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Dielectric properties of conducting polymer composites at microwave frequencies

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The dielectric properties of conducting polymer composites containing polypyrrole (PPy) crushed films, PPy powder, polyaniline (PAn) base and acid powders as the dispersants and silicone rubber and vinyl ester as matrix materials have been investigated in the frequency range 2–18 GHz. The dielectric parameters such as the real part, ε', and imaginary part, ε'', of the permittivity and loss tangent, $\tan\delta$, increase with increasing conductivity and concentration of the dispersant. The geometrical shape of the dispersant governs the ability of conductive network formation which is indicated by a large drop in the resistivity of the composite. Also, dispersant/matrix interactions and physical properties of the matrix influence the agglomeration of the dispersant phase which, in turn, affects the dielectric properties of the composites. Flakes of PPy obtained by crushing highly conductive films and large PAn powder aggregates were unable to form a conducting network. The composites without a network of dispersant exhibit low dielectric parameters. On the other hand, high values of $\tan\delta$ ranging from 0.7–1.1 were achieved for the PPy powder (15 parts)/silicone rubber composites where a conducting network was observed.

1. Introduction

Considerable effort has been focused on applied research in the field of conducting polymers to turn these materials into useful products. This is mainly because conducting polymers possess interesting inherent electrical and optical properties which can be utilized to develop novel microelectronic and optical devices. Although many related potential applications such as Schottky junction, electrostatic charge protection, electromagnetic interference (EMI) shielding and non-linear optical materials have been proposed, a fundamental understanding of optical and dielectric properties of these materials is still lacking.

The dielectric properties of polymers at microwave frequencies have been extensively investigated over the years and summarized in Bur's excellent review [1]. With respect to conducting polymers, the dielectric behaviour of polyacetylene was first investigated by Feldblum *et al.* [2] in search of a lightweight organic microwave absorber. Since then a number of studies of dielectric properties of conducting polymers have been reported [3–7]. It is important to examine the dielectric parameters at a suitable range of frequencies, both from the viewpoint of applications as well as to provide a better understanding of electronic properties and response of the materials to electromagnetic radiation. Recently, the dielectric behaviour of polypyrrole (PPy) doped with tosylate anions was investigated by Hasegawa *et al.* [3] in the range 20–35 000 cm^{-1} and by Phillips *et al.* [4] at far infrared and microwave frequencies. The values of the real and imaginary parts of the complex permittivity were determined to be of the order of 10^4 for fully doped PPy at microwave frequencies [4]. Using impedance spectroscopy, van de Leur and de Ruiter [5] measured the dielectric properties of poly(3-*n*-hexylthiophene) powder over a wide range of temperature and frequencies between 0.01 Hz and 65 kHz. Javadi *et al.* [6] found that the loss tangent of protonated emeraldine polyaniline (PAn) at 6.5 GHz increases from 0.8 to 2.3 with degree of protonation. Recently, Unsworth *et al.* [7] investigated the microwave transmission, reflection and dielectric properties of semiconducting and conducting PPy. They indicated that the real and imaginary parts of the permittivity increased with doping level.

Conductive carbon black composites have been extensively researched in many respects including electrical conduction properties and dielectric behaviour [8–14]. Although it is believed that conducting polymers are promising complementary or alternative materials to carbon black, data on dielectric properties of the composites containing conducting polymers are scarce. In the present study, the dielectric parameters of conducting polymer composites, of which the matrix was either an elastomer or a vinyl ester, were determined at microwave frequencies by analysis of reflection/transmission intensities at normal incidence using a network analyser. The effect of matrix materials and the morphology, concentration and

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conductivity of the dispersant phase on the dielectric behaviour of the composites are discussed.

2. Experimental procedure

2.1. Preparation of polypyrrole powder

The conducting PPy powder was prepared by oxidizing pyrrole monomer with ferric chloride. Anhydrous ferric chloride, $FeCl_3$ (40 g), was added to chilled distilled water (100 ml). Freshly distilled pyrrole (6.4 g) was added slowly under constant stirring while the PPy precipitate formed. The precipitate was filtered from the mixture and washed with water and then dried under vacuum. The dried precipitate was ground into a powder.

2.2. Preparation of polypyrrole film flakes

The PPy films were prepared by electropolymerization in an aqueous solution. Sodium *p*-toluene sulphonate (*p*-TS) and sodium dodecylsulphate (DDS) were used as the dopant. The polymerization solution was freshly distilled pyrrole (0.1 M) and dopant (0.1 M) in distilled water. Stainless steel electrodes were used and the films were grown under a nitrogen blanket for 2 h at a current density of 2.8 mA cm^{-2}. Some *p*-TS doped PPy (PPy/*p*-TS) films were dedoped for 1 h by reversing the polarity of the potentiostat. The films were washed in acetonitrile/water (1:1) to remove excess dopant. After drying, the films were crushed into flakes in a hammer mill for 15 min under liquid nitrogen.

2.3. Preparation of polyaniline acid powder [15]

The conducting PAn acid powder (protonated emeraldine form of PAn) was prepared by oxidizing aniline monomer with ammonium persulphate, $(NH_4)_2S_2O_3$, to give an emeraldine salt. Freshly distilled aniline (20 ml) was added to 300 ml 1 M hydrochloric acid (HCl). $(NH_4)_2S_2O_3$ (11.5 g) was dissolved in 200 ml 1 M HCl. This solution was then added slowly to the aniline solution and stirred for 1.5 h in an ice bath. The PAn acid precipitate was collected and washed with 1 M HCl solution until the residue solution was clear. The precipitate was dried under vacuum and ground into powder.

