Theoretical Notes
Note 269

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X-ray photoemission from the following types of coated and low-z surfaces was measured: dielectric films backed by a conductor, painted metal foils, and low-z conductors. Steady-state irradiation was provided by an x-ray tube operated at 60 and 100 kV. The total emission current from front and/or back surfaces was measured for photons incident at 0° and 60°. Surfaces studied included: Be, CH_2, graphite, Mylar, Teflon, glass, PVC, and Saran plus various coatings of low-z and high-z paints on aluminum.

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19. KEY WORDS (Continued)

20. ABSTRACT (Continued)

Low-energy secondary electrons, which contributed up to 30 percent of the total emission, were retarded with a biased grid. On a few higher-z dielectrics, charge buildup depressed the measured photoemission currents around zero bias. Photoemission yields increased approximately as the cube of an effective atomic number, and CH_2 had the lowest observed photoemission yield. The yield from beryllium was two to three times that of CH_2; this large yield is several times higher than expected for pure Be and is attributed to a thick oxide layer. Present measured values plus previously-measured values of photoemission from metals are compared with results obtained from the QUICKE2 computer code, and agreement within 20 percent was found for most materials. Photoemission from a copper foil coated with low-z varnish was also measured as a function of varnish thickness. The observed emission decreased approximately exponentially with coating thickness to a minimum value corresponding to emission from varnish only. A simple model was developed to estimate such photoemission from a high-z metal coated with a thin low-z layer.

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FIGURES

TABLES

I. INTRODUCTION

Several experimental studies have been done on the x-ray photoemission from metals whose atomic number z ranged from 13 on up.[1-5] Results of theoretical computations are in fair agreement with most of these measurements.[5,6] But for comparison with theory, there is little data for x-ray photoemission from low-z surfaces, particularly from dielectrics.

In one earlier study, photoemission was measured from moderate-z surfaces of NaCl, KCl, etc.[7] During two other previous studies, pulsed x-ray sources were used with parallel-plate diode geometries to measure the photoemission from various dielectrics and low-z surfaces.[8-10]

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[1] I. M. Izrailev, "Photoelectric Yields for Soft X-Rays," Zh, Tekh. Fiz. 32, 1382 (1962) [Sov. Phys. - Tech. Phys. 7, 1020 (1963)]

[2] J. N. Bradford, "Absolute Yields of X-Ray Induced Photoemission from Metals," IEEE Trans. Nucl. Sci., NS-19, 167 (1972)

[3] M. J. Bernstein and K. W. Paschen, "Forward and Backward Photoemission Yields From Metals at Various X-Ray Angles of Incidence," IEEE Trans. Nucl. Sci., NS-20, 111 (1973)

[4] M. J. Bernstein and K. W. Paschen, Forward and Backward Photoemission Yields From Metals at Various X-Ray Angles of Incidence, TR-0076(6250-30)-The Aerospace Corporation, El Segundo, Calif. (Being Published)

[5] K. W. Dolan, "X-Ray-Induced Electron Emission from Metals," J. Appl. Phys. 46, 2456 (1975)

[6] R. R. Schaefer, "Simple Model of Soft X-Ray Photoemission," J. Appl. Phys. 44, 152 (1973)

[7] V. N. Schemelev and M. A. Rumsch, "X-Ray Photoeffect in Dielectric Cathodes," Fiz. Tverd. Tela. 5, 66 (1963) [Sov. Phys. - Solid State 5, 43 (1963)]

[8] F. Hai and M. J. Bernstein, "Photoemission from Polymers," IEEE Trans. Nucl. Sci., NS-18, 178 (1971).

[9] F. Hai and M. J. Bernstein, Photoemission from Polymers, TR-0172(2220-60)-2, The Aerospace Corporation, El Segundo, Calif. (15 March 1972)

[10] R. H. Barlett, et al., "Photon Induced Charge Transfer and Surface Emission," Proc. IEEE, 62, 1215 (1974)

Although different bias voltages were applied between the plates to help differentiate between the primary and low-energy secondary electrons, the results were distorted by emission from the second facing surface. In general, the photoemission from a dielectric layer backed by a conductor was similar to that from a conductor, but some differences seemed important. In particular, the energy spectrum of secondary electrons from some insulators, such as PVC, appeared quite different from that of metals, such as aluminum.

In this study photoemission currents from various paint coatings and from dielectric sheets backed by a conductor were measured using the same steady-state x-ray source employed previously for determining the emission from metals at various angles of incidence.[3] These photoemission currents were measured in detail as a function of bias voltage on a retarding grid. Measured values of primary photoemission from the dielectric and metal surfaces were then compared to values obtained from a widely-used computer code. There were also measurements of photoemission from metal foils coated with low-z paints of various thicknesses, and the results were compared to analytical values.

