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Electric field-induced aligned multi-wall carbon nanotube networks in epoxy composites

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Abstract

CVD-grown multi-wall carbon nanotubes were dispersed as an electrically conductive filler in an epoxy system based on a bisphenol-A resin and an amine hardener. The application of both AC and DC electric fields during nanocomposite curing was used to induce the formation of aligned conductive nanotube networks between the electrodes. The network formation process and resulting network structure were evaluated by in situ optical microscopy and current density measurements as a function of curing time. Parameters such as field strength and nanotube weight fraction were varied. The carbon nanotube agglomeration mechanism was dominated by the electric field-induced forces acting on the nanotubes, which have a negative surface charge after processing in the epoxy. The network structure formed in AC fields was more uniform and more aligned compared to that in DC fields. The specific bulk composite conductivity of fully processed composite samples reflected the differences in the nanotube network structure. Perhaps surprisingly, the network efficiency was not enhanced by this processing method, although the approach does offer the possibility of achieving bulk conductive nanotube–polymer composites with anisotropic electrical properties and a degree of optical transparency.

Keywords: Carbon nanotubes; Epoxy matrix; Electrical properties

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1. Introduction

Current and projected markets for carbon nanotube-based polymer composites are aimed at the exploitation of the high electrical conductivity of carbon nanotubes coupled to their unique geometry. Such composites offer mechanical property advantages over alternatives based on equiaxed fillers (carbon blacks), macroscopic fibres, intrinsically conductive polymers or thin conductive coatings. Due to the low filler loading fractions required, the mechanical properties and the surface finish of the composite matrix can be maintained. In contrast, conventional surface coatings (e.g. ITO) offer a high electrical conductivity of more than $10^5\,\mathrm{S\,m^{-1}}$ coupled with an optical transmittance exceeding 80% in the visible spectrum [1–3]; a combination which cannot, at present, be achieved with bulk, nanotube-filled composites. Nevertheless, a simple nanocomposite approach promises a unique combination of processing ease and comparatively low cost. Indeed, these materials are increasingly used in the electronics sector, as well as for automotive and aerospace applications, where an electrical conductivity exceeding $10^{-6}\,\mathrm{S\,m^{-1}}$ is required for the dissipation of electrostatic charges.

In previous studies we have shown that the use of CVD-grown multi-wall carbon nanotubes as a conductive filler in an epoxy matrix leads to sufficient electrical conductivity for anti-static applications at filler concentrations as low as 0.005 wt% [4,5]. By adjusting the processing parameters, such as shear rate and resin viscosity, macroscopic agglomeration of carbon nanotubes can be induced. This

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agglomeration process leads to useful electrical properties, but the optical transparency of the polymer is degraded. To date, only chemically functionalised single-wall carbon nanotubes have allowed the manufacture of transparent polymer thin film [6] and bulk glass [7] nanocomposites containing randomly dispersed bundles of nanotubes. The thermoplastic nanocomposite films had a reduced optical transparency with increasing nanotube content, but retained 68% of the optical transmission ($\lambda = 500$ nm) of the unfilled matrix at a loading fraction sufficient for anti-static applications [6]; however, the film thickness was only 34 μm.

Studies on carbon black–epoxy composites have shown that process-induced negative surface charges on carbon black particles can be exploited to produce initially charge-stabilised dispersions, which subsequently form oriented filler networks in both direct current (DC) and alternating current (AC) fields [8,9]. The resulting structure of the filler networks was found to depend on the properties of the electric field, with AC fields generally leading to more homogeneous networks. Although electric fields have recently been applied to induce aligned vapour-grown carbon nanofibre networks in a fully processed epoxy matrix [10], this particular system showed poor results with regard to both the loading fraction necessary to form a conductive filler network and the maximum composite conductivity that could be achieved, compared to carbon black in the same epoxy matrix [9].

Electric fields have not yet been applied to achieve oriented conductive carbon nanotube networks in fully processed nanotube–polymer composites. Experiments aimed at aligning carbon nanotubes have, so far, been carried out only on dispersions of nanotubes in various liquid media. Multi-wall carbon nanotubes dispersed in isopropyl alcohol or ethanol, respectively, have been successfully aligned in sinusoidal AC fields [11,12]. Similar effects were observed for single-wall carbon nanotubes in ethanol [13].

The present work investigated the influence of electric fields on dispersions of CVD-grown multi-wall carbon nanotubes in an epoxy matrix during curing. The expectation was that more efficient networks would form if the aggregation process was guided in this way, and that interesting anisotropic conductivities and optical properties might emerge. Nevertheless, the previous literature on carbon nanofibres suggested that network formation might not be straightforward [10].

2. Materials and experimental details

The multi-wall carbon nanotubes used in this study were grown at the University of Cambridge, using injection chemical vapour deposition (CVD). This process produces aligned arrays of carbon nanotubes with well-defined dimensions that can be controlled as a function of production parameters [14,15]. As previously described [5], the aligned and pure nanotube material made for this study had an average diameter of around 50 nm and a length of 43 ± 3 μm. The polymer matrix was based on a bisphenol-A resin (ARALDITE LY 556, Vantico) and an amine hardener (ARALDITE HY 932, Vantico).

