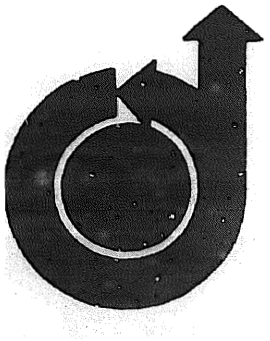


AIAA Paper
No. 70-63

TOTAL EMITTANCE MEASUREMENTS OF THIN METALLIC FILMS AT CRYOGENIC TEMPERATURES

by
C. R. CUNNINGTON and G. A. BELL
Lockheed Palo Alto Research Laboratory
Palo Alto, California
and
B. F. ARMALY and C. L. TIEN
University of California
Berkeley, California

AIAA 8th Aerospace Sciences Meeting

NEW YORK, NEW YORK / JANUARY 19-21, 1970

TOTAL EMITTANCE MEASUREMENTS OF THIN METALLIC FILMS AT CRYOGENIC TEMPERATURES*

G. R. Cunnington and G. A. Bell
Lockheed Palo Alto Research Laboratory
Palo Alto, California

and

B. F. Armaly** and C. L. Tien
University of California
Berkeley, California

ABSTRACT

Calorimetric measurements were performed to determine the total hemispherical emittance of thin gold films as a function of temperature in the range from 60 to 300°K. The effect of film thickness on the emittance is demonstrated through comparison of data for thin (~400 Å) and thick (~2500 Å) metallic films. It is found that for the type of thin metallic films used in cryogenic multilayer insulation the emittance of these films is not as strongly dependent on the film thickness as is predicted by theory. The present experimental data are compared with theoretical values and also with experimental results of other investigators.

I. INTRODUCTION

Thermal radiation characteristics of highly reflective metal surfaces at cryogenic temperatures have become an important factor in the development of thermal isolation systems for cryogenic applications[1] and in the area of thermal control of spacecraft[2] and planetary experiments.[3] The increasing use of the multilayer type of insulation has placed particular importance on the knowledge of the thermal radiation properties of thin metallic films (~400 to 1000 Å) on plastic substrates.[4] The most efficient multilayer insulation reflective shield concept uses vacuum deposited gold or aluminum (~500 Å) on a very thin plastic sheet (0.15 to 0.25 mils thick) such as Mylar or Kapton which greatly reduces system weight[5] and thermal anisotropy[6] of the insulation over one using metal foils. For the analysis of radiative heat transfer in the cryogenic temperature region, it is important to know the total hemispherical emittance of the film as a function of temperature and how emittance varies with film thickness.

The first theoretical work on the radiation properties of thin metallic films[7] was based upon the Drude single (or free) electron (DSE) theory of optical constants. However, it later became apparent that the anomalous skin effect (ASE) theory[8] should be used rather than the DSE theory for the cryogenic temperature range. The recent work of Domoto et al.[5] showed that the predicted values of total hemispherical emittance based upon the ASE theory could be an order of magnitude greater than those predicted by the DSE theory. It has been shown[10,11] for the condition when the film thickness becomes smaller than the electron mean free path that the electrical and thermal properties of the film will differ from those of the bulk metal. To incorporate the size effect (i.e., influence of film thickness) into the skin effect, Dingle[12] established the theoretical framework for evaluating the radiative properties of thin metallic films on the basis of the ASE theory. An extensive analysis of size and skin effect, at cryogenic temperatures has recently been done by Armaly and Tien.[13]

The purpose of the present work is to report the experimental results of total hemispherical emittance of several thin (~400 to 600 Å) and thick (~2500 to 4000 Å) vacuum deposited high purity gold films on plastic and metal substrates over the temperature range of 60 to 300°K. Effect of film thickness on total hemispherical emittance is demonstrated and comparisons are made between experimental and theoretical results for both thickness effects and absolute values of emittance.

