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Thermal Diffusivity of Liquids Determined by Flash Heating of a Three-Layered Cell

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An investigation of the applicability of the flash heating of a three-layered cell for determining the thermal diffusivity of a central liquid layer at temperatures between its melting and boiling points is described. Two different cell designs were developed. In one, the outside layers were brazed to a ring-shaped central spacer; in the other, bolted flanges held the outside layers against a central spacer. Test units were fabricated from type 304 stainless steel. Thermal diffusivity results for water obtained with two brazed cells and one bolted cell were all within ±1% of standard reference values. Results obtained with a high temperature salt (Hitec) with two brazed cells were within ±2.5% of each other over the temperature range of 149–427°C. In contrast, bolted cell measurements were inconsistent.

KEY WORDS: flash heating method; thermal diffusivity of liquids.

1. INTRODUCTION

Since the origination of the flash technique [1], a considerable amount of research has been carried out on its use for the measurement of the thermal diffusivity of pure metals, alloys, ceramics, semiconductors, two-layer composites [2–4], rocks, and expoxy resins. Only limited effort, however, has been made to utilize the technique for the determination of the thermal diffusivity of liquids and liquid metals [2, 5, 6, 7]. One of the main reasons for this is that for most liquids, the incident energy will penetrate well below the surface and some may even be transmitted. As a result, the accurate specification of the heat input is extremely difficult. Interest in applying the flash method to liquids has therefore led to consideration of a three-layered

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cell in which the test liquid is enclosed between two thin solid layers with known thermal properties [2, 7]. A hazardous material could also be safely mounted as the middle layer in such an arrangement.

Some initial work with a three-layered liquid-filled cell in a flash system was carried out by Araki and Taylor [7]. This included the design of a low-temperature cell in which water and some organic liquids were tested at room temperature. The analytical solution developed by Lee [2] for the rear surface temperature response to an instantaneous heat pulse input at the front surface was used to calculate values of the thermal diffusivity from rear surface temperature measurements. On the basis of the results obtained by Araki and Taylor [7], the flash method with a three-layered system appeared promising for measurements of the thermal diffusivity of liquids at high temperatures. The present study was undertaken to investigate this application.

2. THERMAL DIFFUSIVITY FROM THREE-LAYERED CELL TEMPERATURE RESPONSE

Determination of a thermal property by the flash method test involves evaluating by an iterative procedure (for two- and three-layered composites) a value that yields agreement between the temperature predicted from the appropriate solution and the experimentally measured response. Common practice is to measure the temperature at the rear surface of a cell and to match predicted and measured values at the time when this temperature has reached 50% of its maximum rise. Detailed derivations of solutions for applicable heat input functions to a three-layered cell as illustrated in Fig. 1 are given in refs. [2] and [8]. In ref. [2], the rear surface response to an instantaneous front surface pulse is used in the convolution integral to obtain relations for representative time varying front surface heat inputs. An alternate procedure followed in ref. [8] is to use the solution for a unit-step

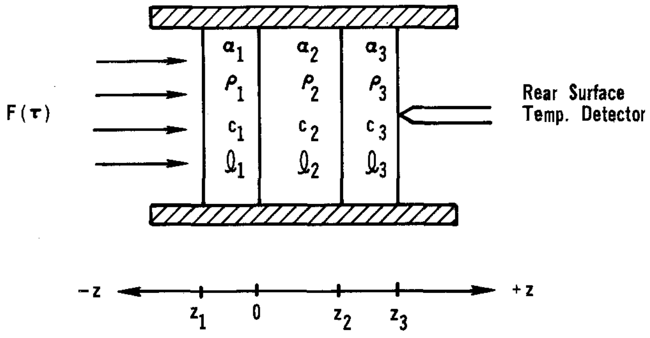

Fig. 1. Diagram of a three-layered cell.

function in a Duhamel integral. Expressions representing the heat input from a laser pulse and the resulting rear surface temperature variation are summarized below.

Assuming the fraction of energy absorbed by the front surface of a cell to be independent of the pulse amplitude, the rate of energy absorbed by the front surface irradiated by a typical heat pulse from a laser powered by discharging capacitors through a xenon flash lamp can be shown to be

$$Q(t) = \frac{q_0 A t}{t_p^2} e^{-t/t_p} \tag{1}$$

where $Q(t)$ is the rate of energy absorption at the front surface of the cell, q_0 is the total energy absorbed by unit area of the front surface, and A is the area of the front surface normal to the heat pulse, t is the time, and t_p is the time when $Q(t)$ reaches a maximum.