II. EXPERIMENT

The setup used for this study, shown in Figure 1, is basically the same as previously reported.[3] Photons from an x-ray tube entered the 27-cm-diameter vacuum chamber through a thin aluminum window, and photoelectrons were emitted from target foils hung on axis from a rotatable feedthrough. In most cases the thin sheets were mounted flat on a 0.15-cm-thick graphite sheet that caused negligible attenuation of the x radiation. Electron emission currents were measured using an electrometer with an input resistance of 10^{10} ohms, and signals of 1 to over 100 millivolts were obtained. Polyethylene was placed on the inside of the entrance and exit windows on the vacuum chamber to minimize photoemission from these surfaces. A lead collimator at the entrance window limited the diameter of the radiation beam to 3.2 cm on axis, and the target foils were 6.4 cm wide so that the x-ray beam matched the target width at an angle of incidence of 60°. The x-ray source was 28.5 cm from the axis. A 22-cm diameter bias grid with an average transparency of about 95 percent was used to retard (or attract) low-energy secondary electrons emitted from the foils. Bias potentials ranged from 0 up to ±500 volts. The grid wires were oriented so that no radiation was incident on them, but a tiny fraction of the photons hitting the target and support were scattered enough to strike the grid wires and emit a few electrons. Similarly, when primary electrons struck the grid wires, some secondary electrons were generated. In both cases, some of these electrons traveled back to the target when the grid was biased negatively and reduced the observed emission current by a very small amount.

The radiation source was a 160-kV Machlett x-ray tube connected to a 120-kV power supply. At the end of these latest measurements, it was discovered that the currents and voltages on the x-ray tube were slightly higher than the dial settings, so that the photon flux was about 10 percent greater than previously assumed. For the present measurements, the

actual operating parameters of the x-ray tube were 62 kV at 2.2 mA and 104 kV at 1.1 mA after accounting for the voltage drop across the ballast resistance. The photon flux was counted using a NaI-photomultiplier combination with a pulse-height spectrometer. The radiation passing from the x-ray tube anode to the inside of the vacuum chamber was attenuated by the equivalent of 0.72 gm/cm^2 aluminum; an additional filter of 0.25 gm/cm^2 copper was also employed during operation at the higher voltage. The resulting three photon flux spectra are shown in Figure 2 along with a corrected harder spectrum for the filter of 0.59 gm/cm^2 copper used in the previous study.

III. MATERIALS TESTED

Samples used for these photoemission measurements were thin sheets of polyethylene, Mylar, beryllium, glass, Saran, Teflon, and polyvinyl chloride that were available in our laboratory. Compositions of these materials, their densities, and thicknesses are listed in Table I. No special surface preparation was used other than to wipe with acetone. Ideally a sample thickness should be comparable to an electron range at the highest photon energy while x-ray attenuation is negligible at the lowest photon energy. As noted in our earlier study, there was some attenuation in the metal foils at large angles of incidence, but this effect could be accounted for by comparing the forward and backward emission currents at oblique incidence. Photoemission was also measured from sheets of aluminum foil coated with various Krylon spray paints; some paints had low-z pigments while others had compounds containing metals up to zinc. The estimated composition of the coatings is also given in Table I.

In contrast to our previous study, most samples of low-z target films were placed on both sides of the graphite support so that the sum of forward and backward emission currents was measured for the same material. In this way the contribution from one side of the support did not have to be subtracted. Radiation angles of incidence were 0° and 60°. During the second part of this study, a sheet of copper coated with thin layers of low-z varnish (G.E. insulation varnish 7031) was mounted on only one side of the graphite support. This varnish was dissolved in alcohol, and uniform layers were built up on the copper sheet by dip coating followed by oven baking. The thickness of the layer for each measurement was determined by weighing the target sample after dip coating and subtracting the weight of the foil itself. It was determined that the areal density was accurate to ±10 percent and uniform to ±5 percent. An additional thick sample of varnish was prepared by painting a heavy layer onto an aluminum foil.

IV. RESULTS

Measured values of emission currents as a function of bias voltage on the retarding grid are shown in Figure 3a for many of the materials tested. All these data exhibit somewhat similar S-shaped curves as the bias was varied from -500 volts through zero to +500 volts, and the secondary electrons appear to have energies well below 100 eV. The rather large increase in the observed emission current at positive grid potential has not been definitely explained. It probably arises from field extraction of additional secondary electrons at the surface, since even the conductors will have some type of thin (5-50 nm) oxide coating or other dielectric surface. No other explanation is apparent at present .... space-charge effects are totally negligible for the magnitudes of emission currents used here. A somewhat similar increase in observed emission with positive bias was observed by others.[7,*]

Although the results of this study are directed principally at the magnitudes of the primary electrons above 1 keV in energy, it is of interest to note the characteristics of the secondary-electron emission. First of all, the secondary-emission coefficient (ratio of secondaries to primaries) appears highest for the lowest-z surfaces and decreases as the effective z becomes larger. Secondly, there appear to be definite differences in the energy spectrum of the secondaries for the different materials.

Photoemission data for three dielectric samples did not follow the typical S-shaped curve as the bias voltage was increased, but exhibited dips in the emission near zero bias as shown in Figure 3b. The values measured around zero bias were also not very reproducible as indicated by the scatter of plotted data during many readings taken over a period of time. These

*A. R. Fredericson (private communication)

dips in the curves are attributed to a buildup of surface charge, and we observed such effects only for the following two conditions:

a. The volume resistivity through the sheet to ground had to be larger than the input resistivity of the electrometer, and

b. The magnitude of the emission had to be at least comparable to that of aluminum.

