Diamond & Related Materials 26 (2012) 55–59

Contents lists available at SciVerse ScienceDirect

Diamond & Related Materials

journal homepage: www.elsevier.com/locate/diamond

Adhesion enhancement of polyolefins by diamond like carbon coating and photografting polymerization ☆

J. Takahashi, A. Hotta *

Department of Mechanical Engineering, Keio University, Yokohama 223-8522, Japan

ARTICLE INFO

Available online 24 April 2012

Keywords:
Diamond-like carbon (DLC)
Polymers
Adhesion
Surface treatment
Photografting polymerization

ABSTRACT

Polyolefins, such as polyethylene (PE) and polypropylene (PP) are widely used materials with low adhesion property. Diamond-like carbon (DLC)-deposited polyolefins are especially attractive for the effective control of the surface and the mechanical properties of the polyolefins, where the adhesion between the DLC and the polyolefins is the key parameter that determines the major properties. Recently we have found that the low-adhesion surfaces of some polyolefins such as high-density PE (HDPE) and linear low-density PE (LLDPE), could be drastically modified to show a good adhesion after diamond-like carbon (DLC) coating, although some other polyolefins such as low-density PE (LDPE), isotactic polypropylene (iPP), and syndiotactic polypropylene (sPP) could not be surface-modified for higher adhesion property by DLC coating, which was due to the low adhesion between polyolefins and DLC. To overcome the problem, photografting polymerization was introduced to improve the low adhesion property of LDPE, iPP, and sPP. The adhesive strengths between the polyolefins and the DLC films were remarkably increased by photografting acrylic acid (AA) onto the polyolefin surface before DLC coating. In addition, the fracture of the base polymer was observed for AA-grafted polyolefins synthesized at the grafting time of ~15–30 min, indicating the strong adhesive force between photografted polyolefins and DLC.

Furthermore, the adhesion forces between DLC and other widely-used polymer materials, such as polystyrene (PS), polymethylmethacrylate (PMMA), polyethyleneterephthalate (PET), polyurethane (PU), and polydimethylsiloxane (PDMS) which were all rather not easy to directly deposit DLC, were also found remarkably increased by photografting AA onto the polymer surfaces before DLC coating.

© 2012 Elsevier B.V. All rights reserved.

1. Introduction

Polyolefins, such as polyethylene (PE) and polypropylene (PP) are widely used and cost-effective materials with processability, recyclability, and chemical resistance. Polyolefins, however, possess low adhesion property, which occasionally prevents polyolefins from being widely used especially in the application fields demanding higher adhesion. In order to improve the low adhesion property of the polyolefins, various researches have been carried out for the surface modifications of the polyolefins [1–14]. Steluta et al. addressed the improvement of the adhesive property of ultrahigh molecular weight PE by the continuous plasma treatment to achieve sufficient surface modification of PE [15]. Lu also reported on the surface modification of PET by the ultrasonic-assisted electroless plating for higher adhesion force [16]. Both surface treatments, however, required rather lengthy processes, producing less sustainable high adhesion surfaces.

Diamond like carbon (DLC) deposition by the plasma enhanced chemical vapor deposition (CVD) method was introduced for the modification of the low adhesion surfaces of the polyolefins. DLC is an amorphous carbon with graphite structures composed of sp^2 bond and diamond structures composed of sp^3. DLC is characterized by its toughness, lubricating property, abrasion resistance, chemical stability, and biocompatibility. The good adhesion between DLC and polymers can be effectively used for e.g. drug-eluting polymers targeting stents or artificial blood vessels in the biomedical field [17] as well as gas barrier polymers for food trays and beverage containers [18]. In fact, DLC-coated polymers have already been industrially used for e.g. food packaging, food trays, and bottles for drink with high gas barrier property. Our group has recently found that the low-adhesion surface of HDPE and LLDPE could be drastically modified to show sufficient adhesion after diamond-like carbon (DLC) coating. Other polyolefins such as low-density PE (LDPE), isotactic polypropylene (iPP), and syndiotactic polypropylene (sPP), however, could not be surface-modified for the higher adhesion property by DLC coating, which was due to the low adhesion between the three polyolefins and DLC.

