

ELECTRICAL AND MECHANICAL PROPERTIES OF NEW RECYCLABLE POWER CABLE INSULATION MATERIALS BASED UPON POLYETHYLENE BLENDS

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Abstract: Chemically crosslinked polyethylene (XLPE) has been used as electrical insulation for power cables since the 1970s due to its favourable combination of electrical and mechanical properties. However, as the electrical engineering community has become increasingly aware of the life cycle environmental impacts, XLPE has come under scrutiny for its lack of recyclability and the high process energies used in its manufacture. Although technologies are being developed to facilitate the re-use of XLPE at the end of its initial service life, the use of this material is inferior to fully recyclable and low process energy alternatives.

In this investigation, we concentrated on the use of binary blends of linear and branched polyethylene (LPE / BPE) as potential replacement materials for XLPE, since such systems have the potential to combine comparable mechanical properties and enhanced breakdown strength with good recyclability. We compare the thin film AC ramp breakdown behaviour of blends as a function of temperature up to 97 °C. These consist of the same BPE in virgin and crosslinked states and in a blend with 20wt% LPE. These data are augmented with dynamic mechanical analysis. In concert, these data indicate that with appropriate morphological control the blended thermoplastic material exhibits superior properties to XLPE under conventional operating conditions and may even be suitable for higher temperature operation than XLPE.

The paper will discuss the importance of polymer blending and blend physical properties in the context of the process requirements and the implications for cable manufacture and on cable electrical and environmental performance in comparison with XLPE.

1 INTRODUCTION

Chemically crosslinked polyethylene (XLPE) has been used as the electrical insulation of choice for many power cable designs since the 1970s, due to its favourable combination of electrical and mechanical properties. However, as the electrical engineering community has become increasingly aware of the life cycle environmental impacts, XLPE has come under scrutiny for its lack of recyclability and the high process energy burden involved in its manufacture. Although technologies are being developed to facilitate the re-use of XLPE at the end of its initial service life, such an approach is inferior to fully recyclable and low process energy alternatives [1].

Environmental consciousness has therefore led to the development of XLPE recycling technologies and to the consideration of novel thermoplastic alternatives [2]. Although such thermoplastic systems are generally based upon propylene copolymers, blends of polyethylene can also exhibit desirable combinations of properties. For example, isothermally crystallized polyethylene blends containing ~20% high density (HDPE) and ~80% low density polyethylene (LDPE), when crystallized at a suitable isothermal temperature,

have been shown to exhibit an increase in AC ramp breakdown strength of ~15%, and significantly retarded electrical tree growth, provided crystallization of the HDPE generates a space-filling lamellar texture [3-6]. The origin of these improved properties is related to both the lamellar crystal distribution that is formed and the location of defective chain segments and impurities, which remain uniformly distributed throughout the system rather than being concentrated at spherulite boundaries as usually happens when a single component system is crystallized [8-9].

In this paper, we discuss the importance of polymer blending on electrical and mechanical behaviour of a polyethylene blend system that had been subjected to non-isothermal crystallization. The AC ramp electrical breakdown strength is reported as a function of temperature, together with changes in mechanical response, as determined by dynamic mechanical thermal analysis (DMTA). Provided the HDPE crystals are able to prevent excessive long-term creep, the materials technology described could constitute a thermoplastic alternative to propylene-based systems.

2 EXPERIMENTAL

2.1 Sample Preparation

The HDPE used in this study was Dow HDPE 40055E and the LDPE used was a non-commercial material grade related to a commercial cable-grade system. Pellets for small-scale laboratory testing were generated using a laboratory twin screw lab extruder.

Thin film samples for breakdown testing were prepared by melting the above pellets between aluminium foils in a Graseby Specac hydraulic press equipped with a thin film making accessory to give disks ~85 μm in thickness. These were transferred into a Mettler Toledo FP82 hotstage, held at 200 $^{\circ}\text{C}$ for 2 min for residual stresses to relax and, finally, either quenched into cold water or crystallized in the hotstage from 130 $^{\circ}\text{C}$ to 90 $^{\circ}\text{C}$ at the required cooling rate..