2.4. Preparation of polyaniline base powder [15]

The non-conducting PAn base powder (emeraldine base form of PAn) was prepared from the PAn acid powder. 500 ml 0.1 M ammonium hydroxide, NH_4OH, solution was added to the moist PAn acid precipitate. The pH of the solution was maintained at 8. The solution was left stirring for 15 h before being collected and washed with 0.1 M NH_4OH. The precipitate was dried under vacuum and ground into powder.

2.5. Preparation of composite samples

Silicone rubber (Dow Corning 3110 RTV) and vinyl ester (Derakane® 8084) were used as the matrices. The dispersant (powder or flakes) was mixed thoroughly into the matrix material and cured at room temperature (the silicone rubber was cured by the supplied catalyst and the vinyl ester by 1.5% methyl ethyl ketone peroxide and 0.5% a cobalt salt accelerator). The concentration of the dispersant was based on parts per 100 parts by weight of the matrix.

2.6. Density and conductivity measurements

Density was obtained by weight and volume of the samples. The volume of the film was obtained from a 5 × 5 cm^2 sample and the film thickness was measured by a travelling microscope with an accuracy of 5%. Density and conductivity of the polymer powders were obtained from pressed pellets. The conductivity of the films and compacted powders (compacting pressure 7 t cm^{-2}) were measured by the four-probe method and the resistivity of the composites by the two-probe method.

2.7. Dielectric measurements

A Hewlett–Packard network analyser (HP 8510) was employed to determine the reflection parameter, S_{11}, and transmission parameter, S_{21}. The real part, ε', and the imaginary part, ε'', of the permittivity, ε^*, were calculated from S_{11} and S_{21} values according to the procedure described in the Hewlett–Packard Product Note [16]. Samples were moulded or fabricated into a "doughnut" shape to fit closely into a coaxial measurement cell. Teflon was used as a standard sample (5 mm thick) and was tightly fit into the coaxial cell. The dielectric measurements were performed on samples only when ε' and ε'' of the Teflon standard agreed with the data shown in the Hewlett–Packard Product Note [16]. The measuring frequency range was 2–18 GHz.

3. Results and discussion

3.1. Effect of dispersant geometrical shape

Table I shows conductivity and density of the dispersants and matrix materials. The permittivity, ε^*, is defined by

$$\varepsilon^* = \varepsilon' - j\varepsilon'' \qquad (1)$$

TABLE I Properties of the matrices and dispersants

Material	Conductivity ($\Omega^{-1}m^{-1}$)	Density (g cm^{-3})
PPy powder	80	1.4
PPy/*p*-TS (fully doped film)	1500	1.6
PPy/*p*-TS (dedoped film)	30	1.6
PPy/DDS	200	1.2
PAn acid powder	4	1.4
PAn base powder	Insulating	1.4
Silicone rubber	–	1.2
Vinyl ester	–	1.2

The imaginary part, ε'', is expressed by

$$\omega\varepsilon_0\varepsilon'' = \sigma(\omega)_m - \sigma \qquad (2)$$

where ω is the angular frequency, ε_0 is the permittivity of free space, $\sigma(\omega)_m$ is the a.c. conductivity and σ is the d.c. conductivity. The terms "conductivity" and "d.c. conductivity" are used interchangeably in this study and "a.c. conductivity" is specifically identified. The loss tangent, $\tan\delta$, is defined as the ratio of the imaginary to the real parts ($\varepsilon''/\varepsilon'$). Fig. 1 shows the values of ε' and ε'' as functions of frequency for the PPy powder/silicone rubber composites at various powder concentrations. The silicone rubber and vinyl ester matrices exhibit similar ε' and ε'' which are approximately 2.9 and 0.1, respectively, over the measured frequency of 2–18 GHz. The values of ε' and ε'' of samples with PPy concentration below 4 parts are independent of frequency. At higher concentrations (> 10 parts), ε' and ε'' decrease slowly with increasing frequency. A similar tendency is also observed in PPy powder/vinyl ester composites of this study and ferrite/polystyrene composites [17]. However, in the PPy flakes/silicone rubber composites, ε' and ε'' are practically independent of frequency at concentrations of dispersant as high as 75 parts. This reflects a weak interaction among the PPy flakes which will be discussed below.

Fig. 2 shows the values of ε' and ε'' for the PPy powder/silicone rubber and PPy flakes/silicone rubber composites at 10 GHz. The loss tangent, $\tan\delta$, of the PPy powder and flakes composites is plotted as a function of concentration (Fig. 3) and of frequency (Fig. 4). From Figs 2–4, the values of ε', ε'' and $\tan\delta$ are higher for the powder composites, although the powder is much less conductive than the film (Table I).

