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NOMENCLATURE

ΔU_{298} = heat of solution at constant volume and at 298° K., kcal./mole
ΔH_{298} = heat of solution at constant pressure and at 298° K., kcal./mole
$\Delta H_{f\,298}$ = enthalpy of formation at 298° K., kcal./mole
q = electrical energy supplied to the calorimeter, calories
E_s = voltage drop across standard resistor, volts
R_s = 0.500 ohm standard wire-wound resistor
R_h = heater resistance = 23.96 ohms
$t_{sec.}$ = heating time, seconds
E = energy equivalent, calories/ohm
ΔR = change in the resistance of the thermistor

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Thermal Conductivities of Aluminum and Zinc Powder Suspensions

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Thermal conductivities are presented for suspensions of powders that are highly conducting compared with the continuous phase. Aluminum and zinc in varying particle sizes and shapes dispersed in lubricating grease in amounts of up to 80% by weight comprised the suspensions. A comparison of experimental results with values predicted from a number of correlating expressions showed generally satisfactory agreement at the lower concentrations but considerable disparity at the higher concentrations. The experimental apparatus and technique are briefly described.

THE THERMAL CONDUCTIVITY of two-phase systems is a function of numerous variables and is difficult to predict, particularly when the thermal conductivity of the two phases differ by over two orders of magnitude. Experimental data are therefore desirable. Results are reported herein for several systems consisting of metal powder dispersions in lubricating grease and comparisons are made with analytical expressions.

EXPERIMENTAL

Materials. Aluminum and zinc powders with varying size and shape particles were employed as the dispersed phase and Marfak No. 1 lubrication grease of the Texas Company served as the continuous phase. One aluminum powder was composed of irregular particles having a mass mean diameter of 40μ and a geometric standard deviation of 2.0. Three other aluminum powders, hereafter referred to as aluminum A, aluminum B, and aluminum C, had mass mean diameters of 33, 40, and 53μ and geometric standard deviations of 2.5, 1.8, and 1.5, respectively. Two zinc powders had irregular particles of 60 and 80μ in mean diameter with standard deviations of 1.4 and 1.6, respectively. The thermal conductivities of zinc and aluminum are 64 and 118 B.t.u./hr. ft. ° F. (*2*), respectively, while the grease had a conductivity of approximately 0.16 B.t.u./hr. ft. ° F.

Apparatus. A steady state, parallel-disk type of conductivity apparatus was employed, and with it, as nearly as possible, a constant heat flux was maintained across the test material. Figure 1 shows the principal features of the calorimeter. The upper plate was maintained at a constant and higher-than-room temperature by the circulation of hot water from a constant temperature bath through an attached coil of copper pipe. This coil was dual-wound and attached to a copper plate, as shown in Figure 2, so that the heat from the water would be distributed evenly over the plate creating an essentially isothermal surface. The bottom portion of the hot plate was separated with a thin ring of transite into two sections so that the center part of the plate would be guarded against adverse temperature effects at the outer periphery of the plate. The chamfer at the bottom of the hot plate provided an escape for entrained air from beneath the hot plate during assembly with suspension in place. It was essential that the heat conducting medium contact the hot plate at all points.