The normalized expression for the rear surface temperature variation of a three-layered cell heated at the front surface by an exponential heat pulse described by Eq. (1) can be written as

$$V_3(z_3, \tau)_{\text{exp}} = 1 - e^{-\tau/t_p} - \frac{\tau}{t_p} e^{-\tau/t_p} + 2 \sum_{k=1}^{\infty} \cdot \frac{(x_1\omega_1 + x_2\omega_2 + x_3\omega_3 + x_4\omega_4)\, Q_{\text{exp}}}{x_1\omega_1 \cos(\omega_1\gamma_k) + x_2\omega_2 \cos(\omega_2\gamma_k) + x_3\omega_3 \cos(\omega_3\gamma_k) + x_4\omega_4 \cos(\omega_4\gamma_k)} \tag{2}$$

in which

$$Q_{\text{exp}} = \frac{(\gamma_k^2 \tau/\eta_3^2) e^{-\tau/t_p} - (\tau/t_p) e^{-\tau/t_p} - e^{-\tau/t_p} + e^{-(\gamma_k^2/\eta_3^2)\tau}}{[(\gamma_k^2 t_p/\eta_3^2) - 1]^2} \tag{3}$$

where τ is the time, the X_i's, η_i's and ω_i's are convenient groupings of the thermal properties of the cell materials, and the γ_k's are the poles of a function that must be inverted from the Laplace to the real time domain (a detailed derivation of this result is given in ref. [8]). When the heat pulse from the laser is very rapid, its effect is equivalent to an instantaneous or Kronecker delta function input. That is, $Q(t) \rightarrow \delta(t)$ for $t_p \rightarrow 0$, and Eq. (2) reduces to

$$V_3(z_3, \tau)_{\text{inst}} = 1 + 2 \sum_{k=1}^{\infty} \cdot \frac{(x_1\omega_1 + x_2\omega_2 + x_3\omega_3 + x_4\omega_4)\, e^{-(\gamma_k^2/\eta_3^2)\tau}}{x_1\omega_1 \cos(\omega_1\gamma_k) + x_2\omega_2 \cos(\omega_2\gamma_k) + x_3\omega_3 \cos(\omega_3\gamma_k) + x_4\omega_4 \cos(\omega_4\gamma_k)} \tag{4}$$

Note that the above expressions involve the thermal properties of all the three layers along with the eigenfunctions (γ_k's) in a summation form. Hence it is necessary to use an iterative procedure to evaluate the unknown value of α_2, i.e., the thermal diffusivity of the middle layer. A data reduction computer program to calculate α_2 and k_2 based on Eq. (2) is given in ref. [8].

3. DESIGN OF A THREE-LAYERED LIQUID CELL

3.1. Effects of Errors in Cell Wall Thermal Properties

The unknown value of α_2 that will be calculated using Eq. (2) depends on the accuracy of values specified for the thermal properties of the side walls and two thermal properties of the central layer. Although the latter two properties will influence results, the quantities over which the cell designer has control are the side wall materials and thicknesses and the central layer thickness. Calculations were therefore first made to investigate the relative importance of each of these factors. The general approach was to determine the error in the central layer thermal diffusivity α_2 for side walls of specified thickness and varying errors in thermal properties.

The influence of side wall thermal conductivity was found to be negligibly small. This is due to the fact that the thermal diffusivities of liquids are low relative to metals. Hence the conduction process in a cell is dominated by

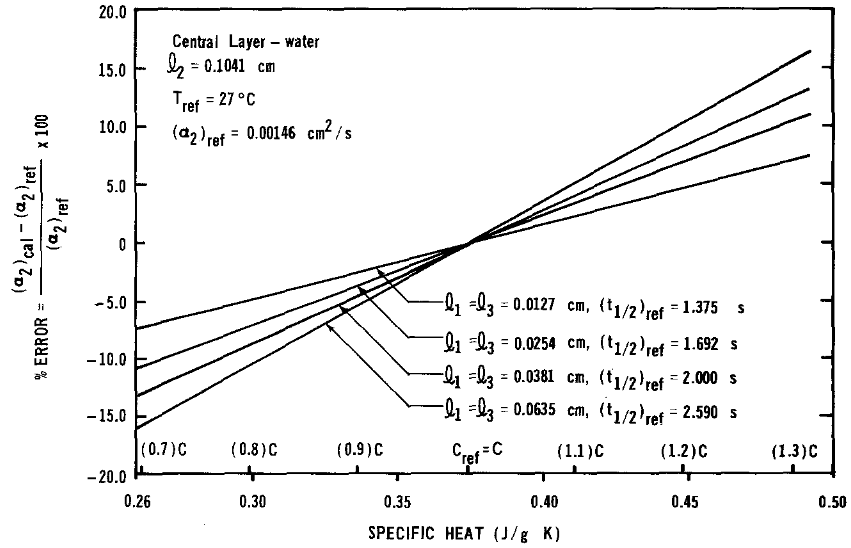

Fig. 2. Effect of error in specific heat capacity of side layers on calculated diffusivity α_2.

the central layer and temperature variation in the side layers at any time is comparatively small.

