High-temperature emissivities of copper, aluminum, and silver

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Using a transient calorimetric technique, the total hemispherical emissivities \( \varepsilon_h \) of very pure copper, aluminum, and silver have been measured at various temperatures in the range 335–1000 K. By carrying out measurements on a sphere, a thin circular disk, and a thin square plate all made of the same copper sample, it is shown that the measured \( \varepsilon_h \) is practically independent of the shape of the specimen. Measured \( \varepsilon_h \) values of Cu, Al, and Ag show considerable departure from the theoretical predictions of Davison and Weeks and the data of previous investigators. \( \varepsilon_h \) data are presented in tabular form at 20 K intervals for all three materials in the range 400–1000 K.

I. INTRODUCTION

Past investigations of the propagation of an electromagnetic wave inside a pure metal may be broadly subdivided into two categories depending upon whether the mean free path \( l \) of the conduction electrons is long or short compared with the penetration depth \( \delta \) of the wave into the medium. Thus at low temperatures at which \( l \) is very long, we observe such phenomena as anomalous skin effect and superconductivity with which we are not concerned in this paper. The former has been extensively studied by London, Pippard, Reuter and Sondheimer, and Chambers. At high temperatures \( l \) is short in comparison with \( \delta \), even for such short waves as in the near-infrared region of spectrum. Consequently, the classical wave propagation equations combined with Ohm’s law should lead to a correct prediction of the behavior of the optical properties of the conductor when its absolute temperature \( T \) is well above its Debye characteristic temperature.

Accordingly, by taking into account the angular dependence of emission of monochromatic radiation predicted by Fresnel’s law, Davison and Weeks derived for the total hemispherical emissivity of a metal the expression

\[
\varepsilon_h = 0.751(Tp)^{1/2} - 0.632(Tp) + 0.670(Tp)^{3/2} - 0.607(Tp)^2, \tag{1}
\]

where the electrical resistivity \( \rho \) is in \( \Omega \) cm. Thus, the thermal radiation emitted by the polished surface of the metal at classical temperatures depends primarily on its \( \rho \) and \( T \). The spectral emissivity \( \varepsilon_{\lambda m} \), in a direction perpendicular to the surface of a conductor for wavelength \( \lambda \), has been expressed in terms of \( \rho \) by Foote in the relation

\[
\varepsilon_{\lambda m} = 0.365(\rho/\lambda)^{1/2} - 0.067(\rho/\lambda), \tag{2}
\]

where \( \lambda \) is in cm.

Because the resistivity of a good conductor is of the order of \( 10^{-8} \) \( \Omega \) cm above 300 K, the first terms on the right of Eqs. (1) and (2) are the major contributors to \( \varepsilon_h \) and \( \varepsilon_{\lambda m} \). At these temperatures, the existing experimental data of \( \rho \) vs \( T \) for pure metals are very nearly proportional to \( T \) in accordance with the prediction of Bloch. Therefore, to a first order of approximation, we may expect \( \varepsilon_{\lambda m} \) and \( \varepsilon_h \) to be proportional to \( T^{1/2} \) and \( T \), respectively, under the specified conditions.

The temperature variations of \( \varepsilon_{\lambda m} \) of polished surfaces of Cu, Ag, Al, Au, and Pt have been studied by several investigators at different wavelengths in the spectral range of 0.55–14 \( \mu \)m. Most of these investigators reported that \( \varepsilon_{\lambda m} \) varied strongly with surface roughness, extent of oxidation, and also slightly with temperature. Because the total emission from a solid is made up of a continuous spectrum of wavelengths, mostly in the infrared, the work of these investigators suggests that \( \varepsilon_h \) of pure metals may also be well described by the dominant term of Eq. (1), which is proportional to \( T \).

Early investigation of the temperature dependence of \( \varepsilon_h \) of copper and aluminum by Best were carried out on hollow, spherical shells fabricated from thin sheets of metals, by a steady-state technique in which the power lost by radiation from the surface is determined by the electrical power required to maintain the surface at a constant temperature. Large conduction losses through power leads and thermocouple leads, the liberal use of a silver cement in the specimen assembly, and nonuniformity of the temperature inside the sphere are three of the many sources of error in this technique. Therefore, it is not surprising that the results obtained for copper by Best, in the range 373–673 K, showed hardly any temperature dependence. His results for aluminum were very much higher than the theoretically predicted values. The measurements on copper by Butler and Inn, Gaumer et al., and Thaler et al., and those on silver by Zimmermann, Butler et al., and Thaler et al., were also unsatisfactory for one or more of several reasons. Some of these are (a) use of low-purity commercial grade materials with high intrinsic \( \rho \) values, (b) use of a coating of an absorbing material like Parson’s black on the inner wall of the vacuum chamber, which results in a considerable increase in outgassing and in a higher ultimate pressure, (c) formation of a high-resistivity surface layer on the metal due to prolonged annealing at \( T > 1000 \) K in an inadequate vacuum, and (d) use of large hollow specimens with seams, enclosing electrical heating assemblies which could produce a nonuniform specimen temperature, a large correction term on account of the power leads, and a large systematic error. Therefore, the present work was undertaken with a view to (i) obtain accurate measurements of \( \varepsilon_h \) on polished surfaces of very pure Cu, Al, and Ag at small intervals of temperature in the range 335–1000 K, (ii) compare...
our results with Eq. (1), and (iii) investigate whether
a change in the polishing technique produces any sig-
ificant change in the measured values of $\varepsilon_a$.

