

## Increase of Gold-Teflon FEP Joint Strength by Removal of Deposited Aluminum Prior to Gold Deposition for Electret Applications

R. F. ROBERTS, F. W. RYAN, H. SCHONHORN, G. M. SESSLER,\* and J. E. WEST, *Bell Laboratories, Murray Hill, New Jersey 07974*

### Synopsis

A new method is described for bonding gold to Teflon FEP, yielding high joint strength without affecting the electrical properties of the virgin polymer material. The method consists of depositing a 1000-Å aluminum layer onto the Teflon by evaporation, removing it by washing the sample in dilute sodium hydroxide, followed by deposition of gold by evaporation. The tensile shear strength of this composite is in excess of 80 kg/cm<sup>2</sup>, approaching the cohesive strength of Teflon. The desirable electrical properties of Teflon FEP, as measured by isothermal charge decay and thermally stimulated currents methods, are preserved by this method. X-Ray photoelectron (ESCA) spectra suggest that the modified wetting characteristics and enhanced joint strengths can be attributed in part to the existence of a thin layer of oxygen-containing hydrocarbon material on the surface of aluminum-treated Teflon FEP.

### INTRODUCTION

A number of methods are available to improve the generally weak adhesive joint strength between gold and Teflon FEP. Among the methods for modifying the polymer surface are chemical treatments,<sup>1</sup> sputtering processes,<sup>2</sup> CASING treatment,<sup>3,4</sup> and electron irradiation.<sup>5</sup> While all of these procedures yield acceptable joint strengths (in excess of 60 kg/cm<sup>2</sup>), most result in an electrical degradation of the polymer material, as evident from the decay of charge injected into the polymer and from measurements of open-circuit thermally stimulated currents (TSC) generated upon heating of the charged samples. The only exception is a method<sup>5</sup> utilizing irradiation of the polymer material with 5 to 20 keV electron beams and intercepted charge densities of about  $5 \times 10^{-6}$  C/cm<sup>2</sup>. Subsequent gold deposition results in samples which, apart from having joint strength, can be annealed to exhibit the same favorable charge retention properties typical for untreated Teflon with an aluminum electrode deposited by evaporation.

While it is thus possible to achieve the desired joint strength and charge retention behavior, the procedure necessary to accomplish this involves irradiation and heat treatment. This makes desirable simpler processes not requir-

\* Present address: Technische Hochschule Darmstadt, 61 Darmstadt, Merckstrasse 25, West Germany.

ing separate steps to ensure acceptable joint strength and good charge retention characteristics.

In this paper, a new method yielding the desired increase in joint strength without affecting the electrical properties of virgin Teflon FEP is described. After a brief description of the procedure for gold deposition and adhesive joint formation, results of joint strength and wettability measurements are presented. Next, the electrical properties of the material after joint formation are investigated using isothermal charge decay and thermally stimulated current techniques. Finally, the surface composition of the treated Teflon FEP, as determined by x-ray photoelectron spectroscopy (ESCA), is compared to that of the virgin material, and conclusions are drawn concerning the chemical changes which are instrumental in affecting the enhanced joint strengths.

### PROCEDURE FOR GOLD DEPOSITION AND ADHESIVE JOINT FORMATION

The substrate used in the present study was Teflon FEP film, 25  $\mu\text{m}$  type A, (E. I. du Pont de Nemours & Co., Inc.). The method for mounting the substrate prior to deposition has been described previously.<sup>5</sup> Deposition of aluminum by evaporation was accomplished in a Veeco 770 High Vacuum Thin Film Evaporator. The substrate was placed 12.0 cm from the tungsten filament (this distance is convenient rather than crucial). The filament was loaded with sufficient high-capacity wire (0.6 cm  $\times$  0.13 cm) to yield an evaporated film on the substrate of approximately 1000  $\text{\AA}$ . This required a vacuum of  $2 \times 10^{-6}$  torr and a current of 35 amperes. The films were coated with metal on both sides. The aluminum was then removed by immersing the metal-coated fluoropolymer in a 0.1N sodium hydroxide solution. The specimens were washed with deionized water and dried between fresh filter papers. Examination of the resulting films with x-ray fluorescence revealed no detectable amount of aluminum. Gold deposition was then achieved by a second evaporation process.