The calorimeter was encased in a transite shell, the ends

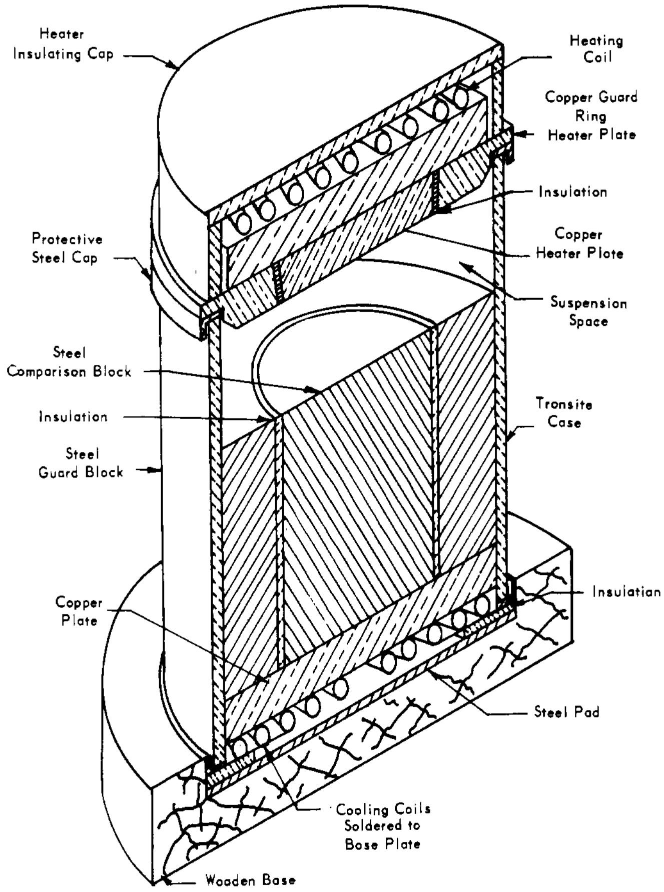

Figure 1. Calorimeter detail

of which were capped with steel rings to prevent mechanical damage. Transite was selected because its thermal conductivity is much lower than metallic construction materials. Furthermore, neither its strength nor its thermal expansion changes appreciably with temperature as do most other construction materials. It is also machinable. The case was 9.75 inches long with an inside diameter of 6.09 inches and a wall thickness of 0.25 inch. The case was insulated externally with approximately 2 inches of asbestos fiber.

The test material occupied the region, designated as the suspension space, between the hot plate and a comparison block. This space had a maximum thickness of 2 inches; it could be altered by inserting machined spacers to elevate the comparison block.

The comparison block of cold rolled steel afforded a measure of the heat flux through the calorimeter. The axial temperature gradient through it was determined, and then the heat flux was calculated from its thermal conductivity. The block was 6.0 inches in diameter and 6.0 inches in length; it had a thermal conductivity of 19.3 B.t.u./hr. ft. ° F., which was determined by experiment. As with the hot plate, the comparison block was separated with a thin layer of transite ⅛ inch thick into the inner test section and an outer guard ring section.

Temperature measurements were taken with a precision potentiometer (Leeds and Northrup Co.) and copper-constantan thermocouples. The thermocouples were each calibrated by checking against a National Bureau of Standards certified thermometer at 5 degree intervals over a temperature range from 20° to 75° and also at the ice point of water. After calibration, the thermocouples, were installed. At the top of the calorimeter, five thermocouples were imbedded near the undersurface of the hot plate. The thermocouples, three of them being situated within the protected center section of the plate and two being in the guard ring, were positioned one at the center and four along a radius ¾, 1½, 2, and 2¾ inches from the center of the plate. The purpose of this layout was to determine the constancy of temperatures over the hot plate. Glyptal (General Electric Co.), a dielectric varnish, secured the thermocouples in position with the junctions 1/32 inch from the surface and in contact with it.

Thermocouples, having the same radial distributions as those in the hot plate, were installed at two levels in the comparison block, 5 inches apart.

Seven thermocouples were positioned along the central axis of the test space for measuring the temperature gradient there. These were supported by a cylindrical plastic sleeve of slightly smaller diameter than that of the calorimeter space. Each thermocouple was displaced angularly relative to the one immediately above to minimize thermal interference. This assembly was placed in the test space first and then the suspension to be tested was spread over it.

The temperature of the water which heated the hot plate, was controlled electronically, while an ice bath served as the heat sink. Variations in the temperature of the hot and cold plates were usually maintained within $\pm 0.05°$ C., but in extreme cases 0.10° C. differences were observed. The over-all error in precision of the instrument, including the calibration of the comparison block, was assessed to be less the 4% by calorimetric measurements with pure water, the thermal conductivity of which is well established over a considerable temperature range. This information, together with the following analysis, was employed to evaluate the performance of the calorimeter.

Since heat transfer through the fluid space is principally by conduction, Fourier's relation applies, and:

$$q = k\,dT/dX \tag{1}$$

where q is the heat flux, k is the thermal conductivity of the material, and dT/dX is the thermal gradient. The thermal conductivity of water, being temperature dependent, is described by the expression:

$$k = aT + b \tag{2}$$

where a and b are constants. Combination and rearrangement of Equations 1 and 2 yield:

$$q\,dX = (aT + b)\,dT \tag{3}$$

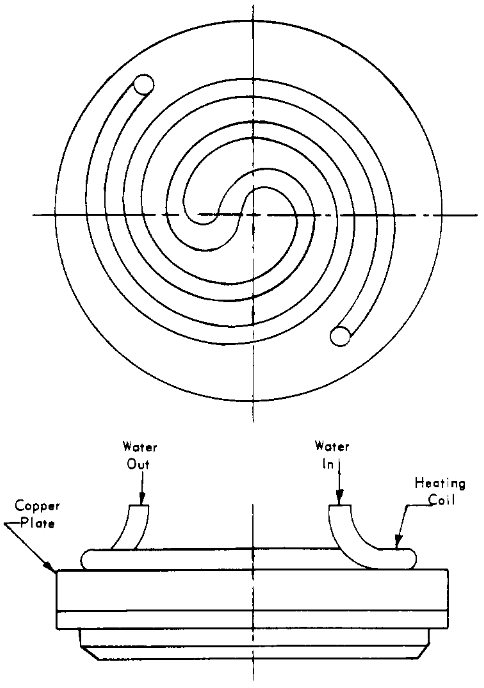

Figure 2. Schematic drawing of the dual-wound heating coil

which, upon integration, gives:

$$X = \frac{a}{2q} T^2 + \frac{b}{q} T + C \qquad (4)$$

where C is the constant of integration.

