



# Effect of modified polypropylene on the interfacial bonding of polymer–aluminum laminated films



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## ARTICLE INFO

### Article history:

Received 29 October 2014

Revised 6 May 2015

Accepted 7 May 2015

Available online 8 May 2015

### Keywords:

Interfacial bonding

Al foil

Modified PP

Polymer–aluminum laminated films

## ABSTRACT

The interfacial bonding between functionalized polymers and chromate–phosphate treated aluminum (Al) foil were investigated in this study. Glycidyl methacrylate (GMA), acrylic acid (AA) and maleic anhydride (MAH) were grafted onto polypropylene (PP) to improve its adhesion strength with the treated Al foil. The interfacial peel strength was evaluated by the T-peel test, and the results showed that modification of PP resulted in a significant improvement in the interfacial peel strength from 1 N/15 mm for pure PP to 10–14 N/15 mm for the modified PP. The surface chemistry, topography and surface energy of the modified PP and Al foil after peeling were characterized by time-of-flight secondary ion mass spectrometry (TOF-SIMS), X-ray photoelectron spectroscopy (XPS), attenuated total reflectance Fourier transform infrared spectroscopy (ATR-FTIR), scanning electron microscopy (SEM) and contact angle measurement. The treated Al foil could react with the functional groups of PP, resulting in the formation of new carboxylates. The new chemical bonding rather than the mechanical interlocking contributed to the improvement of adhesion strength.

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## 1. Introduction

Lithium-ion batteries have attracted much research interest because of their excellent performance and broad application prospects [1–4]. To meet the increasing demand of high energy density and good cycle characteristics, many attempts have been made to develop new electrode materials [5,6], electrolytes [7] and separators [8]. However, the packing materials that play an important role in improving the safety and reducing the weight of lithium-ion batteries have received only sporadic attention [9].

Al–polymer has recently emerged as a promising packing material for lithium-ion batteries due to its good plasticity, light weight and excellent film-forming properties. In this study, an Al–polymer laminated film consisting of six layers was prepared (Fig. 1), where the nylon film (Fig. 1 (1)) was laminated to one side of the Al foil (Fig. 1 (3)) with polyurethane (Fig. 1 (2)) as an adhesive, and the other side of the Al foil was laminated to the cast polypropylene (CPP) film (Fig. 1 (6)) with modified PP (Fig. 1 (5)) as an inner adhesive. Among these six layers, the Al foil and inner adhesive are most important in improving the bonding strength of Al–polymer. To obtain a strong and stable bonding, the surface of Al foil should

be treated firstly. Chromate conversion treatment has so far been the most effective and widely used method for this purpose [10–14]. Grafting modification is also often used to improve the bonding strength between polymer and metal. The polymer grafted with a polar monomer can have a higher surface polarity, which can improve the adhesion strength between metal and polymer. Modified polymers are often used as adhesives to improve the adhesion strength between Al and PP [15–17]. For example, the shear strength of Al–PP lap joints was improved from 3.44 to 10.30 MPa with the grafting of maleic anhydride onto PP [15]. However, although much progress has been made in this field [18], the T-peel strength is not high enough to meet the demand of packing materials used for lithium ion batteries.

In recent years, there have been a number of studies investigating the interfacial bonding between metal and polymer [19–24]. Three adhesion mechanisms have been proposed in the literature: molecular bonding, mechanical coupling, and thermodynamic adhesion [25]. However, there is no evidence to indicate which one contributes to the interfacial bonding between Al and polymer.

In this study, we successfully prepared an Al–polymer laminated film with high T-peel strength. The Al foil was treated with chromate–phosphate conversion solution, and then a multilayered laminate was prepared using PP modified by glycidyl methacrylate (PP-g-GMA), acrylic acid (PP-g-AA) or maleic anhydride (PP-g-MAH) as the inner adhesive. This laminated film consisted

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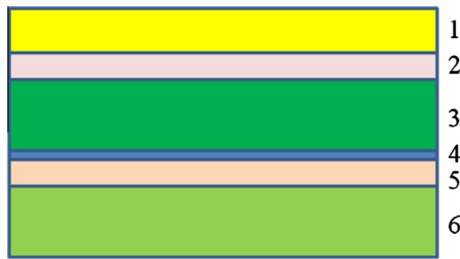


Fig. 1. Structure of an Al-polymer laminated film (1 – Nylon, 2 – Polyurethane, 3 – Al foil, 4 – Chemical conversion coating, 5 – Modified PP, 6 – CPP).

of six layers (from the outer layer to the inner layer: nylon/polyurethane/Al foil/chemical conversion coating/modified PP/PP). The interface properties between Al and modified PP were characterized using time-of-flight secondary ion mass spectrometry (TOF-SIMS), X-ray photoelectron spectroscopy (XPS) and attenuated total reflectance Fourier transform infrared (ATR-FTIR) spectroscopy. The morphologies of Al foil before and after peeling were observed by scanning electron microscopy (SEM). The hydrophilicity and surface energy of the coated modified PP were evaluated by the contact angle measurements. The results showed that the new chemical bonding played a major role in improving the adhesion strength.

