CALORIMETRY METHODS FOR NUCLEAR WASTE MEASUREMENT

J. A. Mason and N. Bainbridge
A. N. Technology Ltd.
TTC, Silwood Park, Ascot, Berkshire, SL5 7PW, England

J. A. Cookson and B. Metcalfe
ANMCO, AEA Technology, Harwell Laboratory, OX11 0RA,
England

Abstract

The calorimetry technique is used for the measurement of nuclear materials by virtue of the heat evolved from nuclear decay. It is widely employed for the measurement of large samples of plutonium for safeguards and accountancy and for the measurement of tritium. Although measurement sensitivity is relatively low compared to other measurement methods calorimetry is an absolute technique and it is largely unaffected by sample inhomogeneity and matrix effects. Despite longer measurement times the technique has a role in the measurement of waste for management and to a lesser extent for safeguards. The present paper presents the results of a preliminary heat-transfer study in which different calorimetry measurement methods have been examined. A time-dependent heat-transfer analysis has been performed using models of both heat flow and isothermal calorimeters for 208 l waste drums. End point prediction has been studied as a method for reducing measurement times.

1. Introduction

This paper considers the application of the calorimetry technique to the measurement of concreted medium activity waste (MAW) drums, and by implication, to the measurement of high activity waste (HAW). Although a role for calorimetry in radioactive waste management and in safeguards for waste has not been defined, the technique has considerable potential and the present preliminary study is intended to explore some possible approaches to the application of calorimetry in this area. Calorimetry would appear to be the most effective method for measuring waste drum heat output prior to final disposal, and this possibility needs to be explored further. When combined with isotopic data a calorimetry measurement provides an upper bound for the plutonium content of a waste drum. As an absolute technique, calorimetry may be used for safeguards purposes and it can also be used as a means of criticality assessment prior to transport or storage.

2. Measurement Methods and Characteristics

Although many variants of the calorimetry method exist, three basic modes of operation relevant to the measurement of nuclear decay may be identified/1,2/. The adiabatic method has been used for the measurement of radiation absorbed dose as well as in many other applications. In the model calorimeter represented in Fig. 1 adiabatic conditions are achieved by matching the surface temperature of the calorimeter measurement chamber to the surface temperature of the drum. An automatic control system is used to maintain the measurement chamber temperature at the drum surface temperature. In this way there is negligible heat exchange between the waste drum and its surroundings and any rise in temperature of the waste drum is due solely to the internal generation of thermal energy. The process is represented mathematically as follows:

$$\frac{dH}{dt} = m\,c_p\,\frac{dT}{dt} \qquad (1)$$

where:

$\frac{dT}{dt}$ is the rate of rise of the temperature of the drum (after initial perturbations have died away),

m is the mass of the drum,

c_p is the specific heat of the drum, and

$\frac{dH}{dt}$ is the rate of thermal energy generation in the drum.

The resulting linear rate of temperature rise of the drum is proportional to the rate of thermal energy generation. Unfortunately the mass and the specific heat must be known and the absence of this information in the case of waste makes this calorimetry method generally inapplicable.

The heat flow calorimetry method is the most commonly used for nuclear materials measurement. In heat flow mode the calorimeter measurement chamber in Fig. 1 is maintained at constant temperature. As thermal energy is generated in the sample or waste drum, the drum temperature rises until the rate of heat loss to the measurement chamber wall matches the rate of thermal energy generation. The process is described by the following equation:

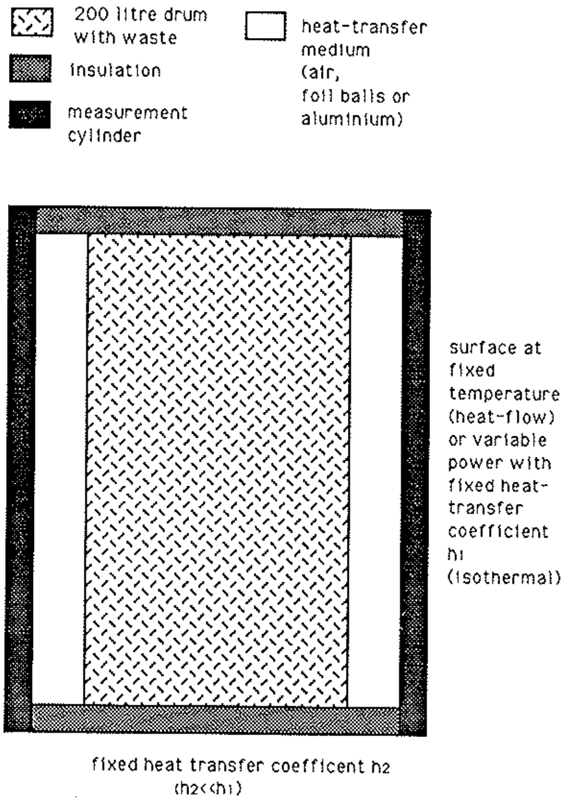

Fig1 Schematic Diagram of Waste Calorimeter for Heat-transfer Analysis

$$\frac{dH}{dt} = K\ (T_S - T_C) \qquad (2)$$

where:

K is the thermal conductivity of the coupling medium between the wall of the drum and the inner wall of the calorimeter measurement chamber,

T_S is the surface temperature of the waste drum, and

T_C is the temperature of the measurement chamber wall.

