The Dielectric Effect of Xylene on an Organoclay-Containing Composite

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ABSTRACT
The influence of a non-polar solvent, xylene, on the electrical properties of a composite system containing polypropylene, copolymer ethylene/(vinyl acetate) and an organoclay has been considered since the synthesis of composites often involves a solvent. Although in isolation neither the organoclay nor the xylene has a marked effect on the dielectric response of the system, when combined synergistic effects can be seen. We interpret this in terms of solvent-mediated relaxation of amphiphilic compatibiliser moieties introduced into the organoclay to aid processability. These local relaxation processes appear to adversely affect the breakdown strength of the system, which is recoverable upon the removal of xylene from the system.

Index Terms —**Composite insulation, Dielectric losses, Dielectric Breakdown**

1 INTRODUCTION

THE electrical properties of nanocomposite materials has been an active area of research for almost three decades. While numerous reports exist of improved properties on addition of a nanofiller, many others indicate that the inclusion can have negative consequences. For example Zilg et al. [1] reported an increase to the DC breakdown strength when an organophilic layered silicate was added to polypropylene and a slight decrease when the same filler was added to ethylene/(vinyl acetate) (EVA). Montanari et al. [2] report on a similar nanofiller and, although a comparable trend was seen, the overlapping error bars mean that the addition of the filler cannot be said significantly to affect the breakdown strength. Elsewhere, Montanari et al. [3] report that the addition of an organophilic clay nanofiller introduces a new relaxation process to the dielectric response of an EVA matrix, due to the interface created between the matrix and the filler. This was also found in a study involving silica nanoparticles and polyethylene[4]. In contradiction to both aforementioned papers concerning the dielectric response of nanocomposites, Roy et al. [5] found there to be a decrease in the permittivity of their nanocomposite at 1 kHz upon the addition of the nanofiller. While the precise origins of these various results are uncertain, what is clear is that small changes in processing or composition can exert a major effect on macroscopic electrical properties [6].

While the bulk of the literature concerning the dielectric response of nanocomposite materials has focused on particulate systems, such as silica, alumina, titanium dioxide, and gold, in other areas, plate-like nanofillers, such as clays, NiO, boron nitride, and graphite, have received great attention. The current interest in nanocomposites is frequently attributed to pioneering work on systems based upon organoclays, where the initial interest concerned the production of material systems with improved mechanical performance. While dispersion of such systems into polar polymers such as polyamides is relatively straightforward, the incorporation of these nanofillers into non-polar host matrices is problematic. One solution, which has seen much work, involves altering the chemical properties of the matrix system through grafting groups, such as maleic anhydride or glycidyl methacrylate, to non-polar polymers [2, 6, 7]. An alternative strategy, used herein, employs a second polymer that is compatible with both the non-polar polymer and the organoclay as a means of creating a homogeneous blend [8], [9].

Compatibilizing agents allow the polymer chains to closely intercalate between the stacks/sheets present in the clay. When the intercalation reaches a point whereby the sheets are completely separated and dispersed into the continuous polymer matrix, the clay is exfoliated [7]. In doing so, the filler transitions from the micro- to the nano-scale, which can fundamentally change the properties of the system due to the massively increased interfacial area combined with minimizing the phase separation of the overall system [10]. When considering the melt processing of nanocomposites by extrusion, the level of dispersion has been related to the order of material addition and the number of times the material was passed through the extruder [7]. Processing via a solvent phase is a common alternative strategy, whereupon the solvent can mediate the mixing of the materials, such that the final extent of dispersion is

influenced by factors such as the filler loading level, temperature and length of solution reflux time [11].

The work described here set out to examine a factor that is intrinsic to solution processing of materials, namely the influence of retained labile species on the electrical characteristics of a polymeric nanocomposite. While the influence of absorbed water has been considered in detail [12, 13] and shown to be capable of greatly influencing dielectric parameters, charge transport and breakdown strength, the effect of non-polar moieties has not attracted comparable attention. Specifically, the work concerns a tertiary polymer blend system based on isotactic polypropylene (iPP) and an EVA copolymer combined with an organically functionalized nanoclay (organoclay). The objective of the work was to examine the influence of xylene, used for processing, on the electrical response of the system.

