

Novel Insulation Materials for HV Cable Systems

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The article reviews the limitations of current XLPE based high voltage cable insulation and outlines the benefits of transition towards sustainable thermoplastic insulation materials.

Introduction

Cross-linked polyethylene (XLPE) has replaced oil-paper insulated systems as the primary solution for medium and high voltage ac cables decades ago, since they enable marginally higher operating temperatures and can be produced with high throughput and well-controlled extrusion technology [1]. The base for this insulation is polyethylene (PE), which can be crosslinked either with peroxide cure (involving thermal decomposition), or by grafting silane onto the polymer chains, and the use of moisture based cure [2]. Crosslinking is deemed necessary, since commercially available, branched low density polyethylene (LDPE) has more significant melting at temperatures around 100 °C and the material loses all of its mechanical stability. In contrast, linear high density PE (HDPE) has a higher melting point and can achieve higher operating temperatures. But while HDPE found some success in medium voltage cables, it has not managed to establish a foothold in the high voltage cable sector [3]. After decades of research, conventional XLPE is at the limit of its capabilities, as outlined below, and further development is bound to have diminishing returns. Further, the costs of large extrusion and catenary crosslinking manufacturing facilities and the costs and time of degassing larger cross-section HV and EHV cables present significant sustainability issues for cable manufacturers.

Limitations of XLPE

While the base PE (either linear high density or branched low density PE) is a thermoplastic, the resulting XLPE compound is a rubber (or thermoset when thermal crosslinking takes place) [4]. The gel content is a measure of the extent of the three dimensional, cross-linked polymer network. This content depends on the type of crosslinking agent used and the crosslinking temperature, and the amount can be determined by measurement of the fraction of material not extracted by a solvent [5]. The transition to a rubber has its advantages, such as improved thermal stability compared to LDPE as the melting point of the crystal phase is approached. While XLPE expands significantly when operating at temperatures close to 100 °C, it will retain adequate structural integrity if the gel content is high enough (typically above 60%) [6].

This thermal expansion is a major issue when operating a HV cable above 90 °C. Over the full low to high temperature operation of a cable thermal expansion differences of up to 20 times may occur, due to differences in the thermal expansion coefficient of the metals in the conductor and screens and shields (copper, aluminium, lead) encasing the main insulation over the full operational temperature range. This does not mean the molecular conformation changes much, but the molecules move further apart from another [4]. As an example, typical XLPE used in cables expands by about 15 % when going from room temperature to 105 °C, while copper or aluminium expands by less than 3 % in the same temperature range [7, 8]. The resulting pressure may affect the dielectric properties [9] and can cause damage to sheath and joints [10]. Heat-resistant XLPE has been developed by changing the base PE and developments of different types of

peroxides [2, 11]. While peroxides as used in some cases [11] increase the operating temperature by up to 20 °C, such measures can result in a slower curing process, if slower curing peroxides are used, this may result in increased scorching [2] which produce oxidised gels which create a dielectric defects.

At the same time XLPE undergoes such thermal expansion, the thermal conductivity of the polymer is also reduced [7]. Ethylene propylene rubber (EPR) has been suggested as an alternative insulation material in the past, since it retains higher thermal conductivity while expanding less than XLPE above 90 °C. However, EPR has significantly higher permittivity and dielectric losses over the entire range of potential operating temperatures, and can resist less strain and elongation before breaking than XLPE [12].

As mentioned above, the main reason behind the move to crosslinking PE is the improved thermomechanical stability. However, the modulus drops by orders of magnitude in the range between 90 to 100 °C [13]. So, if the insulation system is subjected to mechanical forces in this temperature range, irreversible mechanical damage can result, especially near cable joints [10, 14]. XLPE might be able to recover some of its stiffness after cooling from temperature excursions above 90 °C as determined by the gel content, but despite its crosslinked nature, there are strict limits regarding the temperature range over which an XLPE system can operate. Indeed, short excursions to 120 °C might be tolerated in some cable designs but this is really the limit of XLPE technology. Knowing this limit enables cable and network operators to set operational limits to secure the long term integrity of their cable systems.

