Mechanical recycling of XLPE from cable production waste

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SUMMARY

The growing environmental awareness has raised up some new issues in the cable industry: the recycling of cables should now not only consider its metallic part but also its polymeric one. Cross-linked polyethylene (XLPE) is the most widely used polyolefin in this part, but also one of the most difficult to recycle, because of its thermoset nature. This master thesis aimed at defining a mechanical method to recycle the silane-cross-linked polyethylene from the lumps of production.

The solution studied was to blend the lumps of XLPE, previously ground into a 0.6mm diameter powder, with pellets of linear low density polyethylene (LLDPE). The technical feasibility of blending the two materials with either injection-moulding or extrusion was validated at laboratory scale for contents in XLPE ranging up to 70wt-%. In the case of extrusion of tapes, a pre-compounding was necessary only if a two-feeder extruder was not available. The blends obtained showed interesting mechanical properties (stiffness, toughness, hardness), mainly dependent on the content of XLPE and on the nature of LLDPE. The recycled material has then been considered for two applications by performing trials at production scale: insulator of wires and inner-jacket of a power cable. In both cases, the surface roughness limited the maximum content of XLPE accepted, but the tube-tooling extrusion worked out successfully and the mechanical properties obtained were very encouraging: the mechanical requirements were fulfilled, before and after ageing.

Keywords: cross-linked polyethylene, XLPE, PEX, recycling, mechanical recycling, polymer blend, linear low density polyethylene, LLDPE, cables, polyethylene, silane-XLPE, primary recycling
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1. Introduction

1.1. The objectives

To date, the recycling of cables has been driven by economic considerations based on the high volume of use and the high value of the metal part, letting the remaining polymeric part generally regarded as waste.

However, as economic conditions and regulation change (WEEE Directive), inspired by a growing environmental awareness, the recycling of the polymeric part is being more and more considered. New investigations have then started to determine ways of recovering these cables parts, based on the valuable energy/material resource potential of polymers.

Consequently, a Research and Development programme called “Wire and Cable” has been launched in the Research Institute Swerea IVF, focused on the recycling of the different polymers in cable waste. Eight companies have expressed their interest and support in the project: Borealis AB, Ineos ChlorVinyl AB, VCC AB, Volvo 3P AB, Nexans IKO AB, Draka Kabel AB, ABB Power Systems AB and Stena Recycling AB.

As cross-linked polyethylene (XLPE) is a widely used polyolefin in energy cables and its recycling has always been challenging due to its thermoset nature (not remelttable), a project centred on the recycling of XLPE has been started.

The objectives within the XLPE recycling project is to develop a method to separate and recycle XLPE from cable waste and investigate suitable applications for the recycled compounds. As a Master Thesis Student, I have then been asked to investigate on possibilities of achieving a mechanical recycling of XLPE stemming from cable production waste and propose some possible applications for the recycled material.

The objectives of this Master Thesis are the following:

- Evaluate the technical feasibility of blending XLPE with Linear Low Density Polyethylene (LLDPE) by injection moulding. From the trials, determine the mechanical properties of the blend and the main parameters influencing the properties.
- Evaluate the technical feasibility of blending XLPE with LLDPE by extrusion. From the trials, determine the mechanical properties of the blend and the main parameters influencing the properties.
- Investigate the possible applications of the recycled material. If possible, perform some trials at production scale in specialised companies for these considered applications.

1.2. Cable materials

1.2.1. The structure of cables

A cable consists of two or more wires running side by side, which can be bonded, twisted or braided together to form a single assembly \[^1\].
Usually, two main types of cables are distinguished:
- Cables which aim at transmitting information (signals) or energy
- Cables which supply electricity. [2]

There are hundreds and thousands different designs of construction of cables, depending on the application and on the country of use. The national design is standardized by national organisations, such as the the Svenska Elektriska Kommissionen (SEK) for Sweden. But some international and european standardizations also exist, such as the International Electrotechnical Commission (IEC) and the European Committee for Electrotechnical Standardization (Cenelec).

Generally speaking, the electrical cables are always based on a conductor (except for optical cables), which can be single-wired or stranded, made of aluminium or copper. It is then wrapped by insulators, which protect the cable from environmental external damages, optimize its transmission properties and make the use of them safer. These insulations layers can be numerous (bedding, sheathing, jacket,...), and most of them are based on thermoplastics, such as cross-linked polyethylene (PEX or XLPE) or polyvinylchloride (PVC) with halogen free flame retardants (HFFR). For certain applications, some additional layers of metal are added, to improve the safety and electric properties of the cable (screen).

The power cables are a useful example for this project. These cables, which aim at transporting electricity at a voltage higher than 240V are generally composed of 7 layers [3]:

![Figure 1: Basic construction of a power cable (with permission from A.Boss)](image)

The jacket and shield aim at protecting the cable from the hazards and environment, by assuring respectively a physical and mechanical barrier.

### 1.2.2. Cross-linked Polyethylene

Cross-linked polyethylene, commonly abbreviated PEX or XLPE, is a low density polyethylene (LDPE) or high density polyethylene (HDPE) which has undergone some cross-linking, changing it into a thermoset. The first XLPE was prepared in the 1930s.
1.2.2.1. Synthesis \textsuperscript{[1,3,4,5,6]}

As three methods of cross-linking exist nowadays, the XLPE has been classified into three different types:

- PEX-A

PEX-A is cross-linked by using peroxides ("Engel method"), which initiate the formation of radicals on carbon atoms along the chains and the consecutive creation of carbon-carbon bonds by recombination. This can be performed thanks to a high temperature (above the melting temperature) and high pressure extrusion, with a very good control of the heating (rather tight operating window). The crosslinking obtained is usually very constant and uniform, adapted to LDPE and HDPE, but is expensive and takes long time.

![Figure 2: Schematic representation of the peroxide cross-linking](image)

- PEX-B

This silane cross-linking (PEX-B) is nowadays seen as the most common and economic process. The polymer is firstly chemically modified by a grafting of vinyltrimethoxysilane with a small amount of peroxides. The condensation of silanol between these units connects then the chains together through C-C-Si-O-Si-C-C bonds. The incorporation of the reactive silane groups can also be achieved by polymerisation of ethylene in presence of a vinylsilane as a comonomer. The cross-linking is usually achieved in a secondary post-extrusion process, resorting to a cross-linking agent and a catalyst and accelerated by heat and moisture.

![STEP t Grafting](image)
PEX-C is manufactured through an electron irradiation: when the highly energetic electrons hit the PE chains, they break some carbon-hydrogen or carbon-carbon bonds, what leads to the formation of radicals. These radicals usually react with another radical in its vicinity, creating locally bonds between the main chains. The beaming is usually performed on highly amorphous LDPE materials, as the irradiation is less effective on rigid crystalline regions (hard to penetrate). The process requires costly tools and machines and the crosslinking is less uniform than PEX-A, but it is the most environmentally friendly method (no chemicals or solvents).

Figure 3: Schematic representation of the silane cross-linking

Figure 4: Schematic representation of the radiation cross-linking

1.2.2.2. Properties and applications [1,3,4,5,6]

At a significant degree of cross-linking, the XLPE turns into a thermoset, consequently displaying, in comparison with a non cross-linked material, a better dimensional stability and better mechanical properties. Indeed, it is usually stronger and tougher, with an improved Environmental Stress Crack resistance. Its chemical, electric and thermal resistances are also higher, and it withstands more easily corrosion and abrasion. But the most interesting improvement is its increased high temperature resistance, which allows higher service temperatures.

All these properties make PEX particularly used in potable-water pumbling, hydronic radiant heating systems and natural gas and offshore oil pipes. But it is also commonly found in chemical transportation means, and in insulation of high voltage electrical cables (energy cables).

1.2.3. Linear Low Density Polyethylene

The linear low-density polyethylene (LLDPE) is another member of the family of polyethylene, based on the simplest monomer: ethylene.
Particularly, the LLDPE is a linear polyethylene, with a significant number of short branches.

1.2.3.1. Synthesis $^{[1,7,8,9]}$

The synthesis of this thermoplastic is closely related to the methods used for the other polyethylenes, namely HDPE and LDPE. Indeed, it is commonly synthesized by copolymerisation of ethylene with short-chain alpha-olefins (mainly butene, hexene or octene) and in the presence of a catalyst (transition metal catalysts: Ziegler-Natta or Philips type). This is achieved at lower temperature and pressure than LDPE thanks to either a gas phase reaction (ethylene with butene or hexene) or solution reaction (ethylene with octene).

This conveys a rather linear structure, with small chain branching, differing it distinctly from the long-chain branched LDPE.

Another major difference when compared with LDPE is its narrower molecular weight distribution. All these structure differences lead to distinctive properties.

1.2.3.2. Properties and applications $^{[1,7,8,9]}$

The LLDPE shows some interesting advantages over LDPE: it has a higher tensile strength and a higher impact and puncture resistance. In addition, it is highly flexible and can be more elongated. LLDPE shows as well a better Environmental Stress Cracking Resistance, and a puzzling ability to retain structural integrity at lower thickness, what allows the manufacturing of thinner films.

However, its processing is not as easy as that of LDPE: as is has a narrower molecular weight distribution and shorter chain-branching, the LLDPE is less shear sensitive. Consequently, the material is harder to process in extrusion, and shows a faster stress relaxation. This also lead to a lower viscosity in the melt state, with a rather few chain entanglements, allowing an easy elongation as the chains slide passed each other.
Besides, the material is inert and exhibits some good chemical resistance properties, such as a good resistance to UV radiation. Although it is not as glossy as LDPE, it is relatively transparent, and displays some interesting electrical insulating properties.

The LLDPE has nowadays penetrated the whole market of LDPE, turning into the polyethylene of higher growth rate of consumption. It is commonly used in plastic bags, toys, pipes, containers, cabling and tubing. But the major area of application consists in packaging, for which the great flexibility and structural integrity of the material is determinant.

1.3. Recycling and cables

1.3.1. Recycling: why and what?

In this globalisation era, the need to convey information and energy all around the world is more than ever tremendous, requiring a huge cable production and... waste.

In the past, after use, only the metallic parts of cables were considered for recycling (as they offered the highest value), letting the polymeric part as a waste sent to the landfill sites. But, as we are facing growing environmental concerns, changes in legislation and energy savings, the question of recycling of these cables gained attention, to be now considered as inevitable.\(^{[2,10,11]}\) Indeed, cable manufacturers have realised that, nowadays, the scrap of production can range up to a loss of 4 wt-% of the production.

A differentiation has first to be made between the recycling of plastic scrap from manufacturing, called **Primary Recycling** and the one from cables after use (at their “end of life”), called **Secondary Recycling**. The first method is based on unused/unaged materials, usually of known composition and already pre-sorted, whereas the latter one is based on a mix of aged cable of different types, with a different degree of aging/damage and of unknown properties and composition. We can directly figure out that the difficulties for achieving the Secondary Recycling are much higher than for the Primary one. The researches have consequently started by investigating the Primary Recycling, drawn by its relative easiness.\(^{[2,11]}\)

It is commonly known that an **intern/closed Recycling**, namely a reuse of the material in the same industry as the one that has manufactured it, is the most intelligent and effective solution. In most of the projects, it is therefore considered that the scrap of cables should be reused in new cables. This elegant solution is also the one considered in this Research Programme. This can be practically achieved by a classical process of subcontracting the waste from the cable manufacturer to a specialised recycling company, but also internally in the manufacturing company. This last solution is effectively the most reliable and cost saving one, but a substantial difficulty usually stems from displaying the adapted equipment.

Indeed, recycling is not a “one-way process”, and three different steps are usually considered.
1.3.2. The Recycling steps \cite{2,10,11,12,13}

The recycling begins with a first step of **collection** of the targeted materials, usually stored in-house in the case of scrap from manufacturing.

It is followed by the step of **separation** of the lumps, which is usually divided into 3 stages:

Firstly, the stored scraps undergo a coarse separation, based on two parameters:
- The size of the cable: the power cables (PE, PEX based), used in air and ground, are distinguished from the thin cables (PVC based) used in buildings and vehicles.
- The nature of the metal part: aluminium or copper.

Secondly, in order to separate more accurately the polymeric and metal parts, the cables are granulated into 10 to 20 cm long pieces and ground into a 2 mm powder before being sent to the magnetic separation table. Two fractions, with a rather good purity are obtained: the metal part and the polymeric part.

However, as the polymeric part still consists of a mix of different polymers, a third separation step is needed to sort out each polymer. Mainly two processes, based on different physical parameters, were developed:
- Density separation (“Swim – sink”): the polymers are separated according to their density, by being transferred from one float-sink tank to another. This method is particularly accurate for separating PVC from other polyolefins (PP, LDPE, XLPE).
- Triboelectric or electrostatic separation: the mix of plastic is charged triboelectrically, before going through an electric field for separation. This method is particularly well adapted for separation of plastics of similar densities (XLPE / HDPE / LDPE), but very sensitive to the presence of additives.

In addition, a process specially designed for the sorting of cable plastics has been installed at Stena Recycling and evaluated within the R&D Program “Wire and
Cables”: PlastSep. This process allows an effective separation of the light plastic fraction (polyolefin: PE, XLPE) and the heavy fraction (PVC), with a small amount of remaining metal.

Needless to say, this Separation step is the determining step of the whole recycling process, governing the success of the coming recovery. Therefore a lot of studies aim at designing the most efficient, low cost separation process, at any stage: Coarse separation, Metal Separation or Polymer Separation.

The last step is the recovery of the sorted materials, which can be of three different kinds:

- Material or mechanical recovery: the sorted materials are pelletized then reprocessed in order to form new products. This is particularly successful for PVC, still on development for polyolefins.
- Chemical recovery or “Feedstock recycling”: degradation of polymeric materials into low molecular weight compounds. Since the depolymerisation is technically possible only for condensation polymers and PVC, the PEs are mostly pyrolysed to produce fuel oils and synthesis gases.
- Energy recovery: as polymers present a high-energy content, incineration is an interesting recovery solution, which reduces the volume and recovers energy from the waste. However, this is a bit tricky for PVC wastes, as a particular attention is drawn on the chlorine content which must be below 1%, to avoid hydrochloric acid and dioxins.

In this step, the challenge is to define an economic and environmentally-friendly process, which yields to end-use applications of economic value.

1.3.3. Mechanical recyclability of plastics: Polymer blends [1,8,11]

Compared to glass or metals, plastics are more difficult to recycle mechanically. Indeed, all along the two main steps of the mechanical recovery (pelletizing, reprocessing), numerous issues have to be faced.

First of all, after being used, the collected and sorted scraps usually have rather unknown properties: it consists of one kind of polymer, but with different degrees of aging/damage. Even if a Primary Recycling is considered, there is still a mix of grades in this polymer fraction, with different additives, leading to difficulties in finding the adapted processing conditions. Some attempts have been made to remove the additives, but this has turned out not be economically interesting (the polymer is too viscous), and often the polymer was even damaged during the processing.

However, the biggest issue concerns the second step of the mechanical recovery: in this step, the pelletized flakes can be directly reused alone, but most of the time, the properties of the recycled material alone being not satisfying enough, the pellets are mixed with other materials. Unfortunately, as it is commonly known, polymers have a low entropy of mixing. This means that most polymer blends will turn into a two phase system: there is immiscibility.

This property directly stems from the unusually high molecular weight of polymers. Because of this, and of thermodynamic laws, the miscibility can only be achieved if
the enthalpy of mixing is very small or negative, that is to say that the mixed polymers have similar solubility parameters or that specific intermolecular interaction (hydrogen bonds, dipoles interaction,...) exist between them. As this is not the case for most of the considered polymers, they tend to phase separate in a melt state.

This phase separation leads to the existence of phase boundaries, i.e. weaknesses in the material, so that polymer blends are usually considered for limited applications. However, this does not mean that interesting properties cannot be obtained in this way.

When the polymers are immiscible, the two phases (A and B) organise themselves in order to minimize their interfacial area, what is done into three possible ways, depending on the volume fraction and viscosity of each:

![Figure 8a, b, c: Morphology of immiscible blends: (a) B is dispersed in the continuous phase A, (b) A and B are co-continuous, (c) A is dispersed in the continuous phase B](image)

It can be logically deduced that the properties of the (a) blend largely depend on the polymer A, as it will absorb all the stress and energy when the material is stressed, but as no strong bond between the two phases exist, the ultimate strength will be limited by these weak bonds, so that the mechanical properties will be finally poorer than the ones of A. The same reflexion can be made for the (c) case, for the continuous phase B.

Nevertheless, these properties can be improved by processing in a one flow direction: the spheres of the dispersed phase turn into rods, reinforcing the continuous phase such as in a composite.

In addition, when the proportion of the two polymers is well chosen, the morphology (b) can be obtained, in which the surface contact of the two phases is high, so that the load will be equally divided on both phases, making the material stronger.

In certain cases, a compatible blend is also obtained: it is an immiscible blend, but which has the properties of a miscible one (the properties of a miscible blend are often between the ones of each unblended constituent, even sometimes better in a case of synergy).

Please note that other possibilities exist for improving the properties of polymer blends, but they are usually too costly and not used in the case of recycling:

- Use of compatibilisers
- Enhancement of the interactions between the two phases by creation of bonds between them (cross-linking, surface reactions,...).
It is consequently usually said that plastics should be of nearly identical composition in order to mix efficiently, and that a knowledge of the morphology of the mix, of its dependence on the processing conditions and of the compatibility of the phases is required enhance the properties.

### 1.4. Recycling of XLPE: From the first studies until now

In the case of the thermoset XLPE, which is neither remeltable nor soluble because of the cross-links, some investigations for finding an economically viable recycling method have been running for more than 15 years, considering any kind of recovery.

#### 1.4.1. Feedstock recycling

At the beginning of the studies, a feedstock recovery was considered: by heating the XLPE to 430°C with the presence or not of a catalyst, the thermoset degrades to become light fuel oils or waxes. Particularly, a catalytic degradation (with a silica-alumina solid catalyst) turns out to give liquid olefins (mostly from n-C_5 to n-C_{12}) at a good rate.\(^\text{[14]}\)

#### 1.4.2. Material recovery

The investigations have then rapidly focused on mechanical recycling. To enable this, two main methods to go around the processing difficulties stemming from the thermoset character were investigated:

- the cross-links are saved but the material is ground in small particles and reprocessed alone or in a blend.
- the cross-links are destroyed in order to enhance the flowability and processability of the material, either mechanically or by use of supercritical alcohol.