Since side wall thicknesses can be accurately measured and candidate material densities are quite well known, the most probable source of error was considered to be the specific heat capacity of the side wall materials. Taking thicknesses of 0.0127, 0.0254, 0.0381, and 0.0635 cm (0.005, 0.010, 0.015, and 0.025 in.) as representative, calculations of the error in the diffusivity of water layers 0.0762 and 0.1041 cm thick at room temperature were made for assumed errors in the specific heat capacity of a number of possible side wall materials. Representative results are shown in Fig. 2. As anticipated, the error decreases with decreasing wall thickness. It is less sensitive to central layer thickness, but was found to be higher for thinner layers. An encouraging point is that approximately 5% accuracy can be achieved even if the specific heat capacity data is in error by as much as 10%.

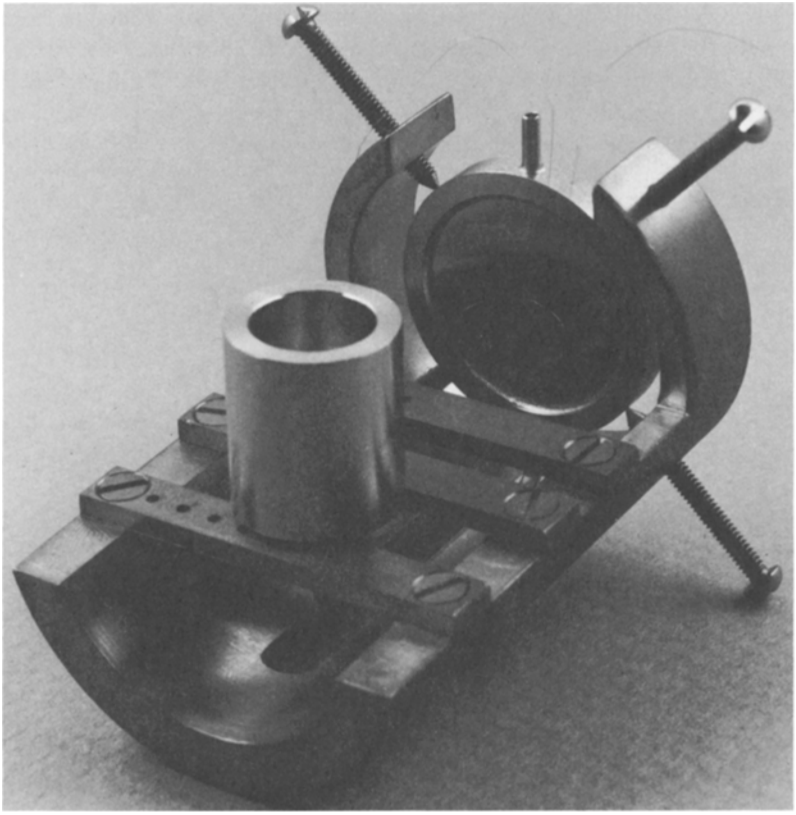

Fig. 3. Brazed cell with reservoir mounted in furnace fixture.

After consideration of a number of materials, use of stainless steel was decided upon. It is readily available in the form needed, thermal property data variation with temperature is relatively well documented, and it can be readily machined and fabricated. Two different cell designs were developed. They will be referred to as (a) brazed, and (b) bolted, since these terms describe distinguishing features.

3.2. Brazed-Cell Design

An example of this design is shown in Fig. 3. It is mounted in a brass holder which fits inside the central ceramic tube in the furnace in which tests were conducted. Typical dimensions of the three-layered section are given in Fig. 4.

Central layer thicknesses and side wall thicknesses were selected partly on the basis of the preliminary calculations described and partly from practical considerations of handling and fabrication. The cell body was first machined from bar stock. This included forming one side wall. The second side wall, fill, and overflow tubes were then brazed to the cell body using a nickel-gold braze, which required heating to above 900°C. Rear surface temperatures were measured with 0.005 in. diameter Chromel-Alumel thermocouple wires spot welded to the rear wall.

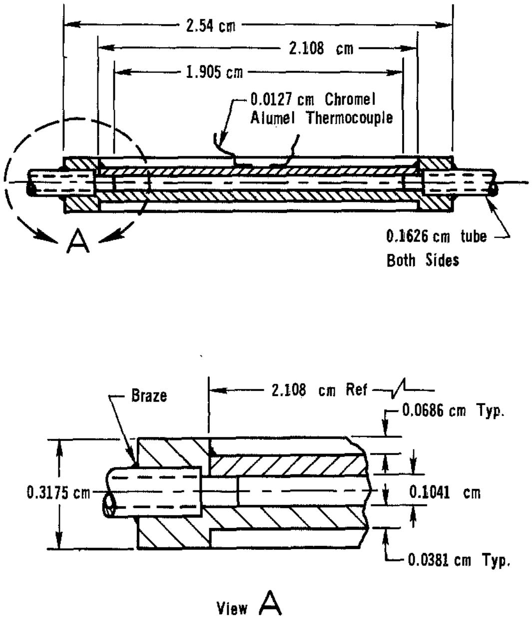

Fig. 4. Cross-section of a brazed cell.