Composites of gold-Teflon or aluminum-Teflon for joint strength measurements were structured as shown in Figure 1. A description of the tensile

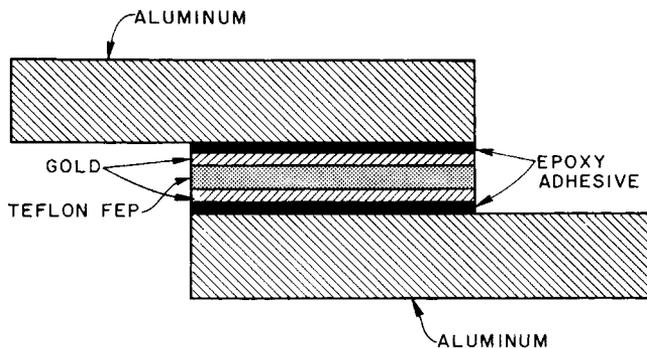


Fig. 1. Schematic cross section of aluminum-Teflon-aluminum composite, employing gold-metallized Teflon.

TABLE I  
Surface Region Modification of Teflon FEP by Metal Deposition

| Film   | Tensile shear strength<br>of composite<br>in Fig. 1, kg/cm <sup>2</sup> |
|--|---|
| Teflon FEP<br>(as received, no<br>metallization)   | ~0  |
| Teflon FEP<br>(untreated except for<br>sufficient exposure to<br>dil. NaOH which is<br>required to dissolve<br>aluminum, no metallization) | ~0  |
| Teflon FEP<br>(1000 Å gold deposited)  | ~0  |
| Teflon FEP<br>(1000 Å aluminum<br>deposited)   | 94  |
| Teflon FEP<br>(1000 Å film of<br>aluminum dissolved<br>from substrate by dil.<br>NaOH) <sup>a</sup>  | 124   |
| Teflon FEP<br>(after removal of aluminum<br>from above, 1000 Å of<br>gold deposited)   | 83  |

<sup>a</sup> No aluminum detected on Teflon FEP by x-ray fluorescence.

shear specimen and conditions for forming and breaking the joint is given elsewhere.<sup>5</sup>

### ADHESIVE JOINT STRENGTH AND WETTABILITY

The data in Table I indicate that the joint strength for the gold deposited-untreated Teflon FEP composite is essentially zero. A considerable enhancement of the joint strength results when aluminum is deposited by evaporation onto previously untreated Teflon FEP. This suggests that some surface modification of the fluoropolymer may have occurred during the deposition of aluminum. Since Teflon FEP is a difficult material to join, unless the surface region has been modified to enhance the critical surface tension of wetting<sup>6</sup> ( $\gamma_c$ ), it is unlikely that just placing a thin film of aluminum in intimate contact with the polymer film would be adequate for forming a strong adhesive joint.

Examination by x-ray fluorescence of the failed gold-untreated Teflon FEP composites revealed that no gold remained on the polymer; however, the low  $\gamma_c$  of the gold surface that had been in contact with the fluoropolymer indicated transfer of fluorocarbon during failure of the joint. This would imply that the gold adequately wets the Teflon FEP when deposited by evaporation but that the surface region of the polymer in the composite is unable to support a large shear stress.

TABLE II  
Wettability of Teflon FEP Modified by Aluminum Treatment<sup>a</sup>

| Liquid                     | $\gamma_{LV}$ , dynes/cm | Contact angle      |               |                    |               |
|----------------------------|--------------------------|--------------------|---------------|--------------------|---------------|
|                            |                          | Aluminum treated   |               | Untreated          |               |
|                            |                          | $\theta$ , degrees | $\cos \theta$ | $\theta$ , degrees | $\cos \theta$ |
| Octane                     | 21.8                     | 0                  | 1.00          | 29                 | 0.875         |
| Dodecane                   | 25.4                     | 17                 | .956          | 42                 | 0.743         |
| Tetradecane                | 26.7                     | 18.5               | .948          | 47                 | 0.682         |
| Hexadecane                 | 27.6                     | 23                 | .921          | 51                 | 0.629         |
| $\alpha$ -Bromonaphthalene | 44.6                     | 59                 | .515          | 68                 | 0.375         |
| Methylene iodide           | 50.8                     | 55                 | .574          | 78                 | 0.208         |
| Formamide                  | 58.2                     | 78                 | .208          | 90                 | 0.000         |
| Glycerol                   | 63.4                     | 86                 | .070          | 95                 | 0.087         |
| Water                      | 72.8                     | 90                 | .000          | 110                | 0.342         |

<sup>a</sup> 1000 Å aluminum vapor deposited, then dissolved off the polymer by 0.1*N* sodium hydroxide. Washed until no traces of sodium and aluminum detected by x-ray fluorescence.

