# Low-Temperature Graft Copolymerization of 1-Vinyl Imidazole on Low-Density Polyethylene Films with Simultaneous Lamination of Copper Foils

# H. S. HAN,<sup>1</sup> K. L. TAN,<sup>1</sup> E. T. KANG,<sup>2</sup> K. G. NEOH<sup>2</sup>

<sup>1</sup>Department of Physics, National University of Singapore, Kent Ridge, Singapore 119260

<sup>2</sup>Department of Chemical Engineering, National University of Singapore, Kent Ridge, Singapore 119260

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ABSTRACT: Surface modification of argon plasma-pretreated low-density polyethylene (LDPE) films by graft copolymerization with 1-vinyl imidazole (VIDz) and with concurrent lamination of copper foils at room temperature and at an elevated temperature were carried out. The adhesion strengths were reported as lap shear adhesion strengths and T-peel strengths. The surfaces of the graft copolymerized films and the mechanically delaminated LDPE and Cu surfaces were characterized by X-ray photoelectron spectroscopy (XPS). It was found that plasma pretreatment of LDPE alone, and in the absence of VIDz, could give rise to strong lap shear adhesion between the polymer and copper. Significant T-peel strengths, however, were obtained only for LDPE/Cu laminates obtained from the simultaneous graft copolymerization and lamination technique. © 1998 John Wiley & Sons, Inc. J Appl Polym Sci 70: 1977–1983, 1998

Key words: LDPE; 1-vinyl imidazole; copper; lamination; grafting; peel strength

## INTRODUCTION

As an exemplary polymer, polyethylene, which is easy to process and very inexpensive, has many desirable mechanical properties and good resistance to weathering. It has been used in huge quantities in many applications. For example, polyethylene finds application, in conjunction with metals, as a corrosion-resistant surface coating and as a hot melt adhesive.<sup>1</sup> However, due to its low surface energy, it normally adheres poorly to other materials. To increase the adhesion of the polymer to other materials, several methods for polymer surface modification have been developed.<sup>2–8</sup>

Surface graft copolymerization is an effective and viable adhesion improvement technique for

Correspondence to: E. T. Kang.

polymer surfaces.<sup>9</sup> Rånby et al.<sup>10-12</sup> demonstrated that initiator promoted surface graft copolymerization when carried out at the interfaces between two contacting polymer films is accompanied by the simultaneous lamination of the polymer films. More recently, we have also demonstrated that simultaneous grafting and lamination of polymer films can be achieved in the complete absence of an added polymerization initiator when the polymer substrates were pretreated or preactivated to generate the peroxide or hydroxyl peroxide species.<sup>13</sup> Earlier studies<sup>14,15</sup> have also demonstrated that the ozone and corona-discharge pretreated LDPE and related films are capable of adhering to one another in the absence of an adhesive. In the present work, surface modification of plasma pretreated low-density polyethylene (LDPE) films by graft copolymerization with 1-vinyl imidazole (VIDz) and the simultaneous adhesion between the modified LDPE films and copper foils at room temperature and at an

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elevated temperature are reported. Good adhesion strengths, measured either as the lap shear strengths or as the T-peel strengths, were obtained for samples grafted and laminated both at room temperature and at elevated temperatures. The surfaces of the modified films were characterized by X-ray photoelectron spectroscopy (XPS). The adhesion strengths of the polymer and copper interfaces obtained from the simultaneous grafting and lamination process were compared to those obtained from the autoadhesion of plasmapretreated LDPE films and copper foils.

## **EXPERIMENTAL**

#### **Materials**

Commercial LDPE films having a thickness of 0.125 mm and a density of 0.92 g/cm<sup>3</sup>, and copper foils having a thickness of 0.1 mm were purchased from Goodfellow Inc. of Cambridge, UK. The surfaces of the LDPE films were cleaned by extraction with acetone prior to plasma pretreatment. The copper foils were washed with a mixture of aqueous 0.01 M HCl/0.01 M HNO<sub>3</sub>/0.01 M H<sub>2</sub>SO<sub>4</sub> acids in the volume ratio of 6:1:1, respectively, for 3 min, rinsed with distilled water several times, and then cleaned with acetone. After drying under reduced pressure, the cleaned copper foils were used immediately for the bonding experiments with the LDPE films. The monomer, 1-vinyl imidazole (VIDz) was purchased from Aldrich Chemical Co., Milwaukee, WI, and was used as received. Acetone was of reagent grade and was purchased from Mallinckrodt Baker, Inc. of Kentucky.