II. EXPERIMENTAL TECHNIQUE

The transient calorimetric technique used in this re-
search is shown schematically in Fig. 1. In this tech-
nique, a vacuum-enclosed, thermally isolated specimen
of mass $M$ and surface area $A$ is preheated to a tem-
perature $T$, which is well above that of the surrounding
walls. It is then allowed to cool by radiating to the
walls maintained at $T_0$. Under these conditions the
total power lost by the specimen may be considered as
the sum of the powers lost by radiation $Q_r$, by thermo-
couple conduction $Q_c$, and by conduction $Q_g$, through
the residual gas surrounding the specimen. Hence we
have

$$-M C_p \frac{dT}{dt} = \varepsilon_a A (T^4 - T) + Q_c + Q_g,$$

where $C_p$ is the specific heat at constant pressure,
$\frac{dT}{dt}$ is the time rate of change of temperature, and $\sigma$
is the Stefan-Boltzmann constant. The mass and the
area of the specimen are measured accurately at room
temperature. The areas at higher temperatures are
calculated by applying a correction for thermal expan-
sion of the metal. $\varepsilon_a$ is then calculated using values of
$C_p$ for the pure materials, obtained from published
literature, and using values of $T$ and $\frac{dT}{dt}$ mea-
sured with the thermocouple. The method by which
$\frac{dT}{dt}$ was obtained is described later.

The high-purity copper, aluminum, and silver
specimens used in this investigation were made from
polycrystalline samples with percentage purities of
99.999. The copper and aluminum samples were pur-
chased from the Materials Research Corp. and the sil-
ver from the Ventron Corp.

Because results of previous investigators, obtained by using specimens of different shapes, showed
some disagreement, it was felt necessary to investi-
gate first the effect, if any, of the shapes of the speci-
mens on the measured emissivities. Accordingly,
measurements were made on three different copper
specimens with the shapes described in Table I. The

TABLE I. Description of the specimens of 99.999% pure metals used in this research.

<table>
<thead>
<tr>
<th>Specimens</th>
<th>Shape</th>
<th>Mass, g</th>
<th>Diameter or side, cm</th>
<th>Thickness or alt, cm</th>
<th>Surface area, cm²</th>
<th>Surface preparation</th>
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</thead>
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<td>Circular disk</td>
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</tr>
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<td>0.9755</td>
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<td>2.9139</td>
<td>M</td>
</tr>
</tbody>
</table>

$^a$CuII and CuIV refer to the same sphere but differ in polishing technique.

$^b$Masses include those of pins used for fixing thermocouples.

$^c$All dimensions were measured at room temperature.

$^d$M, mechanical polishing. E, electropolishing.

FIG. 1. Schematic drawing of experimental arrangement: S, specimen; B, Plexiglas support; C, rf coil; D, Pyrex glass bell jar; E, reference junction in ice bath; F, water-cooled power leads to rf generator; G, electrical leads to recorder; H, connection to cold trap and diffusion pump.


The procedure used in each experimental run was first to evacuate the bell jar and to locate the specimen $S$, inside the water-cooled rf coil $C$, by moving the Plexiglas support without disturbing the vacuum. The power for the coil was supplied by a 1 kW rf generator. After the specimen had been heated to a predetermined temperature, the generator was turned off. The specimen was then moved away from the coil and set approximately in the center of the bell jar allowing it to cool by radiating to the surroundings maintained at a constant room temperature. The output of the thermocouple, compensated by a reference junction $E$, at 0 °C, was continuously recorded by a Leeds and Northrup Speedomax-W recorder. Temperature versus time curves were then obtained from the emf versus time curves.