The results in Table I indicate that the deposition of aluminum by evaporation has resulted in a successful surface treatment for the adhesive bonding of Teflon FEP. Removal of aluminum followed by the deposition of gold yields joint strengths of about 80 kg/cm<sup>2</sup>, in excess of those achieved in the previous study<sup>5</sup> where we resorted to electron bombardment to modify the surface region of the Teflon FEP.

Measurement of the critical surface tension of wetting ( $\gamma_c$ ) offers an interesting approach to the question of surface layer changes. Table II lists the liquids used for measuring  $\gamma_c$  and the contact angles obtained with untreated and treated films (both films were exposed to dilute sodium hydroxide and washed with copious amounts of water). A Zisman plot of these data is shown in Figure 2. It is clear that there is a significant difference between the two differently treated Teflon samples in terms of their wetting characteristics. The  $\gamma_c$  increases to a value of 25 dynes/cm for the treated specimen, but the method offers no indications as to what may have occurred during the evaporation procedure. Visual examinations of the surface revealed

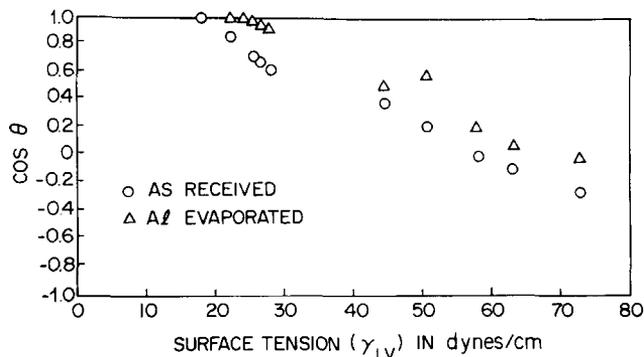


Fig. 2. A Zisman plot of  $\cos \theta$  vs.  $\gamma_{LV}$  for a variety of liquids on Teflon FEP. The extrapolated values ( $\cos \theta = 1$ ) yield the critical surface tension of wetting for the surfaces shown in Fig. 3: (O) corresponds to Fig. 3a; ( $\Delta$ ) corresponds to Fig. 3b.

no apparent roughening to account for these marked differences in wettability. Scanning electron microscopy (SEM) photographs at 5000 $\times$ , shown in Figure 3, reveal a suggestion of a surface roughening which is clearly below that expected to produce a significant change in contact angle.<sup>7</sup>

### ELECTRICAL PROPERTIES

The electrical properties of metallized Teflon FEP are of interest with respect to electret and other applications of this material. Since Teflon films metallized by aluminum evaporation show the most favorable charge retention and thermally stimulated current (TSC) characteristics<sup>8</sup> known, such films will be used as control samples for comparison with gold-coated samples.

For charge decay and TSC testing, the Teflon samples metallized on one side were first negatively charged through the polymer face by bombardment with monoenergetic beams of 20 keV electrons<sup>9</sup> under vacuum. The charging was carried to a level of about  $3 \times 10^{-8}$  C/cm<sup>2</sup>. The spatial depth of the charge is about 4  $\mu$ m under these conditions.<sup>8</sup> After charging, the samples were removed from vacuum for the performance of charge decay and TSC testing.

Results of isothermal charge decay at 150°C for gold-coated Teflon FEP samples are compared in Figure 4 with results for the control samples. For polymer foil-electrets contained in an oven at elevated temperatures, it has been determined<sup>10</sup> that an ion absorption mechanism, which results in charge compensation, can predominate if the polymer surface of the electret is unshielded. In the present study, the possibility of such ion absorption was eliminated by placing the samples in a metal container within an oven. Stor-