II. EXPERIMENTAL APPARATUS AND PROCEDURE

The calorimetric emittance apparatus consists of a 2-1/2411. diameter by 3/16-in. thick aluminum, brass, or copper sample substrate mounted to a liquid nitrogen or liquid hydrogen cooled vacuum chamber. (See Figure 1.) Sample temperature is controlled from cryogen temperature to 300°K using a spiral wound heater imbedded in a 2-1/4-in.-diameter copper heating block, Temperature readout is provided using a Rosemount Engineering Company 104 AH1 platinum resistance thermometer.

Radiated sample energy is collected using a blackbody absorber mounted to the cryogenically cooled supporting structure within 30 mils of the sample front face. This absorber consists of a series of thin wall metal tubes (3/16-in. o.d. by 10-mil wall by 1-in. long) fitted to the interior of an aluminum shell. The inside of the shell and tubes are coated with Cat-a-Lac flat black epoxy paint. The geometry of the blackbody coupled with the high absorptance of the paint leads to a calculated theoretical absorptance of 0.97. The blackbody thermal link is selected so that absorbed sample energy raises blackbody temperature between 5 and 10°K above that of the bath once thermal equilibrium is reached. For the present experiments, two thermal links were needed: three 20-mil-diameter stainless steel wires were used for sample temperatures between 60 and 160°K, while a 1/4-in. o.d. by 20-mil wall stainless tube was used for sample temperatures between 160 and 300°K. Blackbody temperature is monitored using a platinum or germanium resistance thermometer.

To define the thermal energy reaching the blackbody from the sample, it is necessary to calibrate the blackbody absorber. This is done by allowing the sample to cool to cryogenic bath temperature, and supplying heat energy to the absorber through a carbon resistance heater. Once blackbody thermal equilibrium is attained, the power dissipated in the carbon resistor is measured and the temperature (thermometer resistance) is recorded. The process is then repeated for different

*Work at the University of California conducted under National Aeronautics and Space Administration Grant NGR-05-003-295, Ames Research Center.
**Present address: Department of Mechanical and Aerospace Engineering, University of Missouri, Rolla, Missouri.

power levels, and a curve of blackbody temperature versus power input to the blackbody can be generated. This technique produces a very reproducible curve since the blackbody thermal link and all the leads to the resistances mounted on its back face are thermally grounded to the cryogenic bath.

Emittance Calculations

Nomenclature:

- Q_l = heat flux to blackbody thermal link
- Q_c = calibration power input
- A_s = A_b = sample and abeorber frontal area
- σ = Stefan-Boltzmann's constant
- T_c = sample temperature during calibration ≈20°K or 77°K
- T_s = sample temperature
- T_b = blackbody absorber temperature
- F = sample to blackbody view factor = blackbody to sample view factor
- ϵ_s = sample emittance at T_s
- a_b = blackbody absorptance ≈0.98
- ϵ_b = blackbody emittance at $T_b \approx 0.98$
- a_s = sample absorptance for blackbody radiation at T_b and sample at T_s
- Y_s = blackbody radiation exchange with wall during sample run
- a'_s = sample absorptance for blackbody radiation et T_b and sample at T_c
- Y_c = blackbody radiation exchange with wall during calibration

With a radiating sample present, the heat flux reaching the blackbody thermal link is equal to the radiated sample energy, which is absorbed by the blackbody, minus the radiated blackbody energy, which is either absorbed by the sample or the chamber walls.

