

DIELECTRIC RESPONSE OF POLYETHYLENE LOADED WITH SiO₂ NANOPARTICLES

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ABSTRACT

The incorporation of metallic oxide nano-particles in a polymeric matrix has been shown to improve the material's dielectric performances. In this work, the dielectric response of polyethylene based samples filled with nanosilica particles was investigated. These measurements were carried out for a broad frequency range at different temperatures and after different thermal treatments and compounding procedures. Frequency domain dielectric spectroscopy was found to be a power tool to assess the electrical properties of the interfacial layer between the nanoparticles and the polymeric matrix. In particular, it was shown that the conductivity of this layer, which is significantly higher than the bulk conductivity of both the matrix and the filler, has a large impact on the material dielectric response, and particularly at the power frequency.

KEYWORDS

Nanodielectrics, Dielectric Response, Interfacial layer.

INTRODUCTION

Nanoparticle-filled polymers have been known to provide advantages over micron-filled or unfilled polymers because they improve endurance and resistance to degradation. This has been shown to be true for the case of polyethylene loaded with metallic oxide nanoparticles such as SiO₂ [1] or Al₂O₃ [2]. The addition of nanoparticles has also been shown to cause significant changes in the dielectric response of the composite material and most of the time these changes are closely related to the dielectric properties of the interaction zones between the particles and the matrix and not so much to the bulk dielectric properties of the filling material [3].

For classic micrometric composites, most of the dielectric data provided in the literature for non-conductive fillers, such as metallic oxides, in a non-conductive polymeric matrix, will feature, in addition to the relaxation mechanisms proper to each phase, an interfacial polarization peak, known as Maxwell-Wagner-Sillars polarization, and an increase of both the real and in imaginary parts of the complex permittivity as the frequency is lowered. Another universal observation for ordinary composites is that none of the existing mixing laws [4, 5] can fit the experimental data on a broad range of frequencies [6, 7]. For hydrophilic fillers, the existence of absorbed water in materials and particularly in the interfacial region between the filler and the polymer matrix is almost inevitable and will invariably affect the composite dielectric response, often leading to a interfacial relaxation peak at much higher frequency than what would be expected based on values of the filler bulk conductivity. This effect is expected to be even more pronounced for nanocomposites as the volume fraction of interfaces is much higher.

In this work, the dielectric response of polyethylene based samples filled with nano-silica particles was investigated.

These measurements were carried out for a broad frequency range at different temperatures and after different thermal treatments and compounding procedures. The experimental data clearly showed the hydrophilic and unstable nature of the interface.

SAMPLE PREPARATION

Nanocomposites with 2 and 4 wt% of SiO₂-nanoparticles were prepared using extrusion melt compounding as a dispersive method using a twin screw extruder Rheomix 600. Two different suppliers provided the nanoparticles. 15 nm SiO₂ spherical nanoparticles, labelled SiO₂-1, were supplied by Nissan Chemical Industry in the form of a colloidal solution of isopropanol. The size and the spherical nature of these particles were assessed by a direct observation of the particles by the means of an atomic force microscope (AFM) [8]. SiO₂ spherical nanoparticles of 14 nm, labelled SiO₂-2, were also supplied by Degussa in the form of a powder. Two different PE resins without any additives were provided by DOW Chemical in the form of a powder. The first one, labelled PE-1, has a melting temperature, as measured from differential scanning calorimetry (DSC), of 130°C, a density of 0.95 g/cm³ and a melt-flow index of 0.35 g/min. The second one, labelled PE-2, has a melting temperature of 110°C, a density of 0.91 g/cm³ and a melt-flow index of 0.46 g/min.

Two different types of nanocomposites (NC) were compounded. NC-1 was a mixture of PE-1 and SiO₂-1 while NC-2 was a mixture of PE-2 and SiO₂-2. In both cases, the polyethylene powder and nanoparticles were mechanically premixed for 15 min before their introduction into the extruder. The mixture was fed into the extruder and the compounding was conducted using a rotating speed of 100 rpm and a temperature profile of 200°C, 220°C, 250°C, 250°C, 250°C, 220°C, and 200°C in the sequential heating zones from the hopper to the die. Temperature and pressure were continuously recorded during compounding. After compounding, the material was extruded from a die that had a cylindrical nozzle of 4 mm in diameter in order to produce cylindrical extrudate. The extrudate was then cooled at room temperature and pelletized with an adjustable rotating knife located after the water bath into 5 mm long cylinders. In order to improve the dispersion of the particles, a second run into the extruder was used for NC-2. Infrared measurements were conducted on the various samples obtained by this procedure and no noticeable oxidation occurring during processing was observed. Pellets obtained after extrusion were then hot pressed at 160°C for 5 min under 5 MPa into plates of various thickness between 500 µm and 1 mm.

DIELECTRIC RESPONSE

The dielectric spectroscopy measurements were performed on plane-plane samples using a broadband dielectric spectrometer and a controlled oven. The