This study mainly focuses on the effects of electric fields on epoxy composites containing 0.01 wt% carbon nanotubes, a value slightly above the bulk percolation threshold under optimal aggregation conditions [5]. The nanotubes were first dispersed in about 15 g of epoxy resin using a dissolver disk, rotating at 2000 rpm, for 60 min at room temperature. The mixture was then equilibrated at 60 °C for a further 10 min of stirring at 2000 rpm, before adding the hardener (32:100 hardener:resin ratio). Maintaining 60 °C, the mixture was stirred for a further minute at 500 rpm, and then left standing for 4 min to remove excess air.

Optical microscopy slides were sputtered with gold to produce electrodes with a spacing of 400 μm for in situ observation of the nanotube network formation. These slides were placed on a Linkam hot stage set at 80 °C and drops of the nanotube-epoxy dispersions containing 0.01 wt% multi-wall carbon nanotubes were exposed to DC and sinusoidal AC fields of 100 V/cm. Here, given field strengths refer to the peak values of the AC fields, which were applied at a frequency of 1 kHz throughout the experiments. An Olympus light microscope coupled with a digital video camera was used to observe the network formation as a function of time during the curing process.

A further set of specimens was cast in silicone rubber moulds to produce bulk nanocomposites. In order to apply the electric fields during curing of these bulk samples, rectangular copper electrodes with a cross-sectional area of about 2 cm^2 and a spacing of 1 mm were dipped into the dispersions. These bulk composites were cured at 80 °C for 4 h in an oven. Throughout the full curing cycle, DC and AC fields of 50, 100 and 200 V/cm, respectively, were applied. For DC fields, a Wayne Kerr autoranging power supply and a Solartron 1278 Frequency Generator were used both to apply the potential and to measure the current passing through the sample during the curing process. The 1 kHz sinusoidal AC fields were supplied by a low-frequency signal generator developed in-house, and the currents were measured with a Fluke multimeter. In order to explore the effects of loading fraction, bulk nanocomposites containing from 0.005 to 0.02 wt% nanotubes were cured in AC and DC fields of 100 V/cm.

AC impedance spectroscopy was carried out on fully cured bulk specimens, using a Solartron 1278 Frequency Generator and 1260 Impedance/Gain Phase Analyzer at voltage amplitudes of 0.4 V DC and 1 V AC, and frequencies ranging from 1 to 10^5 Hz. Using the measured real and imaginary parts of the complex impedance $Z^*(f)$, the conductivity σ was calculated as a function of frequency, f, of the samples according to $\sigma(f) = (1/|Z^*(f)|)t/A$. Here, A is the embedded electrode surface area and t is the distance between them. The bulk samples were cut into slices perpendicular to the electrodes and examined by transmission light microscopy.

3. Results and discussion

As stated previously, the shear-intensive mechanical stirring process leads to well-dispersed CVD-grown multi-wall carbon nanotubes in the pure epoxy component [4,5]. After the addition of the hardener, process conditions can be adjusted either to induce or to prevent nanotube agglomeration [5]. The quality of the nanotube dispersion was verified by SEM fractography. Quantification is difficult, and a direct measurement of the true average nanotube separation is not possible; however, measurements on fracture surfaces of well dispersed 0.01 wt% samples yielded a mean in-plane distance of 9.5 ± 5.3 μm. This value is consistent with an estimate of the mean specific epoxy volume associated with each nanotube, calculated from the loading fraction and the properties of both materials (nanotubes: mean length 43 ± 3 μm, mean diameter 50 nm, density 1.7 g/cm^3; epoxy: density 1.175 g/cm^3). Thus, the volume associated with each nanotube is roughly 2430, 1220, and 610 μm^3 per nanotube for loadings of 0.005, 0.01, and 0.02 wt%, respectively; the expected separation for the 0.01 wt% sample is thus 10.7 μm for a random distribution of nanotubes, in close agreement with the experimental value.

The initial stability of the nanotube dispersions can be attributed to a negative surface charge on the nanotubes [5], arising from interactions with the solvent medium. Whilst Yamamoto et al. reported a positive charge for multi-wall carbon nanotubes in isopropyl alcohol [12], our results agree with work published by Prasse et al. who observed that carbon black is negatively charged in the Araldite LY556/HY932 epoxy system [8]. The negative charge was explained by the basic character of the epoxy system which leads to a hydrogen-transfer from the carbon black to the polymer [9]. Although it is likely that CVD nanotubes have suitable surface hydrogens, other effects cannot be ruled out at this stage.

The in situ optical microscopy study revealed that both AC and DC fields induce the formation of oriented carbon nanotube networks during curing of the epoxy. These networks form conductive pathways through the initially liquid samples. Percolation behaviour was apparent from the sharp increase in the current density with increasing time. After the completion of the epoxy cross-linking reaction, the increased current density compared to the starting value provided evidence for the formation of permanent electric field-induced nanotube networks.

3.1. Theoretical considerations

For low filler loading fractions, such as the ones used in this study, dispersions of multi-wall carbon nanotubes in the epoxy can be regarded as dilute solutions of metallic cylinders. The dynamics of similar solutions under the influence of external electric fields have been studied [12]. Although the electrical behaviour of nanotubes is, in principle, complex due to the varying conductance of each shell [16] and the dependence on defects [17], the average properties of relatively large nanotubes are expected to be semi-metallic, and similar to those of graphite.

