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Volume and surface resistivity of low-density polyethylene filled with stainless steel fibres

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D.c. electrical properties of low-density polyethylene filled with stainless steel fibres have been studied at various concentrations above the percolation threshold. The volume fractions used were 2, 5 and 10% of stainless steel fibre. The volume resistivity of polyethylene varied between 6.15×10^7 Ω cm at 2% loading level and 3.71×10^5 Ω cm at 10%. Corresponding values for surface resistivity varied between 2.77×10^7 Ω at 2% and 3.94×10^4 Ω at 10%. The value of the critical exponent for percolation was estimated to be around 2.4 for volume resistivity and 3 for surface resistivity.

1. Introduction

There is considerable interest in conductive polymers because of their potential usefulness in applications like shielding against electromagnetic/radio-frequency interference (EMI/RFI), electrostatic discharge (ESD) and as heating elements for moderate temperatures [1, 2].

Polymers in general are good insulators with volume resistivities ranging from 10^{15} Ω cm to 10^{22} Ω cm, with polyethylene residing at the high end [3]. One method of rendering an insulating polymer conductive is to blend it with a conductive filler. Fillers in the form of powders, fibres or flakes can be chosen from different conductive materials including carbon black, graphite, copper, aluminium, nickel and stainless steel [1, 2, 4]. There are two primary factors that determine the degree of conductivity of the composite material:

(i) volume fraction of the filler and
(ii) geometrical properties of the filler, shape, aspect ratio and size distribution.

It is well known from the theory of percolation that the volume fraction of the filler necessary to achieve a certain targeted conductivity value decreases with increasing aspect ratio of the fibres embedded in the matrix [4]. Obviously, it is not sufficient to start with the correct aspect ratio since the average aspect ratio of the filler might degrade during processing. Thus, it is also desirable for the conductive fillers to maintain their high aspect ratios under the intensive conditions of the mixing process. Stainless steel fibres satisfy both of these requirements. They are readily available with average lengths of 5 mm and average diameters of 0.005 mm, yielding a mean aspect ratio of about 1000 [5]. At this high aspect ratio the threshold volume fraction needed to induce bulk conductivity is about 1% [4]. In processing studies where the attrition behaviour of stainless steel has been compared with that of other fillers like graphite fibres or metal-coated glass fibres, the aspect ratios of stainless steel fibres were shown to be better preserved than the others [4].

A survey of the literature shows that stainless steel fibres have been introduced into various binders like polypropylene [6], acetylonitrile–butadiene–styrene [7], nylon 6/6, polyphenylene sulphide and polycarbonate [4]. The objective of this study is to study the d.c. electrical properties, namely the volume and surface resistivity, of stainless steel fibres in a low-density polyethylene (LDPE) matrix. It was also anticipated that the results would provide additional experimental data to compare with the theoretical considerations and numerical simulations pertaining to percolation phenomena in two and three dimensions.

2. Experimental procedure

2.1. Materials

The low-density polyethylene resin used was Petrothene PEV-007 obtained from USI Chemicals, Cincinnati, Ohio. The stainless steel fibres were procured from Bekaert, Marietta, Georgia (Beki-Shield product code 75/6/C10).

The suspension was prepared by mixing the ingredients in a Haake Rheocord batch mixer, model 40. The mixing was carried out at 150 °C at 60 r.p.m. Three different volume loading levels of 2, 5 and 10% stainless steel fibre were incorporated. Three different samples were prepared for each concentration to verify the reproducibility of the measurements. The samples were compression-moulded using a Carver Glove Box press at 150 °C and under a pressure of 1910 kPa. The resulting samples were disc-shaped with a thickness of 0.3 cm and a diameter of 8.25 cm.

2.2. Apparatus

For the measurement of volume and surface resistivity, ASTM standard D257 (d.c. Resistance or Conductance of Insulating Materials) [8] was followed. The experimental set-up used is shown in Fig. 1. A d.c. voltage is applied to the sample, which is placed inside a Keithley resistivity adapter, model 6105. The voltage