2.2 Structural Characterisation

Non-isothermal crystallization studies of the above materials were conducted by differential scanning calorimetry (DSC), using a Perkin Elmer DSC7 system running Pyris software. Block specimens with equivalent thermal histories to those described above were also prepared in the DSC; these were cut open using a cryo-ultramicrotome and etched using standard procedures [10] before being examined in the scanning electron microscope (SEM).

2.3 Dielectric Strength

Dielectric breakdown testing was conducted using a testing chamber containing 20 cs silicone fluid that was, in turn, placed in an oven and partly immersed in silicone oil so as to provide a stable temperature environment. Vertically opposed ball bearings, 6.3 mm in diameter, were used as electrodes. These were regularly replaced in order to eliminate possible electrode pitting effects.

A 50 Hz signal was applied to the sample with an RMS amplitude ramp of 50 V s^{-1} until breakdown occurred. The resulting data were analyzed assuming a two parameter Weibull distribution [11,12] and appropriate parameters were calculated using the maximum likelihood estimation method using Weibull ++7 software from Reliasoft Corp

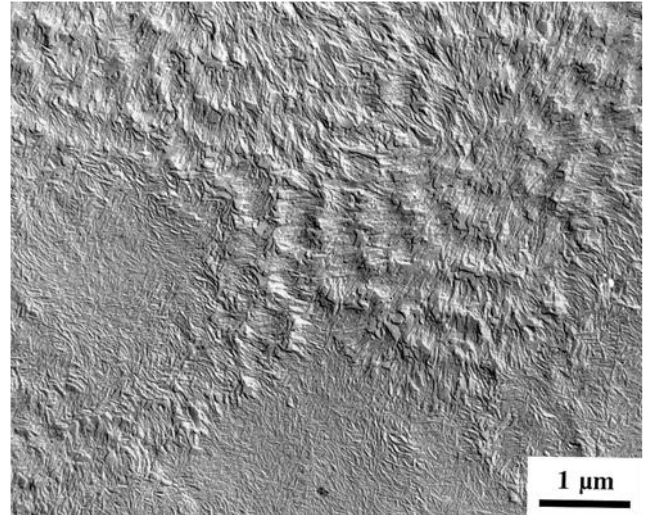
3 RESULTS

3.1 Isothermal Crystallization of Polyethylene Blends

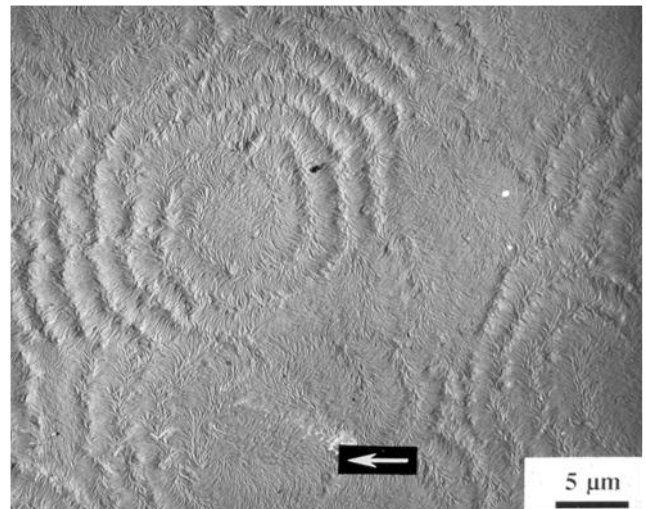
The isothermal crystallization of blends of LDPE and HDPE provides a means of designing materials with very different morphologies. Although the precise details depend on the

molecular characteristics of the two polymers, the composition of the blend and the crystallization temperature, early work by Hosier et al [3-5] showed that they can nevertheless be broadly divided into three categories.

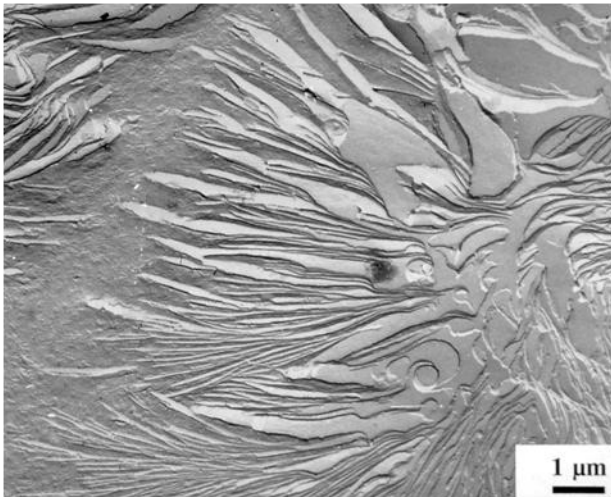
These are illustrated in Figure 1 for a system containing 20% of HDPE.