In this first method, the goal is to keep the interesting thermoset properties of XLPE (dimensional stability, chemical resistance,...), but diminish the length of the network by cutting it into pieces. This is practically achieved by grinding or pulverising the scraps collected. The chips or powder obtained are then reprocessed. Reprocessing XLPE alone does rarely give an interesting material, as the mechanical properties are satisfying only if the material is mechanically cohesive, namely if elevated temperature and pressure are used, and an adequate level of chain entanglement is assured by the process. The injection moulding turns out to be the most adapted process for this 100%-reprocessing, as the mechanical cohesion of the material is higher than the one obtained after extrusion or compression moulding. In addition, almost the same results were observed for injected-moulded crumbs (1,2 mm diameter) and pre-heated large chunks (as large as the feeding opening), what means that the grinding step would then not be required (cost saving). However, no trials for real applications were investigated.\(^\text{[15]}\)

A more common reprocessing way is to blend the XLPE with another material, mostly HDPE: after being pulverised in granules of an average 0,6 mm size, the XLPE was successfully blended by injection moulding with HDPE. The processability was satisfying up to 40 wt.-% of XLPE, and the higher the content of thermoset, the better
the impact strength. It was consequently concluded that the XLPE could be a useful impact modifier of virgin resins.\textsuperscript{16}

Some further experimentation has actually been performed in Swerea IVF within the Research Programme. This time, the scraps were reduced into a 0.5mm powder and injection-moulded with HDPE up to a concentration in XLPE of 70 wt-\%. The processability was validated for a content up to 70\%, and the samples obtained were nicely homogeneous and smooth. It was noticed that the higher the content of XLPE, the stiffer and the more viscous the injected-moulded material was. These blends also corroborate the concept of “impact modifier role” of XLPE, as its observed interaction with the PE fraction was mechanically analogous to thermoplastic elastomers (impact modified thermoplastics), such as vulcanized EPDM particles in a PP matrix. Some practical knowledge has been gained, and the need of resorting to a two feeder injection moulding machine for more easiness was questioned. The best set of properties was obtained with a 60 wt-\% blend: the stiffness, ductility and impact resistance of the recycled material were quite high, so that the properties were roughly the same as those of LDPE. Consequently, it was envisaged to use this blend in applications similar to those of LDPE: bowls, tanks, pipes, sheets, jacket of cables, ... but no trials at larger scale were performed.\textsuperscript{17}

In the second method, the goal is to get rid of the thermosetting structure by breaking the cross-links, so that the meltability of the material is improved, and the viscosity reduced.

The degradation of these cross-links can be achieved mechanically, with an adapted melt and shear kneading method. In this thermoplasticizing process (process of obtaining a thermoplastic material from a crosslinked material by breaking the cross-linked structure and hence reducing the molecular weight), the XLPE has first been chopped in chips of 5 to 10 mm\(^2\) before being thermoplasticized into pellets of diameter of 2-3 mm. A first attempt was made to reprocess it alone, by press moulding or extrusion. The processability was good, as easy as for LDPE. The recycled material shown a few differences in comparison with LDPE, as only a few cross-links and constituents of high molecular weight remained, and the control of the level of reduction of the molecular weight was easily assured. However, in the case of silane-crosslinked polyethylene, the recycled material obtained had a rough surface.\textsuperscript{18} It was also realised that the mechanical properties of XLPE with reduced molecular weight were not so interesting, so that further investigation were consequently performed. The XLPE with reduced molecular weight was blended with virgin PE for a special wire sheathing application. It was noticed that for a content in XLPE lower than 25\%, the material resulting was almost the same as the virgin material, and that it could even be recross-linked with traditional processing method, to get a material usable in wires.\textsuperscript{19}

The latter method of degradation of silane cross-links developed consists of a selective decomposition of the siloxane bound using supercritical alcohol. Indeed, the siloxane bond can theoretically be decomposed by water or alcohol, but, at room temperature and pressure, alcohol/water has a very different polarity than XLPE chains, so that the liquids cannot dissolve into the thermosets and reach the siloxane bonds. On the contrary, used in supercritical conditions, the fluids can have the adequate polarity to reach the bonds, decompose them selectively and reduce the
viscosity of the material. A continuous process was established for that purpose, which might be extendable to industry, and the siloxane bond were successfully decomposed selectively by supercritical methanol. The structure obtained is close to the one of a silane-grafted polyethylene, so the material can then be recross-linked. Some attempts to recycle the XLPE of insulation of cables by this way turned out to be successful.[20]

Finally, between all these possibilities, some mixed solutions have been tested, such as an extrusion of a blend of XLPE/LDPE performed under supercritical methanol. The processability was validated for contents up to 40% and the strand obtained had a better tensile strength and elongation at break than before. However, the samples were irregular, and no trials at larger scale were performed.[21]

1.4.3. Project objectives: investigation of blends with LLDPE

Bearing these previous investigations in mind, it was decided to investigate the processability and the properties of the recycled material stemming from a mechanical recycling of XLPE when mixed with LLDPE.

A mechanical recovery by grinding and reprocessing is economically the most interesting method for the cable manufacturers: it requires no complicated/particular equipment, contrary to a feedstock recycling or a mechanical recycling with uncross-linking (for both supercritical methanol and shear processing). Therefore was considered.

Since the reprocessing of XLPE alone displays a rather limited interest, a blend was envisaged. A lot of possible blends could be considered. Until now mostly a XLPE/HDPE blend was studied, and Swerea particularly had experience of that. However, as the results obtained were interesting, but not good enough to be considered for a scale up development, it was decided to investigate another virgin material.

As XLPE is non-polar, some non polar material would favourably be considered. Furthermore, because of the thermodynamic rules of blends, it seemed sensible to consider another polymer from the same family of PE. Considering economic issues, the cheapest ones were targeted: LDPE or LLDPE. The latter being commonly used in cables, and never investigated until now, it was then chosen. The macromolecular structure of LLDPE should also be the most suitable one for a blend with the thermoset XLPE: as explained before, it is nicely linear with short-chain branching, so that it should be easier to mix with XLPE than the long-branched LDPE.

For all these reasons, the subject of this master thesis was focused on the investigation of a possible mechanical recycling of XLPE from cable and manufacturing waste by blending with LLDPE.
2. Experiments

2.1. Raw materials

2.1.1. XLPE

The lumps of XLPE used in this work come from the manufacturing of the insulation layers of power cables, by the cable producer Nexans IKO AB. It consists of a silane cross-linked polyethylene, based on the silane/polyethylene copolymer Visico/Ambicat® produced by Borealis. This special copolymer is nowadays preferred to the classical grafted polymer presented above, as it avoids the (not always healthy) contact with silane products.

The manufacturing of the XLPE with this copolymer is performed in two steps. 95% of pellets of the copolymer are firstly extruded with 5% of catalyst, and the cross-linking is then achieved in a sauna, at high temperature and moisture conditions. The final degree of cross-linking obtained is between 60 and 70%, and this process assures that the whole cross-linking is completed by the end of the manufacturing, so that no cross-linking can take place in the following reprocessing steps.

According to the ASTM Standard F 876-93, the cable tubing of cross-linked polyethylene requires effectively a degree of cross-linking in the range from 65% to 89%. Indeed, by considering the method of manufacturing, an ideal degree of cross-linking can be found, which assures the best compromise between strength and flexibility and the better stress cracking resistance. Note that the XLPE should never be fully cross-linked, as it would lead to tubes too brittle and sensitive to stress cracking, but too little cross-linking can lead to properties not good enough for the application (hot set test not passed).

In the present case, a mix of two grades was received: Roughly 80% of a material of density \( d_1 = 0.923 \text{ g/cm}^3 \) Almost 20% of a material of density \( d_2 = 0.930 \text{ g/cm}^3 \).
In total, 250 kg were collected.

The residues were first granulated into flakes of a few millimeters at Hedins Plaståtervinning in Timmele.

![Granulated flakes from lumps of XLPE](image)

They were then sent to the company Pallmann Maschinenfabrik GmbH Co. KG, located in Germany, in order to be ground into two fractions:
- Five big bags of 20 kg, displaying a particle size \( \leq 0.6 \) mm. They would be called in the following as the “0,6 mm” powder.
- Four big bags of 20 kg, with a particle size \( \leq 0.4 \) mm, called in the following as the “0,4 mm” powder.

Both diameters will be used in the project, the dimension being stated in brackets. In the case that the diameter is not mentioned, it would be, by default, considered as 0,6 mm. Indeed, this material is economically the most interesting one: its grinding is less expensive than the one of the 0,4-diameter material, so it corresponds more to the recycling cost-saving objective.

The visual appearance of the XLPE is the one of a light powder, which flies quite easily and has different colours (blue, red, green, black,...)

![0,6mm powder of XLPE](image)

2.1.2. LLDPE

Two grades of LLDPE were used for this work:
The “a” type: LL 6201 Series from Exxon Mobil
The “b” type: LL 1004YB Film from Exxon Mobil.

The main difference between them is their molecular weight: the “b” type has a higher molecular weight than the “a” type, making it commonly used for extrusion.

<table>
<thead>
<tr>
<th>Denomination</th>
<th>Commercial Name</th>
<th>Producer</th>
<th>MFI (g/10min ASTM D1238)</th>
<th>Density (g/cm³)</th>
<th>Peak Melting Temperature (°C)</th>
<th>Hardness Shore D</th>
</tr>
</thead>
<tbody>
<tr>
<td>LLaDPE</td>
<td>LL 6201 Series</td>
<td>Exxon Mobil</td>
<td>50</td>
<td>0.926</td>
<td>123</td>
<td>62</td>
</tr>
<tr>
<td>LlbDPE</td>
<td>LL 1004YB Film</td>
<td>Exxon Mobil</td>
<td>2.8</td>
<td>0.913</td>
<td>121</td>
<td>51</td>
</tr>
</tbody>
</table>

*Table 1: Technical data for the two types of LLDPE*

![Pellets of LLaDPE (left) and of LlbDPE (right)](image)
2.2. Analyses

2.2.1. Notched Charpy Impact

As the impact strength of a material depends greatly on the geometry of the sample and on the temperature, some standard procedures have been fixed in order to obtain comparable results. One of the standard flexed-beam impact tests is called the Charpy test, and is commonly used for all kinds of material.

In this test, a beam of material (notched or not) is struck by a swinging pendulum at the bottom of its arc, so as to cause fracture. The amount of energy lost in the process, called impact energy, is then obtained from the height to which the pendulum rises after the impact. The sample is held horizontally but not clamped. (see Appendix 1 for details about the apparatus used)

The test is performed according to the standard ISO 179/1eA.

A rectangular sample, with a length of 80 mm, a width of 10 mm and a thickness of 4 mm was firstly notched in its middle point. The notch depth was of 2 mm, and the radius of the tip of 0,25 mm. The sample was then carefully placed on the support (62 mm span), so that the centre of the notch was located directly in the plane of the impact, but not on the side directly hit. After releasing the pendulum, if the break was complete or hinge, a percentage of energy was displayed and recorded.

The impact strength is calculated from:

\[ \alpha_{CU} = \frac{W \times 10^4}{h \times b_N} \]

With:

- \( \alpha_{CU} \) the notched Charpy impact strength (kJ/m²)
- \( W \) the corrected energy absorbed by breaking the specimen (J). Obtained by multiplying the recorded value by the intrinsic energy of the pendulum.
- \( h \) the thickness of the test specimen (mm)
- \( b_N \) the remaining width at the notch base of the test specimen (mm)

For each trial, 10 samples were tested.

2.2.2. Tensile testing

Tensile testing is one of the most fundamental test for evaluating the mechanical properties of plastics. Indeed, it unveils the stress-strain behaviour of the material, allowing a possible classification and an easy comparison with other materials. With it, an evaluation of the stiffness, strength, toughness and ductility is rather easily obtained.

The test was performed following the ISO 527. The stress-strain curves are obtained by measuring the force needed to elongate the sample at a constant rate of extension. This is practically achieved by pulling apart the two grips which clamp the
specimen, and record the extension with an extensometer attached to the gage section. (see picture of the apparatus in Appendix 1)

The specimens used were all of the dumbbell shape, but of different proportions. For each sample, five specimens were analysed.

The analysis of the curves is focused on the yield point and break/fracture point. This first one, delimiting the zone of elastic response to the plastic one, gives an idea of the stiffness of the material. The latter one, delimiting the maximum strain and stress at break, gives an idea of its toughness. For each analysis, the two points were detected automatically by the computer, but in most of the case, some adjustments of the real observed point were performed by hand.

Three (constant) rates of extension were used: 1mm/min, 50 mm/min or 250 mm/min.

A very small rate of extension (1 mm/min) ensures a high precision, what is important in the case of determination of the elastic modulus. This intrinsic parameter is determined by calculating the slope of the curve between 0,05 and 0,25% of elongation.

In order to obtain the whole curve, higher speeds are required. Different standard rates are then advised, depending on the processing method used for producing the samples. In the case of injection moulding, a rate of 50 mm/min is usually recommended, whereas extruded samples are rather studied at 250 mm/min. In addition, since the cable industry has its own standard rates, in most cases, both elongation rates were used.

2.2.3. Melt Flow Index (MFI)

This analysis aims at determining the melt flow index: the mass of a thermoplastic resin (in grams) that flows in 10 minutes through a capillary of specific diameter and length under a specific pressure and constant temperature.

The experiments followed the ISO 1133, Method A (1999), such as the Cable Standard IEC-60811-41. Under these conditions, the melt index is the amount in grams of a thermoplastic that can be forced through a 2,0955mm orifice when subjected to 20N (2,160 kg) in 10 min at 190°C.

Practically, the trials are achieved by pouring the powder/pellets (roughly 4 grams) in the pre-heated compartment, put them under load and wait for 4 minutes of warming. The capillary is then open, and the quantity of falling material in a cut-off time (15/30/45 sec) is weighed for at least 5 samples. (see apparatus in Appendix 1)

By measuring the ease of flow of the melt, this analysis gives us an indirect measure of the molecular weight: a high melt flow rate corresponds to a low molecular weight. In our case, dealing with a blend of two materials, it evaluates rather the viscosity of the recycled material at a certain shear rate, what is useful when considering its processing and applications.
2.2.4. Hardness

This test aims at determining the relative hardness of materials, using a similar method as the one of metals and ceramics. It consists of a measurement of the load needed to penetrate with a special indentor (shore D) the material under a specified time. (see detail of the apparatus in Appendix 1)

2.2.5. Scanning Electron Microscopy

The Scanning Electron Microscope is a new generation of microscopes developed in the 1950’s. Instead of resorting to light to form an image, it uses electrons, allowing a larger depth of field, a higher resolution and quality of the images than the traditional ones.

When a beam of electrons hits a sample, different X-rays and electrons are emitted. Depending on the level of interaction of the sent electrons with the atoms of the surface, three kinds of electrons can be emitted: the primary back-scattered electrons, the secondary electrons and the Auger electrons. In this case, only the back-scattered electrons were considered by the detectors, and converted into a signal sent to the screen. As these ones are resulting from the interaction of the electrons of the beam with the nucleus of the surface atoms, the signal obtained is proportional to the weight of the scanned element: the heaviest the element, the brightest the signal on the image.

Practically, the beam of electrons is produced by an electron gun and guided by electromagnetic fields in vacuum conditions to the sample.

The SEM with back-scattered electrons can be equipped with a special detector: the energy dispersive spectrometer (EDS). This particular detector records the characteristic energies/wavelengths emitted by the impact of the highly energetic electron beam. By comparing this spectrum with a data base, an identification of the chemical composition of the sample can then be obtained.

Because the SEM resorts to vacuum conditions and uses electrons to form the image, a special preparation of the sample must be done. The cross-sections are firstly roughly ground in surface and immersed in an epoxy resin. After 24 hours, the sample is polished in different steps. The goal of this stage is to obtain an even polished surface of the cross-section in order to optimise the quality of the pictures. In our case, as one phase is soft, at least 4 polishing steps are needed, ranging up to a last step with a diamond, 1µm, polisher. Finally, a layer of carbon, assuring conductivity, is applied on the sample.

Figure 12: Sample prepared for SEM analysis
2.2.6. **Differential Scanning Calorimetry**

The Differential Scanning Calorimetry is a classical method for characterising the thermophysical properties of polymers. It allows the determination of the temperature of the main phase transitions (melting or glass temperature), the heat capacity of the material, the heat of crystallisation and the degree of crystallinity, but we can also estimate the temperature history of a sample and follow directly some reactions (cross-linking).

The method is based on a recording of the difference of power required to keep the sample and the reference at the same temperature. Supplied by the heaters, this amount of heat aims at compensating the phase changes or reactions of the sample that might take place as the global temperature is raised. Indeed, some reaction taking place might be endothermic, such as melting or exothermic, such as a crystallisation. (See apparatus in Appendix 1)

This analysis has the advantage of requiring a small quantity of the sample, but it is a destructive one. The instrumentation used is sensitive and responds rapidly, but a calibration needs to be performed frequently. The quality of the baseline (presence of noise or jumps) is checked up by running an analysis with two empty pans and a trial with a calibrant (indium in our case).

2.2.7. **Environmental Stress Cracking Resistance: Bell Telephone Test**

The Bell Telephone Test is one of the most common Environmental Stress Cracking Resistance tests in the cable industry and it is often required before considering any possible application of a material.

This test consists of an evaluation of the time of resistance to crack/craze initiation of samples subjected to a constant strain while being immerged in a solution of alcohol.

The test was performed according to the Cable Standard IEC 60811-4-1, method B. Rectangular test specimens (38 x 13 x 3 mm) were notched and bent with the notch pointing upwards in a metal U-shaped specimen holder. The holder was then placed in a glass tube containing a 10 vol-% Igepal CO-630 water solution (2-[2-(4-nonylphenoxy)ethoxy]ethanol). The tubes were sealed and placed in a water bath at 50°C for 48h. Failure of the samples was then recorded, given by the appearance of cracks visible by a naked eye after 48h.

*Figure 13: Bell Telephone Test according to the IEC 626/04*
2.2.8. Surface roughness 3D-Analysis

During the trials, we have realised the importance of controlling the surface roughness of the samples, as a rather smooth surface is required for some applications. However, up to now, no standards for the cable industry advising a method of measurement of this roughness exist, and the only assessment is based on a simple touching feeling.