The thin Saran exhibited such behavior only when the film was just taped to the graphite support so that there was limited electrical contact. When a thin conducting coating of Aquadaq was applied to the inside surface of the Saran touching the graphite, the emission data appeared normal as given in Figure 3a. Although polyethylene had a large value for the volume resistivity, only a slight dip in the bias curve data was observed on the thicker sample even when the emission was enhanced by a coating of high-z paint on the surface. It now appears that our previously-reported large difference in the energy spectrum of secondary electrons from PVC compared to that from aluminum[8,9] was mainly caused by charge buildup on the PVC and the random nature of taking a few data points. We found that the dip in the photoemission data for PVC could be enhanced (lowered) by the deposition of positive ions onto the surface from an ionization-gauge tube.

Values of the primary photoemission current densities were taken to be the average of forward and backward emission at 60° angle of incidence with an applied negative bias. These average values are tabulated in Table II and are also plotted in Figure 4 as a function of effective atomic number $\bar{z}$ defined by

$$\bar{z} = (\Sigma_i A_i z_i^3 / \Sigma_i A_i)^{1/3}$$

where z_i and A_i are the atomic number and fractional weight of each element in a compound. A plot of photoemission data for higher-z metals was given in a paper documenting our previous study.[3] The height of each symbol in Figure 4 represents the estimated overall experimental uncertainty; measurements taken a year apart on different samples of Al and Cu gave results

that agreed within 10 percent. Widths of the symbols in Figure 4 correspond to uncertainties in $\bar{z}$ that were largest for a high-z paint pigment in a low-z binder. Overall chemical composition of the paints after drying were not known and no laboratory analyses were made.

Typical sets of data for forward photoemission from the coated copper foil as a function of varnish thickness are shown in Figure 5 for irradiation at normal incidence. Additional forward photoemission data taken for irradiation at 60° gave similar curves with magnitudes about 80 percent higher (a small fraction of the diverging beam missed the target at 60°). Backward photoemission at normal incidence differed from the forward emission in the following respects: emission from the uncoated copper was about 30 percent lower while the emission from the thick varnish was down by at least a factor of two, so that the photoemission curves decreased slightly faster with varnish thickness.

V. ANALYSIS

In our previous study, the photoelectric yield at each photon energy was represented by the following expression:

$$n_e/n_x = G_e \sum_{K,L,M} \mu_a S_e \quad \text{electrons/photon}$$

where G_e is a constant for each material independent of photon energy over the present range of interest, μ_a is the photon energy-absorption cross section,[11] S_e is the computed mean path length,[12] and the summation is for photoelectrons emitted from the different atomic shells. Auger electrons were neglected since their creation efficiency was low because the photon energies were much higher than the K shells of lower-z materials or the material was high-z. Values of G_e were found to decrease with higher atomic number consistent with the effects of greater electron scattering. Since the photon flux was found to be 10 percent greater than originally assumed, the G_e values are correspondingly reduced. Representative revised values of G_e for photon energies of 20 to about 80 keV are: C(graphite), 0.34 ± 0.07; Al, 0.27 ± 0.03; Cu, 0.20 ± 0.02; Ta, 0.16 ± 0.02.

Values of photoemission current densities were calculated for the various materials using the analytic computer code QUICKE2.[13] Computed values

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[11] W. H. McMaster, et al., "Compilation* of X-Ray Cross Sections," UCRL-50174, Lawrence Radiation Laboratory, Livermore, Calif. (May 1969)

[12] M. J. Berger and S. M. Seltzer, "Tables of Energy-Losses and Ranges of Electrons and Positrons," Studies of Penetration of Charged Particles in Matter, National Research Council Publ 1133, 205 (1964)

[13] T. A. Dellin and C. J. MacCullum, "Photo-Compton Currents Emitted from a Surface," J. Appl. Phys. 46, 2924 (1975)

*Other tabulated values agree to better than 10 percent.

of forward and backward emission for photons incident at 60° were averaged and are presented in Table II for comparison with the measured values. There is generally very good agreement. Experimental uncertainties include the magnitude of the photon flux and slight attenuation of the low-energy photons in some of the thicker samples. Computed sums of forward and backward emission from the metals at normal incidence are 5 to 10 percent lower than at oblique incidence which is in agreement with our earlier experimental results. The ratios of forward to backward emission at normal incidence also agreed with the measured values within experimental error. It was stated by the authors of QUICKE2 that the computed backward emissions from low-z surfaces are probably low, but this was not observed since only the sum was measured.

The QUICKE2 code also computes the emitted number of secondary electrons that yield secondary emission coefficients. For our spectra, the computed coefficients differed from the measured values in two respects. First, the computed values were around 2 to 3 times larger than observed. Secondly, the computed coefficients for each spectrum increased with atomic number from Al to Ta, while the observations indicated the reverse trend.

Computations of photoemission from coated metal surfaces require use of a long-running Monte Carlo code such as SANDYL[14] or POEM[15] that can handle multiple layers. For comparison with experimental results, such as the coated copper used here, a separate computer run would be needed for each varnish thickness of interest. This would use an unreasonable amount of computer time. As an alternative, a simple empirical model was developed that used the electron-energy distributions $N_e(E_e)$ generated by QUICKE2 for emission from metals. This model provides an estimate of the fraction

[14] H. M. Colbert, SANDYL: A Computer Program for Calculating Combined Photon-Electron Transport in Complex Systems, Report SLL-74-0012, Sandia Laboratories, Albuquerque, N.M. (1974)

[15] W. L. Chadsey, POEM: A Fast Monte Carlo Code for the Calculation of X-Ray Photoemission and Transition Zone Dose and Current, TR-0075-0324, Air Force Cambridge Research Laboratory, Hanscom Field, Mass. (1975)

of electrons emitted by a higher-z metal that pass through various thicknesses of low-z coatings. In our case, we wish to analyze the emission from copper through the layers of varnish. Therefore, the experimental results in Figure 5 were corrected for the emission from the varnish itself and the adjusted curves are shown in Figure 6.