From Equation 4, the temperature distribution through the water can be calculated, provided a, b, and q are known. Values of a and b were taken (*2*) as 8.036×10^{-3} cal./hr. cm. °C.^{2} and 5.103 cal./hr. cm. °C., respectively, and the heat flux, q, calculated from the experimental data, was 57.778 cal./hr. cm.^{2} for each experiment. With these values, temperature distributions were calculated from Equation 4 and compared with the experimental data; these data, presented in Table I, are in agreement with those calculated. These results are typical of the experiments performed with pure water.

Equation 4 shows that distance *vs.* temperature is a second degree function, and comparison of Equations 2 and 4 shows that the second degree term is due to the variation of thermal conductivity with temperature. Over the temperature range considered, the variation in the case of water is about 3%, and on a plot of distance *vs.* temperature, causes a very slight deviation from a straight line. Thus it is considered that the conductivities were detected within 3%, since this precision is required to detect the nonlinearity predicted by Equation 4. When the experimental error introduced by use of the comparison block is included, over-all precision is thought to be about 4%.

Table I. Experimental and Calculated Temperature Distributions for Pure Water as Heat Transfer Medium

Temp., °C.	Distance Above Cold Plate: Measd., cm.	Distance Above Cold Plate: Calcd.[a], cm.
	Experiment I	
52.20	4.39	4.342
45.40	3.69	3.694
39.70	3.15	3.157
32.70	2.51	2.503
26.00	1.87	1.899
18.40	1.19	1.189
11.55	0.57	0.570
	Experiment II	
52.70	4.39	4.357
45.60	3.69	3.682
39.55	3.15	3.111
33.00	2.51	2.500
26.15	1.87	1.849
18.75	1.19	1.190
11.90	0.57	0.570

[a] Calculated from Equation 4: $X = (6.954 \times 10^{-5})\ T^2 + (8.833 \times 10^{-2})\ T - 0.4595$. [b] Calculated from Equation 4: $X = (6.954 \times 10^{-5})\ T^2 + (8.833 \times 10^{-2})\ T - 0.4909$.

Measurements. The calorimeter was first filled with grease and its thermal conductivity determined. Thereafter, weighed portions of metal powder were added to produce increasing weight fractions of the dispersed phase. The phases were hand mixed to uniform consistency before charging into the calorimeter. Special care was taken to avoid air entrainment during mixing. The suspensions were sufficiently viscous to avoid any perceptible settling of the metal particles. Not all of a preceding mixture was used in any given subsequent preparation to reduce the powder requirement. Thermal conductivity data for the suspensions so prepared are tabulated in Tables II, III, and IV.

AGREEMENT WITH ANALYTICAL PREDICTIONS

Numerous investigators have studied two-phase dispersed systems and have developed analytical models for calculating suspension thermal conductivities. Among them are Maxwell (*4*), Rayleigh (*6*), Bruggerman (*1*), Meredith (*5*), Weiner (*7*), and Jefferson, Witzell, and Sibitt (*3*). The arguments rendered vary from the simple concept of alternating laminations to highly complicated statistical formulations. Calculations with several of these developments were made corresponding to the suspensions investigated. Tables II, III, and IV also give these results.

A comparison of the experimental and analytical values shows that a considerable disparity exists in many cases. Of course, agreement can be improved by considering the limiting thermal conductivities a suspension can have and also by using an equation more appropriately derived for a specific case in question. Even then, apparently valid arguments give widely different results. For instance, equations presented by Rayleigh, Bruggerman, and Jefferson

Table II. Experimental Results and Analytical Computations for Zinc-Grease Suspensions

Data Source	Thermal Conductivity (B.t.u./hr. ft. °F.) for Weight and, in Parentheses, Corresponding Volume Fractions of: 0.20 (0.02)	0.40 (0.08)	0.60 (0.16)	0.70 (0.22)	0.80 (0.33)
			Experimental		
Irregular Zn particles	0.171	0.186	0.220	0.243	0.322
			Analytical Calculations[a]		
Maxwell (spheres)	0.178	0.205	0.257	0.301	0.413
Bruggerman (spheres)	0.177	0.209	0.271	0.338	0.533
Rayleigh (spheres)	0.178	0.205	0.256	0.301	0.413
Meredith					
Rods randomly oriented in three dimensions	0.178	0.207	0.264	0.316	0.447
Rods randomly oriented in two dimensions	1.41	6.57	22.2	47.7	120.0
Rods, long axes perpendicular to temp. field	0.174	0.192	0.227	0.260	0.338
Dispersion of spherical doublets	0.178	0.208	0.261	0.311	0.429
Weiner					
Disk model	2.07	5.10	9.97	13.7	20.5
Filament model	0.168	0.177	0.194	0.209	0.243
Jefferson, Witzell, Sibitt (spheres)	0.173	0.188	0.229	0.265	0.390