It was therefore decided to investigate a way of characterizing the surface roughness of the samples, in order to be able to compare it quantitatively. As the Textile Department often resorts to three dimensional pictures of their samples, we have considered attempting a digitization. These analyses were performed by the company Cascade Computing AB.

- Description of the measuring system

The optical measuring system used for digitizing the cable parts is named ATOS III SO (SO= Small Object). It is manufactured by GOM mbH in Braunschweig, Germany, and is especially adapted for digitization of small parts. The system is using two high performance CCD cameras with 4 MPixel each to collect data. A light insensitive projector projects a fringe pattern on the surface of the cables and the cameras collect measuring points with help of the fringe pattern. The system is furthermore equipped with lenses giving a measuring volume of 30 x 30 x13 mm (the measuring volume is the volume within the system can collect data). The achieved accuracy is around ± 0,005 mm.

![Figure 14: Optical measuring system ATOS (left), with written permission of E. Larsson - Schematic representation of the analysis (right)](image-url)
• Preparation of the samples

A 4cm long piece of the cable was firstly cut and almost 2cm of it was stripped to let only the aluminium conductive base, which can be easily fixed on the reference frame.

Secondly, the samples were sprayed with Titanium Oxide (in solution into an alcoholic solvent), thanks to an airbrush. A very fine layer, approximately of a thickness of some microns, was applied on the surface, assuring a better contrast for the analysis. Indeed, as the plastic jacket can display a rather transparent surface, the optical measurement systems may “look through” the surface (rays are not reflected by the outer surface, but by the opaque surface of the aluminium base), so that the analysis is distorted. The powder can, after measuring, be removed from the object.

• Performing the digitization

After preparation, the cable piece was fixated in a reference frame. This reference frame was measured before with a photogrammetric system called TRITOP, to get a reference point file which will be used by the ATOS system, allowing the whole object to be 3D-scanned in the frame with highest accuracy.

The frame was placed on a rotation table that is controlled by the ATOS system. The system started to collect data from different angles of the cable part. When the whole surface was covered on upper side of the cable part, the frame was turned upside down and the other side of the cable part is digitized. After digitizing, the results were calculated and a 3D-scan of the cable part was produced.

• Analysis of the scans

Firstly, a model of the reference (sample of LLbDPE alone) was scanned, giving us the “master model” which will be used for all the calculations. The master model consists of a perfect cylinder of the diameter of the reference sample (4mm).

As the scans were not 100% straight in shape, we needed to select a part of them that could be used for the comparison with the master model. In order to evaluate the quality of the method, two areas, of approximately 3mm wide, situated roughly 1 mm far from each end of the specimen were chosen. Each area was then aligned to a cylinder with the diameter of 4mm (master model), using a best-fit alignment of the selected points. Finally, a deviation colour plot was created, and the minimum, maximum and average deviations values were collected (these values will not be used for the conclusions).

Besides, some more calculations were performed. Indeed, if we consider a cross section of the 3D-digitized sample, we get a typical two dimensional representation of the roughness, with a mean line and sinusoidal-like fluctuations.
Consequently, the classical ways of calculating a 2D-roughness have been considered, according to the standard JIS B 0061 and JIS B 0031. Upon all the existing parameters, three of them (the most common ones) were chosen: Ra, Ry, Rz.

- The arithmetical mean roughness, Ra, is the most common parameter for defining roughness. Its formula (see below) takes into consideration not only the height of the mountains and valleys, but also their frequency. As the feeling of roughness comes both from the existence of humps and hollows, and from the quantity of them, it is consequently the most adapted parameter to describe the roughness in our case.

\[ Ra = \frac{1}{\ell} \int_{0}^{\ell} |f(x)| \, dx \]

- The maximum peak (Ry) gives the value of the highest deviation: from the lowest valley, to the highest mountain. It consequently gives an idea of the maximum difference we can observe between mountains and valleys, without taking into consideration their periodicity.
The Ten-point mean roughness (Rz) follows the same idea as Ry, but instead of taking two points, it is based on 10 points: the 5 tallest mountains and the 5 lowest valleys.

![Figure 18: Calculation of Rz](image)

The values of Ry and Rz are consequently going to be close to each other, and much higher than the values of Ra.

Please note that all these calculations were done on excel, and based on the mean line.

### 2.3. Investigation of the processability of the blend

#### 2.3.1. Injection moulding

A first processing way to mix XLPE and LLDPE is injection moulding.

2.3.1.1. Description of the process

Injection moulding is one of the most common manufacturing techniques of plastics. Indeed, adapted to both thermoplastics and thermosets, it offers a wide possibility of shapes, a very good repeatability and a high speed production (automated process). However, as the costs of the equipment and of the mold are high, it requires a large investment.
The equipment is usually based on a reciprocating “screw extruder”, which assures the injection of the melt, and on a mold attached to a clamping part.

Four main zones can be distinguished in the extruder:
- the feed zone, where the material is conveyed forward,
- the compression zone, where the melting takes place,
- the metering zone, where additional pressure and mixing ensures a good homogeneity of the melt,
- the injection zone, which consists of a pool at the end of the screw, where the molten material waits before being pushed in the mould by a forward translation of the screw.

Arriving into the moulding section, the melt will flow through the nozzle and the sprue, followed by different runners and gates within the mould tool, to form finally the moulding.

The injection moulding is a cyclic process, divided into four main steps:
- Melting: the screw is turning, melting more and more of the fed material, which is then collected in the injection zone.
- Injection of the melt: the screw stops turning and advances forward, pushing the melt through the nozzle to the mould.
- Cooling: the screw retracts and the moulding is cooled.
- Removal of the part: the solidification being reached, the mould opens and the part is removed, as the process starts again.

The time of this cycle is the key element of the process, as it has to be reduced to the minimum but intelligently, in order to keep a good quality of the moulded parts. This is a difficult task since a lot of parameters are involved.

2.3.1.2. Trials performed

The trials were performed on the 18/09/2008 and the 23/09/2008, with the equipment of Swerea IVF (see Appendix 1 for the reference of the machines). The moulding chosen was a dumbbell specimen (tensile test bar), which allows further analyses following the standards.

![Figure 19: Geometrical shape of the dumbbell specimen](image)

The goal of these trials is firstly to evaluate the technical feasibility of an injection-moulded blend. The effect of the content of XLPE was furthermore investigated, in order to determine some minimal or maximal content accepted.
The trials planned and achieved are summarized in the following table:

<table>
<thead>
<tr>
<th>Number</th>
<th>Name</th>
<th>Processing</th>
<th>% of XLPE (wt.)</th>
<th>% of LLDPE (wt.)</th>
<th>Pre compounding</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>LLa30l</td>
<td>Injection moulding</td>
<td>0</td>
<td>100</td>
<td>No</td>
</tr>
<tr>
<td>2</td>
<td>LLa40l</td>
<td>Injection moulding</td>
<td>40</td>
<td>60</td>
<td>No</td>
</tr>
<tr>
<td>3</td>
<td>LLa50l</td>
<td>Injection moulding</td>
<td>60</td>
<td>40</td>
<td>No</td>
</tr>
<tr>
<td>4</td>
<td>LLa60l</td>
<td>Injection moulding</td>
<td>70</td>
<td>30</td>
<td>No</td>
</tr>
</tbody>
</table>

Table 2: Summary of the Injection Moulding Trials

It has to be noticed that:
- No “pre-compounding” was achieved: the two materials (XLPE and LLDPE) were simply mixed by hand before being poured in the hoper. The mixing was always achieved a few minutes before feeding, in order to ensure a relative homogeneity of feed. It consisted of a straightforward shaking of the bag during 30 seconds. Roughly 2 kg of material were needed for each trial, in order to obtain enough specimens for the planned analyses.
- The XLPE particles used for these trials were only of particle size ≤ 0.6 mm.
- Only one type of LLDPE was studied, the “a” type: LL6201 Series (low molecular weight). Indeed, it turns out to be the most adapted grade of LLDPE for this kind of process.

The processing conditions are summarized in Appendix 2. It was particularly noticed that the properties of the feed changed with time, leading to a necessary change of the parameters during the trials. **The homogeneity of the feed was not perfect, so that we might expect a certain drift of composition in the samples.** It would be interesting to analyse this expected drift of composition, in order to conclude about the necessity of a pre-compounding which will assure a more homogeneous feed.

Concerning the analyses, each sample was subjected to the following analyses:
- Tensile testing and determination of the elastic modulus
- Notched Charpy impact strength at room temperature (RT) and -30°C
- Hardness Shore D
- Melt Flow Index (MFI) determination.

In addition, for the samples containing 40% and 70% of XLPE, some further analyses were made:
- A Scanning Electron Microscopy (SEM) analysis of cross-sections, in order to observe the morphology of the specimens and their homogeneity.
- Differential Scanning Calorimetric analysis (DSC) of particular parts of the samples, in order to determine their physical properties and chemical composition.
- The Bell Telephone Test (BTT), to investigate their cracking resistance.

### 2.3.2. Tape extrusion

#### 2.3.2.1. Description of the process \(^{[1,6]}\)

The extrusion is a widely used method for processing thermoplastics, as it efficiently mixes, melts and pumps them to produce objects of a fixed cross-sectional profile with good surface finish. It is furthermore a non-expensive and straightforward
method, which is adapted to high production, due to the continuous forming of profiles.

The equipment required for extrusion is based on the extruder, which gives the adequate compressive and shear stresses to melt, mix and transport the melt without degradation. In addition, a cooling stage and puller are needed.

The polymer is initially put into the hopper, and fed the extruder channel by gravitation through the feed throat. It is then conveyed by the screw through its three zones (feeding, compression, metering), where it becomes a melt, and pass through the screen pack and breaker plate, before entering the die. The die provides the shaping of the final part, which is then cooled after exiting the extruder, typically by a water bath, before being pulled to the removal.

The screw and the die of the extruder are the most important parts of the process. The screw (and the barrel) should provide steady state processing conditions. The die should be designed to give the exiting flow the required shape, taking into consideration the problems of die swell and possible shark skin.

2.3.2.2. Description of the trials

The trials were performed between the 14/10/2008 and the 28/11/2008, using the equipment of the Department of Materials and Manufacturing Technology of Chalmers (see Appendix 1 for reference of the machines).

The main goal of these analyses was to evaluate the technical feasibility of a tape extruded blend. In addition, three parameters were investigated. The effect of the content of XLPE was one more time under investigation, but starting this time at a lower content. Since most of the extruders used in industry only rely on one feed, it might be necessary to resort to pre-compounded pellets when considering applications. The effect of pre-compounding on the mechanical properties was consequently studied. Finally, the two kinds of LLDPE were used, in order to determine the most suitable one.

The trials planned and achieved are summarized in the following table:

<table>
<thead>
<tr>
<th>Name</th>
<th>Processing</th>
<th>% of XLPE (wt.)</th>
<th>% of LLDPEa (wt.)</th>
<th>% of LLDPEb (wt.)</th>
<th>Pre compounding</th>
</tr>
</thead>
<tbody>
<tr>
<td>LLa0E</td>
<td>Tape Extrusion</td>
<td>0</td>
<td>100</td>
<td>100</td>
<td>No</td>
</tr>
<tr>
<td>LLb0E</td>
<td>Tape Extrusion</td>
<td>0</td>
<td>70</td>
<td>70</td>
<td>No</td>
</tr>
<tr>
<td>LLa30E</td>
<td>Tape Extrusion</td>
<td>30</td>
<td>70</td>
<td>70</td>
<td>Yes</td>
</tr>
<tr>
<td>LLa30EPC</td>
<td>Tape Extrusion</td>
<td>30</td>
<td>70</td>
<td>70</td>
<td>Yes</td>
</tr>
<tr>
<td>LLb30E</td>
<td>Tape Extrusion</td>
<td>30</td>
<td>50</td>
<td>50</td>
<td>No</td>
</tr>
<tr>
<td>LLb30EPC</td>
<td>Tape Extrusion</td>
<td>30</td>
<td>50</td>
<td>50</td>
<td>Yes</td>
</tr>
<tr>
<td>LLa50E</td>
<td>Tape Extrusion</td>
<td>50</td>
<td>50</td>
<td>50</td>
<td>Yes</td>
</tr>
<tr>
<td>LLb50E</td>
<td>Tape Extrusion</td>
<td>50</td>
<td>50</td>
<td>50</td>
<td>No</td>
</tr>
<tr>
<td>LLb50EPC</td>
<td>Tape Extrusion</td>
<td>50</td>
<td>50</td>
<td>50</td>
<td>Yes</td>
</tr>
</tbody>
</table>

Table 3: Summary of the Tape Extruded Trials
In addition, the extrusion of a **standard material** used by the cable producer Nexans was performed with the usual processing parameters. This copolymer is a typical material used for inner-jacketing power cables, based on PE and Polyvinyl Acetate (PVA). This will allow us to have a reference for the analysis of the mechanical properties and to be able to conclude about a possible reuse of the test material as an inner-jacket.

### 2.3.2.3. Observations

The processing conditions are detailed in Appendix 3. Generally speaking, the extrusion of the different tapes was performed without problems, showing the **technical feasibility of the process for this blend**.

- **Tape extrusion**

![Figure 20: Rolls (left) and extruder (right) during extrusion](image1)

![Figure 21: Die of the extruder during tape extrusion](image2)

A single screw extruder was used for the tape extrusion, with a cooled roller system (note that the cooling is necessary for good results). The extruded tape obtained had a thickness ranging from 0.5 to 2.5 mm, and were about 9 cm wide. The targeted thickness was however of 2 mm, and we had difficulties in some cases to control this parameter.
Particularly, the **LLa0I sample was somewhat difficult to extrude**: the melt did not exhibit a real viscous flow but a rather liquid one. Unfortunately, this entailed the extrusion of very thin tapes (0,45 mm), which cannot be analysed.

**The maximum amount of XLPE in the blend that can be processed by this method is 70%**. However, the tapes obtained at this level were about to crush into pieces, even though they were quite thick.

However, by working at the highest speed of the extruder (150 rpm) and with a medium roll speed (0,5 rpm), the expected thickness was most of the time reached for the other samples. The temperature of the barrel was also adapted to the content of XLPE, ranging from 130°C to 180°C.

For the trials without pre-compounding, the materials were mixed a few minutes before being poured in the hoper. Nevertheless, **a drift of the feeding composition was visually observed**: after a certain time standing in the feeder, the LLDPE pellets had a tendency to flow first to the extruder as the XLPE powder were sticking a bit more on the edge. This was especially observed in the extreme case of a XLPE content of 70%: after half of the extrusion, the concentration of the “dry” XLPE powder was suddenly increasing at the die, leading to a break in the tape extrusion. These observations justified the need of investigating on the importance of a pre-compounding.

Concerning the influence of the nature of LLDPE used, for the same content of XLPE, the samples blended with LLaDPE displayed a visual appearance close to that of LLbDPE. But a further comparison of the mechanical properties has to be made before drawing any conclusions.

**A significant roughness of the samples was noticed.** It might be interesting to investigate if this would be **an asset or a bane**, and find out which parameters are influencing it. It was also pointed out that the roughness visually depends on the content of XLPE: the more XLPE, the rougher were the tapes. The pre-compounding seemed to reduce the roughness.

**Figure 22: Effect of pre-compounding on the visual aspect of the tapes (left sample is LLb50EPC and right one is LLb50E)**
Figure 23: Effect of the nature of LLDPE on the visual aspect of the tapes: a type (left) / b type (right) – Samples with 70% of XLPE

- Pre-compounding

Figure 24: Pre-compounding in process

For the pre-compounding, a twin screw extruder was fed continuously by two independent feeders. After calibration of each feeder, the extrusion was run at the processing conditions detailed in Appendix 3.

The two “strings” exiting from the die were cooled down in a three meter long water bath (20°C), and then chopped directly into pellets of average size. The manufacturing of the pellets was consequently rather straightforward and quick, almost self-running.

The water-cooling was preferred to air-cooling, as the extruded PE materials are usually sticky and, due to their low thermal conductivity, need a long cooling time. However, some negative aftermath stem from this cooling: in most of the cases, the pellets were still humid after being chopped and put in bags, giving a tendency to stick to the inner surface of the cone-shaped feeder, and not to flow out continuously in the single screw extruder. Consequently, **a drying of the pellets should be performed prior to the final extrusion.** This is particularly necessary when a large amount of material should be used, such as in the coming application trials.
Another negative aspect of this cooling method is that the cooling of the material is not even during the pellets manufacturing: the water contained in the bath was not renewed continuously, so that after a few hours, the temperature of the water bath had significantly increased, being less efficient. The two “extruded strings” were then warmer and consequently softer when they arrived to the chopper, leading to pellets with a distorted geometrical shape. They possessed then a higher roughness, what worsened their flow in the hopper, leading to problems of discontinuous feeding. However, at larger scale, this problem would not be encountered anymore and could then be neglected. At a laboratory level, this problem was circumvented by the use of another hopper: a funnel with a higher slope/inclination.

![Figure 25: Pellets from pre-compounding of the sample LLb50EPC](image)

The first visual observation seemed to encourage the use of a pre-compounding, since the pre-compounded tapes showed a better homogeneity and smoothness as the one without pre-compounding. This has now to be correlated with the results of analyses.

- **Analyses**

Each sample will undergo the following analyses:
- Tensile testing and determination of the elastic modulus
- Hardness Shore D
- Melt Flow Index determination.

In addition, for certain samples, the Bell Telephone Test (BTT) will furthermore be used.

**2.4. Investigation of possible applications**

**2.4.1. Wire extrusion at Draka Kabel**

**2.4.1.1. Description of the trials**

The trials were performed on the 14/01/2009, on the production process EP 90 of the plant of Draka Kabel in Nässjö. Before, the pre-compounding was made at the Department of Materials and Manufacturing Technology of Chalmers on the 08 and 09/01/2009.
The main goal of these trials was to evaluate the processability of a pre-compounded blend for an extrusion of thin aluminium-based wires. A 1,2 mm thick jacket was extruded around the aluminium cable, corresponding to the standard thickness of power cables in the industry.

In addition, four parameters were investigated. The effect of the content of XLPE was one more time studied, ranging from 0 to 50%. It aims at determining the maximum content of XLPE that is allowed for that application. As the four different kinds of pre-compounded pellets manufactured prior to the extrusion did not always offer the desired content of XLPE, in some cases, a “dilution” of pre-compounded pellets into virgin LLbDPE was needed. This end hand-mix, performed just before pouring into the hopper might be heterogeneous during the time of feeding, as observed during our previous processing experiences. Consequently, the effect of the degree of “dilution” on the mechanical properties was considered.