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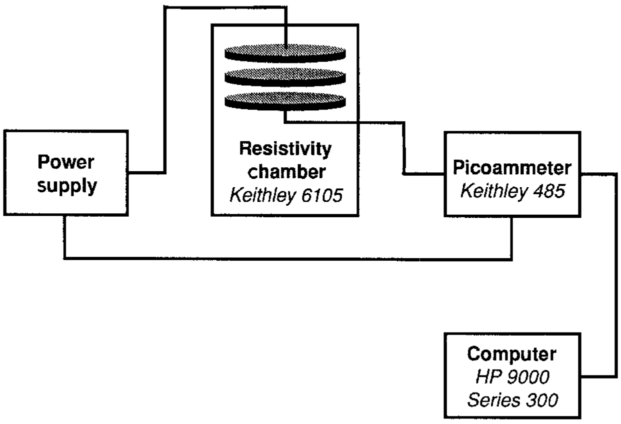

Figure 1 Volume and surface resistivity measurement set-up.

applied to the sample is measured with a Fluke 37 digital multimeter and the current flowing through the sample is measured with a Keithley picoammeter, model 485. The picoammeter is equipped with an IEEE 488 interface through which it is connected to a Hewlett-Packard 9000, Series 300, computer. The collection and statistical analysis of the data are automated.

The resistivity adapter has disc-shaped electrodes. The top electrode applies a 1 lb (0.454 kg) constant force to the sample. The bottom electrode is spring-loaded. This arrangement ensures uniform pressure over the measuring area. The bottom electrode includes a guard ring which eliminates edge effects. The metal enclosure around the electrodes shields against stray pick-up. Dimensions of the electrodes and the configuration of electrical connections for volume and surface resistivities are shown in Figs 2 and 3, respectively.

The volume and surface resistivity values are determined in accordance with the ASTM standard [8] as follows:

$$\text{Volume resistivity } \rho_v = AV/It\,\Omega\,\text{cm} \qquad (1)$$

where V is the applied voltage in volts, I is the measured current in amperes, A is the effective area of the guarded electrode in cm^2, and t is the thickness of the sample in cm;

$$\text{Surface resistivity } \sigma_s = PV/Ig\,\Omega \qquad (2)$$

where P is the effective perimeter of the guarded electrode and g is the distance between the electrodes.

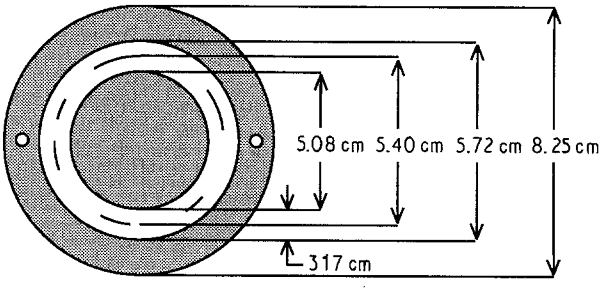

Figure 2 Dimensions of model 6105 electrodes.

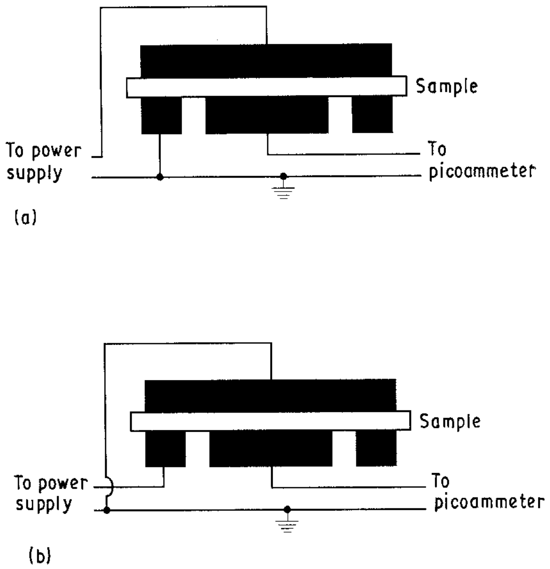

Figure 3 Electrical connections to the electrodes: (a) volume resistivity, (b) surface resistivity.

2.3. Measurement procedures

2.3.1. Sample preconditioning

The electrical properties of insulating materials are influenced by humidity. To obtain reproducible test results, samples were preconditioned as recommended in the ASTM standard D618 (Conditioning Plastics and Electrical Insulating Materials for Testing [9]). Each sample was kept in a dessicator over anhydrous calcium chloride for 48 h before measurements. Surfaces of the samples and the electrodes were cleaned with dehydrated alcohol. The samples were handled with acetate rayon gloves.