The slopes of the temperature-time curve in a particular portion of any cooling curve was determined by using a least-squares method to fit a parabola to a set of seven adjacent experimental points in that interval. The calculated derivative at the midpoint was assumed to be the slope of the curve at that point. The parabola was then shifted along the experimental points to obtain slopes at the other points.

III. ERRORS AND ACCURACIES

In calculating $\varepsilon_s$ from Eq. (3), we assumed that at any instant, the cooling specimen was at a uniform temperature. This assumption is true, within a very small fraction of a degree, for such small specimens with very high thermal conductivities as were used in this investigation. A thermal analysis using the Fourier heat-conduction equation was made to determine the limit of validity of this assumption. Assuming the cooling rate to be the same at all points inside a sphere and the temperature distribution to be spherically symmetric, the temperature difference between the center and the surface was found to be given by

$$\Delta T = \frac{(MC/A)\theta}{h r},$$

(4)

where $h$ is the heat conductivity and $r$ the radius of the sphere. Using the known values of $C_s$ and $h$ (Ref. 25) in Eq. (4), $\Delta T$ was found to be about 0.005 K for both the copper and silver spheres for a specimen temperature of 700 K. $\Delta T$ was even smaller for $T < 700$ K. Similar calculations were made for the plate specimens of copper and aluminum, for which the $\Delta T$'s were estimated to be smaller than the corresponding values for the spheres.

As mentioned previously, in some of the earlier investigations, $\varepsilon_s$ in which the transient calorimetric technique was used, the walls of the evacuated enclosure were coated with some highly absorbing material to ensure that the energy radiated from the specimen was absorbed completely by the walls. Very simple considerations will show that the cooling rate of the specimen and the measured $\varepsilon_s$ are affected only to an insignificant extent by the absence of this coating if the ratio $A/A_w$, of the surface areas of the specimen and the walls, is a very small fraction. If $R_w$ is the nonzero reflectivity of the walls, the power that is first emitted by the specimen and reabsorbed by it after a single diffuse reflection from the walls is approximately given by $Q_p(A/A_w)R_w$. The fractional power reabsorbed by
a specimen temperature of 400 K, the zero of the time scale has been fixed arbitrarily.

The specimen is thus given by \((A/A_w)R_w\epsilon_s\). Under present experimental conditions, \(A/A_w\) is of the order of \(10^{-4}\), which results in an insignificantly small value of \(10^{-6}\) for \((A/A_w)R_w\epsilon_s\). The power reabsorbed after two or more reflections from the walls of the enclosure can be shown to be even less significant. The correction term \(\dot{Q}_r\) was less than 0.5% of \(\dot{Q}_c\) in a vacuum of \(1\times10^{-5}\) torr, maintained during all experiments. For a specimen temperature of 400 K, \(\dot{Q}_r/\dot{Q}_c\) was estimated to be only 0.6% and 3.5%, respectively, for the two sizes, 2 mils (0.0051 cm) and 5 mils (0.0127 cm), of thermocouple wires used. At higher specimen temperatures, \(\dot{Q}_r/\dot{Q}_c\) was even smaller.

The usual formulation of Kirchhoff’s law assumes thermodynamic equilibrium between a radiating object and its surrounding walls. Therefore, the validity of the law is restricted to the special case when \(T = T_0\). Under present experimental conditions in which \(T > T_0\), Eq. (3) should be written

\[
-MC_p \left(\frac{dT}{dt}\right) = A\sigma \epsilon_s T^4 - \alpha(T, T_0) A\sigma T_0^4 + \dot{Q}_c + \dot{Q}_r, \tag{5}
\]

where \(\alpha(T, T_0)\) is the total hemispherical absorptivity of the specimen at temperature \(T\) when the walls are at \(T_0\). However, it has been pointed out by Eckert\(^{27}\) that, for metals, the relation

\[
\alpha(T, T_0) = \epsilon_s(T, T_0) \tag{6}
\]

holds good in the infrared region. Therefore, when \(T/T_0 > 2\), the error involved in replacing \(\alpha(T, T_0)\) by \(\epsilon_s\) at \(T_0\) in Eq. (5) is negligible. When \(1 < T/T_0 < 2\), this substitution is a fairly good approximation because in this classical region, \(\epsilon_s\) of a pure normal metal is continuous and varies only slightly with temperature.

The thermocouple wires were obtained from the Omega Engineering Co. The emf vs \(T\) table for chromel-alumel supplied by the company was used in determining specimen temperatures. The recorder has been calibrated with a precision microvolt potentiometer. Though the precision with which any particular temperature could be determined from a cooling curve was 0.05 K, the actual temperature of the specimen could have been in error by several degrees due to possible systematic errors. However, the cooling rates \(-dT/dt\), calculated from the cooling curve, were certainly accurate to better than \(5\times10^{-5}\) K/s, which causes a fractional error of less than 1%. The overall accuracy in the measurement of \(\epsilon_s\) is estimated to be better than 3% and the maximum absolute error is 6%.