[a] On the basis of a thermal conductivity for zinc of 64 B.t.u./hr. ft. °F. and for grease of 0.163 B.t.u./hr. ft. °F.

Table III. Experimental Results and Analytical Computations for Spherical Aluminum–Grease Suspensions

Data Source	Thermal Conductivity (B.t.u./hr. ft. ° F.) for Weight Fractions and, in Parentheses, Corresponding Volume Fractions of				
	0.20 (0.07)	0.30 (0.12)	0.40 (0.18)	0.50 (0.24)	0.60 (0.32)
			Experimental		
Spherical Al A	0.173	0.180	0.197	0.218	0.259
Spherical Al B	0.167	0.174	0.198	0.218	0.246
Spherical Al C	0.170	0.179	0.192	0.210	0.237
			Analytical Calculations[a]		
Maxwell (spheres)	0.188	0.213	0.249	0.298	0.369
Bruggerman (spheres)	0.190	0.227	0.268	0.349	0.479
Rayleigh (spheres)	0.188	0.213	0.249	0.300	0.379
Meredith					
Rods randomly oriented in three dimensions	0.190	0.221	0.257	0.311	0.410
Rods randomly oriented in two dimensions	15.0	38.5	85.3	159.0	344.0
Rods, long axes perpendicular	0.179	0.194	0.219	0.259	0.310
to temp. field	0.179	0.194	0.219	0.259	0.310
Dispersion of spherical doublets	0.190	0.216	0.256	0.301	0.391
Weiner					
Disk model	7.85	12.8	18.6	25.7	34.2
Filament model	0.165	0.172	0.184	0.200	0.224
Jefferson, Witzell, Sibitt (spheres)	0.182	0.207	0.250	0.326	0.450

[a] On the basis of a thermal conductivity for zinc of 118 B.t.u./hr. ft. ° F. and for grease of 0.152 B.t.u./hr. ft. ° F.

Table IV. Experimental Results and Analytical Computations for Irregular Aluminum–Grease Suspensions

Data Source	Thermal Conductivity (B.t.u./hr. ft. ° F.) for Weight Fractions and, in Parentheses, Corresponding Volume Fractions of						
	0.20 (0.07)	0.30 (0.12)	0.40 (0.18)	0.50 (0.24)	0.60 (0.32)	0.70 (0.43)	0.80 (0.56)
				Experimental			
Irregular Al particles	0.185	0.191	0.202	0.248	0.339	0.416	0.834
				Analytical Calculations[a]			
Maxwell (spheres)	0.202	0.230	0.268	0.321	0.397	0.525	0.785
Bruggerman (spheres)	0.204	0.245	0.289	0.376	0.516	0.848	1.810
Rayleigh (spheres)	0.202	0.230	0.268	0.323	0.409	0.581	1.005
Meredith							
Rods randomly oriented in three dimensions	0.204	0.238	0.278	0.336	0.442	0.624	1.02
Rods randomly oriented in two dimensions	16.2	41.5	92.0	178.6	370	742	1630
Rods, long axes perpendicular to temp. field	0.193	0.209	0.236	0.280	0.334	0.437	0.660
Dispersion of spherical droplets	0.205	0.233	0.277	0.325	0.422	0.421	0.923
Weiner							
Disk model	8.58	13.8	20.1	27.7	36.8	48.4	63.8
Filament model	0.177	0.186	0.197	0.216	0.242	0.284	0.372
Jefferson, Witzell, Sibitt (spheres)	0.196	0.224	0.270	0.351	0.485	0.794	1.96

[a] On the basis of a thermal conductivity for zinc of 118 B.t.u./hr. ft. ° F. and for grease of 0.164 B.t.u./hr. ft. ° F.

should be applicable to suspensions of spherical particles in a continuous medium. Yet for the aluminum suspensions, the disagreement is an acceptable 10% at low concentrations and over 100% at the highest concentrations studied, and there is no criterion to judge which equation is more nearly correct. Better agreement, however, exists for the zinc suspensions because the thermal conductivity of zinc is substantially lower than that of aluminum and, hence, is closer to that of the continuous phase.

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