2 EXPERIMENTAL

2.1 MATERIALS AND PROCESSING

The following commercial materials were used, as supplied, in the study described here. The iPP and organoclay were both purchased from Sigma Aldrich (product numbers 427888 and 682624 respectively), while the EVA was supplied by DuPont (Elvax 750, containing 9 wt. % of VA). All of the samples considered here contained, by weight, 80:20 of iPP to EVA plus 0, 2.5 or 5 wt. % organoclay. The material processing procedure was adapted from published studies [14] and involved the following sequential steps. First, the organoclay was sonicated in xylene for 30 min to promote dispersion, before being added, along with the EVA, into a rotary evaporator and heated to 140 °C. When the polymer had completely dissolved, the iPP was then introduced and the temperature was increased to 155 °C to dissolve the iPP. The mixture was left stirring for 30 min before, finally, the solvent was removed under vacuum.

From the resulting product, samples of the required thickness were melt-pressed using a hydraulic press. After being held in the melt phase for 5 min, the films were crystallized isothermally at 120 °C for 10 min, before being quenched into a room temperature water bath. The required temperature and time for the crystallization process was determined from a preliminary study of the crystallization behavior of this polymer blend by differential scanning calorimetry (DSC), and was chosen to ensure maximum crystallization of the iPP. All of the resulting sample films were then further dried for one week under dynamic vacuum at 70 °C, whilst monitoring the mass change, to minimize residual solvent and absorption of environmental water in the resulting specimens. These initial specimens will hereafter be referred to as dried samples. In order to explore any potential effects relating to the presence of unwanted labile species, further batches of samples were subjected to exposure to xylene followed by a subsequent re-drying phase. This involved immersion of in xylene for one week (termed immersed samples), followed by drying under dynamic vacuum at 70 °C for two weeks (termed re-dried samples). Variations in sample mass throughout this process

were monitored. For convenience, the above samples are hereafter referred to using the following nomenclature: PP/EVA9-20/X/Y, where PP/EVA9-20 indicates a blend system based on iPP with 20 parts by weight of EVA containing 9% VA, X indicates the nominal clay loading level (0, 2.5 or 5 wt. %) and Y indicates the imposed treatment (D, dried; I, immersed; R, re-dried).

2.2 MATERIAL CHARACTERISATION

Thermogravimetric analysis (TGA) was performed in air up to 800 °C at a heating rate of 10 °C min^{-1}, using a Perkin Elmer Pyris 1 instrument. Fourier-transform infrared (FT-IR) spectra were acquired using a Thermo Scientific Nicolet iS5 instrument; data were acquired with the instrument operating in both transmission mode and attenuated total internal reflection (ATR) mode. AC breakdown strength was measured in accordance with ASTM D149-97a. For this, the sample was immersed in silicone oil between vertically opposed spherical electrodes (6.3 mm in diameter). Each system was tested twenty times using a ramp rate of 500 V/s and the results were analyzed using two-parameter Weibull statistics with 90 % confidence limits, within the Origin software package. Dielectric spectra were acquired using a Solartron 1296 dielectric interface together with a Schlumberger SI1260 impedance/phase gain analyzer system. For this, an AC voltage of amplitude 1 V RMS was applied across electrodes 30 mm in diameter with a guard electrode; a frequency range of 10^{-1} to 10^5 Hz was considered. In the case of samples tested following xylene immersion, these were removed from the solvent bath and allowed to dry naturally in the fume cupboard for 30 min before being tested, such that no residual xylene remained on the sample surface during testing. We found the mass change of the sample after 30 min was slow and therefore unlikely to change significantly over the duration of the 20 min data acquisition.