Fig. 5 indicates that at 5 parts of PPy powder in a silicone rubber matrix, the resistivity of the composite begins to decrease sharply. This conductivity behaviour is typical for all composite materials which are prepared by mixing a conducting material into an insulating matrix. The beginning of this transition, termed the "percolation threshold", is known to be dependent on particle shape, size and the nature of the insulating matrix. For example, Blaszkiewicz *et al.* [18] found that the percolation threshold for carbon black particles in epoxy was 7.4 vol% compared with 42 vol% for graphite flakes in epoxy. Geometric considerations suggest that flakes will have a lower percolation threshold than spherical particles [19]. If conducting spheres are randomly placed in an insulating matrix then a volume fraction of 0.637 would be required before a continuous conducting network

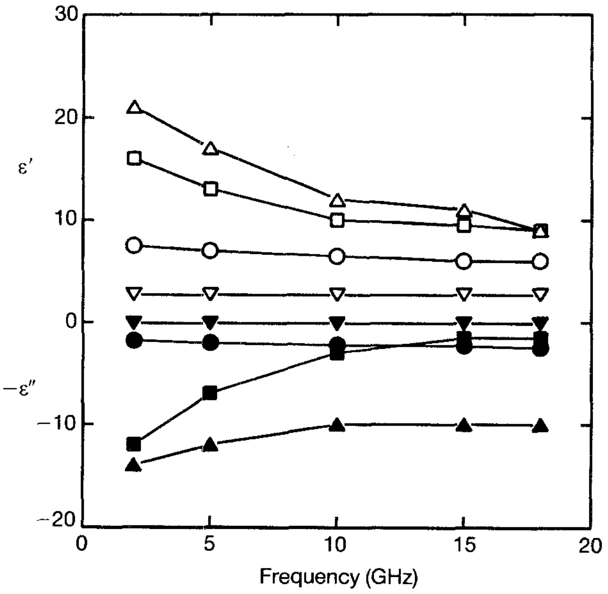

Figure 1 The real part, ε', and imaginary part, ε'', of the complex permittivity as a function of frequency and concentration of PPy powder in silicone rubber. (▽, ○, □, △) ε', (▼, ●, ■, ▲) ε''. (▽, ▼) 0 parts; (○, ●) 4 parts; (□, ■) 10 parts; (△, ▲) 15 parts.

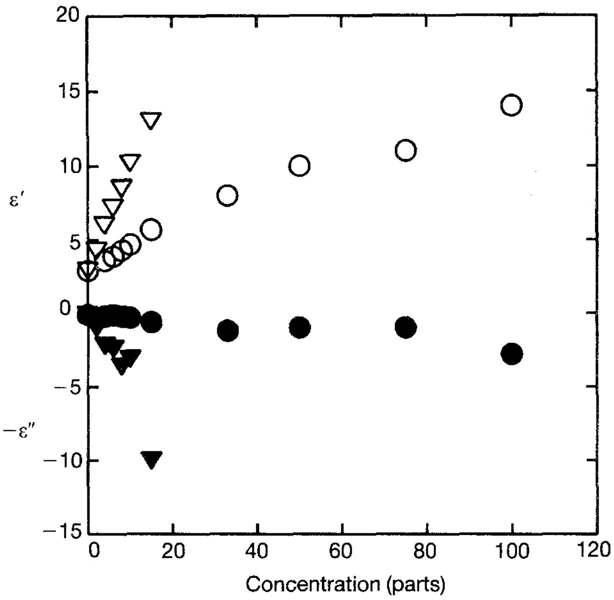

Figure 2 Comparison of the real part, ε' (open symbols), and imaginary part, ε'' (closed symbols), of the complex permittivity between the PPy powder/silicone rubber and PPy flakes/silicone rubber composites at 10 GHz. (▽, ▼) PPy powder/silicone rubber composite, (○, ●) fully doped PPy/*p*-TS flakes/silicone rubber composite.

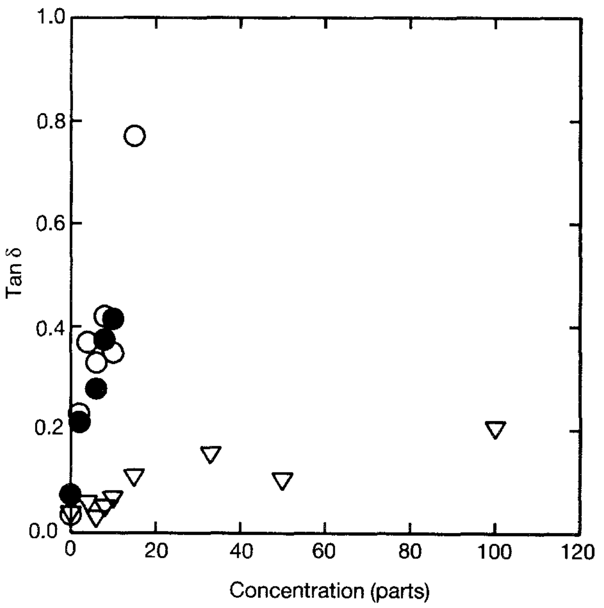

Figure 3 Loss tangent, $\tan\delta$, versus concentration of dispersants. (○) PPy powder/silicone rubber composite; (●) PPy powder/vinyl ester composite; (▽) fully doped PPy/*p*-TS flakes/silicone rubber composite.