As shown in Fig. 3, cells were designed for testing in a vertical position. Operation with test cells mounted in a horizontal position was initially considered. However, this would have complicated the optical system and its alignment, and available information suggested that the convection effects in a vertical cell would be negligible. For a 1 mm thick central layer filled with water at 65°C, the Grashof number based on layer thickness is only about 300 for a temperature difference of 10°C. Experiments indicate that for Grashof numbers (based on thickness) less than 2000, heat transfer is by simple conduction [9]. In view of this, it was decided that testing of cells in a vertical position should not introduce any error. Results indicated that this assumption was satisfied.

3.3. Bolted-Cell Design

The special fixtures and handling required for brazing of the cell just described prompted the development of a bolted design which also eliminated the fill reservoir (the small vertical cylindrical vessel seen in Fig. 3). As illustrated in Fig. 5, this consisted of a central spacer with two outer flanges with knife edges. Disks (side walls) were placed on either side of the central spacer. The knife edges were forced against the disks to achieve sealing by tightening eleven bolts spaced uniformly around the cell body. The V-shaped void in the central spacer served both for filling and as a small reservoir for the test liquid.

4. EXPERIMENTAL APPARATUS AND PROCEDURE

The experimental system employed is shown schematically in Fig. 6. For specimen pulse heating, a xenon flash lamp powered neodymium laser was used. The liquid test cells were mounted in air in the central ceramic tube of an electrically heated furnace. During testing, both ends of the ceramic tube were plugged (except the forward opening during the laser pulse) to reduce free convection effects. The laser and tube furnace were mounted atop an optical bench for convenience in handling and alignment of the laser beam with the cell. Uniform heating of the front surface of the test cell was achieved by placing it in the central region of the expanded laser beam. This required aligning the central axis of the tube furnace with the central axis of the neodymium laser rod, and this was accomplished with a small helium-neon laser mounted behind the furnace, as shown in Fig. 6. The continuous beam from this laser was directed through the furnace and neodymium laser rod. The reflected beam from the rear spherical mirror of the neodymium laser was then made coincident with the incident beam by small adjustments in the alignment of the neodymium laser. To further improve uniformity, a concave lens of 250 mm focal length was introduced approximately halfway

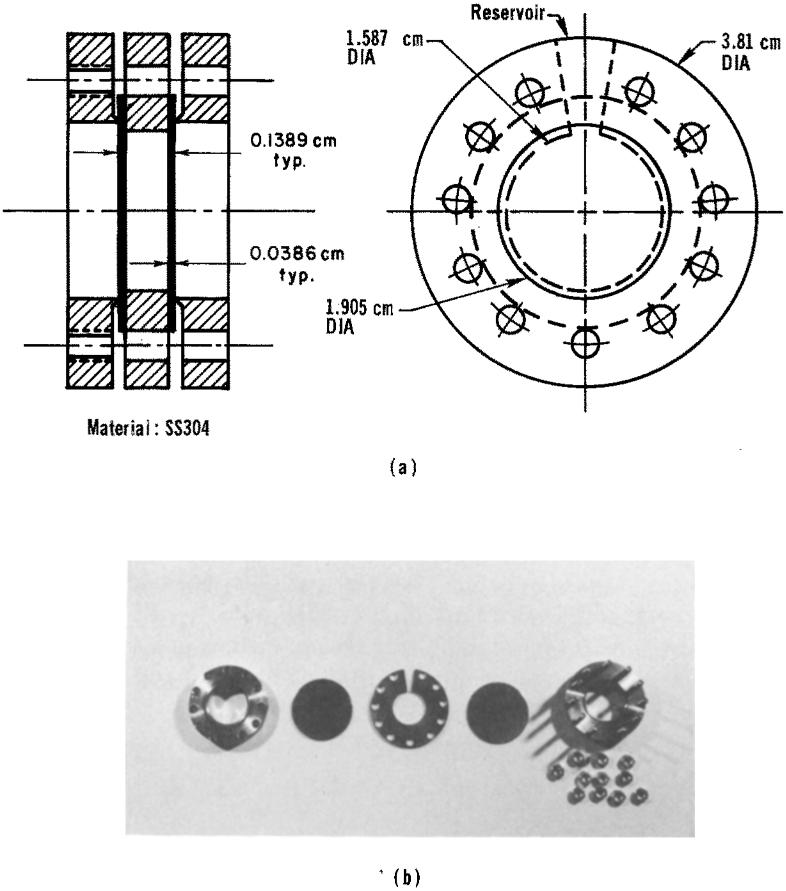

Fig. 5. Bolted cell; (a) section and (b) components.

