Lamination of Polytetrafluoroethylene Films via Surface Thermal Graft Copolymerization with Ionic and Zwitterionic Monomers

Y. X. LIU,[1] E. T. KANG,[1] K. G. NEOH,[1] K. L. TAN,[2] C. C. HUANG,[3] D. J. LIAW[3]

[1] Department of Chemical Engineering, National University of Singapore, Kent Ridge, Singapore 119260

[2] Department of Physics, National University of Singapore, Kent Ridge, Singapore 119260

[3] Department of Chemical Engineering, National Taiwan University of Science and Technology, Taipei, Taiwan 106, Republic of China

ABSTRACT: Surface thermal graft copolymerization with concurrent lamination was carried out between two argon plasma-pretreated polytetrafluoroethylene (PTFE) films in the presence of aqueous zwitterionic solutions of *N,N*-dimethyl-*N*-methacrylamido-propyl-*N*-(3-sulfopropyl)ammonium betain (DMASAB), *N,N*-dimethyl(methacryloyl-ethyl)ammonium propansulfonate (DMAPS), and 1-(3-sulfopropyl)-2-vinylpyridinium betaine (SVPB), as well as an aqueous ionic solution of potassium-2-sulfopropylacrylate (SPA) and potassium-2-sulfopropyl methacrylate (SPM), under atmospheric conditions and in the complete absence of an added initiator and system degassing. The lap shear adhesion strength between the PTFE films from simultaneous grafting and lamination depended on the argon plasma pretreatment time of PTFE films, the thermal lamination temperature, the concentration of the monomer solution, and the ionic nature of the grafted chains. Lap shear adhesion strength greater than 120 N/cm^2 and exceeding the yield strength of the PTFE substrate used could be readily obtained in most PTFE/zwitterion/PTFE assemblies after simultaneous thermal graft copolymerization and lamination. The chemical compositions of the graft-copolymerized surfaces were studied by X-ray photoelectron spectroscopy (XPS). © 1999 John Wiley & Sons, Inc. J Appl Polym Sci 74: 816–824, 1999

Key words: zwitterion; graft copolymerization; lamination; PTFE; adhesion strength; XPS

INTRODUCTION

Surface modification of polytetrafluoroethylene (PTFE) and other fluoropolymers has been of particular interest, as these materials belong to one of the most important families of engineering polymers well known for their high thermal stability, excellent chemical inertness, low friction coefficient, and low dielectric constant.[1,2] However, the low surface energy of these polymers makes it very difficult to bond their surfaces with other substrates, such as adhesives and paints, which generally consist of polar groups or components. This surface inertness thus makes fluoropolymers unsuitable for use in areas involving adhesive bonding and coating.

A variety of different treatments have been employed to activate PTFE surfaces for adhesion

improvement. The most widely used method for improving the bondability of fluoropolymers is treatment with a reducing etchant,[3–5] such as sodium naphthalide, which defluorinates the surface typically to a depth of about 100 nm. Although chemical etching is effective, it also presents considerable disadvantages.[6,7] Plasma treatment has also been used extensively in the surface modification of fluoropolymers. It has been reported that the hydrophilicity of the PTFE film can be considerably enhanced by surface plasma treatment and further improved by graft copolymerization.[8–15]

In a parallel development, Rånby et al.[16–18] showed that initiator-promoted surface photograft copolymerization, when carried out at the interface between two contact polymer films, was accompanied by the simultaneous lamination of polymer films. The lamination phenomenon arises from the entanglement and interaction, at the lapped interface, of the grafted chains from the two polymer film surfaces. More recently, we also demonstrated that simultaneous photografting and lamination of polymer films can be achieved in the complete absence of an added polymerization initiator when the polymer substrates are pretreated or preactivated to generate the peroxide or hydroxyl peroxide species.[19] We further demonstrated that the simultaneous graft-copolymerization and lamination process can be carried out thermally, with the same efficiency, to result in strong adhesion between two polymer films[20] or between a polymer film and a metal.[21]

