

Thermal graft copolymerization with concurrent lamination of low density polyethylene and poly(tetrafluoroethylene) films

M.C. Zhang^a, E.T. Kang^{a,*}, K.G. Neoh^a, H.S. Han^b, K.L. Tan^b

^aDepartment of Chemical Engineering, National University of Singapore, Kent Ridge, Singapore 119260

^bDepartment of Physics, National University of Singapore, Kent Ridge, Singapore 119260

Received 7 April 1998; accepted 20 May 1998

Abstract

A simple technique of thermal graft copolymerization of glycidyl methacrylate (GMA) on argon plasma pretreated poly(tetrafluoroethylene) (PTFE) films or low density polyethylene (LDPE) films with simultaneous lamination of the two films was demonstrated. The effect of crosslinking agent on the adhesion strength of the PTFE/PTFE interface was also investigated. The simultaneous thermal grafting and lamination process was carried out under atmospheric conditions and in the complete absence of a polymerization initiator. The surface structure and composition of the graft copolymerized PTFE and LDPE films from either self-delamination in acetone or mechanical delamination were studied by X-ray photoelectron spectroscopy (XPS). The efficiency of surface graft copolymerization and lamination was found to be strongly dependent on the plasma pretreatment time of the substrate, graft concentration, thermal grafting/lamination temperature and time. T-peel adhesion strengths of about 0.9 and 1.7 N cm⁻¹ were achieved for the PTFE/GMA/PTFE assemblies in the absence and presence, respectively, of the crosslinking agent, triallyl-1,3,5-benzenetricarboxylate (TBTC). The T-peel adhesion strength of the LDPE/GMA/LDPE assembly, on the other hand, can reach about 2.6 N cm⁻¹ in the absence of TBTC. XPS analysis of the mechanically delaminated surfaces suggests that the failure mode of the PTFE/GMA/PTFE assembly and the PTFE/GMA + 2 vol% TBTC/PTFE assembly is adhesional in nature, and that of the LDPE/GMA/LDPE assembly is cohesive. © 1998 Elsevier Science Ltd. All rights reserved.

Keywords: Thermal graft copolymerization; Lamination; Low-density polyethylene

1. Introduction

Polymers with desirable bulk properties have found extensive applications. In most engineering applications, the surface property of a selected polymer is also an important factor to be considered. However, the surface properties of most engineering polymers in use today are less than optimum for many applications. To improve the surface property, different physicochemical methods have been employed, e.g. treatment with high-energy irradiation, ozone, corona discharge and plasma. Another way to alter the surface properties of polymers is to graft copolymerize with specific monomers. New functionalities, such as hydrophilicity [1], biocompatibility [2], adhesion [3] and lubricative properties [4], can thus be incorporated. In the last few years, a considerable amount of work has been done in this area. For example, Rånby et al. [5,6] developed an approach for surface photografting of polymers. In their process, the polymer substrates were either UV-irradiated in a mixture of

photoinitiator and monomer vapour, or after presoaking on-line in a solution of the monomer and initiator [7,8]. The effect of photografting on adhesion was also investigated for low density polyethylene plate surface graft copolymerized with various monomers [6]. Other researchers have also successfully incorporated different functional groups onto polymer surfaces via graft copolymerization [9–13].

More recently, a novel surface photografting with concurrent lamination method has been reported [14–16]. Strong adhesion between two polymer films could be achieved simultaneously during the photografting process when a small quantity of monomer solution containing a photoinitiator was introduced between two contacting films [17]. We also demonstrated that simultaneous photografting and photolamination could be achieved under atmospheric conditions and in the complete absence of an added photoinitiator in the monomer when the polymer substrates were pretreated with ozone [18,19].

In the present work, we further demonstrate that the simultaneous grafting and lamination process can be carried out thermally with the same efficiency. Thus, simultaneous

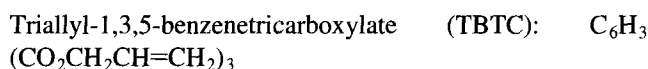
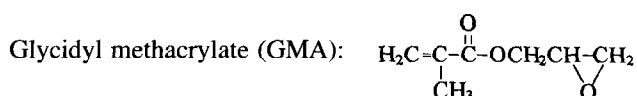
* Corresponding author.

thermal grafting and lamination under atmospheric conditions and in the complete absence of an added polymerization initiator has been carried out between two poly(tetrafluoroethylene) substrates, as well as between two low density polyethylene substrates.

