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Polyethylene/octa-(ethyl octadeca-10,13 dienoamide) silsesquioxane blends and the adhesion strength to paperboard

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ABSTRACT

Octa-(ethyl octadeca-10,13 dienoamide) silsesquioxane or bio-POSS was used in the fabrication of polyethylene (PE)/bio-POSS blends by melt mixing. These PE/bio-POSS blends were applied to paperboard by compression moulding coating. The *T*-peel test was used to determine the adhesion of the blends to paperboard. A FTIR-ATR spectroscopic study was performed to identify the interfacial interaction between PE/bio-POSS blends and paperboard. The *T*-peel test showed that the adhesion of PE to paperboard was enhanced when there was less than 10 wt% of bio-POSS in the blends. The best adhesions were achieved at 5 wt% and 3 wt% bio-POSS, coated at 200 °C/20 °C and 300 °C/20 °C, respectively. The increase in adhesion strength was attributed to the contribution of mechanical interlocks and probable interfacial interaction between amide groups of bio-POSS and hydroxyl groups of paperboard, as specified by FTIR-ATR. Bio-POSS content above 10 wt% led to a decrease in adhesion between PE/bio-POSS blends and paperboard because the melt flows and mechanical properties of the blends were dramatically decreased.

1. Introduction

The most widely used plastic, polyethylene (PE) is often used in laminated packaging products, *e.g.* PE-coated paperboard. A requirement for this product is good adhesion between PE and paperboard. The adhesion depends on the characteristics of the paperboard (surface roughness and porosity), PE (melt flow rate, meting point, wettability and thickness), and the coating conditions (temperature, pressure and rate of coating). It is also affected by surface chemical factors and the intermolecular forces across interface [1]. Adhesion properties may be enhanced by treating paperboard and PE before coating. Improvement in adhesion properties by surface treatment of paperboard was successfully obtained by Kemppi [1], Fredholm and Westfelt [2]. Our work has only focused on the chemical modification of the PE-containing coating layer to improve the adhesion between coating and paperboard.

Polyethylene (PE) has low surface free energy (31 mJ/m^2) and low wettability [1]. When it is coated on the paperboard, PE shows low adhesion. In order to improve the adhesion properties, it is necessary to modify the PE prior to coating. Modifying PE was performed by treating the surface (surface treatment of PE) [3,4]. In those works, the oxygen-containing polar groups such as C=O, OH, COOH are introduced to PE surface, leading to an increase in surface polarity of PE without influencing on the properties of PE. When coated on the paperboard, the interfacial attraction force is increased, thereby improving the adhesion. Besides, the properties of PE can be modified by using additives or reinforcing agents.

Inorganic/organic hybrid polymers based on polyhedral oligomeric silsesquioxanes (POSS) have been much studied in the recent years. POSS is a class of silicon compounds with an empirical formula $R(SiO_{1.5})_n$ having an inorganic core cage and organic groups attached at the vertex, with a size of 1–3 nm. The organic groups allow POSS to be easily incorporated into polymers either by copolymerisation, or physical mixing, whereupon POSS/polymer blends having significantly improved properties such as mechanical properties, thermal stability, surface hardening, and rheological properties will be made [5–9]. Recently, a considerable study on the fabrication of polymer/POSS with improved properties by using a mixing method has been published [10–16]. There is a lack of work on applying polymer/POSS to paperboard as well as determination of adhesion between the layers in such laminates.

Polymer materials fully or partly based on renewable resources are a topic of great interest. The use of such materials can significantly contribute to a sustainable development. Octa-(ethyl octadeca-10,13 dienoamide) silsesquioxane is a POSS compound derived from a natural unsaturated fatty acid (octadeca-10,13

Abbreviations: PE, Polyethylene; POSS, Polyhedral oligomeric silsesquioxanes; Bio-POSS, Octa-(ethyl octadeca-10,13 dienoamide) silsesquioxane; CTMP, Chemithermomechanical pulping; MFI, Melt flow index; FTIR, Fourier transform infrared spectroscopy; SEM, Scanning electron microscopy; DSC, Differential scanning calorimetry; IaI, IaII, Peak intensity of amide I, II; Tm, Melting temperature

