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On the influence of xylene on the dielectric response of an organoclay-containing nanocomposite

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Abstract: The dielectric effect, investigated using dielectric spectroscopy and DC dielectric breakdown strength measurements, of introducing xylene into a composite system containing polyethylene, a co-polymer of ethylene and vinyl acetate and an organoclay can be understood in light of X-ray diffraction data. In dielectric spectroscopy, although organoclay alone changes the dielectric response of the polymer blend and xylene has no effect on the unfilled polymer blend, when both xylene and organoclay are present a synergistic response is revealed at the 1 V_{rms} amplitude voltage used to acquire the dielectric data. In contrast to this, in DC dielectric breakdown strength measurements revealed that, under high field conditions, both the xylene and organoclay, independently, caused a decreased breakdown strength. This work was undertaken in order to examine the generality of the possible effects of labile, low molar mass impurities on electrical properties of comparable systems, which may be processed through solvent-based routes.

1. Introduction

Nanocomposites have attracted much attention as a consequence of the desirable combination of properties that such systems may exhibit. Although much of the early interest focused on mechanical performance [1], since the concept of nanometric dielectrics was first discussed by Lewis in 1994 [2], interest in the influence of nanofillers on electrical parameters has grown massively [3]. While it is generally agreed that the mechanisms by which the presence of a nanofiller may influence overall macroscopic properties are related to local interactions, the high specific interfacial area that is a characteristic of all nanocomposites can lead, indirectly, to adverse consequences. For example, the exposure of material systems to environmental water during their lifetime has led a number of researchers to consider the influence of this on dielectric properties [4–7]. Hosier *et al.* [4] reported that the presence of water in polyethylene (PE) nanosilica nanocomposites reduced the direct current (DC) dielectric breakdown strength significantly more than in their control sample without nanosilica. Elsewhere, Fabiani *et al.* [5] investigated the effect of water on organoclay-ethylene/vinyl acetate (EVA) nanocomposites and found that the nature of the organoclay was significant in influencing the considered electrical properties, which included dielectric relaxation, DC conduction and DC breakdown strength. Overall, such findings have provided a better understanding of how the materials will perform *in-situ* for their respective applications and, in particular, have demonstrated that the presence of small quantities of water – which can be considered as a labile *polar* impurity – can markedly affect the properties of nanocomposites.

One commonly used strategy for the production of nanocomposites involves dispersion of one or more of the components in a suitable solvent and, as a result of this, it is conceivable that some residual solvent may be retained within the system after nanocomposite recovery. Indeed, where prior dispersion of a nanofiller is undertaken in a solvent, favourable interactions are a requirement, such that retention of adsorbed solvent is not unreasonable on thermodynamic grounds. Nevertheless, unlike in the case of water, the influence of retained organic molecules has seen little investigation, despite the fact that, in other contexts [8], it is well appreciated that organic moieties can markedly influence electrical factors, notably, charge transport dynamics. Our previous investigation of the effect of xylene on a polypropylene (PP) based system revealed a synergistic relationship between the organoclay and the xylene, despite the uptake of xylene being independent of the organoclay presence [9]. Herein, we seek to extend this work to examine the effects of xylene on a PE-EVA-organoclay nanocomposite, in order to establish whether the results seen in the aforementioned study are principles that can be applied to other material systems or, rather, are material specific and determined by factors such as the precise phase structure of the matrix or the dispersion of the nanofiller.

2. Experimental

In this investigation, the following materials were used as supplied: low density PE (LDPE: LD100BW, Exxon Mobil Chemicals); high density PE (HDPE: Rigidex HD5813EA, BP Chemicals); EVA containing 9 wt. % of vinyl acetate (Elvax 750, DuPont); a montmorillonite (MMT) organoclay containing 35-45 wt. % of an organic compatibiliser (682624, Sigma Aldrich). The ratio of LDPE:HDPE was always 80:20 and will be referred to, for simplicity, as PE. The blend of LDPE and HDPE was employed herein, as elsewhere, due to its ability to form a continuous space-filling lamellar morphology where key properties are independent of variations in nucleation density [10–12]. The ratio of PE:EVA was always 80:20.

