum liners have a measured volume of 0.196 cubic meters.

These packaging densities are not implied to be typical or usual but are reported because thermal conductivity is a function of density, and the values would be meaningless without them.

EXPERIMENTAL TECHNIQUE

Methods of determining thermal conductivity fall into two broad categories: transient and steady state methods. The article "Heat Transmission in Low Conductivity materials" featured in Thermal Conductivity, A.W. Pratt, 1969, provides a summary of experimental methods applicable to low conductivity materials.

Transient methods simulate the variable state term in the general solution of the energy equation with the appropriate boundary conditions. Transient probe methods are normally referred to as line source or probe methods; the basic configuration consists of a thin wire or foil embedded in the test material (material of unknown thermal conductivity). Transient probe techniques generally determine the thermal conductivity of materials more efficiently than steady-state methods. In addition, the probe method usually is more suitable for nondestructive, in-situ measurements. As a result, the transient probe technique was the method chosen to determine the thermal conductivity of the waste forms.

ANALYTICAL BACKGROUND

As stated previously, the thermal conductivity probe is based on the transient line source technique, where power is supplied to the probe at a constant rate. The thermal response of the probe as a function of time is used to determine the effective thermal conductivity of the test material.

The analytical solution for the response of an infinite medium to a line source is represented by Eq. 1 from Conduction of Heat in Solids, Carslaw and Jaeger, 1984:

$$T(r,t) = \frac{q}{4\Pi k} \ell n \left[\frac{4\alpha t}{r^2}\right] - \frac{\gamma q}{4\Pi k} \qquad (1)$$

where

T = temperature rise of medium, K

q = constant heat input per unit length of wire starting at zero time, W/m

k = thermal conductivity of the medium, W/m-K

t = time, s

r = distance from wire, m

α = material diffusivity, $k/\rho c_p$, m^2/s

ρ = material density, kg/m^3

c_p = specific heat of material, J/kg-K

γ = Euler's constant, 0.5772

Π = 3.142.

If higher order terms of the solution are neglected, the temperature rise of the probe for points where $r^2/\alpha t$ is small may be described by:

$$(T_2 - T_1) = \frac{q}{4\Pi k} \ell n \left[\frac{t_2}{t_1}\right] \quad (2)$$

where

T_i = temperature at time t_i.

When the approximation described by Eq. 2 is valid, the probe temperature rise is linear with the natural logarithm of time. The slope of the temperature rise is then proportional to the probe heater input power and inversely proportional to the effective thermal conductivity of the material. When the linear temperature rise is fully developed, as shown in Fig. 1, this straight line method is readily employed as the basis for probe data analysis techniques.

To obtain a complete understanding of the probe, the following interrelated factors should be included:

1. Finite probe length and diameter.
2. Thermal properties of the probe material.
3. Contact resistance at the probe sample.
4. Effect of the sample boundaries.
5. Location of the temperature measurement device.

These factors were discussed in detail in "Analysis of Thermal Conductivity Probes for High Temperature Applications," J. A. Koski, Sandia National Laboratories, 1981 and "Thermal Conductivity of Aqueous Foam," Drotning et al., Sandia National Laboratories, 1982.

EXPERIMENTAL DETAILS

The probe was constructed from a Watlow Electric Manufacturing commercial heater, Model No. RGS3910, and the heater is 1 m long and 1.2 cm in diameter. The heated length is 68 cm. The sheath material is mild steel. The electrical rating of the heater is 1000 watts at 240 volts. A chromel-alumel thermocouple was welded to the heater sheath at the midpoint of the heated length. Additional thermocouples were welded at one-half the distance from the midpoint to the ends of the heated length, making a total of three thermocouples per probe. The probe had a larger diameter than is typical (Fig. 2) because strength was required when the simulated waste was packed around the probes. The probes were operated at low power settings (3 to 10 watts per meter).

A portable automated data acquisition and control system was originally designed for measurements of conductivity using the line source probe method for UO_2 powders in inert atmosphere at elevated temperatures ("Thermal Conductivity Probe System," Drotning and Tormey, Sandia National Laboratories, 1984). The same data acquisition system and the thermal probe described above were used for this application.

Fig. 3 illustrates the portable thermal conductivity probe system. The hardware components were comprised of a Hewlett-Packard HP-85 personal computer, an HP3497A data acquisition/control unit, an HP6266B constant voltage DC power supply, a customized probe switching system, and the thermal conductivity probe.

