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# Closing the plastic waste material cycle in the power industry

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#### **Summary:**

Cables and wires are widely used in electronic equipment and power industry for transmission of both data and electricity, where the rapid development of power lines and constant update of electronic devices have resulted in an increase of waste cables. However, recycling of cables and wires is mainly focused on the recovery of metallic fraction due to their higher market value, rather than other plastics used in insulation. Polymeric materials from the scrap cables are often disposed in landfills or incinerated, which results significant risks to health and the environment due to the release of hazardous substances. Among different recycling techniques, the pyrolysis process is a promising alternative for waste recycling, where plastics are trasformed into light gases and oils which can be useful raw materials for fuels and/or chemical/polymer production.

This study aimed to analyze the problems related to cable waste recycling in detail, especially to the chemical recycling of the main polymers, which is an alternative route to incineration and landfill. An extensive literature search has therefore been carried out on the pyrolysis of the main polymeric materials used in cables, such as polyethylene, cross-linked polyethylene, and polyvinyl chloride. Finally, a method was proposed to analyse different polymers from cables in a pyrolysis reactor on a laboratory scale.

The University of South-Eastern Norway takes no responsibility for the results and conclusions in this student report.

Preface

# Preface

This report covers the work done during the master thesis "*Closing the plastic waste material cycle in the power industry*", carried out at the Faculty of Technology, Natural Science and Maritime Sciences of the University of South-Eastern Norway in Spring 2021, within the European Erasmus program. The thesis was offered by the University of South-Eastern Norway in collaboration with the external company Skagerak Energi AS. The primary focus was on recycling of plastic wastes from cable industry to produce sustainable fuels and consequently to reduce the incineration and landfilling of potentially hazardous waste. Further, this can be considered as the starting point of a wider work aimed for simulating the pyrolysis process in order to make an analysis of its environmental sustainability, which will be carried out at Polytechnic University of Milan, in Italy.

I would like to thank my supervisors, Professor Marianne S. Eikeland and Dr. Janitha Bandara for their support, advice and giving me this opportunity for working on this project. I would also thank Skagerak Energi AS and in particular Finn Werner Bekken for their help and providing the required equipment and materials.

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# Contents

| P | eface   | 3   |
|---|---|---|
| C | ontents   | 4   |
| N | omenclature   | 5   |
| 1 | Introduction  | 6   |
| 2 | Literature review   | 8   |
|   | <ul> <li>2.1 Generation of WEEE</li></ul>   | 8<br>10<br>13<br>16<br>17<br>20<br>23<br>24<br>25<br>26<br>27<br>27<br>30<br>34<br>35 |
| 3 | Experimental study  | . 39  |
|   | <ul> <li>3.1 Materials.</li> <li>3.2 Laboratory scale pyrolysis reactor and procedure.</li> <li>3.3 Mass balances and energy balances.</li> <li>3.4 Health and environmental safety.</li> <li>3.5 Determination of heating values.</li> </ul> | . 39<br>. 41<br>. 45<br>. 46<br>. 47  |
| 4 | Results and discussion.   | . 50  |
|   | 4.1 Feedstock properties<br>4.2 Effect of temperature on relative product yields  | . 50<br>. 53  |
| 5 | Conclusion  | 55  |
| R | eferences   | . 56  |
| A | opendices   | .62   |

Nomenclature

# Nomenclature

| AC       | Alternating current                        |
|----------|--|
| CV       | Calorific value                            |
| EEE      | Electrical and electronic equipment        |
| GC       | Gas chromatography                         |
| GHG      | Greenhouse gases                           |
| HCl      | Hydrochloric acid                          |
| HDPE     | High-density polyethylene                  |
| HHV      | Higher heating value                       |
| HV       | High voltage                               |
| ICTs     | Information and communication technologies |
| IPA      | Isopropyl alcohol                          |
| LDPE     | Low density polyethylene                   |
| LHV      | Lower heating value                        |
| LLDPE    | Linear low-density polyethylene            |
| LV       | Low voltage                                |
| NaOH     | Sodium hydroxide                           |
| PE       | Polyethylene                               |
| PP       | Polypropylene                              |
| PTF      | Plastic-to-fuel                            |
| PVC      | Polyvinyl chloride                         |
| TGA      | Thermogravimetric analysis                 |
| USN      | University of South-Eastern Norway         |
| WEEE     | Waste electrical and electronic equipment  |
| wt%      | Weight percentage                          |
| XLPE/PEX | Cross linked polyethylene                  |
|          |  |

Introduction

# **1** Introduction

Cables and wires are widely used in both electronic equipment and in power industry for the data transmission and distribution of electrical energy. When cable systems reach the end of their functional life, the discarded cables must be handled in a responsible manner. Cable and wire wastes are defined as e-waste according to the Directive 2012/19/EU [1], and must be treated separately because of the possible presence of hazardous materials. Different materials used in cables can be distinguished as shown in Figure 1.1. While the metallic fraction is recycled in different fields, the plastic insulation is considered as a residue, which is therefore disposed in landfill or incinerated [2].