2.3.2. Time of electrification

When a d.c. voltage is applied across a polymer specimen, current starts to flow. However, the current does not remain constant. It usually decreases with time [8]. This phenomenon is due to the polarization of charges at the interfaces and the sweep of mobile ions in the sample. Therefore, a standard has been established by ASTM to measure the current only after a certain time period, called the time of electrification, is elapsed. This time is usually taken as 60 s. In the measurement of the samples prepared for this study this standard was observed. The value of the current was measured only after the elapsing of 1 min.

2.3.3. Interfacial charges

It is necessary to eliminate the influence of the interfacial charges before the sample is tested again [9]. This is accomplished by removing the voltage source and shorting the electrodes for a duration of about four times the time of electrification. This precaution was observed and the samples were allowed to neutralize for a duration of 4 min between measurements.

3. Results and discussion

Results for the volume and surface resistivity measurements are shown in Tables I and II respectively. The resistivity of LDPE changes dramatically upon

TABLE I Volume resistivity versus volume percentage of stainless steel fibres in LDPE

Fibre content (vol %)	Sample No.	Volume resistivity (Ω cm)
2	1	6.91×10^7
	2	6.07×10^7
	3	5.46×10^7
	Average	6.15×10^7
5	1	1.20×10^6
	2	1.39×10^6
	3	0.95×10^6
	Average	1.18×10^6
10	1	3.90×10^5
	2	4.38×10^5
	3	2.86×10^5
	Average	3.71×10^5

TABLE II Surface resistivity versus volume percentage of stainless steel fibres in LDPE

Fibre content (vol %)	Sample No.	Surface resistivity (Ω)
2	1	0.58×10^7
	2	7.09×10^7
	3	0.63×10^7
	Average	2.77×10^7
5	1	3.20×10^5
	2	5.17×10^5
	3	0.87×10^5
	Average	3.08×10^5
10	1	4.14×10^4
	2	3.04×10^4
	3	4.65×10^4
	Average	3.94×10^4

the addition of only 2% by volume of stainless steel fibre. The resistivity values decrease further with increasing volume concentrations of stainless steel fibres. The observed decrease is more pronounced for the surface resistivity values.

The surface resistivity and volume resistivity of stainless steel fibre-reinforced LDPE serve as good examples of percolation phenomena in two and three dimensions, respectively. The concept of percolation was first investigated by Broadbent and Hammersley [10]. They introduced the notion of percolation probability which refers to the availability of zones of the medium which are "sufficiently" connected to the rest for conduction. These conducting regions possess dimensions which are significantly greater than the mean free path of the electrons of the conductive metal fillers. An example of a conduction threshold is the report of Gurland [11] which suggests that at around 30% by volume of silver, Bakelite powder exhibits a pronounced threshold. It is known that the value of the concentration threshold is very sensitive to the mixing, processing and compaction conditions employed [12]. Experimental results summarized in the previous paragraph agree qualitatively with the expectations of percolation theory, where a sharp drop in resistivity is observed at a volume concentration slightly above the reported 1% by volume for fibrous conductive fillers with aspect ratios of about 1000. The conjectured power-law dependence of resistivity on volume fraction beyond the percolation threshold is given as [13]

$$\rho_v = \rho_{v0}\,(f - f_{cv})^{-t_v} \qquad (3)$$

$$\sigma_s = \sigma_{s0}\,(f - f_{cs})^{-t_s} \qquad (4)$$

where f is the volume fraction of the stainless steel fibre and f_{cv}, f_{cs} are percolation thresholds for volume and surface resistivities, respectively; t_v, t_s are the critical exponents for volume and surface resistivities, respectively, and ρ_{v0} and σ_{s0} are the prefactors for volume and surface resistivity values, respectively.

Assuming that the percolation threshold is around 1% by volume, our results given in Tables I and II generate a power-law exponent (i.e. critical exponent) t of 2.4 for volume resistivity and 3 for surface resistivity data. These t values generally agree with the Monte Carlo simulation-based critical exponent determined by Kirkpatrick [14]. It is interesting to note that the critical exponent t assumes a higher value for surface resistivity data than the t value based on the volume resistivity data.

4. Conclusions

D.c. electrical properties of LDPE filled with stainless steel fibers have been studied. The volume and surface resistivity of LDPE decreased by more than ten orders of magnitude by the addition of 2% by volume of stainless steel fibres. The resistivity of the composite continued to decrease with further increases in the volume percentage of fibres. The best fit of the experimental results to the numerically expected power-law type of equations generated critical exponents for percolation in agreement with the literature values which were generated by Monte Carlo simulations. The critical exponent was determined to be higher for surface resistivity than for volume resistivity.

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