3 RESULTS AND DISCUSSION

3.1 SAMPLE/XYLENE INTERACTIONS

Figure 1 shows TGA data obtained from both dried and immersed samples. Consider first the results obtained from the dried samples, where three aspects of the data are significant. At low temperature, the absence of any detectable changes in mass indicates that the concentration level of any volatile species that remained entrained within the systems after drying is below the sensitivity of TGA. The bulk of the decomposition then occurs within the temperature range 300-500 °C, which corresponds to the decomposition of the iPP, the EVA and the organic compatibiliser within the organoclay. In line with previous work, the displacement of decomposition to higher temperatures in the presence of organoclay was as expected [15]. At high temperatures (>600 °C), residual mass occurred only when organoclay had been present in the sample and corresponds to the remnant inorganic components of the organoclay. By considering the decomposition of the organoclay in isolation it is possible to evaluate the included organic fraction and, from this, we

conclude that the true initial loading level of the organoclay equates to 1.5 and 4.7 wt. % of the system for the composition goals of 2.5 and 5 wt. % respectively. Nevertheless, for simplicity, the paper will still refer to the samples by the nomenclature described above, which was based on the nominal target compositions. The inset in Figure 1 shows low temperature TGA data obtained from the samples that had been immersed in xylene, from which it is evident that all three systems lose about 5% of their initial mass on heating from room temperature up to 150 ºC. Since the reported boiling temperature of the various isomers of xylene range from 138-144 ºC, this feature, together with the absence of any comparable mass losses in the PP/EVA9-20/X/D sample set, strongly implies that this change in mass is related to desorption of xylene, which occurs progressively up to ~150 ºC.

Indeed, FT-IR data (see Figure 2) obtained from the PP/EVA9-20/5/Y sample set indicates the presence of absorption peaks associated with xylene in PP/EVA9-20/5/I, which are absent in both PP/EVA9-20/5/D and PP/EVA9-20/5/R. Since comparison of the FT-IR spectra obtained from these three systems revealed no other differences, we conclude that the process of xylene immersion did not affect the system in any way other than to introduce this species.

Finally, Figure 3 shows the variation in sample mass with immersion time in xylene, which reveals a number of features of the xylene absorption process. First, in equilibrium, the mass of all three samples can be seen to have increased by 5-7 %. Second, the initial data points acquired after just 4 hr reveal a comparable mass increase to that seen after 7 days. As such, the key aspects of these data is that xylene absorption occurs rapidly and that the mass of the absorbed xylene is largely independent of the amount of organoclay present. While the former necessarily indicates that any associated measurement of sample mass will be subject to uncertainties derived from the process used to remove xylene from the sample surface prior to measurement of the sample mass, the TGA and mass change measurements are, nevertheless, still in broad quantitative agreement.

3.2 DIELECTRIC RESPONSE

Figure 4 shows dielectric spectroscopy data acquired from PP/EVA9-20/0 after different immersion times in xylene. For such a composite system (propylene, ethylene and vinyl acetate units), it is possible to estimate the anticipated value of the real part of the relative permittivity, ε', using any one of a number of effective medium mixing theories [16]. Using the Lichtenecker logarithmic mixing rule [17], results in a value of 2.4, based upon values of 2.3 for iPP [18] and 2.7 for EVA containing 9 % of VA, approximating the latter to be the same as EVA containing 12 % VA [19]. Evidently, the precise permittivity values shown in Figure 4a fall somewhat below this value, which is a consequence of uncertainties/variations in the sample thickness and the impact this has on deriving an effective permittivity value from the measured capacitance. Nevertheless, the key feature of these data and the complementary imaginary permittivity data shown in Figure 4b is that neither parameter is affected by absorption of xylene. The xylene used here contains a mixture of the xylene isomers, but, for a representative
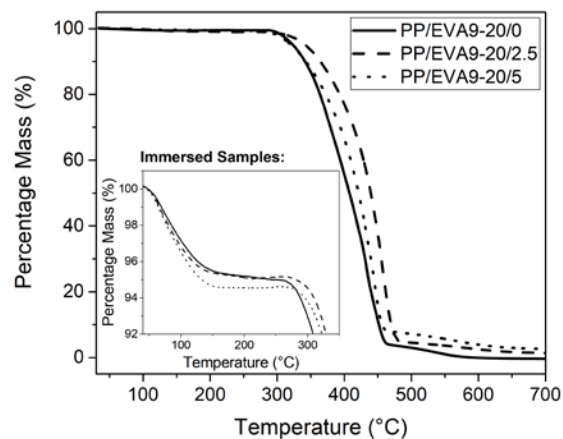