Thermoplastics

The process of crosslinking PE and the presence of the molecular network within the material can have a significant effect on the performance of XLPE insulation. First, peroxide crosslinking – the most commonly used process – inevitably results in the production of small molecular species as reaction by-products which, if retained within the material, can adversely affect electrical properties.

Dicumyl Peroxide (DCP) is the most commonly used crosslinking agent. It was established as early as 1953 by General Electric, followed by silane crosslinking introduced by Dow Corning in the 1960ies, resulting in silane crosslinked PE (SXLPE) [15]. SXLPE has a few advantages over DCP cured materials, such as a simpler curing process, but with the downside of potential water retention in the insulation system. Most cable manufacturers prioritize DCP, since reactions using silane crosslinking agents occur at 90 °C in a water bath, which can lead to increased risk of water tree formation in the final product [16]. In either case, there will be both labile small molecule and gaseous by-products of peroxide cure remaining in the cable system, with compounds such as acetophenone, cumyl alcohol, methylstyrene and methane being the most significant in HV cables [17]. These by-products are not only responsible for the distinctive smell of a new XLPE cable, they also alter the electrical properties, like reducing the impulse breakdown strength [18]. It has been shown that the space charge behavior also changes depending on the type of cure, where DCP by-products tend to increase accumulation of heterocharge, while silane favors homocharge [16]. In terms of mechanical properties, the tensile strength, elongation and threshold fracture energy all decrease with increasing DCP content [19].

Crosslinking of polyethylene by DCP occurs, typically, at temperatures in the range of 150-200 °C and, results in the formation of a molecular network within the polyethylene melt. This is significant in that subsequent crystallization of the polymer occurs within the crosslinked network, which serves both to increase the nucleation density and suppress the formation of extensive morphological features. Figure 1 shows a transmission electron micrograph of a sample of XLPE cable in comparison with a typical LDPE microstructure. While the XLPE reveals the presence of lamellar crystals, these are thin (low melting) and limited to approximately 100 nm in their lateral extent. This is very different from the structure that develops in the absence of crosslinking, where the PE molecules are arranged into lamellae which, themselves, form spatially organised polycrystalline spherulites, which, in extreme cases, can approach 1 mm in diameter.