Photoelectrons from a metal, which enter a low-z coating with an initial energy E_e, can penetrate a distance up to the mean pathlength $S_e(E_e)$ as computed from the average stopping power. Most electrons penetrate a much smaller distance because of both their initial angular distribution and the effects of scattering. To account for this range of penetration, we divided the value $N_e(E_e)$ at each by E_e by the corresponding value $S_e(E_e)$. The resulting curves for coated copper are also plotted in Figure 6 where the functional dependence of these analytical results was changed from E_e to S_e so that all curves would have common units on the abscissa. The analytical curves were normalized to the experimental curves near zero thickness for the varnish. Considering the simplicity of the model, there is reasonable agreement for the three spectra. Some of the uncertainty in the analytical curves arises from the discrete limited number of points used during the QUICKE2 calculations.

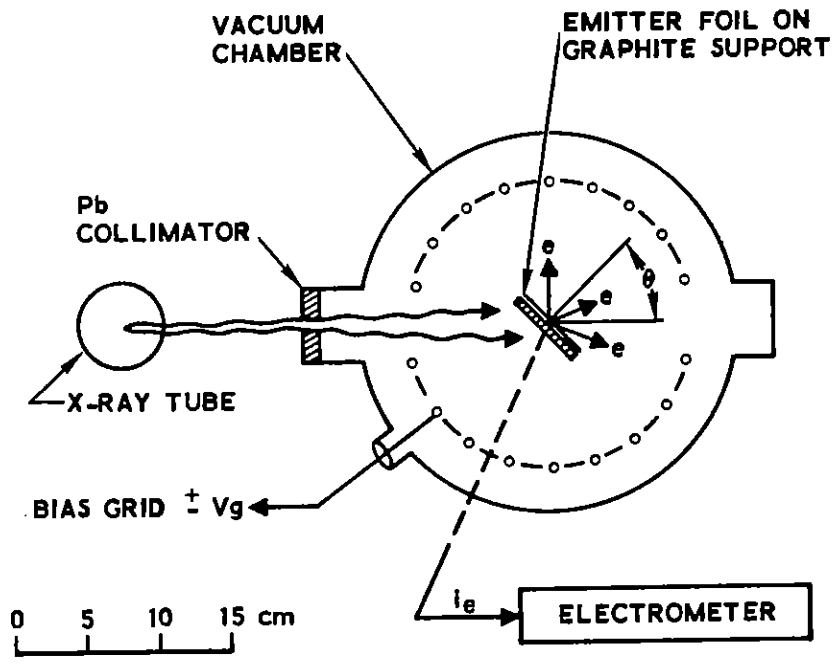


Fig. 1. Schematic Diagram of Apparatus to Measure X-Ray Photoemission from Coated Surfaces in Forward and/or Backward Directions