(a)



(b)

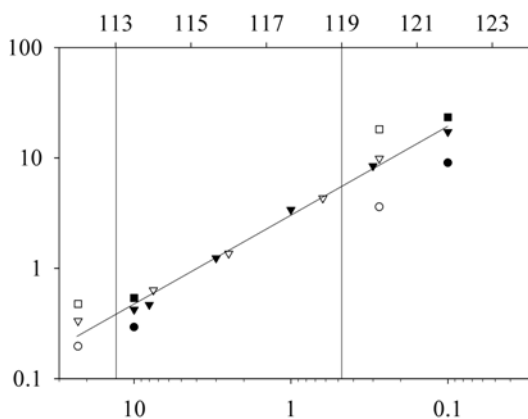


(c)

Figure 1: Transmission electron micrographs showing samples of a polyethylene blend containing 20% HDPE crystallized (a) by rapid quenching, (b) isothermally at 115 °C and (c) isothermally at 124 °C.

When crystallization occurs very rapidly, the two components of the blend crystallize intimately to give a morphology based upon small crystals; in this case, it is not possible from micrographs like Figure 1a to differentiate the two components of the blend. At high isothermal crystallization temperatures (above about 120 °C), crystallization occurs slowly to give distinct inclusions of HDPE crystals within a matrix that is, predominantly, composed of LDPE. This morphology can be seen in Figure 1c, which shows two isothermally crystallized (124 °C) aggregates of HDPE lamellae separated by quenched LDPE.

Isothermal crystallisation temperature / degC (hollow symbols)



Nonisothermal cooling rate / degC per minute (filled symbols)

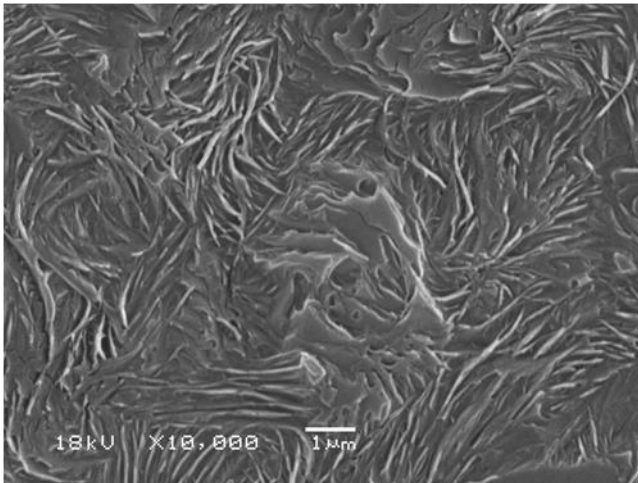
Figure 2: Crystallization time comparison for isothermal and non-isothermal crystallization for an 20:80 wt% HDPE:LDPE blend. Circles, triangles and squares: 10, 50, 90 % conversion.

At intermediate temperatures (~113-119 °C) a space filling morphology forms that is composed of thick, isolated lamellae of HDPE separated by inclusions of LDPE that are tens of nanometres in size. In this respect, this structure can be considered as a nanostructured bi-polymeric composite. A sample crystallized at 115°C is shown in Figure 1b, in which all of the visible features correspond to the minority HDPE phase; the majority LDPE phase is so well distributed that it is hardly visible at this magnification. The value of this morphology is that it enhances the breakdown strength of the system by ~15% compared with either the quenched blend or the LDPE material alone [3].