In the presence of an electric field $\vec{E}$, each conductive nanotube experiences a polarisation $\vec{P}$. This polarisation can be divided into two contributing components, i.e. one parallel to the tube axis ($\vec{P}_{\parallel}$) and one in the radial direction ($\vec{P}_{\perp}$). The magnitude of both components depends on the polarisability tensor of the nanotube. For single-wall carbon nanotubes, it has already been suggested that the static polarisability in the direction of the tube axis is much larger than across the diameter [18]. To a first approximation a similar behaviour might be expected for multi-wall carbon nanotubes exposed to low-frequency electric fields.

This polarisation leads to a torque N_E acting on the nanotube. Under the given conditions, this torque aligns the nanotube against the viscous drag of the surrounding medium in the direction of the electric field. A comparable behaviour has already been observed for multi-wall carbon nanotubes in different solvents [11,12]. An illustration of the behaviour of a cylindrical particle exposed to a homogeneous electric field is given in Fig. 1.

Apart from this rotation and alignment, the behaviour of a carbon nanotube depends on the type of the applied electric field and its surface charge. In case of a DC field, charged carbon nanotubes move according to their electrophoretic mobility μ [19], where

$$\mu = \frac{v}{E} = \frac{\varepsilon\zeta}{\eta}, \tag{1}$$

towards the electrode with the opposite sign, where they discharge. Here, ε is the dielectric constant and η the viscosity of the epoxy resin, respectively, and ζ is the zeta potential of the carbon nanotube, which is a measure of the surface charge [19].

In case of an AC field, the net electrophoretic mobility due to the zeta potential equals zero. However, aligned metallic carbon nanotubes cause inhomogeneities in the electric field, as shown schematically in Fig. 2. One effect is

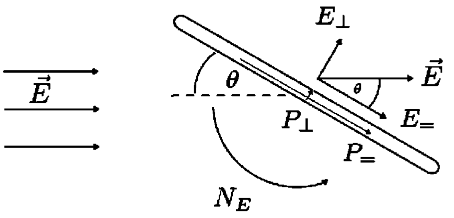


Fig. 1. Schematic illustration of a polarised cylindrical particle in an electric field.

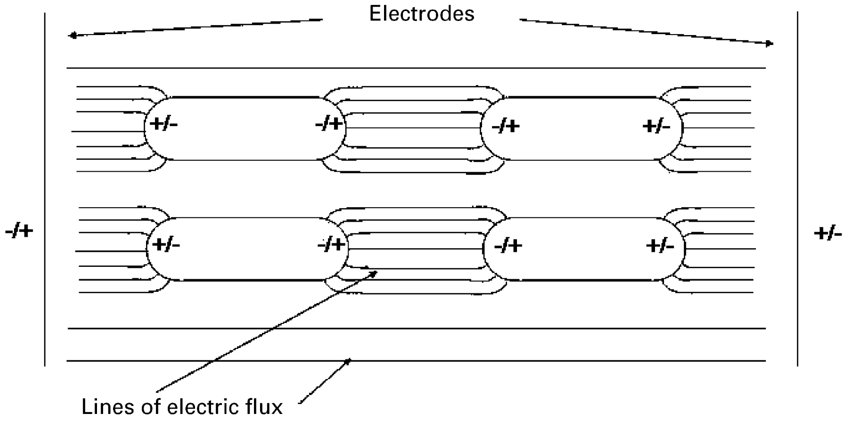


Fig. 2. Schematic illustration of the interaction between aligned and polarised metallic cylindrical particles in an electric field.

a Coulombic attraction between oppositely charged ends of the nanotubes. In addition, the non-uniform electric field in the vicinity of the nanotube tips results in the movement of induced dipoles towards the area with the highest field strength, a behaviour which is called dielectrophoresis [20, 21]. This effect can also induce nanotube movement towards the electrode for those nanotubes in close proximity to the electrodes.

3.2. Carbon nanotube network geometry

Closer analysis of the optical micrographs recorded during the in situ electric field-induced nanotube agglomeration allowed the characterisation of the resulting nanotube network structures as a function of field type and time.

During the application of a DC electric field, a fraction of the carbon nanotubes was observed to move towards the anode, under electrophoresis, verifying the presence of negative surface charges. As soon as these nanotubes are close enough to the electrode to allow charge transfer, the nanotubes discharge and adsorb onto the anode. Tips of nanotubes connected to the electrode then become sources of very high field strengths and the location for adsorption of further filler particles. As a result, ramified nanotube network structures extend away from the anode, eventually reaching the cathode and providing conductive pathways throughout the sample.

Fig. 3 shows two transmission light micrographs of a dispersion of 0.01 wt% nanotubes exposed to a DC field of 100 V/cm. The dendritic agglomerates were observed at the anode after 5 min. With increasing time during curing, more carbon nanotubes were incorporated into the existing oriented agglomerates. After 240 min, the epoxy is fully cured and the final inhomogeneous nanotube network is clearly visible; see the middle image in Fig. 3. As expected, the nanotube network is denser near the anode. In contrast, the volume close to the cathode is only bridged by a small number of thin, aligned, nanotube agglomerates.

