Effect of flyash addition on dielectric properties of polypropylene

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Dielectric properties of flyash filled polypropylene have been measured at different frequencies, and temperatures ranging from ambient to 160°C. It has been found that the dielectric constant of flyash filled and unfilled polypropylene decreases with increasing frequency up to 100 kHz. The dissipation factor of pure polypropylene decreases with increasing frequency. Dielectric constant of flyash filled polypropylene increases as compared to pure polypropylene. With increasing temperature, dielectric constant (ε′) and dissipation factor (tan δ) of flyash filled and unfilled polypropylene have been measured at all frequencies between 4-100 kHz. The β' peak position has been found to shift towards higher temperature side for flyash filled polypropylene with increase in frequency. It has also been found that incorporation of flyash in polypropylene shifts the α peak towards the higher temperature side.

Polypropylene (PP) is becoming increasingly popular for a wide variety of applications in automobiles, electric appliances, containers, capacitor films, indoor and outdoor carpets. It has good resistance to heat and chemicals. It has negligible water absorption and excellent electrical properties. The compound has very high volume electrical resistivity of the order of the 10^15 ohm/cm and its dielectric strength is 10-22 kV/mm. Polypropylene films have been widely used as dielectric materials in oil impregnated power capacitors because of their low dielectric loss.

Large number of fibres and fillers are incorporated in pure polypropylene to improve its impact, flow and mechanical properties and to reduce the cost. Mica is incorporated in polypropylene to increase its flexural strength and heat deflection temperature. It is reported that incorporation of mica in polypropylene improves the electrical properties, dimensional stability, and flame retardancy. Gupta et al. have made an attempt to identify the energy absorbing mechanism in a stress strain test on short glass fibre reinforced polypropylene composites. Sato et al. have studied the tensile fractured fibre composites and found that the fibre firstly gets separated from matrix and then interfacial cracks occur. Chand and Gautam have developed composites of flyash and glass fibre with polyester resin and reported their improved structural and thermal stability.

In the present studies, flyash waste filler is incorporated in polypropylene in different weight fractions. Dielectric properties such as dielectric constant (ε′) and dissipation factor (tan δ) of these composites have been determined at different temperatures and frequencies.

Materials and Methods

Materials

The commercial grade isotactic polypropylene was supplied by M/s Shri Ramdev Plastics, India. Flyash particles of size less than 45 microns were used in the studies. The main chemical constituents of fly ash were: SiO₂ (65.1%), Al₂O₃ (25.1%), Fe₂O₃ (4.2%), CaO (1.4%) and other metal oxides (4.2%).

Different amounts of flyash (5 to 20 wt%) were incorporated in polypropylene matrix. Composition of flyash filled polypropylene (PP/FA) used in the investigations was: 100/00; 95/05; 85/15; 80/20.

Mixing of polypropylene and flyash

Weighed amounts of PP and flyash (FA) were mixed in the air-circulating oven. These materials were mixed for 10 min using the mixing method reported in our previous paper at 280°C on a single screw extruder, rotating at 20 rpm.

Compression molding

Sheets of 2 mm thickness were made on a compression-molding machine. Composite samples were pressed at 190°C.

Dielectric sample preparation

Circular samples of 10 mm diameter were cut from the above sheets.
Coating

Graphite type air-drying conducting paint was applied using a brush on both the sides of the circular pallets.

Dielectric Measurements

$\varepsilon'$ and tan $\delta$ of the composites were determined by measuring capacitance ($C$) and dissipation factor ($D$) using Hewlett-Packard LCR meter model number 4274A. Measurements were made for determining $C$ and $D$ for filled and unfilled PP in the temperature range ambient to 160°C. Heating rate of the furnace was kept constant at 1°C/min.