Figure 1. Data from TGA conducted in air on the dried samples and the immersed samples (inset).
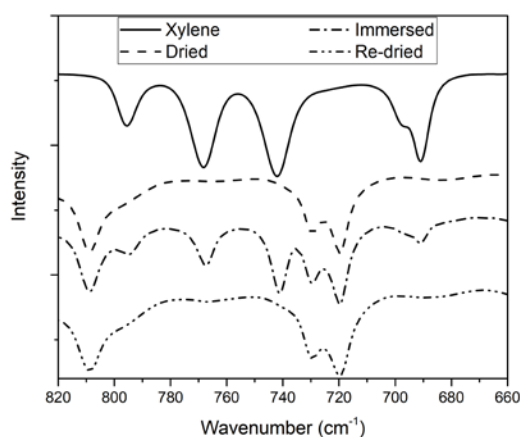

Figure 2. FT-IR data from the fingerprint region acquired in transmission mode from PP/EVA9-20/5/Y samples containing 5 wt % organoclay compared to xylene. The various spectra are off-set for clarity.
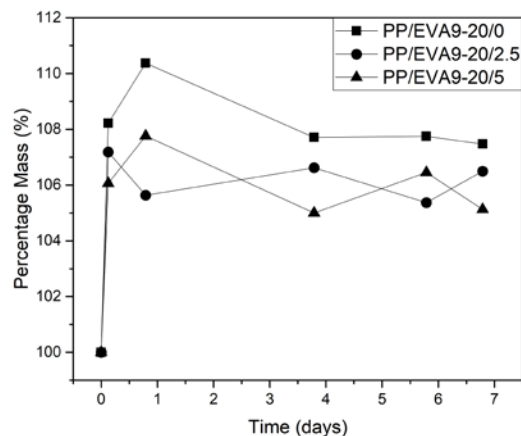

Figure 3. Xylene uptake monitored through mass change, measured 30 min after being removed from the xylene bath.

example, para-xylene has a real relative permittivity value of 2.26 at 1 kHz [20] and, therefore, the inclusion of a small quantity of this solvent would not be expected to dramatically affect ε'. Some dielectric relaxation studies of systems containing chemically similar solvents to xylene have been undertaken: in the case of solutions of polystyrene in toluene, a weak β relaxation is reported and attributed to
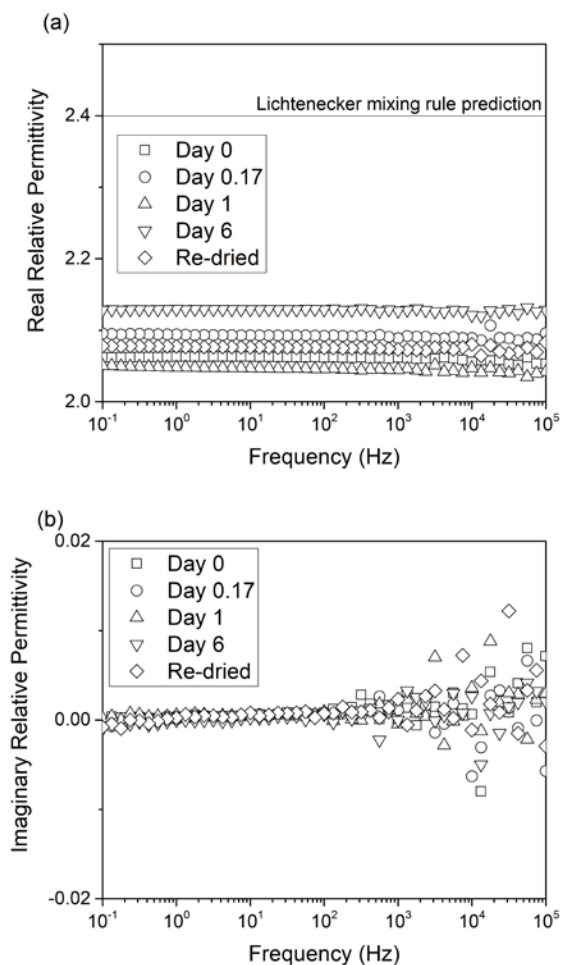
Figure 4. The (a) real and (b) imaginary parts of the relative permittivity of PP/EVA9-20/0 after different time intervals of immersion in xylene.

rotation of toluene molecules [21]; a great number of dielectric studies of systems containing benzene have been undertaken, but these focus on the microwave region. As such, the absence of any detectable relaxations in the systems containing absorbed xylene is unsurprising.