Therefore:

$$Q_l = A_s\sigma T_s^4 F \frac{\epsilon_s a_b}{1 - F^2(1-\epsilon_s)(1-a_b)} - A_b\sigma T_b^4 F \frac{\epsilon_b a_s}{1 - F^2(1-\epsilon_b)(1-a_s)} - Y_s \quad (1)$$

and during calibration the heat flux reaching the link is:

$$Q_l = Q_c - Q_r - Y_c \quad (2)$$

where:

$$Q_r = A_b\sigma T_b^4 F \frac{\epsilon_b a'_s}{1 - F^2(1-\epsilon_b)(1-a'_s)} - A_s\sigma T_s^4 F \frac{\text{'8\%}}{1 - F^2(1-\epsilon_s)(1-a_b)} \quad (3)$$

Thus, Q_l has been defined with a radiating sample present, and with a calibration heat input Q_c; so, for corresponding blackbody temperatures we have by combining Eqs. (1), (2), and (3):

$$A_s\sigma T_s^4 F \frac{\epsilon_s a_b}{1 - F^2(1-\epsilon_s)(1-a_b)} - A_b\sigma T_b^4 F \frac{\epsilon_b a_s}{1 - F^2(1-\epsilon_b)(1-a_s)} - Y_s = Q_c - A_b\sigma T_b^4 F \frac{\epsilon_b a'_s}{1 - F^2(1-\epsilon_b)(1-a'_s)} + A_s\sigma T_s^4 F \frac{\epsilon_s a_b}{1 - F^2(1-\epsilon_s)(1-a_b)} - Y_c \quad (4)$$

The second term on the left-hand side and the second and third terms on the right-hand side of Eq. (4) can all be neglected since $T_B << T_s$. This introduces an error estimated at less than 1%. Also, $Y_c = Y_s$ and thus

$$Q_l = A_s\sigma T_s^4 F \frac{\epsilon_s a_b}{1 - F^2(1-\epsilon_s)(1-a_b)} \quad (5)$$

The emittance ϵ_s can now be calculated since all the terms in Eq. (5) are known including a_b, which can be calculated using an iterative process and data obtained by substituting a blackbody radiator identical to the blackbody absorber for the sample. The emittance, or absorptance, of the blackbody from 80 to 260°K was found to be 0.98 ± 0.01.

Error Analysis

The probable errors listed with the emittance values found in Table 2 were calculated as follows:

$$\delta\epsilon_s = \frac{\Delta\epsilon_s}{\epsilon_s} \times 100\% = \left[\left(\frac{4\Delta T_s}{T_s}\right)^2 + \left(\frac{\Delta Q_c}{Q_c}\right)^2 + \left(\frac{\Delta a_b}{a_b}\right)2 + \left(\frac{\Delta F}{F}\right)_2\right]^{1/2} \times 100$$

Table 1 summarizes typical emittance measurement errors at sample temperatures of 60, 100, and 300°K. As shown in this table, measurements at the lowest sample temperature are subject to the greatest uncertainty, principally because the ΔT between the blackbody absorber and the cryogen bath is approaching the same order of magnitude as the barometric pressure shifts at cryogenic liquid temperature. At the higher sample temperatures, the principal source of error is attributable to the ΔT's across the 2-1/2-in.-diameter sample.

A source of error which was not listed in Table 1 but which can be very significant, is heat transfer to or from the blackbody absorber due to residual gas conduction. In the present experiments, great care was taken in designing and fabricating the experimental vacuum chamber so that pressures less than 10^{-8} Torr could be realized. At the lowest sample temperature

it can be shown that heat transfer by residual gas conduction is of the same order of magnitude as heat transferred by radiation if the chamber pressure is of the order of 10^{-5} Torr.

Preparation of Specimens

Total hemispherical emittance measurements were performed over the temperature range of 60 to 300°K on four specimens of vacuum deposited gold films on plastic substrates and four specimens of vacuum deposited gold films on copper substrates. The three gold on Mylar and one gold on Kapton specimens were taken from commercially.prepared metallized materials; the Mylar specimens were metallized on both sides and the Kapton on only one side. For these the purity of the gold used for evaporation, the deposition rate, and the chamber pressure during evaporation are not known. The 400, 520, and 4000 Å gold on copper specimens were prepared at Lockheed from 99.999% purity gold. Deposition rate was 20 to 50 Å/sec and chamber pressure ~10^{-6} Torr. The substrate was highly polished brass over which 1000 Å of copper was deposited. The 2500 Å specimen* was prepared using gold of 99.999% purity; chamber pressure was 10^{-5} Torr; deposition rate was 180Å/sec. No determinations were made of the composition of the "as deposited" films.