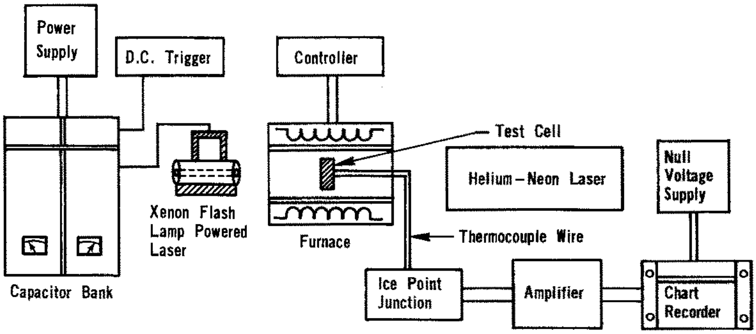

Fig. 6. Schematic of apparatus arrangement.

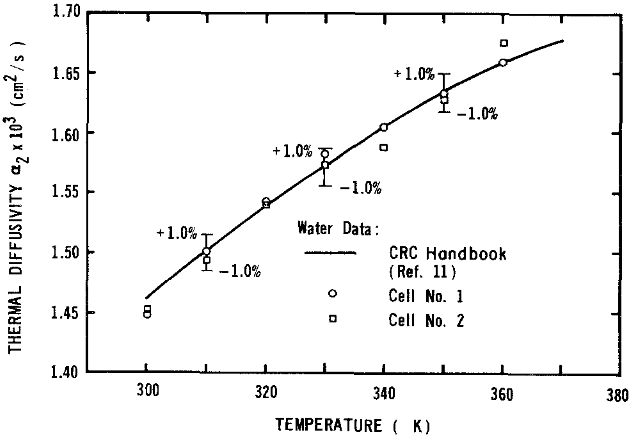

Fig. 7. Comparison of measured with reference values of α for water (brazed cell).

between the neodymium laser and the furnace. This expanded the beam to approximately 3.2 cm in diameter, making it larger than the cell diameter, which provided even heating of the front surface. Test exposures of sensitive papers mounted at the cell location in the furnace indicated a uniform central region of 2.8 cm in diameter. A more detailed description of the system components and test procedures is given in ref. [8].

5. EXPERIMENTAL RESULTS

Data were first taken using deionized water to check the operation of the system and verify the accuracy of the cell dimensions and thermal properties.

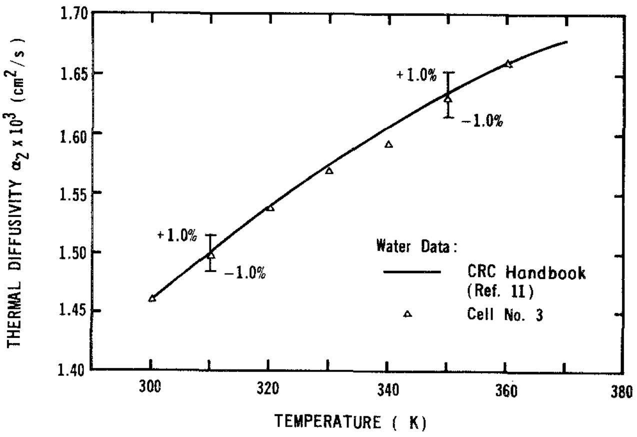

Fig. 8. Comparison of measured with reference values of α for water (bolted cell).

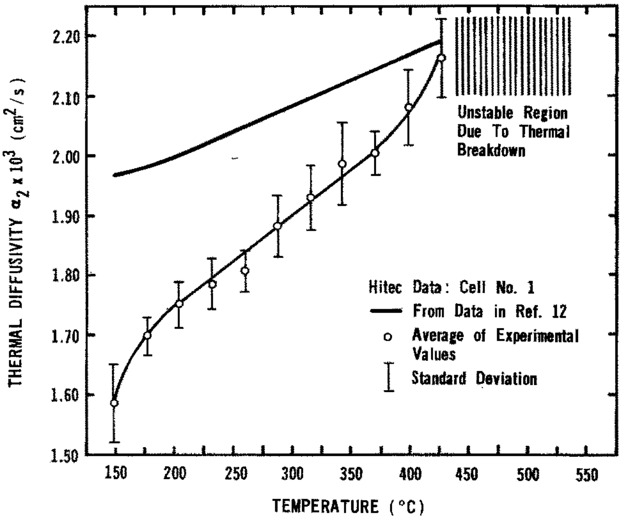

Fig. 9. Thermal diffusivity of Hitec salt vs. temperature from cell No. 1.