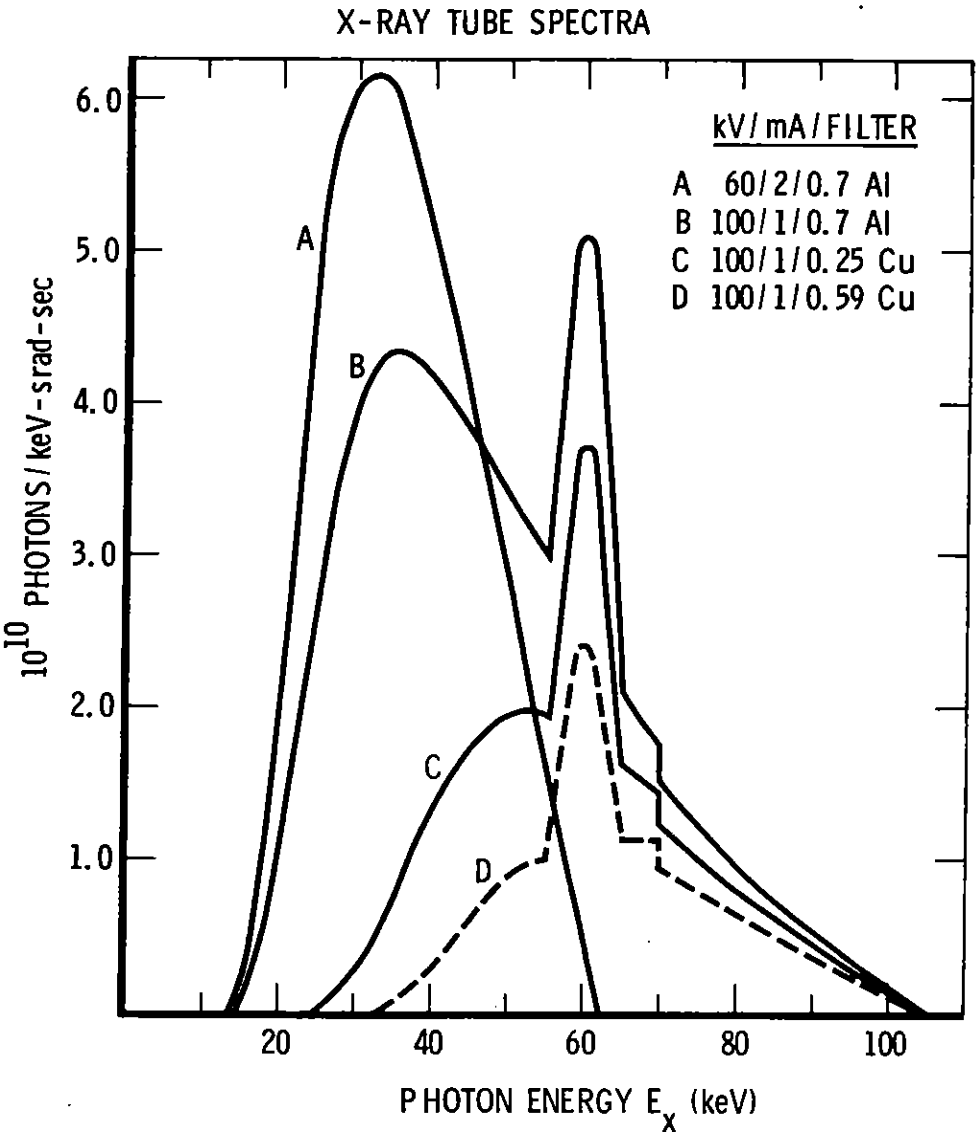
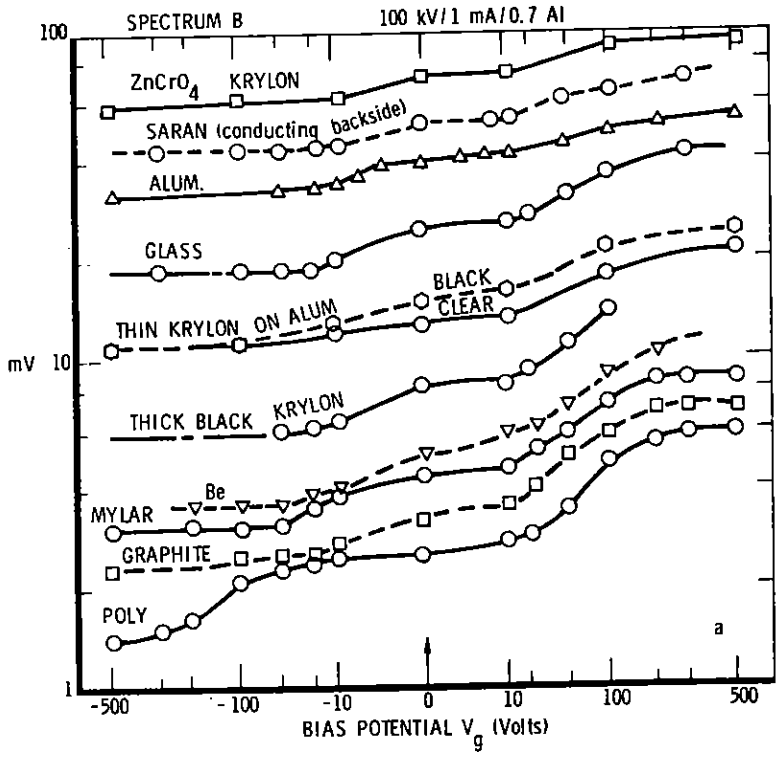
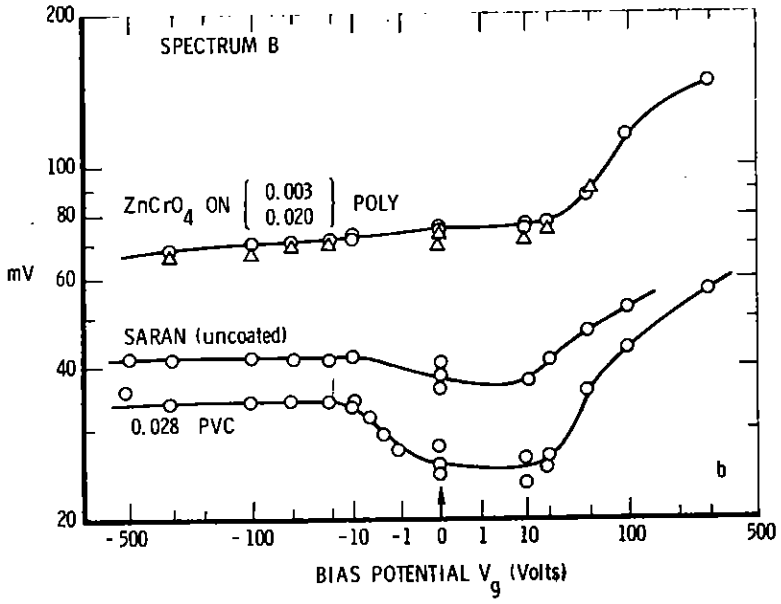


Fig. 2. Filtered X-Ray Spectra Used for Photoemission Studies. (Peak voltages on x-ray tube were actually 62 and 104 kV.)



a. Data for Materials Acting as Conductors.



b. Data Affected by Charge Buildup on the Dielectric.