In the present work, we demonstrate that improved adhesion between the PTFE films can be obtained through a simple, one-step simultaneous surface graft-copolymerization and lamination process. The reactive peroxides and hydroxyl peroxides for the initiation of surface graft copolymerization were generated by an Ar plasma pretreatment. The effects of the plasma pretreatment time, lamination temperature, and concentration of monomer solutions on the lap shear adhesion strength between the two PTFE films were studied. The surface compositions of the graft-copolymerized surfaces were studied by XPS. The monomers used for graft copolymerization include *N,N*-dimethyl-*N*-methacryl-amiopropyl-*N*-(3-sulfopropyl)ammonium betain (DMASAB), *N,N*-dimethyl(methacryloylethyl)ammonium propansulfonate (DMAPS), 1-(3-sulfopropyl)-2-vinylpyridinium betaine (SVPB), potassium-2-sulfopropylacrylate (SPA), and potassium–2-sulfopropyl methacrylate (SPM).

EXPERIMENTAL

Materials

PTFE films having a thickness of about 0.01 cm and a density of 2.18 g/cm^2 were purchased from Goodfellow Inc. (Cambridge, UK). The surfaces of the film were cleaned by extraction with acetone and methanol before use.

The water-soluble ionic monomers used for graft copolymerization, that is, DMASAB, DMAPS, SVPB, SPA, and SPM, were prepared according to the methods reported earlier.[22,23] They have the following chemical structures:

DMASAB:
$CH_2\!\!=\!\!C$ (CH_3)—$CONH$—$(CH_2)_3$—$N^+(CH_3)_2$—$(CH_2)_3$—SO_3^-

DMAPS:
$CH_2\!\!=\!\!C$ (CH_3)—COO—$(CH_2)_2$—$N^+(CH_3)_2$—$(CH_2)_3$—SO_3^-

SVPB:
$CH_2\!\!=\!\!CH$—$C_5H_5N^+$—$(CH_2)_3$—SO_3^-

SPA:
$CH_2\!\!=\!\!CH$—COO—$(CH_2)_3$—SO_3K

SPM:
$CH_2\!\!=\!\!C(CH_3)$—COO—$(CH_2)_3$—SO_3K

The solvents and other reagents were of analytical grade and were used without further purification.

Plasma Pretreatment and Thermal Graft Copolymerization

PTFE film strips of about 2.0 × 5.0 cm were pretreated with Ar plasma before thermal graft copolymerization. Ar plasma pretreatment was carried out in an Anatech SP 100 plasma system, equipped with a cylindrical quartz reaction chamber. The glow discharge was produced at an Ar pressure of 0.45 Torr. In the simultaneous thermal grafting and lamination experiment, two plasma-pretreated PTFE strips were lapped together in the presence of a small quantity of an aqueous monomer solution of a predetermined concentration. The assembly was then pressed and sandwiched between two glass plates using mechanical clips before being subjected to thermal treatment. The assembly was kept at a predetermined temperature for a predetermined period of time. Two lapping areas were used: 0.1 × 0.5 cm and 1.0 × 1.0 cm. The simultaneous graft copolymerization and lamination process is shown schematically in Figure 1. The thermally

Figure 1 Schematic representation of the simultaneous thermal graft-copolymerization and lamination process.

laminated films with a larger lapped area were separated by immersing the lapped films in doubly distilled water. The self-delaminated films were subjected to repeated rinsing and soaking in a vigorously stirred hot water (60°C) bath for at least 24 h to remove unreacted monomers and the adsorbed residual homopolymer. These self-delaminated films were used for surface graft composition measurements by XPS.

Surface Characterization

The surface-modified polymer films were characterized by X-ray photoelectron spectroscopy (XPS). XPS measurements were made on a VG ESCALAB MKII spectrometer with a MgKα X-ray source (1253.6 eV photos) at a constant retard ratio of 40. The polymer films were mounted on the standard sample studs using double-sided adhesive tape. The core-level signals were obtained at a photoelectron take-off angle (α, with respect to sample surface) of 75°. The X-ray source was run at a reduced power of 120 W (12 kV and 10 mA). All binding energies (BEs) were referenced to the C1s neutral carbon peak at 284.6 eV. In the peak synthesis, the line width [full width at half-maximum (fwhm)] for the Gaussian peaks was maintained constant for all components in a particular spectrum. Surface elemental stoichiometries were determined from peak-area ratios, after correcting with the experimental determined sensitivity factors, and were accurate to within ±5%. The elemental sensitivity factors were determined using stable binary compounds of well-established stoichiometries.