Results and Discussion

The dielectric loss $\varepsilon''$ of non-polar polymers such as PP is generally ascribed to impurities, oscillation of C-CH$_3$ side groups about the chain backbone, or dipoles arising from the antioxidant. Among these various factors, impurities such as residual catalyst and antioxidant have been reported to affect the dielectric loss.$^{15}$

Fig. 1a shows the variation of dielectric constant ($\varepsilon'$) with temperature for unfilled polypropylene at different frequencies. It has been found that increase in temperature increases the magnitude of $\varepsilon'$. Fig. 1b shows that the dissipation factor (tan $\delta$) of unfilled PP decreases with increasing frequency when plotted against increasing temperature. Dissipation factor was maximum at 4 kHz frequency and minimum at 100 kHz. In the tan $\delta$ vs temperature plot, $\beta'$-peak appears around 50°C which is similar to the previously reported $\beta'$-peak due to antioxidant present in PP$^1$. Another peak appears around 107°C which corresponds to $\alpha$-relaxation and may be due to the C-CH$_3$ group present in the crystalline region.

Figs 2a, 3a and 4a show the variation of dielectric constant ($\varepsilon'$) with temperature between 30-160°C for PP/FA compositions having weight fraction ratio 95:05, 85:15 and 80:20. It has been observed that increase in frequency decreases the magnitude of dielectric constant ($\varepsilon'$); and increase in flyash content increases the dielectric constant. Also,
increase in temperature increases dielectric constant ($\varepsilon''$), which is similar to other filled polymers.

Figs 2b, 3b, and 4b show the variation of dissipation factor (tan $\delta$) with temperature for the PP/FA compositions (95:05, 85:15 and 80:20). The $\beta''$-peak has been found to shift to higher temperature side with increase in frequency. These results show that the filler shifts $\beta''$-peak. It can be explained on the basis of the results reported in literature that impurities affect the tan $\delta$ peak in PP. Another peak ($\alpha$) shifts to higher temperature side with change of flyash ratio (5, 15, and 20 wt%). At 4 kHz, the peak has been found to shift from 117, 120 and 121°C respectively for 5, 15 and 20 wt% flyash filled PP compositions as compared to pure PP.

Fig. 3(a) — Variation of $\varepsilon''$ with temperature in PP/FA (85/15); (b) Variation of tan $\delta$ with temperature in PP/FA (85/15); [4 kHz (ϕ); 10 kHz (Δ); 20 kHz (○); 100 kHz (□)]

Fig. 4(a) — Variation of $\varepsilon''$ with temperature in PP/FA (80/20); (b) Variation of tan $\delta$ with temperature in PP/FA (80/20); [4 kHz (ϕ); 10 kHz (Δ); 20 kHz (○); 100 kHz (□)]

Fig. 5 — Variation of log $f$ (Hz) vs 1000/T (K) for $\beta'$ and $\alpha$ peaks for PP (○) and PP/FA compositions (95:05 (ϕ); 85:15 (Δ); 80:20 (Δ))
Fig. 5 shows the plots for log of frequency vs reciprocal of absolute temperature which are linear for \( \alpha_c \) (crystalline) and \( \beta' \) (antioxidant) peaks. Slope of the straight line for \( \beta' \)-peak of flyash filled PP samples has been found to change as compared to pure PP showing modification in relaxation time. Peak position of the \( \beta' \)-peak in pure polypropylene has been found to shift towards the higher temperature side with the increase in frequency. For flyash filled polypropylene also, \( \beta' \)-peak has been found to shift towards higher temperature side with increasing frequency. Also, the \( \alpha_c \) crystalline peak for different weight fractions of flyash filled polypropylene is affected more with the increase in frequency as compared to pure PP. This is similar to the findings on salt-filled Nylon-6\(^4\) explained on the basis of lower crystallite degree of the salted samples as compared to pure polymer regarding: on increasing the temperature, tan \( \delta \) curve of Nylon-6 goes up faster at higher temperatures and the dielectric constant \( (\varepsilon') \) for the salt/nylon-6 mixture becomes higher than that of almost amorphous state of the salted samples.

Conclusions

Incorporation of FA in polypropylene increases the dielectric constant of PP. An increasing of temperature, dielectric constant of all PP samples (filled and unfilled) increases at all frequencies. Upon incorporation of FA in polypropylene, \( \beta' \)-peak is shifted to the higher temperature side at all the frequencies. Two types of relaxation peaks (\( \beta' \) and \( \alpha_c \)) are observed in unfilled and FA filled PP compositions.

References