Figure 5 shows dielectric data obtained from the two systems containing the organoclay. First, consider the data obtained prior to exposure to the xylene, from which it is evident that the addition of the organoclay has little effect on the measured absolute value of ε', with high frequency values falling in the range 2.0 to 2.2. At low frequencies, a small increase in ε' is evident for both PP/EVA9-20/2.5/D and PP/EVA9-20/5/D; this is accompanied by an associated increase in the imaginary permittivity, ε'' and may be associated with additional Maxwell-Wagner polarization processes that result from the presence of the polymer/organoclay interfaces. Nevertheless, it is evident that, like the xylene, addition of the organoclay does not, in itself, markedly affect the dielectric response of the system.

The effect of organoclays on the dielectric response of a wide range of different polymer systems has been reported in the literature. Izci *et al.* [22] report that, at 10^{-2} Hz the, permittivity of their polypropylene-based nanocomposite increases with organoclay loading – a peak that had already been shifted to higher frequency due to the polymer-clay

interface. Elsewhere, Zazoum *et al.* [23] report similar findings on a polyethylene-based nanocomposite. In both cases, without the organoclay, the base polymer showed no dielectric relaxations.

However, when both xylene and the organoclay are present, dramatic changes in the dielectric response of the system are evident, despite the fact that the results presented above demonstrate that neither, in isolation, exerts a marked influence. Specifically, a pronounced increase in ε' is evident with decreasing frequency which, from the imaginary data, appears to be associated with two processes, a distinct dielectric loss process around $10-10^3$ Hz, plus a low frequency dispersion which may either be related to Maxwell-Wagner polarization or to increasing DC conduction. From the non-monotonic temporal changes seen in the strength of these processes, we suggest that such variations are an artefact of the chosen sample handling methodology. As discussed above in connection with Figure 3, absorption (and therefore presumably desorption) of xylene occurs relatively rapidly in these systems; consequently, by ensuring that no xylene remained on the sample surface prior to making measurements, varying degrees of desorption occurred during the 30 min drying time over which the sample was exposed to the uncontrolled ambient laboratory environment. Comparison of the dielectric data in Figure 5 obtained from the PP/EVA9-20/X/D and PP/EVA9-20/X/R samples indicates the presence of the increased low frequency losses implying that some xylene, nevertheless, remained entrained within the latter systems.

