



Macromolecular Nanotechnology

Morphology studies and ac electrical property of low density polyethylene/octavinyl polyhedral oligomeric silsesquioxane composite dielectrics

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ABSTRACT

This paper presents the results of morphological and ac electrical investigations on low density polyethylene (LDPE) composites with octavinyl polyhedral oligomeric silsesquioxane (POSS). It has been shown that at low loadings, the frequency dependence of dielectric constant and dielectric loss for the LDPE/POSS composites showed unusual behaviors when compared with conventional (micro-sized particulates) composites. The ac breakdown strength was measured and statistical analysis was applied to the results to determine the effects of POSS loadings on the dielectric strength of LDPE. The morphological characterization showed that the presence of POSS additives apparently altered the supermolecular structure of LDPE and resulted in more homogeneous morphology when compared with the neat LDPE. The structure–property relationship was discussed and it was concluded that the final dielectric properties of the composites were determined not only by the incorporation of POSS additives but also by the supermolecular structure of LDPE. Rheological analyses of LDPE/POSS composite were also performed and the results showed that the octavinyl-POSS had good compatibility with LDPE.

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1. Introduction

In the past decades, the mixing of organic polymers with nano-fillers to form so-called nanocomposites with improved properties attracted considerable attentions [1–6]. One interest of the researchers is to seek the design rules that would allow the composites to be engineering materials with the desirable electrical properties. The purpose of this paper is to investigate the ac electrical properties of low density polyethylene (LDPE)/polyhedral oligomeric silsesquioxane (POSS) composites and to evaluate the possibilities of using LDPE/POSS composites as potential dielectrics or insulating materials in high voltage application.

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POSS is a class of cage-shaped molecules represented by the formula $(RSiO_{1.5})_n$ with an silica-like core (Si_6O_9 , Si_8O_{12} or $Si_{10}O_{15}$) surrounded by 6, 8 or 10 organic corner groups, which are becoming one class of versatile inorganic fillers for preparing polymer composites with desirable properties [7]. POSS can be dispersed in polymer matrix by copolymerization, chemical grafting or blending and consequently lead to some dramatically improved properties, such as, increase of thermal stability [8] and mechanical modulus [9], as well as reduction in dielectric constant [10], crystallization time [11] and flammability [12]. Although, POSS have been proved to be efficient to decrease the dielectric constant of some polymers, little is known about the dielectric properties of polymer/POSS composites. This paper constitutes a contribution to the understanding of the effects of POSS on the dielectric properties of LDPE. For materials used as dielectric applications, the dielectric strength is one of the properties that must be

taken into account in order to check the ability to withstand high electric field [13–15]. Therefore, we paid much attention to the investigations on dielectric breakdown strength of the composites.

Dielectric strength is defined as a relationship between the breakdown voltage and the sample thickness, representing the maximum field what the materials can withstand. There exist many factors influencing the ac dielectric strength of polymers [16,17]: environment conditions (temperature, moisture), applied voltage frequency, electrode conditions and the polymers themselves involving morphology, additives, defects and so on.

In the following a brief review is given of early work in which the dielectric strength of solid polymers has been related to various factors.

(i) Temperature: the temperature dependence of breakdown mechanisms for most thermoplastic polymers like LDPE has been extensively investigated and could be divided into three categories [18]: low temperature region, middle high temperature region and high temperature region. In low temperature region, the polymers show a glass-like state and the electron avalanche breakdown is supposed to be dominant, on the basis of the results of breakdown time lag, the thickness dependence of breakdown and the effects of impurity etc. The main mechanisms in middle high temperature region where the polymers show a rubber-like state, are considered to be thermal and free volume breakdown. In higher temperature region, the polymers are in melting or softening states and the breakdown characteristics were explained by thermal and electromechanical breakdown.

(ii) Applied voltage frequency [16,17]: charge carriers in practical polymers are trapped in localized electronic states and a higher frequency voltage can yield higher electrical conductivity resulting in lower breakdown strength when thermal breakdown takes place.

(iii) Structure and morphology: over the last three decades, many speculations have been put forward to explain the relationship between structure and morphology of semicrystalline polymers and their dielectric strength [6,19–25]. The majority of authors suppose that the damage of semicrystalline polymers in a high electric field is related to macroscopic inhomogeneity of structure [26]. However, clearly there exist some controversies in understanding the effects of the supermolecular structure on the breakdown strength: (i) it has been shown by Ceres and Schultz that the electrical lifetime of polypropylene decreased with increasing spherulite size [25]; (ii) results of Ishida and Okamoto from cross-linked polyethylene (XLPE) specimens indicated that the improvement in dielectric strength was probably due to the increase in spherulite size [21]; (iii) Vaughan et al. mentioned that no clear trends emerged to indicate that spherulite size variations were reflected in the short-term electrical failure processes of a series of LDPE systems [6]. In the case of the effects of lamellar thickness and crystallinity on the dielectric breakdown strength, however, it seemed that different authors have the similar conclusions: (i) a semicrystalline morphology consisting of extensive thick lamellae produces the highest dielectric strength results; (ii) high degree of crystallinity can result in improvement in dielectric strength of polyolefin.

(iv) Fillers: the introduction of fillers can significantly modify the dielectric strength of polymers depending on the electrical characteristic of fillers [15,27,28]. They appear in polymer matrix as a separate phase, and in this case the dielectric strength of the polymer is determined by the dielectric constant or electrical conductivity of the fillers because the local field distortion and enhancement around the fillers are caused by the difference in dielectric constant or electrical conductivity between the fillers and the base polymer.

(v) Defects like voids and pores: experiment [28,29] and simulation [30] results have confirmed that the breakdown strength of solid samples is related to the void introduced in the sample preparation process. The existence of voids in insulators produces local electrical failures, resulting in partial discharges and thereby reducing the electrical breakdown strength of the insulating material and the lifetime of the insulation apparatuses [29].

In a quasi-homogeneous field configuration, the dielectric breakdown of solid polymers is usually controlled by the presence of major flaws such as fillers and voids found within the polymer matrix.

The molecular structure of an octavinyl-POSS is shown in Fig. 1. Octavinyl-POSS are selected because that they contain C=C bonds, which can offer an opportunity to tailor the morphology and properties of LDPE/POSS composites by physical blending or reactive blending. In this work, we focused on the morphology and ac electrical properties of LDPE/octavinyl-POSS composites prepared with physical blending.

It was well understood from previous work that, for polymer/particle systems, the particle–particle interaction, particle–polymer interaction and the compatibility between polymer matrix and particles can be sensitively reflected by the rheological characterization [31]. On the other hand, polyethylene is a semicrystalline polymer which consists of crystallites and disordered regions between crystallites [32,33]. The electrical properties of the polyethylene could be significantly influenced by its semicrystalline nature and rheological characteristics. In addition, the incorporation of fillers can result in changes in degree of crystallinity, supermolecular structure and crystalline phases [33]. Therefore, the morphological, rheological and thermal characteristics were also investigated in order to understand the structure–dielectric property relationships for the composite systems.

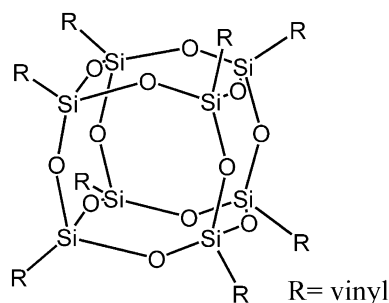


Fig. 1. Chemical structure of an octavinyl-POSS molecule.

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