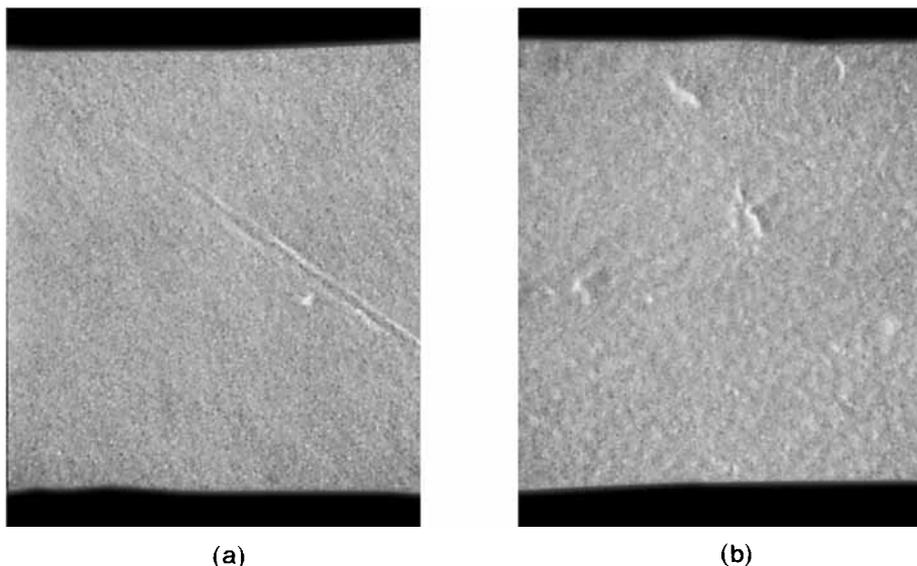


Fig. 3. (a) Scanning electron microscopy (SEM) photograph (5000 $\times$ ; 45°) of untreated Teflon FEP which had been exposed to 0.1N NaOH for 10 min. (b) SEM photograph (5000 $\times$ ; 45°) of Teflon FEP which had a 1000-Å film of aluminum deposited by evaporation and subsequently dissolved in 0.1N NaOH.

age within the container was such that the electret did not contact any surface, thus eliminating the possibility of charge decay due to radiation induced conductivity.<sup>12</sup> Figure 4 indicates the first stages of the double-exponential decay typically observed<sup>8</sup> at elevated temperature for negatively charged Teflon FEP. The decay on the gold-coated samples is slightly less than that on the control samples, but the difference is probably not significant.

Typical TSC curves for gold-coated and control samples are shown in Figure 5. The data were obtained by heating the negatively charged samples in open circuit (between evaporated electrode and second electrode at 0.5 cm distance from polymer face of sample) applying a linear heating rate of 1°C/min. The TSC peaks for the gold-coated and the control samples are both at

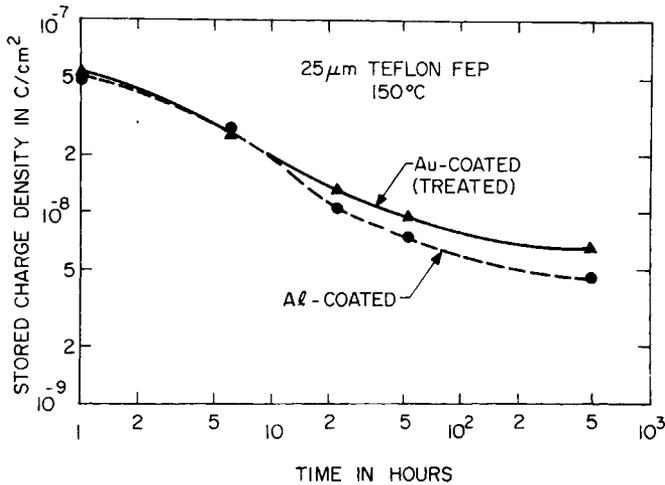


Fig. 4. Decay of stored charge density of gold-coated and aluminum-coated 25- $\mu\text{m}$  Teflon FEP samples as a function of storage time at 150°C. Pretreatment of gold-coated sample: (1) 1000 Å aluminum deposited; (2) aluminum removed with sodium hydroxide; (3) 1000 Å gold deposited.