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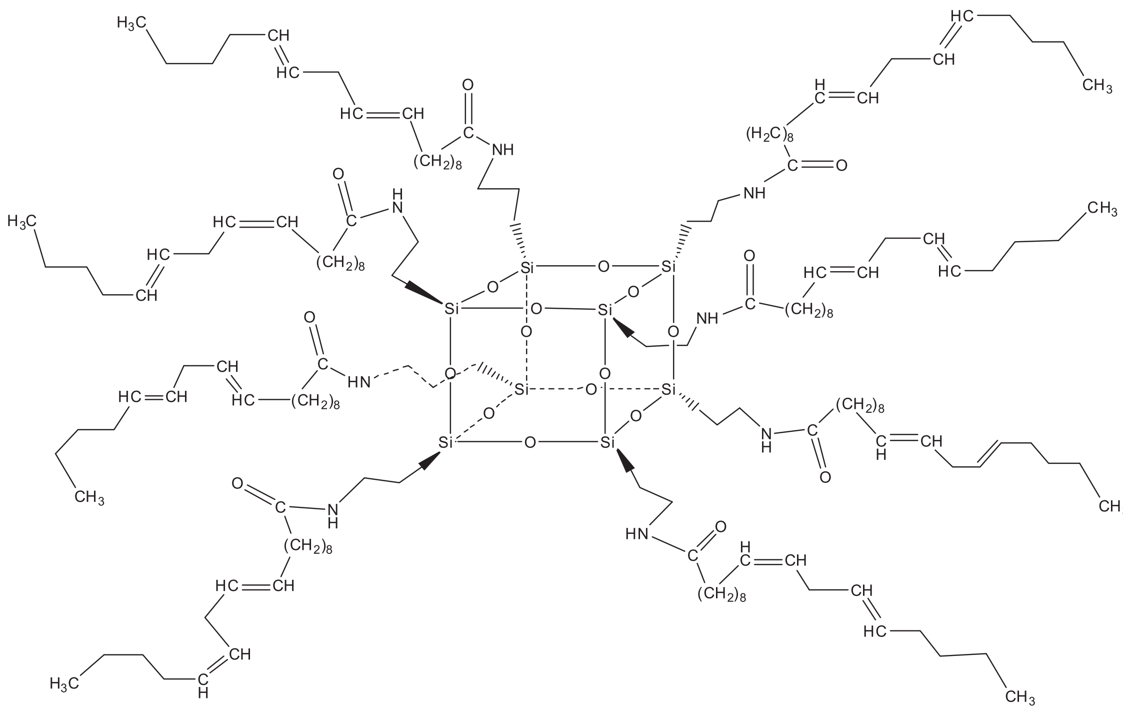

Fig. 1. Typical structure of octa-(ethyl octadeca-10,13 dienoamide) silsesquioxane (bio-POSS).

dienoic acid). This POSS compound is therefore a biological and renewable material, and called bio-POSS. The molecule of bio-POSS is typically composed of a Si_8O_{12} cubic cage and eight (ethyl octadeca-10,13 dienoamide) groups peripherally attached at eight Si vertexes, as can be seen in Fig. 1. The core cage is inorganic, giving thermal stability, while the peripheral organic groups allow bio-POSS to be incorporated into organic polymers to form POSS/polymer blends. Among the methods for fabricating polymer blends, melt mixing is efficient, cheap and simple to scale up to industrial level. Furthermore, fabricating blends by mixing will consume less time in comparison with copolymerisation that requires more time to develop new polymerisation routes. The properties of polymer/bio-POSS blends can be easily tailored by combining polymer and bio-POSS components and varying the blend composition.

The aim of this work is to investigate the effects of bio-POSS on the adhesion of polyethylene (PE) to paperboard. Various PE/bio-POSS blends have been fabricated by melt mixing in a micro-extruder. After that, the resultant blends are applied on the paperboard by compression moulding coating. The adhesion between polyethylene/bio-POSS blends and paperboard is evaluated by the *T*-peel test. The melting temperature of PE/bio-POSS blends is determined by differential scanning calorimetry (DSC). To observe the morphology of PE/bio-POSS blends, the samples are examined using scanning electron microscopy (SEM). Fourier transform infrared attenuated total reflection (FTIR-ATR) spectroscopy is used to identify the surface characteristics of the blends and paperboard.

2. Materials and methods

2.1. Materials

Polyethylene (PE) was purchased from Ineos Polyolefins and Polymers Europe, with melting point of 120 °C and the melt flow index (MFI) of 0.5 g/10 min (190 °C/2.16 kg). Norsilika8002 is a yellowish viscous liquid which was supplied by Jotun AS, Norway, containing octa-(ethyl octadeca-10,13 dienoamide) silsesquioxane (bio-POSS) dissolved in a hydrocarbon solvent, with a solid content of 40–44%. Paperboard was provided by Korsnäs AB, Sweden.

2.1.1. Paperboard

The paperboard is a much used fibre-based material for packaging due to its good stiffness, its low production cost, its lightness, its good and multiple printing opportunities and its recyclability. The paperboard has two different sides (uncoated and coated side) with different colours (brown and white), respectively. It is composed of four different layers:

(i) the bottom layer is unbleached kraft pulp of a pine and spruce softwood mix,
(ii) the middle layer is unbleached kraft pulp and chemithermo-mechanical pulping (CTMP),
(iii) the dense layer is bleached kraft pulp and
(iv) the top layer is coated by white pigments and binders (the pre-coated side).

Fig. 2 shows scanning electron microscope (SEM) images of paperboard: (a) shows the uncoated side (brown colour) and (b) shows the cross-section of the paperboard which illustrates four different layers (i, ii, iii and iv). As can be observed, the uncoated side of paperboard is very rough, containing large fibres. The rough surface of the uncoated side of paperboard is very advantageous for polymer coating mechanical adhesion.

2.2. Sample preparation

Before blending, bio-POSS was dried at 150 °C in vacuum in order to remove the hydrocarbon solvent completely. After that, the blends were prepared by melt mixing PE and dried bio-POSS at various compositions in a twin screw 15cc micro-extruder DSM