**IV. RESULTS AND DISCUSSION**

Typical \(T\) vs \(t\) and \(-dT/dt\) vs \(T\) curves obtained for Cu\(iv\) are shown in Figs. 3 and 4, respectively. The specific-heat values of copper, required for calculating \(\epsilon_s\), were those of Pawel\(^{19}\) and of Lyusternik.\(^{20}\) The data of these authors were chosen because each of them had obtained results for the entire range of 400–1000 K from measurements on single specimens with purities of 99, 99.9% and 99.9%, respectively. In spite of the difference in purities of the two specimens used by these two authors, their data show good agreement indicating that specific heat is a property which is not very sensitive to traces of impurities in this temperature range.

The data obtained for the copper sphere, Cu\(iv\) and Cu\(iv\), are compared in Fig. 5 with the results of previous investigators and with the theoretical predictions of Eq. (1) evaluated by using resistivity data for this metal from the AIP Handbook.\(^7\) It is seen that the \(\epsilon_s\) vs \(T\) curves of previous researchers are considerably higher than ours. However, the two curves obtained by us show remarkable agreement even though the surfaces were prepared by different techniques. Another noteworthy feature in Fig. 5 is that our results are only about 50% of the theoretically predicted values at \(T > 600\) K. A large difference is also observed between the slopes of the experimental and theoretical curves. As a consequence, the difference between the theoretical and experimental values of \(\epsilon_s\) decreases steadily as \(T\) is decreased.

In Fig. 6 our \(\epsilon_s\) vs \(T\) curve for aluminum is compared with the data of Best\(^{13}\) and with Eq. (1) evaluated by using the published resistivity data of Simmons.
The specific-heat data used in obtaining our results for the entire range were taken from the work of Pochapsky who used a specimen of 99.9% purity. It is clear that the results of Best are almost 100% higher than ours. The theoretical curve is also considerably above our experimental curve which is shown to have linearity with $T$. The discrepancy between the theoretical and experimental slopes is also obvious.

The $C_\text{p}$ values of silver used in calculating $\varepsilon_\text{h}$ from Eq. (3) are those of Eastman et al. and Bronson et al. Our measured $\varepsilon_\text{h}$ values are compared in Fig. 7 with those of Butler et al. and with the predictions of Eq. (1) evaluated by using the resistivity values obtained from the AIP Handbook. Again, the theoretical curve given by Eq. (1) is far above our curve in the entire temperature range. For ready reference, $\varepsilon_\text{h}$ vs $T$ data obtained by curve fitting all experimental points are presented in Table II for the three metals Cu, Al, and Ag at 20 K intervals.

The optical properties of a metal are known to be TABLE II. Comparison of averaged $\varepsilon_\text{h}$ values of present research on Cu, Al, and Ag with the theoretical predictions of Eq. (1) in the range 400-1000 K.

<table>
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<tr>
<th>Temperature, K</th>
<th>Cu</th>
<th>Cu IV</th>
<th>Cu IV</th>
<th>Eq. (1)</th>
<th>Al I</th>
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<td>2.82</td>
<td>5.60</td>
<td>4.83</td>
<td>7.07</td>
<td>2.73</td>
</tr>
<tr>
<td>980</td>
<td>2.93</td>
<td>2.82</td>
<td>2.87</td>
<td>2.87</td>
<td>5.73</td>
<td>4.92</td>
<td>7.22</td>
<td>2.76</td>
</tr>
<tr>
<td>1000</td>
<td>3.00</td>
<td>2.87</td>
<td>2.92</td>
<td>2.92</td>
<td>5.86</td>
<td>5.01</td>
<td>7.37</td>
<td>2.79</td>
</tr>
</tbody>
</table>

aColumn Cu IV gives our best values for copper.
affected significantly by the physical condition of its surface. Therefore the resistivity which must be used
to evaluate \( \epsilon_e \) in Eq. (1) should be the resistivity \( \rho \) of
the metal at the surface layer. This can be expressed
by the equation

\[
\rho = \rho_L + \rho_i + \rho_c,
\]

(7)

where \( \rho_L \) is the resistivity caused by thermal vibrations
of the lattice, \( \rho_i \) is the resistivity caused by scattering
of the electron waves by impurity atoms, and \( \rho_c \) is the
resistivity arising from cold working and the intro-
duction of foreign atoms in the surface layer during its
preparation. As has been shown by Pawlek and Rogal-
la, \( \rho_L \) is the dominant contributor to \( \rho \) for a specimen
at temperatures around room and above. This con-
clusion is strongly supported by the results of high-
frequency surface resistivity measurements of Cham-
boss's at room and at very low temperatures.