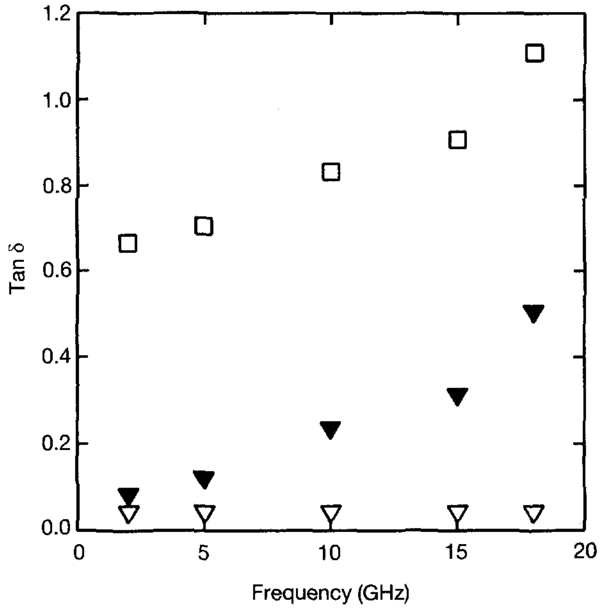

Figure 4 Loss tangent, tan δ, versus frequency. (□) PPy powder (15 parts)/silicone rubber composite; (▼) fully doped PPy/*p*-TS flakes (100 parts)/silicone rubber composites; (▽) silicone rubber.

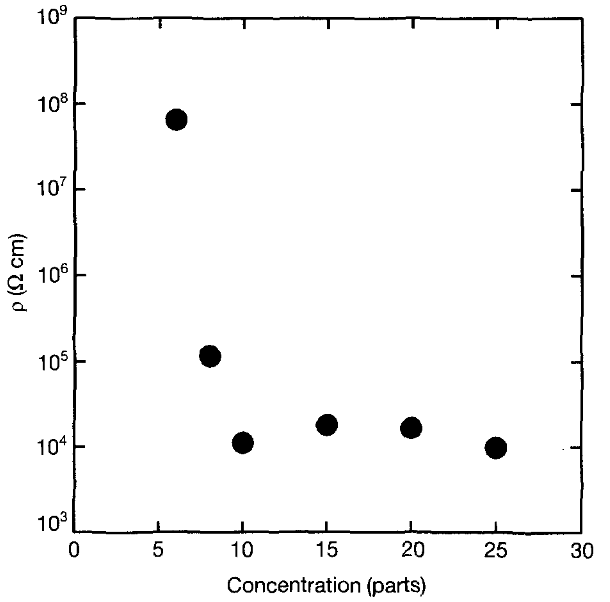

Figure 5 Resistivity, ρ, versus concentration of PPy powder for the PPy powder/silicone composites.

would form [18]. This theory, however, neglects all attractive interactions. Carbon black particles actually aggregate. These aggregates can be in the form of long chains with a resulting high aspect ratio (length to cross-sectional area). The conducting filler can then be thought of as being fibrillar and the probability of two fibril chains contacting is higher than two flakes or two non-interacting spheres.

Previous explanations for the lower than predicted percolation threshold have included the effect of tunnelling between conductive particles [20]. However, it is difficult to see why a tunnelling phenomenon would be affected by the properties of the matrix. In this work, for example, the silicone composites have a measurable resistivity (10^4 Ω cm at 10 parts PPy powder, Fig. 5) while the composites prepared with a vinyl ester matrix show a resistance greater than 10^8 Ω. Wessling [21] has suggested that carbon blacks and intrinsically conducting polymer particles will "flocculate" at some critical concentration and form "seams". Such seams can form a conducting network within an insulating matrix even though the concentration of conductive filler is substantially below the geometric percolation threshold. Similar flocculation phenomena are well-documented for metallic powder/polymer blends [22–24].

The percolation threshold concentration of the PPy powder/silicone rubber composite is substantially lower than that of polyacetylene/polystyrene copolymer (20 vol %) [25]. Obviously, this is a direct consequence of the aggregation of powder particles to form a segregated distribution which generates electrically conductive networks leading to an insulator–conductor transition at a low concentration. A PPy network in the PPy powder/silicone rubber system is clearly seen from Fig. 6, whereas such a network is not visible in the vinyl ester system (Fig. 7). Fig. 8 shows the micrographs of the polypyrrole flakes/silicone rubber system where a continuous PPy network is not formed even at 100 parts of PPy flakes. It is clear that in this system, aggregation does not occur. Addition of