2. Experimental

2.1. Materials

Poly(tetrafluoroethylene) (PTFE) and low density polyethylene (LDPE) films of thickness 0.1 and 0.125 mm, respectively, were used in the present study and were purchased from Goodfellow (Cambridge, UK). The surface of the film was cleaned with acetone and doubly distilled water in an ultrasonic water bath before used. Glycidyl methacrylate (GMA) of 97% purity and triallyl-1,3,5-benzenetricarboxylate (TBTC) of 98% purity were purchased from Aldrich (Milwaukee, USA) and was used as received. The chemical structures of GMA and TBTC are shown as follows:



2.2. Surface pretreatment

The PTFE and LDPE film strips of about 3 mm × 10 mm were pretreated with argon plasma before the thermal graft copolymerization and lamination experiment. The Ar plasma pretreatment was carried out in an Anatech SP100 plasma system, equipped with a cylindrical quartz reactor chamber. The glow discharge was produced at a plasma power of 35 W, an applied oscillator frequency of 40 kHz and an argon pressure of 0.6 torr. The plasma pretreated film strips were subsequently exposed to the atmosphere for about 5 min before the thermal grafting and lamination experiment.

2.3. Thermal graft copolymerization with concurrent lamination

In the simultaneous graft copolymerization and lamination experiment, a small quantity of pure GMA monomer or GMA monomer containing 2 vol% of TBTC was introduced between two identical Ar plasma pretreated PTFE films. For the grafting and lamination of the LDPE films, only pure GMA monomer was used. The crosslinking agent was not used in this case as TBTC is effectively activated only at temperatures above 100°C, which is higher than the maximum grafting and lamination temperature used for the LDPE films. Each assembly was sandwiched between two

glass plates by means of two mechanical clips before being transferred into a constant temperature oven for a pre-determined period of time. After the thermal grafting and lamination experiment, the samples were returned to room temperature by annealing in the well-insulated oven over a period of no less than 6 h. Since the so-prepared assemblies did not readily self-delamination in solvents, an alternative method was applied to produce samples for the determination of surface graft concentration. Thus, one of the films in the assembly was replaced by an inert pristine film. The interface of the assembly after the graft copolymerization and lamination experiment readily underwent self-delamination in acetone.

2.4. Adhesion strength measurement

The adhesion strengths were determined by measuring the T-peel adhesion strengths on an Instron 5540 tensile strength tester from Instron Corp. of USA. All measurements were performed at a crosshead speed of 0.5 cm min⁻¹. Each adhesion strength reported was the average of at least three sample measurements. The lapping area was kept at about 3 mm × 10 mm for all T-peel strength measurement.

2.5. Surface characterization

The films from self-delaminated assemblies were cleaned with copious amounts of acetone and deionized water to remove the surface adsorbed homopolymer and unreacted monomer before the X-ray photoelectron spectroscopy (XPS) measurements. XPS measurements were made on a VG ESCALAB MKII spectrometer with an Mg K_α X-ray source (1253.6 eV photon) at a constant retard ratio of 40. The LDPE or PTFE films were mounted on standard sample studs by means of double-side adhesive tape. The X-ray source was operated at a reduced power of 120 W (12 kV and 10 mA). The pressure in the analysis chamber was maintained at 7.5 × 10⁻⁹ torr or lower during the measurements. The core-level spectra obtained at the photoelectron take-off angle of 75° were used for analysis. All binding energies (BEs) were referenced to the C 1s neutral carbon peak at 284.6 eV, so as to compensate for the effect of surface charging. In 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 of the sample. Surface compositions were determined from peak-area ratios, corrected with the experimentally determined sensitivity factors, and were reliable to ±5%. The elemental sensitivity factors were determined using stable binary compounds of well-established stoichiometries. The surface graft concentration of each polymer was expressed as the number of repeating units of the grafted chain per substrate carbon atom and was determined from the

XPS-derived surface stoichiometries:

[Graft concentration]

$$= \frac{\text{Number of repeating units from graft chains}}{\text{Total number of substrate carbon atoms}}$$

3. Results and discussion

3.1. Graft copolymerization of PTFE films with concurrent lamination

The dependence of the [O]/[C] and [F]/[C] atomic ratios, as determined from the C 1s, O 1s and F 1s core-level spectra peak–area ratios at the photoelectron take-off angle of 75°, on the Ar plasma treatment time for the PTFE film is shown in Fig. 1. An increase in the [O]/[C] ratio and a monotonous 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 [20]. However, after 60 s of Ar plasma treatment, the [O]/[C] ratio approaches an asymptotic value. The Ar plasma treatment can cause the breakage of some C–F bonds in the surface layer of the PTFE, resulting in the defluorination of the film surface. The subsequent exposure of the activated surface to air causes oxygen to be incorporated on the PTFE surface, leading to surface oxidation and the formation of peroxide and hydroxyl peroxide species [1].