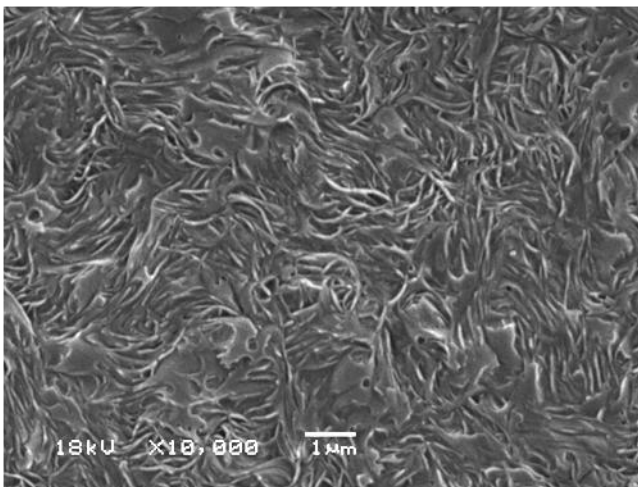
3.2 The Non-isothermal Crystallization Rate Window

While prolonged isothermal crystallization at a defined temperature constitutes a well defined and reproducible means of preparing specimens in the laboratory, it is not appropriate to a manufacturing environment. Consequently, if the concept of morphological design outlined above is to find technological application, then some less onerous manufacturing method needs to be devised.

In the knowledge that, for the blend, there exists a temperature window of ~6 °C within which isothermal crystallization leads to enhanced electrical properties, it was decided to consider the viability of non-isothermal analogues. An initial estimate was obtained by comparing crystallization times for isothermal and non-isothermal crystallization. Samples of the blend, ~5 mg in mass, were analyzed by DSC. Each sample was held at 200 °C for 2 min to erase its thermal history, before being crystallized to completion at a chosen isothermal temperature or subjected to the required non-isothermal temperature profile. The resulting crystallization exotherms were integrated with respect to time and compared in terms of their 10th, 50th and 90th percentiles; these data are summarized in Figure 2. From this analysis, a non-isothermal cooling rate range window of 0.5 – 10°C min⁻¹ was estimated to correspond to the isothermal temperature window of 113 – 119 °C, based upon thermal factors alone. Indeed, Figure 3 shows SEM images of systems crystallized using cooling rates of 0.1 and 10 °C min⁻¹, from which it is evident that a space-filling lamellar texture develops when the specimen is crystallized throughout this cooling rate range. As such, both the enhanced short term breakdown behaviour and the improved treeing resistance that have previously been associated with the existence of a space-filling lamellar texture should be accessible when crystallization occurs anywhere within this two orders of magnitude range, assuming that this morphological characteristic is indeed the source of enhanced performance.



(a)



(b)

Figure 3: SEM micrograph showing the morphology of the polyethylene blend prepared using a cooling rate of (a) of 0.1 °C min^{-1} and (b) 10 °C min^{-1} .

3.3 Breakdown Behaviour of Non-isothermally Crystallized Specimens

Initially, the effect of cooling rate on breakdown was explored at room temperature. The Weibull plots of Figure 4 compare data from quenched samples to results obtained from samples crystallized at 0.5 °C min^{-1} and 10 °C min^{-1} , while Figure 5 shows derived contours at the 90 % certainty level through the parametric likelihood surfaces, which is a convenient means of testing for differences in parameter values following the assumption of Weibull behaviour. Since, from this, the data obtained from samples prepared at 0.5 °C min^{-1} and 10 °C min^{-1} are indistinguishable from each other, yet distinct from the quenched data, we can infer that the true non-isothermal rate window is at least as wide as that represented by the data shown in Figures 4 and 5. This conclusion is consistent with the SEM micrographs shown in

Figure 3. In concert, all these data support the concept that it is this space-filling lamellar morphology that is the source of the enhanced electrical performance.

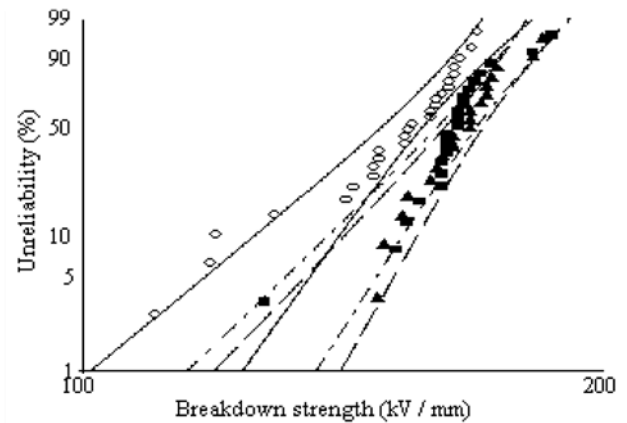


Figure 4: Weibull plots for the chosen blend following quenching (hollow circles,) and non-isothermal crystallization at 10 K min^{-1} (squares) and 0.5 °C min^{-1} (triangles). Two-sided 90 % confidence bounds in breakdown strength: dash-dot: 10 °C min^{-1} , long dash: 0.5 °C min^{-1} , unbroken line: quench.