The material preparation procedure was adapted from published work [13]. First, the organoclay was sonicated in xylene for 30 min before being added, with the EVA, into a rotary evaporator and heated to 140 °C. When the polymer had completely dissolved, the LDPE and HDPE were added

and mixed until these components had also dissolved. After stirring for 10 min, the resulting mixture was put under vacuum to remove the xylene, before the product was finally dried under dynamic vacuum at 70 °C for 6 d. A hydraulic press set at 140 °C was used to produce sample films nominally 100 μm in thickness, which were quenched from the melt phase in order to generate as simple a matrix morphology as possible.

All of the resulting specimens were then further dried for one week under dynamic vacuum at 70 °C, to minimise residual solvent and to prevent absorption of environmental water. These initial samples will hereafter be referred to as "dried". Further batches of identical samples were then prepared as follows: (a) by immersion in a room temperature xylene bath for 4 d (termed "immersed" samples); (b) by immersion in xylene for 4 d in a room temperature bath followed by drying under dynamic vacuum at 70 °C for 4 d (termed "re-dried" samples). The resulting samples will hereafter be referred to using the following nomenclature: PE/EVA/X/Y where X is the organoclay loading (either 0 or 5 wt. %) and Y is the conditioning (D, dried; I, immersed; R, re-dried).

3. Material Characterisation

Fourier-transform infrared (FT-IR) spectra were acquired using a Thermo Scientific Nicolet iS5 instrument operating in transmission mode. X-ray diffraction (XRD) data were obtained on a Rigaku SmartLab instrument, operating in reflection mode, from $2\theta = 1.5°$ to $8°$ in steps of $0.02°$. DC breakdown strength was measured as follows: the sample was immersed in silicone oil between vertically opposed spherical electrodes (6.3 mm in diameter) and the applied voltage was increased at a rate of 350 V s^{-1}. The resulting breakdown data were analysed assuming 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 analyser system. For this, an AC voltage of amplitude 1 V$_{rms}$ was applied across electrodes 30 mm in diameter with a guard-ring; a frequency range of 10^{-1} to 10^5 Hz was considered. When testing the immersed samples, 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.

4. Results

4.1. Xylene Absorption and Desorption

Fig. 1 shows the variation of sample mass with xylene immersion time for material systems PE/EVA/0/D and PE/EVA/5/D. From these data, it is evident that absorption occurs rapidly before plateauing at levels equating to around 9 wt. % of the initial sample mass in the case of PE/EVA/0/I and 10 wt. % for PE /EVA/5/I. However, the uncertainties in these values are such that these numerical differences are not statistically significant, indicating that the absorption of xylene is not significantly affected by the presence of the organoclay. Xylene, being a good solvent for PE and EVA, is readily compatible with both these polymers and, presumably,
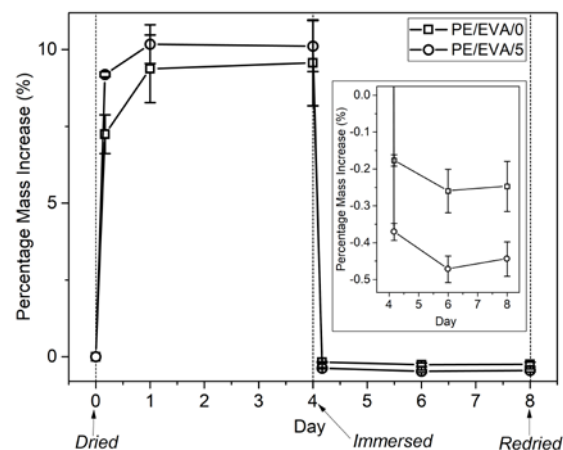

Fig. 1. *Variation of sample mass for the PE/EVA/0 and PE/EVA/5 systems during immersion in xylene and subsequent drying; the indicated error bars correspond to ± one standard deviation. The inset highlights the different behaviour of the two systems upon re-drying*

with the organic fraction of the organoclay. This assertion is consistent with comparable published work [9].