Fig. 2a–c show the respective C 1s core-level spectra of a pristine PTFE film, a graft-modified PTFE film from the self-delaminated PTFE/GMA/PTFE assembly and a graft-modified PTFE film from the self-delaminated PTFE/GMA + 2 vol% TBTC/PTFE assembly. Both assemblies were prepared under the following conditions: Ar plasma treatment time of 60 s for one of the two PTFE films, thermal grafting/lamination at 180°C for 4 h. The C 1s spectrum of pristine PTFE film consists of a main component at the binding energy of 291.4 eV, attributable to the CF_2 species,

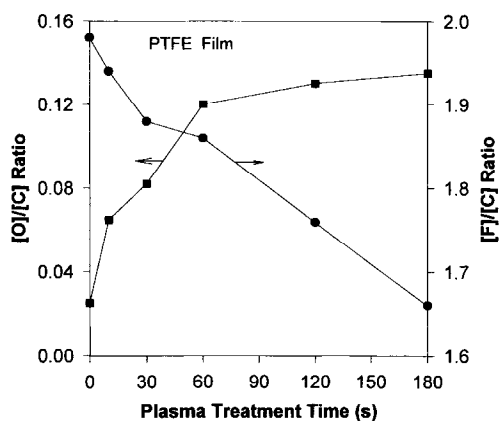


Fig. 1. Effect of Ar plasma treatment time on the [O]/[C] and [F]/[C] atomic ratios of the PTFE film.

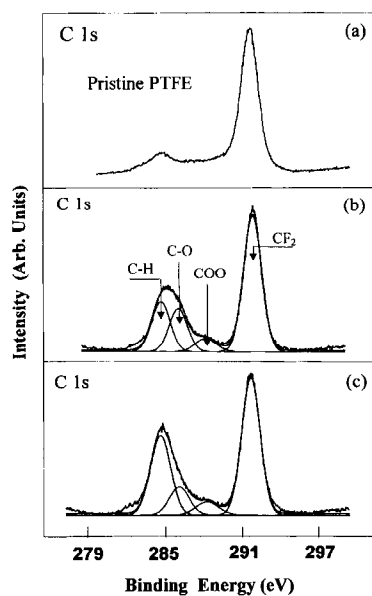


Fig. 2. C 1s core-level spectra for (a) a pristine PTFE film, (b) a PTFE film from a self-delaminated PTFE/GMA/PTFE assembly and (c) a PTFE film from mechanically delaminated PTFE/GMA + 2 vol% TBTC/PTFE assembly. Both assemblies were prepared with plasma pretreatment time of 180 s for the PTFE film followed by thermal grafting/lamination at 180°C for 2 h.

and a broad minor component ($\sim 10\%$ 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 structures of the CF_2 species arising from the Mg $K_{\alpha 3,4}$ radiation. From Fig. 2b, it can be seen that the low BE component in the C 1s core-level spectrum is significantly enhanced after surface graft copolymerization. In comparison with the C 1s core-level spectrum of the GMA homopolymer [18], this low BE component can be attributed to the presence of surface grafted GMA polymer. Three C 1s components with BEs similar to those of the GMA homopolymer can be deduced, 284.6 eV for the CH species, ~ 286.2 eV for the CO species and ~ 288.5 eV for the COO species. The relative peak-area ratio for the three species is about 3.2:3.1:1.0, in good agreement with the theoretical ratio of 3.0:3.0:1.0 based on the chemical structure of GMA. For simplicity, the contribution of the X-ray satellite structures of the CF_2 species to this portion of the C 1s spectrum has been neglected in the peak synthesis. In the presence of the crosslinking agent during graft copolymerization, the lineshape of the C 1s core-level spectrum and the ratio of the [CH]:[CO]:[COO] species, as shown in Fig. 2c, are significantly different from that of the pure GMA graft copolymerized PTFE surface shown in Fig. 2b (see also below).