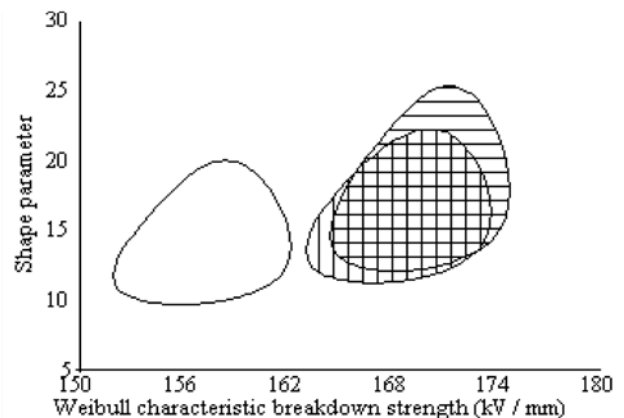
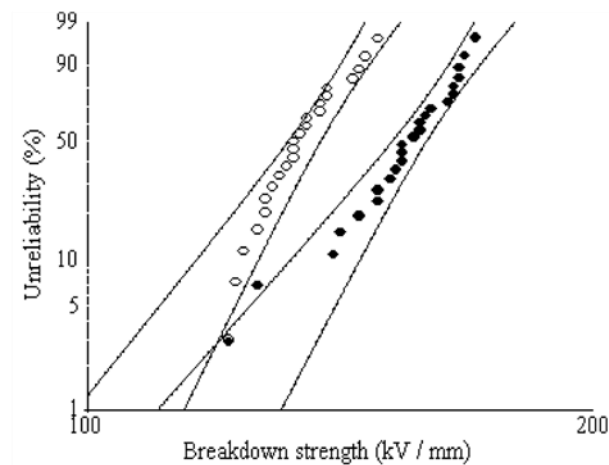


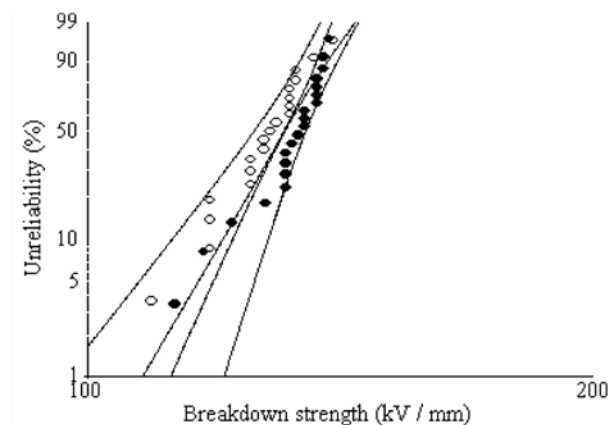
Figure 5: Contour plots (sections of the likelihood surface at a 90 % level) for the data in Figure 2. Open contour: quench. Vertical fill: 10 °C min^{-1} . Horizontal fill: 0.5 °C min^{-1} . Substantial overlap of the nonisothermal data means that they are statistically indistinguishable at this level.

Figure 6 compares the breakdown behaviour at elevated temperatures of the blend prepared under conditions of controlled crystallization (10 °C min^{-1}) with that of equivalent samples prepared by quenching. From this figure and the related data shown in Figure 4, it is evident that the Weibull shape parameter, β , increases with testing temperature while the scale parameter, α , decreases. Nevertheless, under all conditions considered here, the performance of the blend is improved compared with that of the quenched

analogue; from these data, controlled crystallization provides the greatest benefit at temperatures around 60 °C.



(a)



(b)

Figure 6: Weibull plots from testing the blend at (a) 64 °C and (b) 97 °C. In both cases, the left and right hand data sets correspond to: quenched and controlled-crystallized ($10\text{ }^{\circ}\text{C min}^{-1}$) samples respectively.