Metal film thickness measurements were made for the three coated Mylar specimens and three of the gold-copper specimens. These were performed using a proton transmission technique which measures transmission of a proton beam through the gold film as a function of the incident proton energy. The film thickness is determined from accurately calibrated energy versus thickness relationships for a proton/gold system.[14] This method itself yields thicknesses that are accurate to ±50Å; however, the actual emittance samples were not used to make the measurements. The measurements of the Mylar specimens were made using an adjacent piece of the same section of film from which the emittance samples were taken. Since thicknesses across the section have been observed to vary by 200 Å or more, one should be careful in drawing conclusions concerning film thickness versus emittance on films whose thickness difference is less than 200 Å. For the 400, 520, and 4000 Å gold on copper specimens, film thickness was inferred from measurements made on quartz plates placed on two sides of the substrate during the deposition process. As these were immediately adjacent to the emittance specimen, it is believed these thickness values are representative of the actual emittance specimen. Thicknesses for the Kapton and the 2500 Å gold on copper specimens were provided by the supplier.

III. DISCUSSION OF EXPERIMENTAL RESULTS

Table 2 and Figures 2 and 3 present the total hemispherical emittance versus temperature data for the eight specimens investigated. Measurements on the 400A film on Kapton and the 2500 Å film on copper specimens were performed at the University of California, Berkeley, California. All other data were obtained at the Lockheed Palo Alto Research Laboratory. While the experimental data seem to deviate appreciably from the theoretical results (possible causes for this discrepancy will be discussed later), it should be noted that the general trend as exhibited by the experimental data does agree qualitatively with the prediction and the data are self-consistent among themselves. The consistency of the experimental data is indeed very impressive in view of the agreement between low temperature data of two independent investigators (Lockheed and Berkeley) and the agreement among room temperature data of three independent investigations (Lockheed, Berkeley, and that of Ruccia and Hinckley). The experiment of Ruccia and Hinckley[15] was limited to room temperature. The measured effect of film thickness is not as pronounced as that predicted by the theory, as is shown in Figure 4. However, their study as well as this one clearly demonstrates a noticeable effect of film thickness on the total hemispherical emittance and, in addition, confirms the predicted trend of this effect.[13]

It is interesting to note that the substrate material (i.e., plastic or copper) does not appear to have a significant influence on emittance of the gold surface, even in the thin film data shown in Figure 2. This implies that gold films of thickness ~400 Å are nearly opaque for the primary wavelength range corresponding to the sample temperature. Therefore, the effect of film thickness on emittance is not due to the change in transparency but rather due to the restriction of electron free paths at the boundaries.

The present thick-film data are limited to those of 2500 and 4000 Å films. The size effect for films of thickness greater than this range should be negligible since the film thickness will become much larger than the electron mean free path.[13] For instance, at room temperature the electron mean free path for gold is approximately 600 Å and the size effect becomes negligible for films of thicknesses larger than 1500 Å. This fact is further confirmed by the experimental result shown in Figures 3 and 4.

Another set of low-temperature emittance data for gold has been reported by Caren,[16] but his data are much higher than those reported here. The gold used in his experiment was chemically deposited and no thickness was specified. The large discrepancy is probably due to the quality of the film used and possible differences in the experimental procedure.