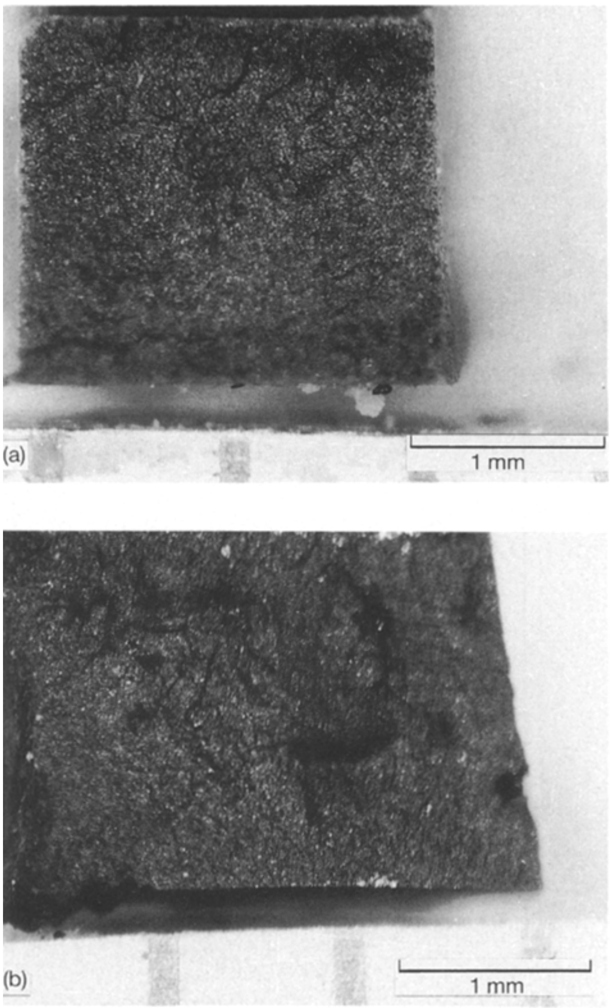

Figure 6 Optical micrographs showing a network of conducting dispersant (dark phase) in PPy powder/silicone rubber composite. (a) 4 parts PPy powder; (b) 15 parts PPy powder.

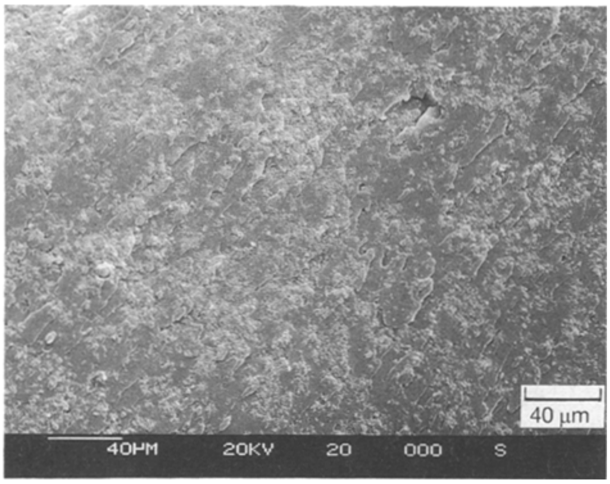

Figure 7 Scanning electron micrograph of PPy powder (15 parts)/vinyl ester composite.

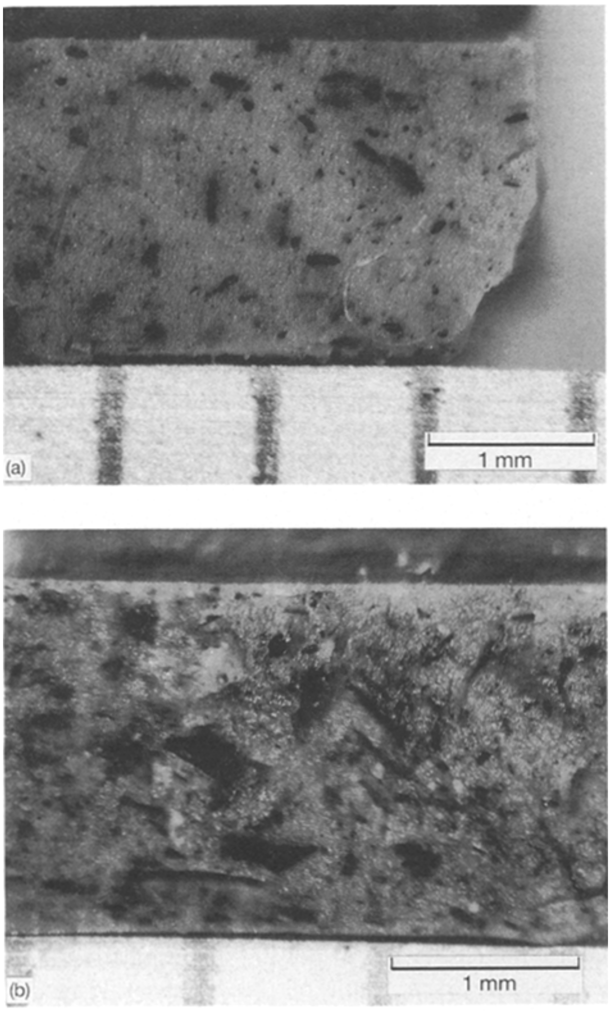

Figure 8 Optical micrographs of fully doped PPy/*p*-TS flakes/silicone rubber composite. (a) 4 parts of PPy flakes; (b) 15 parts of PPy flakes.

these flakes to a conducting polypyrrole powder/silicone rubber sample appears to disrupt the network that was originally formed. However, powder particles do not always lead to the formation of a contiguous network. The large size (up to 300 µm) of PAn aggregates formed during chemical polymerization may prevent effective formation of the network in the silicone rubber matrix (Fig. 9), even when PAn concentration is as high as 20 parts. The effect of the size of dispersant on the formation of network is currently under investigation for a variety of conducting polymers. In this study, the PPy powder/silicone rubber composite is the only system exhibiting the formation of a conductive network (Table II).