One possible explanation for the evident reduction in breakdown strength with increasing temperature is that it is an artefact associated with softening of the samples such that the 50 g upper electrode assembly deforms the specimen during testing, thereby increasing the effective field within the material.

To test this hypothesis, a series of masses up to 500 g were placed on the upper electrode; all were pre-heated to maintain thermal equilibrium. Figure 7 shows that up to 250 g, increasing the load on the specimen does not influence the measured breakdown strength, suggesting that the variation seen in Figure 6 is a genuine material effect, rather than being an artefact of the testing protocol.

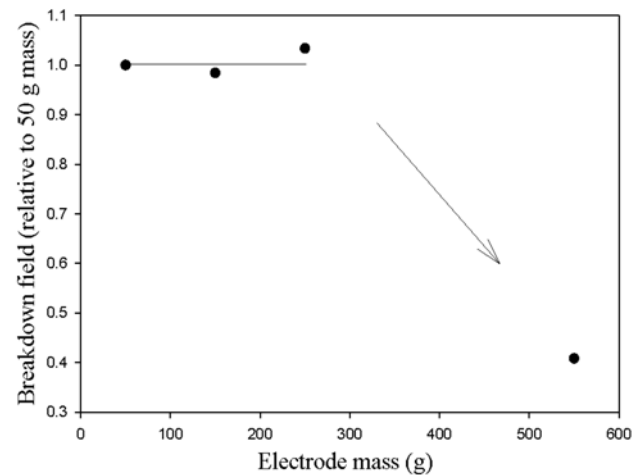


Figure 7: Effect of electrode mass on measured breakdown strength at 97 °C, each point corresponding to an average of four breakdown events

4 DISCUSSION

A major limitation of this study is that the breakdown data are highly sensitive to electrode geometry and testing protocol. For example, 60 Hz measurements on LDPE using moulded parallel-plane electrodes obtained a room temperature breakdown strength 230% higher than in this study, with a value at 100 °C only 50 % higher [13]. Nonetheless, the value of this particular breakdown technique is in the ability to generate large amounts of data relatively quickly. Although it may not provide an indication of how a real cable system may perform, it does serve as a useful comparative indicator for materials assessment.

The implications of these data for real cable systems are clear. At the temperatures considered, the critical engineering concern will be whether or not the geometrical integrity of the cable can be sustained. In contrast, the apparent problem of non-isothermal crystallization not necessarily imparting improved electrical breakdown properties to the system at high temperatures is insignificant if structural integrity cannot be maintained.

Thermo-mechanical data obtained from equivalent materials are informative as they show that controlled cooling raises the temperature for a given modulus by an equivalent of $\sim 20\text{ }^{\circ}\text{C}$ [14], such that the modulus of the controlled cooled sample at $\sim 90\text{ }^{\circ}\text{C}$ is comparable to that of the quenched material at $\sim 70\text{ }^{\circ}\text{C}$. This remains true even near the melting point of the BPE. It would therefore seem that the same space-filling lamellar texture that leads to improved breakdown strength at lower temperatures provides a mechanically stiff reinforcing structure at higher temperatures. Furthermore, it succeeds in achieving this without disastrously increasing the low temperature stiffness.

5 CONCLUSIONS

The effect of imparting a controlled thermal history on a blend comprising 20 wt% HDPE in an LDPE matrix is to improve the breakdown strength by up to 15 %. As the LDPE softens, the benefit of controlled crystallization is less obvious: the advantage is rather to impart extra mechanical stiffness to the blend via the reinforcing framework of space-filling HDPE lamellae. It is likely that this same framework will include an increased level of inter-lamella tie molecules which will also structurally reinforce the material.

Such a blend could therefore be considered for use in HV cable applications, it would need to undergo extensive mechanical creep testing and model cable trials. The effects on mechanical and electrical properties of longer term annealing, possible ageing at higher temperatures and of recrystallization following high temperature short circuit transients would require more research. Although this blend would be completely molten at the XLPE overload temperature of 130 °C, initial trials by the authors indicate that the material may be acceptable at temperatures as high as 120 °C. Performance optimization of the actual blend studied and of related blend materials should also be considered.

6 ACKNOWLEDGMENTS

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