At each temperature, between 15 and 20 tests were made. The final thermal diffusivity values were calculated by first taking an average of all the experimental half-time ($t_{1/2}$) values per reference ambient temperature and then reading these values along with other known thermophysical properties of the cell into the data reduction computer program. The thermophysical properties of the side wall materials were taken from TPRC handbook [10].

The diffusivity values of water so obtained using two cells of the brazed design shown in Figs. 3 and 4 are plotted in Fig. 7. The cell designated No. 1 had outer layers 0.038 cm (0.015 in.) thick; the central layer was 0.104 cm (0.041 in.) thick. Cell No. 2 had outer layers 0.063 cm (0.025 in.) in thickness and a central layer 0.114 cm (0.043 in.) thick. Deviations of the measurements in Fig. 7 from standard reference values [11] are within ±1%.

The thermal diffusivity values of water obtained from a representative bolted cell (see Fig. 5) are plotted in Fig. 8. These results are contained within −0.8% and +0.1% by comparison with thermal diffusivity values of ref. [11]. This cell is designated No. 3; it had side layers 0.039 cm (0.014 in.) thick and a central layer 0.138 cm (0.055 in.) thick.

A high temperature heat transfer fluid was selected to investigate the applicability of the three-layered cell concept for thermal diffusivity measurements of liquids at high temperatures. The particular one chosen is a eutectic mixture of sodium nitrite (40%), sodium nitrate (7%), and potassium nitrate (53%), and is known as Hitec[4] [12].

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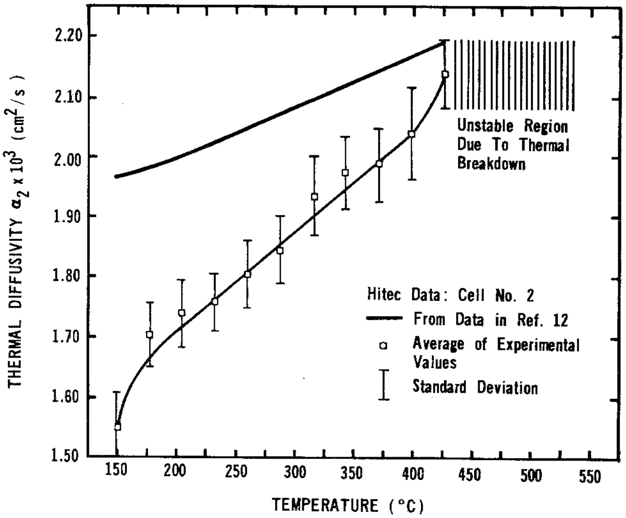

Fig. 10. Thermal diffusivity of Hitec salt vs. temperature from cell No. 2.

In the graphical presentation of results from cells No. 1 and 2, in Figs. 9 and 10, each data point represents the average of between 30 and 40 experimental half-time values per reference temperature per cell. Figure 11 presents a combined plot of the thermal diffusivity results for both cells versus temperature. A straight line drawn through these points appears to be a satisfactory representation of the variation of the experimental average values with temperature.

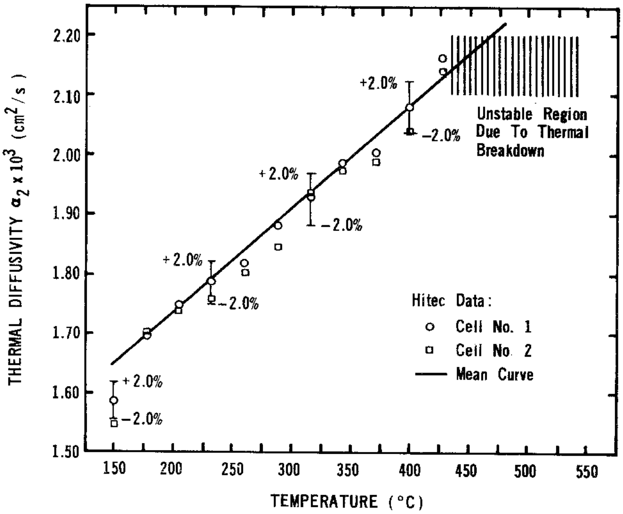

Fig. 11. Comparison of measured values of α for Hitec.