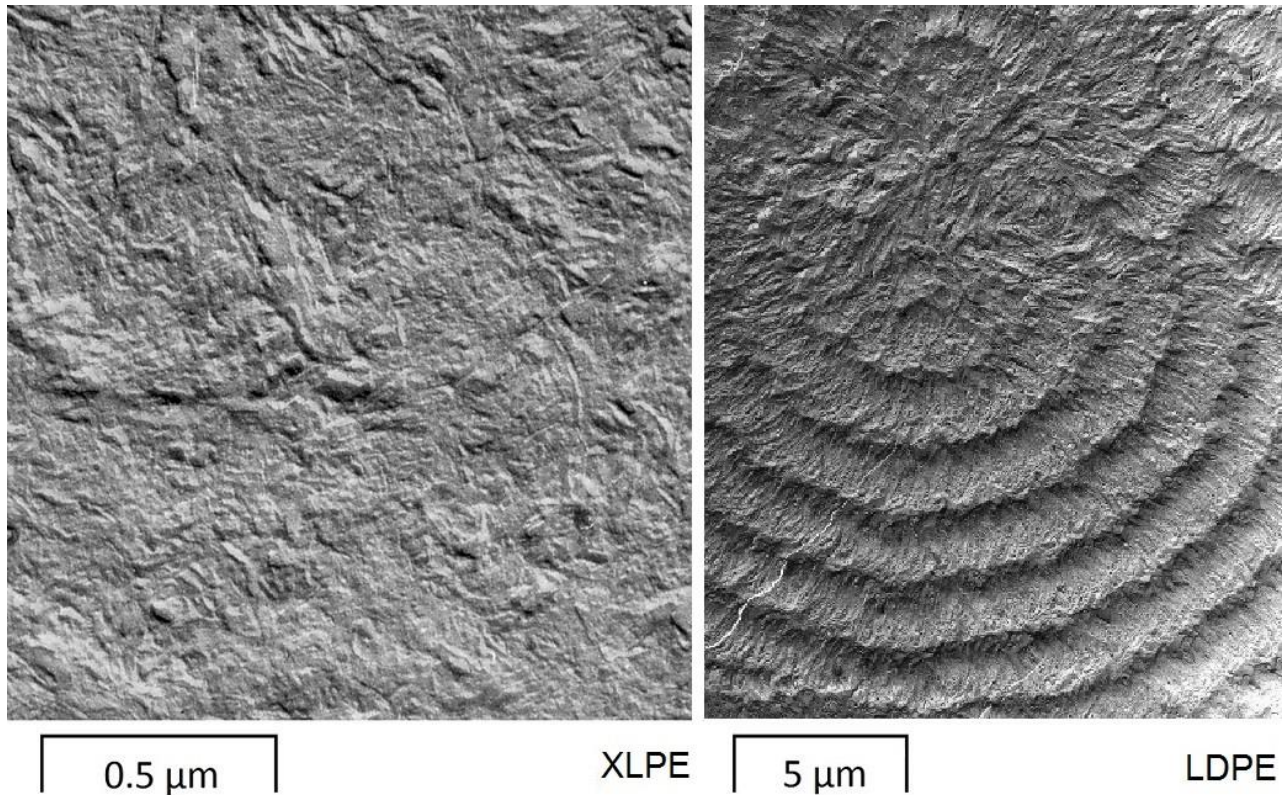


Figure 1. Transmission electron micrograph of typical XLPE structure in contrast to LDPE (in this case metallocene catalyzed LDPE).

The effect of this structural complexity on the electrical properties of semicrystalline polymers is a topic that has received considerable attention over many decades. An early study of particular relevance to the topic of novel thermoplastic insulation systems was reported by Kolesov in 1980 [20]. This work considered the effect of spherulite size on the breakdown strength of thin films of PE and polypropylene and revealed a form of behavior that is represented schematically in Figure 2. From this, it is evident that breakdown strength initially decreases with increasing spherulite size up to the point where the spherulite diameter equates to the film thickness and, thereafter, remains at a constant value. The explanation for this is that breakdown occurs via interspherulitic regions, which constitute electrically weak pathways through the structure and therefore, simplistically, the morphology shown in Figure 1 may be considered optimal, since it equates to one in which the spherulite size can be considered to be vanishingly small. However, this fails correctly to recognize the reason why interspherulitic regions are weak.

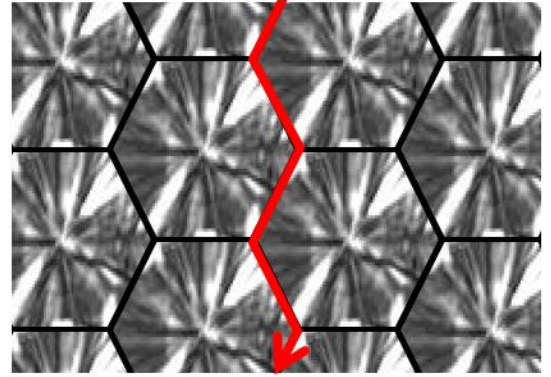
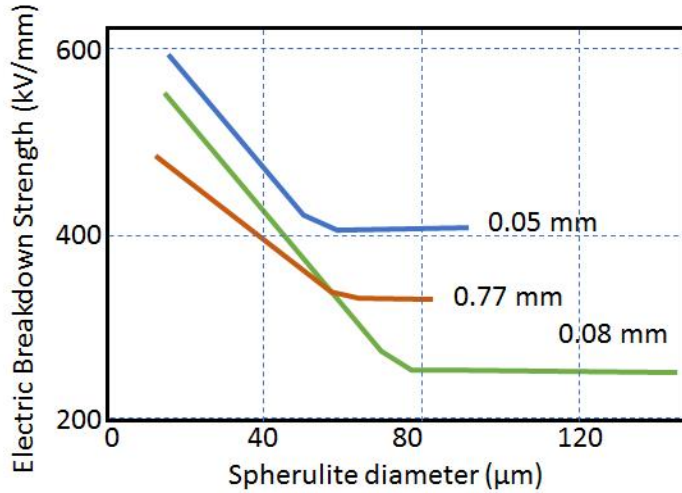