Transmission light microscopy of thin sections taken from the bulk samples confirmed these observations. The bottom image in Fig. 3 shows a typical micrograph of the nanotube network in a bulk sample cured in a DC field of 100 V/cm. Note that due to polishing, there is a slight variation in sample thickness near the electrodes, which appears as a change in contrast. Compared with the glass slide experiment, even stronger nanotube agglomeration on the anode can be observed. This effect may be attributed to lower steric hindrance in three dimensions.

In AC fields, more uniform, aligned nanotube agglomeration was achieved, although the initial onset was slower (around 10 min); these agglomerates are also ramified and grow roughly along the electric field lines, as shown in Fig. 4, until they touch to form conductive connections between the electrodes. The field-induced nanotube network formation can be attributed to a polarisation of the carbon nanotubes. This polarisation leads to an additional attractive interaction (dielectrophoresis) between dispersed individual nanotubes and/or existing network branches.

Similar structures were observed in the bulk samples. The resulting nanotube network is shown in the bottom image of Fig. 4, a transmission light micrograph of this sample cut perpendicular to the electrodes. As in the DC case, the apparent gap between the electrodes and the epoxy can be attributed to polishing artefacts. In addition, the bubble-shaped light spots located near the image borders are produced by light passing through the support of the copper electrodes.

Similar network shapes have also been reported for AC and DC electric field-induced networks of carbon black [8, 9] and carbon nanofibres [10] in epoxy.

3.3. In situ current density measurements

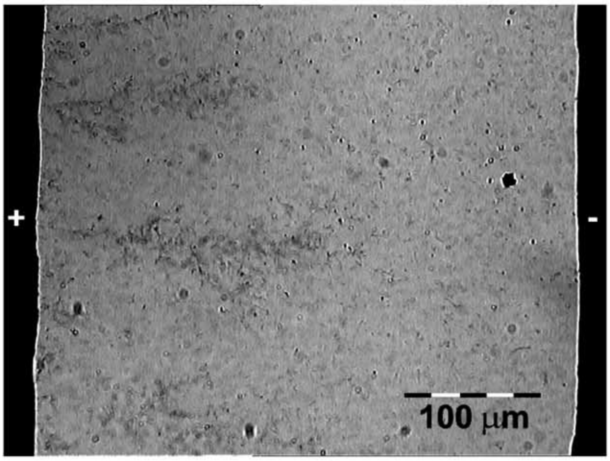
Current density measurements collected during the curing of the bulk samples were consistent with the in situ optical microscopy observations; results are shown in Fig. 5. For comparison, current density values for the pure resin–hardener mixture without carbon nanotubes are included. In addition, the viscosity of the unfilled epoxy system as a function of time at 80 °C is included to indicate the progress of the curing reaction.

The process of cross-linking of the pure epoxy system is characterised by both a sharp increase in viscosity and corresponding decrease in current density after about 4000 s. The curing reaction not only leads to a thickening of the system, but also reduces the concentration of unreacted amine and epoxy molecules, i.e. potential charge carriers, in the dispersion. Both effects contribute to a decreasing ionic conductivity, σ_{ion}, since $\sigma_{\mathrm{ion}} \propto c/\eta$, where c is the concentration of ions and η is the viscosity [22]. The ionic conductivity of the system can be estimated from the current density, $\mathbf{J}$, and the field strength, E, as $\sigma_{\mathrm{ion}} = J/E$. At the onset of the curing reaction, the ionic conductivity is around 10^{-7} Sm^{-1} for both the pure epoxy and the samples containing 0.01 wt% of carbon nanotubes. This value agrees well with the ionic conductivity previously reported for this epoxy system [9]. Although additional capacitive effects might have influenced the root mean square (r.m.s.) value of the AC measurement, the resulting ionic conductivity is in the same order of magnitude. This observation indicates that a loading fraction of 0.01 wt% of dispersed carbon nanotubes does not significantly influence the electrical properties of the uncured epoxy.

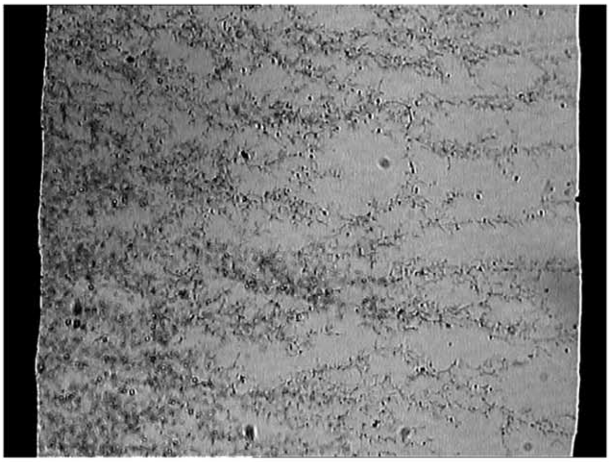
In contrast, the application of electric fields and the subsequent nanotube network formation led to pronounced changes in the current density as a function of time. After about 600 s, the current density of the sample cured in a DC field starts increasing. In the AC field, nanotube network formation can be seen to occur even earlier since network growth starts at both electrodes, even though the onset of agglomeration was slower. Due to the more homogeneous network structure formed in AC fields, the maximum current density is also higher than in the DC case.