## 2. Experimental section

### 2.1. Materials

Al foil (AA8021), nylon, polyurethane and CPP were provided by Jiangsu Zhongjin Matai Medicinal Packaging Co. (Jiangsu, China). PP grafted with GMA (PP-g-GMA), PP grafted with MAH (PP-g-MAH) and PP grafted with AA (PP-g-AA) were home-made in our laboratory.

### 2.2. Sample preparation

A semi-finished product, nylon/polyurethane/Al foil (Al foil and nylon film laminated with polyurethane), was provided by Jiangsu Zhongjin Matai Medicinal Packaging Co. The Al foil was treated with chromate-phosphate conversion solution [26], and then the semi-finished product was hot pressed with the modified PP and CPP film. Three types of samples with the same structure were fabricated (Fig. 1): (1) nylon/polyurethane/Al foil/chemical conversion coating/PP-g-AA/PP (Al/PP-g-AA system), (2) nylon/polyurethane/Al foil/chemical conversion coating/PP-g-MAH/PP (Al/PP-g-MAH system), and (3) nylon/polyurethane/Al foil/chemical conversion coating/PP-g-GMA/PP (Al/PP-g-GMA system).

### 2.3. Characterization and tests

The adhesion strength between Al foil and CPP was measured to evaluate the adhesion performance of the modified PP. A T-peel test was conducted according to GB8808-88 using a tensile tester CMT 2203 (SANS, China) at room temperature with a peeling speed of 300 mm/min. The peel strength was averaged over 5 samples.

The modified PP was analyzed using a Nicolet 6700 FT-IR spectrometer (Thermo Electron Corporation, America). After the peel test, the surface composition changes of Al foil were determined by ATR-FTIR and TOF-SIMS (ION-TOF GmbH, Germany).

The surface wettabilities of PP-g-GMA, PP-g-AA, PP-g-MAH were determined by measuring the static water contact angle using a contact angle goniometer JC2000D3 (Shanghai Zhongchen Digital Technic Apparatus, China). Once the water drop contacted

the sample surface, timing was started and images were collected, and the contact angle was measured at 15 s. All experiments were performed at 25 °C. The surface energy was calculated using Eq. (1) [27].

$$\cos \theta + 1 = 2(\gamma_S/\gamma_L)^{\frac{1}{2}} e^{-\beta(\gamma_L - \gamma_S)^2} \quad (1)$$

where  $\theta$  is the contact angle,  $\beta$  is equal to  $0.0001247 \text{ (mN/m)}^{-2}$ , and  $\gamma_S$  and  $\gamma_L$  are the surface free energies of solid and liquid, respectively.

Surface morphologies of Al foil before and after peeling were observed using a scanning electron microscope S-4800 (Hitachi Limited, Japan) operated at 10 kV.

To analyze the chemical state of carbon and oxygen species on the Al foil surface after peeling, XPS analysis was conducted on a ESCALAB 250 spectrometer equipped with an Al anode. The spectra were shifted to set the C–C/C–H components of the C1s peak at a binding energy of 284.6 eV to correct sample charging. The XPS spectra were curve-fitted and decomposed using XPSPEAK software.

## 3. Results and discussion

The T-peel strength between Al foil and CPP film laminated by different adhesives is shown in Fig. 2. Three functionalized PPs were prepared under the same conditions (i.e., molar of monomer, concentration of initiator, reaction time and temperature). The maximum peel strength between Al foil and CPP film (11 N/15 mm) was achieved with the use of PP-g-AA as an adhesive, followed by PP-g-MAH (10 N/15 mm), PP-g-GMA (10 N/15 mm), and pure PP (1 N/15 mm), respectively. It is obvious that the peel strength can be improved greatly by the modification

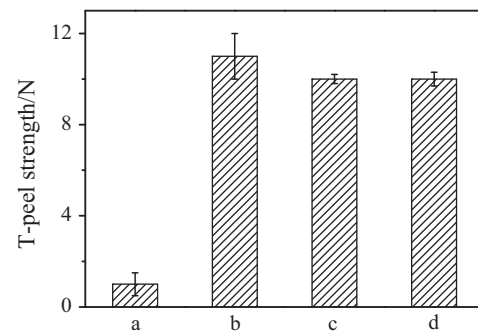


Fig. 2. T-peel strength between Al foil and CPP film with different adhesives: (a) pure PP, (b) PP-g-AA, (c) PP-g-MAH, and (d) PP-g-GMA.

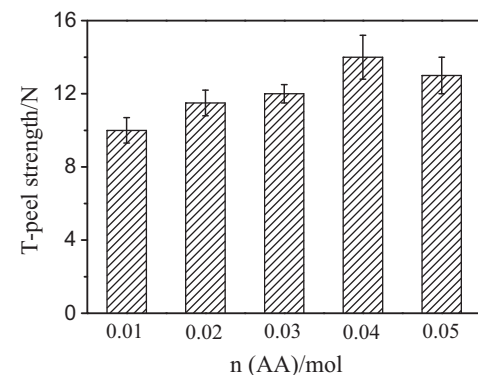


Fig. 3. Effect of AA content on the T-peel strength of PP-g-AA.

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