Figure 2. Electric breakdown strength of PP and HDPE samples of different thickness as function of spherulite mean diameter size (left, original data from [20]); right hand side: illustration of the breakdown path along the weak boundaries.

The process of polymer synthesis is inherently statistical in nature, such that all factors that relate to the architecture of a polymer molecule are distributed. For example, consider the branched molecular structure of LDPE, where pertinent factors include: the molecular length; the number of branches; the separation between branches; the length of the branches. These factors all impact upon the morphology that is formed and, hence, the properties of the material. For example, the side branches cannot be included within the crystalline structure and, consequently, the separation between neighboring branches determines the maximum crystal thickness that can be formed and, hence, the melting point of the lamellar crystals. In this way, the branches tend to inhibit crystallization, such that the overall crystallinity is relatively low, as is the consequent density. HDPE, on the other hand, contains relatively few branches such that crystallization occurs more readily to give a material that is high melting and has a higher crystallinity and higher density. A convenient way to consider such a polymer system is therefore to think of molecular features such as branches, which hinder crystallization, as molecular defects, such that a sample of any crystallizable polymer contains a range of different molecules, some of which form crystals more readily than others. Detailed studies of crystallization in the late 1970s [21-23] demonstrated that the first molecules to crystallize are those that are both long and defect free, which form so-called dominant lamellae. Between these crystals, longer, but more defective molecules then crystallize into subsidiary lamellae. The shortest and most defective fraction diffuses away from the spherulite, eventually, to become concentrated between neighboring spherulites. Whilst the complete range of different molecular forms present in the material are intimately mixed in the melt, once crystallization begins, they undergo a self-assembly process that concentrates different molecules at different sites within the microstructure. The weakness of the interspherulitic regions reported by Kolesov is a consequence of this segregation process.