#### **Graft Polymerization and Adhesion**

Prior to graft copolymerization, LDPE films were treated with 40 kHz Ar plasma in a glow discharge quartz reactor manufactured by Anatech Co. Ltd. of USA (Model SP100 Plasma System). The plasma power supply was set at 32 W. The film was exposed to the glow discharge at an argon pressure of about 0.58 Torr for a predetermined period of time. The plasma-pretreated films were subsequently exposed to atmosphere for about 30 min before the graft copolymerization and lamination experiments.

For lamination without graft copolymerization, a plasma-pretreated film and a copper foil were pressed and sandwiched between two microscope glass slides by means of two mechanical clips, completing an LDPE/Cu assembly. In the case of graft copolymerization with concurrent lamination, a drop of the VIDz monomer was introduced between a plasma-pretreated LDPE film and a copper foil, followed by pressing the polymer film and the copper foil between two glass slides as before, completing the LDPE/VIDz/Cu assembly. Both types of assemblies were kept for a predetermined period of time either at room temperature (25°C) or at an elevated temperature (80°C). For lamination carried out at the elevated temperature, the assembly was cooled gradually to room temperature over a period of 12 h in a wellinsulted oven to minimize thermal stresses at the polymer-metal interface.

## **Surface Characterization**

The LDPE films after surface graft copolymerization with concurrent lamination of the copper foils, were either immersed in distilled water for self-delamination or were delaminated by mechanical shearing. The delaminated surfaces were characterized by X-ray photoelectron spectroscopy (XPS). The surface-modified films from self-delamination in water were washed with copious amounts of water for 24 h to remove the adsorbed homopolymer and unreacted monomers before the XPS measurements to determine the graft concentration. XPS measurements were made on a VG ESCALAB MKII spectrometer with a MgK $\alpha$  X-ray source (1253.6 eV photons) at a constant retard ratio of 40. The films were mounted on the standard sample studs by means of double-sided adhesive tapes. The core-level signals were obtained at photoelectron take-off angle of 75°. The X-ray source was run at a reduced power of 120 W (12 kV and 10 mA). The pressure in the analysis chamber was maintained at 7.5  $\times$  10<sup>-9</sup> Torr or lower during each measurement. All binding energies (BEs) were referenced to the neutral carbon C1s peak at 284.6 eV. Surface elemental stoichiometries were determined from XPS peak area ratios, after correcting with the experimentally determined sensitivity factors, and were reliable to  $\pm 5\%$ . The elemental sensitivity factors were determined using stable binary compounds of well-established stoichiometries.

The graft concentration within the probing depth of the XPS technique<sup>9</sup> in each case was expressed as the number of repeating units of the grafted VIDz polymer per repeating unit of the substrate LDPE, and was determined from the XPS-derived surface stoichiometries according to:



**Figure 1** C1s and O1s core-level spectra of (a) and (b) a pristine LDPE film, and (c) and (d) a LDPE film with 10 s of Ar plasma pretreatment.

[Graft Concentration]

$$= [(N1s)/2]/\{[(C1s) - 5 \times (N1s)/2]/2\}$$
$$= (N1s)/[(C1s) - 5 \times (N1s)/2]$$

where the N1s and C1s XPS spectral areas have been corrected with the experimentally determined sensitivity factors. The factors 2 and 5 are introduced to account for the fact that there are 2 carbon atoms per repeating unit of LDPE chain, and 2 nitrogen atoms and 5 carbon atoms per repeating unit of the grafted VIDz polymer.

#### **Adhesion Strength Measurements**

The adhesion strengths were determined by measuring the lap shear force and the T-peel adhesion force at room temperature (25°C) with an Instron 5544 tensile tester. All measurements were carried out at a crosshead speed of 10 mm/min. For each adhesion strength reported, at least three sample measurements were averaged. For the lap shear strength measurement, the contact area of the two films in an assembly was kept at 0.5 × 0.1 cm, and for the peel strength measurement, at 1.0 × 0.5 cm.