Adhesion Strength Measurements

The lamination strengths as a function of the various experimental parameters were evaluated by measuring the lap shear tensile strength with an Instron 5544 tensile tester from Instron Corp. All measurements were carried out at a crosshead speed of 0.1 cm/min. For each lap shear strength reported, at least three measurements were averaged.

RESULTS AND DISCUSSION

Surface Characterization

Argon plasma pretreatment followed by atmospheric exposure causes a gradual increase in the surface concentration of the oxygen species. The changes in the O/C and F/C atomic ratios of the PTFE film, as determined from the C1s, O1s, and F1s core-level spectral peak area ratios obtained at the photoelectron take-off angle (α) of 75°, as a function of the Ar plasma treatment time, are shown in Figure 2. An increase in the O/C ratio and a decrease in the F/C ratio were observed upon increasing the Ar plasma treatment time, in agreement with the results generally reported in the literature.[24,25] It can also be clearly seen that the O/C ratios approach an asymptotic value at a plasma treatment time longer than 60 s, under the glow-discharge conditions used in the present work. This observation is in agreement with the previous report that prolonged plasma and corona treatment of polyethylene or polypropylene sur-

Figure 2 O/C and F/C ratios of the PTFE films surface as a function of plasma pretreatment time.

faces does not result in the introduction of a larger amount of oxygen atoms or peroxides.[26,27] The Ar plasma causes breakage of C—F bonds, resulting in the abstraction of fluorine atoms from the surface. However, for the plasma energy and frequency used in the present work, the decrease in the F/C ratio is significantly less than that reported in our earlier work in which high-energy and high-frequency plasma was used.[19] In the present case, the surface F/C ratio decreased only from about 2.0 to 1.75 after 120 s of plasma exposure.

Figure 3(a,b) shows, respectively, the C1s and wide-scan spectra, obtained at a photoelectron take-off angle(α) of 75° of the pristine PTFE film used in the present work. The C1s core-level spectrum consists of a main component with a binding energy of 291.4 eV, attributable to the $\underline{C}F_2$ species,[28] and a broad minor component ($\approx$10% of the main component area) at about 8 eV below the main peak. The minor component can be attributed mainly to the contribution of the X-ray satellite peaks of the CF_2 species, arising from the MgK$\alpha_{3,4}$ radiation. The wide-scan spectrum demonstrated that there exist only two elements, carbon and fluorine, which is consistent with the molecular structure of PTFE: —$[CF_2—CF_2]_n$—.

As an example, Figure 3(c,d) shows the corresponding C1s and wide-scan spectra of a PTFE film after 60 s of Ar plasma pretreatment, followed by 5 h of the simultaneous thermal graft copolymerization and lamination with 20 wt %

Figure 3 (a) C1s core-level and (b) wide-scan spectra of a pristine PTFE film; (c) C1s core-level and (d) wide-scan spectra of a 40-s Ar plasma-pretreated PTFE film after the simultaneous thermal copolymerization and lamination with 20 wt % aqueous DMASAB solution at 85°C for 5 h.

aqueous DMASAB solution. The presence of surface-grafted DMASAB polymer can be deduced from the appearance of a prominent C1s component at the BE of about 284.6 eV, associated with the neutral carbon species of the DMASAB polymer and the diminished CF_2 component intensity. The presence of the surface-grafted DMASAB polymer is further confirmed by the appearance of the S2p and N1s signals in the wide-scan spectrum of Figure 3(d). The covalently bonded sulfonate species ($—SO_3^-$) of DMASAB has a characteristic S2$p_{3/2}$ BE at about 168 eV.[28] The N1s peak at the BE of about 402 eV is attributed to the positively charged nitrogen group of DMASAB.[29] The surface characteristics of the PTFE films graft-copolymerized with other monomers under similar conditions as that of the DMASAB monomer were also studied by XPS. The appearance of the major C1s component at about 284.6 eV together with the N1s and/or S2p core-level signals all suggested that these monomers have been graft-copolymerized onto the surfaces of PTFE films.