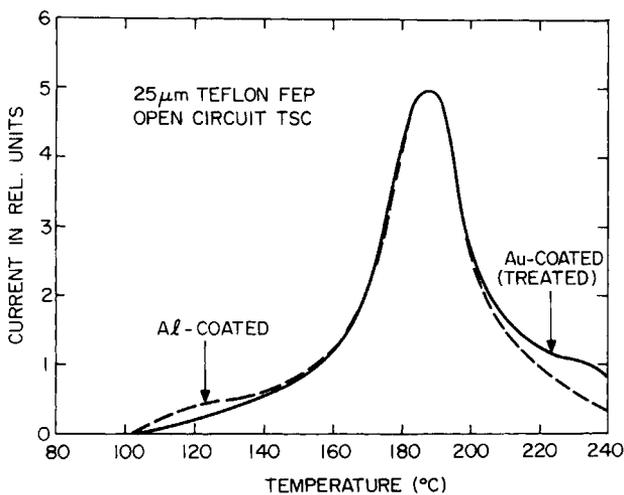


Fig. 5. Thermally stimulated current curves for gold-coated and aluminum-coated 25- $\mu\text{m}$  Teflon FEP samples. Pretreatment of gold-coated sample as in Fig. 4.

about 190°C, as has also been observed before for untreated negatively charged Teflon.<sup>11-13</sup> This has to be compared with peak temperatures of 130° to 150°C for Teflon samples with sputtered gold electrodes or CASING-treated samples with evaporated gold electrodes.<sup>6</sup>

The lower peak temperatures indicate a difference in the electrode properties of evaporated aluminum or the present gold coating as opposed to the gold coating achieved by sputtering or after CASING. While the former coatings probably represent blocking electrodes, the latter are injecting contacts.<sup>14</sup> Since the Teflon FEP hole mobility is much larger than electron mobility,<sup>13,15,16</sup> an injecting electrode will result in a faster isothermal charge decay or a lower TSC peak temperature due to hole injection.

The charge retention behavior and the TSC experiments show that the electrical properties of Teflon FEP gold coated by the present method are equal to those of aluminum-evaporated samples. Thus, charged Teflon FEP coated by application of the new process exhibits the most favorable electret properties presently achievable.

### SURFACE CHARACTERIZATION USING ESCA (Electron Spectroscopy for Chemical Analysis)

In an attempt to determine whether a change in surface composition of Teflon FEP was being induced by deposition and removal of aluminum, ATR infrared spectroscopy was employed initially. Examination of the polymer film before and after the deposition and removal of aluminum by ATR revealed essentially identical surfaces. Considering the fact that ATR samples a relatively thick surface layer along with the realization that changes induced by the aluminum deposition might reside in the uppermost region of the surface prompted an examination of similar samples with a truly surface sensitive technique.

X-Ray photoelectron spectroscopy<sup>17</sup> (XPS or ESCA) is the technique of measuring the kinetic energy spectra of core and valence electrons ejected from a surface when an essentially monoenergetic x-ray beam impinges on that surface. Energy conservation for the photoemission process allows one to calculate the binding energy of particular electrons *in the sample* using the relationship

$$E_b = E_{x\text{-ray}} - E_{\text{kin}} - \varphi_{\text{sp}}$$

where  $E_b$  is the binding energy of an electron with respect to the Fermi level in the sample,  $E_{x\text{-ray}}$  is the x-ray energy,  $E_{\text{kin}}$  is the kinetic energy of the photoelectron in space after it has entered the spectrometer, and  $\varphi_{\text{sp}}$  is the work function of the spectrometer, which remains constant. The values of  $E_b$  obtained not only serve to identify particular elements but are also a function of the chemical environment or oxidation state of a given element. The intensity of a binding energy peak is, of course, a measure of the concentration of a particular element in the surface region. The photoelectrons which give rise to ESCA signals are those which escape from the surface region without energy loss. Tracy has compiled data<sup>18</sup> which indicate that the mean free path or escape depth of such electrons in a wide range of mainly inorganic materials

is on the order of 4–30 Å depending upon the electron's kinetic energy. Thus, ESCA is, by nature, essentially a surface-sensitive technique.

ESCA spectra were obtained using a Varian VIEE-15 spectrometer system with  $MgK\alpha$  radiation (1253.6 eV) as the excitation source. The pressure in the spectrometer during measurements was  $\sim 10^{-7}$  torr, and the temperature was  $\sim 35^\circ C$ .

Sample preparation consisted of depositing a layer approximately 1000 Å thick of either vacuum-evaporated gold or aluminum onto one side of 25- $\mu m$ -thick Teflon FEP. (Metallization of only one side of a polymer sample allowed the remaining side to be used as a control.) The metal was then removed from the polymer by immersion in mercury (for gold) or 0.1*N* sodium hydroxide (for aluminum). The Teflon FEP films were examined in the spectrometer after being mounted on cylindrical holders with double-coated tape.