## **RESULTS AND DISCUSSION**

Figures 1(a)–(d) show the respective the C1s and O1s core-level spectra of a pristine LDPE film and a 10-s Ar plasma-pretreated LDPE film. A small quantity of oxygen was detected on pristine LDPE film surface, as indicated by the weak O1s core-level signal in Figure 1(b). The presence of oxygen

on pristine LDPE film surfaces as a result of compounding and thermoplastic processing<sup>16</sup> had also been demonstrated in other studies.<sup>17</sup> A relative high level of bonded oxygen species was detected on the plasma-pretreated LDPE film [Fig. 1(c) and (d)], The observation is consistent with the earlier reports on plasma treated LDPE films.<sup>18–21</sup> Furthermore, the formation of peroxides on the Ar plasma treated LDPE films has been determined with good accuracy using 1,1-diphenyl-2-picrylhydrazyl.<sup>18</sup>

Figure 2 shows the effect of Ar plasma pretreatment time on the surface concentration of oxygen. The presence of different oxygenated functional groups, such as the C-O, C=O, O—C=O species, were detected by XPS [see Fig. 1(c)]. The oxygenated functionalities, for example, the carboxylic acid groups, play a significant role in the promotion of adhesion.<sup>22-24</sup> A earlier study<sup>25</sup> also demonstrated that oxidation of polyethelene with an oxidaizing agent prior to bond formation gave good adhesion of polyethylene to copper. It was known that corona treatment markedly enhanced the autoadhesion of LDPE sheets or films.<sup>14,15</sup> The self-adhesion property can also arise from hydrogen bonding between the carbonyl groups and the enolic hydrogens. In the present study, it was found that when a plasmapretreated LDPE film was brought into intimate contact with a copper foil, a significant lap shear adhesion strength was observed. Figure 2 also shows the dependence of lap shear adhesion strength for the LDPE/Cu assembly as a function of plasma pretreatment time of the LDPE film for laminations carried out at room temperature



**Figure 2** Effect of plasma pretreatment time on the oxygen concentration of LDPE film surface and on the lap shear adhesion strength of the LDPE/Cu assembly.

(25°C) for 24 h. The adhesion strength of an assembly consisting of a pristine LDPE film (i.e., without any plasma pretreatment) and a copper foil was infinitely low (Fig. 2), because copper could not undergo chemical interaction with the low energy surface of pristine polyethylene.<sup>26</sup> The lap shear adhesion strength of the assembly consisting of a plasma-pretreated LDPE film and a copper foil arises sharply with plasma pretreatment time. The optimum strength is achieved for LDPE film with 10 s of Ar plasma pretreatment time. The lap shear adhesion strength decreases substantially with further increase in the plasma pretreatment time. This result is similar to that observed during the surface graft copolymerization of the Ar plasma-pretreated LDPE film.<sup>18</sup> Prolonged plasma treatment may produce an etching effect, resulting in the removal of the activated and polar species from the polymer surface.

The C1s and N1s core-level spectra of the LDPE film with surface grafted VIDz polymer obtained from a self-delaminated LDPE/VIDz/Cu assembly in water, together with those of the LDPE surface and the copper surface obtained from delamination by peeling of a LDPE/VIDz/Cu assembly, are shown in Figure 3(a)-(f). The appearance of the N1s core-level signal in Figure 2(b) readily suggests that the VIDz polymer was indeed grafted onto the LDPE film surface. The molar ratio  $[VIDz]/[C_2H_4]$ , i.e., number of repeating units of the VIDz polymer per repeating unit of the substrate LDPE, for the LDPE surface obtained from delamination by peeling of the LDPE/ VIDz/Cu assembly is higher than that of the LDPE film with surface grafted VIDz polymer, because the LDPE film with surface grafted VIDz polymer was obtained through self-delamination of the graft copolymerized and laminated LDPE/ VIDz/Cu assembly after prolonged soaking and washing in water to remove the adsorbed VIDz homopolymer.