Figure 1.1 Metals and plastic separation in cable processing [3]

Landfilling and incineration pose major health and environmental risks, as hazardous substances may be released into the environment, contaminating air, water, and soil [4]. At the same time, the recovery of energy and resources is not optimized: plastics are made up of petroleum-based products, so they can be chemically transformed into liquid oils and gases with a high calorific value, comparable with the commercial fuels [5]. In particular, pyrolysis is an efficient process, which can directly degrade plastic materials into number of valuable

Introduction

products such as monomer for plastic synthesis, fuels, and intermediates feedstock for petrochemical industry. It involves the breaking of long polymer chains into smaller hydrocarbons under heat and in the absence of oxygen. Reactor type, plastic type and the operating parameters determine the yield and the quality of pyrolysis products. During the process optimization, it is important to know the variables affecting most, and the process sensitivity over those parameters [6].

The main objective of this study is to analyze the problems related to the recycling of waste cables, which is an increasingly important part of electronic wastes. Structure and materials commonly used in the cable industry are analyzed, with particular emphasis on polymeric materials. An extensive literature search was therefore carried out on the pyrolysis of main polymeric materials used in cables, such as polyethylene, cross-linked polyethylene, and polyvinyl chloride. The original plan included experimental studies of cable waste pyrolysis at different temperatures using a laboratory-scale batch reactor, in order to assess the relative yields of the different fractions and the main components of the gas phase. Unfortunately, this was not possible due to technical shortcomings of the reactor and however, the procedure to be used for future work is described. In addition, a detailed comparison between conventional pyrolysis and microwave-assisted pyrolysis was planned. However, as it was not possible to compare the different performance of two reactors experimentally, the problems associated with cable recycling and conventional pyrolysis were analyzed using the literature data.

This chapter presents a theoretical framework of relevant topics for this study. In particular, the problem of electronic waste, the structure of cables and the problems concerning their recycling, the polymeric materials used in cable industry and the different recycling methods for plastics are addressed. Finally, the process of pyrolysis in general and in the specifics of the polymers used in cables is analysed.

### 2.1 Generation of WEEE

According to the definition given by the Directive 2012/19/EU, electrical and electronic equipment (EEE) "means equipment which is dependent on electrical currents or electromagnetic fields in order to work properly and equipment for the generation, transfer and measurement of such currents and fields and designed for use with a voltage rating not exceeding 1000 Volts for alternating current and 1500 Volts for direct current" [1].

EEE has become indispensable in contemporary society, improving the living standards of people all over the world. Technological progress, industrialization and increased disposable income have boosted the production and use of electronical and electronic goods [7]. At the same time, changes in information and communication technologies (ICTs) and rapid product innovation have significantly reduced the average useful life of most electronic equipment [8]. Furthermore, electronic devices are generally designed for a certain operating period, after which they stop working (end of life) or perform sub-optimally due to prolonged use (obsolescence).

As the average lifespan of electronic items decreases, obsolete products are replaced faster and faster, increasing the amounts of used electronics which may be reused, stored, recycled, or disposed of. Some devices are reconditioned or remanufactured, reducing their environmental impact and the demand for new devices, saving earth's raw materials. However, most electronic products are recycled for material recovery, discarded or exported, generating a waste stream containing hazardous and valuable materials [9].

This waste stream is referred to as e-waste, or Waste Electrical and Electronic Equipment (WEEE) and comprises the electronic devices that have become obsolete, nonworking, or unwanted which reached an end point of its functional life. Actually, there is no official definition of the term WEEE. According to the one provided by the Solving the E-waste Problem organization, "EEE becomes e-waste once it has been discarded by its owner as waste without the intent of reuse" [10].

WEEE is one of the fastest growing waste streams in the world. In 2019, the world generated 53.6 Mt of e-waste, an increase from the 45 Mt reported in 2016 [11], with an average of 7.3 kg per capita. As can be seen in Figure 2.1, the upward trend in global e-waste generation is expected to continue in the coming years and is predicted to produce 74.7 Mt by 2030 [7].