The nuclear heat generation rate is determined by establishing a calibration linking the thermal energy generation rate to the temperature difference $T_S - T_C$.

In the heat flow method the size of the temperature difference (the measured quantity) and the time constant are dependent on the value of the thermal conductivity of the coupling medium. Measurements using this method generally take a long time as the time constant is also a function of the thermal conductivity and heat capacity of the materials (and packaging) in the waste drum.

In the isothermal calorimetry method the calorimeter measurement chamber is again maintained at constant temperature. In this case, however, as heat is evolved from the sample the automatic control system reduces the electrical power supplied to the measurement chamber and the applied electrical power is replaced by thermal power from the sample. In its simplest form the thermal power of the sample is determined by noting the difference in applied electrical power before and after sample (drum) insertion. This method is generally faster than the heat flow method as the time constant is dominated by the time constant for the calorimeter, not for the waste drum.

End point power prediction is a method for reducing measurement times for both heat flow and isothermal calorimeter methods. Both the rise of drum temperature (heat flow) and the decay of measurement chamber applied electrical power (isothermal) can be approximated by a single exponential function as follows:

$$Y = A(1 \pm e^{-\lambda t}) \qquad (3)$$

where:

A is the final temperature (heat flow) or the final power (isothermal), and

λ is the coefficient of the exponential function (decay constant).

The prediction process involves fitting the appropriate exponential function to the measured data once perturbations from the insertion of the drum have died away. Significant reductions in the measurement time (often of greater than a factor 20 or more) are possible using the prediction approach.

3. Modelling and Analysis

Calorimeters for measuring 208 l concreted waste drums have been modelled employing both the heat flow and isothermal modes of operation. The cylindrical geometry two dimensional time dependent conduction heat-transfer

Table 1
Heat Flow Calorimeter Simulations

Sample Power (Watt)	Thermal Coupling Medium	K (W/m°C)	T rise (°C)	Predicted T rise (°C)	Msmt Time (hrs)	Error (%)
14	air	0.025	14.18	13.86	6	-2.2
14	foil	0.6	0.5719	0.5661	6	-1.0
14	Al coupling	30	0.0114	0.217	6	-
1.4	air	0.025	1.418	1.330	7	-6
1.4	foil	0.6	0.0572	0.058	7	<1
1.4	Al coupling	30	0.0011	0.022	6	-
0.14	air	0.025	0.1418	0.0310	3	-
0.14	foil	0.6	0.0058	0.0047	3	-
0.14	Al coupling	30	0.0001	-	-	-

computer code Teach-C has been used to simulate measurements for drums with different thermal energy generation rates and different thermal coupling media. Temperature and power data, from the heat flow and isothermal calorimeter models respectively, have been analysed using an exponential regression prediction procedure to estimate possible reductions in measurement times.

A number of assumptions have been made in this preliminary study. First of all the drum matrix is assumed to be homogeneous, with a uniform distribution of thermal energy generation, a density of 2.6 g/cm^3 and a specific heat capacity of 0.49 W s/g °C, representing average values for a medium activity waste (MAW) steel drum containing compacted spent fuel element hulls distributed in a cement mortar/3/. It has been demonstrated experimentally /3/ that 'hot' spots in a drum do not have a significant effect on the calorimetry result. A further assumption is that the drum and calorimeter are initially at the same temperature and that no thermal perturbation results from transferring the drum into the calorimeter.

The results of heat flow calorimeter simulations are displayed in Table 1. Simulations have been undertaken for three different sample powers and three different types of thermal media coupling the drum to the calorimeter measurement chamber. The table also displays the final equilibrium drum temperature, the predicted temperature, the time required to obtain a prediction, and the percentage error in the predicted result. Fig. 2 displays the relevant temperature rises as a function of time for drum heating rates of 1.4 W and 140 mW. Fig. 3 illustrates the temperature response of a heat flow calorimeter with a 1.4 W sample (drum heating rate) for two different thermal coupling media. It can be observed that the temperature response is dramatically altered when the coupling medium is changed.