The N1s core-level spectra can be curve-fitted with four components. The two high BE components are attributable to the amine  $(-\underline{N}-\underline{C}-)$ and imine  $(=\underline{N}-)$  nitrogen of the VIDz ring, while the two low BE components is attributable to the nitrogen of the VIDz ring charge transfer complexed with Cu atoms or ions.<sup>27–29</sup> The transfer of electron from copper to the imidazole ring would result in a more electron rich environment for the nitrogen species  $(-\underline{N}-\underline{C}-)$  and  $=\underline{N}-)$ and, thus, a negative BE shift. The presence of a similar low BE peak has been suggested for the



**Figure 3** C1s and N1s core-level spectra of (a) and (b) the LDPE film with surface grafted VIDz polymer obtained from the self-delaminated LDPE/VIDz/Cu assembly in water; (c) and (d) the LDPE surface, and (e) and (f) the copper surface obtained from delamination by peeling of the LDPE/VIDz/Cu assembly, which exhibits a peel strength of 2.1 N/cm. Ar plasma pretreatment time of the LDPE film is 10 s, grafting/lamination temperature for the LDPE/VIDz/Cu assembly is 25°C and the grafting/lamination time is 24 h.

charge transfer transfer complex resulted form the deposition of copper on poly(vinylimidazole).<sup>29</sup>

It was known that the adhesion strength of a metal thin film on a polymeric substrate will depend on the chemistry across the interface and the strength of chemical interaction between the metal and the polymer, i.e., on the chemical reactivity and nature of both constituents of the interface.<sup>26,30</sup> When VIDz was introduced into the interface between the LDPE film and the copper foil for the simultaneous graft copolymerization and lamination, the lap shear adhesion strength of the so-formed LDPE/VIDz/Cu assembly was insignificant if the LDPE film had not been pretreated with plasma.

Plasma pretreatment increased the lap shear adhesion strength significantly, with the lap shear adhesion strength reaching the optimum value in just 10 s of Ar plasma pretreatment. Figure 4 shows the dependence of the lap shear adhesion strength for the LDPE/VIDz/Cu assembly as a function of plasma pretreatment time for grafting and lamination carried out at room tem-



**Figure 4** Effect of plasma pretreatment time on the graft concentration of the LDPE film surface and on the lap shear adhesion strength of the LDPE/VIDz/Cu assembly laminated at room temperature for 24 h.

perature for 24 h. Comparison of the dependence of lap shear adhesion strength of the LDPE/Cu assembly to that of the LDPE/VIDz/Cu assembly on the plasma pretreatment time (Fig. 2 vs. Fig. 4) suggests that the adhesion strength is improved by the simultaneous presence of graft copolymerization with VIDz at plasma pretreatment time greater than 10 s. However, it should be emphasized that the mechanisms of interfacial adhesion in the presence and in the absence of VIDz are probably different, as suggested by the distinctive differences in adhesion rate (Fig. 6) and T-peel strength between the two types of laminates. Plasma pretreatment followed by exposure to air not only introduces oxygenated functionalities into the surface, but also creates macromolecular radicals that can be mostly converted into peroxides.<sup>18,29</sup> Labile peroxides can initiate graft copolymerization in the presence of a vinyl or acrylic monomer.<sup>17,31</sup> The fact that a long period of lamination time ( $\geq 12$  h, see also Fig. 6) is required for the development of a substantial adhesion at 25°C suggest that a long induction period is required for the peroxide-induced graft copolymerization to be initiated at this low temperature. XPS analysis of the self-delaminated LDPE film from the LDPE/VIDz/Cu assembly showed that VIDz was graft polymerized onto the plasma-pretreated LDPE film surface, as indicated by the appearance of a fairly strong N1s core-level signal [Fig. 3(b)]. The concentration of surface grafted VIDz polymer as a function of Ar plasma pretreatment time is also summarized in Figure 4. The significant adhesion strength is believed to be due to the formation of Cu complex with N in the grafted VIDz polymer.

The contribution of graft copolymerization to the adhesion strength of the polymer-metal interface is best reflected in the T-peel strength of the interface. The peel strengths of the LDPE/Cu assemblies are very low even when the LDPE films have been plasma pretreated before being bonded to copper foils. When the lamination is accompanied by graft copolymerization with VIDz, the T-peel strength of the polymer/metal interface is significantly enhanced. Figure 5 shows the dependence of the peel strengths of the LDPE/Cu and LDPE/VIDz/Cu assemblies on the plasma pretreatment time of the LDPE substrates.