The concentration of surface-grafted polymer from the present thermal graft copolymerization and lamination process can be determined directly from the relative intensities of the two C1s components, taking into account the carbon stoichiometries of the graft and the substrate chains and the contribution of the MgK$\alpha_{3,4}$ satellite structure of the CF_2 peak component to the intensity of the peak component at 284.6 eV. Thus, the concentration of the surface-grafted polymer, defined as the number of repeating units of the graft chain per repeating unit of the PTFE chain, can be calculated from the following expression:

Graft concentration

$$= 2 \times \frac{[(\text{C1}s \text{ area of } 284.6 \text{ eV peak}) - 0.1 \times (\text{C1}s \text{ area of } 291.4 \text{ eV peak})]/n}{\text{C1}s \text{ area of } 291.4 \text{ eV peak}}$$

The factors 2 and n are introduced to account, respectively, for the fact that there are two CF_2 units per repeating unit of PTFE and there are n carbon atoms per repeating unit of the grafted polymer. Alternatively, the surface concentration of the grafted polymers can also be determined from the [S]/[F] or [N]/[F] ratio, based on the sensitivity factor-corrected S2p, N1s, and F1s spectral area ratios. The graft concentrations determined from the carbon and heteroatom stoichiometries agree to within 10%.

Figure 4 Lap shear adhesion strength and graft yield of the DMASAB polymer as a function of plasma pretreatment time of the PTFE films. [DMASAB] = 20 wt %; lamination time = 5 h; lamination temperature = 85°C.

Effect of Plasma Pretreatment Time

Thermal lamination arising from surface graft copolymerization can be achieved when a small quantity of aqueous solution of the monomer, without degassing and in the absence of an added initiator, is sandwiched between two plasma-pretreated PTFE films, followed by thermal curing. The effect of the Ar plasma pretreatment time of the PTFE films on the concentration of surface-grafted DMASAB, SVPB, SPA, SPM, and DMAPS polymers on the PTFE films and the corresponding lap shear adhesion strength are shown in Figures 4–8, respectively. The simultaneous lamination and graft copolymerization were carried out in the presence of 20 wt % of the respective monomer solution at 85°C for 5 h. In each case, the concentration of the surface-grafted polymer increases initially with increasing plasma pretreatment time, but exhibits a decrease at a plasma pretreatment time greater than 60 s under the glow-discharge conditions used in the present study. This phenomenon is consistent with the fact that the surface O/C ratio of the PTFE film reaches an asymptotic value at 60 s of plasma treatment time, as shown in Figure 2. Prolonged plasma treatment will cause further oxidation of the peroxide carbon species and also give rise to etching of the PTFE surface, resulting in a decrease in the overall content of peroxide or hydroxyl peroxide species at the surface. In com-

Figure 5 Lap shear adhesion strength and graft yield of the SVPB polymer as a function of plasma pretreatment time of the PTFE films. [SVPB] = 20 wt %; lamination time = 5 h; lamination temperature = 85°C.

Figure 7 Lap shear adhesion strength and graft yield of the DMAPS polymer as a function of plasma pretreatment time of the PTFE films. [DMAPS] = 20 wt %; lamination time = 5 h; lamination temperature = 85°C.

parison with the previous work,[13] which involves graft copolymerization by UV illumination after the removal of oxygen in the solution, the overall graft yield via the present simultaneous thermal graft copolymerization and lamination process is lower. This phenomenon probably resulted from the fact that no system degassing had been carried out prior to thermal polymerization. In the

absence of degassing, the trace amount of dissolved oxygen, which is deleterious to radical polymerization, must have caused a significant decrease in the graft yield or graft chain length.