With regard to the quantitative comparison between theoretical and experimental results, the present data are about 50 or 100% higher (room temperature and 60°K cases, respectively) than the predicted values. This difference which is much larger than the experimental errors of 3 to 8%, must be due to factors other than the thickness of the film. One factor which is most likely to affect the results is the quality of the film used for testing, It has been shown by Bennett and Bennett[17] that the spectral emissivity of metallic films is very much dependent on the quality of the surface, the quality and type of the substrate material, the purity of the film, and the vacuum and the rate under which the film was deposited,

The effect of the first three items could be neglected when discussing the total emissivity for the 2500 Å thick sample, The smoothness and the flatness of the film surface and the substrate will affect the spectral measurements much more than the total measurements. Also, the ratio of the length characterizing the roughness of the surface to the characteristic wavelength of the incident radiation will decrease rapidly as the temperature decreases. Thus, the effect of roughness is usually negligible at cryogenic temperatures. The effect of sample purity, for puritiea higher than 99.99%, was also found to be negligible.[17] The remaining item, i.e., the vacuum and the rate at

*Sample supplied by Alan P. Bradford, Department of the Army, Night Vision Laboratory, Fort Belvoir, Virginia.

which the film was deposited, however, is very important. For example, the spectral emissivity of metallic films deposited at a rate of 7 Å/sec and under high-vacuum conditions, 10^{-5} Torr, could be two or three times higher than the emissivity of a similar film deposited under ultrahigh-vacuum conditions, 10^{-9} Torr. The rate at which the film is deposited is also an important factor. The higher the rate of deposition, the better and purer is the film. Measurements determining the effect of deposition rate on the film emissivity, however, are not available.

ACKNOWLEDGMENTS

The authors are grateful to M. McCargo and S. A. Greenberg who provided the proton transmission measurements, and to D. A. Vance for preparation of gold on copper specimens.

IV. REFERENCES

[1] Caren, R. P. and Cunnington, G. R., "Heat Transfer in Multilayer Insulation Systems," Advances in Cryogenic Heat Transfer, Chem. Eng. Prog. Symp. Series, Vol. 64, No. 87, 1968, pp. 67 – 81.

[2] Tawil, M. N. and Caloger, P., "Use of Multilayer Insulation on the LM Vehicle," AIAA Paper No. 69-609, June 1969.

[3] Zierman, C. A. and Calimbas, A. T., "Radiative Thermal Control System for a Lunar Surface Experiment," in Aviation & Space Progress & Prospects ASME, June 1968.

[4] Cunnington, G. R. and Tien, C. L., "A Study of Heat Transfer Processes in Multilayer Insulations," AIAA Paper No. 69-607, June 1969.

[5] Ruccia, F. E., Hinkley, R. B., and Reid, R. C., "Thermal Performance of Tank Applied Multilayer Insulation," in Advances in Cryogenic Eng., Vol. 12, Plenum Press, New York, 1967, pp. 216 – 24.

[6] Vliet, G. C. and Coston, R. M., Thermal Energy Transport Parallel to the Laminations in Multilayer Insulation," in Advances in Cryogenic Eng., Vol. 13, Plenum Press, New York, 1968, pp. 671–679.

[7] Barnes, R. B. and Czerny, M., "Concerning the Reflection Power of Metals in Thin Layers for the Infrared," Physical Review, Vol. 38, 1931, pp. 338 – 345.

[8] Reuter, G, E. H. and Sondheimen, E. H., "The Theory of the Anomalous Skin Effect in Metals," Proc. Roy. Soc., London, Ser. A, Vol. 195, 1948, pp. 336 – 364.

[9] Domoto, G. A., Boehm, R. F., and Tien, C. L., "Predictions of the Total Emissivity of Metals at Cryogenic Temperatures," in Advances in Cryogenic Eng., Vol. 14, Plenum Press, New York, 1968.

[10] Fuchs, K., "The conductivity of Thin Metallic Films According to Electron Theory of Metals," Camb. Phil. Soc., Vol. 34, 1938, pp. 100 – 108.

[11] Tien, C. L., Armaly, F. G., ana Jagannathan, P. S., "Thermal Conductivity of Thin Metallic Films and Wires at Cryogenic Temperatures," Proc. of Eighth Thermal Conductivity Conference, Plenum Press, New York, 1969, pp. 13 – 20.