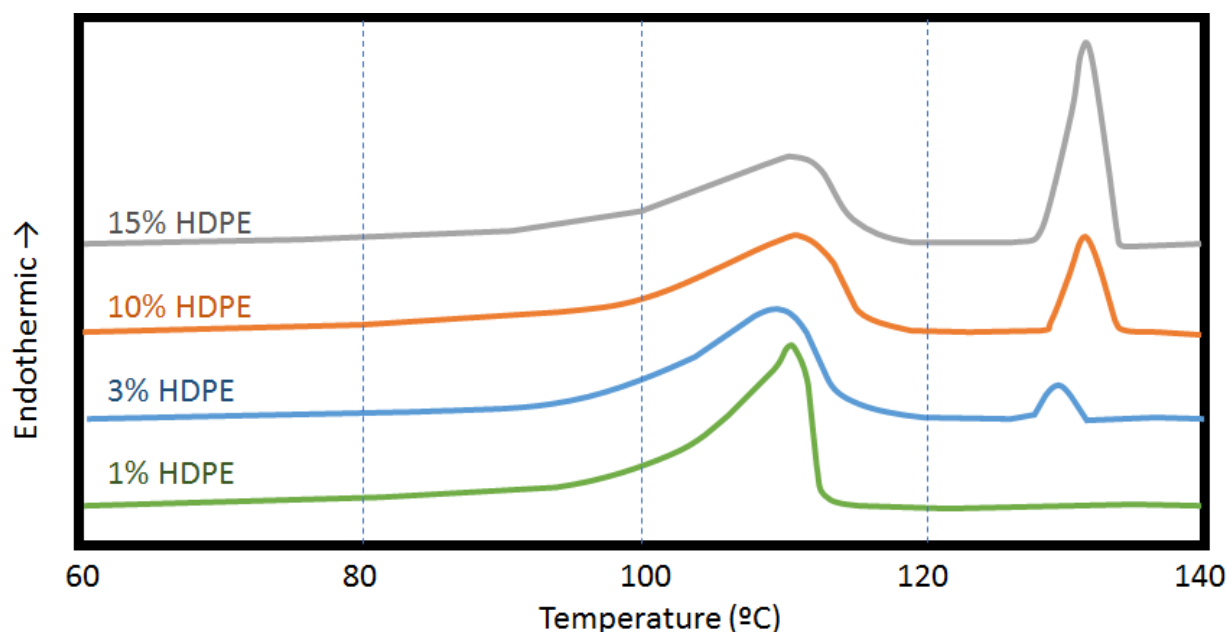


Figure 3. DSC melting behavior of samples isothermally crystallized at 124°C for different HDPE/LDPE blend compositions (original data from [24]).

The account given above would appear to indicate that degraded electrical properties are a necessary consequence of spherulite formation. This is not the case, as was demonstrated by Hosier et al. [24, 25] in a series of papers. This work took a fundamentally different approach that involved blending together two different polymers (LDPE and HDPE) and crystallizing these in a specific way that exploited the difference in their propensity to crystallize. Figure 3 shows a differential scanning calorimetry trace of the melting behavior of such blends, which clearly shows that melting occurs in two stages.

First, there is a significant melting peaking at about 110 °C which, primarily, corresponds to melting of the LDPE component of the blend. Second, there is a peak near 130 °C, which relates to melting of the HDPE. Hence a temperature window exists, where the HDPE is able to crystallize but where the presence of the branches in the LDPE prevents it from doing so. The significance of this work lies in the finding that, if crystallization occurs initially in this window, then a framework of thick HDPE crystals forms within molten LDPE. The fact that the LDPE remains molten is significant, since it prevents the concentration of defective molecules at particular sites within the morphology, thereby overcoming the process that gave rise to reduced breakdown strength in the work of Kolesov. Figure 4 shows such a morphology, which reveals the presence of a “skeleton” of individual HDPE crystals separated by nanoscale inclusion of LDPE. As such, this structure could quite reasonably be described as a nanocomposite, where the properties are a consequence of both the constituent elements and their spatial distribution. Electrically, such continuous space filling morphologies have been shown to give rise to an increase in breakdown strength of 24% compared with a material of the same molecular composition but with a fine scale morphology more akin to that of XLPE. The improvement in electrical treeing resistance is much more dramatic, with tree growth rates being significantly reduced in PE blends compared to XLPE [26]. Also, mechanically, the nanoscale inclusion of the LDPE provide low temperature flexibility while the continuous framework of thick, high melting HDPE crystals gives good high temperature mechanical integrity. This integrity is retained to some 30 °C above the melting temperature of XLPE. Albeit, that higher temperature does result in some melting and corresponding volume expansion of the LDPE and, in the absence of the XLPE gel, some flow – as we will see this can be further improved by consideration of propylene based systems.