We have, up to now, always resorted to the use of XLPE powder of a diameter ≤ 0,6 mm, as it was economically the most interesting one. Anticipating some roughness problems, we have for the first time also used some 0,4mm diameter XLPE powder. The influence of the size of the XLPE particles was consequently investigated during these trials.

Finally, as most of the standard jacketing materials used for this application contain additives, some antioxidants and carbon black were added in some cases, allowing us to investigate the influence of the presence of additives on the mechanical properties.

The trials performed are summarized in the following table:

<table>
<thead>
<tr>
<th>Name</th>
<th>% XLPE, φ=0,4</th>
<th>% XLPE, φ=0,6</th>
<th>Additives</th>
<th>End mix</th>
</tr>
</thead>
<tbody>
<tr>
<td>LLb0KE</td>
<td>0</td>
<td>0</td>
<td>No</td>
<td>No</td>
</tr>
<tr>
<td>LLb30(0,4)KE</td>
<td>50</td>
<td>0</td>
<td>No</td>
<td>No</td>
</tr>
<tr>
<td>LLb5(0,4)KED</td>
<td>5</td>
<td>0</td>
<td>Yes</td>
<td>Yes with KE 30</td>
</tr>
<tr>
<td>LLb5(0,4)KEAD</td>
<td>15</td>
<td>0</td>
<td>Yes</td>
<td>Yes with KE 30</td>
</tr>
<tr>
<td>LLb15(0,4)KEAD</td>
<td>15</td>
<td>0</td>
<td>No</td>
<td>Yes with KE 30</td>
</tr>
<tr>
<td>LLb15(0,4)KED*</td>
<td>15</td>
<td>0</td>
<td>No</td>
<td>Yes with KE 50</td>
</tr>
<tr>
<td>LLb50(0,4)KE</td>
<td>50</td>
<td>0</td>
<td>No</td>
<td>No</td>
</tr>
<tr>
<td>LLb30(0,6)KE</td>
<td>0</td>
<td>30</td>
<td>No</td>
<td>No</td>
</tr>
<tr>
<td>LLb5(0,6)KED</td>
<td>0</td>
<td>5</td>
<td>No</td>
<td>Yes with KE 30</td>
</tr>
<tr>
<td>LLb5(0,6)KEAD</td>
<td>0</td>
<td>5</td>
<td>Yes</td>
<td>Yes with KE 30</td>
</tr>
<tr>
<td>LLb15(0,6)KED</td>
<td>0</td>
<td>15</td>
<td>No</td>
<td>Yes with KE 30</td>
</tr>
<tr>
<td>LLb15(0,6)KED*</td>
<td>0</td>
<td>15</td>
<td>Yes</td>
<td>Yes with KE 50</td>
</tr>
</tbody>
</table>

Table 4: Summary of the wire extruded trials

In addition, a sample of a standard material used for this application was analysed, in order to give us a reference. It is a material from Borealis, called Borstar® LE8706, a linear low density polyethylene, UV-stabilized.
2.4.1.2. Observations

The processing conditions are detailed in Appendix 4. Generally speaking, the extrusion of the different wires was performed without problems, showing the processing feasibility of the material for this application.

- Wire extrusion

The production process EP 90 relies on a one-feeder single-screw extruder, a 15m long water bath for cooling, a puller and different bobbins and reel-up machines. For this thin cable jacketing extrusion, the extruder is using a tube tooling process. The extruded wires had a diameter of roughly 4mm and a roughness highly dependent on the content of XLPE.

The first trial performed, the extrusion of LLbDPE alone, gave us a reference: a very regular, glossy and smooth cable.

With the second trial, which consisted of a feed containing 30% of XLPE, we discovered the main issue of this processing: the roughness.

The roughness of the wires extruded during this second trial, assessed only by visual and sensitive impressions, was clearly too high, and would not be accepted for such an application. To reduce the roughness, some changes in the processing conditions can be done, such as increasing the temperature of the barrel or the speed of the
screw. As the latter was already at its maximum value, the heating of the extruder was increased (see Appendix 4). However, this did not have a large influence on the roughness and using our previous processing experience (extrusion of tapes), it was concluded that this problem might stem from the content of XLPE directly: the higher the content of XLPE, the higher the roughness. Consequently, the next trial was performed with a lower content in XLPE, what was achieved by “diluting” the pre-compounded pellets with pure LLbDPE. This end hand-mix, performed just before being poured into the hopper leads to a final content of 5%-weight of XLPE.

This third trial led to smoother cables, just as expected. The content of XLPE had, as expected, a strong influence on the roughness of the cables and **the more XLPE, the rougher**. It was consequently decided that the following trials should be made with a content of XLPE lower than 30%. We centred the trials to a low concentration of XLPE, namely on 15 and 5%-XLPE. This was achieved by diluting the pre-compounding pellets of 0,4 and 0,6 mm with pure LLbDPE.

However, as it was previously observed, a last hand-mix performed just before pouring might lead to a heterogeneous feeding. Consequently, the effect of the degree of “dilution” on the mechanical properties was studied, by comparing the properties of samples of the same final content of XLPE (15 weight-%), but coming from either KE 30 or KE 50 pre-compounded pellets. The visual appearance of the cables was very similar in both cases, so that we need to resort to the analyses results to conclude about this parameter.

No visual difference was noticed between cables of the same content of XLPE, the same dilution, but different XLPE size (0,4 and 0,6 mm). The analyses are required for drawing any conclusion.

Finally, since most of the power cables are based on jacket containing additives, it was decided to perform a few trials with additives. A classical masterbatch, named Polycable FC 7303 LD/P was added to the 5%- and 15%-XLPE trials. Commercialised by Polyplast Müller, it contains antioxidants and carbon black and consequently gives a black colour. A standard final content of 6,2 wt-% was used for each trial.

No changes in the processing conditions were made, and the cables obtained had a nice appearance, a little uneven but more glossy and smooth than without additives. Consequently, additional analyses of the mechanical properties after aging will be done.

Note that some trials containing 50wt-% of XLPE were achieved, even though their properties might not be particularly interesting. These trials gave us an important indication concerning the range of XLPE content that can be processed for this application: **the material was suitable for this application process for a XLPE content ranging from 0 to 50%**.
Figure 28: Extruded wires LLb5(0,4)KEAD

- **Pre-Compounding**

Prior to these trials, 72 kg of pre-compounded material were extruded at Chalmers, with the same equipment as the one used for the tape extrusion, which consists of a two feeders twin screw extruder, a water-cooling bath and a chopper (see reference of the apparatus in Appendix 1). Indeed, as concluded from the tape extrusion trials, in the case of a rather large scale use of the material, a pre-compounding is advised for easing the trials.

The pre-compounding was carried out without any problem with the water-cooling. Setting a feeding of, in average, 200g/min, and the extruder-speed at 130 rpm, the production of each material group has taken 2h, working almost without the need of human control (see details of processing conditions in Appendix 4).

As decided, all the pellets were based on the LL 1004 YB Film from Exxon Mobil (so called “LLbDPE”). Four groups of pellets of 18 kg each were extruded, and then pre-dried for 3h at 60°C:
- KE 30 0,4 : Material containing 30%-weight of cross-linked polyethylene, with particles of average diameter of 0,4mm.
- KE 30 0,6 : Material containing 30%-weight of cross-linked polyethylene, with particles of average diameter of 0,6mm.
- KE 50 0,4 : Material containing 50%-weight of cross-linked polyethylene, with particles of average diameter of 0,4mm
- KE 50 0,6 : Material containing 50%-weight of cross-linked polyethylene, with particles of average diameter of 0,6mm.

It was noticed that the pellets containing 30% of XLPE had a better appearance (shape, roughness, softness) than the ones containing 50%.

- **Analyses**

Most of the samples were subjected to the following analyses:
- Tensile testing and determination of the elastic modulus, before and after aging.
- Surface roughness analysis by 3-dimensional digitization.
- Bell Telephone Test (BTT).
2.4.2. Cable-jacket extrusion at Nexans

2.4.2.1. Description of the trials

The trials were performed on the 25/02/2009, on the production process Nr.23 of the plant of Nexans in Grimsås. Before, the pre-compounding was performed at the Department of Materials and Manufacturing Technology of Chalmers on the 12 and 13/02/2009.

The main goal of these trials is to evaluate the processability of the use of a pre-compounded blend for an extrusion of an inner-jacket of power cables. This inner-jacket was extruded over stranded conductors and covered by an extruded PVC-jacket a few weeks later.

But the interest in these trials was even greater, since a replacement of the usual material, a copolymer of LLDPE and Vinyl Acetate, was looked for. If the recycled material is adapted to the process and if the mechanical requirements are fulfilled, it would be considered as a possible substitute.

In addition, three parameters were investigated: The effect of the content of XLPE was one more time studied, ranging from 10 to 20%. This rather narrow interval was a direct consequence from the roughness problems encountered during the trials.

As for the wire extrusion, the pre-compounded pellets manufactured prior to the extrusion did not always offer the desired content of XLPE, so that in some cases, a “dilution” of pre-compounded pellets into virgin LLDPE was needed. Consequently, the effect of the degree of “dilution” on the mechanical properties was studied.

Finally, as the standard jacketing material used for this application contains some additives, particularly some lubricants, the influence of the presence of lubricants on the mechanical properties was investigated.

The trials performed are summarized in the following table:

<table>
<thead>
<tr>
<th>Name</th>
<th>Weight content in XLPE (%)</th>
<th>Lubricant</th>
<th>Dilution</th>
</tr>
</thead>
<tbody>
<tr>
<td>LLb10JED</td>
<td>10</td>
<td>No</td>
<td>Yes from 50%</td>
</tr>
<tr>
<td>LLb15(0,6)JFAD</td>
<td>15</td>
<td>Yes</td>
<td>Yes from 30%</td>
</tr>
<tr>
<td>LLb20(0,6)JE</td>
<td>20</td>
<td>No</td>
<td>No</td>
</tr>
</tbody>
</table>

*Table 5: Summary of the Cable Extruded Trials*

Please note that, in comparison to the wire extrusions, a rather small number of trials was performed. This is due to the difference in quantity of material required: as three kilos of material were needed to obtain one sample (150m) at Draka Kabel, here, 50 kilos were needed per trial.
2.4.2.2. Observations

- **Pre-compounding**

Prior to the jacket extrusion, roughly 150 kg of pre-compounded material were manufactured at Chalmers (usual unit – see Appendix 1 for references of the apparatus), and pre-dried for 2 hours at 60°C. They were all based on the LL1004 YB from Exxon Mobil (so called “LLbDPE”), and contained different amounts of XLPE:

- **CLLb30(0,6)**, which consisted of a compound containing 30% of XLPE, the pellets of XLPE being used were of the size 0,6mm. Quantity = 50 kg.
- **CLLb20(0,6)**, compound containing 20% of XLPE of the diameter of 0,6mm. Quantity = 60 kg.
- **CLLb50**, compound containing 50% of XLPE, from which 11,2 kg have been produced by using the XLPE pellets of diameter 0,4 mm and 19 kg resorting to the pellets of diameter 0,6mm. Quantity = 30,2 kg.

The pre-compounding was done without any problem. Setting a feeding of, in average, 200g/min, and the extruder-speed at 145 rpm, the manufacturing of **CLLb20(0,6)** and **CLLb30(0,6)** took 9h, working almost without the need of human control (see details of processing conditions in Appendix 5).

- **Inner-jacket extrusion**

![Figure 29: Extrusion of the inner-jacket on the production unit Nr.23, plant of Nexans](image)

The production process Number 23 consists of a one-feeder single screw extruder, a 100m long U-shaped water bath for cooling, a puller and different bobbins and reel-up machines (see Appendix 1 for details of the apparatus).

The inner-jacket was extruded over stranded conductors in a tube tooling process. The conductors, which were manufactured a few days before the trials, consist of 4 groups of stranded insulated aluminium wires. The inner jacket obtained had a thickness ranging from 1,4 to 1,8 mm, and an outer-layer of PVC was extruded a few days after these trials, giving the final power cable:
The design of these power cables is a typical Norwegian one, as it is manufactured for Norway. Two jackets are used, the outer one essentially for its flame retardant properties and the inner one as a good barrier to moisture. In addition, the 4 groups of stranded conductors are insulated and separated from the inner-jacket by a thin film of PP. In between the four groups, a filler in PE and polyester is added, in order to assure a global nice spherical shape of the cable (which cannot be clearly seen in the picture). Finally, a thin red string, coding for the manufacturer, is enclosed in the construction.

Using our previous processing experiences, it was decided to start with the pellets of lower content in XLPE; 50 kilos of the CLLb20(0,6) were poured in the hopper of the extruder, heated to 180°C (see Appendix 5 for more processing details). The extrusion took place without any problems, showing the processing feasibility of the material for this application.

The cables obtained were a little stiffer than the usual ones. This can actually be a problem as adequate mechanical properties are required for the inner-jacket, particularly concerning its tensile strength and strain at break. Some further mechanical analyses are needed to determine if the requirements are fulfilled or not.

In addition, this first trial gave a rather high surface roughness (evaluated by feeling), in comparison to the one of the copolymer (LDPE/VA) usually used for this application. It is the second time that this notion of roughness of the cable is underlined as an issue.

As before, we envisaged different solutions to solve this technical problem.

Firstly, we tried to find an optimisation of the processing conditions: the heating of the barrel and the speed of the screw were increased as much as possible, bringing no notable improvements. An attempt to enhance the smoothness of the extruded jacket by changing the die was performed as well, without success.

We secondly thought about resorting to additives to smoothen the cables. Indeed, the classical material used for this application does not only consist of the copolymer, but some additives and particularly lubricants are added, which might decrease the roughness.
Consequently, a trial with 1 wt-% of lubricants was performed by mixing by hand the 50 kg of feed. The lubricant consists of white pellets of fluorinated polyethylene commercialised by DuPont under the brand name Viton®.

However, as this solution did not turn out to be so efficient, a last parameter was finally considered: the effect of the content of XLPE. It is known from our previous trials that this solution is a successful one for diminishing the roughness (the less XLPE, the smoother), but it should always be considered as a “last resort”, as it diminishes the recycling interest. Consequently, the next trials were performed with a lower content of XLPE: respectively 15 and 10%.

These contents were technically achieved by diluting the pre-compounding pellets with pure LLbDPE, what gave the possible problem of heterogeneity of the feeding, and the need to consider the dilution as a possible influencing parameter on the results.

![Figure 31: Views of (left) a freshly extruded inner-jacket, (right) final power cables](image)

- Analyses

Each sample was subjected to **tensile testing before and after Aging**.
3. Results and Discussion

3.1. Injection-moulded samples

3.1.1. Notched Charpy Impact

<table>
<thead>
<tr>
<th>Name</th>
<th>Content in XLPE (weight %)</th>
<th>Notched Impact strength RT</th>
<th>Notched Impact Strength -30°C (kJ/m²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>LLa01</td>
<td>0</td>
<td>no break</td>
<td>5.25</td>
</tr>
<tr>
<td>LLa401</td>
<td>40</td>
<td>no break</td>
<td>2.349</td>
</tr>
<tr>
<td>LLa501</td>
<td>50</td>
<td>no break</td>
<td>2.498</td>
</tr>
<tr>
<td>LLa601</td>
<td>60</td>
<td>no break</td>
<td>2.633</td>
</tr>
<tr>
<td>LLa701</td>
<td>70</td>
<td>no break</td>
<td>2.741</td>
</tr>
</tbody>
</table>

*Table 6: Results of the Charpy impact tests*

The samples all had a good impact strength, as no break was observed at RT. It was therefore decided to make this test at -30°C. This was practically achieved by putting the specimens (prepared at room temperature) in a freezer at -30°C, at least one day before the experiments, and took them quickly out for the test.

At this low temperature, the specimens broke and the lowest impact strength recorded was 2.35 kJ/m²:

![Figure 32: The notched impact strength at -30°C as a function of the content of XLPE](image)

The notched impact strength exhibited a minimum, located before or next to 40% of XLPE. After this minimum, the strength increased slowly, almost steadily. The impact toughness of samples containing more than 40% of XLPE was thus quite independent of the XLPE-content.

3.1.2. Tensile testing at 50 mm/min

For this particular analysis, the dumbbell specimens stemming from the injection moulding were directly used, and the measurements were performed under a rate of 50 mm/min as recommended by the ISO 527.
Globally, the tensile testing curves observed are, as expected, typical of ductile plastics:

![Stress-strain curve of LLa60I](image)

*Figure 33: Stress-strain curve of LLa60I*

The slope at the beginning of the curve was quite high, revealing a **rather stiff material**. The yield point and break point were both located at around 9.4 MPa, which indicates a **relatively soft material**. The yield point was usually reached at around 19.8% of strain, and the fracture point at around 197%. This entails that the **toughness of the material was rather high**. This last observation corroborates the ones stemming from the previous notched Charpy impact test: globally, the samples displayed a high impact strength.

<table>
<thead>
<tr>
<th>Name</th>
<th>Content in XLPE (weight %)</th>
<th>Yield Stress (MPa)</th>
<th>Yield strain (%)</th>
<th>Break Stress (MPa)</th>
<th>Break Strain (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>LLa0l</td>
<td>0</td>
<td>10.38</td>
<td>16.80</td>
<td>7.80</td>
<td>382.94</td>
</tr>
<tr>
<td>LLa40l</td>
<td>40</td>
<td>9.16</td>
<td>18.31</td>
<td>9.49</td>
<td>193.16</td>
</tr>
<tr>
<td>LLa50l</td>
<td>50</td>
<td>9.23</td>
<td>20.71</td>
<td>9.83</td>
<td>148.82</td>
</tr>
<tr>
<td>LLa60l</td>
<td>60</td>
<td>9.15</td>
<td>21.45</td>
<td>9.92</td>
<td>145.28</td>
</tr>
<tr>
<td>LLa70l</td>
<td>70</td>
<td>9.01</td>
<td>21.49</td>
<td>9.88</td>
<td>114.40</td>
</tr>
</tbody>
</table>

*Table 7: Results of the Tensile Testing, at 50 mm/min*

The values of yield stress and yield strain of LLa0l were closed to the one given in the data sheet.