Following 4 d of xylene immersion, the samples were dried under vacuum, whereupon the mass of the PE/EVA/0/R and PE/EVA/5/R samples fell, respectively, to 0.25 wt. % and 0.45 wt. % *below* the initial values and were statistically separable (see error bars on inset in Fig. 1). This suggests that some fraction of each system was extracted during the solvent exposure and that this occurred to a greater extent in the system containing the organoclay. Given that xylene is chemically comparable to toluene, which has been used elsewhere as a solvent etchant for EVA [14], it is plausible that xylene could remove some fraction of the EVA. Additionally it is possible that this prolonged immersion in xylene has extracted some of the intercalant organic modifications from the organoclay, despite the organoclay previously being exposed to xylene during sample processing. Indeed, Ho *et al.* [15] investigated the extraction of excess organic moieties reported to be present by the industrial provider of their organoclay and found that removal took 3 d under ethyl alcohol reflux, conditions not matched by our solution blending synthesis but more likely to parallel prolonged xylene-immersion. Elsewhere, Elban *et al.* [16] studied washing procedures to remove un-bound organic moieties and reported results consistent with the removal of weakly- and un-bound moieties at clay edge surfaces and/or external basal planes.

Figs. 2a and 2b contain FT-IR results obtained from the PE/EVA/0/Y and PE/EVA/5/Y sample sets respectively; in both figures, the FT-IR spectrum of xylene is also included for reference purposes. From the traces labelled 1, obtained from xylene, it is evident that the FT-IR spectrum of this solvent contains a number of characteristic peaks within the spectral region shown, which fall within relatively featureless regions of the spectrum of both PE/EVA/0/D and PE/EVA/5/D (traces labelled 2). Consequently, the presence of xylene in the PE/EVA/0/I and PE/EVA/5/I samples (traces labelled 3) is readily detected. In the case of PE/EVA/0/R and PE/EVA/5/R (traces labelled 4), weak absorption around 690 cm^{-1} and between 740 cm^{-1} and 820 cm^{-1} suggest that a small amount of xylene is retained, despite the samples being stored
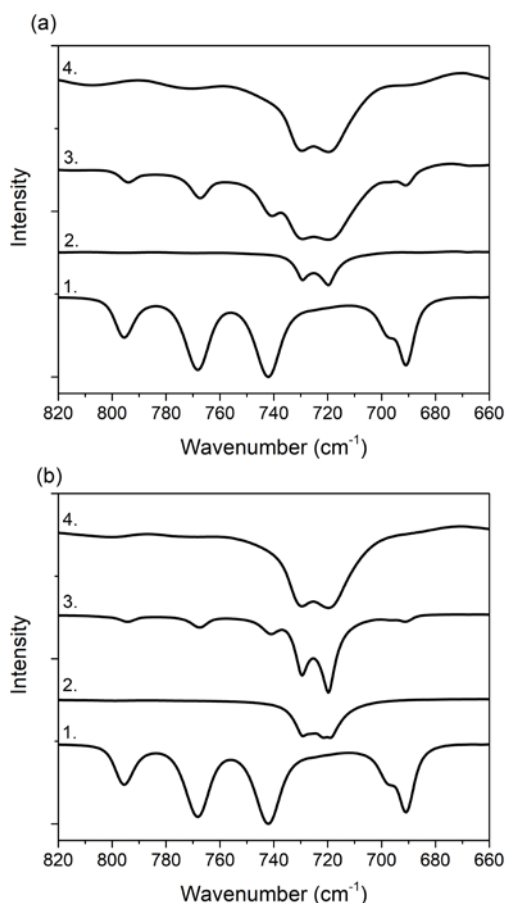
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Fig. 2. *FT-IR data obtained from (**a**) the PE/EVA/0/Y and (**b**) the PE/EVA/5/Y sample sets. In both, the four traces shown are offset for clarity and correspond to: 1, xylene solvent; 2, the initial, dried samples; 3, the immersed samples, after removal from the xylene; 4, the final, re-dried samples*

Fig. 3. *Offset XRD data obtained from the PE/EVA/5/Y sample set, compared with equivalent data obtained from the organoclay itself, and a vertical line to aid interpretation*

under dynamic vacuum at 70 °C for 4 d and Fig. 1 suggesting that the mass change had stabilised. However, careful examination reveals shifts in these xylene-related peak maxima in the re-dried samples compared with both the xylene solvent itself and the immersed specimens. We associate this effect with local molecular interactions that influence the relevant vibrational modes; elsewhere, effects such as hydrogen bonding, variations in local disorder and vibronic coupling have been reported to shift and/or broaden FT-IR peaks in a manner akin to that seen here [17,18]. This is further supported by the absence of any shifts in the peaks corresponding to PE and EVA.