Fig. 4 illustrates the software developed for the HP-85 for data acquisition and analysis ("Thermal Conductivity Probe System," Drotning and Tormey, Sandia National Laboratories, 1984). The program uses the special function keys on the HP-85 to direct the operator to the various functional sections of the program. These functional sections include data acquisition (ACQUIRE), graphical data analysis (ANALYZE), and visual readout of the instrumentation parameters (VIEW).

Once loaded and initialized, the program waits for the operator to select one of the desired functions. During this period, the system periodically samples the probe temperature to determine a linear approximation to the baseline temperature drift of the probe. The VIEW function provides the operator with system parameter information, such as time, probe temperature, voltage, current, and resistance of the load (either the dummy load or the resistive probe heater).

The ACQUIRE function is used to acquire probe data. The operator may optionally specify a file name for data storage on cassette tape following data collection. Once acquisition has begun, the system switches power from the dummy load to the probe heater, and readings of the time and probe temperature are recorded. The power (current and voltage) applied to the probe is also recorded. Acquisition of data may be terminated either by operator intervention or by automatic control after a specified run duration. The applied power is then switched back to the dummy load. The program either returns to the function selection menu or proceeds to the analysis section, depending on the software configuration.

In the ANALYZE function data are obtained from storage on tape or in memory for subsequent graphical data analysis using the straight line analysis method. The data are plotted on the CRT screen as temperature versus the natural logarithm of time. The operator selects the linear region using a movable cursor, a least squares fit is made to the data, and the thermal conductivity is computed. Additional hardware and software documentation may be found in "Thermal Conductivity Probe System," Drotning and Tormey, Sandia National Laboratories, 1984 and "An Automated Thermal Conductivity Probe and Applications to Powders," <u>Thermal Conductivity 18</u>, W.D. Drotning, 1983.

EXPERIMENTAL RESULTS AND DISCUSSION

The probe response (temperature rise versus ln time) for the disposable clothing at room temperature is shown in Fig. 5. The linear region used in the analysis is bound by the vertical dashed lines. The conductivity values for the three simulated waste forms and the reference material are given in Table II.

Table II. Effective Thermal Conductivity of Simulated CH-TRU Waste Forms

Waste Form (-)	k (W/m-K)	Test Duration (Sec)	Temp(1) (°C)	ΔTemp(2) (°C)	Probe Power (W/M)
Clothing	0.062	1800	36.6	21.5	7.72
Clothing	0.060	3600	31.1	15.5	4.28
Clothing	0.061	10800	38.6	35.5	8.05
Wipes	0.047	600	29.8	13.5	4.26
Wipes	0.046	1800	39.9	23.5	7.7
Wipes	0.039	10800	37.7	31.5	4.94
Plastic	0.052	1800	32.9	14.7	4.95
Plastic	0.050	10800	36.5	26.0	4.95
Reference	0.190	1800	29.6	12.0	10.0
Reference	0.188	3600	32.8	14.5	10.0

[1] Average temperature of the straight line portion of the curve.
[2] Temperature rise of the probe.

Apart from some solid materials of low thermal conductivity, most normal low conductivity materials may be considered mixtures of gases and solid bodies. Their insulation value is due to the low thermal conductivity of air which, when dry and free of convective movement, transmits heat at the rate of 26.2×10^{-3} W/mK at 273°K. In general, the higher the fractional void volume or porosity, the lower the conductivity of the material will be. This effect is illustrated in Table II (densities are given in Table I), which shows the increase in effective thermal conductivity with density at atmospheric temperatures.

These results should be interpreted as conservative—that is, having a lower value than a real waste form. The conservatism arises from the fact that none of the waste forms in this investigation contained any appreciable moisture. They were packed in the drums at a relative humidity of less than 40 percent and were dry. At normal temperatures water conducts heat at approximately 25 times the rate in air. The presence of moisture will reduce the insulation value of the waste forms.

SUMMARY AND CONCLUSIONS

The thermal conductivity of simulated CH-TRU waste was successfully measured at ambient conditions using the line source thermal conductivity probe technique. The "atypical" probe (i.e., larger in diameter) used for this application was successfully adapted to existing hardware and software. The portable automated instrumentation system is ideally suited for remote field experiments. Results of the experimental program indicate the line source thermal conductivity probe technique can provide a reliable, nondestructive, and easily implemented method for determining the thermal conductivity of actual CH-TRU waste. However, further experimental study is warranted to develop a disposable-piercing type of probe optimized for measuring the conductivity of CH-TRU waste at generating sites.