As clearly shown in Figures 4–8, the dependence of the lap shear adhesion strength on the plasma pretreatment time for each system coincides approximately with the dependence of the

Figure 6 Lap shear adhesion strength and graft yield of the SPM polymer as a function of plasma pretreatment time of the PTFE films. [SPM] = 20 wt %; lamination time = 5 h; lamination temperature = 85°C.

Figure 8 Lap shear adhesion strength and graft yield of the SPA polymer as a function of plasma pretreatment time of the PTFE films. [SPA] = 20 wt %; lamination time = 5 h; lamination temperature = 85°C.

graft yield on the plasma pretreatment time. This result, in turn, is consistent with the dependence of adhesion strength on diffusion and entanglement of the graft chains. Despite the low graft yields for the DMASAB, DMAPS, and SVPB polymers, the observed strong adhesions suggest that the amphoteric interactions of the zwitterionic polymer must have also played an important role in addition to the chain entanglement.

The adhesion data in Figures 4–8 also suggest that the maximum attainable lap shear adhesion strength appears to differ substantially for each monomer used. When the amphoteric DMASAB, DMAPS, or SVPB monomer is used for graft copolymerization, the maximum lap shear adhesion strength can reach about 120 N/cm^2, which also exceeds the yield strength of the 0.1-mm-thick PTFE substrate used in the present study. Substrate yielding in this case occurs when the shear strength experienced by the 0.1×0.5 cm lapped junction exceeds about 100 N/cm^2. When the ionic SPA or SPM monomer is used, the maximum lap shear strength achieved is only in the order of 80 N/cm^2 under the present lamination conditions. This maximum attainable lap shear adhesion strength is much smaller than that achievable in the presence of the amphoteric monomers. This phenomenon may be related to the ionic and molecular structures of the graft chains and will be discussed again below.

Effect of Lamination Temperature

The influence of lamination temperature on the lap shear adhesion strength is shown in Figure 9. Increase in the lamination temperature has a positive effect on adhesion strength over a wide temperature range, but exerts a negative effect at temperatures above 120°C. Generally, it is necessary to elevate the lamination temperature for adhesion improvement through increase in the surface graft-copolymerization efficiency. In addition to the thermal decomposition of surface peroxides, the graft-copolymerization process can be represented by the following three relevant reactions:[17]

$$P \cdot \; + M \rightarrow PM \cdot \quad \textbf{(I)}$$

$$P \sim \, \cdot \; + M \rightarrow \text{ grafted polymer} \cdot \quad \textbf{(II)}$$

$$\sim \cdot \; + M \rightarrow \text{ homopolymer} \cdot \quad \textbf{(III)}$$

where P and M denote polymer and monomer, respectively. Of the initiation reaction and the

Figure 9 Lap shear adhesion strength as a function of lamination temperature. Monomer concentration = 20 wt %; lamination time = 3 h; plasma pretreatment time = 40 s.

two propagation reactions, species (**I**) and (**II**) both have a much lower reactivity than that of species (**III**), because (**I**) is a surface free radical located on the substrate surface and (**II**) is a macromolecular free radical tethered on the polymer substrate. Their mobilities and vibrational frequencies are much lower than those of the homopolymer chain free radical. As a consequence, species (**I**) and (**II**) are expected to show a greater sensitivity to the reaction temperature, as they have a higher activation energy than that of reaction (**III**). Therefore, elevating the reaction temperature not only favors the peroxide decomposition, but is also beneficial to the surface polymerization, resulting in enhanced adhesion between the polymer films. At an even higher temperature, the reduction in the lap shear adhesion strength can be due to thermal degradation of the functional groups introduced by the plasma pretreatment and the grafted polymer owing to their low thermal stability, as well as to the surface migration of the mobile PTFE segments.[15] Alternatively, the water molecules of the monomer solution at the lapped interface will evaporate more rapidly at elevated temperature, resulting in a reduction in both the extent of polymerization and the extent of chain diffusion and entanglement.

Figure 10 Lap shear adhesion strength as a function of monomer concentration. Plasma pretreatment time = 20 s; lamination temperature = 85°C; lamination time = 3 h.