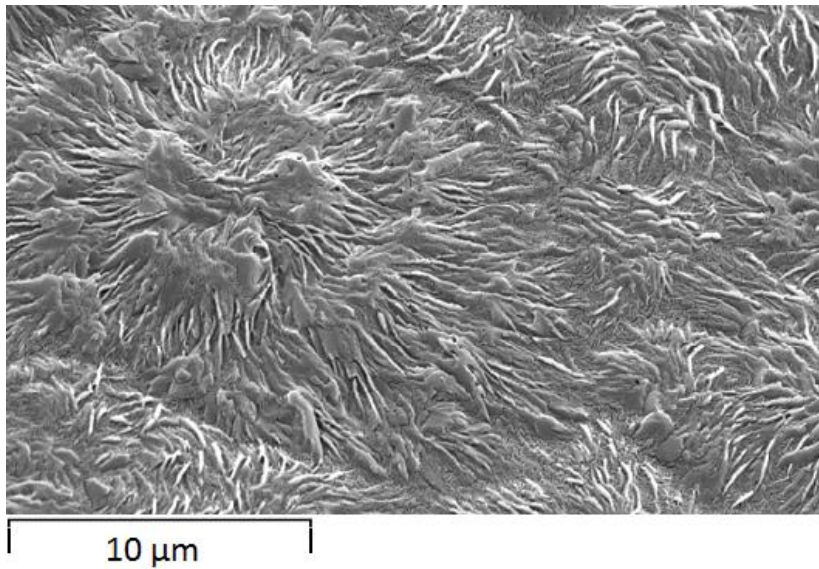


Figure 4. SEM graph showing the crystalline HDPE “skeleton” structure, which is embedded in the non-crystalline majority component LDPE.

Propylene based systems

The previous section has sought to illustrate how self-assembly of polymeric systems, in which the molecular composition has been specifically designed, can be used to manipulate the morphology that develops, leading to control of macroscopic physical properties. In the case of polyethylene, this is relatively simple to achieve, both in the laboratory [27] and in more technological scenarios [28]. In other systems, the necessary control is much more difficult to achieve.

Polyethylene and polypropylene (PP) are bulk commodity polymers that are comparably priced and which both have an intrinsically non-polar molecular structure. However, several significant differences exist which include:

- The melting temperature of isotactic polypropylene homopolymer (iPP) is much higher than that of HDPE.
- The morphology of iPP is based on a number of different crystal structures (notably α , β , and γ).
- iPP tends to form particularly large spherulites, which has often been interpreted as being a consequence of few secondary nucleation sites. The benefit is that the material is intrinsically clean.
- Large spherulites however, mean that inter-spherulitic regions are particularly weak in this polymer.
- The synthesis method of PP does not produce branched molecular structures akin to LDPE. However, copolymerization of propylene with another olefin is widely used to produce copolymers with various molecular structures, which could accommodate limited branching, and different properties.
- Miscibility of iPP and copolymers can be limited, such that a range of phase separated structures can form.

In view of the above factors, manipulating the structure and properties of PP is much more difficult than in PE. Even though, superficially, blending can be used in a comparable way with readily crystallized iPP used in place of HDPE, and with a less crystallizable copolymer replacing LDPE.

Interest in the use of propylene-based systems in cable applications is driven by two factors: first, the relatively high melting temperature of PP (above 160 °C) provides the potential for much higher cable current ratings, if the increase in conductor temperature and Joule losses can be tolerated. The apparent cleanliness

of PP means that, if correctly designed, such systems should exhibit excellent electrical properties – c.f. the use of PP in capacitor and supercapacitor applications. Broadly speaking, a widely-accepted strategy for the design of propylene-based systems for use in cable applications [29, 30] is to blend together iPP (relatively stiff and with a relatively high glass transition temperature) with a more flexible, lower crystallinity copolymer. Whilst this approach can adequately address the need for material systems that are mechanically comparable to XLPE – but with a much higher melting temperature – the electrical consequences of this can be problematic. So, it is clear that trade-offs are required in optimizing the molecular and structural architecture to optimize the range of physical properties required for robust cable operation.

Figure 5 shows the two structural factors that need to be controlled to generate propylene-based blends with good electrical properties. Figure 5a shows a sample of iPP after isothermal crystallization at 120 °C, which clearly reveals the presence of distinct interspherulitic boundaries, which are electrically weak. Figure 5b shows the morphology of a system containing 12% of ethylene which undergoes extreme phase separation, even when the sample is quenched. The resultant two phase structure contains electrically weak regions, even in the absence of any iPP. Consequently, the objective of blending is to eradicate weakened interspherulitic regions through the addition of an appropriate low crystallinity copolymer, as in the case of the HDPE/LDPE blend discussed above, while minimizing the dimensional scale of any phase separation effects.