The concentration of the grafted GMA polymer can be expressed as the [COO] to $[\text{CF}_2]$ ratio and is calculated from the corresponding peak area ratios in the C 1s core-level spectra. Fig. 3 shows the effect of Ar plasma treatment time of the PTFE film on the GMA graft concentration

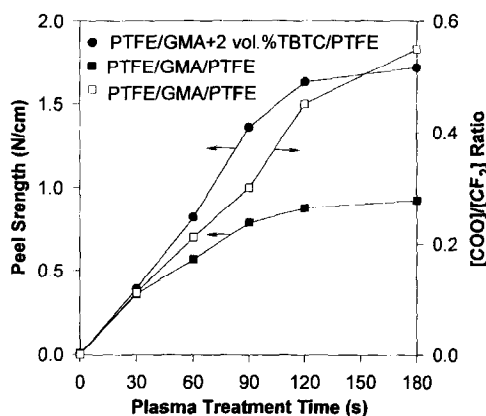


Fig. 3. Effect of Ar plasma treatment time on the concentration of the grafted GMA polymer and the peel adhesion strength between two PTFE films under the condition of grafting/lamination at 180°C for 4 h.

and the adhesion strength of the PTFE/GMA/PTFE and PTFE/GMA + 2 vol% TBTC/PTFE assemblies for grafting/lamination carried out at 180°C for 4 h. The concentration of the grafted GMA polymer increases monotonously with the plasma pretreatment time up to a plasma pretreatment time of 120 s, after which the graft concentration increases at a much slower rate, consistent with the change of the [O]/[C] ratio shown in Fig. 1. It is likely that increasing the time of exposure to plasma will generate more active sites on the film surface, resulting in a higher graft concentration. For both type of assemblies, the peel adhesion strength increases rapidly with the plasma treatment time up to the plasma pretreatment time of 120 s, after which the adhesion strength increases at a much slower rate. The latter phenomenon is also consistent with the fact that the graft concentration approaches an asymptotic value after 120 s of plasma pretreatment. For the PTFE/GMA/PTFE assembly with plasma pretreatment time of 180 s for the films, the peel adhesion strength reaches about 0.9 N cm⁻¹ at a graft concentration ([COO]/[CF₂]) of about 0.56. The data in Fig. 3 also show that the adhesion strength of the assembly is significantly

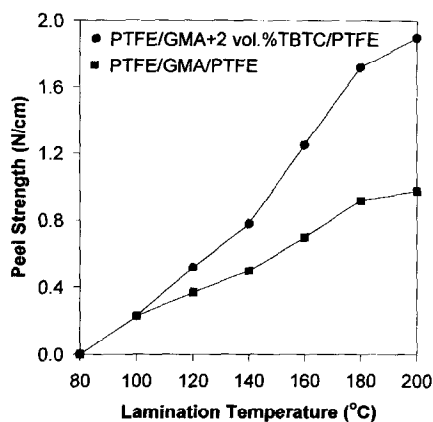


Fig. 4. Effect of thermal lamination temperature on the peel adhesion strength under the conditions of 180 s of plasma pretreatment time for the PTFE films and thermal grafting/lamination for 4 h.

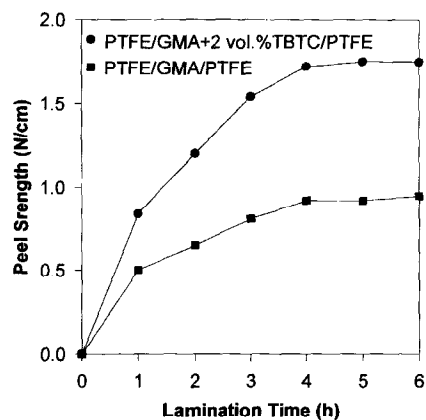


Fig. 5. Effect of thermal lamination time on the peel adhesion strength under the conditions of 180 s of plasma pretreatment time for the PTFE films and thermal grafting/lamination at 180°C.

increased by the addition of TBTC in the monomer solution. The peel adhesion strength of the PTFE/GMA + 2 vol% TBTC/PTFE assembly can reach about 1.7 N cm⁻¹ for the 180 s plasma pretreated substrates.

Fig. 4 and Fig. 5 summarize, respectively, the effects the thermal grafting/lamination temperature and grafting/lamination time on the adhesion strength of the PTFE/GMA/PTFE and PTFE/GMA + 2 vol% TBTC/PTFE assemblies. Increasing grafting/lamination temperature results in a monotonous increase in the adhesion strength (Fig. 4). The adhesion strength of each assembly, however, reaches an asymptotic value after 4 h of grafting/lamination time (Fig. 5). In all cases, the PTFE/GMA + 2 vol% TBTC/PTFE assembly always exhibits a higher adhesion strength than that of the corresponding PTFE/GMA/PTFE assembly. The TBTC molecule has three vinyl groups. It can react with GMA chains in the interface or directly graft copolymerized onto the PTFE film surface resulting in an increase in the degree of entanglement and crosslinking of the polymer chains at the interface. The data in Figs. 3–5 thus suggest that it is entirely possible to achieve a one-step, graft copolymerization with concurrent lamination between two surface pretreated PTFE films under atmospheric conditions and in the complete absence of a polymerization initiator.