A similar behaviour was observed for samples containing different carbon nanotube loading fractions. Fig. 6 shows the resulting current densities as a function of time for samples containing 0.005, 0.01 and 0.02 wt% of carbon nanotubes, respectively. During curing at 80 °C, AC and DC fields of 100 V/cm were applied. In all cases, the electric fields led to an increase in the current density, which can be attributed to the nanotube network formation. With increasing carbon nanotube loading fraction, the onset to network formation occurs earlier. At higher densities of filler particles, the probability of nanotube contact and agglomeration is increased, leading to a greater number of more conductive pathways and a higher maximum current density. The steeper rise in current density for the DC field relates to the network growth process, which tends to build from one electrode, rather than gradually assembling throughout the sample. After the initial steep increase in current density, all samples showed a plateau behaviour, indicating that the network is stabilised even though curing is incomplete. This effect is more pronounced in the case of DC fields, especially for a nanotube content of 0.01 wt%,

Slide



5 min



240 min

Bulk

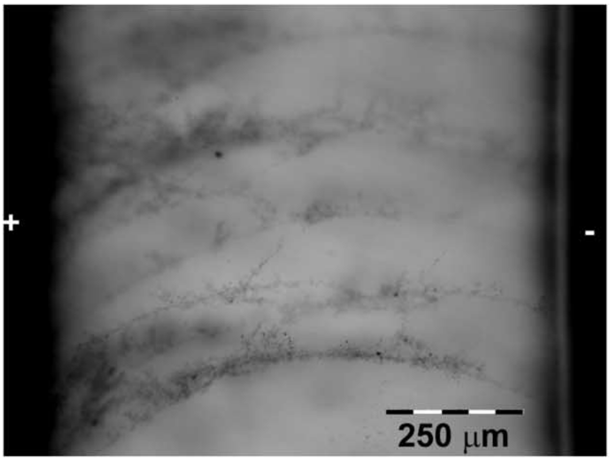


Fig. 3. Transmission optical micrographs of epoxy composites containing 0.01 wt% multi-wall carbon nanotubes during curing at 80 °C in a DC field of 100 V/cm. The bottom image shows the resulting nanotube network structure in a fully processed bulk composite cured under these conditions.

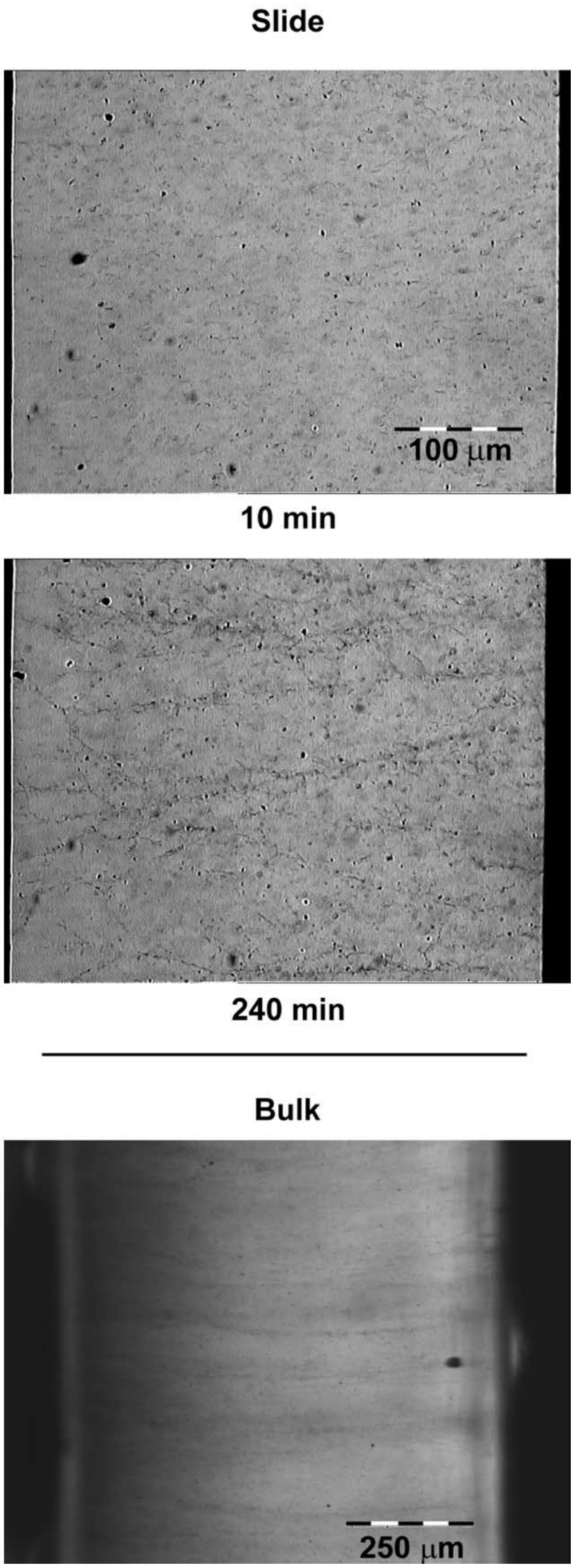


Fig. 4. Transmission optical micrographs of epoxy composites containing 0.01 wt% multi-wall carbon nanotubes during curing at 80 °C in an AC field of 100 V/cm. The bottom image shows the resulting nanotube network structure in a fully processed bulk composite cured with the same parameters.

providing further evidence for faster nanotube incorporation into network branches.