Many different workers have considered the effect of water absorption on the dielectric response of nanocomposites, since the consequences of exposure to atmospheric water is an important practical consideration [12, 13]. For example, Zhang *et al.* [12] exposed a range of nanocomposites containing nanoalumina to ambient conditions and found that in both the epoxy and polyethylene based systems, a water-related relaxation process resulted at the polymer-filler interface. Most notably, in the absence of nanoalumnina, the relaxation of water was significantly slower in the epoxy matrix and not observed in the polyethylene matrix, a change that is ascribed to the nature of the polarity of the polymer – in polyethylene, being, non-polar the concentration of water will be very low. Elsewhere, Lau *et al.* [13] considered the effect of water uptake in a morphologically-designed polyethylene blend containing nanosilicas with different surface chemistries that revealed, with increasing water uptake: (a) increased permittivity values; (b) the development of dielectric loss processes that increased in strength and frequency. These effects were ascribed to accumulation of water molecules at nanoparticle/matrix interfaces, whereupon the variations seen are a consequence of changes in the aggregation state of the interfacial water and the different interactions that consequently result. That is, initially, water molecules are tightly bound to the polar nanoparticle surface through hydrogen bonding while, as the water content increases, the influence of direct interfacial interactions decreases as more loosely bound water structures form. These are,
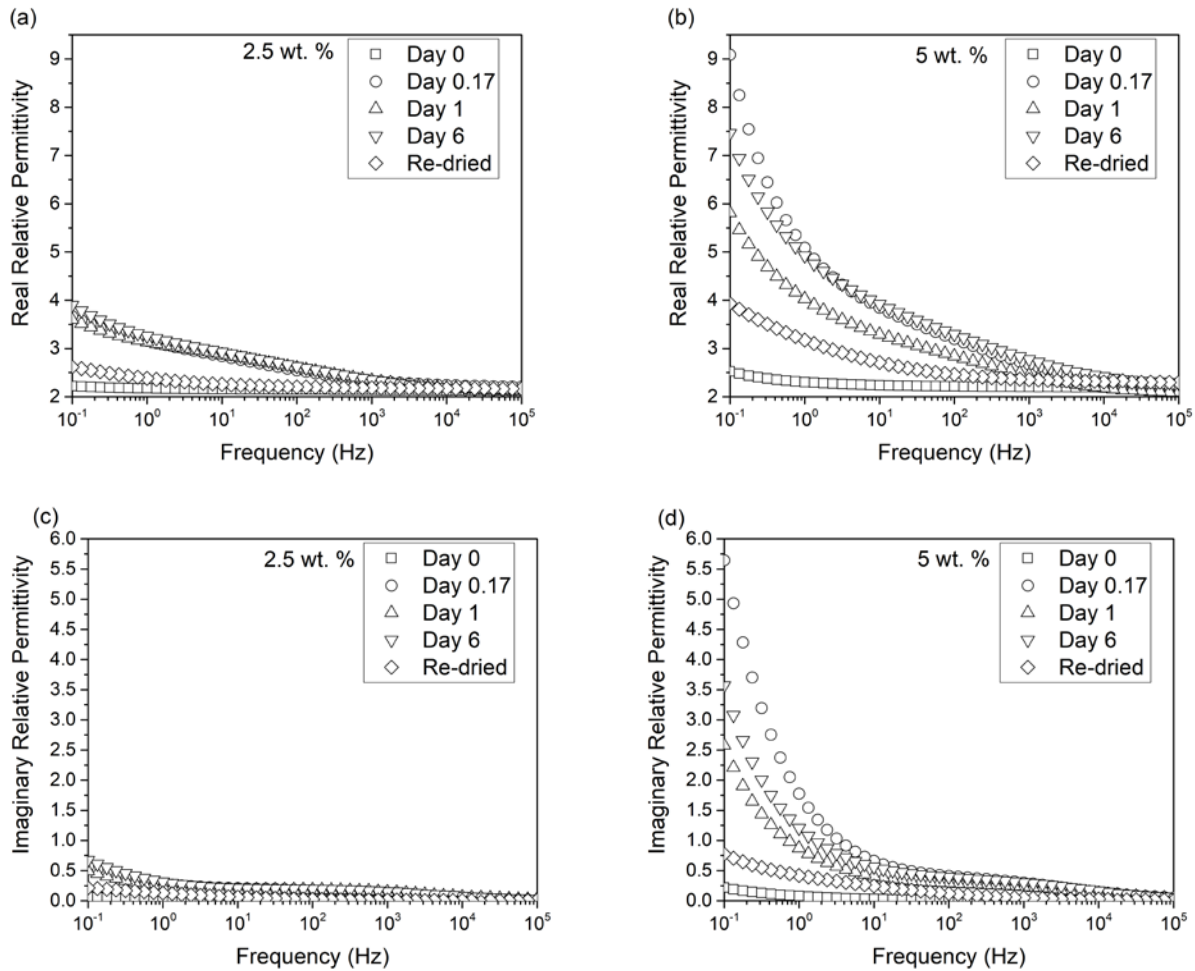
Figure 5. The real relative permittivity of the composites containing (a) 2.5 wt. % and (b) 5 wt. % organoclay and the imaginary permittivity of the composites containing (c) 2.5 wt. % and (d) 5 wt. organoclay after different time intervals of immersion in xylene.