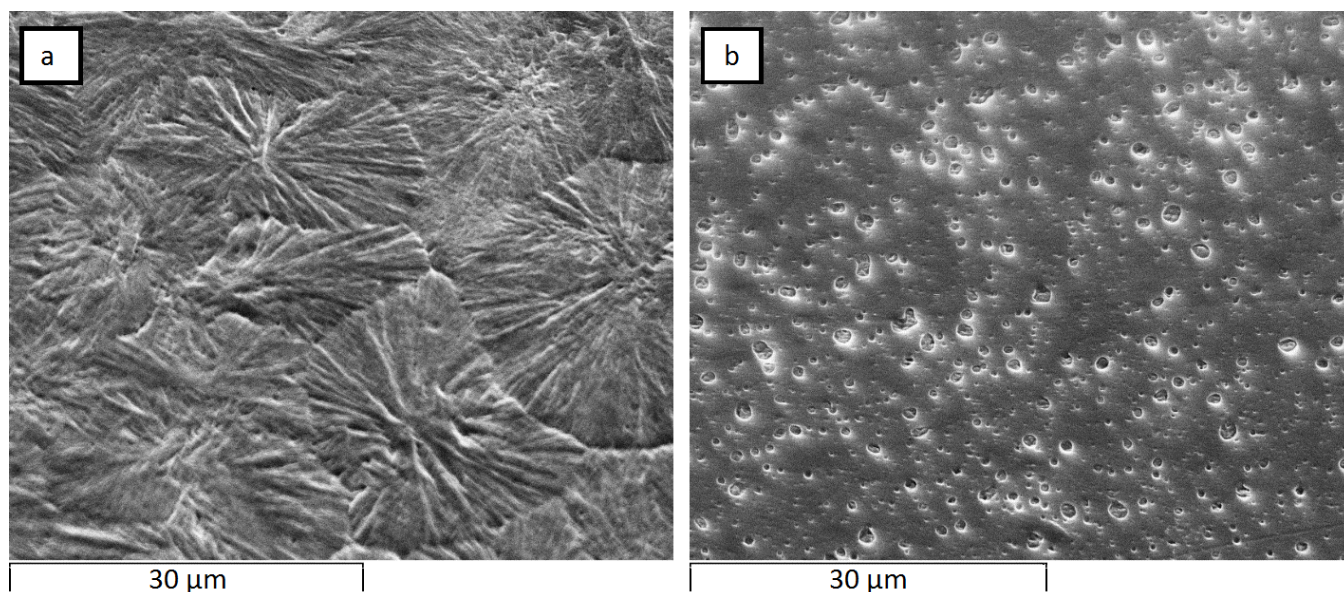


Figure 5. SEM graph showing (a) iPP after isothermal crystallization at 120 °C, showing clear interspherulitic boundaries; (b) PP blend with 12% ethylene, showing clear phase separation, resulting in electrically weak regions.

Figure 6a shows a blend system in which weak interspherulitic regions have been successfully eliminated through the incorporation of an ethylene/propylene copolymer into iPP (equal masses of each). However, the resulting system is still characterized by a degree of phase separation.