We are now going to investigate the effect of the content of XLPE on the yield and break point (stress at fracture).
It can be observed that, as the content of XLPE in the specimen increased, the yield stress decreased somewhat and the yield strain increased, quite sharply. We can then conclude that the more XLPE we have in the specimen, the smaller the toughness. Furthermore, the slope of the stress-strain curve decreased with an increasing content in XLPE, which means that the stiffness of the material decreased.
- Break/Fracture point

**Figure 36: The break stress as a function of the content of XLPE**

As the concentration of XLPE in the specimen increased, the break stress increased slightly and the break strain decreased sharply. Consequently, the more XLPE, the lower the toughness.

**Figure 37: The Strain at break as a function of the content of XLPE**

3.1.3. **Tensile testing at 1 mm/min: determination of the elastic modulus**

The test was performed according to the ISO 527 and using the dumbbell specimen from the injection moulding.

The average value of the elastic modulus (E-modulus) was of the order of 240 MPa, which indicated that the material was rather flexible.
In addition, the tensile modulus gradually decreased as the content of XLPE increased. Consequently, we can conclude that a higher XLPE-content leads to a lower stiffness.

This decrease followed furthermore a linear relation of the type:

\[
\text{Tensile Modulus} = -1.6927 \times \text{content XLPE (weight)} + 317.13
\]

This allows us to calculate a theoretical value of the E-modulus of pure XLPE, giving a value of 147.9 MPa.

Thus, for injection moulded samples we have:
- \( \text{E-mod(XLPE)} = 148 \text{ MPa} \)
- \( \text{E-mod(LLaDPE)} = 317 \text{ MPa} \)

### 3.1.4. Melt Flow Index (MFI)

The results obtained for each trial are given in figure 39.

**Figure 38: The tensile modulus as a function of the content of XLPE**

**Figure 39: The MFI as a function of the content in XLPE**
A dramatic drop of the MFI is noticed as the content of XLPE increased. We can consequently deduce that the more XLPE, the higher the viscosity.

### 3.1.5. Hardness

![Graph showing the hardness Shore D as a function of the content in XLPE](image)

Figure 40: The hardness Shore D as a function of the content in XLPE

The hardness seems to be independent of the content of XLPE. The average value is close to the one of LLDPE.

#### Summary of the analyses of mechanical and flow properties:

Globally, the material obtained is rather flexible and tough. When considering the impact of the content of XLPE on the properties, the following conclusions were drawn:

<table>
<thead>
<tr>
<th>Analysis</th>
<th>According to</th>
<th>Average value</th>
<th>As the content of XLPE</th>
<th>Conclusion</th>
</tr>
</thead>
<tbody>
<tr>
<td>Notched Charpy Impact Strength</td>
<td>ISO 179/1-eA</td>
<td>3,1 kJ/m²</td>
<td>Independent</td>
<td>High Impact Strength</td>
</tr>
<tr>
<td>Yield stress (TT at 50 mm/min)</td>
<td></td>
<td>9,384 MPa</td>
<td>Roughly</td>
<td></td>
</tr>
<tr>
<td>Yield strain (TT at 50 mm/min)</td>
<td></td>
<td>19,75%</td>
<td>Slight</td>
<td></td>
</tr>
<tr>
<td>E-modulus (TT at 1 mm/min)</td>
<td>ISO 527</td>
<td>242.65 MPa</td>
<td>Linear</td>
<td></td>
</tr>
<tr>
<td>Break stress (TT at 50 mm/min)</td>
<td></td>
<td>9,383 MPa</td>
<td>Slight</td>
<td></td>
</tr>
<tr>
<td>Break strain (TT at 50 mm/min)</td>
<td></td>
<td>196,92%</td>
<td>High</td>
<td></td>
</tr>
<tr>
<td>MFI</td>
<td>ISO 1133</td>
<td>12,34 g/10 min</td>
<td>Dramatic</td>
<td>Viscosity</td>
</tr>
<tr>
<td>Hardness</td>
<td>Shore D</td>
<td>48.8</td>
<td>Independent</td>
<td>Hardness of LLDPE</td>
</tr>
</tbody>
</table>

* = Significant fluctuations between the different samples

*Table 8: Summary of the conclusions concerning the effect of the content of XLPE on the material properties*
In addition, the values of the E-modulus of pure material have been estimated after injection moulding:
- E-mod(XLPE) = 148 MPa
- E-mod(LLaDPE) = 317 MPa

We are now going to have a look at the particular analyses: SEM, BTT and DSC.

3.1.6. Scanning Electron Microscopy

This analysis provides an opportunity to investigate at a microscopic level the organisation of the blend, which would help us to estimate the degree of interaction between the two materials, fundamental parameter in polymer blends.

In addition, by performing these analyses on different cross-sections along the injection-moulded sample, we may observe any traces of heterogeneity in the structure of the material.

Consequently, the following cross-sections have been chosen in the case of the sample LLa70I:

![Figure 41: Positions of the cross-sections for SEM analysis](image)

The pictures obtained are the following:

![Figures 42 a and b: SEM pictures of LLa70I, cross section 1 (close to the sprue)](image)
Figures 43 a and b: SEM pictures of LLa70l, cross-section 2 (next to sample)

Figures 44 a and b: SEM pictures of LLa70l, cross-section 3 (middle of the sample)

Figures 45 a and b: SEM pictures of LLa70l, cross-section 4 (far from the sprue)

Along the y axis of the moulding, the same type of organisation of the material was observed. This shows the homogeneity of morphology of the two blended polymers all along the injected sample, revealing the good adaptability of the process for this blending case, without pre-compounding.
The observed organisation of the two polymers is particular, and was rather unexpected. Actually, as the polymers are usually known for having low affinities and leading to immiscible blends, the phase separation was expected, but this kind of “co-continuous structure” is a surprise. We were much expecting a classical “isolated-islands” like picture resulting from a blend of two immiscible polymers, such as the case (a) and (c) in the figure 8. But here the flow field during the injection moulding may be responsible for the observed structure.

This well-distributed organisation, which looks like a delta of a river, indicates a large interfacial surface, what might be the reason for the good mechanical properties observed: the blend is immiscible but compatible.

Besides, the brighter phase does not spread continuously through the whole sample, but is hold in an area that within about 0,5 to 1 mm from the surface. This consequently leads to a schematic representation of the morphology as a “delta-type” network of the bright phase, coated by an outer layer of the darker phase.

This outer-coating is interesting, as it might be very useful in some special applications: we can easily imagine a great advantage of having an inner-reinforced polymer with an outer homogeneous layer.

The existence of this “coating” is not totally unusual in the case of injection-moulded samples. We can easily figure out that big particles introduced in this process might not be able to remain on the edges of the injected flow, so that they concentrate in the middle of the sample, letting the surfaces free.

To complete our understanding of these observations, an identification of the nature of each phase was achieved by coupling the SEM to an Energy Dispersive spectrometer (EDS) detector.

This analysis was at first made on the cross-section situated in the middle of the dumbbell specimen (cross-section 3).

![Figure 46: Position of the two analysis point for the SEM-ESD of sample LLa70I, cross-section 3](image)
Two points from each phase were chosen for performing an isolated identification, see figure above.

The first one, as shown above, was located in the dark phase, building the outer-coating. The spectrum (see figure 47) showed that only the presence of Carbon atoms is revealed, what made us conclude that the dark phase should then be LLDPE.

Figure 47: SEM-ESD spectrum of point 1 (dark phase)

On the other hand, the spectrum of point 2, in the bright phase, revealed a more intense oxygen peak, which is an evidence of the presence of siloxane bound, what made us conclude that it should be the XLPE-phase.

Figure 48: SEM-ESD Spectrum of point 2 (bright phase)

However, it is still surprising that no silicon atoms were revealed: indeed, as both oxygen and silicon atoms form the cross-links of XLPE, we should logically find some traces of them in the samples.
To straighten out this question, the same analysis was performed on samples containing different contents of XLPE, namely 0 or 100%, and the spectrum were compared.

Figure 49: SEM-ESD Spectrum of LLaDPE

Figure 50: SEM-ESD Spectrum of LLa40I

Figure 51: SEM-ESD Spectrum of XLPE
The comparison of these three spectrums reveals immediately where the problem is: it is the problem of significance of a small signal versus the highest signal (resolution of the method), such as the problem of limit of detection (in terms of signal-to-noise ratio).

Indeed, as one can observe in the XLPE spectrum, the peaks of silicon or oxygen atoms are "erased" by the huge peak of carbon, leading to some difficulties to figure them out from the noise. Due to the overweight of the carbon peak, the silicon/oxygen peaks are hard to detect, they disappear in the background.

It should furthermore be noticed that in this analysis, the intensity of the peaks is not directly proportional to the concentration of the material in the area analysed. It may give a qualitative idea of the presence of the different elements, but no quantitative conclusion can be drawn. Consequently, no further discussions are needed concerning the limit of detections and resolution of the apparatus; the SEM-ESD was not the most adapted method of identification of the elements of the samples. (Please note that some quantitative analyses may be performed by the instrument, but in the case of an organic material, very long times are required)

The two major conclusions stemming from these SEM analyses are:
- The injection moulding without pre-compounding is a suitable processing method for the blend. The samples are at a microscopic scale homogeneously organised.
- The morphology of the blend leads to a large interfacial surface and to the existence of an LLDPE skin, wrapping the sample.

### 3.1.7. Differential Scanning Calorimetry

- Conditions of analysis

All the analyses were performed under diazote conditions (to avoid the presence of O₂ and the oxidation of the samples), with in average 12 mg of material for each sample. The following programme was applied for each sample, leading to an average time of analysis of one hour:

- Hold for 1 min at 50°C
- Heat from 50°C to 170°C at 10°C/min
- Hold 1 min at 170°C
- Cool from 170°C to 50°C at 5°C/min
- Hold for 1 min at 50°C
- Heat from 50°C to 170°C at 10°C/min

The first four steps aim at getting rid of aging effects in the sample, and rejuvenate it in order to get more reliable results. Consequently, only the curve obtained from the sixth step of the program is considered in each thermogram.

The main goal of these analyses is to determine if any significant drift in the chemical composition of the samples takes place along its y axis. Indeed, by a simple look at an injection-moulded sample, one can observe the change of appearance of the blend along the dumbbell specimen and knowing that no drift in
the microscopic morphology along the y-axis was observed in the last analyses, we can logically expect that a drift of chemical composition takes place along the injection path.

Actually, during the performance of the trials, we have already noticed that the feed was not perfectly homogeneous (see paragraph Experiments - Injection moulding). In addition, it is not so unusual that an injection-moulded blend of polymer with high difference of viscosity and specific volume gives heterogeneous samples. Indeed, it can be easily imagined that as the melt is injected into the moulding at a constant rate, the flow arriving at the neck of the dumbbell specimen (part of decreased thickness) undergoes a pressure increase, which might hinder the flow of big particles, as the less viscous ones might go through easily. But the question is whether the drift is big enough to be considered as a problem.

Consequently, it was decided to analyse different parts of the samples all along the injection path, as it was done for the SEM analyses:

![Figure 52: Place of analysis for the DSC](image)

It has been furthermore decided to compare the composition along the z-axis. Indeed, as the SEM revealed the existence of a coating of almost pure LLaDPE around the sample, it would be interesting to see if the DSC analysis corroborates this observation.

These analyses were performed for two blends: LLa40I and LLa70I. In addition, the thermograms of LLaDPE and XLPE were needed as references (piece from the middle of the sample analysed).

- Results
The thermogram of the sample of LLa0I (or LLaDPE) showed one distinct peak and a second more "hidden" one. The clear endothermic peak, standing at $T = 121.5\, ^\circ C$, corresponds to the melting peak. This value is close to the one given by the producer in the data sheet, $123\, ^\circ C$, which was obtained by an ExxonMobil method. The second unclear peak reveals that the LLDPE was synthesised by using some Ziegler-Natta catalysts: since the two catalytic sites do not have the same activity, two natures of chains were obtained, leading to two melting temperatures. As this second peak is very discrete, we will consider in the following paragraph that the melting temperature of the LLaDPE is $T = 121.5\, ^\circ C$.

In the case of XLPE, one broad melting peak is observed, at $T=103.7\, ^\circ C$. 

---

**Figure 53: Thermogram of LLaDPE**

**Figure 54: Thermogram of XLPE**
Let us now compare the thermograms obtained for the different cross-section of the sample LLa70I:

![Figure 55: Thermogram of the four different points of analysis of LLa70I](image)

The curves all had the similar appearance: a first broad peak at roughly 104°C, corresponding to the melting of XLPE, followed by the thinner melting peak of LLaDPE at roughly 122°C. Only the base line was not at the same level for the different specimens, and it was consequently difficult to conclude visually if there was any major change in composition along the samples.

To analyse this, we decided to apply the following area comparison in each case:

- The area of the melting peak of XLPE was determined by selecting the two limiting points around the peak, even if the second one was not coming back to the level of the baseline.

- The area of the melting peak of LLaDPE was determined by starting from the second limiting point of the XLPE, going back to the baseline.

The ratio of these two areas was then calculated in order to have an idea of the content of XLPE.

This method was considered since the two polymers exhibit a significant difference in melting temperature and melting enthalpy ($\Delta H_{\text{XLPE}} = 84 \text{ J/g}$, $\Delta H_{\text{LLaDPE}} = 132 \text{ J/g}$).
The results obtained are:

<table>
<thead>
<tr>
<th>Cross-section number</th>
<th>Position</th>
<th>Area peak XLPE (mJ)</th>
<th>Area peak LLaDPE (mJ)</th>
<th>Ratio XLPE/LLaDPE</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>close to nozzle</td>
<td>760,484</td>
<td>86,6</td>
<td>8.782</td>
</tr>
<tr>
<td>2</td>
<td>top of specimen</td>
<td>742,077</td>
<td>117,103</td>
<td>6.337</td>
</tr>
<tr>
<td>3</td>
<td>far from nozzle</td>
<td>656,269</td>
<td>109,428</td>
<td>6.016</td>
</tr>
<tr>
<td>4</td>
<td>close to nozzle - on surface</td>
<td>361,575</td>
<td>137,275</td>
<td>2.557</td>
</tr>
<tr>
<td>4</td>
<td>close to nozzle - on surface</td>
<td>468,707</td>
<td>178,234</td>
<td>2.630</td>
</tr>
</tbody>
</table>

By comparing the first three ratios of XLPE/LLaDPE, it can be concluded that the further the cross-section from the nozzle is, the smaller the ratio is. There seems to be a real drift of the chemical composition along the moulding, which consists in a decrease of the XLPE-content along the y axis.

This decrease in concentration can be easily understood: as the XLPE constitutes the most viscous and voluminous phase of the blend, it might have some troubles to flow up to the very end part of the specimen, through the winding of the runners, the narrow gates and necks of the dumbbell.

But a more important issue is now to clarify if this drift of composition is serious enough to be considered as a source of trouble with regard to the properties of the mouldings.

If we compare the values of the ratio XLPE/LLaDPE of the cross-sections 2 and 3, which is equivalent to comparing the content of XLPE at the beginning of the
dumbbell specimen to the one at its end, we can see that the ratio diminishes only by 0.3, what represents roughly 5.2% of the variation. This seems to be a minor deviation, but it could be interesting to compare these values with the corresponding for LLa40I.

A second interesting observation results from a comparison of the average value of the ratio for the surface to the ratio for a classical cross-section: the content of XLPE is roughly three times smaller on surface. This observation correlates the previous conclusions obtained from the SEM analyses: there seems to be less XLPE in surface of the samples, supporting the existence of a coating of LLaDPE.

We now need to check these conclusions for the sample LLa40I.

As for LLa70I, each thermogram shows the expected same pattern: a first broad peak at roughly 103°C, corresponding to the melting of XLPE, followed by the thinner melting peak of LLaDPE at roughly 122°C.

By resorting to the same method as before, the area comparison analysis was performed.

![Figure 57: Thermogram of the four points of analysis of LLa40I](image-url)

<table>
<thead>
<tr>
<th>Cross-section number</th>
<th>Situation</th>
<th>Area peak XLPE (μJ)</th>
<th>Area peak LLaDPE (μJ)</th>
<th>Ratio XLPE/LLaDPE</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>close to nozzle</td>
<td>240,498</td>
<td>220,558</td>
<td>1.090</td>
</tr>
<tr>
<td>2</td>
<td>top of specimen</td>
<td>310,136</td>
<td>293,299</td>
<td>1.057</td>
</tr>
<tr>
<td>3</td>
<td>far from nozzle</td>
<td>246,349</td>
<td>288,856</td>
<td>0.853</td>
</tr>
<tr>
<td>4</td>
<td>close to nozzle - on surface</td>
<td>2,609</td>
<td>97,035</td>
<td>0.027</td>
</tr>
</tbody>
</table>

*Table 10: Area measurements on LLa40I*
First of all, the ratios obtained in that case are overall smaller than the ones obtained for LLa70I: the content of XLPE is obviously lower.

It can be observed that as the point of analysis moves further from the nozzle, the content of XLPE decreases. This is in agreement with the observations for LLa70I.

From the beginning of the dumbbell specimen to its end, the ratio diminishes by 20%. This value is much higher than in the previous case, what make us questioning the possibility of neglecting the drift in composition. However, as the LLa40I stands as the lower limit of concentration (content of XLPE studied between 40 and 70%), it seems reasonable to assume that in most of the samples, the drift of chemical composition can be neglected.

Besides, the comparison of the ratio XLPE/LLaDPE between a sample situated on surface and a classical cross-section leads to the same conclusion as before.

Consequently, the two major conclusions stemming from these DSC analyses are:

- There was a drift of the chemical composition of the sample along the y-axis. However, in most cases, the drift was only of a few percent and can be neglected.
- The existence of a skin constituted mainly of LLDPE, wrapping the sample, is corroborated by these analyses.

A last remark should be added concerning the method. The reliability of this method can be questioned as the areas calculated are not based on two limiting point situated on the base line. However, the difference in the ratio values of the two cross-section number 4 of the sample LLa70I gives already an idea of a reasonable repeatability of the method.

3.1.8. Environmental Stress Cracking Resistance: Bell Telephone Test

The samples LLa40I and LLa70I were, as before, chosen for the test. The moulded test specimen were produced at Swerea, and then sent to Borealis for testing.

Since the samples LLa40I exhibited no cracks after being in the solvent for 48h, the test was passed. However, it was failed for LLa70I.

It would be necessary to investigate further about the maximum content of XLPE at which the test is passed.