[12] Dingle, R. B., "The Anomalous Skin Effect and the Reflectivity of Metals, IV, Theoretical Optical Properties of Thin Metallic Films," Physica, Vol. 19, 1953, pp. 1187 – 1199.

[13] Armaly, F. G. and Tien, C. L., "Emissivities of Metals at Cryogenic Temperatures," to be presented at the International Heat Transfer Conference, Paris, 1970,

[14] McCargo, M. and Greenberg, S. A., "Range of Low Energy Protons in Metals and Metal Oxides," submitted for publication.

[15] Ruccia, F. E. and Hinckley, R. B., "The Surface Emittance of Vacuum-Metallized Polyester Film," in Advances in Cryogenic Eng., Vol. 12, Plenum Press, New York, 1967, pp. 300 – 307.

[16] Caren, R. P., "Low-Temperature Emittance Determinations," in Progress in Astronautics and Aeronautics, Vol. 18, Academic Press, New York, 1966, pp. 61 – 73.

[17] Bennett, H. E. and Bennett, J. M., "Precision Measurements in Thin Film Optics," Physics of Thin Films, Vol. 4, Academic Press, New York, 1967, pp. 1 – 96.

	60°K			100°K			300°K		
	N	ΔN	% Error	N	ΔN	% Error	N	ΔN	% Error
T_s °K	60	0.5	3.3	100	0.7	2.8	3.00	2.5	3.3
Q_c-Watts	2.0×10^{-5}	0.15×10^{-5}	7.5	1.60×10^{-4}	0.01×10^{-4}	0.6	3.60×10^{-2}	0.018	0.5
a_b	0.98	0.01	1.0	0.98	0.01	1.0	0.98	0.01	1.0
F	Neglect	Neglect	Neglect	Neglect	Neglect	Neglect	Neglect	Neglect	Neglect
ϵ_s	0.009	0.001	8.3	0.0105	0.0003	3.0	0.021	0.0007	3.4

Table 1 HEMISPHERICAL EMITTANCE DATA – PROBABLE ERRO VARIOUS SAMPLE TEMPERATURES

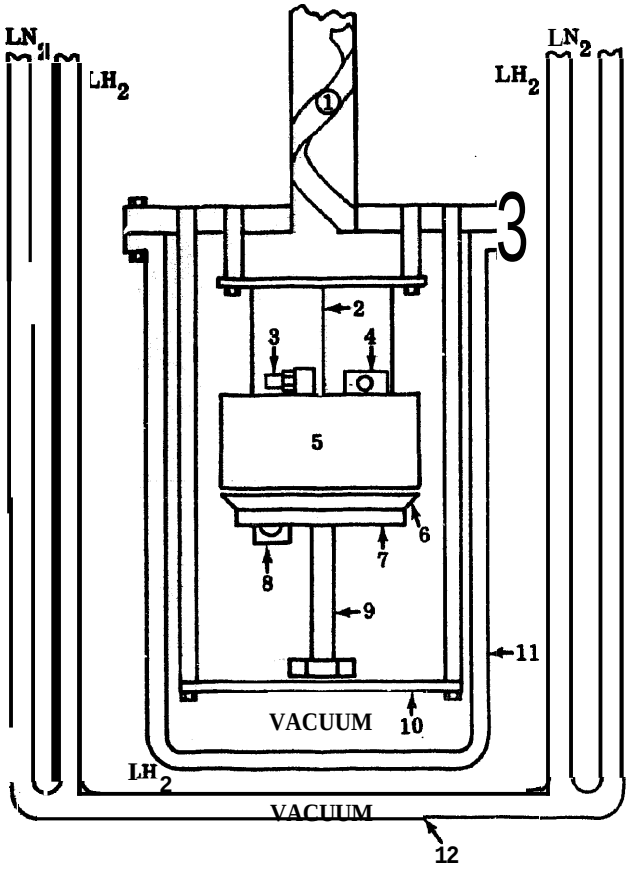
Gold on Mylar						Gold on Kapton	
470		550 Å (a)		720 Å (b)		400 Å (c)	
ϵ	Temperature (°K)	ϵ	Temperature (°K)	ϵ	Temperature (°K)	ϵ	Temperature (°K)
0.0109 ± 8.3%	61	0.0114 ± 8.3%	63	0.0083 ± 8.3%	63	0.0188 ± 5%	196
0.0119 ± 3 %	88	0.0129 ± 3 %	91	G.0102 ± 3 %	90	0 0204 ± 5%	255
0.0131 ± 3 %	129	0,0152 ± 3 %	129	0..0120 ± 2 %	132	0.0212 ± 5%	314
0.0154 ± 3 %	162	0.0156 ± 3 %	149	0.0132 ± 3 %	166		
0.0203 ± 3 %	253	0.0212 ± 3 %	253	0.0175 ± 3 %	259		
0.0220 ± 3 %	307	0.0244 ± 3 %	302	0.0210 ± 3 %	312		