The relationship between the lap shear adhesion strength and room temperature lamination time, as shown in Figure 6, also suggests that the adhesion mechanism of the LDPE/VIDz/Cu assembly was different from that of the LDPE/Cu assembly. Under the atmospheric conditions, the lap shear adhesion strength of the LDPE/Cu assembly develops rapidly with the lamination time, whereas there exists an induction period before the commencement of the increase in lap shear adhesion strength of the LDPE/VIDz/Cu assembly. Chemisorption, which took place between the active groups on the surfaces of plasma-pretreated LDPE films and copper atoms in the copper foil, presumably accounts for the rapid increase in the lap shear adhesion strength of the LDPE/Cu assemblies. The inhibition in the increase in lap shear adhesion strength of the



**Figure 5** Effect of plasma pretreatment time on the T-peel strength of plasma pretreated polymer/metal assemblies laminated in the absence and presence of VIDz at room temperature for 24 h.



**Figure 6** Lap shear adhesion strength of the Ar plasma pretreated LDPE/Cu assemblies laminated in the absence and presence of VIDz as a function of lamination time at room temperature. The time of plasma pretreatment for the LDPE films was 10 s.

LDPE/VIDz/Cu assemblies probably resulted from the loss of chemisorption between the polymer and the metal in the presence of VIDz and the presence of an induction period for the polymerization of VIDz under atmospheric conditions and at room temperature. XPS analyses of the delaminated LDPE films from the corresponding assemblies show that the concentration of the grafted VIDz polymer on the LDPE film surface also exhibits a sluggish increase with the lamination time during the initial stage. An abrupt increase in graft concentration is observed only after 10 h of lamination time (Fig. 6).

The inhibition of graft copolymerization was relieved for grafting and lamination carried out at elevated temperature. When the LDPE/Cu assemblies or the LDPE/VIDz/Cu assemblies were kept at 80°C for only 2.5 h, relatively high lap shear adhesion strengths were obtained in both cases, as shown in Figure 7. It seems that the interaction between copper foils and the oxygenated groups on the surfaces of plasma-pretreated LDPE films, and the removal of the inhibition in the rate of polymerization of VIDz are facilitated by high temperature. Furthermore, lamination and grafting at the elevated temperature also results in an improvement in the adhesion strength between the LDPE and copper. The T-peel strengths for both types of assemblies also develop within a much short period of lamination  $(\sim 2.5 \text{ h})$  at the elevated temperature. Nevertheless, a significant peel strength was observed again only for the assembly with interfacial graft



**Figure 7** Lap shear adhesion strength of the LDPE/Cu assemblies laminated in the absence and presence of VIDz at 80°C for 2.5 h as a function of the Ar plasma pretreatment time for the LDPE films.

copolymerization (Fig. 8). In addition to contribution from the grafted chains at the interface, other processes, such as salt formation between COOH groups on the LDPE film and VIDz on the copper surface, may also have contributed to the observed adhesion strength of the LDPE/Cu laminate.

Finally, the failure mode of the LDPE/VIDz/Cu laminate was briefly studied. The C1s and N1s core-level spectra of delaminated LDPE and Cu surface from a LDPE/VIDz/Cu assembly exhibiting a peel strength of 2.1 N/cm are shown in Figure 3(c)–(f), respectively. Comparison of the



**Figure 8** Peel strength of the LDPE/Cu assemblies laminated in the absence and presence of VIDz at 80°C for 2.5 h as a function of the Ar plasma pretreatment time for the LDPE films.

intensities of the N1s core-level spectra on the mechanically delaminated LDPE and Cu surfaces readily suggests that most the grafted VIDz polymer chains remain on the LDPE surface. This observation, together with fact that a strong Cu signal is detected form the mechanically delaminated Cu surface, readily suggests that the present LDPE/VIDz/Cu assembly fails by mechanical delamination at the graft/Cu interface.

# CONCLUSION

Surface modification of Ar plasma pretreated LDPE films by graft copolymerization with 1-vinyl imidazole (VIDz) in the presence of a contacting copper metal foil is accompanied by the simultaneous lamination of the metal to the polymer. The grafting and lamination process can be carried out at room temperature and under atmospheric conditions. The induction period required for the development of adhesion strength at room temperature is circumvented by lamination at elevated temperature. The adhesion strengths are reported as lap shear adhesion strengths and Tpeel strengths. Although plasma treatment of the LDPE film alone can also result in strong lap shear adhesion between the LDPE film and copper foil, significant T-peel strength is obtained only for the polymer/metal interface laminated in the presence of simultaneous graft copolymerization. Strong adhesion between plasma pretreated LDPE and Cu presumably arises from the charge transfer interaction between the grafted VIDz polymer and Cu.

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