The present adhesion phenomenon can be attributed to a large extent to the intermolecular interactions at the interface of the two contacting films [21,22]. At elevated temperature, the peroxides generated by Ar plasma on the film surface underwent decomposition to initiate the graft copolymerization on the two PTFE surfaces. As the reaction proceeds, the developing chains diffuse across the interface and become entangled with one another [23,24]. In other words, the observed adhesion strength arises mainly from the entanglement and diffusion of the grafted chains at the interface. The fact that the peel adhesion strength is enhanced significantly in the presence of a small quantity of added crosslinking agent provides further support for the proposed mechanism.

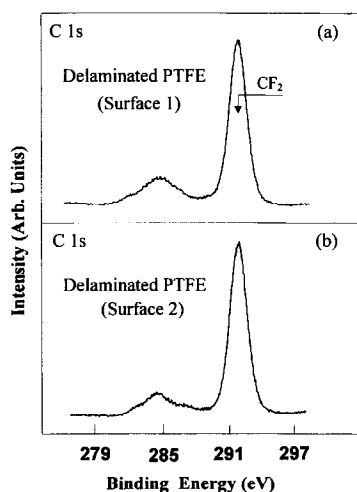


Fig. 6. C 1s core-level spectra of the delaminated PTFE film surfaces from a mechanically delaminated PTFE/GMA/PTFE assembly and a PTFE/GMA + 2 vol% TBTC/PTFE assembly having a peel adhesion strength of about 0.9 and 1.7 N cm⁻¹, respectively.

The failure modes of the graft copolymerized and laminated PTFE/GMA/PTFE assembly and the PTFE/GMA + 2 vol% TBTC/PTFE assembly were also investigated. Fig. 6a and Fig. 6b show the C 1s core-level spectra for a pair of mechanically delaminated PTFE films after thermal graft copolymerization at 180°C for 4 h and having a peel adhesion strength of about 0.9 N cm⁻¹. Before the graft copolymerization and lamination process, the films were pretreated with plasma for 180 s. In comparison with the C 1s core-level spectrum of the pristine PTFE film shown in Fig. 2a, trace amounts of GMA polymer can be detected on both of the delaminated film surfaces. This result suggests that the adhesion failure of the PTFE/GMA/PTFE assembly must have involved the fracture of the graft chains at the lapped interface. Thus the failure mode is adhesional in nature. A similar failure mechanism was also observed for the PTFE/GMA + 2 vol% TBTC/PTFE assembly with a peel adhesion strength of about 1.7 N cm⁻¹, as suggested by the presence of a strong C 1s core-level signal arising from the graft chains and TBTC on the delaminated PTFE surface (Fig. 2c).

3.2. Graft copolymerization of LDPE films with concurrent lamination

The change in the surface [O]/[C] atomic ratio of the LDPE film, as determined from the C 1s and O 1s core-level spectra peak–area ratios at the photoelectron take-off angle of 75°, as a function of the Ar plasma treatment time is shown in Fig. 7. The [O]/[C] ratio increases abruptly with the plasma treatment time, and reaches an asymptotic value after 10 s of plasma treatment.

Fig. 8a–c show the C 1s core-level spectra of a pristine LDPE film, a 10 s Ar plasma treated LDPE film, and a 10 s plasma pretreated film after graft copolymerization with

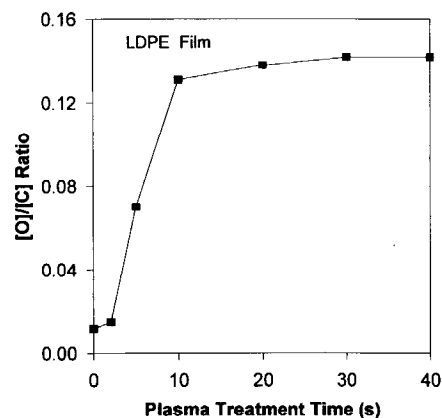


Fig. 7. Effect of Ar plasma treatment time on the [O]/[C] atomic ratio of the LDPE film.