Gold on Copper							
400		520 Å (d)		4000 Å (d)		2500 Å (c)	
E	Temperature (°K)	ϵ	Temperature (°K)	E	Temperature (°K)	ϵ	Temperature (°K)
0.0097 ± 8.3%	59	0.0108 ± 8.3%	59	0.0085 ± 8.3%	59	0.0135 ± 5%	201
0.0121 ± 3 %	96	0.0118 ± 3 %	96	0.0096 ± 3 %	97	0.0153 ± 5%	255
0.0158 ± 3 %	161	0.0153 ± 3 %	163	0.0118 ± 3 %	161	0.0164 ± 5%	302

Notes:

(a) Thickness measured on film samplea, adjacent to emittance specimen, 200 Å thickness variation observed across sheet, ± 100 Å.
(b) Thickness meaeured on 1-in. square area from center of emittance specimen;. *i50* Å.
(c) Thickness per supplier, measurement method not known.
(d) Thickness measured an monitors adjacent to emittance specimen, ± 50 Å.

Table 2 CALORIMETRIC TOTAL HEMISPHERICAL EMITTANCE DATA OF VACUUM DEPOSITED GOLD FILMS



1 COPPER RADIATION STRAP
2 BLACKBODY THERMAL LINK (THREE 20-mil STAINLESS WIRES)
3 GERMANIUM RESISTANCE THERMOMETER
4 BLACKBODY HEATER
5 BLACKBODY ABSORBER
6 SAMPLE SUBSTRATE
7 SAMPLE HEATER
8 PLATINUM RESISTANCE THERMOMETER
9 SAMPLE THERMAL LINK
10 COPPER SUPPORTING STRUCTURE
11 VACUUM-TXGHT EXPERIMENTAL CHAMBER
12 DOUBLE WALL DEWAR