3.2. Tape extruded samples

3.2.1. Tensile testing at 250 mm/min

This particular analysis was performed following the Cable Standard IEC 60811-1-1, at a rate of 250 mm/min, with dumbbell specimen punched from the tapes. However, for convenience, the dimension of the dumbbell shape was different from the ones of the standard: instead of $L = 75 \text{ mm}$, $L' = 25 \text{ mm}$, $l = 12,5$, we used: $L = 115 \text{ mm}$ and $L' = 35 \text{ mm}$, $l = 25 \text{ mm}$.
In addition, since the specimens were directly punched from the extruded tapes, the thickness of the specimen varied between each sample. This might be a source of error: when the specimen was not strong enough (too thin), it bent when applying the extensometer and the precision of the analysis was then poorer. Particularly, in the case of LLa0E, this too low thickness did not allow at all the analysis. Unfortunately, this problem could not be eliminated after the extrusion of the tapes, since remelting and moulding the tapes would lead to a different structure of the material and to different mechanical properties. Therefore, in the following tables, no values are given for LLa0E.

Three different shapes of the stress-strain curves are observed for these samples, see Figures 58,59,60.

![Figure 58: Stress-strain curve of LLa30EPC](image)

![Figure 59: Stress-strain curve of LLb30EPC](image)

![Figure 60: Stress-strain curve of Nexans’ reference](image)
These three curves all followed the expected behaviour of ductile plastics, but the position of the yield and fracture points differed significantly.

Particularly, the overall shape of the stress-strain curve of the LLbDPE was closer to the one of Nexans’ jacket than for the “a” type, with a fracture situated at a higher stress than the yield. Besides, for Nexans’ reference material and LLbDPE, the same little fluctuations were observed close to the yield point. These first observations indicated that the “b” material would the most suitable one for this process. We however needed more investigations to be able to affirm this clearly.

It is furthermore interesting to note that the standard deviations of the values of the “a” type were much higher than the ones linked to the values of the “b” type: the overall homogeneity of the “b” type samples was better than the “a” type. This higher repeatability is another positive point for the LLbDPE.

At a same content of XLPE and conditions of processing, the LLbDPE samples showed a higher break strain and break stress than the “a” type, what reveals a higher toughness. The location of the yield points was however not dependant on the nature of LLDPE.

<table>
<thead>
<tr>
<th>Name</th>
<th>Strain at yield (%)</th>
<th>Stress at yield (MPa)</th>
<th>Break stress (MPa)</th>
<th>Break strain (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>LLa0E</td>
<td>25.16</td>
<td>5.84</td>
<td>10.72</td>
<td>885.20</td>
</tr>
<tr>
<td>LLb0E</td>
<td>13.36</td>
<td>13.22</td>
<td>11.17</td>
<td>36.72</td>
</tr>
<tr>
<td>LLa30E</td>
<td>14.84</td>
<td>10.65</td>
<td>9.13</td>
<td>19.98</td>
</tr>
<tr>
<td>LLb30E</td>
<td>23.62</td>
<td>9.04</td>
<td>11.52</td>
<td>450.17</td>
</tr>
<tr>
<td>LLb30EPC</td>
<td>25.41</td>
<td>8.94</td>
<td>12.79</td>
<td>442.43</td>
</tr>
<tr>
<td>LLb50E</td>
<td>14.71</td>
<td>8.80</td>
<td>8.41</td>
<td>27.04</td>
</tr>
<tr>
<td>LLb50EPC</td>
<td>25.28</td>
<td>9.79</td>
<td>9.28</td>
<td>90.12</td>
</tr>
<tr>
<td>LLb50E</td>
<td>26.77</td>
<td>7.80</td>
<td>11.13</td>
<td>377.43</td>
</tr>
<tr>
<td>LLb50EPC</td>
<td>25.48</td>
<td>8.40</td>
<td>11.68</td>
<td>300.67</td>
</tr>
<tr>
<td>LLa70E</td>
<td>16.06</td>
<td>4.57</td>
<td>4.37</td>
<td>23.63</td>
</tr>
<tr>
<td>LLb70E</td>
<td>25.26</td>
<td>4.33</td>
<td>4.42</td>
<td>142.27</td>
</tr>
<tr>
<td>Nexans reference</td>
<td>26.35</td>
<td>7.01</td>
<td>18.10</td>
<td>732.20</td>
</tr>
</tbody>
</table>

*Table 11: Results of the tensile testing of extruded tapes, at 250 mm/min*

- Effect of the content of XLPE on the yield and break point

Yield point
For the “a” type, it was observed that, as the content of XLPE in the specimens increased, the yield stress decreased quite sharply and the yield strain remained constant. We can then conclude that the more XLPE we have in the specimen, the lower is the slope of the stress-strain curve, namely that the lower the stiffness of the material is.

For the “b” type, as the content of XLPE in the specimen increased, the yield strain remained constant but the yield stress passed through a maximum at around 30% of XLPE before decreasing.
Consequently, we can distinguish two domains:

- For a content from 0 to 30%, **the more XLPE we have in the specimen, the higher is the slope of the stress-strain curve**, namely that **the higher the stiffness of the material is**.
- For a content of XLPE higher than 30%, **the more XLPE we have in the specimen, the lower the stiffness of the material is**.

Please note that the existence of this maximum is due to the values of yield stress and strain of the sample LLb0E, i.e. one measurement. It would consequently be wise to investigate further on the domain 0 to 30%, to obtain more data points.

**Break/Fracture point**

![Break Stress vs Content XLPE](image1)

*Figure 63: The break (fracture) stress as a function of the content of XLPE for “a” and “b” type, without pre-compounding*

![Strain at Break](image2)

*Figure 64: The Strain at break as a function of the content of XLPE for the “a” type, without pre-compounding*

![Strain at Break](image3)

*Figure 65: The Strain at break as a function of the content of XLPE for the “b” type, without pre-compounding*

For the “a” type, as the content of XLPE in the specimen increased, the break stress and strain at break decreased. Consequently, **the more XLPE we have, the weaker the toughness.**
For the “b” type, as the content of XLPE in the specimen increased, the break strain decreased sharply as the break stress passed through a maximum at around 30% before decreasing. However, the increase of break stress in the content range of 0 to 30% was much smaller than the decrease of the strain. Consequently, we can generally conclude that (as for the “a” type) **the more XLPE we have, the weaker the toughness.**

- **Effect of nature of XLPE on the yield and break points**

<table>
<thead>
<tr>
<th>Name</th>
<th>Strain at yield (%)</th>
<th>Stress at yield (MPa)</th>
<th>Stress at break (MPa)</th>
<th>Strain at break (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>LLa30E</td>
<td>13.36</td>
<td>13.22</td>
<td>11.17</td>
<td>36.72</td>
</tr>
<tr>
<td>LLb30E</td>
<td>23.62</td>
<td>9.04</td>
<td>11.52</td>
<td>450.17</td>
</tr>
<tr>
<td>LLa30EPC</td>
<td>14.84</td>
<td>10.65</td>
<td>9.13</td>
<td>19.98</td>
</tr>
<tr>
<td>LLb30EPC</td>
<td>25.41</td>
<td>8.94</td>
<td>12.79</td>
<td>442.43</td>
</tr>
<tr>
<td>LLa50E</td>
<td>14.71</td>
<td>8.80</td>
<td>8.41</td>
<td>27.04</td>
</tr>
<tr>
<td>LLb50E</td>
<td>25.77</td>
<td>7.98</td>
<td>11.13</td>
<td>377.43</td>
</tr>
<tr>
<td>LLa50EPC</td>
<td>25.28</td>
<td>9.79</td>
<td>9.26</td>
<td>90.12</td>
</tr>
<tr>
<td>LLb50EPC</td>
<td>25.48</td>
<td>8.40</td>
<td>11.58</td>
<td>300.67</td>
</tr>
<tr>
<td>LLa70E</td>
<td>15.05</td>
<td>4.57</td>
<td>4.37</td>
<td>23.63</td>
</tr>
<tr>
<td>LLb70E</td>
<td>25.26</td>
<td>4.33</td>
<td>4.42</td>
<td>142.27</td>
</tr>
</tbody>
</table>

**Table 12: Comparison of the values of tensile testing between the “a” and “b” type of LLDPE**

Yield point

At any content of XLPE and processing method (except at 50% with pre-compounding), the yield strain of the “b” type was higher than that of “a”, and its yield stress lower than the one of “a”. **Consequently, the LLaDPE samples were generally stiffer than the “b” ones.**

Break/Fracture point

At any content of XLPE and processing method, the strain at break of the “b” type was much higher than the “a”, and its break stress a little higher as well. **Consequently, the LLbDPE samples were generally much tougher than the “a” ones.**

This conclusion was corroborated by the visual following observation: during the tensile testing, the behaviours of the “a” and “b” samples after the yield point were totally different: the LLbDPE presented a better ability to harmonize the cold drawing, so that higher elongations were reached before break.

- **Effect of pre-compounding on the yield and break points**
Table 13: Comparison of the values from the tensile testing for specimens prepared with or without pre-compounding

<table>
<thead>
<tr>
<th>Name</th>
<th>Strain at yield (%)</th>
<th>Stress at yield (MPa)</th>
<th>Stress at break (MPa)</th>
<th>Strain at break (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>LLa30E</td>
<td>13.36</td>
<td>13.22</td>
<td>11.17</td>
<td>36.72</td>
</tr>
<tr>
<td>LLa30EPC</td>
<td>14.84</td>
<td>10.66</td>
<td>9.13</td>
<td>19.96</td>
</tr>
<tr>
<td>LLb30E</td>
<td>23.62</td>
<td>9.04</td>
<td>11.52</td>
<td>460.17</td>
</tr>
<tr>
<td>LLb30EPC</td>
<td>25.41</td>
<td>8.24</td>
<td>12.79</td>
<td>442.43</td>
</tr>
<tr>
<td>LLa50E</td>
<td>14.71</td>
<td>8.80</td>
<td>8.41</td>
<td>27.04</td>
</tr>
<tr>
<td>LLa50EPC</td>
<td>25.28</td>
<td>9.79</td>
<td>9.28</td>
<td>90.12</td>
</tr>
<tr>
<td>LLb50E</td>
<td>25.77</td>
<td>7.89</td>
<td>11.13</td>
<td>377.43</td>
</tr>
<tr>
<td>LLb50EPC</td>
<td>25.49</td>
<td>8.40</td>
<td>11.58</td>
<td>300.67</td>
</tr>
</tbody>
</table>

Yield point

Generally speaking, the values of strain and stress at yield were not that much affected by the pre-compounding. No real trend/influence could be detected.

Break/Fracture point

The values of break stress and strain with or without pre-compounding were not as close as the previous ones, but no real trend could be observed.

3.2.2. Tensile testing at 1 mm/min: determination of the E-modulus

The test was performed according to the standard IEC 60811-1-1 and with special dumbbell specimens punched from the tapes.

Table 14: Results of the tensile testing of extruded tapes, at 250 mm/min

<table>
<thead>
<tr>
<th>Name</th>
<th>Tensile modulus - ISO 527 (MPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>LLa0E</td>
<td>too thin samples</td>
</tr>
<tr>
<td>LLb0E</td>
<td>219.49</td>
</tr>
<tr>
<td>LLa30E</td>
<td>229.37</td>
</tr>
<tr>
<td>LLa30EPC</td>
<td>242.39</td>
</tr>
<tr>
<td>LLb30E</td>
<td>203.39</td>
</tr>
<tr>
<td>LLb30EPC</td>
<td>157.23</td>
</tr>
<tr>
<td>LLa50E</td>
<td>161.76</td>
</tr>
<tr>
<td>LLa50EPC</td>
<td>188.96</td>
</tr>
<tr>
<td>LLb50E</td>
<td>161.24</td>
</tr>
<tr>
<td>LLb50EPC</td>
<td>145.81</td>
</tr>
<tr>
<td>LLa70E</td>
<td>165.59</td>
</tr>
<tr>
<td>LLb70E</td>
<td>145.97</td>
</tr>
<tr>
<td>Nexans reference</td>
<td>112.76</td>
</tr>
</tbody>
</table>

- Effect of the content of XLPE
Figure 66: The tensile modulus as a function of the content of XLPE for the “a” and “b” extruded samples, without pre-compounding.

For both “a” and “b” types, the tensile modulus was gradually decreasing as the content of XLPE in the material increased. Consequently, we can conclude that the more the material contains of XLPE, the lower the stiffness is.

Please note that for the “a” type, this observation corroborates the one made at the tensile testing at 250mm/min. For the “b” type, we are not able to justify the previous observation (existence of two domains), because of the absence of the point at 0%, however, the same trend is observed for a XLPE content higher than 30%.

As for the injection-moulded samples, it could be interesting to figure out a linear relation of the E-modulus as a function of the content of XLPE.

Figure 67: Linearisation of the tensile modulus as a function of the content of XLPE for the “b” type.
The equation of the linear relation obtained is:

\[ \text{Tensile modulus} = -1.1371 \times \text{content XLPE (%weight)} + 226.66 \]

This allows us to calculate a theoretical value of the E-modulus of pure XLPE, giving 113 MPa.

This point, at 100%, can now be used as a fourth point in the curve representation of the “a” samples, which had fewer available data.

![Figure 68: Linearization of the tensile modulus as a function of the content of XLPE for the “a” type](image)

An approximate value of the E-modulus of pure LLaDPE of 260 MPa was then obtained.

**From now on, we can consequently consider that for the extruded samples:**

- \( E-\text{mod}(\text{XLPE}) = 113 \text{ MPa} \)
- \( E-\text{mod}(\text{LLbDPE}) = 220 \text{ MPa} \)
- \( E-\text{mod}(\text{LLaDPE}) = 260 \text{ MPa} \)

These values are all lower than the one calculated for injection moulded samples, as expected.

- Effect of the nature of XLPE
Table 15: Comparison of the values of tensile modulus between the “a” and “b” type of LLDPE

At any content of XLPE and processing method, the tensile modulus of the “b” type was somewhat lower than that of “a”. Consequently, the LLaDPE samples were generally stiffer than the “b” ones. This corroborates the conclusion from the previous tensile testing.

- Effect of the pre-compounding

Table 16: Comparison of the tensile modulus with or without pre-compounding

For the “a” type, it seems that the pre-compounding somewhat increased the E-modulus. The opposite conclusion can be drawn concerning the “b” type. Consequently, we can conclude that a pre-compounding will enhance somewhat the stiffness of the “a” samples but has a negative influence on the stiffness of the “b” samples.

General conclusions from the tensile testing

The main conclusions stemming from the tensile testing are:
- For “a” samples and “b” samples containing more than 30% of XLPE, the higher the content of XLPE, the lower the stiffness and roughness. For the “b” samples, at contents lower than 30%, the trend might be the opposite (to be checked up).
- Some experimental and calculated values of the three considered materials were obtained: E-mod(XLPE) = 113 MPa; E-mod(LLbDPE) = 220 MPa; E-
mod(LLaDPE) = 260 MPa. Therefore, the “a” samples were generally stiffer but less tough than the “b” ones for the same content of XLPE and processing method.

- The pre-compounding gave a slightly positive effect on the stiffness of the “a” samples, and a somewhat negative one for the “b” samples.
- The overall mechanical behaviour of “b” samples in tension was closer to the one of the reference (Nexans’ material), and it had a better repeatability.

Please note the uncertainty concerning the effect of the content of XLPE on “b” samples containing less than 30% of XLPE. In a few cases, the optimal conditions of analysis were not obtained as the thickness of the tapes sometimes did not offer enough stiffness to avoid a bending of the specimen.

### 3.2.3. Hardness

The determination of the hardness was performed at the same conditions as described in the paragraph “Analyses – Hardness”.

- **Effect of the content of XLPE**

![Graph showing the hardness of extruded “a” and “b” samples as a function of the content of XLPE, without pre-compounding.](image)

*Figure 69: The hardness of the extruded “a” and “b” samples as a function of the content of XLPE, without pre-compounding*

In comparison to the injection-moulding trials, the extruded samples exhibited a dependence on the content of XLPE. **For both “a” and “b” types, the hardness decreased as the content in XLPE increased.**

It seems consequently as if the blend had not a skin of LLDPE on surface.

- **Effect of the nature of XLPE**
Table 17: Hardness of the extruded “a” and “b” samples

Since the hardness of LLaDPE (62) is higher than the one of LLbDPE (51), it is not surprising to observe that the “a” samples present a higher hardness than the “b” samples.

- Effect of the pre-compounding

Table 18: Hardness of the extruded “a” and “b” samples with or without pre-compounding

The pre-compounding had no significant influence on the hardness of the tape extruded samples.

3.2.4. Melt Flow Index

The determination of the Melt Flow Index was performed at the same conditions as described in the paragraph “Analyses – MFI”.

- Effect of the content of XLPE / Effect of the nature of XLPE

Table 19: MFI of the extruded “a” and “b” samples without pre-compounding
As for the injected moulded samples, the “a” extruded samples showed a dramatic drop in MFI as the content of XLPE increased. So, it can be concluded one more time that the more XLPE, the more viscous the sample were.

This conclusion fits also to the “b” samples. Of course, as the LLbDPE had a lower MFI than LLaDPE, the MFI values of the “b” samples were always lower than the ones of the “a” samples. However, even though the “starting points” between the two types of LLDPE were different, the rate of decrease was very different between the “b” and “a” samples: the MFI of “a” samples decreased twice as fast as the “b” ones.

This last observation is of interest with regard to the processing: the “a” type might show some large rheological differences between two trials, as soon as the content of XLPE varies. Used at a production scale, this might then be tricky to handle, needing a lot of flexibility and adaption of the process. We can easily imagine the problems of production if, between two loads varying a little in their XLPE content, the temperature and pressure conditions have to be reset.

This important difference between “a” and “b”, in addition to the previous observations and conclusions, make us conclude which material is the most adapted for tape-extruding our blend. It was decided that the “b” material was the most suitable LLDPE for extrusion of the blend.

- Effect of pre-compounding
The pre-compounding had no influence on the MFI of the tape extruded samples.

### 3.2.5. Environmental Stress Cracking Resistance: Bell Telephone Test

As for the two injection-moulded samples before, the Bell Telephone Test moulded test sheets were manufactured at Swerea, and then sent to Borealis for the testing. The test is performed at the conditions described in the paragraph “Analyses – BTT”.

The samples LLb30EPC and LLb30E were chosen for this analysis. Indeed, thanks to the previous observations during processing and analysis, the “b” type has turned out to be the most interesting kind of LLDPE. A content of 30% represents as well a reasonable content for future applications.