GMA at 80°C for 2 h. The original narrow C 1s peak of the pristine LDPE becomes skewed towards the high BE side after plasma treatment. Four different peak components are discernible in the spectrum: a peak with BE at 284.6 eV for the neutral carbon species, 286.1 eV for the C–O species, 287.7 eV for the C=O species and 288.9 eV for the O=C–O species [25]. Comparison of Fig. 8b and Fig. 8c, reveals that the C–O and O=C–O component peak areas are significantly increased on the GMA graft copolymerized LDPE film. For simplicity, the number of GMA units per unit carbon of the LDPE substrate or the [COO]/[C]_{LDPE} ratio, after taking into account of the small amount of COO species generated by the Ar plasma treatment, is used to represent the concentration of the grafted GMA polymer on the LDPE film. The [COO]/[C]_{LDPE} ratios, derived from the [COO] to [C]_{LDPE} component intensity ratios in the C 1s core-level spectra by taking into account

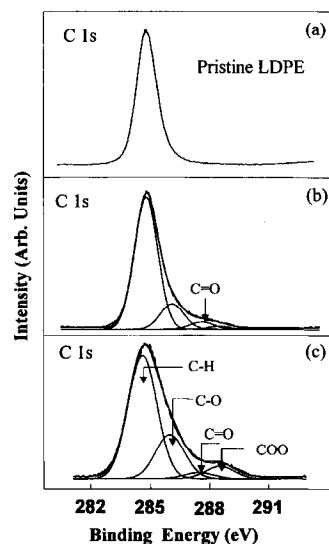


Fig. 8. C 1s core-level spectra for (a) a pristine LDPE film, (b) a 10 s Ar plasma treated LDPE film, and (c) a GMA graft copolymerized LDPE film from the mechanically delaminated LDPE/GMA/LDPE assembly prepared from the conditions of 10 s of plasma pretreatment for the LDPE film and thermal grafting/lamination at 80°C for 2 h.

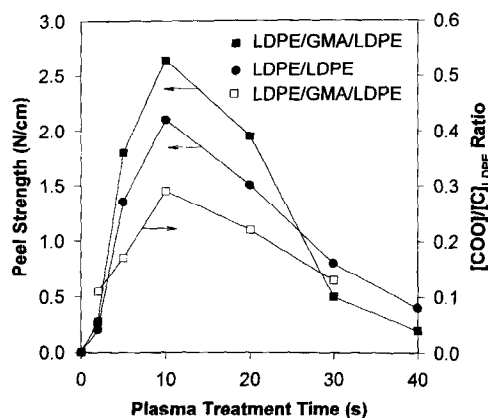


Fig. 9. Effect of Ar plasma treatment time on the concentration of grafted GMA polymer and the peel adhesion strength under the condition of grafting/lamination at 80°C for 2 h.

of the fact that each GMA unit also contributes six carbon atoms to the rest of the C 1s spectrum, as a function of the Ar plasma pretreatment time are shown in Fig. 9. The value of $[\text{COO}]/[\text{C}]_{\text{LDPE}}$ ratio reaches about 0.29 under the present experimental conditions. Theoretically, the $[\text{COO}]/[\text{C}]_{\text{LDPE}}$ ratio will approach infinity at the complete coverage of the LDPE surface by the GMA polymer within the probing depth of the XPS technique. Thus, the GMA graft copolymerized LDPE surface is below the onset of complete surface coverage under the present experimental conditions. Also, it can be seen that the amount of grafted GMA polymer increases rapidly with the Ar plasma treatment time, and reaches a maximum at the treatment time of 10 s. It had been reported that the concentration of the carboxyl groups on the LDPE film increases with plasma treatment time and the concentration of the carboxyl groups is related to the extent of scission of the polymeric chain [25]. The formation of the low-molecular-weight species will contribute to the formation of a weak boundary layer. This weak boundary layer, even after further modification by graft copolymerization, can be readily removed during the subsequent exhaustive washing process to remove adsorbed homopolymer. Alternatively, prolonged Ar plasma treatment of the LDPE surface does not result in the further oxidation of the film surface or in the formation of more surface peroxide species, as suggested by the data in Fig. 7, but is accompanied by the etching of the polymer surface. An earlier study [1] has also shown that the surface peroxide concentration of LDPE film reaches a maximum within a short time (~10 s) of Ar plasma treatment.