As shown in Fig. 3, tan δ and concentration of the dispersant exhibit a linear relationship. The value of tan δ for the PPy powder (15 parts)/silicone rubber composite is approximately 0.65 at 2 GHz and approaches 1.1 at 15 GHz (Fig. 4). The results in Fig. 3 imply that higher values of tan δ can be obtained if

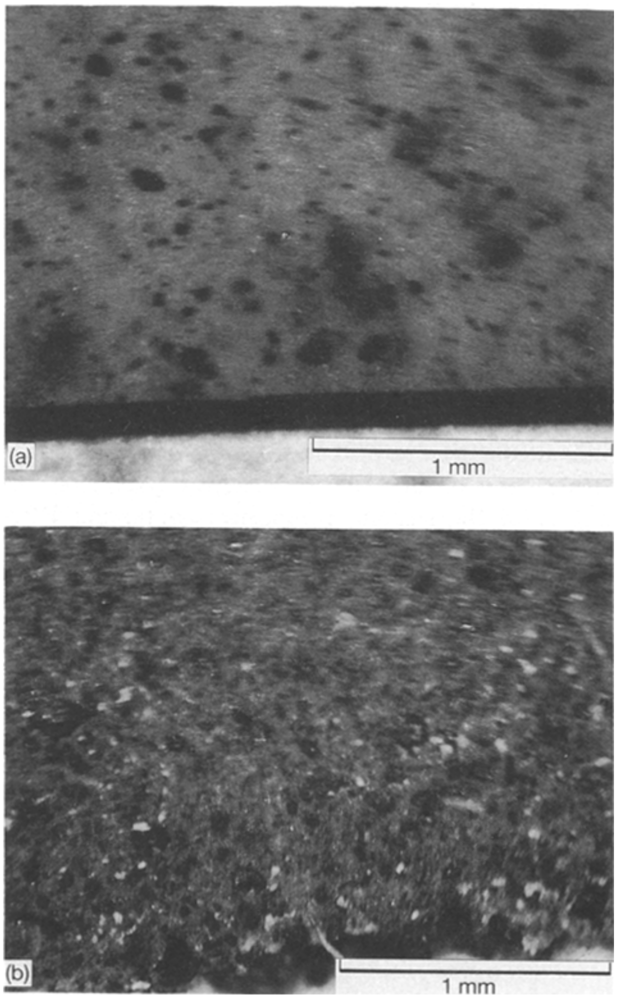

Figure 9 Optical micrographs of PAn acid powder/silicone rubber composite. (a) 4 parts PAn acid powder; (b) 15 parts PAn acid powder.

TABLE II Conductive network formation

Dispersant	Matrix	
	Silicone rubber	Vinyl ester
PPy powder	Yes	No
PPy flakes	No	N/A
PPy powder/flakes[a]	No	N/A
PAn acid powder	No	N/A
PAn base powder	No	N/A

[a] 13 parts powder and 10 parts flakes.

composites of higher concentrations of the dispersant can be achieved. In this study, because of the poor wettability of powder particles and weak binding force of silicone rubber, it was difficult to produce composites with a dispersant concentration higher than 20 parts.

The classical laws of mixtures have been customarily used to express the dielectric relationship between the composite, matrix and dispersants. However, these laws can break down at high dispersant concentrations. For example, according to Lichtenecker [26], the magnitude of permittivity $|\varepsilon^*|$ can be related by

$$\log(|\varepsilon^*|_c) = V \log(|\varepsilon^*|_d) + (1 - V) \log(|\varepsilon^*|_m) \quad (3)$$

where the subscripts c, d and m refer to the composite, dispersant and matrix, respectively, and V is the volume fraction of the dispersant. As shown in Fig. 10, a linear relationship is observed for PPy powder and flakes composites only up to a volume fraction of 5% and 10%, respectively. A marked deviation from this law is obvious at greater concentrations. This indicates that Equation 3 is not appropriate for these composites because the law deals only with random distribution of the dispersant where particle shapes, particle interactions, complex aggregate structure of conductive components and network formation are not taken into account.

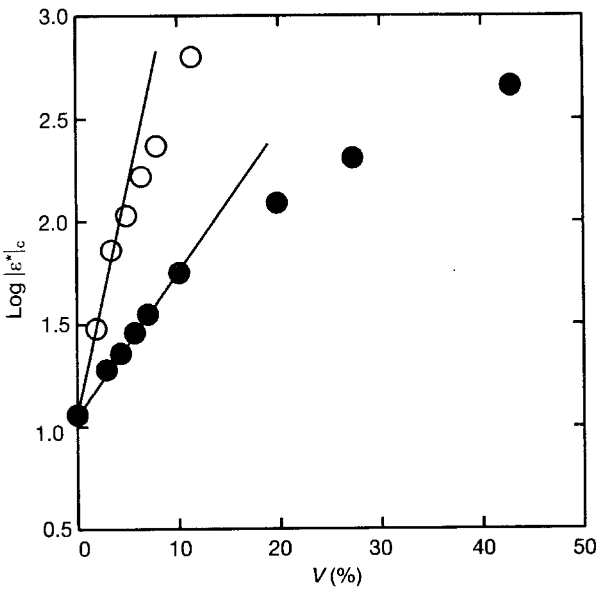

Figure 10 Magnitude of permittivity versus volume fraction. (○) PPy powder/silicone rubber composite; (●) fully doped PPy/p-TS flakes/silicone rubber composite.