Fig. 3. Photoemission Current Signals as a Function of Bias Voltage on Retarding Grid for $\theta_i = 60°$. (Electrometer input was 10^{10} ohm.)

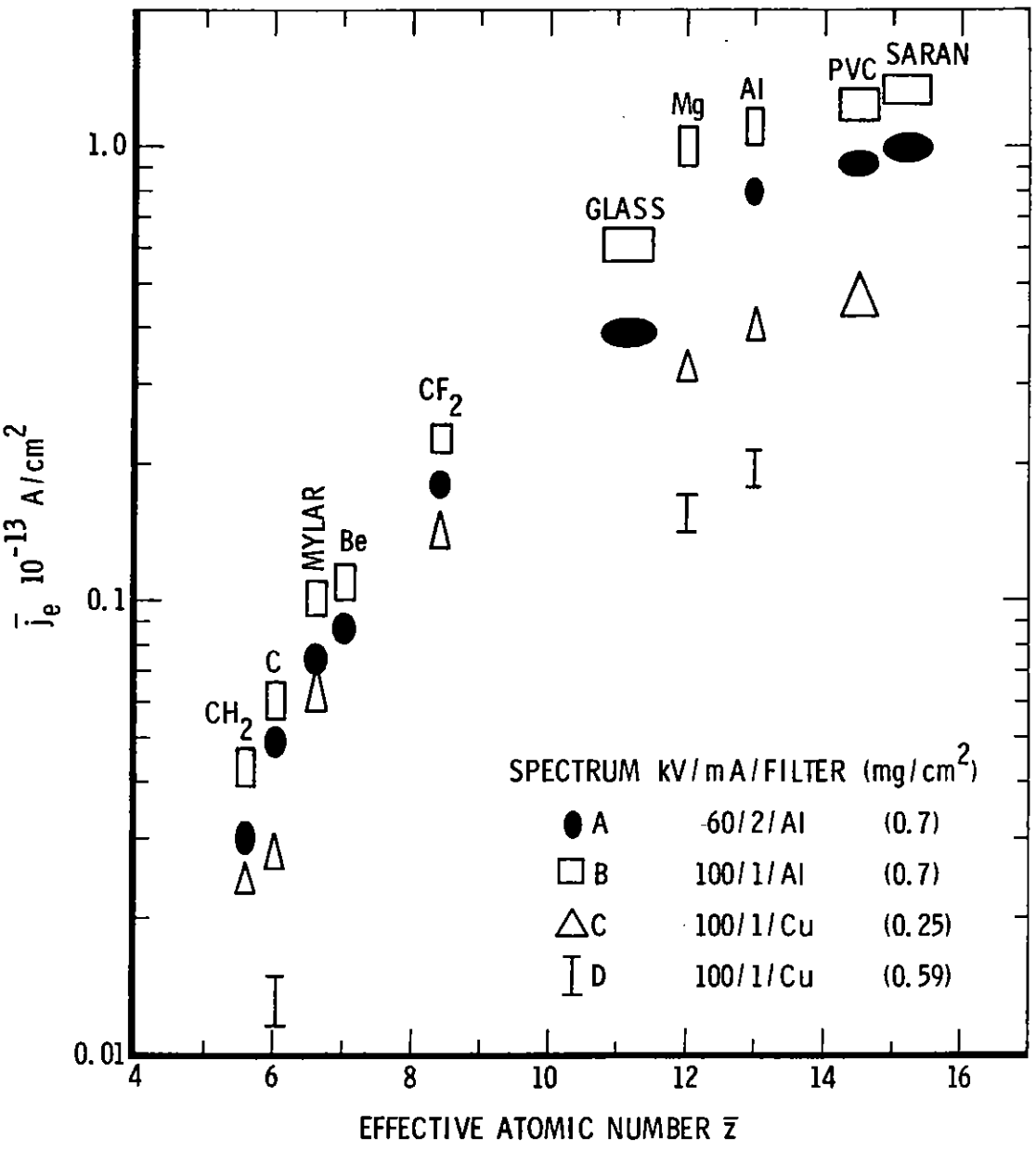


Fig. 4. Average Photoemission Current Densities j_e from Surfaces as a Function of Effective Atomic Number