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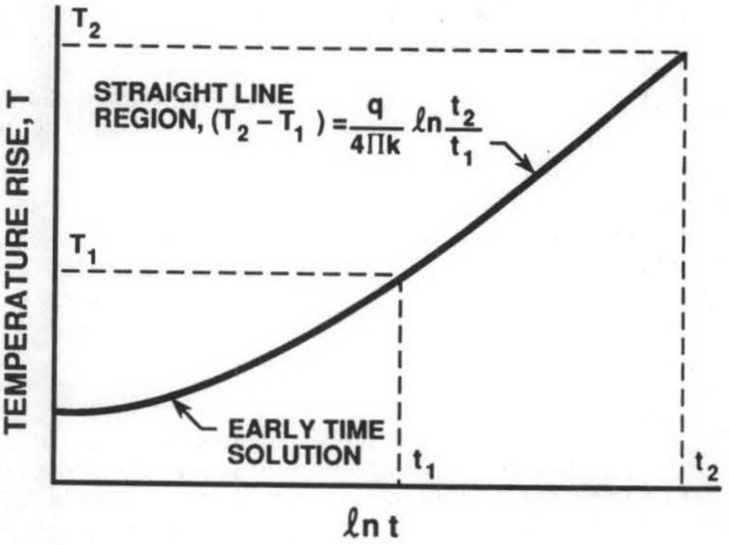

Figure 1. Temperature rise near a line heat source in an infinite medium as a function of log-time.

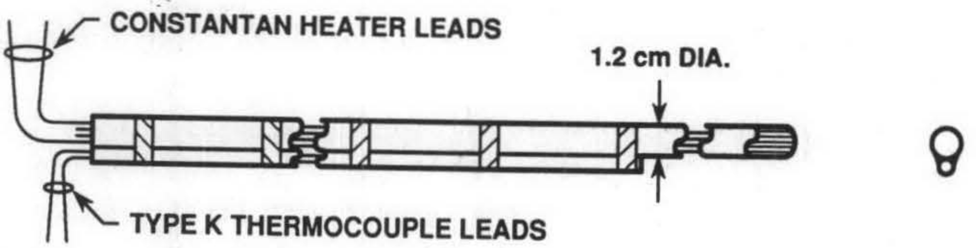

Figure 2. Construction details of thermal probe.

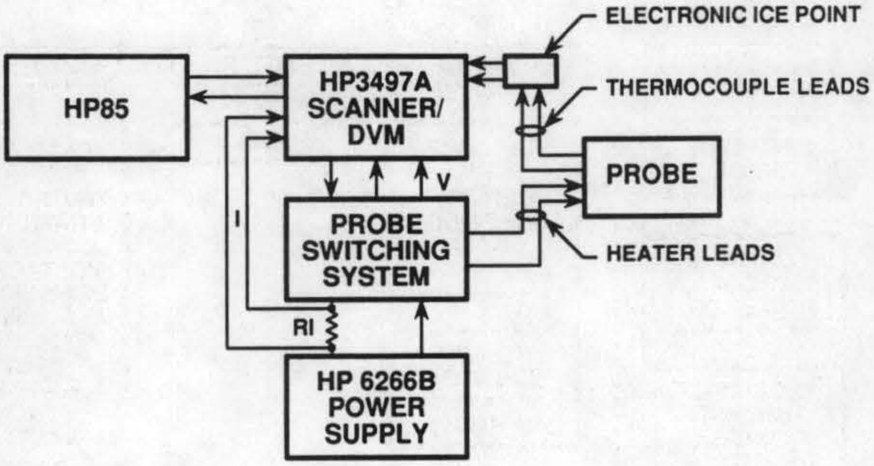

Figure 3. Block diagram of thermal conductivity probe measurement system.