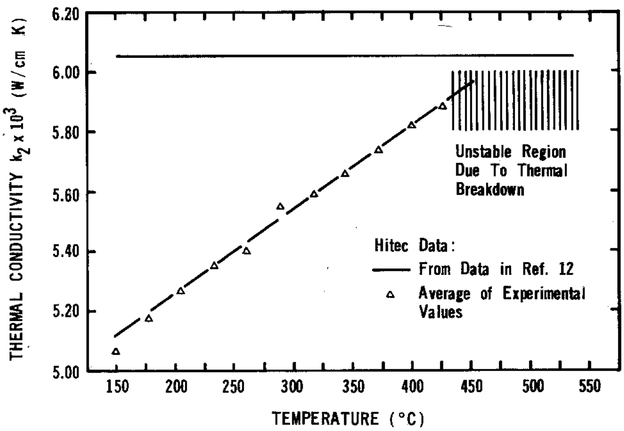

Fig. 12. Thermal conductivity of Hitec salt vs. temperature from $k = \alpha_{avg}\rho c$, where α_{avg} values are taken from Fig. 11.

The temperature dependent thermal conductivity values of Hitec were calculated ($k_2 = \alpha_2\rho_2 c_2$) by using the average of experimental thermal diffusivity $(\alpha_e)_{avg}$ values from Fig. 11 and values of ρ_2 and c_2 from ref. [10]. The results are plotted in Fig. 12 along with a curve showing the constant thermal conductivity value given in ref. [12]. Table I compares the final experimental thermal diffusivity values from Fig. 11 with values given in ref. [12] and [13]. It is interesting to note that the deviation is negative and

Table I. Comparison of Thermal Diffusivity Results With Reference Values

Temp. (°C)	$(\alpha_e)_{avg}$ [a] ($cm^2 s^{-1}$)	$(\alpha_R)_1$ [b] ($cm^2 s^{-1}$)	$(\alpha_R)_2$ [c] ($cm^2 s^{-1}$)	Deviation (from ref. [12]) (%)	Deviation (from ref. [13]) (%)
149	0.00165	0.00197	0.00144	−16.4	+14.8
177	0.00170	0.00199	0.00144	−14.5	+18.1
204	0.00174	0.00200	0.00144	−13.0	+21.1
232	0.00179	0.00203	0.00144	−11.6	+24.5
260	0.00183	0.00205	0.00142	−10.8	+29.2
288	0.00189	0.00207	0.00137	−9.0	+37.6
316	0.00194	0.00210	0.00133	−7.7	+46.0
343	0.00198	0.00212	0.00128	−6.6	+54.9
371	0.00203	0.00214	0.00123	−5.3	+65.2
399	0.00208	0.00217	0.00118	−4.0	+76.3
427	0.00213	0.00219	0.00113	−2.9	+87.7

[a] Average value of experimental α from Fig. 11.
[b] Calculated from ref. [12].
[c] Calculated from ref. [13].

decreases linearly with increasing temperature from the data in ref. [12], but exactly the reverse is true for the data from ref. [13]. Reasons for these differences along with an explanation for the unstable thermal region above 450°C shown in Figs. 9–12 will be advanced in the following section.

Tests were also conducted with Hitec salt in bolted-design cells. Experimental half-time values for early tests at temperatures less than 100°C above melting appeared consistent with values from cells No. 1 and 2. However, with increasing temperature, half-time values increased to 20% and more than the values obtained with the brazed-design cells. Inspection of the bolted cell revealed that in some cases some outward bowing of one of the side walls had occurred. In others, it appeared that heat loss through the knife-edges to the relatively heavy outer rings of the cell may have been responsible. Redesign of the central spacer so as to cause the side walls of the bolted-design cell to be in tension was considered. However, in view of the relatively large thermal capacity of the outer rings, it was decided that even though this design had several attractive features (ease of filling, simplicity of changing side walls), further development was not merited.

6. DISCUSSION AND CONCLUSIONS

On the basis of the accuracy and consistency of the experimental results obtained in this investigation, it is concluded that the flash method is suitable for measuring the thermal diffusivity or thermal conductivity of liquids at elevated temperatures using the three-layered cell concept. For water (Figs. 7 and 8), the maximum deviations of −1.0% at 300K for cell No. 1, −1.1% at 340K for cell No. 2, and −0.8% at 340K for cell No. 3 from reference values [11] are considered to show excellent agreement of the experimental values with reference values and also among the three test cells. For temperatures up to 450°C, it can be seen that the experimental results obtained for the Hitec salt using cells No. 1 and 2 are also quite consistent and in good agreement with one another over the complete experimental temperature range. The scatter of average data values about the mean values is a maximum of +2.5% and a minimum of −0.3%. Variations at a given test condition are thought to be primarily due to small but differing convection effects from the air in the ceramic tube surrounding the test cell. These could be eliminated by conducting tests in a vacuum.

In addition to random errors, it is recognized that heat leakage from the front layer of a brazed cell through the outer ring to the rear layer could introduce a systematic error. The excellent agreement between measured and reference diffusivity values for water suggests that this effect was negligible. As an explanation for this, it is first noted that the thermal capacity (ρc) of the outer ring is approximately the same as that of water or Hitec, and this region is exposed to the same heating pulse as the central three-layer region.