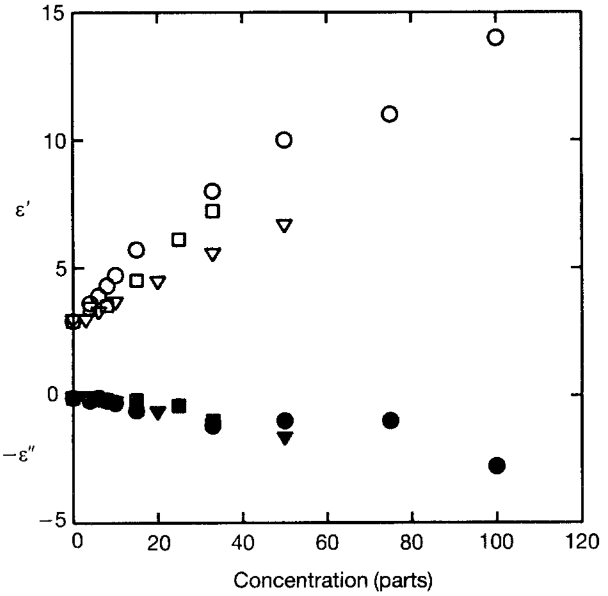

Figure 11 The real part, ε′ (open symbols), and the imaginary part, ε″ (closed symbols), versus concentration of the PPy flakes in silicone rubber. (○, ●) Fully doped PPy/p-TS; (□, ■) PPy/DDS; (▽, ▼) dedoped PPy/p-TS.

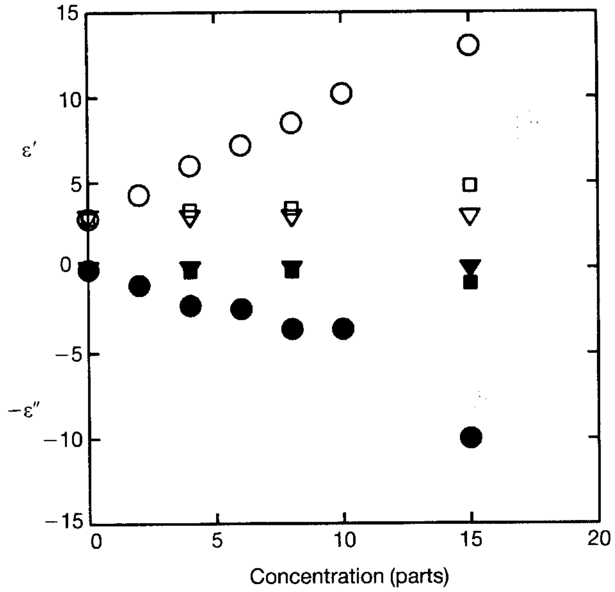

Figure 12 The real part, ε′ (open symbols), and the imaginary part, ε″ (closed symbols), versus concentration of powder in silicone rubber. (○, ●) PPy powder; (□, ■) PAn acid powder; (▽, ▼) PAn base powder.

3.2. Effect of the conductivity of dispersant

Figs 11 and 12 illustrate the dielectric parameters ε′ and ε″ of the silicone composites containing PPy flakes, PPy and PAn powders. For the composites containing PPy flakes, ε′ increases with the dispersant conductivity, σ (Table I). Also, sufficiently high conductivity of the PPy powder ($\sigma = 80\ \Omega^{-1}\ \mathrm{m}^{-1}$) results in a substantial increase in ε′ in the PPy powder composites (Fig. 12). In comparison, low conductivity of the dispersant results in a marginal increase in dielectric properties of the PAn acid composite, and the insulating PAn base powder does not alter dielectric properties of the matrix (Fig. 12). Obviously, the ε′ values increase with dispersant concentration and conductivity as a result of the Maxwell–Wagner–Sillars (MWS) interfacial polarization between the dispersant particles and the insulating matrix (e.g. [27]). A similar result has been observed in carbon black/polyethylene systems [11].

The dispersant conductivity, σ, has little influence on ε″ in the composites containing PPy flakes or PAn powders (Figs 11 and 12), whereas a substantial increase in ε″ is noted for the PPy powder composites (Fig. 12). This again suggests that the formation of conductive contiguous networks or long chains, which are favoured in PPy powder, would have a profound effect on enhancement of ε″. At high frequencies, the loss tangent, tan δ, of the PPy powder/silicone rubber composite exceeds unity (Fig. 4). On the other hand, tan δ of the carbon black/polyethylene composites reaches a maximum at the percolation threshold and declines with increasing carbon black loading. The decline is attributed to reduction of the induced charges by the interfacial polarization effects [11]. This contrasts with the data obtained from the PPy powder/silicone rubber composite where tan δ continues to increase with dispersant loading beyond the threshold (Fig. 3). Although it has been recognized that the MWS polarization always develops in heterogeneous dielectrics, this problem has not been studied in detail for electrically conductive powder/flakes composites. The apparent conflict of the tan δ data between the carbon black/polyethylene and PPy powder/silicone rubber composites suggests that the MWS effect may not be the only factor affecting the dielectric properties of the conducting polymer composites. Indeed, the high value of ε'' (or tan δ) of the PPy powder/silicone rubber composite indicates a high a.c. conductivity, $\sigma(\omega)_m$, at microwave frequencies (Equation 2). At high frequencies, the polarization of the matrix between two conductive clusters acting like a capacitor network can substantially contribute to the a.c. conductivity due to the displacement current [28].