The thickness of surface layer removed from the
copper sphere by electropolishing was estimated to be
about 10 \( \mu \)m. By removal of such a thick layer, it is
believed that much of the strains and impurity atoms
introduced in the surface layer during any mechanical
polishing process should have been removed. If the
high-temperature values of \( \rho_L \) had been comparable to
\( \rho_i + \rho_c \) in the mechanically polished sphere, a signi-
ficant lowering of \( \epsilon_e vs T \) curve could have been ex-
pected after electrolytic polishing. The fact that very
little difference has been observed is, therefore, a
verification of Pawlek and Rogalla's view that at high
temperatures \( \rho_L \gg \rho_i + \rho_c \). This also enables us to con-
clude that any further improvement in the technique of
surface preparation will perhaps produce only an in-
significant change in the \( \epsilon_e vs T \) curve in this tempera-
ture range.

An examination of all the results obtained in this re-
search, therefore, leads us to the inescapable conclu-
sion that Eq. (1) is not adequate to represent the true
behavior of the emissive properties of copper, alumi-
num, and silver at high temperatures. In order to

![FIG. 8. Continuous plot of high- and low-temperature \( \epsilon_e vs T \) data for electrolytically polished copper: *, best data, Cu IV, obtained in this research; *, best data obtained by Estalote and Ramanathan\(^2\), for the range of 150–300 K; —, Davison and Weeks.\(^5\)](image)

... further substantiate this conclusion the best \( \epsilon_e vs T \) data for the range of 150–300 K for electrolytically
polished Cu and Al, published by Estalote and Ramana-
than, \(^2\) elsewhere in this number of the journal, are
plotted continuously along with our best high-tempena-
ture data for these metals in Figs. 8 and 9, re-
spectively. It is seen that the low- and high-temperature
data fit together as well as may be expected, without
any significant discontinuity. It is also seen that in
both these figures the unbroken lines representing Eq.
(1) are higher than the experimental curves in the en-
tire range of 150–1100 K. Further, at any one tem-
perature, the slopes of the theoretical curves are lar-
gers than the corresponding experimental slopes. Con-
sequently, the largest disparity between theory and ex-
periment observed at the highest temperature decreases steadily as \( T \) decreases and nearly vanishes around
150 K.

The results of a recent investigations of the tem-
perature variation of \( \epsilon_e \) of Ni and Fe seem to indicate
that ferromagnetic metals might perhaps have a dif-
f erent emissive behavior. Before any attempt to modi-
fy theory\(^3\) is made, it is necessary to carry out mea-
surements on other metals. A careful investigation
of the temperature dependence of the spectral emis-
sivities of pure metals may also be expected to elucidate
theory.

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indebted to Bud Schuler and J. H. Buckley for the con-
struction of apparatus and preparation of specimens, and to R. L. Pontheir for constant help during the
course of this research.

\(^1\)H. London, "The high frequency resistance of superconducting
\(^2\)A. B. Pippard, "The anomalous skin effect in normal metals,"

![FIG. 9. Continuous plot of high- and low-temperature \( \epsilon_e vs T \) data for electrolytically polished aluminum: *, present re-
search; o, Estalote and Ramanathan\(^2\); —, Davison and
Weeks.\(^5\)](image)
K. G. Ramanathan and S. H. Yen


After submitting this paper for publication the present authors came across an article entitle “Theoretical and experimental studies of the total emittance of metals,” by W. J. Parker and G. L. Abbott, published in Symposium on Thermal Radiation of Solids edited by S. Katzoff (U. S. GPO, Washington D. C., NASA SP-55, 1965), pp. 11–38. In this article the authors have modified the theory of Davisson and Weeks by taking into account the finite, nonzero, relaxation times of the conduction electrons in metals. For the particular case of a zero relaxation time the numerical result of Parker and Abbott is practically the same as Eq. (1) for good conductors for which $\rho T < 0.1$. The effect of a nonzero relaxation time is to bring down the $\xi$ values at all temperatures. However, Parker and Abbott concluded that their experimental data for the metals Ta, Nb, W, and Mo qualitatively agreed with Eq. (1) and with their theoretical result for a zero relaxation time. On the other hand the results reported in this paper for Cu, Al, and Ag appear to lend support to the relaxation theory of Parker and Abbott. It is hoped that a detailed analysis of the effect of finite relaxation times of conduction electrons on the emissive behavior of metals will form the subject matter of a future communication.