Towards the end of the curing reaction, the current density can be observed to decrease, as shown in Fig. 5. For the pure epoxy system, the current density drops by a few orders of magnitude, whereas it reaches a much higher constant value in all the nanotube-filled systems. As can be seen in Fig. 6, the remaining current density value is generally higher for AC fields for a given nanotube content.

This general decrease in current density in the composite samples at the end of the curing reaction must be associated with the cross-linking process. Since the conductivity associated with the charge carriers in the resin system is orders of magnitude lower than that of the nanotubes, their immobilisation can have only a negligible contribution to the overall conductivity of the composite at this stage. Rather, the loss of conductivity can be attributed to distortion of the nanotube network as a result of the shrinkage associated with the final cross-linking of the matrix. With increasing nanotube loading fraction, the magnitude of the current density decrease becomes smaller, as shown in Fig. 6; greater redundancy in the network helps to maintain integrity during cross-linking.

3.4. Characteristics of bulk composites

Following similar trends to the in situ current density measurements described above, the specific conductivity of fully-cured bulk samples was influenced by the three parameters: frequency of the electric field applied during curing, field strength, and the loading fraction of carbon nanotubes.

The resulting specific bulk composite conductivity increases with the strength of the applied field, as shown in Fig. 7. Here, the specific composite conductivity of fully-processed samples, containing 0.01 wt% carbon nanotubes, cured under the influence of varying electric fields, is plotted as a function of frequency on a log–log scale. A reference sample, cured without the application of an electric field, shows typical dielectric behaviour, indicated by the frequency-dependent conductivity increase with a slope of unity on the double-logarithmic scale. In contrast, all samples exposed to AC and DC fields show a frequency-independent conductivity of more than $10^{-7}\ \mathrm{Sm}^{-1}$ at low frequencies. This constant conductivity is maintained up to a specific knee frequency, above which a transition to a dielectric behaviour can be observed. This observation is typical for nanotube-epoxy composites [5]. Furthermore, AC fields lead to significantly higher conductivity values than DC fields of similar strength, by as much as an order of magnitude, reflecting the more homogeneous nanotube network structure. This result is in excellent agreement with

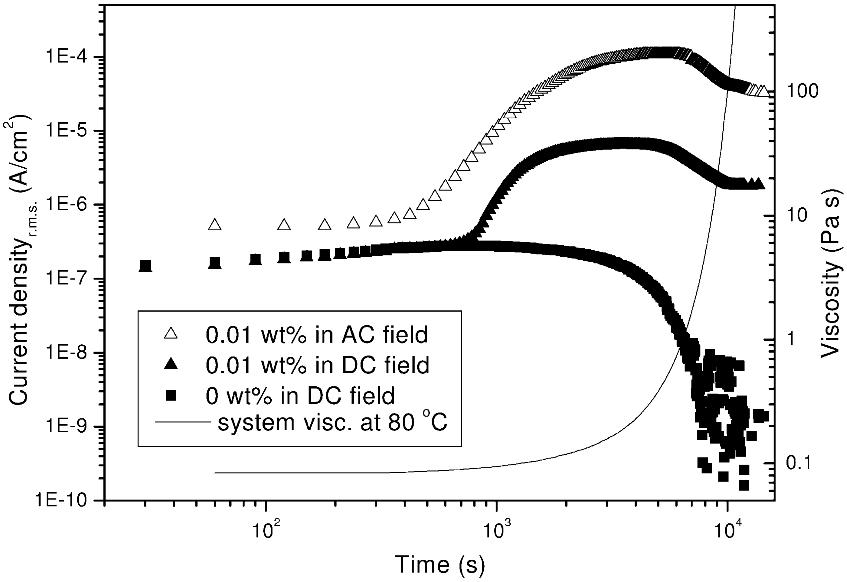


Fig. 5. In situ current density measurements of bulk epoxy composite samples containing 0.01 wt% multi-wall carbon nanotubes compared to the pure epoxy system during curing at 80 °C in electric fields of 100 V/cm. The viscosity of the pure epoxy system at 80 °C as a function of time is included as a reference.

the differences in the final current density values observed in the in situ measurements.

As previously observed for carbon nanotube–epoxy composites [4,5,23] processed without external electric fields, the static conductivity of the cured composites increases with increasing loading fraction of multi-wall carbon nanotubes, as more nanotubes are incorporated into the network. Fig. 8 shows a comparative plot of AC impedance spectroscopy results on bulk samples cured under AC and DC fields of 100 V/cm.

It is worth noting that the electrodes have to be brought into direct contact with the nanotube–epoxy dispersion in order for agglomeration to occur. Whilst AC fields of up to 100 V/cm were able to induce network formation between two copper electrodes dipped into the dispersion, the same electric fields did not lead to nanotube agglomeration when