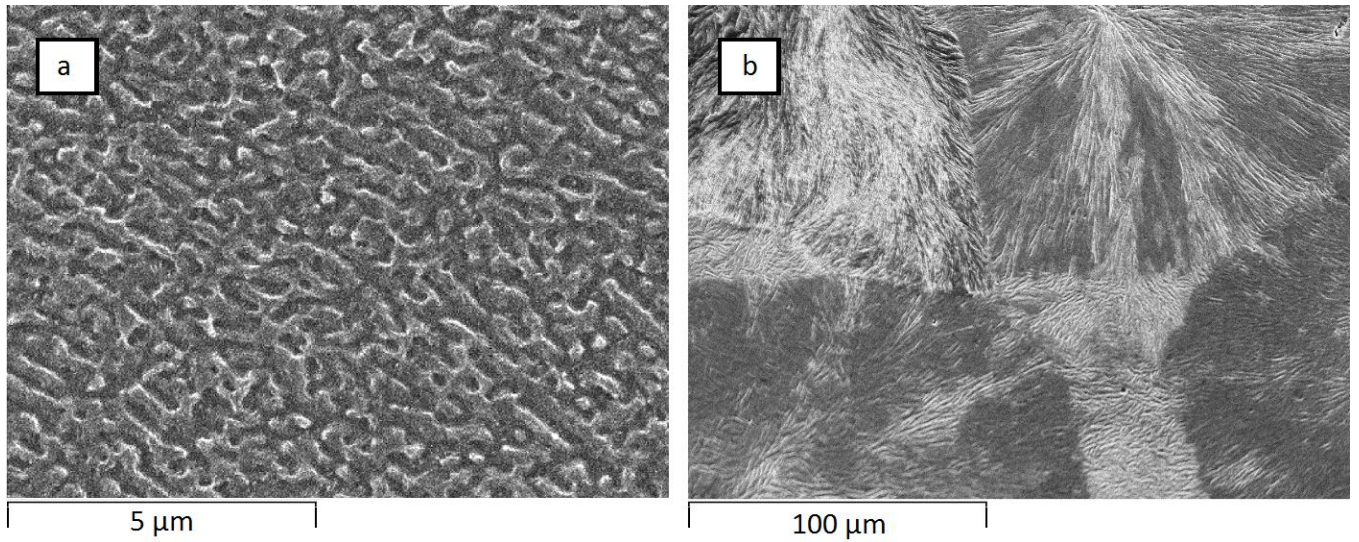


Figure 6. SEM graph showing (a) iPP blend system with ethylene/propylene copolymer, virtual elimination of weak boundaries; (b) example of successful PP blend without phase separation.

In contrast, the blend system shown in Figure 6b contains no evidence of weak interspherulitic regions or phase separation and is characterized by excellent breakdown performance that is largely invariant to processing variations.

Nevertheless, even the material system shown in Figure 6a provides excellent electrical performance when extruded onto a mini-cable, as shown in Table 1, which compares the breakdown performance of two sets of propylene-based mini-cables with that of a reference XLPE-insulated mini-cable (insulation thickness ~ 4 mm). In this case, 6 m lengths were subjected to a stepped DC test and the failure voltage was noted. In the case of the XLPE-insulated cables, all samples broke down around 200 kV.

None of the equivalent propylene-based systems failed. Indeed, while subjecting the propylene-insulated mini-cables to an extreme bend deformation prior to testing did reduce their performance somewhat, even the lowest breakdown voltage observed was still considerably above that of the best performing XLPE.

Table 1. Breakdown strength of PP-based mini-cables, compared to XLPE reference mini-cables of same dimensions [31].

	No. 1: V_b (kV)	No. 2: V_b (kV)	No. 3: V_b (kV)	No. 4: V_b (kV)	No. 5: V_b (kV)
<i>Straight propylene-based</i>	>400	>400	>400	>400	>400
<i>Bent propylene-based</i>	>400	396	388	270	>400
<i>Straight XLPE reference</i>	184	168	224	196	196

Conclusions

We commented at the start that after decades of research, including more recent development on low DCP containing XLPE formulations, conventional XLPE is at the limit of its capabilities. These restrictions are a result of the intrinsic limits of purity that can be obtained with the most commonly adopted crosslinking chemistries, and the thermal performance of the materials in regard to their relatively low melting points. These constraints due to the base polymer have an impact on the performance of both HVAC and HVDC cable designs, whose performance may be compromised by the impurity content, and whose manufacture is challenging and contains risks that are difficult to mitigate. Hence, further development is bound to have diminishing returns and may not be economically sustainable in the long term, as new cable manufacturing investment becomes necessary in order to maintain production in the long term. Nonetheless, XLPE technologies are widely adopted and are expected to remain a key cable insulation system into the future.

In contrast, it is now possible to formulate thermoplastic polymer blends that are able to overcome the limitations of XLPE. These new materials promise more sustainable systems in the long term, both economically and environmentally, while providing the inherent ability to provide greater design freedom for HV and EHV AC and DC cable developments, by making use of their intrinsic purity, higher thermal stability, simpler and more economic processing without degassing, and significantly lower investment costs for next generation cable manufacture.

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