Although the results of this study indicate that the dielectric behaviour of the conducting polymer composites is strongly related to the dispersion of the second phase, i.e. aggregation or random distribution, the d.c. conductivity cannot be considered as the only parameter controlling the dielectric properties. Future studies should be focused on how the formation of a conducting network and the proximity of conductive clusters will effect the matrix polarization and thus the a.c. conductivity in a wide range of frequencies.

3.3. Effect of matrix materials

Figs 3 and 13 show the dielectric properties of PPy powder/silicone rubber and PPy powder/vinyl ester composites. When the silicone rubber is used as a matrix the values of ε' and ε'' significantly increase, although tan δ is apparently independent of matrix materials (Fig. 3). In contrast to the PPy powder/silicone rubber composites where the insulator–conductor transition occurs at a concentration of 5 parts (Fig. 5), the PPy powder/vinyl ester maintains insulating characteristics to a maximum 15 parts achieved in this study. During curing, vinyl ester can prevent the agglomeration of the powder particles due to increasing viscosity of the matrix and thus the conductive networks or long chains cannot effectively develop. This suggestion is supported by the study of Ghofraniha and Slovey [10] which revealed the relationship between the resistivity of carbon black/polymer blends and their rheological behaviour. The degree of cross-links in the matrix can also affect the electrical properties of the carbon black composites [8]. Furthermore, it is possible that insulating vinyl ester layers are formed on the particle surface disrupting the network contiguity.

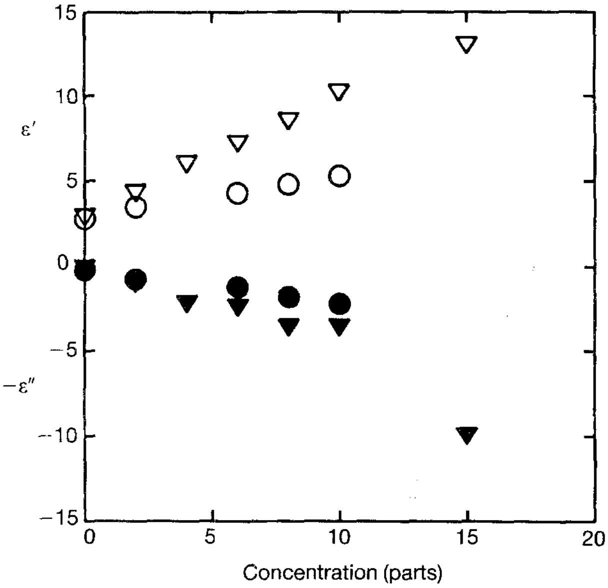

Figure 13 The real part, ε' (open symbols), and the imaginary part, ε'' (closed symbols), versus concentration of PPy powder in the composites. (▽, ▼) Silicone rubber as matrix material; (○, ●) vinyl ester as matrix material.

Wessling [21] and Miyasaka *et al.* [29] have shown that the surface tension of the insulating matrix plays an important role in determining the percolation threshold of carbon black composites. When carbon black is mixed into a matrix with low surface tension such as polypropylene, it prefers to aggregate in order to minimize the interfacial energy. If nylon or esters are used, the carbon black is dispersed more efficiently because the difference in the surface tension of the matrix and the conducting dispersant is not as great. (Surface tensions of carbon black and polyaniline have been recorded as approximately 55 and 70 mN m^{-1}, respectively [21].) Therefore, when the mismatch in surface tensions between the conducting particles and the insulating medium is minimized, the interfacial energy is minimized and dispersion can readily occur with the insulating polymer being completely adsorbed on to the conducting particle. If, however, the matrix has a low surface tension, as in the case of silicones [30], flocculation at low concentrations of carbon or conducting polymer can be achieved. However, the size of the dispersant particles is also important and its effect on flocculation has been previously investigated [23, 29, 31]. When the dispersant phase is made up of large aggregates, the excess interfacial energy obtained upon complete dispersion is considerably less than the corresponding interfacial energy gained upon dispersing smaller particles. Thus, the drive towards flocculation or network formation will be greater in a finely divided system. This would account for the differences observed between the dispersing behaviour of PPy and PAn powders in this study.

4. Conclusions

The formation of a conductive network in the conducting polymer composites was found to be the main factor in increasing ε'' or tan δ of the composite. The network formation, in turn, is controlled by the shape and aggregate size of conducting polymer particles dispersed in an insulating matrix. The PPy powder produced by chemical polymerization exhibits various particle configurations which facilitate the formation of the network at a concentration as low as 5 parts. On the other hand, large PAn aggregates or PPy crushed films, which are two-dimensional flakes of high conductivity, cannot develop networks or contiguous long chains, and effectively destroy the formation of networks when added to composites containing PPy powder. Therefore, conductivity of the composites is low in the absence of a contiguous network of the conducting dispersant even if the content and the conductivity of that dispersant are high.

To date, matrix materials have been selected as required by mechanical and environmental criteria. However, this study revealed that the curing process, physical properties of the matrix and interaction between matrix and dispersed particles can affect the agglomerating characteristics of the particles which determine the resistivity and dielectric behaviour of the composites.

The ability to design the dielectric properties of conducting polymer composites is one of the significant outcomes of this study. Depending on the choice of matrix materials, and the particle geometry and conductivity of the dispersants, the dielectric properties of the resultant composites can be tailored for a particular application.

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