consequently, characterized by the associated peaks being displaced to higher frequencies. In the work of Lau *et al.* [13], the absorbed species directly interacts with the applied electric field and the reduction in the frequency of the response, from the characteristic dielectric response of free water in the GHz range, to values in the hertz to kilohertz range due to local environmental effects. While the data shown in Figure 5 exhibit certain parallels, the data shown in Figure 4 clearly demonstrate that, in these results, the dielectric response of the overall system is not due to the presence of any single component. Rather, since the observed dielectric loss processes require the presence of both xylene (dielectrically inactive, in isolation, from Figure 4,) and the organoclay (dielectrically inactive from Day 0 data in Figure 5), we therefore suggest that the dielectric effects shown above occur as a result of organoclay-xylene interactions. That is, the presence of the xylene effectively solvates dipolar species in the clay so facilitating a molecular response to the applied field and, thereby, leading to the increased losses that are observed. In general, organoclays contain amphiphilic molecules that are introduced within the galleries, both to increase compatibility with the host matrix and to increase the interlayer spacing and, thereby, ease exfoliation [10]. In such systems, the VA from the copolymer EVA interacts with the silicate clay layers, while the non-polar chains on the organoclay promote compatibility with non-polar matrix polymers. As such, we suggest a potential molecular explanation for increased losses seen in Figure 5 relates to solvation of the compatibilizer by the absorbed xylene.

3.3 DIELECTRIC BREAKDOWN

Figure 6 contains representative AC breakdown data obtained from the three PP/EVA9-20/2.5/Y specimens, presented in the form of Weibull plots, while parameters derived from such an analysis for all systems are presented in Table 1; the quoted uncertainties in the Weibull scale parameter, correspond to 90 % confidence intervals.

In the case of the unfilled samples, where the variation in breakdown strength seen is comparable to the uncertainties in these measurements, which indicates that, in isolation, the presence of the xylene in PP/EVA9-20/0/I does not significantly affect the breakdown strength. However, in the systems containing the organoclay, the presence of the xylene, which in the unfilled system is benign, leads to a significant reduction in breakdown strength, indicating that interactions between the xylene and the organoclay serve to promote a mechanism that causes breakdown to occur at a reduced electric field. This phenomenon is similar to work
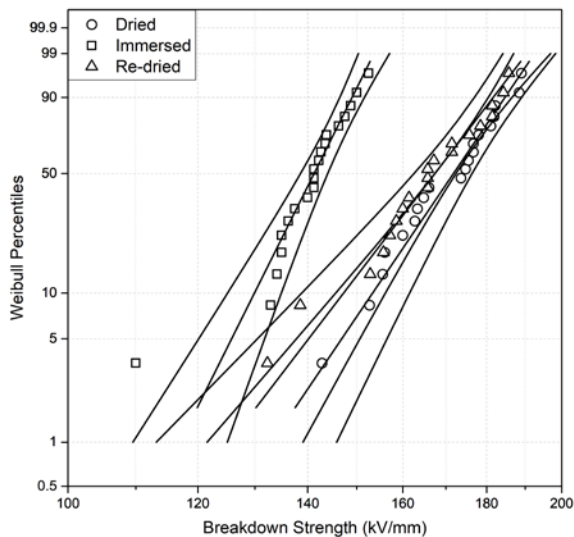
Figure 6. Representative Weibull plots, here comparing data obtained from the PP/EVA9-20/2.5/Y sample set.

described elsewhere, which reported on the effects of absorbed water on breakdown strength [4]. In the case of water, it is possible to propose a number of mechanisms by which its presence could adversely affect breakdown strength, involving dipolar and/or ionic processes; here, the critical role played by the nonpolar xylene combined with the mediating effect of this species in increasing dielectric losses at 50 Hz (the frequency of the dielectric breakdown testing), suggests that the former is most pertinent. In all cases, the re-dried samples exhibited a breakdown strength that was somewhat lower than their respective, initial, dried samples which is again consistent with the marginally increased dielectric losses seen in Figure 5 for the PP/EVA9-20/X/R systems.

To explore further the origins of the marginally reduced breakdown strength and increased dielectric losses seen in PP/EVA9-20/X/R materials, these systems were subject to further analysis. Measurements of the change in sample mass during drying following immersion in xylene showed that, after one week, the mass had returned to the original level, which indicates that the mass of any retained xylene is less than the associated experimental uncertainties. FT-IR analysis of the re-dried sample also revealed no evidence for retained xylene. In concert, these observations lead to another possibility, namely that the electrical effects seen are not associated with retained xylene but are, rather, caused by the xylene immersion process inducing some irreversible

Table 1. Summary of AC Breakdown data of the samples.