Table 2 contains data for isothermal drum calorimeter simulations for two different heating rates and three different thermal coupling media. Again predicted results, the time to obtain a predicted result and the error in the predicted result are tabulated. Fig. 4 displays the results of three isothermal drum calorimeter simulations for a 700 mW heating rate and three different thermal coupling materials.

4. Conclusions

The results of the simulations indicate that it should be possible to measure 208 l waste drums at power levels equivalent to a plutonium content of the order of 50 g with an uncertainty of a few percent on the heat measurement. Assuming that the perturbation effect of drum insertion is small and reasonable temperature matching is possible, rapid measurements of a few hours are indicated, typically 6 or 7 hours. Even more rapid measurements may be possible if larger errors in the predicted results are

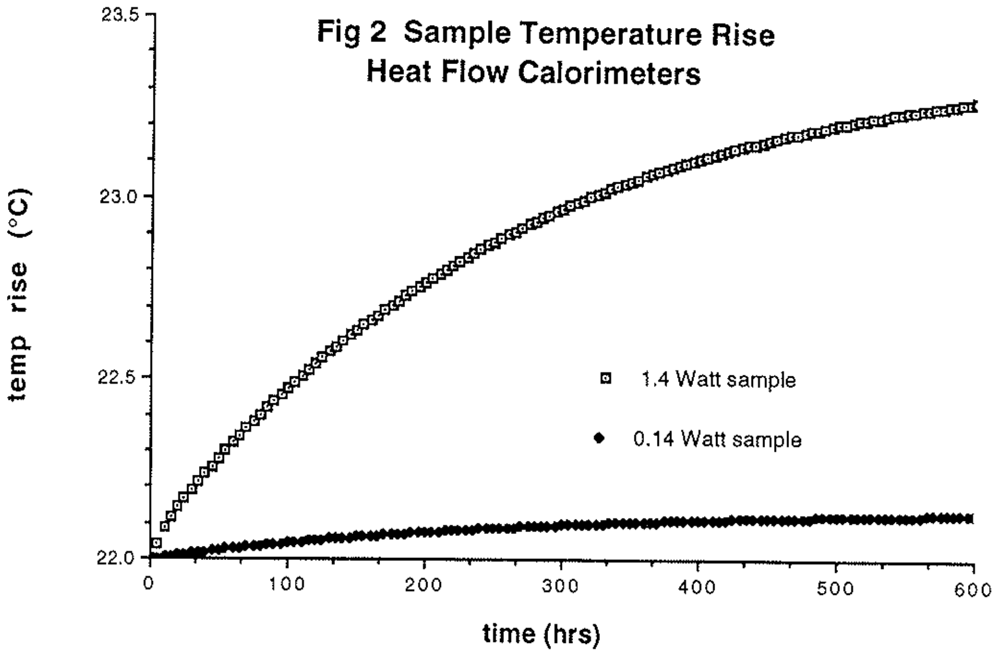
Fig 2 Sample Temperature Rise Heat Flow Calorimeters

tolerated. It should be noted that the results of these model simulations are in keeping with earlier experimental results/4/.

The variations in calorimeter response suggest that for both types of instrument an optimum value of heat-transfer coupling medium may exist which provides a measurable temperature or power difference and at the same time a sufficiently rapid predicted result. Although of a more complicated design, isothermal calorimetry would appear to be more appropriate for rapid measurements at low power levels as this method avoids the need to change the drum temperature and thus avoids invoking the thermal time constants of the waste drum.

In conclusion the results of the present preliminary study indicate that concreted waste drums can be measured by calorimetry with a heat measurement precision of a few percent and that with end point power prediction the measurements can be completed in a few hours. Further and more detailed modelling is necessary to confirm these conclusions.

5. References

/1/ J.A. Mason, "The Use of Calorimetry for Plutonium Assay", Report SRDP-R100, Safeguards R and D Project, United Kingdom Atomic Energy Authority, 1982.