Figs 9, 10 and 11 summarize the dependence of the peel adhesion strength of the LDPE/GMA/LDPE assembly on the Ar plasma pretreatment time of the LDPE film, the thermal grafting/lamination temperature, and the thermal grafting/lamination time respectively. The adhesion strength increases rapidly with plasma treatment time, and reaches a maximum at 10 s of plasma treatment. Again, the observed adhesion strength exhibits a close dependence on

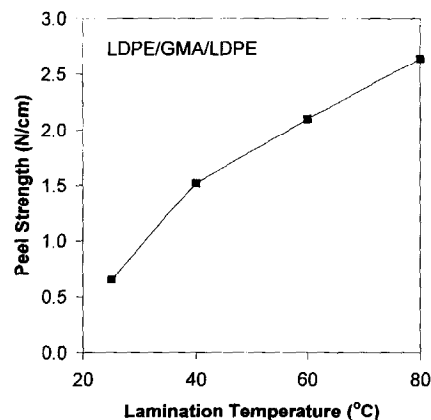


Fig. 10. Effect of thermal lamination temperature on the adhesion strength of the LDPE/GMA/LDPE assembly under the conditions of 10 s of plasma pretreatment for the LDPE film and 2 h of thermal lamination time.

the concentration of surface grafted GMA polymer. Also, the adhesion strength increases monotonously with temperature up to about 80°C. A peel adhesion strength of 2.6 N cm⁻¹ was achieved for LDPE films with 10 s of plasma pretreatment and with grafting/lamination at 80°C for 2 h.

It is well known that corona or plasma treated polyethylene films are capable of exhibiting self-adhesion properties when two contacting films are heated under pressure [26–28]. For comparison purpose, the peel adhesion strength for self-adhered LDPE/LDPE assembly as a function of plasma treatment time is also shown in Fig. 9. The adhesion results are in good agreement with those reported by Sapiuha et al. [25]. The peel adhesion strength for the LDPE/GMA/LDPE laminate is higher than that of the LDPE/LDPE laminate in the range of 5 to 20 s of plasma treatment time or when the value of $[\text{COO}]/[\text{C}]_{\text{LDPE}}$ exceeds 0.17. Thus, the adhesion strength resulting from the entanglement and diffusion of the grafted GMA chains is higher than that resulting from the self-laminated interface arising from hydrogen bonding between carbonyl groups and enolic hydrogens [27] generated by plasma treatment.

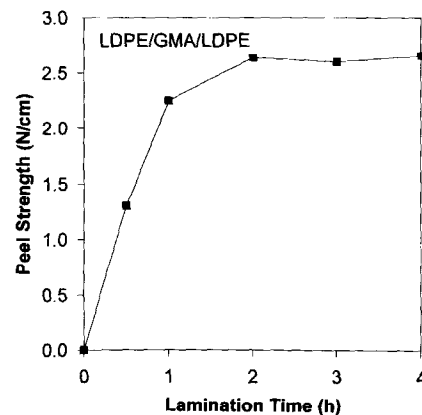


Fig. 11. Effect of thermal lamination time on the adhesion strength of the LDPE/GMA/LDPE assembly under the conditions of 10 s of plasma pretreatment for the LDPE film and followed by lamination at 80°C.

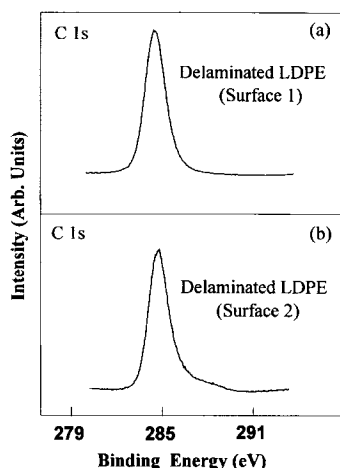


Fig. 12. C 1s core-level spectra of the delaminated LDPE film surfaces from the mechanically delaminated LDPE/GMA/LDPE assembly having a peel adhesion strength of about 2.6 N cm^{-1} .

The failure mode of the thermally grafted and laminated LDPE/GMA/LDPE assembly was investigated. Fig. 12a and Fig. 12b show the XPS C 1s core-level spectra of a pair of mechanically delaminated LDPE films after thermal graft copolymerization with simultaneous lamination at 80°C for 2 h and having a peel adhesion strength of about 2.6 N cm^{-1} . Before the graft copolymerization and lamination process, the LDPE films were pretreated with Ar plasma for 10 s. The two C 1s core-level spectra in Fig. 12 are slightly dissimilar in lineshape. One is identical to that of the pristine LDPE film as shown in Fig. 8a, while the other one is skewed slightly towards the high BE side, suggesting the presence of a trace amount of grafted GMA chains. The appearance of a fresh LDPE surface after delamination would readily suggest that the interface has delaminated by cohesive failure within one of the LDPE substrates, as a result of strong entanglement of the graft chains. As the interfacial graft layers or graft chains are still within the probing depth of the XPS technique, the graft signal associated with the high BE C 1s component is detected below the surface of the other LDPE substrate.