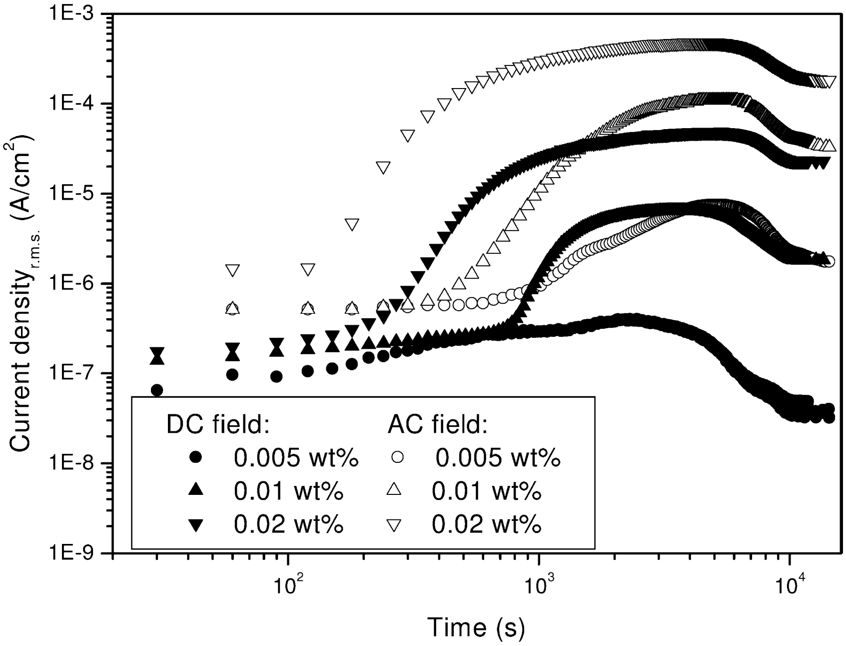


Fig. 6. In situ current density measurements of bulk epoxy composite samples containing different weight fractions of multi-wall carbon nanotubes. AC and DC electric fields of 100 V/cm were applied during curing at 80 °C.

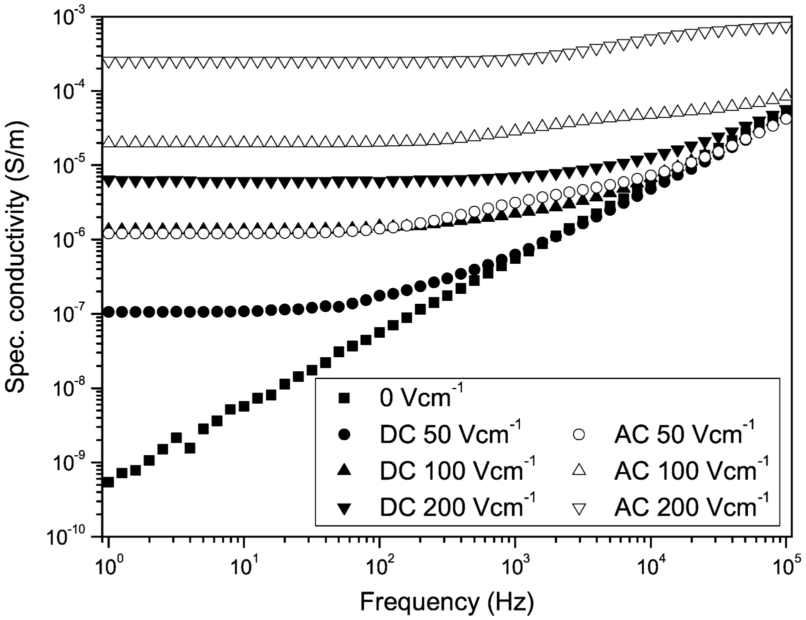


Fig. 7. Specific AC conductivity as a function of frequency for fully processed bulk epoxy nanocomposites containing 0.01 wt% multi-wall carbon nanotubes exposed to AC and DC electric fields during curing.

such a direct contact was prevented. This observation is consistent with the mechanism of the network formation suggested earlier, in which the surface charges present on individual carbon nanotubes are overcome by direct electron transfer.

As discussed, nanotube aggregation in bulk epoxy composites can be adjusted by varying the processing conditions, such as stirring temperature, stirring rate, and curing temperature [5]. The initial sample preparation conditions used in this study were aimed at preventing nanotube aggregation in the bulk epoxy during curing in the absence of electric fields. Fig. 9 highlights the success of this approach. Here, a cross-section of a bulk composite sample containing 0.01 wt% carbon nanotubes cured in a DC field of 100 V/cm is shown. The total sample length is 30 mm and the sample thickness is about 1 mm; the electrode gap is also about 1 mm. A marked difference in the optical transparency of the composite between the electrodes compared to the outer sample regions can be seen. No nanotube agglomeration has occurred in the outer regions, but the presence of the dispersed nanotubes has led to a darkening of the matrix. In contrast, the composite region between the electrodes remains clear and almost fully transparent.

Optical transmission measurements of this sample at a thickness of 2.6 mm using UV–VIS–NIR spectroscopy verified that the transmission at 600 nm, normalised to the value of pure epoxy, is about 25% for the bulk composite outside the electrode gap, and 66% within the electrode gap. The higher transparency within the gap can be mainly attributed to the reduced number of independent scattering centres after agglomeration. However, a small fraction of the filler particles may also be removed to the anode by (di)electrophoresis. No significant dependence of the optical absorption on polarisation was observed, probably due to the relatively large dimensions of the agglomerated filaments. Nevertheless, these measurements highlight the potential of using electric fields to induce aligned nanotube networks in epoxy composites, leading to an electrical conductivity sufficient for anti-static dissipation, whilst retaining a significant proportion of the optical transmission; the degree of transparency is greater than for random networks of similar nanotubes [4,5,23].