Sample	Weibull Scale Parameter / kV/mm	Shape Parameter
PP/EVA9-20/0/D	144 ± 8	6.9
PP/EVA9-20/0/I	140 ± 6	9.3
PP/EVA9-20/0/R	136 ± 6	8.6
PP/EVA9-20/2.5/D	176 ± 4	16.6
PP/EVA9-20/2.5/I	144 ± 4	22.5
PP/EVA9-20/2.5/R	171 ± 6	14.7
PP/EVA9-20/5/D	160 ± 6	10.5
PP/EVA9-20/5/I	133 ± 4	13.9
PP/EVA9-20/5/R	153 ± 6	11.9

change in the system. In view of the proposal that absorption of xylene can solvate components within the systems, a possible implication of this is that extraction of such species occurs during immersion in xylene. To explore this possibility, the xylene was examined by FT-IR after removal of the above specimens. In addition, pristine organoclay was also soaked in xylene before FT-IR analysis specifically to consider the possibility of compatibiliser extraction. None of these experiments provided any evidence for changes in the chemical composition of the xylene.

4 CONCLUSIONS

The influence of a labile, nonpolar molecule on dielectric response and breakdown behavior has been examined in a polymer blend system containing varying loading levels of an organoclay. In the absence of xylene, addition of the organoclay has little effect on the dielectric response of the system and, at low loading levels, leads to a significant increase in the breakdown strength of the system. Equally, in the absence of the organoclay, the presence of xylene has an insignificant effect on both dielectric response and breakdown. However, when both the organoclay and the xylene are present together, strong synergistic effects are seen. Specifically, the dielectric losses are markedly increased and, in parallel, the AC breakdown strength is reduced. We propose that these effects are associated with solvation of amphiphilic species that are added to the virgin clay in order to render it more compatible with the host polymer and ease dispersion during processing.

While it is generally accepted that in nanocomposite systems optimal properties require good dispersion of the nanoclay, the work presented here, we suggest, indicates that where dielectric and electrical properties are concerned, improved dispersion is not the panacea it may be elsewhere. Rather, the mechanisms that determine bulk properties can be influenced by local physical and chemical factors. While it is conceptually simple to envisage how absorbed water can couple with an applied electric field and, thereby, affect dielectric response, it is less obvious that the presence of a nonpolar species can serve to facilitate comparable effects. Here, the presence of the nonpolar xylene leads, indirectly, to phenomena that mirror the direct effects of absorbed water. While both are, practically, deleterious, this does not eliminate the possibility of other labile species acting in some advantageous way, by which macroscopic electrical properties are enhanced.

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BIOGRAPHIES

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Thomas Andritsch (M'11) was born in Innsbruck, Austria in 1980. He received the Dipl.-Ing. Degree in Electrical Engineering from Graz University of Technology in 2006 and his Ph.D. degree in the same field from Delft University of Technology in 2010. He is currently working as a Lecturer at the University of Southampton with focus on advanced materials for high voltage applications and plant. He has extensive experience with preparation and testing of polymer-based electrical insulation materials, including nanodielectrics, electroactive polymers and syntactic foams, as well as liquid insulation systems and nanofluids. Thomas is currently the chair of the UK & Ireland IEEE DEIS Chapter. He is member of the IEEE DEIS working group on nanodielectrics, developing recommended practices for nanocomposite synthesis and preparation, and was contributing to Cigre WG D1.40 on the subject of functional nanomaterials for the electrical power industry.

Alun Vaughan (SM'07) has a B.Sc. degree in chemical physics and a Ph.D. degree in polymer physics. After working at the UK's Central Electricity Research Laboratories and spending a period as an academic at The University of Reading, he is now Professor of Dielectric Materials at the University of Southampton. He is a former chair of The Dielectrics Group of the Institute of Physics, a Fellow of the Institute of Physics, a Fellow of the IET and delivered the 2016 IEEE DEIS Eric O. Forster Memorial Lecture.