Figure 2.1 WEEE generation between 2019 and 2030. Adapted from [7]. Note: figures from 2020 onwards are projections; weight unit is in million tons (Mt)

WEEE is a complex, heterogeneous mixture of high-value materials, plastics, and hazardous components that make it difficult and costly to treat in an environmentally appropriate manner[9]. It includes a wide variety of products, so its composition varies significantly, depending on the design, composition, functionality of the devices and the origin of the waste

stream. E-waste includes huge quantities of metals, which can be divided in ferrous or nonferrous metals, and plastics; other components can be glass and composites [12].

Because of its hazardous content, e-waste can cause major environmental and health damage if not disposed in an appropriate way. Hazardous materials include heavy metals, chemicals such as ozone depleting substances and various flame retardants. All of these can potentially threaten healthy lives through the contamination of water, soil and air if not properly treated in the recycling process [4]. Due to heterogeneity and hazardousness of the waste stream and the costs involved, it is estimated that only 17.4% of e-waste is formally collected and recycled, the rest ends up in landfills or is incorrectly recycled in developing countries, leading to hazardous to worker and environmentally damaging situations [7].

In addition to the ecological, human health and social aspects, the recycling of e-waste has an enormous potential as alternative source of valuable metals and material considered "critical" due to the increasing demand, the low geological presence, and the technological difficulty of the recovery process. In fact, among the components of e-waste are materials such as iron, copper and aluminum, precious metals like gold, silver, platinum and palladium and rare earth elements which can be recycled [4]. Therefore, from a resource perspective, e-waste provides a huge amount of resources, reducing pressure on virgin materials and its recycling enables countries to reduce demand for raw materials in a sustainable way.

### 2.2 Cables and wires: features and role in WEEE

E-waste contains all categories of electronic products, such as electronic office equipment, entertainment devices, and household appliances. This includes, for example, computers, mobile phones, printers, televisions, washing machines, hoovers, refrigerators and so on. In addition to these, small devices such as computer keyboards, headphones, printed circuit boards, wires and cables also contribute to the e-waste stream. It is estimated that WEEE consists of 41% by weight of small equipment and IT devices, 24.5% by weight of large equipment, 20% by weight of temperature exchange equipment. The rest includes screens, monitors and light bulbs [7]. Figure 2.2 shows the e-waste totals estimated per category in 2019, including data for screens and light bulbs.





Figure 2.2 Estimates of e-waste totals per category in 2019. Adapted from [7]

Cables and wires are widely present in electronic equipment and the rapid progress of ICTs has increased their production and application. This development, with the constant update of electric and electronic devices, has resulted in an increase of waste cable. Cables and wires are also extensively used in power industry for the transmission and distribution of electrical energy. In recent decades, population growth, urbanization and the associated increasing demand for energy have resulted in the development of power lines. This has led to a growth in the market for power cables and consequently increased the amount of scrap cables produced.

The structure of cables depends on their application and influences their performance. In general, they consist of one or more metallic conductors, insulation, shielding, sheaths, and jacket, as can be seen in Figure 2.3, which shows the typical structure of underground cables. Cable conductors carry the electric current to the intended device or location and are typically made of aluminum or copper wires that can be solid or stranded. Copper and aluminum are widely used as a conductive material thanks to their high electrical conductivity [13]. Solid wires are the cheapest, stranded wires are more flexible, but they increase the maximum electrical stress by 20% [14].



Figure 2.3 Structure of underground cable [15]

Each metallic conductor is coated with an insulation material. The insulation is typically made from a dielectric material (e.g., plastic, rubber) and is provided to isolate the conductor from other paths through which the current might flow [16]. The dielectric compound must exhibit high dielectric strength, high insulation resistance, good mechanical properties, high chemical resistance and high temperature withstand capability [17]. Different materials are used depending on their application, but the most commonly ones are impregnated paper, butyl rubber, polyvinyl chloride, polyethylene, and cross-linked polyethylene [13]. The main insulation may be sandwiched between two semiconducting shields. They may be placed to prevent partial discharges at the interfaces between insulation and conductor and between insulation and external shielding layer. They are generally used in medium and high voltage cables, to provide a uniform electric field around the cable insulation and minimize the electrostatic stresses [16]. Ethylene copolymers filled with conductive carbon black, such as ethylene vinyl acetate, are typically used as semiconductor materials [14]. To protect cable from moisture and other aggressive chemicals present in the environment a metallic sheath could be inserted. It is usually made of lead alloy or aluminum. For protecting the metallic sheath from corrosion, layers of fibrous material like paper, jute, or polyvinyl chloride are used[13].