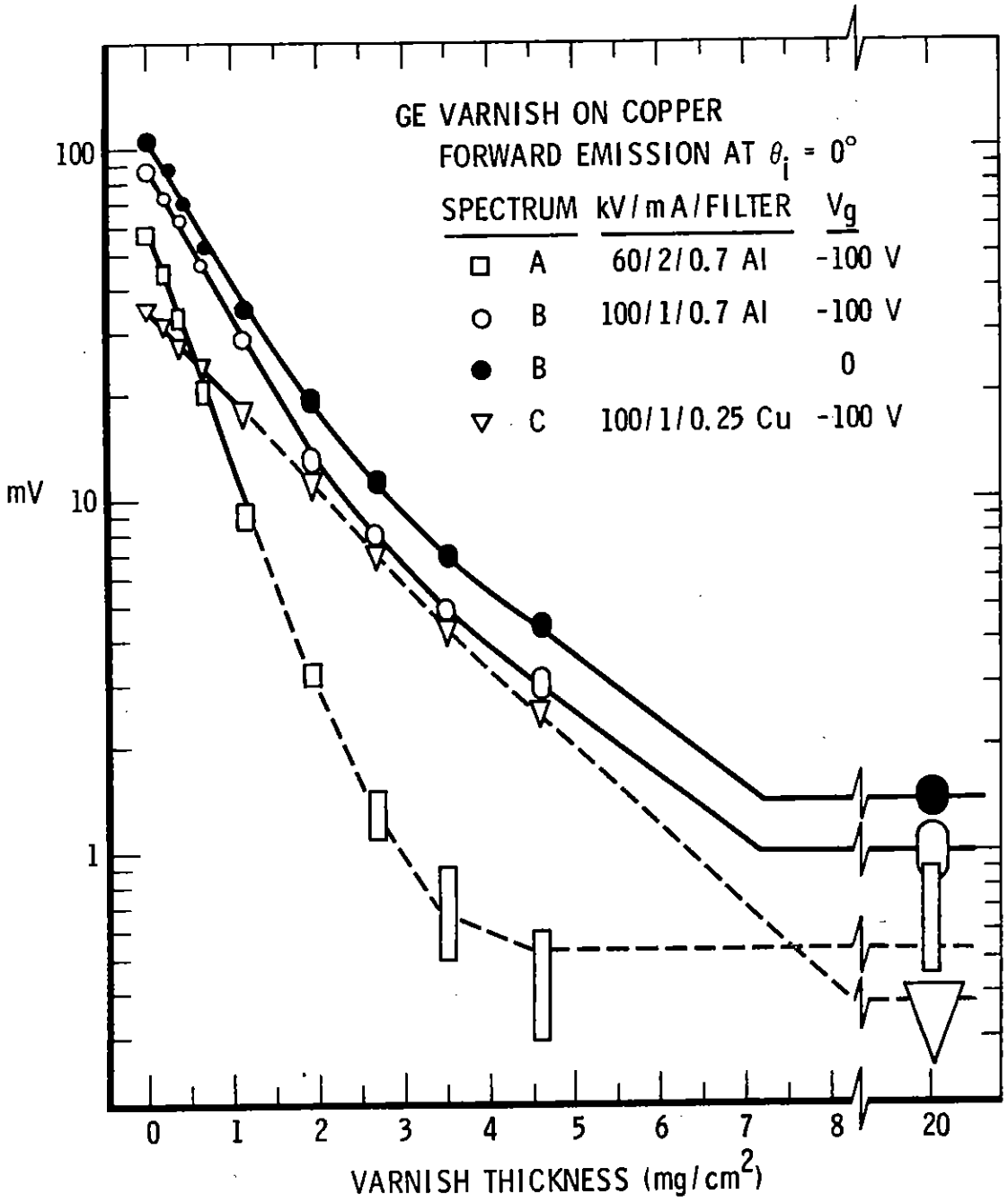


Fig. 5. Photoemission Signals from Coated Copper as a Function of Varnish Thickness. [Open symbols for primary electrons only (negative bias on grid), while closed symbols for one spectrum shows additional contribution of secondaries.]