Hence sufficient energy is supplied to raise it to about the same final temperature as the main part of the cell. Due to the much higher diffusivity of the outer ring material, however, the incident energy will diffuse throughout it in a relatively shorter time, and some heat flow from the front to the rear layer can occur. The energy involved will come from the outer or large radius regions of the front layer. Because of this, the effect on temperatures at the center, where measurements are made, is a minimum.

An additional reason for the heat leakage through the cell outer ring being negligible can be seen in Fig. 13. Here it can be seen that the calculated front surface temperature based on one-dimensional flow decreases very rapidly after reaching its maximum. Within 1 s after the heat pulse, it has already dropped to about 0.4 to 0.5°C above the final equilibrium temperature of the cell. This means that the temperature potential for heat leakage was small during a major portion of the approximately 2.5 s period in which one-half the rear surface temperature rise occurred. Recognizing that the above discussion of heat leakage is qualitative, a computer analysis of the multidimensional heat flow is planned to obtain quantitative information.

The experimental thermal diffusivity results differ from values calculated ($\alpha = k/\rho c$) from the data for k, ρ, and c given in ref. [12], by a maximum of -16% at the lower end of the temperature range (149°C). This difference can be attributed to the suggested constant thermal conductivity value of 0.00605 W/cm K over a temperature range of 200 to 400°C. A constant value of k for a liquid salt is probably not correct over this temperature range.

Thermal property data for Hitec salt are also given in ref. [13]. These

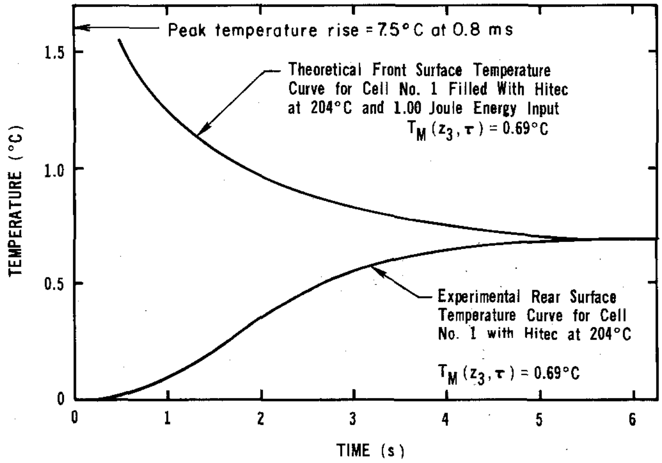

Fig. 13. Theoretical front surface and experimental rear surface temperature variations with time.

values differ significantly from the present experimental results by 15% at 149°C, increasing to a maximum of 88% at 427°C. This difference is attributable to the fact that the thermal conductivity decreased with increasing temperature. Since the values for ρ and c in refs. [12] and [13] are in good agreement, the differences noted are considered to be due to the thermal conductivity values. Also, since values given for ρ and c appear to be appropriate, the mean curve in Fig. 11 is recommended for the variation of thermal diffusivity of Hitec salt with temperature.

In the operating instructions for Hitec given in refs. [12] and [13], it is indicated that small changes in the composition of this salt occur and result in an increase of its melting point, when used in an open system in contact with air. Results attributable to this were observed when taking data at 149°C, which is 7°C above its specified melting temperature (142°C). Rear surface temperature variations showed a slow rise followed by an unusually long equilibrium time after the heating pulse. This suggested that the salt was not in a complete liquid form, and the diffusivity value determined may be a little lower than the true value at this temperature.

Another region of interest lies above 450°C, shown as unstable in Figs. 9–12, where a slow thermal breakdown of nitrite to nitrate, alkali metal oxide, and nitrogen gas occurs [12]:

$$5\,\mathrm{NaNO_2} \rightarrow 3\,\mathrm{NaNO_3} + \mathrm{Na_2O} + \mathrm{N_2}\uparrow \qquad (5)$$

At temperatures above 450°C, the rear surface temperature rose more rapidly after the heating pulse and did not remain at a maximum value for even a short time. These trends suggest increased heat transfer through the liquid layer, which could be caused by chemical changes occurring in the liquid.

The measurements on water above approximately 85°C were not considered reliable. At this point, it appears that some boiling along the inner side of the heated wall occurs. This is to be expected, because the energy to raise all three layers to a readily measurable amount must initially be deposited in the front layer. Since this layer is relatively thin, its initial temperature rise will be several degrees. Recognizing that this effect will be encountered at even lower temperatures, if a very thin front layer is used, leads to the conclusion that the thinner the cell side layers, the smaller will be the applicable temperature range of the cell.

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