Sample charging<sup>19</sup> is a problem inherent to ESCA measurements on insulators. In cases where referenced binding energies were desired, gold wire<sup>20</sup> was secured around the sample and all peak positions were referenced to the gold( $4f_{7/2}$ ) signal which was assigned a binding energy of 84.0 eV.<sup>19</sup> Repetitive measurements indicated that a binding energy obtained in this manner was reproducible to only approximately  $\pm 0.3$  eV.

Fluorine(1s), oxygen(1s), and carbon(1s) electron spectra of various Teflon FEP samples are shown in Figure 6. Line 1 of this figure shows that untreated Teflon FEP gives rise to intense fluorine and carbon signals at  $\sim 689$  and  $\sim 292$  eV, respectively. The difference in the relative intensities of these sig-

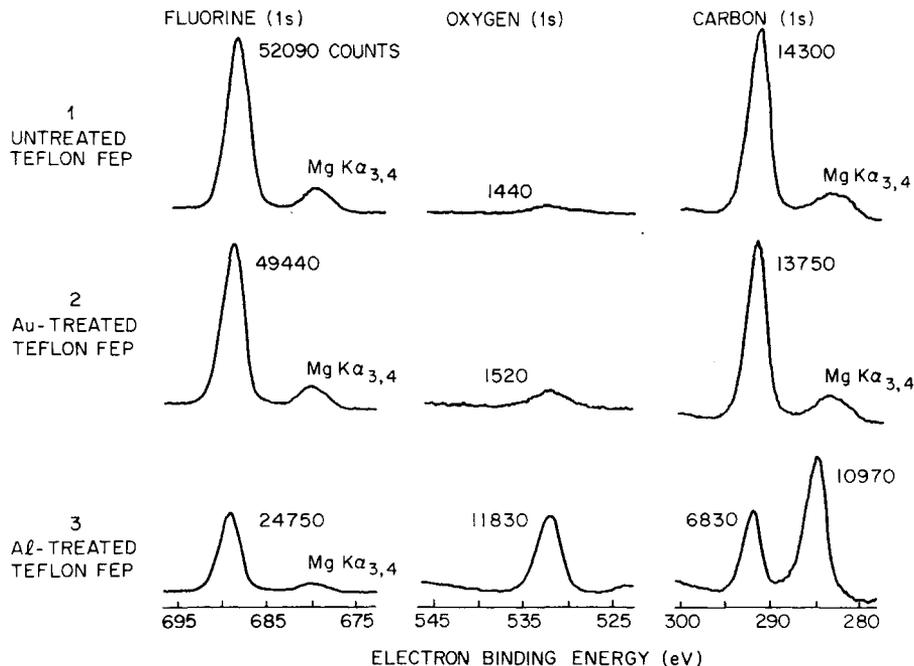


Fig. 6. Core level ESCA spectra of Teflon FEP before and after gold or aluminum treatment. The F(1s), O(1s), and C(1s) signals were accumulated in 30 10-sec, 100 10-sec, and 50 10-sec scans, respectively.

nals  $\{I[F(1s)]/I[C(1s)] = 3.64\}$  and those given for the same fluoropolymer  $\{I[F(1s)]/I[C(1s)] = 8.24\}$  in Figure 5, line 1 of reference 22 is probably due to different signal accumulation times. In the present study, the fluorine(1s) and carbon(1s) signals were accumulated in 30 10-sec and 50 10-sec scans, respectively. The relatively high carbon(1s) binding energy reflects the highly electronegative fluorine environment of carbon atoms in Teflon FEP. (Hydrocarbon materials typically give rise to carbon(1s) electron binding energies of about 285 eV.) The very weak oxygen signal of this sample probably originates from adsorbed oxygen-containing species.

The absence of gold(4f) and mercury(4f) signals in the electron spectrum of gold-treated Teflon FEP indicates that no gold or mercury was retained in the surface region of this sample. The core level spectrum (not shown) of the control side of this polymer film is essentially identical to that (Fig. 6, line 1) of the untreated polymer while the spectrum (Fig. 6, line 2) of the side on which gold had been deposited and removed is not significantly different from that of the untreated polymer. There appears, however, to be a slight reduction in the intensity of the fluorine signal and a slight enhancement in the intensity of the oxygen signal. Furthermore, a very weak carbon(1s) signal is now apparent at about 285 eV on the high-binding-energy side of the satellite arising from  $MgK\alpha_{3,4}$  radiation. Considering the essentially identical joint strengths ( $\sim 0$  kg/cm<sup>2</sup> shown in Table I) of untreated and gold-treated Teflon FEP, it is unclear at this point whether the subtle spectral changes of these two samples just described are a manifestation of changes brought about by the gold deposition and removal or not.