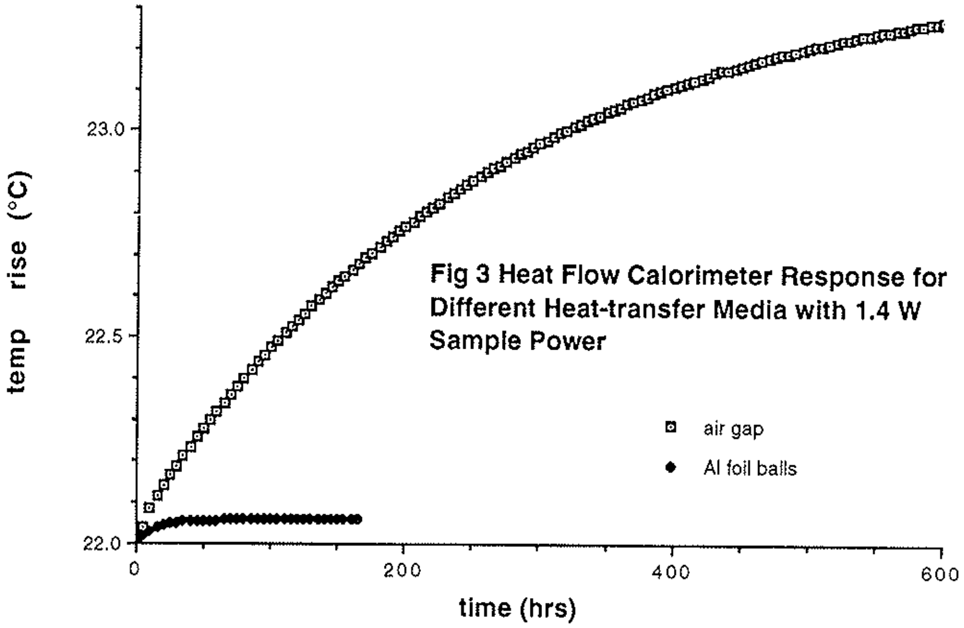
Fig 3 Heat Flow Calorimeter Response for Different Heat-transfer Media with 1.4 W Sample Power

Table 2
Isothermal Calorimeter Simulations

Sample Power (Watt)	Base Power (Watt)	Thermal Coupling Medium	K (W/m°C)	Predicted Power (Watt)	Time (hrs)	Error (%)
7	111.198	air	0.025	104.293	6	-1.3
7	111.198	foil	0.6	104.137	7	0.9
7	110.987	Al coupling	30	104.029	4	-1.2
0.7	11.119	air	0.025	10.434	7	-2.1
0.7	11.119	foil	0.6	10.422	7	-0.3
0.7	11.099	Al coupling	30	10.406	3	0.9

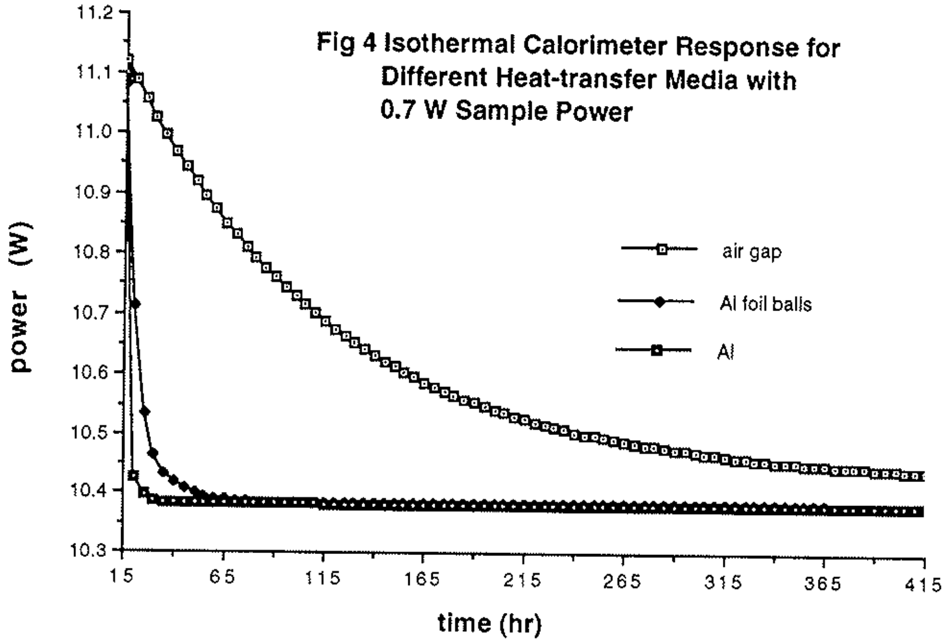
Fig 4 Isothermal Calorimeter Response for Different Heat-transfer Media with 0.7 W Sample Power

/2/ American National Standards for Nuclear Materials, "Plutonium-Bearing Solids Calibration Techniques for Calorimetric Assay", Report ANSI N 15.22-1987, American National Standards Institute, Inc., 1987.

/3/ H. Kapulla, "A Calorimeter for the determination of sources of nuclear heat in MAW (Medium Active Waste) waste drums containing cemented cladding and structural components from the reprocessing of LWR fuel elements", Report KFK 3785, Kernforschungszentrum Karlsruhe GmbH, Karlsruhe, 1984.

/4/ B. L. Taylor, B. Metcalfe, C. Wilkins and H. Devonshire, "Calorimetry for the quality checking of encapsulated ILW", Report AERE R 13415, UKAEA, Harwell Laboratory, 1990.