Single or multiple insulated conductors with their shields and sheaths are enclosed in a single jacket. Cable jackets are used to prevent moisture penetration and provide resistance to mechanical damage, UV light, flame, or chemical attack. They are commonly composed of polyethylene (low, medium, or high density), polypropylene or polyvinylchloride, and may also contain small quantities of carbon black, which is not conducting and provides the resistance to light [16].

Cables are usually classified according to the voltage for which they are designed. They can be divided in the categories shown in Table 2.1.

| Categories                 | Voltage                                  |
|----------------------------|--|
| Low-tension cables         | Maximum operating voltage of 1 kV        |
| High-tension cables        | Maximum operating voltage of 11 kV       |
| Super-tension cables       | Operating voltage from 22 kV to 33 kV    |
| Extra high-tension cables  | Operating voltage is from 33 kV to 66 kV |
| Extra super voltage cables | Voltage requirement beyond 132 kV        |

Table 2.1 Classification based upon voltage rating of the cable [18]

These classes can be combined into three broad categories: low-voltage, medium-voltage, and high-voltage cables. However, the exact demarcations have never been very precise because they vary between different countries and over time [19]. Internationally, it can be assumed that up to 1 kV there are low-voltage cables, up to 30 kV there are medium-voltage cables and beyond that there are high-voltage cables [16].

## 2.3 Cable recycling problems and technologies

Scrap cables that are collected from different sources consist of different cable types and material components, such as metals, plastics, rubber, and glass fibers. The heterogeneous nature of waste cables makes it difficult the reuse and recycling of cables, but nowadays for economic and environmental reasons the recovery of waste cable has become an important

issue. Wires recycling has mainly focused on the recovery of metallic fraction due to its higher market value, while polymeric materials have been considered as a residue, being landfilled or incinerated after metal separation [2]. However, nowadays increasingly stringent new regulations and growing concern for the environment make it necessary to reduce landfilling as much as possible. Thus, with a focus on resource recycling, improved methods for recycling both metals and polymers in cable waste need to be implemented. Generally, cables are first pre-sorted, then they are grinded, and metals are separated using electrostatic plates or dense medium separation as can be seen in Figure 2.1, which shows the general process of scrap copper cable recycling process. The remaining material (up to 70% of the total cable) is a mixture of different polymers and contains metal residue (1-2%) [20].



Figure 2.4 Schematic representation of scrap copper cable recycling process [21]

Actually, different techniques for cable recycling and metal recovery can be exploited. The most common waste cable recycling technology is the mechanical method, which is economical and eco-friendly, as it avoids the secondary pollution associated with toxic emissions from burning plastics. It involves cable stripping and crushing and presents several challenges such as loss of metal, high energy consumption and a tendency to generate dust. Thus, other physical recycling techniques can be used, such as freezing, high- pressure water

jet technology process or ultrasonic separation, which are linked to high recycling efficiency, excellent environmental benefits and high-quality products [22].

To separate the different components also energy recovery techniques, such as incineration or thermal decomposition, can be used. Incineration has been the most widely used technique in recent decades, but in addition to thermal energy it produces toxic gases from the combustion of plastics, due to the presence of halogen-containing flame retardants. Moreover, in the incineration process, the surface of the metal is severely oxidized, largely decreasing its purity. As a consequence, recycled metal cannot be used directly and has to undergo numerous treatments, increasing costs [22]. On the other hand, thermal decomposition allows efficient separation of metals and plastics by heating the materials to high temperatures in the absence of oxygen. Organic fuels are obtained from plastics, and the oxygen-free environment reduces the possibility of metal oxidation and the formation of toxic gases [23].

Metals can also be recovered by chemical methods that allow their dissolution in organic solvents that dissolve the plastic sheaths preserving the metals. However, the recycled metal is in liquid or gaseous form, rather than in the desired metallic form and there are problems associated with the disposal of impure organic solvents, as well as the risks of environmental pollution by the solvents [23].

Separation of plastic and metals can therefore be achieved by various processes, each with its own advantages and disadvantages. After that, metals are reused in different fields depending on the purity obtained. Separated polymeric components can be used for further recycling procedure, but since they are less clean than virgin material, the recycled material is typically used in applications with lower quality requirements.