In order to enhance the adhesion between DLC and polyolefins, we introduced a photografted layer for the intermediate layer between

---

☆ Presented at the Diamond 2011, 22st European Conference on Diamond, Diamond-Like Materials, Carbon Nanotubes, and Nitrides, Budapest.

* Corresponding author.
E-mail address: hotta@mech.keio.ac.jp (A. Hotta).

0925-9635/$ – see front matter © 2012 Elsevier B.V. All rights reserved.
doi:10.1016/j.diamond.2012.04.006

DLC and polyolefins achieved by photografting polymerization. Only a few studies on the intermediate layers for the enhancement of the adhesion between DLC and specific polymers (polycarbonate (PC) and polymethylmethacrylate (PMMA)) have been reported so far [18,19].

Photografting polymerization is a type of radical polymerization, where a synthetic reaction starts with generated radicals of monomers. Photografting polymerization was investigated originally for a surface modification method [20–23], which was actively used for the improvement of the wettability of polymer surfaces by photografting hydrophilic monomers [24–31]. The surface wettability of the polymers grafted by acrylic acid (AA) was substantially improved with a marked change in the surface property from hydrophobic to hydrophilic. Controlled hydrophilic surface of polymers could be effectively used for e.g. biomedical purposes such as antithrombogenic surfaces for guide wires, stents and artificial organs. Also, raising the hydrophilicity of the polymers could result in developing self-cleaning polymer surfaces. Adhesion would be another interesting surface property, where high adhesion force would lead to the creation of high-quality complex or composite polymeric materials.

In this study, we investigated the effects of AA-photografting polymerization on the adhesion between DLC and polymers. It was found that the adhesion force between DLC and polyolefins was remarkably increased by introducing an AA-intermediate layer. Moreover, the adhesion forces between DLC and other widely-used polymeric materials, such as polystyrene (PS), polymethylmethacrylate (PMMA), polyethyleneterephthalate (PET), polyurethane (PU), and polydimethylsiloxane (PDMS) that are difficult to directly deposit DLC, were also examined and found remarkably increased by photografting AA onto the polymer surfaces.

2. Experimental

2.1. Materials

Polyolefins (HDPE, LLDPE, LDPE, iPP, and sPP) and other polymers (PS, PMAA, PET, PU and PDMS) have been purchased in the form of pellets from Sigma-Aldrich. Pellets of each material were thermally pressed and molded for 15 min into thin films with a thickness of 0.5 mm at temperatures 10 °C higher than each melting point of the polymers with the molding pressure of 5 MPa, using heat press equipment (A-H2003, As One Corp.). The polymeric samples were then cut into 7.0 cm in length and 1.4 cm in width for a polymer substrate for photografting polymerization. Before the T-peel experiments, the fracture strength and Young's modulus of polymers were measured by a tensile tester (AG-50NIS MS of Shimadzu Co., Ltd). The fracture strength and Young's modulus were the key mechanical properties that would determine the adhesive force. The fracture strengths and Young's moduli of HDPE, LLDPE, LDPE, iPP, sPP, PS, PMMA, PET, PU, and PDMS were calculated as 41, 16, 30, 23, 26, 30, 23, 140, 100, and 2.8 MPa for the fracture strengths respectively, and 1.3, 0.3, 0.2, 1.2, 1.6, 1.0, 0.4, 2.4, 0.3, and 0.04 GPa for the Young's moduli, respectively.

In order to obtain high adhesion surface, the surface should normally be highly hydrophilic. For that purpose, acrylic acid, methacrylic acid, and methacrylamide could be prospective candidates for graft monomers. Among them, by adhesion testing, it was found that the fracture of the base polymer, which was only found when the adhesive force was significantly high, could be observed only for the polymers grafted by acrylic acid, and thus we used AA for our experiments [32,33].