4. Conclusion

Interfacial thermal graft copolymerization induced laminations of PTFE/PTFE films and LDPE/LDPE films in the presence of pure GMA monomer and GMA monomer containing 2 vol% of the crosslinking agent triallyl-1,3,5-benzene-tricarboxylate were investigated. The graft modified surfaces were studied by XPS. Efficient thermal grafting with concurrent lamination can be achieved under atmospheric conditions and in the absence of any polymerization

initiator. The T-peel adhesion strength depends on graft concentration, plasma pretreatment time, thermal grafting/lamination temperature, thermal grafting/lamination time, and the presence of the crosslinking agent (for the PTFE/PTFE assembly). The T-peel adhesion strength can be enhanced by a higher graft concentration and the presence of a small quantity of the crosslinking agent. XPS results of the delaminated surfaces suggest that the failure mode of the PTFE/GMA/PTFE assembly and PTFE/GMA + 2 vol% TBTC/PTFE assembly is adhesional in nature, and that of the LDPE/GMA/LDPE assembly is cohesive in nature.

References

- [1] Suzuki M, Kishida A, Iwata H, Ikada Y. *Macromolecules* 1986;19:1804.
- [2] Ikada Y. *Biomaterials* 1994;15:725.
- [3] Mittal KL, editor. *Polymer surface modification: relevance to adhesion*. Utrecht: VSP, 1995.
- [4] Ikada Y, Uyama Y. *Lubricating polymer surface*. Lancaster, PA: Technomic, 1993.
- [5] Rånby B, Gao ZM, Hult A, Zhang PY. *ACS Symp Ser* 1988;364:168.
- [6] Rånby B. *J Adhes Sci Technol* 1995;9:599.
- [7] Allmer K, Hult A, Rånby B. *J Polym Sci, Part A: Polym Chem* 1988;26:2099.
- [8] Zhang PY, Rånby B. *J Appl Polym Sci* 1990;40:1647.
- [9] Uchida E, Uyama Y, Ikada Y. *J Polym Sci, Part A: Polym Chem* 1989;27:527.
- [10] Mori M, Uyama Y, Ikada Y. *J Polym Sci, Part A: Polym Chem* 1994;32:1683.
- [11] Kang ET, Neoh KG, Chen W, Tan KL, Huang CC, Liaw DJ. *J Adhes Sci Technol* 1996;10:725.
- [12] Kang ET, Tan KL, Kato K, Uyama Y, Ikada Y. *Macromolecules* 1996;29:6872.
- [13] Zhang X, Kang ET, Neoh KG, Tan KL, Kim DY, Kim CY. *J Appl Polym Sci* 1996;68:625.
- [14] Yang WT, Rånby B. *Macromolecules* 1996;29:3308.
- [15] Yang WT, Rånby B. *J Appl Polym Sci* 1996;62:533.
- [16] Yang WT, Rånby B. *J Appl Polym Sci* 1996;62:545.
- [17] Yang WT. *Lamination by photografting*. Ph.D. thesis, Department of Polymer Technology, Royal Institute of Technology, Stockholm, Sweden, 1996.
- [18] Wang T, Kang ET, Neoh KG, Tan KL, Cui CQ, Lim TB. *J Adhes Sci Technol* 1997;11:679.
- [19] Wang T, Kang ET, Neoh KG, Tan KL, Liaw DJ. *Langmuir* 1998;14:921.
- [20] Badey JP, Espuche EU, Jugnet Y, Sage D, Tran M-D, Chabert B. *Polymer* 1994;35:2472.
- [21] Pritchard HW. *Acta Polym* 1983;34:112.
- [22] Voyutskii SS. *J Adhesion* 1971;3:69.
- [23] Chen KS, Uyama Y, Ikada Y. *J Adhes Sci Technol* 1992;6:1023.
- [24] Chen KS, Uyama Y, Ikada Y. *Langmuir* 1994;10:1319.
- [25] Sapietha S, Cerny J, Klemberg-Sapieth JE, Martinu L. *J Adhesion* 1993;42:91.
- [26] Kim CY, Evans J, Goring DAI. *J Appl Polym Sci* 1971;15:1365.
- [27] Owens DK. *J Appl Polym Sci* 1975;19:265.
- [28] Strobbel M, Strobel JM, Lyons CS. *SPE Technical Paper* 1991;37:1096.