In discussing the XRD data presented in Fig. 3, first consider the dominant feature that falls between $2\theta = 2.0°$ and $2\theta = 4.0°$ in all diffraction patterns, which stems from the interlayer periodicity of the smectic organoclay structure. The uppermost diffraction pattern shown in Fig. 3 was obtained from the as-received organoclay. The presence of the strong peak around $2\theta = 3.0°$ indicates an interlayer periodicity, d_{001}, of 2.9 nm, which is in line with expectations [19,20]. Organoclays generally contain amphiphilic ionic moieties, frequently taking the form of alkyl ammonium salts [21,22], which are introduced into the clay galleries to increase the interlayer separation in order to ease exfoliation/dispersion and to improve compatibility with the matrix polymer. As such, this value is consistent with the quoted presence of
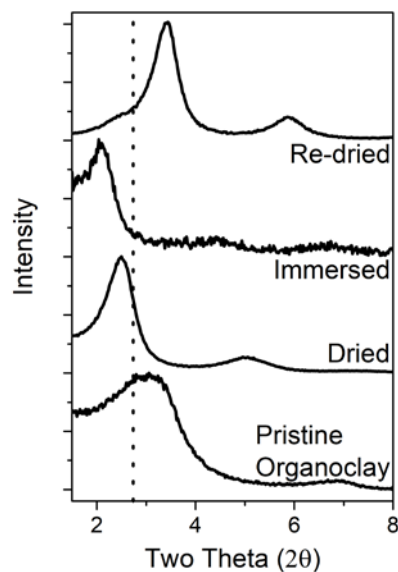
~40% of compatibiliser in this system; in the absence of such species, MMT (i.e. the base smectic phyllosilicate) exhibits an interlayer spacing around 1.5 nm [23].

In the case of PE/EVA/5/D, the equivalent peak is displaced to around $2\theta = 2.5°$, indicating a basal spacing, d_{001}, of 3.5 nm, which is expanded compared to the organoclay, as expected from published literature [20,24]. The peak of PE/EVA/5/I, around $2\theta = 2.1°$, indicates a further increase in the basal spacing to 4.2 nm, which can be attributed to diffusion of the xylene into the system as a consequence of favourable thermodynamic interactions [25,26] leading to swelling of the organoclay [15]. Comparison of the above XRD data with those obtained from PE/EVA/5/R reveals that the position of the d_{001} scattering is displaced to a higher angle in PE/EVA/5/R. This indicates a reduction in interlayer periodicity to about 2.6 nm, which implies that the xylene immersion has caused some permanent physical change to the structure. In light of published literature showing that an organoclay was preferentially intercalated by EVA in a PE-EVA blend [27] and that EVA can be extracted from a PP/EVA blend by a solvent similar in nature to xylene [14], the above changes can, *in toto*, be interpreted as follows. During immersion, swelling of the organoclay by xylene occurs in parallel with extraction of a fraction of the organic intercalants (EVA and/or a fraction of the amphiphilic compatibiliser), such that the interlayer periodicity increases. However upon removal of the solvent during the subsequent drying process, the interlayer distance decreases and becomes smaller than before the xylene treatment.

In the XRD data obtained from PE/EVA/5/D and PE/EVA/5/R, a weak and broad second peak ($2\theta = 5.0°$ and $2\theta = 5.9°$ respectively) can be seen located at close to twice the scattering angle of their respective d_{001} peak. Such features have been reported in previous studies and associated with second order scattering from the smectic structure of the clay. We, therefore, similarly consider this to be the d_{002} peak [21]. Similarly, and since the graph for the PE/EVA/5/I sample has effectively been shifted to the left, within the 2θ

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range investigated both the d_{002} and d_{003} peaks can be seen at $2\theta = 4.3°$ and $6.7°$ respectively; suggesting long range order of the intercalated clay structure [28,29]. Significantly though, d_{002} and d_{003} scattering in PE/EVA/5/I appears to be weak and therefore present only in small amounts, indicating a greater degree of disorder when the clay is swollen with xylene.