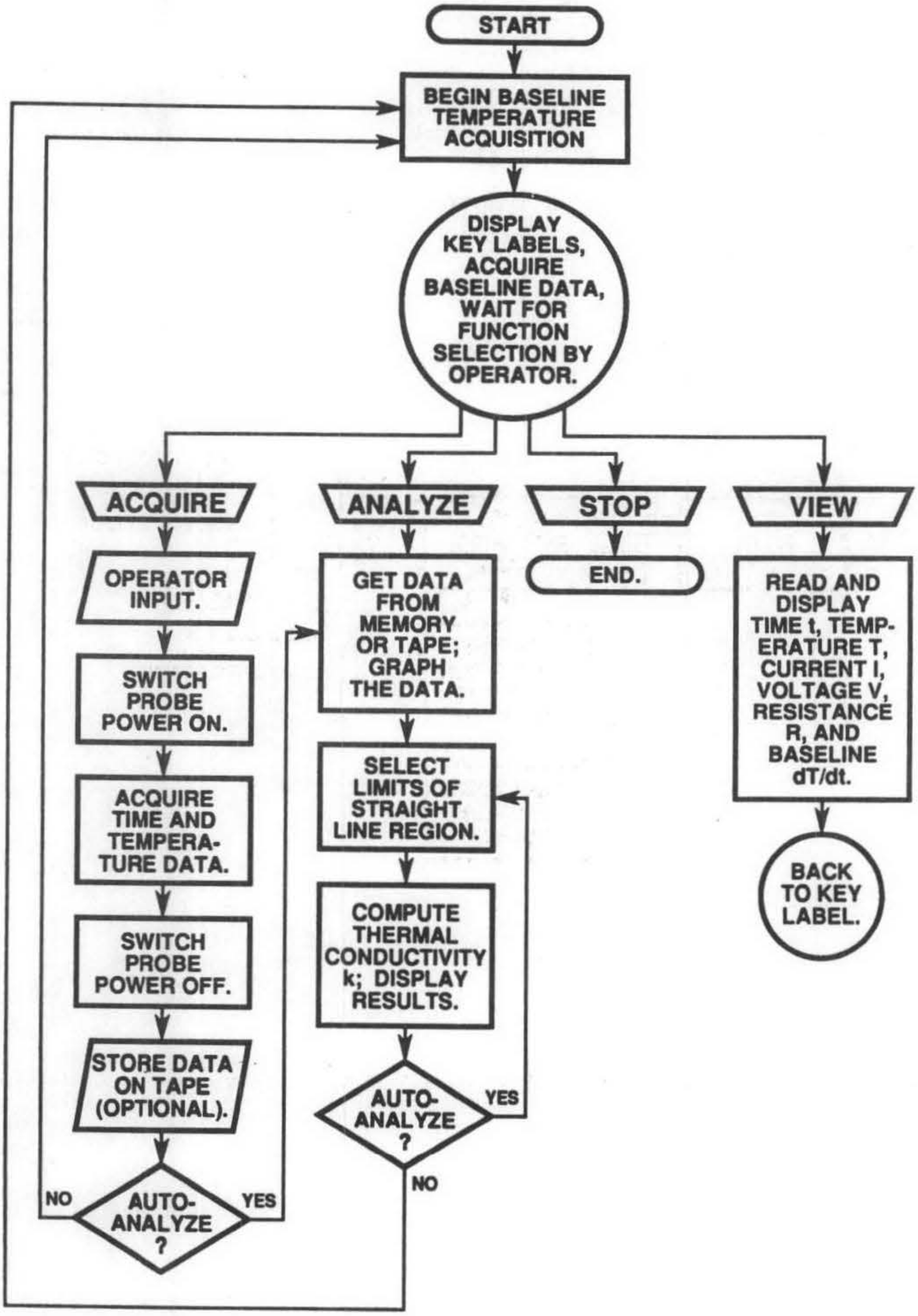

Figure 4. Flow diagram of probe data acquisition and analysis software.

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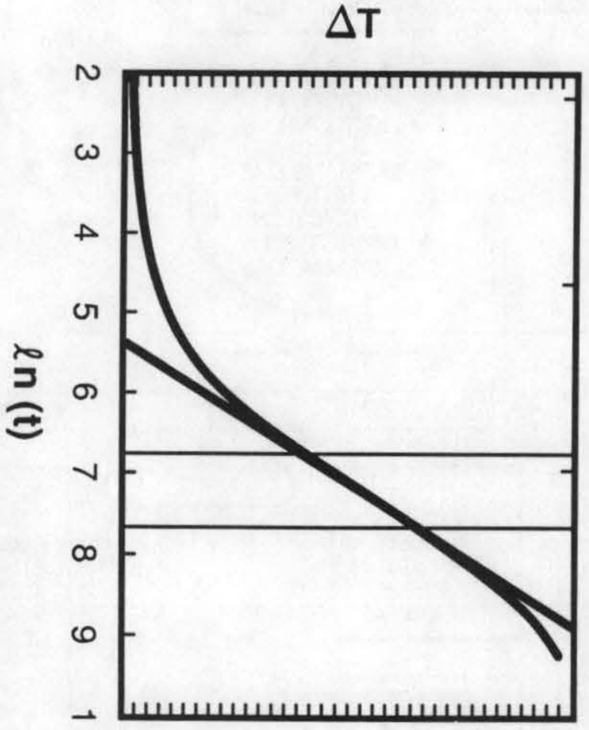

Figure 5. Probe response for CH-TRU simulated waste at ambient temperature.