Although the maximum static conductivity of such field-aligned nanotube networks in the epoxy exceeds the anti-static level, it is nevertheless lower than the maximum conductivities achievable under optimised processing conditions for epoxy composites containing the same weight fraction of nanotubes arranged in random aggregates [4,5, 23]. The reason for this discrepancy is not clear, but may relate to a low network efficiency in the ramified, field-aligned structure; the dendritic structure is likely to produce a large number of dead ends. The maximum conductivity of such nanocomposites is generally limited due to the presence of polymer barriers between individual nanotubes and/or nanotube clusters [24,25] and is a few orders of magnitude lower than the conductivity of pure nanotube mats [26].

4. Conclusions

CVD-grown multi-wall nanotubes were used as a conductive filler in an epoxy system based on a bisphenol-A resin and an amine hardener. The applied processing scheme led to initially well-dispersed carbon nanotubes in

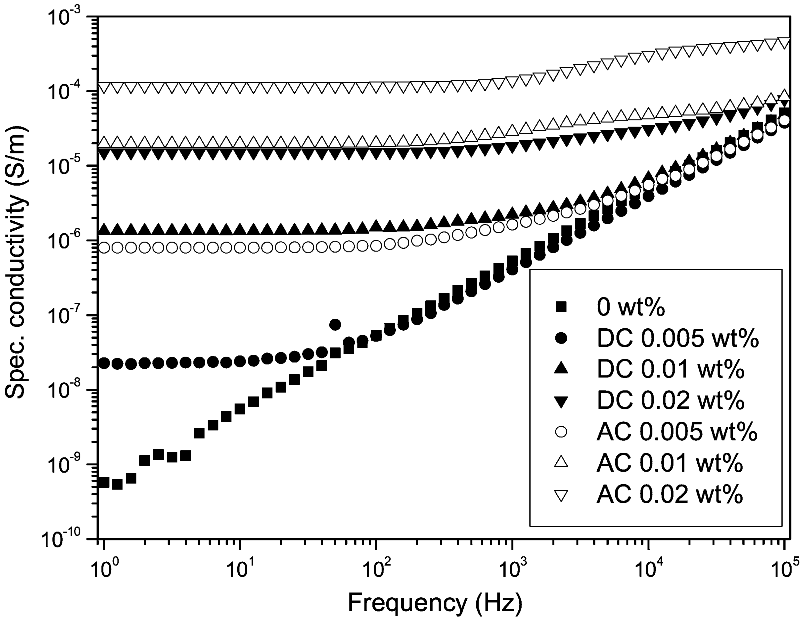


Fig. 8. Specific AC conductivity as a function of frequency for fully processed bulk epoxy nanocomposites containing different weight fractions of multi-wall carbon nanotubes exposed to AC and DC electric fields of 100 V/cm during curing.

the epoxy resin. Further processing was designed to prevent nanotube agglomeration prior to the application of electric fields. It was shown that both AC and DC electric fields can be used to induce the formation of aligned carbon nanotube networks spanning the gap between electrodes in contact with the dispersion.

The success of this approach was demonstrated for both thin film composites and bulk nanocomposite samples. More uniform and more aligned networks can be achieved in AC fields compared to the rather inhomogeneous and branched network structures obtained in the case of DC fields. With increasing field strength, the quality of these networks and the resulting bulk conductivity of the composite material can be enhanced. However, the maximum specific composite conductivity that can be achieved using this approach is still low compared to the conductivity value of pure multi-wall carbon nanotubes and indicates the presence of polymer barriers preventing direct contact between individual filler particles.

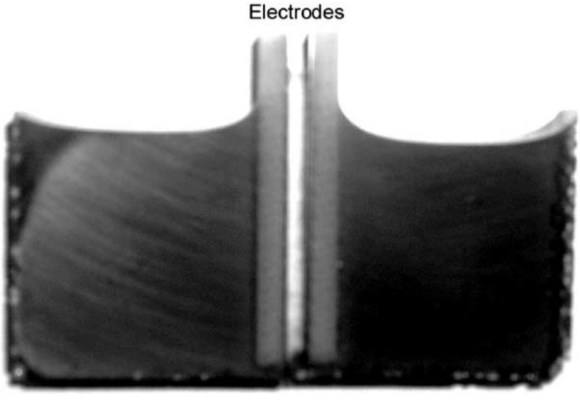


Fig. 9. Transmission optical micrograph of a bulk composite sample containing 0.01 wt% of multi-wall carbon nanotubes cured in a DC electric field of 100 V/cm. The image highlights the optical transparency due to the oriented nanotube network structure between the electrodes. The total sample length is 30 mm, the thickness is about 1 mm and the electrode gap is also 1 mm.

The resulting specific conductivity of bulk composite samples cured in electric fields is suitable for electrostatic dissipation. Furthermore, the observed orientation of the field-induced nanotube networks shows promising optical transparency and should yield anisotropic electrical properties in the cured composite. Although anisotropic conductivity has recently been demonstrated for electric field-aligned vapour-grown carbon nanofibres in the same epoxy matrix [10], the use of nanotubes has produced, in this study, field-aligned networks at dramatically lower filler concentrations and a significant increase in maximum composite conductivity.

The use of series-resistors during the field-induced nanotube aggregation process should allow adjustment of the final conductivity of the composite, following an approach already successfully demonstrated for carbon black-filled epoxy [27].

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