4.2. Dielectric Relaxation

Dielectric spectroscopy data obtained from the PE/EVA/0/Y and PE/EVA/5/Y sample sets are shown in Fig. 4. From Fig. 4a, it is evident that all the systems containing no organoclay are characterised by a real relative permittivity, ε', value in the range 2.27 - 2.32 and a very weak dependence of this parameter on frequency. The reported value of ε' for PE is 2.3 at 1 kHz [30], which is consistent with these data, indicating that the presence of neither the vinyl acetate co-monomers (~2 wt. %), nor the absorbed xylene in the case of PE/EVA/0/I (~10 wt. %), serves materially to affect the value of this parameter. Indeed, the real relative permittivity of para-xylene is close to that of PE, with a value of 2.26 at 1 kHz [31]. Zazoum et al. [32] considered the dielectric response of an 80:20 LDPE:HDPE blend (the same ratio as used herein, albeit that the polymers were obtained from different suppliers) and reported that ε' was, as here, independent of frequency over the range investigated. The results presented in Fig. 4b indicate a slow rise in the imaginary part of the relative permittivity, ε'', and while comparable behaviour has been attributed to the polar carbonyl groups of the vinyl acetate in EVA [33], the fact that the magnitude of these losses is at the sensitivity limit of our measurement equipment makes their statistical significance questionable. To conclude, the data presented in Figs. 4a and 4b are consistent with published results and demonstrate that the presence of xylene has no observable effect on the dielectric response of our polymer system, when investigated at an applied AC voltage of amplitude 1 V_{rms}.

Consider, now, the data obtained from PE/EVA/5/D, which are presented in Figs. 4c and 4d. Comparison of these with the results shown in Figs. 4a and 4b indicate that the presence of the organoclay alone results in a significant modification to the dielectric response, which is most easily seen in Fig. 4d. By plotting Fig. 4d on log/log axes, it is clear that, for PE/EVA/5/D, ε'' increases progressively with decreasing frequency up to a value of 0.1 at 0.1 Hz. In dielectric data, charge transport processes manifest themselves in a dependence of the form:
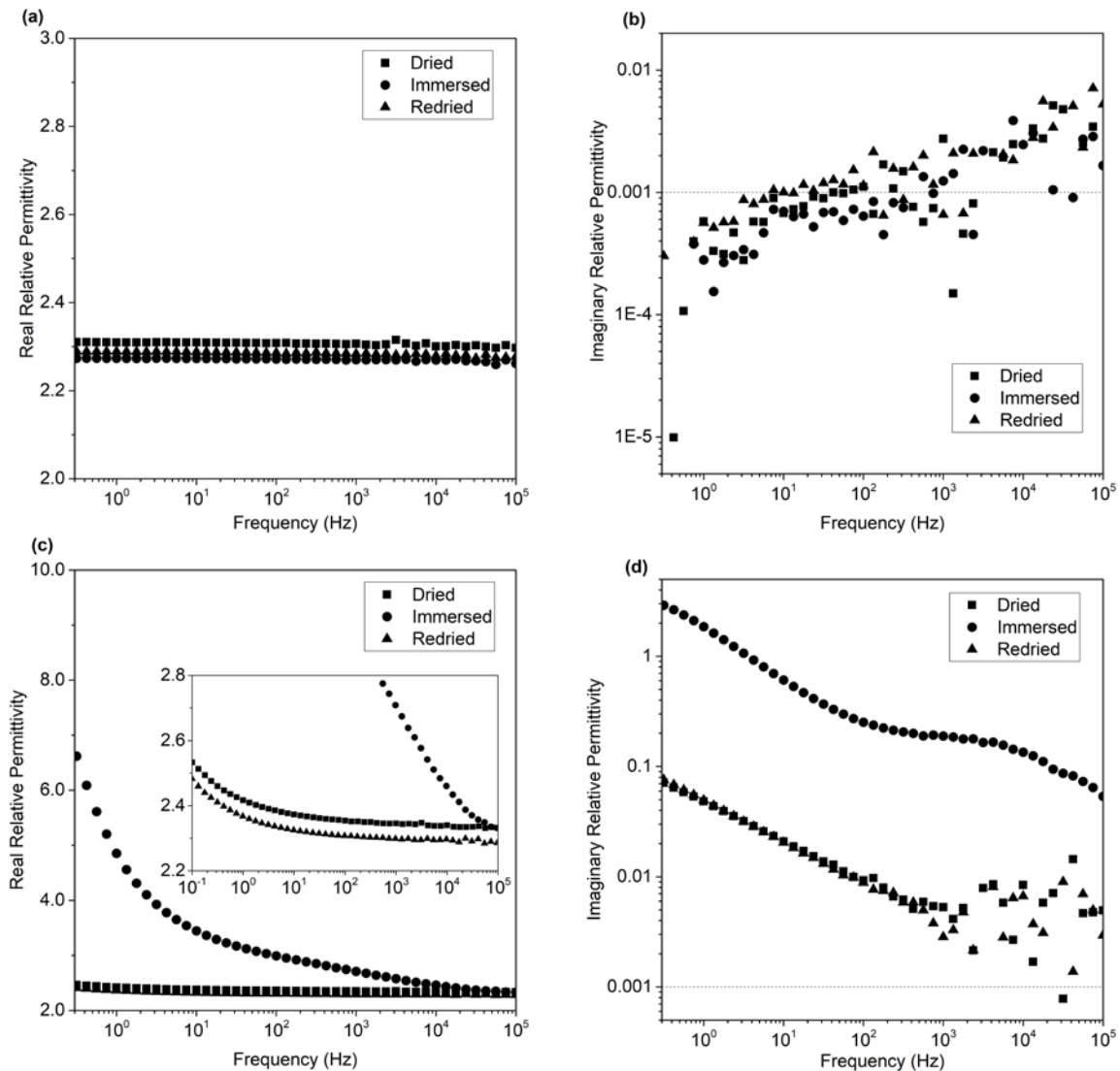
$$\varepsilon'' \propto \omega^{-1}$$