When one compares the electron spectrum of aluminum-treated Teflon FEP (Fig. 6, line 3) with untreated Teflon FEP (Fig. 6, line 1), significant differences are apparent. Both the fluorine(1s) and carbon(1s) signals of the aluminum-treated Teflon FEP at  $\sim 689$  and  $\sim 292$  eV have diminished in intensity by about 50% while oxygen(1s) and carbon(1s) photolines at  $\sim 532$  and  $\sim 285$  eV have appeared. The absence of aluminum(2p) and sodium(1s) signals in the electron spectrum of aluminum-treated Teflon FEP indicates that no aluminum or sodium (from the 0.1N NaOH solution) was retained in the surface region of this sample. As in the case of the gold-treated Teflon FEP, the core level spectrum of the control side of aluminum-treated Teflon FEP is essentially identical to that of the untreated polymer, thus eliminating the aluminum evaporation temperature and pressure conditions and immersion in NaOH solution as possible causes for the change in surface composition of the sample.

Based on this information, it is apparent that a significant amount of fluorine is being lost from the surface region of Teflon FEP during the aluminum treatment. The fate of the lost fluorine has been determined by ESCA examination of Teflon FEP substrates, onto which very thin layers ( $\sim 10$ ,  $\sim 30$ , and  $\sim 60$  Å) of aluminum have been deposited. The results of these experiments indicate that the aluminum vapor deposition process generates new fluorine-containing species in the aluminum/fluoropolymer interfacial region, which are subsequently removed during immersion of the composite in dilute aqueous sodium hydroxide. A subsequent paper<sup>21</sup> will describe these results in detail. It is reasonable to expect that this loss of fluorine might result in the introduction of unsaturation<sup>22</sup> (carbon-carbon double and/or triple

bonds) involving the remaining material. Indeed, carbon(1s) signal(s) from such a material could be expected to appear at a binding energy not significantly different from the  $\sim 285$ -eV carbon peak observed from the aluminum-treated Teflon FEP sample.

In order to test for unsaturation, an electron spectrum was taken of aluminum-treated Teflon FEP film which had been immersed in a bromine-in-carbon tetrachloride solution at room temperature overnight, rinsed in pure carbon tetrachloride, and finally allowed to dry. The exposure of a material possessing carbon-carbon double and/or triple bonds in the surface region to bromine should result in the formation of carbon-bromine species which can be detected using ESCA.<sup>22</sup> Scans of the bromine(3d) electron binding energy region revealed no photoelectron signal, thus eliminating unsaturation from further consideration as being responsible for the appearance of the intense carbon(1s) peak at about 285 eV in Figure 6, line 3. It therefore seems apparent that this peak and the intense oxygen(1s) peak are intimately related and suggests the existence of some oxygen-containing hydrocarbon species in the surface region of the aluminum-treated Teflon FEP. Since the enhanced joint strengths obtained with this material imply the absence of Teflon FEP at the surface, the oxygen-containing hydrocarbon material probably forms a thin and perhaps complete layer over the Teflon FEP. The fact that fluorine(1s) and carbon(1s) signals from underlying fluoropolymer are observed indicates that the oxygen-containing hydrocarbon layer has a thickness which is less than the escape depth of either the fluorine(1s) or carbon(1s) electrons which are ejected (using  $MgK\alpha$  radiation) with kinetic energies of  $\sim 560$  and  $\sim 960$  eV, respectively. In inorganic materials, electrons ejected with such kinetic energies can be expected to have escape depths on the order of 10–13 Å.<sup>18</sup> In contrast, recent findings<sup>23</sup> suggest that in polymers and other organic materials, comparable kinetic energy electrons may have escape depths as great as 50–100 Å.

In summary, x-ray photoelectron spectroscopic data show a difference in surface composition of Teflon FEP onto which aluminum has been vacuum deposited and then removed compared to untreated Teflon FEP or Teflon FEP onto which gold has been vacuum deposited and then removed. With the information available on this system, it therefore seems reasonable to attribute at least some of the enhancement of joint strength obtained with aluminum-treated Teflon FEP to the presence of a thin oxygen-containing hydrocarbon layer at the surface.