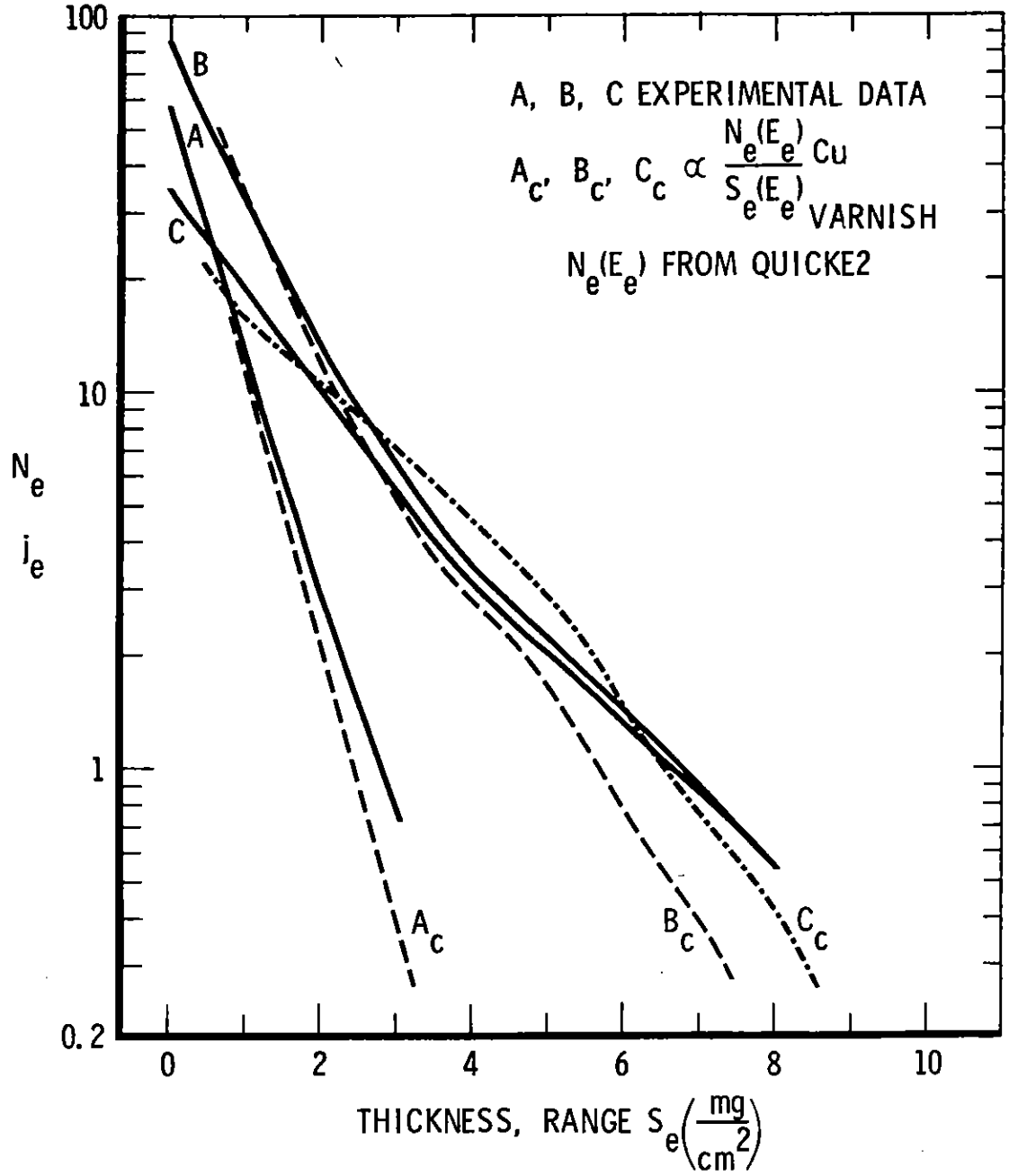


Fig. 6. Magnitudes of Photoelectron Flux from Copper that is Transmitted Through Varnish of Increasing Thickness. (Solid lines are data from Fig. 5 adjusted for emission from varnish itself. Dashed curves are derived from values of electron-energy distribution $N_e(E_e)$ that are divided by $S_e(E_e)$ at each electron energy; these analytical results are normalized to solid lines near zero thickness.)

Table I. Sample Thicknesses of Materials Studied Plus Assumed Composition and Effective Atomic Number of Photoemitting Surfaces

Material Tested	Assumed Composition	Effective at No. $\bar{z}$	Thickness
POLYETHYLENE	CH_2	5.6	0.08 mm
GRAPHITE	C	6.0	1.5
KAPTON	--	6.5	0.03
MYLAR	$C_{10}H_8O_4$	6.7	0.05
SCOTCH TAPE	--	--	0.05
INSULATING VARNISH (G.E. 7031)	--	6.9	0.10
BERYLLIUM	BeO	7.0	0.013
KRYLON PAINTS (Clear and black)	C_2H_2O	7.0	0.17
GLASS	SiO_2	11.3	1.25
ALUMINUM	Al	13.0	0.18
PVC	C_2H_3Cl	14.0	0.71
SARAN	$C_2H_2Cl_2$	15.0	0.013
GREEN KRYLON PAINT ($ZnCrO_4$)	$ZnCrO_4 + C_n$	18 - 22	0.02
COPPER	Cu	29.0	0.025

Table II. Primary Photoemission Currents ($E_e > 1$ keV) in Units of 10^{-13} A/cm^2. (Measured values and QUICKE2 computed results are average of forward and backward emission for $\theta = 60°$. Uncertainty in measured values ranged from ±20% for the low-z surfaces to ±10 percent for the higher-z materials.)

MATERIAL	SPECTRUM A 60/2/0.7 Al		SPECTRUM B 100/1/0.7 Al		SPECTRUM C 100/1/0.25 Cu		SPECTRUM D 100/1/0.59 Cu	
	MEAS.	QUICKE2	MEAS.	QUICKE2	MEAS.	QUICKE2	MEAS.	QUICKE2
Polyethylene	0.026	0.034	0.044	0.054	0.025	0.023	--	--
Graphite	0.050	0.046	0.072	0.070	0.028	0.027	0.013	0.015
Kapton	0.060	--	0.078	--	--	--	--	--
Mylar	0.072	0.070	0.095	0.098	0.063	0.035	--	--
Scotch Tape	0.078	--	0.10	--	--	--	--	--
G.E. Varnish	0.074	--	0.11	--	0.040	--	--	--
Beryllium (BeO)	0.088	0.090	0.11	0.125	--	0.045	--	--
Krylon (low-z)	0.11	--	0.16	--	--	--	--	--
Teflon	0.18	0.16	0.23	0.215	0.14	0.073	--	--
Glass	0.39	0.48	0.59	0.63	--	--	--	--
Magnesium	--	0.55	0.97	0.72	0.33	0.23	0.16	0.11
Aluminum	0.81	0.72	1.10	0.92	0.39	0.30	0.19	0.142
PVC	0.92	--	1.22	--	0.45	--	--	--
Saran	1.03	1.13	1.34	1.52	--	0.50	--	--
Krylon ($ZnCrO_4$)	1.35	--	1.85	--	1.22	--	--	--
Titanium	--	--	5.3	4.5	1.63	1.52	0.76	0.74
Copper	6.5	5.9	9.8	9.0	3.57	3.18	1.75	1.57
Silver	--	--	20.0	16.8	8.7	7.6	4.4	4.1
Tantalum	--	--	26.3	22.2	9.7	8.2	5.0	4.3

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