Effect of Monomer Concentration

The monomers studied in the present work dissolve readily in water. The phenomenon is attributable to their ionic structures and offer a very simple and convenient means for carrying out the simultaneous grafting and lamination process between two polymer surfaces. The concentration of monomers used for graft copolymerization also plays a predominant role on the lap shear adhesion strength. Figure 10 shows the effect of the concentration of the various monomer solutions on the lap shear adhesion strength for the 20-s Ar plasma-pretreated PTFE films after 3 h of grafting and lamination at 85°C. It can be seen that for most monomers there is an optimum concentration which corresponds to the highest lap shear adhesion strength. Furthermore, the optimal concentrations for most monomers are near 20 wt %. At higher concentration, increase in the solution viscosity probably affects the diffusion of the monomer molecules and thus retards the radical-induced graft polymerization, which is diffusion-controlled. At the same time, the increasing presence of residual or unreacted monomer will also form a weak boundary layer, resulting in a decrease in the lap shear adhesion strength. At the other extreme, when a low concentration of the monomer is used for graft polymerization, the concentration of the grafted polymer and the mo-

lecular weight of the grafted polymers will decrease, which will inevitably affect the adhesion between the lapped films.

The lap shear adhesion strength recorded in the present study is the force per square centimeter of the grafted surface required to separate the two adhered films from the equilibrated distance to infinite separation. In an ideal case, this force should be related to the entanglement and diffusion of the grafted polymers, as well as to the electrostatic force between them. Among the monomers used in the present study, the lap shear adhesion strength achieved in the presence of the DMASAB, DMAPS, or SVPB monomer is higher than that in the presence of the SPA or SPM monomer. In the case of the DMASAB, DMAPS, or SVPB graft-copolymerized interface, the adhesion strength is further enhanced by the interchain and intrachain electrostatic interactions arising from the amphoteric nature (i.e., containing both cationic $-CN^+$ and anionic $-SO_3^-$ species) of the grafted chains. Furthermore, the electrostatic interactions are less dependent on, and probably at the same time improve, the diffusion and entanglement among the grafted chains, resulting in respectable lap shear adhesion strengths. The films laminated from graft copolymerization with the SPA or SPM monomer have only anionic–anionic repulsive interaction at the lapped interface, which will also give rise to a decrease in the extent of diffusion and entanglement among the grafted chains. As a result, lower lap shear adhesion strengths were observed when a shear force was applied to the SPA and SPM graft-copolymerized interfaces. In the cases of the amphoteric monomers, the lap shear adhesion strength of films graft copolymerized with SVPB is slightly lower than those graft copolymerized with DMAPS or DMASAB. The phenomenon may be attributed to the presence of the rigid phenyl group of the SVPB polymer which can affect the extent of electrostatic molecular interaction spatially.

As is well known, ionic interaction in aqueous media is greatly influenced by salt ions. The remarkable reduction in the shear strength observed when monomers of SPA or SPM were used for graft copolymerization may be due to the presence of potassium ion in their molecules. The potassium cation moves more efficiently than does the anionic moiety and this may markedly reduce the Coulombic interaction between the polymer chains. The result is also consistent with the earlier work of Zhang et al.,[30] which showed

that the shear strength between an anionic monomer and a cationic monomer–graft copolymerized polyester films decreased significantly with increasing KCl concentration at the lapped interface.

CONCLUSIONS

Effective thermal graft copolymerization with concurrent lamination between two PTFE films can be achieved in the presence of a variety of zwitterionic and ionic monomers under atmospheric conditions and in the complete absence of an added initiator and system degassing, provided that the polymer substrates are pretreated with argon plasma. It was demonstrated that the plasma pretreatment time, the thermal lamination temperature, the concentration of the monomer solution, and the chemical structure of monomers used greatly influence the resulting lap shear adhesion strength and graft yield. The electrostatic force between the grafted polymer, in addition to chain entanglement, played an important role on the adhesion strength. Lap shear adhesion strengths obtained by using such zwitterionic monomers as DMASAB, DMAPS, and SVPB are higher than those obtained using the anionic monomers, such as SPA and SPM, and readily exceed the yield strength of the thin PTFE film used. The composition of the graft-copolymerized PTFE surfaces were characterized by XPS.

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