2.2. Photografting polymerization

The photografting polymerization of AA onto polymer substrates was carried out using photografting equipment (Riko rotary photochemical reactor RH400-10W, Science and Technology Industry Co., Ltd). First, benzophenone, the polymerization initiator, was dissolved in acetone for a sensitizer at a concentration of 0.0275 M. Each polymer substrate was then immersed in the benzophenone solution for 1 min to generate radicals on the polymer substrate. AA-monomer solution was prepared at the concentration of 1.0 M. The polymer substrate immersed in the benzophenone solution was then dipped into the AA-monomer solution using Pyrex reaction tubes. The reaction tubes with each monomer solution and the polymer substrates were irradiated by the ultraviolet (UV) ray emitted from a 400 W high-pressure mercury lamp at 60 °C for 60 min. The photografted polymer substrates were rinsed with pure water to remove unreacted monomers and homopolymers and dried at 50 °C.

2.3. Fourier transform infrared (FT-IR) spectroscopy

FT-IR spectroscopy (FT/IR-4200, JASCO) was carried out to analyze the chemical structures of grafted polymer substrates by analyzing the changes in the FT-IR spectra from the polymer surfaces before and after the photografting polymerization. The film samples were set on the sample stage and the IR spectra ranging from 400 to 4000 cm^{-1} in wavenumbers were analyzed. The averaged data of sixteen scans were collected to construct each FT-IR spectrum at a resolution of 2 cm^{-1} by the attenuated total reflectance (ATR) method. The peak of the carboxyl groups ($-\mathrm{COOH}$) should be observed at 1720 cm^{-1}, which can be used as an indicator of the progress of the AA-photografting polymerization.

2.4. Contact angle measurements

In order to analyze the surface of the substrates, the wettability of the surface was measured at each experimental step by a contact angle tester (DM500, Kyowa Interface Science Corp.). The contact angle measurement is a powerful tool that can clearly determine whether the exposed surface is DLC, polymer, or AA-grafted layer. The contact angle measurement was carried out by dropping pure water of 2.0 mm in diameter on the sample surfaces. The contact angle was determined by the average of ten experimentally-calculated values through water droplets.

2.5. Deposition of DLC

The deposition of DLC was carried out at the pressure of 13.3 Pa with the output of 200 W using a high-frequency plasma CVD device (Hiranokoh-on Corp.). First, the polyolefin films were placed between the two electrodes in the plasma CVD device, and the pressure was reduced to 0.3 Pa using a rotary pump and a mechanical booster pump. $\mathrm{C_2H_2}$ gas was inserted as a source of DLC with the pressure of 13 Pa. DLC was then deposited on the polymer surface for 14 s under the condition of 13.56 MHz and 200 W. The thickness of the deposited DLC was approximately 50 nm.

2.6. T-peel adhesion force testing

T-peel strength measurement was carried out under the Japanese Industrial Standard (JIS) and the International Organization for Standardization (ISO) using the tensile tester (AG-50NIS MS of Shimadzu Co., Ltd). The T-peel strength is defined as the peel adhesive strength of adhesive agent measured by the 180 degree peel test of a specimen that was adhered by putting two L-shaped specimens together to become T-shaped, which was defined by JIS K 6854-2 and ISO 11339. Also, in American Standard for Testing and Materials (ASTM) D1876-08, the T-peel test was explicitly defined as the standard test method for peel resistance of adhesives. Epoxy resin as adhesive was put on the DLC surface of one specimen which was then attached to another specimen surface-treated in the same fashion. The two specimens were bonded together under the pressure of 3.0 MPa

Download English Version:

https://daneshyari.com/en/article/702340

Download Persian Version:

https://daneshyari.com/article/702340

Daneshyari.com