Since the samples did not exhibit any cracks after being in the solvent for 48h, the test was passed.

**The tape extruded material had consequently a good cracking resistance with 30% of XLPE, with or without pre-compounding.**

The resistance of the material to Environmental Stress Cracking was at least as good as the one of the reference (given by Nexans), so that we can reasonably consider using the extruded material as an inner-jacket of power cables.

#### General conclusions

Thanks to these analyses, we have determined the main effects of the investigated parameters:

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Stiffness</th>
<th>Toughness</th>
<th>Hardness</th>
<th>Viscosity</th>
</tr>
</thead>
<tbody>
<tr>
<td>As the content of XLPE</td>
<td>for “a” and “b”,</td>
<td>for “a” and “b”,</td>
<td>for “a” and “b”,</td>
<td>for “a” samples,</td>
</tr>
<tr>
<td></td>
<td>*</td>
<td>*</td>
<td>*</td>
<td>high</td>
</tr>
<tr>
<td>Between “a” and “b” type</td>
<td>“a” stiffer than “b”</td>
<td>“b” tougher than “a”</td>
<td>“a” harder than “b”</td>
<td>“b” more viscous than “a” but easier to process</td>
</tr>
<tr>
<td>Effect of Pre-compounding</td>
<td>lightly positive for “a”</td>
<td>no influence</td>
<td>no influence</td>
<td>no influence</td>
</tr>
<tr>
<td></td>
<td>lightly negative for “b”</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

* = there might be an optimum of stiffness for “b” samples at around 30% of XLPE

**Table 21: General conclusions for tape-extruded samples**
Furthermore, some estimated values of the three considered materials were obtained:

- $E_{\text{mod}}(\text{XLPE}) = 113 \text{ MPa}$
- $E_{\text{mod}}(\text{LLbDPE}) = 220 \text{ MPa}$
- $E_{\text{mod}}(\text{LLaDPE}) = 260 \text{ MPa}$.

By taking into consideration the observations made during the processing, the following main conclusions can be drawn:

- The material can be processed for a content ranging between 0 to 70%-weight of XLPE. However, the higher XLPE content, the poorer the mechanical properties.
- After considering the mechanical properties and the processing aspects, LLbDPE should be chosen for extrusion of the blend.
- The conclusions concerning the last parameter, the need or not of a pre-compounding, are not so straightforward. Since the pre-compounding adds a costly step in the recycling process and does not have a positive influence on the properties, it should rather be avoided. However, if the equipment available does not include a two-feeder extruder, its achievement is obligatory. Consequently, the necessity of pre-compounding relies mainly in the processing equipment.

### 3.2.6. Comparison with the reference material

The standard material given by the cable producer Nexans allow us to evaluate the quality of the different blends, their possible use in the cable industry and figure out an optimal formulation.

<table>
<thead>
<tr>
<th>Name</th>
<th>Strain at yield (%)</th>
<th>Stress at Yield (MPa)</th>
<th>Stress at Break (MPa)</th>
<th>Strain at Break (%)</th>
<th>Tensile Modulus - ISO 527 (Mpa)</th>
<th>MFI (g/10min)</th>
<th>Hardness Shore D</th>
</tr>
</thead>
<tbody>
<tr>
<td>LLa0E</td>
<td>N/A</td>
<td>10.715</td>
<td>885.196</td>
<td>231.491</td>
<td>2.650</td>
<td>61</td>
<td>N/A</td>
</tr>
<tr>
<td>LLb0E</td>
<td>25.158</td>
<td>5.644</td>
<td>10.715</td>
<td>885.196</td>
<td>231.491</td>
<td>2.650</td>
<td>51</td>
</tr>
<tr>
<td>LLb30E</td>
<td>23.615</td>
<td>9.044</td>
<td>11.524</td>
<td>450.171</td>
<td>209.388</td>
<td>1.309</td>
<td>49</td>
</tr>
<tr>
<td>LLb50EPC</td>
<td>25.412</td>
<td>8.938</td>
<td>12.790</td>
<td>442.431</td>
<td>157.234</td>
<td>1.227</td>
<td>47</td>
</tr>
<tr>
<td>LLa50E</td>
<td>14.707</td>
<td>8.796</td>
<td>8.407</td>
<td>27.041</td>
<td>161.760</td>
<td>3.663</td>
<td>48</td>
</tr>
<tr>
<td>LLa50EPC</td>
<td>25.284</td>
<td>9.785</td>
<td>9.280</td>
<td>90.123</td>
<td>188.962</td>
<td>3.509</td>
<td>52</td>
</tr>
<tr>
<td>LLb50E</td>
<td>25.747</td>
<td>7.983</td>
<td>11.127</td>
<td>377.431</td>
<td>161.241</td>
<td>0.477</td>
<td>45</td>
</tr>
<tr>
<td>LLa70E</td>
<td>15.063</td>
<td>4.570</td>
<td>3.373</td>
<td>23.629</td>
<td>165.586</td>
<td>0.370</td>
<td>44</td>
</tr>
<tr>
<td>LLb70E</td>
<td>25.257</td>
<td>4.332</td>
<td>4.421</td>
<td>142.274</td>
<td>145.973</td>
<td>0.045</td>
<td>38</td>
</tr>
<tr>
<td>Nexans reference</td>
<td>26.352</td>
<td>7.013</td>
<td>18.057</td>
<td>732.159</td>
<td>112.763</td>
<td>0.440</td>
<td>44</td>
</tr>
<tr>
<td>Average</td>
<td>21.27</td>
<td>8.32</td>
<td>9.50</td>
<td>254.15</td>
<td>184.29</td>
<td>7.69</td>
<td>48.83</td>
</tr>
<tr>
<td>Min</td>
<td>13.35</td>
<td>4.33</td>
<td>4.37</td>
<td>19.93</td>
<td>145.81</td>
<td>0.05</td>
<td>38.00</td>
</tr>
<tr>
<td>Max</td>
<td>25.77</td>
<td>13.22</td>
<td>12.79</td>
<td>865.20</td>
<td>242.39</td>
<td>42.54</td>
<td>62.00</td>
</tr>
<tr>
<td>Standard deviation</td>
<td>5.42</td>
<td>2.63</td>
<td>2.82</td>
<td>271.24</td>
<td>35.17</td>
<td>12.27</td>
<td>5.77</td>
</tr>
</tbody>
</table>

*Table 22: Overall results of analysis of the extruded samples for comparison with the reference material*

The different properties, bordered by the minimum and maximum values, contain in most of the case the value of the reference. This indicates that the area of study was already well chosen, with the exception of the stress at break (too low) and tensile modulus (too high).
In comparison with the different blends, the reference has a high value of strain at yield, stress at break and strain at break, but a rather low E-modulus, MFI and hardness. So, it is generally less hard and stiff than the average material studied, but rather viscous and tough.

If we then consider that the most important properties of a cable relate to its rather low stiffness and high toughness, the best suitable formulation is the LLb30EPC. This gives us already an indication of the range of content we should work with when considering using the material for cable applications.

The overall mechanical properties and BTT tests finally allow us to consider some future production-scale trials, where the material should be reused as an inner-jacket of power cables.

### 3.3. Extruded wires

#### 3.3.1. Tensile testing at 250 mm/min and 50 mm/min

The main goal of this tensile testing is to determine if the mechanical requirements for this application are fulfilled by the recycled material. Since these requirements only concern the fracture point (tensile strength and ultimate elongation), only the tensile tests at 50 and 250 mm/min were performed according to, respectively, the ISO 57 and the IEC 6081-1-1.

A part of these requirements also concern the properties of the material after ageing, so an ageing of 7 days at 100°C was performed according to the IEC 811-1-2 (samples of 10 cm length hanged in an oven, without the conductive part).

In order to save time, only 5 out of the 14 extruded samples were analysed. However, they are enough to allow us to conclude about the effect of three of the four parameters, the effect of the dilution being considered at first time as negligible.

<table>
<thead>
<tr>
<th>Name</th>
<th>% RLPE-Φ-0.4</th>
<th>% RLPE-Φ-0.6</th>
<th>Additives</th>
<th>End mix</th>
<th>Unaged Break Stress (Mpa)</th>
<th>Unaged Strain at break (%)</th>
<th>Aged Break Stress (Mpa)</th>
<th>Aged Strain at break (%)</th>
<th>Aged Break Stress (Mpa)</th>
<th>Aged Strain at break (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>LLb550,4KED</td>
<td>35</td>
<td>0</td>
<td>No</td>
<td>Yes with KE 30</td>
<td>17</td>
<td>555.2</td>
<td>16.6</td>
<td>563.9</td>
<td>16.2</td>
<td>546</td>
</tr>
<tr>
<td>LLb550,8KED</td>
<td>0</td>
<td>5</td>
<td>No</td>
<td>Yes with KE 30</td>
<td>18.1*</td>
<td>610.8*</td>
<td>17.6</td>
<td>629</td>
<td>14.8</td>
<td>628</td>
</tr>
<tr>
<td>LLd550,0,6KED</td>
<td>0</td>
<td>15</td>
<td>No</td>
<td>Yes with KE 30</td>
<td>16.1</td>
<td>582.2</td>
<td>16.3</td>
<td>542</td>
<td>16.3</td>
<td>531</td>
</tr>
<tr>
<td>LLd550,0,6KEA</td>
<td>0</td>
<td>15</td>
<td>Yes</td>
<td>Yes with KE 30</td>
<td>17</td>
<td>545.3</td>
<td>16.5</td>
<td>532.2</td>
<td>16.1</td>
<td>522</td>
</tr>
<tr>
<td>LLb350,0,6KE</td>
<td>0</td>
<td>30</td>
<td>No</td>
<td>No</td>
<td>16.2</td>
<td>460.7</td>
<td>15.16</td>
<td>407.1</td>
<td>15.8</td>
<td>451</td>
</tr>
</tbody>
</table>

* Two samples did not break before the maximal elongation

Table 23: Summary of results of Tensile Testing of extruded wires, aged and unaged, at 50 and 250 mm/min

Generally speaking, the global values obtained at 250 mm/min, were, as expected, a bit lower than the ones of 50 mm/min, such as the values obtain after ageing are lower than the one unaged.

Furthermore, if we compare the unaged values of stress and strain at break of the extruded tapes at 250 mm/min to the ones of the wires, we can observe that the
strain at break were in the same range, but the global stress at break were much higher in the case of the wires. For example, in the case of a content in XLPE of 30%, the strain at break remained at a similar value (450 MPa), as the break stress increased from 11.5 to 16 MPa. Consequently, the processing conditions of extrusion seem to have an impact on the break stress: in the case of the wire extrusion, the break stress is significantly higher.

- **Effect of the content of XLPE**

By comparing the results of LLb5(0,6)KED, LLb15(0,6)KED, LLb30(0,6)KED, unaged or aged, at any rate of analysis, it was observed that the break stress and strain decreased as the content in XLPE increased: the more XLPE, the weaker the toughness. This conclusion corroborates the previous observations made on Extruded Tapes. Please note that this unveils the strange value of the unaged break stress of LLb15(0,6)KED at 50 mm/min, which is lower than is should be, and will be from now on considered as unreliable (marked in italics in the table).

- **Effect of the size of the XLPE particles**

By comparing the values of LLb15(0,4)KED and LLb15(0,6)KED, excepted the one of the unaged break stress of LLb15(0,6)KED at 50 mm/min as explained before, it was observed that the values of break stress and strain at any ageing level and rate of analysis were very close. Their differences were not higher than 30% in the case of strains, what can be considered as part of the experimental errors when compared to the average values, and neglected. **Consequently, the size of the XLPE particles showed no significant impact on the break point.**

- **Effect of additives**

One more time, by comparing the values of LLb15(0,6)KED and LLb15(0,6)KEAD, (excepted the one of the unaged break stress of LLb15(0,6)KED at 50 mm/min), it was observed that the values of break stress and strain at any ageing level and rate of analysis were very close. Their differences were not higher than 20% in the case of strains, what could be considered as part of the experimental errors when compared to the average values, and neglected. **Consequently, the presence of additives showed no significant impact on the break point.**

- **Fulfilment of the requirements**

The mechanical requirements upon standard low voltage sheath are the following:

- A tensile strength of at least 10 N/mm² (or 10 MPa)
- A minimum of 300% of elongation at break before and after ageing.

As it can be observed, the 5 tested samples fulfil all the requirements. **Consequently, a recycled material with a content of 5 to 30 wt-% of XLPE showed adequate mechanical properties for this application.** Let us see now if their cracking resistance is satisfying.
3.3.2. Bell Telephone Test

The test was performed at the same conditions as described before with the moulded test sheets manufactured at Swerea, and sent to Boréalis.

The samples chosen for this analysis were the ones studied in the tensile testing plus some additional 4 ones:

- LLb5(0,4)KED
- LLb5(0,4)KEAD
- LLb15(0,4)KED
- LLb15(0,6)KED
- LLb30(0,4)KED
- LLb30(0,6)KED
- LLb5(0,6)KEAD
- LLb15(0,6)KEAD
- LLb0KE

Indeed, these 9 samples allow us to study the effect of the size of the particles, of the presence of additives and of the content of XLPE on the Stress Cracking Resistance. The content range has been restricted to 0 to 30%, as that seemed reasonable for this application.

Since the samples did not exhibit any crack after being in the solvent for 48h, the test was passed for all of them.

The extruded wires had consequently a good cracking resistance, within the range of XLPE content studied, independently of the presence or not of additives or of the size of the particles used. This observation enhanced the possibility to use the recycled material for future cable wires.

3.3.3. Surface roughness 3D-Analysis

This digitization was performed on 8 samples, and the calculation of Ra, Ry and Rz was achieved for all of them.

The 3D-pictures obtained were very self-explanatory and we could already conclude about the impact of some parameters, such as the content in XLPE.

![Figure 71: Deviation colour plots of LLb15(0,6)KED, for the area 1 (left) and 2 (right)]
However, for the two other parameters, the differences were not so striking, so that calculations were requested to determine/quantify their impacts.

Table 24: Ra, Ry, Rz calculations on extruded wires

As expected, the values of Ry and Rz were close, and roughly ten times higher than the Ra. However, these three ways of evaluating the roughness (Ra, Ry and Rz) all show the same response to the different parameters, what made us finding out that the value of Ra of LLb15(0,6)KEAD (marked in italics in the table) is much lower than it should be and that it is unreliable: it will not be taken into consideration for the following analysis of the results.

Firstly, it can be noticed that the roughness increased significantly with the content in XLPE: it is hardly doubled when the content is multiplied by three, what is not a surprise. The visual observations during the extrusion have led us to conclude that the roughness was acceptable for a content lower than 30%. Thanks to this quantification of the roughness, we can now affirm that this is equivalent to saying that the material can be considered for this application for a Ra < 0,044 mm.
By comparing the values of LLb5(0,6)KED and LLb5(0,6)KEAD, it can be noticed that the presence of additives does not impact significantly the roughness.

As well, by comparing the values of LLb15(0,6)KED/LLb15(0,4)KED or LLb30(0,6)KE/LLb50(0,4)KE, it can be noticed that the size of the XLPE powder does not impact significantly the roughness. Please note that the size of the powder does not show any positive influence on the properties, so that it can be concluded that it is not worth spending time and money on reducing the particles from 0,6 to 0,4 mm.

These two observations do not stand as a particularly positive result for the project. Indeed, during the processing, it has already been noticed that even the modification of some manufacturing parameters had no significant effect on the roughness, so that we were hoping for finding other parameters that could reduce it. Unfortunately, it turns out that only the content of XLPE of the material has a significant effect on the roughness of the extruded samples.

However, this method of analysis unveils a lot of possibilities for future studies. It can be easily imagined that by analysing some standard materials, some requirements upon the roughness could be fixed and a rigorous scientific comparison between materials achieved. Even a standardization of the method could be considered, which would allow a quantification of the feeling of roughness, upon further investigations and improvement on the repeatability, reliability and degree of precision of the analysis.

### 3.4. Extruded inner-jacket

#### 3.4.1. Tensile testing at 250 mm/min and 50 mm/min

One more time, the tensile testing were performed at two different speeds, according to their respective Standards. The smaller dumbbell specimen of the standard, which is 50mm long, was used. Indeed, the surface of the samples only consists of the unwrapped inner-jacket, what is a rather narrow piece of sample.

Practically, for the analyses, the outer-jacket was removed and the inner-jacket separated from the other constituents of the cable. The aging was however performed on the whole cable, at 100°C during 240h (10 days), according to the IEC 811-1-2.

<table>
<thead>
<tr>
<th>Name</th>
<th>Weight content in XLPE (%)</th>
<th>Lubricant</th>
<th>Unaged Break Stress (Mpa)</th>
<th>Unaged Strain at break (%)</th>
<th>Aged Break Stress (Mpa)</th>
<th>Aged Strain at break (%)</th>
<th>Unaged Break Stress (Mpa)</th>
<th>Unaged Strain at break (%)</th>
<th>Aged Break Stress (Mpa)</th>
<th>Aged Strain at break (%)</th>
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</thead>
<tbody>
<tr>
<td>LLb10(0,6)</td>
<td>10</td>
<td>No</td>
<td>16.1</td>
<td>532</td>
<td>13.3</td>
<td>751</td>
<td>13.7</td>
<td>490</td>
<td>12.8</td>
<td>644</td>
</tr>
<tr>
<td>LLb15(0,6)</td>
<td>15</td>
<td>Yes</td>
<td>14.2</td>
<td>510</td>
<td>11.7</td>
<td>655</td>
<td>12.7</td>
<td>461</td>
<td>10.4</td>
<td>575</td>
</tr>
<tr>
<td>LLb20(0,6)</td>
<td>20</td>
<td>No</td>
<td>15.0</td>
<td>476</td>
<td>12</td>
<td>582</td>
<td>14.4</td>
<td>488</td>
<td>11.1</td>
<td>563</td>
</tr>
</tbody>
</table>

*Table 25: Tensile testing results for the inner-jacket at 250 and 50mm/min*

The results obtained at 250mm/min are a little lower than the one obtained at 50 mm/min. However, it can be noticed that the unaged values of strain at break are
smaller than the aged values, whatever the speed of analysis. This observation is a
real surprise as the influence of ageing observed on the extruded wires was the
opposite. Bearing in mind that the ageing of these power cables is performed on the
whole cable, and knowing its complex construction in comparison to the last wires, it
can be guessed that this strange behaviour under ageing is not due to the material
itself, but rather to an interaction with the constituents of the cable. The most
probable origin of this phenomenon is the migration of plasticizers from the PVC of
the outer-jacket to the inner-jacket.

