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The influence of TiO₂ nanoparticle incorporation on surface potential decay of corona-resistant polyimide nanocomposite films

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1. Introduction

Polymer nanocomposites have attracted wide interest as a method of enhancing polymer properties and extending their applications. Surface potential decay has been used widely as a tool to monitor charge transport and trapping characteristics of insulating materials. Since the early 1960, there has been more interest in the surface potential decay of corona-charged polymeric materials in the open circuit configuration. The earlier surface potential decay research was on corona-charged polyethylene film which was given in some details by Ieda and co-workers [1]. One of the interesting phenomena in the observation of surface potential decay is the cross-over phenomenon, namely, the decay curves starting with different initial surface potentials cross each other during the decay process [2-5]. Over the years, many researchers have focused their studies on surface potential decay of coronacharged polymeric materials and various theories and models have been proposed to explain the cross-over phenomenon with various hypotheses such as field-dependant injection [6], charge

trapping/detrapping process [7] and bulk polarization [8].

ABSTRACT

PI nanocomposite films containing surface modified nanoparticles by employing silane coupling agent were prepared using in-situ dispersion polymerization process. The surface potential decay measurements on films were investigated over the different negative corona-charged voltages and times in a controlled environment where temperature and relative humidity were kept at 21 °C and 45%, respectively. There is a significant change in the surface potential decay characteristics after nano-fillers were introduced into polyimide. The surface potential decay pattern depends also on the amount of nano-fillers. The possible surface potential decay and corona resistance mechanisms responsible for the observed phenomena were discussed.

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Though many experimental results were presented so far, the physical mechanism of charge decay has not clearly been understood. leda et al. explained the decay curves only for low initial potentials using Ohm's law and predicted an exponentially surface potential decay [1,9]. Batra et al. assumed field-independent mobilities and demonstrated that the cross-over phenomenon is not associated with the finite depth of penetration [10]. Wintle et al. has developed theories that include field-independent mobilities of various forms as well as trapping [11].

Generally, due to clear characteristics of charge transporting and trapping in insulating materials under corona charging, the measurement of the surface potential decay has been proved to be a simple and useful technique. The technique involves with depositing charge on the surface of a polymer by means of a dc corona discharge and monitoring charge decay over a period of time using contactless probe. Many experiments on the charge transport in insulating polymers have been carried out using the surface potential decay method [12,13]. However, most of surface potential decay researches were on the single polymers such as polyethylene, epoxy, polypropylene and polyethylene terephthalate, little attention has been paid to surface potential decay in polymer nanocomposites [14–17].

In recent years, polyimide (PI) has received more and more attention in electrical and electronic industries due to its high thermal and chemical stability, good mechanical property and excellent electrical properties in a wide range of temperatures



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[18–21]. However, due to the rapid development of electrical engineering and electronic technology, it imposes more stringent requirements to the materials. The single tradition materials are facing more challenge, especially, they do not always withstand the damaging effects of corona, which can cause ionization and eventual breakdown of an insulation material or system when the voltage stress reaches to a critical level. To improve the insulating lifespan of high-voltage electrical equipment to resist the corona attack, there is strong interest in insulated polymer materials with good thermal stability and excellent corona resistance [22,23]. Due to the development of nano-technology and nano-materials, polymer nanocomposites are put forward as a kind of novel composite materials with improved insulating characteristics [24,25]. In this work, Polyimide/TiO₂ (PI/TiO₂) nanocomposite films containing surface modified nano-TiO₂ particles by employing silane coupling agent were prepared using in-situ dispersion polymerization process. Surface potential decay characteristics of both pure PI film and PI/TiO₂ nanocomposite films (one-layer) were investigated over the different negative corona charging voltages and corona charging times using a needle-grid-ground electrode system under a controlled environment with temperature and relative humidity at 21 °C and 45%, respectively. To investigate the influence of interface on surface potential decay, surface potential decays for four kinds of two-layer samples were also carried out. Results of the repeated experiments show that the potential decay is faster for higher initial potential and longer charging time. It has been found that there is a significant influence of nanoparticles on the surface potential decay characteristics of PI nanocomposite films. The surface potential decay curves depend also on the amount of nano-TiO₂ particles. The schematic depiction of the possible charge transporting in different layer samples were presented.

2. Experiment details

2.1. Fabrication of the polyimide nanocomposite films

Both pure PI films and PI/TiO₂ nanocomposite films were prepared using in-situ dispersion polymerization process in the laboratory. Prior to use, the surface of the nano-TiO₂ particles with a mean diameter of ~ 50 nm was treated with γ -aminopropyltriethoxy silane (KH550) as a coupling agent in order to disperse the nano-TiO₂ particles into PI matrix homogeneously. The PI/TiO₂ nanocomposite films were prepared by addition of the nano-TiO₂ particles (from 5 to 15 wt.%) into N, N-dimethylacetamide (DMAc) with the monomers of pyromellitic dianhydride (PMDA) and 4, 4'-oxy dianiline (ODA). Then the mixture was cast onto a cleaned glass plate and was held in the vacuum oven at room temperature to let the trapped air escape. After polyamid acid (PAA) was converted to PI by the thermal imidization, the PI/inorganic nanocomposite films were obtained. As a control experiment, the pure PI film was also prepared employing the same process. The thickness of all the films was close to 70 µm.

2.2. Corona charging setup and potential decay measurement

A schematic experimental setup in a controlled environment with temperature and relative humidity at 21 °C and 45%, respectively, is shown in Fig. 1a.

The samples were negatively charged on their free surface using a typical needle-grid-ground corona charging setup which consists of a high-voltage needle electrode, a wire mesh grid electrode and a rotatable earthed electrode. The needle electrode which connected to a negative dc high-voltage supply was situated above the grounded metallic plate. The grid electrode which was connected to a different negative dc high-voltage supply was situated between the needle electrode and the grounded plate. The distance between the needle and the mesh and between the mesh and the ground plate was 4.5 cm and 1.5 cm, respectively. The area of the mesh and the sample was 150 cm² and 25 cm², respectively. In this way, a suitable electric field could be generated in the gap between the grid and the surface of the sample. After negative corona charging, the sample was quickly moved with the rotating system toward a compact JCI 140 static monitor to observe the surface potential decay. The duration of moving the rotating system is around 2 s. However, the readings from the JCI 140 static monitor are not the direct values of the surface potential of the samples, a calibration should be made. A film with a thin gold coated is connected to a dc voltage. Readings from the JCI 140 static monitor are taken when varying the applied voltage. A liner relationship was obtained between the readings and the applied voltage as shown in Fig. 1b. According the equation inset in the Fig. 1b, the reading from the JCI 140 static monitor can be converted into the values of surface potential.

3. Results and discussion

Negative polarity was initially applied to corona charging electrode and the absolute value was used in the present paper when surface potential and time characteristics were plotted.

3.1. Surface potential decay of one-layer films

Influence of different corona charging voltages and charging times on surface potential decay characteristic of one-layer films were studied.



Fig. 1. (a) A schematic image of the experimental setup used for corona charging and surface potential measurement, (b) Calibration for the JCI 140 static monitor.

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