Polymeric materials are extensively used in electronic cables industry because they exhibit good insulating properties, high flexibility, shockproof nature and easy processability. Currently the most common polymers present in waste cables are polyvinyl chloride (PVC) and polyethylene, either in crosslinked (XLPE) or thermoplastic (PE) structure. However, cross-linked polyethylene (XLPE) is still difficult for material recycling because of its non-thermoplasticity [20].

### 2.4 Different types of plastics: thermoplastics and thermosets

In order to understand the problems related to the recycling of polymeric materials in cables, a distinction between thermoplastic and thermosetting polymers must be made.

Thermoplastics soften and become viscous by the application of heat and then they turn into glassy or semi-crystalline solids after the subsequent cooling process. The melt-solidification thermal behavior of thermoplastics is a reversible and limited process, whereby multiple heating and cooling cycles can be performed in a limited number without any severe structural or functional changes. However, if in the molten state they are brought to too high temperatures they degrade irreversibly. Used polymers that have become waste can be remolded and recycled thanks to the reversibility of physical softening or melting. Most linear polymers and some branched flexible chain polymers are thermoplastic. Examples of these are polyvinyl chloride, polyethylene, polystyrene [24], shown in Figure 2.5.



Figure 2.5 Types of thermoplastics

Thermosets undergo an irreversible liquid-solid transition when they are heated: during the production process, small molecules are chemically linked and create complex interconnected meshes, resulting in a permanent rigid product. Therefore, further heating of thermosets will result in chemical decomposition and severe structural alteration without its going through a fluid state. Thermosets are generally not considered recyclable due to the relative inability to process these materials by melting because only at very high temperatures the bonds can be broken, and the polymer destroyed. Most cross-linked polymers are thermosetting. They are

Literature review generally harder and more resistant than thermoplastics. Examples of thermosets are urea formaldehyde, epoxy, phenolic resins [24].

### 2.5 Polymeric materials in cable industry

Nowadays different types of polymers such as polyvinyl chloride (PVC), polyethylene (PE), cross-linked polyethylene (XLPE) are being used for insulation purposes. Figure 2.6 shows the main wires and cables polymer market shares, where the others are rubbers, polypropylene, polyamide, polyurethane, silicone, and fluoropolymers.



Figure 2.6 Wires and cables polymer market shares. Adapted from [25]

Polyvinyl chloride (PVC) is a cost-effective and versatile thermoplastic, used for the insulation of cables and wires. It is the third most commonly produced polymer in the world after PE and PP and it is obtained by radical polymerization from vinyl chloride monomer [26][27]. It has good insulating properties, simple installation and maintenance, excellent chemical resistance to acids, alkalis, and corrosion. It is also fire resistant due to its high chlorine content, which corresponds to 57% of the PVC molecular weight and acts as a flame retardant [28]. To improve the flexibility and thermal and mechanical stability of PVC, stabilizers, and plasticizers, such as low molecular weight phthalates, are often added. PVC is available in two

broad categories, flexible and rigid. Actually, flexible PVC is widely used in cables and wires, so in most of them organic additives are present. The material properties depend on the concentration of the added additives but these can greatly influence the PVC recovery process[2]. In cables industry due to its high dielectric constant (5-9) and high dissipation factor (at 20 °C 4-12%), PVC is suitable as insulation for low-voltage application [29]. The main disadvantage of PVC is that its physical characteristics change with temperature, where at low temperatures it becomes hard and difficult to strip, and at high temperatures it becomes very soft. Because of this, the temperature of PVC for general use should not exceed 70°C, remembering also that its glass transition temperature is around 80°C [30]. Furthermore, if PVC is exposed to sunlight, it can be degraded by ultraviolet radiation [14].

Polyethylene (PE) is a long-chain polymer produced through the polymerization of ethylene gas. It is a thermoplastic with good electrical properties (low dielectric constant, low dielectric loss and high breaking strength), mechanical hardness and flexibility, good resistance to chemicals, easy processing and low cost [14]. According to the degree of branching and the molecular weight it can be classified in low density polyethylene (LDPE) with a high number of short and long chain branches, linear low-density polyethylene (LLDPE) with a large fraction of short branches and high-density polyethylene (HDPE) with low branching [31]. The thermal and mechanical properties of polyethylene depend strongly on the degree of branching and the molecular weight of the polymer. Generally, a higher density of the polymer is accompanied by a higher degree of crystallinity and a higher hardness [32]. In industrial applications, polyethylene is often used as electrical insulators for medium and high voltage cables. Polyethylene insulation cable has good dielectric properties, high insulation resistance, good technological properties, easy processing and good moisture resistance, but for higher operating voltage level, special additives must be added [30].