Furthermore, the values of stress at break presented a rather unusual evolution as a
function of the content in XLPE: the values of LLb15(0,6)JEAD were lower than the
ones of the two other samples. The decrease was actually not so pronounced
between the point at 15% and the one at 10 or 20%, so that it could hardly be
interpreted as a real minimum and it should be rather characterized as fluctuations,
but it was different from the classical evolutions observed in all the previous trials, in
which the break stress was chiefly decreasing as the content in XLPE was
increasing. Considering that the effect of dilution of the material can be neglected,
these fluctuations might come from the presence of lubricants. Indeed, the second
sample was the only one which contains additives, so that it might be the reason from
this little decrease. As the number of samples was rather limited, there was no way to
corroborate this trend, so that we could only emit the possibility that lubricants
might have a light negative impact on the break stress.

- Fulfilment of the requirements

But the main goal was to conclude about the possibility of using this recycled material
as a substitute for the present copolymer.
For this, the recycled material has to fulfil the following mechanical requirements:
- An elongation at break higher than 300% before and after aging.
- A tensile strength (break stress) higher than 10 MPa before aging.

As it can be seen in the previous table, the material fulfils all these
requirements. Consequently, from a mechanical point of view, the material stands
as a possible substitute of the present copolymer. However, to be concretely
considered, some further investigations on other aspects have to be done: economic
calculations, acceptability of the rough aspect by the customer...
Conclusions

The investigations have revealed the technical feasibility of a relative straightforward primary recycling of lumps of Si-XLPE. This mechanical recovery is based on a blending of the ground lumps with LLDPE.

The first step consists of the granulation and grinding of the lumps, in order to obtain a powder which can be easily and homogeneously mixed with pellets of LLDPE. The analyses revealed that the size of the powder did not have a significant influence on the mechanical properties, so that a grinding into a 0.6mm diameter powder was sufficient.

The powder could then be blended with LLDPE by either injection-moulding or extrusion. The processability was validated in both cases for contents in XLPE up to 70wt-%. The blends showed encouraging mechanical properties, mainly dependent on the content in XLPE and on the nature of LLDPE. It was observed that the higher the amount of XLPE was, the poorer the mechanical properties (stiffness, toughness, hardness) were. However, in the case of injection-moulding, the hardness was steadily at a high value, since the samples had a particular morphology (LLDPE skin). The choice of the virgin LLDPE was an important parameter: a resin with high molecular weight should be preferred for extrusion (such as the “b” type), as a rather low molecular weight LLDPE (such as the “a” type) was more suitable to injection-moulding. In the case of extrusion, a pre-compounding was necessary only if a two-feeder extruder was not available for the blend.

The recycled material was considered for two applications within the cable industry: insulator of wires and inner-jacket of a Norwegian power cable. For both cases, the tube tooling extrusion done at a production scale worked out successfully. The only issue was the roughness of the samples, which limited the maximum amount of XLPE accepted. Nevertheless, the mechanical properties were very satisfying: the specimens fulfilled the mechanical requirements upon tensile strength and elongation at break before and after ageing.

These encouraging results should lead to further investigations. As sustainability takes into account not only the technical feasibility but also the economical and social interest, this proposed recycling solution should be considered on an economic and marketing point of view by interested companies. The studies were furthermore restricted to an intern recycling, with the hope of obtaining a possible in-house solution, but applications in other industries should be envisaged. It would be particularly interesting to take advantage of the surface roughness of extruded samples, so that we would not be limited in the content of XLPE. Finally, as peroxide cross-linked polyethylene is also used in the cable industry, it could be interesting to study the possibility of adapting this method to lumps of Pe-XLPE.
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[12] V. Massardier, Trends in Recycling of Polymer Materials, IMP UMR CNRS #5223


[21] Lertrojanachusit N., Nunthatonni C., Pongprayoon T., Wannatong L., *Recycling of Silane-Crosslinked Polyethylene by the extruder: effect of active and non-active additives*, King Mingkut’s Institute of Technology North Bangkok, Thailand


<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Definition</th>
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<tr>
<td>ASTM</td>
<td>American Society for Testing and Materials</td>
</tr>
<tr>
<td>BTT</td>
<td>Bell Telephone Test</td>
</tr>
<tr>
<td>CCD</td>
<td>Charge-Coupled Device</td>
</tr>
<tr>
<td>DSC</td>
<td>Differential Scanning Calorimetry</td>
</tr>
<tr>
<td>EDS</td>
<td>Energy Dispersive Spectrometer</td>
</tr>
<tr>
<td>EOL</td>
<td>End Of Life</td>
</tr>
<tr>
<td>EPDM</td>
<td>Ethylene Propylene Diene Monomer</td>
</tr>
<tr>
<td>HDPE</td>
<td>High density Polyethylene</td>
</tr>
<tr>
<td>HFFR</td>
<td>Halogen-Free Flame Retardant</td>
</tr>
<tr>
<td>IEC</td>
<td>International Electrotechnical Commission</td>
</tr>
<tr>
<td>ISO</td>
<td>International Organisation for Standardisation</td>
</tr>
<tr>
<td>JIS</td>
<td>Japanese Industrial Standards</td>
</tr>
<tr>
<td>LDPE</td>
<td>Low Density Polyethylene</td>
</tr>
<tr>
<td>LLDPE</td>
<td>Linear Low Density Polyethylene</td>
</tr>
<tr>
<td>MFI</td>
<td>Melt Flow Index</td>
</tr>
<tr>
<td>PE</td>
<td>Polyethylene</td>
</tr>
<tr>
<td>Pe-XLPE</td>
<td>Peroxide cross-linked Polyethylene</td>
</tr>
<tr>
<td>PP</td>
<td>Polypropylene</td>
</tr>
<tr>
<td>PVA</td>
<td>Polyvinyl Acetate</td>
</tr>
<tr>
<td>PVC</td>
<td>Polyvinylchloride</td>
</tr>
<tr>
<td>RT</td>
<td>Room Temperature</td>
</tr>
<tr>
<td>SEK</td>
<td>Svenska Elektriska Kommissionen</td>
</tr>
<tr>
<td>SEM</td>
<td>Scanning Electron Microscopy</td>
</tr>
<tr>
<td>Si-XLPE</td>
<td>Silane cross-linked Polyethylene</td>
</tr>
<tr>
<td>XLPE, PEX</td>
<td>Cross-linked Polyethylene</td>
</tr>
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Appendix 2: Average conditions of processing of the Injection-Moulding Samples
Appendix 2: Average conditions of processing of the Tape extruded samples
Appendix 3: Average conditions of processing of the Extrusion of wires at Draka Kabel
Appendix 4: Average conditions of processing for the Extrusion of inner-jacket at Nexans
Appendix 1: References of the apparatus

For Processing

- Injection Moulding Machine

  Manufacturer's Name: Engel
  Type: ES 200/110 HL-V
  Number: 44557
  Year of construction: 2001

- Single screw extruder

  Brabender Compact extruder
  Type: 837415
  Nr: 013062
  Screw L/d=25 mm, diam=19mm

- Roll

  Brabender Univex - Take off

- Twin Screw Extruder

  Werner & Pfleiderer (Germany)
  Type: ZSK 30 M912

  Feeder 1:
  kTron (Switzerland)
  Type: T-22

  Feeder 2:
  Iceva AB “Colorfeed 500” (Sweden)

- Chopper

  Dreher (Germany)
  Type: 5G 10 Ni
  Number: G8369
- Dryer
Piovan Technologies
Soc: S Mana di Sala (Italy)
Typ: CR 50
Nbr: IS 152 3222

For the wire extrusion at Draka Kabel, equipment of the process production EP 90:

- Extruder: Maillefer (Lausanne), type BM 45-20D, No 835106A, with screen pack constituted by two meshes of 28.4 mm, mesh number 40.
- Thickness measurer: Zumbach ODAC 75XY
- Puller: Rosendahl
- Buffer: Skatlek SC-3
- Reel-up: Skatlek U12P4K

For the inner-jacket extrusion, at Nexans, production Equipment n. 23

- Extruder:
  Brand Maillefer (Lausanne), type BM 150-18D, No 64-443
  Single feeder and single screw extruder
  Screw Barrier (channel for solid and melt), D/L = 18
  Mesh: Nr 30 / Nr 60 / Nr 30, diameter = 154 mm

- Traction: speed of 85% (19 rpm) during extrusion

For Analysis

- Notched Charpy impact
  Zwick (Germany)
  Type: 5110.100/00
  Number: 112029 019-03/90

- Tensile Testing machine
  MTS Systems (France)
  Number: 273203/05/02/99

  Software Test Works 4, Method ISO 527 Drag
- MFI
Ceast 6540/011

- Hardness
Shore D
Frank

- SEM Machine

JSM 840 A Scanning Microscope
Software : SemAfore

- DSC Machine

Perkin Elmer DSC 7 – Differential Scanning Calorimeter
Software : Pyris Series DSC 7

- Surface Roughness Analysis

ATOS III SO
Company : GOM mbH (Germany)
Appendix 2: Average conditions of processing of the Injection-Moulding Samples

Trials performed the 18/09/2008 and 23/09/2008.

Heating:

<table>
<thead>
<tr>
<th>Temperature</th>
<th>°C</th>
</tr>
</thead>
<tbody>
<tr>
<td>Temperature of the nozzle</td>
<td>210</td>
</tr>
<tr>
<td>Temperature of the cylinder zone 1</td>
<td>180</td>
</tr>
<tr>
<td>Temperature of the cylinder zone 2</td>
<td>200</td>
</tr>
<tr>
<td>Temperature of the cylinder zone 3</td>
<td>210</td>
</tr>
<tr>
<td>Temperature of the material</td>
<td>210</td>
</tr>
</tbody>
</table>

Injection time: 0,8 sec
Cooling time: 25 sec
Holding time: 20 sec

**Cycle time: 50 sec**

Injection Pressure: 106 to 170 bar
Holding Pressure: 70 to 105 bar
Appendix 2: Average conditions of processing of the Tape extruded samples

Trials performed the 14/10/2008 and the 28/11/2008.

<table>
<thead>
<tr>
<th>Name</th>
<th>Temperature zone 1 (°C)</th>
<th>Temperature zone 2 (°C)</th>
<th>Temperature zone 3 (°C)</th>
<th>Temperature zone 4 (°C)</th>
<th>Speed of the screw (rpm)</th>
<th>Speed of the rolls (rpm)</th>
<th>Average Thickness of the tape (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>LLa0E</td>
<td>120</td>
<td>135</td>
<td>135</td>
<td>135</td>
<td>40</td>
<td>0,4</td>
<td>0,45</td>
</tr>
<tr>
<td>LLb0E</td>
<td>161</td>
<td>177</td>
<td>174</td>
<td>163</td>
<td>150</td>
<td>0,51</td>
<td>2,1</td>
</tr>
<tr>
<td>LLa30E</td>
<td>130</td>
<td>140</td>
<td>150</td>
<td>150</td>
<td>50</td>
<td>0,31</td>
<td>0,95</td>
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<tr>
<td>LLa30EPC</td>
<td>120</td>
<td>150</td>
<td>160</td>
<td>160</td>
<td>150</td>
<td>0,7</td>
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<tr>
<td>LLb30E</td>
<td>149</td>
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<td>LLb30EPC</td>
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<td>176</td>
<td>179</td>
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<td>160</td>
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<td>0,5</td>
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<td>LLb50E</td>
<td>151</td>
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<td>172</td>
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<td>0,51</td>
<td>2,4</td>
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<tr>
<td>LLb70E</td>
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<td>183</td>
<td>183</td>
<td>150</td>
<td>0,45</td>
<td>2,35</td>
</tr>
</tbody>
</table>

Note: A speed of 50 rpm represents a flow of 20g/min.

E roll = 2,15 mm at least
Single Screw extruder, with screw : ½

Pre-Compounding of the 25/11/08 and 28/11/08

Twin screw extruder
Average Total output: 100g/min

<table>
<thead>
<tr>
<th>Name</th>
<th>Feeding flow XLIPE (g/min)</th>
<th>Feeding flow LLDPE (g/min)</th>
<th>Screw speed (rot/min)</th>
<th>Heating zone one to six (°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>LLa30EPC</td>
<td>29,9</td>
<td>72</td>
<td>80</td>
<td>180</td>
</tr>
<tr>
<td>LLa50EPC</td>
<td>71</td>
<td>72</td>
<td>80</td>
<td>180</td>
</tr>
<tr>
<td>LLb30EPC</td>
<td>29,4</td>
<td>70</td>
<td>86</td>
<td>180</td>
</tr>
<tr>
<td>LLb50EPC</td>
<td>66,2</td>
<td>67</td>
<td>86</td>
<td>180</td>
</tr>
</tbody>
</table>
Appendix 3: Average conditions of processing of the Extrusion of wires at Draka Kabel

Trials performed the 14/01/2009.

- Trial 1 – Feed of LLbDPE alone

Temperature Profile:

<table>
<thead>
<tr>
<th>Temperature of the barrel zone 1</th>
<th>125°C</th>
</tr>
</thead>
<tbody>
<tr>
<td>Temperature of the barrel zone 2</td>
<td>150°C</td>
</tr>
<tr>
<td>Temperature of the barrel zone 3</td>
<td>180°C</td>
</tr>
<tr>
<td>Temperature of the barrel zone 4</td>
<td>200°C</td>
</tr>
<tr>
<td>Temperature of the output</td>
<td>210°C</td>
</tr>
<tr>
<td>Temperature of the die 1</td>
<td>210°C</td>
</tr>
<tr>
<td>Temperature of the die 2</td>
<td>210°C</td>
</tr>
</tbody>
</table>

Screw speed: 100 rpm (max)

- Trial 2 – KE 30 0,4 alone

Same temperature profile, but the surface is rougher. In order to optimise this, we need either to increase the temperature or the speed of the extruder. The latter being already at his maximum value, we have increased the heating of the barrel:

Temperature Profile:

<table>
<thead>
<tr>
<th>Temperature of the barrel zone 1</th>
<th>125°C</th>
</tr>
</thead>
<tbody>
<tr>
<td>Temperature of the barrel zone 2</td>
<td>160°C</td>
</tr>
<tr>
<td>Temperature of the barrel zone 3</td>
<td>200°C</td>
</tr>
<tr>
<td>Temperature of the barrel zone 4</td>
<td>220°C</td>
</tr>
<tr>
<td>Temperature of the output</td>
<td>220°C</td>
</tr>
<tr>
<td>Temperature of the die 1</td>
<td>240°C</td>
</tr>
<tr>
<td>Temperature of the die 2</td>
<td>250°C</td>
</tr>
</tbody>
</table>

Screw speed: 100 rpm (max)
Same temperature profile and speed for the whole resting analysis.

- Pre-Compounding on the 08/01/09 and 09/01/09

Twin screw extruder

<table>
<thead>
<tr>
<th>Name</th>
<th>Feeding flow XLPE (g/min)</th>
<th>Feeding flow LLDPE (g/min)</th>
<th>Screw speed (rot/min)</th>
<th>Heating zone one to six (°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>KE 30 0.4</td>
<td>66.9</td>
<td>145</td>
<td>124</td>
<td>136</td>
</tr>
<tr>
<td>KE 30 0.6</td>
<td>61</td>
<td>145</td>
<td>126</td>
<td>170</td>
</tr>
<tr>
<td>KE 50 0.4</td>
<td>104</td>
<td>102.6</td>
<td>124</td>
<td>180</td>
</tr>
<tr>
<td>KE 50 0.6</td>
<td>106.9</td>
<td>105</td>
<td>130</td>
<td>180</td>
</tr>
</tbody>
</table>

Note: In the case of a content of 50%, a frequent unstable first phase of extrusion has been observed: after a few first minutes of normal running, the extruder suddenly extruded only the XLPE powder in big clots (what stops the flow of extruded wires), before coming back to a stable and normal flowing state after a few more minutes.
Appendix 4: Average conditions of processing for the Extrusion of inner-jacket at Nexans

Trials performed on the 25/02/2009.

- Trial 1: LLb20(0,6)JE

Heating profile:

<table>
<thead>
<tr>
<th>Zone 1</th>
<th>Zone 2</th>
<th>Zone 3</th>
<th>Zone 4</th>
<th>Neck</th>
<th>Die</th>
</tr>
</thead>
<tbody>
<tr>
<td>Theoretic</td>
<td>125°C</td>
<td>160°C</td>
<td>200°C</td>
<td>210°C</td>
<td>220°C</td>
</tr>
<tr>
<td>Real</td>
<td>125°C</td>
<td>160°C</td>
<td>200°C</td>
<td>215°C</td>
<td>&lt;210°C</td>
</tr>
</tbody>
</table>

* = little problem of heating at the beginning of the trials

Screw speed (rotation) = 19 revolution/min
T sample on the die: 220°C

- Trial 2: LLb15(0,6)JEAD

Heating profile:

<table>
<thead>
<tr>
<th>Zone 1</th>
<th>Zone 2</th>
<th>Zone 3</th>
<th>Zone 4</th>
<th>Neck</th>
<th>Die</th>
</tr>
</thead>
<tbody>
<tr>
<td>Theo</td>
<td>140°C</td>
<td>160°C</td>
<td>200°C</td>
<td>210°C</td>
<td>220°C</td>
</tr>
<tr>
<td>Real</td>
<td>140°C</td>
<td>160°C</td>
<td>195°C</td>
<td>210°C</td>
<td>220°C</td>
</tr>
</tbody>
</table>

Screw speed (rotation) = 20 revolution/min [increasing during the trial]

- Trial 3: LLb10JED

Temperature profile:

<table>
<thead>
<tr>
<th>Zone 1</th>
<th>Zone 2</th>
<th>Zone 3</th>
<th>Zone 4</th>
<th>Neck</th>
<th>Die</th>
</tr>
</thead>
<tbody>
<tr>
<td>Theo</td>
<td>140°C</td>
<td>160°C</td>
<td>200°C</td>
<td>220°C</td>
<td>220°C</td>
</tr>
<tr>
<td>Real</td>
<td>140°C</td>
<td>160°C</td>
<td>200°C</td>
<td>220°C</td>
<td>220°C</td>
</tr>
</tbody>
</table>

Screw speed (rotation) = 21 revolution/min
T sample on the die: 213°C

- Pre-Compounding on the 12/02/09 and 13/02/09