Fig. 4. *Real and imaginary relative permittivity dielectric spectroscopy data for PE/EVA/0/Y samples in (a) and (b) and PE/EVA/5/Y samples in (c) and (d) respectively*

where ω is frequency [34,35]. In a nanocomposite composed of polylactide and an MMT-based organoclay, Pluta *et al.* [34] reported a comparable behaviour to that seen here, in which the exponent in the above equation was between -1 and 0. This, they attributed to a combination of charge transport, electrode polarisation and interfacial polarisation processes. Elsewhere, Fabiani *et al.* [5], described the dielectric behaviour of EVA containing an organically-modified fluorohectorite clay and reported a loss peak at around 10 – 20 Hz, which they attributed to dielectric processes at

Table 1 Summary of the Weibull distribution results of the DC dielectric breakdown data together with the sample thicknesses to aid interpretation of the results

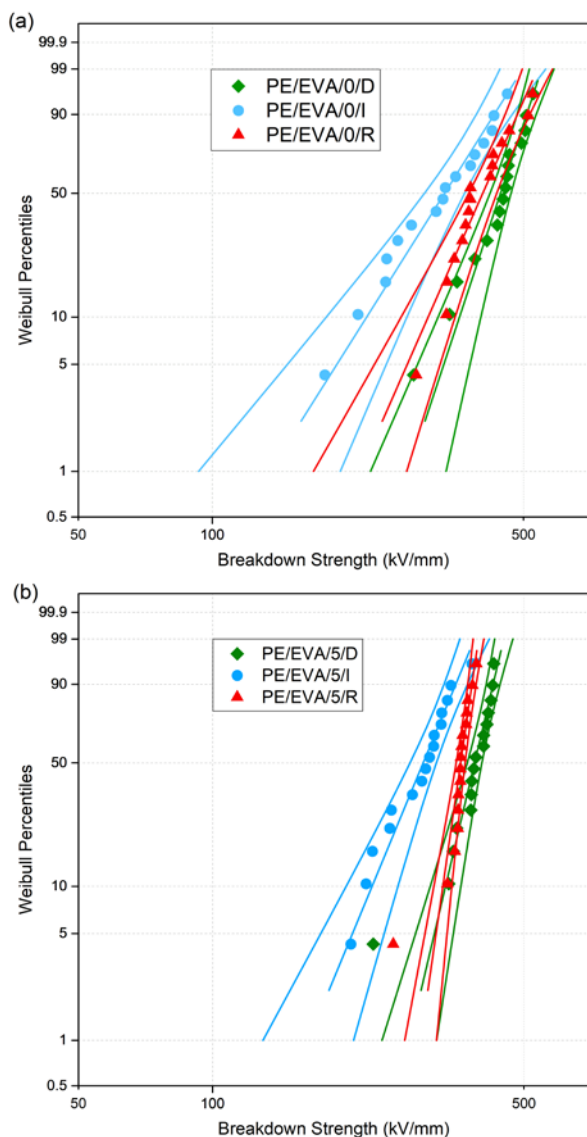| Sample	Scale Parameter / kV/mm	Shape Parameter	Sample Thickness / mm
PE/EVA/0/D	463 ± 22	8.9	0.09
PE/EVA/0/I	360 ± 33	4.7	0.10
PE/EVA/0/R	427 ± 28	6.7	0.09
PE/EVA/5/D	399 ± 13	12.6	0.10
PE/EVA/5/I	313 ± 19	7.1	0.10
PE/EVA/5/R	367 ± 8	20.5	0.09

Fig. 5. *Weibull plots of the DC dielectric breakdown data for (**a**) PE/EVA/0/Y and (**b**) PE/EVA/5/Y*

polymer-filler interfaces. As such, we suggest that, the variations ε' and ε'' that are evident in Figs. 4c and 4d. result from a combination of dipolar and charge transport processes.

Comparison of the dielectric behaviour of PE/EVA/5/D and PE/EVA/5/R reveals equivalent behaviour, which indicates that any differences between these two systems, caused by immersion in xylene, is negligible from a dielectric perspective. In contrast, the behaviour of PE/EVA/5/I is very different, suggesting that the xylene and organoclay act synergistically to influence the dielectric response of the system at the applied AC voltage (1 V_{rms}) used in these measurements. Specifically, data obtained from PE/EVA/5/I is characterised by a much more pronounced increase in ε' with decreasing frequency than for either PE/EVA/5/D or PE/EVA/5/R, a general shift in ε'' to higher values, and the appearance of specific relaxation processes around 0.1 Hz and 10^4 Hz. As discussed above, an increase in ε'' with decreasing frequency is generally associated with charge transport effects and/or Maxwell-Wagner interfacial polarisation and comparison of the behaviour of PE/EVA/5/I with the other two systems, shown in Fig. 4d, reveals that such effects are increased by the presence of xylene by a factor of 30. In the case of organoclays, the presence of the ionic compatibiliser moieties provides a potential molecular origin for the effects identified above. In the presence of xylene, solvation of these ionic species occurs, together with swelling of the galleries, as seen in Fig. 3. The consequence of both these factors is enhanced mobility of compatibiliser ions, which facilitates coupling with the applied electric field more strongly than for samples containing organoclay but no xylene. Furthermore, given the observed reduction in organoclay periodicity shown by the XRD data in Fig. 3 for PE/EVA/5/R compared with PE/EVA/5/D, it is feasible that during the immersion step extraction of some compatibiliser moeities may occur, which may then result in increased ionic conduction. Nevertheless, we suggest that a major contributory factor in the strongly increased losses seen across the complete frequency spectrum when both the organoclay and xylene are present is solvation of ionic/diploar species. This solvation enhances mobility and, hence, serves to facilitate coupling of such moieties with the applied AC voltage (1 V_{rms}). Indeed, the nature and dispersion of the compatibiliser has been shown to produce different permittivity traces, highlighting its role in interfacial polarisation in dielectric spectroscopy [5,36].