## CONCLUSIONS

The metal deposition method described in this paper allows one to bond gold and Teflon FEP with a joint strength approaching the bulk strength of the polymer without degradation of its charge storage properties. Films of this kind have the advantage that their metal coating is completely inert to environmental conditions. This makes such films useful for application in devices like electret transducers. X-Ray photoelectron (ESCA) spectra indicate significant chemical modification of the Teflon FEP, apparently consisting of the generation of a thin and perhaps continuous surface layer of oxygen-containing hydrocarbon material, as a result of the aluminum deposition

procedure. In contrast, scanning electron microscope (SEM) photographs indicate only a insignificant physical modification of the Teflon in terms of surface roughening as a result of the aluminum treatment. It therefore appears that the modified wetting characteristics and enhanced joint strengths obtained as a result of this procedure can be attributed in part at least to the existence of the chemically-modified Teflon FEP surface layer.

### References

1. A. A. Benderly, *J. Appl. Polym. Sci.*, **6**, 221 (1962).
2. D. M. Mattox and J. E. McDonald, *J. Appl. Phys.*, **34**, 2493 (1963).
3. H. Schonhorn and R. H. Hansen, *J. Appl. Polym. Sci.*, **11**, 1461 (1967).
4. H. Schonhorn, F. W. Ryan, and R. H. Hansen, *J. Adhesion*, **2**, 93 (1970).
5. G. M. Sessler, J. E. West, F. W. Ryan, and H. Schonhorn, *J. Appl. Polym. Sci.*, **17**, 3199 (1973).
6. W. A. Zisman, *Advan. Chem. Ser.*, **43**, 1 (1964).
7. J. J. Bikerman, *Physical Surfaces*, Academic Press, New York, 1973, pp. 265-266.
8. G. M. Sessler and J. E. West, *J. Electrostatics*, **1**, 111 (1975).
9. G. M. Sessler and J. E. West, *Appl. Phys. Lett.*, **17**, 507 (1970).
10. E. W. Anderson, L. L. Blyler, G. E. Johnson, and G. L. Link, in *Electrets, Charge Storage and Transport in Dielectrics*, M. M. Perlman, Ed., The Electrochem. Soc., Inc., Princeton, 1973, pp. 424-435.
11. J. Van Turnhout, *ibid.*, pp. 230-251.
12. G. M. Sessler and J. E. West, *Phys. Rev. B*, **10**, 4488 (1974).
13. R. A. Creswell, M. M. Perlman, and M. A. Kabayama, in *Dielectric Properties of Polymers*, F. E. Karasz, Ed., Plenum, New York, 1972, pp. 295-312.
14. M. A. Lampert and P. Mark, *Current Injection in Solids*, Academic Press, New York, 1970.
15. G. M. Sessler and J. E. West, *J. Appl. Phys.*, **43**, 922 (1972).
16. B. Gross, G. M. Sessler, and J. E. West, to be published.
17. K. Siegbahn, C. Nordling, A. Fahlman, R. Nordberg, K. Hamrin, J. Hedman, G. Johansson, T. Bergmark, S. E. Karlsson, I. Lindgren, and B. Lindberg, *ESCA: Atomic, Molecular and Solid State Structure Studied by means of Electron Spectroscopy*, Almquist and Wiksells, Uppsala, 1967.
18. J. C. Tracy, in *Electron Emission Spectroscopy*, W. Dekeyser, L. Fiermans, G. Vanderkelen, and J. Vennik, Eds., D. Reidel Publishing Co., Inc., Boston, 1973, p. 305.
19. W. P. Dianis and J. E. Lester, *Anal. Chem.*, **45**, 1416 (1973).
20. C. R. Ginnard and W. M. Riggs, *Anal. Chem.*, **44**, 1310 (1972).
21. R. F. Roberts and H. Schonhorn, *ACS Polym. Prepr.*, **16**(2), 146 (1975).
22. D. W. Dwight and W. M. Riggs, *J. Colloid Interfac. Sci.*, **47**, 650 (1974).
23. W. M. Riggs, Paper No. 215, 5th Northeast Regional Meeting, American Chemical Society, Rochester, New York, October 1973.

Received April 14, 1975

Revised May 27, 1975