Figure 1. Calorimetric Emittance Apparatus

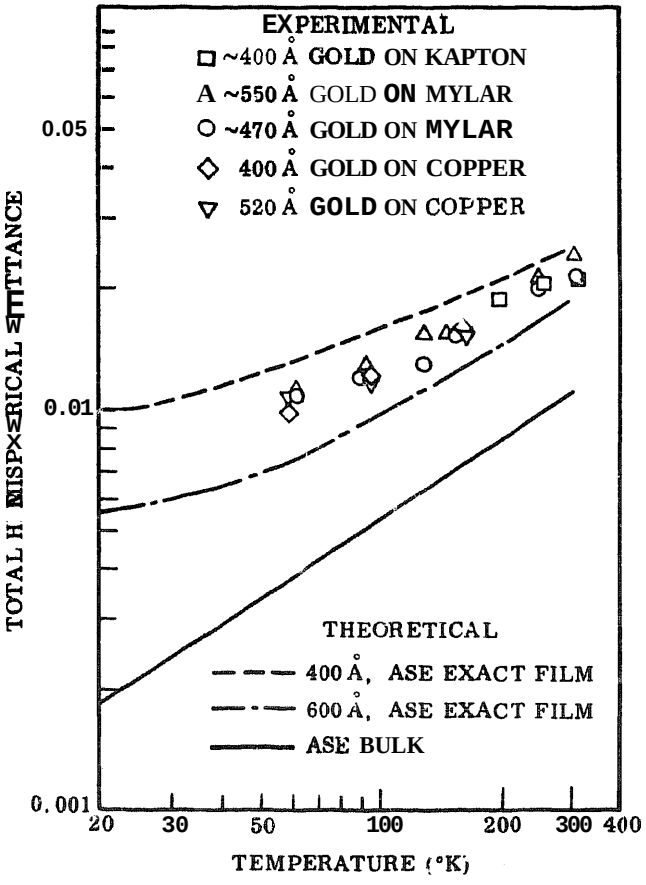


Figure 2. Total Hemispherical Emittance Versus Temperature for Thin Gold Films on Mylar and Copper

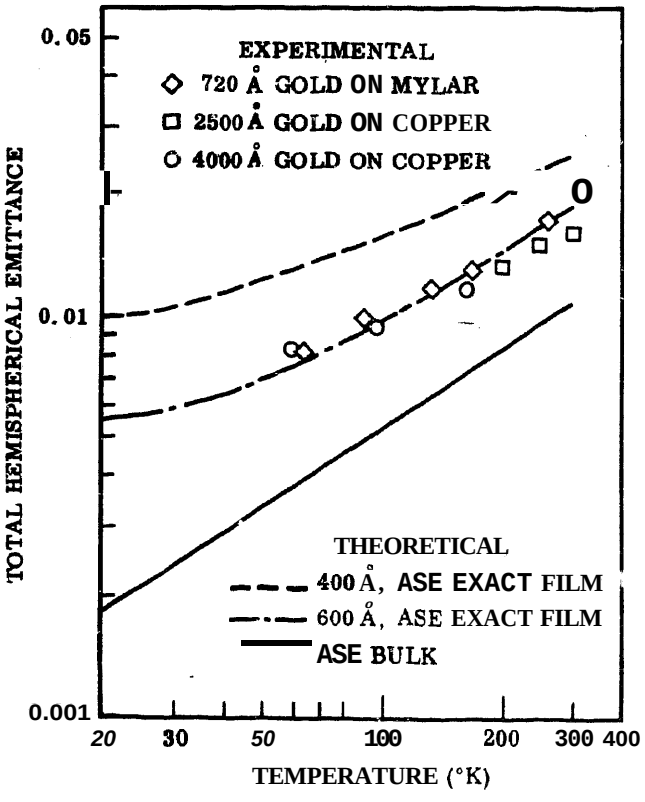


Figure 3. Total Hemispherical Emittance Versus Temperature for Thick Gold Films on Mylar and Copper

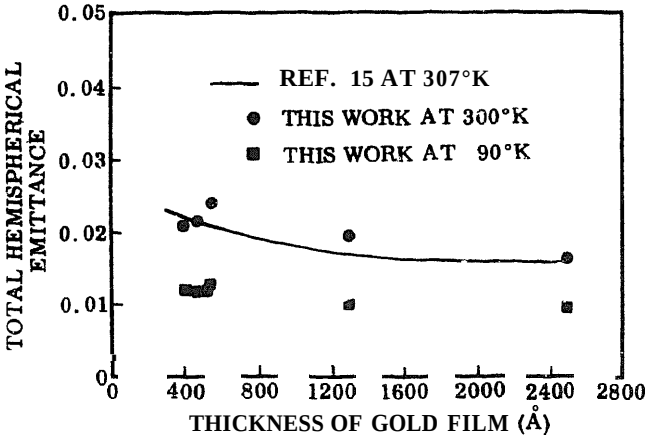


Figure 4. **Total Hemispherical Emittance of Gold Films Versus Film Thickness for 300° anti 90°K**

END