4.3. Electrical Breakdown

To explore the above effects further, the influence of material composition in general, and the presence of xylene in particular, DC dielectric breakdown behaviour was considered. Fig. 5 compares DC breakdown data obtained from all of the systems considered here, presented in the form of Weibull plots; derived data are listed in Table 1. Comparison of the behaviour of the PE/EVA/0/Y sample set with equivalent data obtained from PE/EVA/5/Y reveals that, irrespective of pre-conditioning regime, the inclusion of 5 wt. % of the organoclay consistently results in a reduction in the Weibull scale parameter by ~60 kV/mm. From the dielectric data presented above, it is evident that the inclusion of the organoclay increases the low frequency losses, which we have partially ascribed to enhanced charge transport. As such, we suggest that the observed ~60 kV/mm reduction in DC breakdown results directly from increased DC conduction

in the presence of the organoclay. Comparable relationships between DC conductivity and DC breakdown strength have been reported elsewhere [37].

Comparison of the DC breakdown strength of PE/EVA/0/D and PE/EVA/0/R or PE/EVA/5/D and PE/EVA/5/R demonstrates that subjecting these systems to xylene absorption and subsequent desorption has a statistically insignificant effect on the derived Weibull scale parameter, implying that any structural or compositional changes are electrically insignificant as far as breakdown processes are concerned, alike the permittivity findings presented above. However, the presence of xylene in both PE/EVA/X/I systems results in a reduction in breakdown strength of ~30%, which exceeds the uncertainties in these data. In the case of PE/EVA/5/I, this decrease in DC breakdown strength may be related to xylene facilitating enhanced charge transport through the mechanisms discussed above, which manifest themselves even at low applied voltages. That is, in this system, increased ionic conduction occurs involving solvated ions liberated from the organoclay; this is consistent with both the XRD and mass change data presented above. Furthermore, it is also conceivable that, in addition to those processes that can be detected at low voltages by dielectric spectroscopy, the xylene itself may directly influence charge transport processes at high applied voltages. Indeed, the adverse electrical consequences of acetophenone, cumyl alcohol and α-methyl styrene in cross-linked polyethylene [8,38,39] are well appreciated. By both mechanisms, increased conduction leads to reduced DC breakdown strength, as reported elsewhere [5,37,40]. This suggestion also provides a possible explanation for the comparable reduction in DC breakdown strength exhibited by PE/EVA/0/I. As such, while we do not discount the possible influence of labile solvated compatibiliser molecules in the case of PE/EVA/5/I, we suggest that xylene, like some technologically important organic moieties, also appears to have a direct influence on charge transport processes. In a similar way to [41], we suggest that studying the space charge behaviour and the field dependence of DC conductivity may provide further insights into the breakdown mechanism through an understanding about how various factors (notably, absorbed xylene) influence charge accumulation and transport.

5. Conclusion

This paper compares the effect of a non-polar, labile organic moiety on the electrical behaviour of a polyethylene-based blend and the same matrix system containing an MMT-based organoclay. In the absence of the organoclay, the presence of ~10 wt. % of xylene has no discernible effect on the low field dielectric spectroscopy results; whereas the inclusion of 5 wt. % of the organoclay results in increased low frequency dielectric processes, which we associate with a combination of interfacial polarisation effects and charge transport. Evidently, the presence of both the xylene and the organoclay has a massive impact on the permittivity results, which, we suggest, are related to synergistic effects associated with the solvent-enhanced mobility of the ionic/dipolar species within the system, including compatibiliser moieties in the organoclay. The presence of both the organoclay and the xylene – together and separately – both serve to reduce the measured DC breakdown strength,

presumably, through a range of mechanisms related to enhanced high field charge transport. Nevertheless, it is evident from the work presented that organic moieties retained within nanocomposite systems can markedly affect macroscopic properties, even though the molecules themselves may be nonpolar and appear therefore not to couple strongly with any applied electric field. As such, care needs to be exercised when preparing nanocomposites through solvent-based routes, to ensure that such processes do not affect the behaviour of the resulting system, complicating interpretation. Also, while the presence of xylene in the particular system considered here results in increased dielectric losses and reduced breakdown strengths – effects that are unlikely to be desirable – we suggest that it is conceivable that different solvent molecules and different polymers may act synergistically to give very different effects.

6. Acknowledgments

7. References

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