Thanks to its excellent electrical and mechanical properties, LDPE has been used as an insulating material for high voltage AC cables for about fifty years [33]. However, its low glass transition temperature limits its operating temperature to around 75 °C, so LDPE is not suitable for larger capacity power cables [34]. Linear high-density PE (HDPE), on the other hand, has a higher melting point and can withstand higher operating temperatures. But, although HDPE

found some success in medium voltage cables, it has failed to become the main material in the high voltage cable sector [35].

To improve its performance, polyethylene is cross-linked into XLPE (or PEX) through the creation of intermolecular covalent bonds to form a mesh structure. The cross-linking of polyethylene improves its thermal deformation, wear resistance, chemical resistance, mechanical strength, electrical properties and especially heat resistance [30]. The operating temperature rises to 90°C and unlike non-cross-linked polyethylene, short-term temperature peaks above its melting point are permitted. There are different forms of PEX depending on the crosslinking technique, the main ones include PEX-a prepared by the peroxide method for radical formation, PEX-b by the silane grafting method and PEX-c by a gamma irradiation method for radical formation [36].

Thanks to its many advantages, cross-linked polyethylene (XLPE) is widely used as a cable insulation material. XLPE cable has very good electrical properties (dielectric loss should be small as well as capacity), easy installation, better insulating properties than PVC, which allows for lower thicknesses, lower weight, and consequently lower costs. Excellent anti-ageing properties and high resistance to heat distortion mean that the normal operating temperature is 90°C, the short-term failure temperature is 130°C and the short-circuit temperature is 250°C [30]. As the temperature of the conductor is proportional to the amount of load current through the cable, large current can be transmitted through an XLPE cable than through a non-cross-linked cable of the same dimensions.

Unlike non-cross-linked polyethylene, which is a linear molecular, easy to be deformed at high temperature, XLPE is a thermosetting polymer. This means that it hardens irreversibly during polymerization and cannot be remelted for reshaping and reuse, thus making recycling difficult. This significantly reduces their efficiency and economic viability in recycling. Mechanical recycling of XLPE is not feasible because even at higher temperatures its structures prevent flow [36]. Table 2.2 shows a comparison between PVC, PE and XLPE performance.

| Performance                                       | PVC         | PE        | XLPE      |
|---|-------------|-----------|-----------|
| Density [g cm <sup>-3</sup> ]                     | 1.4         | 0.92      | 0.92      |
| Insulation thickness [mm]                         | 1.0         | 0.70      | 0.70      |
| Maximum temperature [°C]                          | 60 - 70     | 75        | 95        |
| Softening temperature [°C]                        | 120         | 105 - 115 | 127       |
| Instantaneous short-circuit temperature [°C]      | 135         | 150       | 250       |
| Insulation resistance $[m\Omega \text{ Km}^{-1}]$ | 20          | 1000      | 1000      |
| Volume resistivity [ $\Omega$ cm]                 | 1012 - 1015 | 1017      | 1017      |
| Dielectric constant [60 HZ]                       | 6 - 8       | 2.3       | 2.3       |
| Weathering resistance                             | Excellent   | Reduced   | Common    |
| Aging resistance                                  | Reduced     | Common    | Excellent |
| Embrittlement at low temperature                  | Reduced     | Common    | Excellent |

Table 2.2 Comparison of PVC, PE and XLPE performance. Adapted from [30]

# 2.6 Recycling of plastic waste

As mentioned in chapter 2.3, after the separation of metals and polymers in cables, the latter are either disposed in landfills or incinerated. It is estimated that around 70,000 tons of cable sheathing materials are landfilled each year [37]. This information is actually from 2013, but as technological and industrial development has increased the amount of cables used in electronic equipment and the power industry, it can be expected that even more plastics from waste cables are landfilled nowadays.

Plastic waste is bulkier than other organic residues and takes up large volumes in landfill, causing higher costs in the disposal [38]. Furthermore it takes up to hundreds years to degrade naturally due to the presence in its molecular bonds of hydrogen, carbon and a few other elements such as nitrogen and chlorine that make plastics very durable [5]. Landfilling poses major health and environmental risks, particularly in causing soil and groundwater contamination, increased greenhouse gas (GHG) emissions, fire and public health problems [39]. It is therefore necessary to improve recycling methods to manage plastics and reduce landfill disposal. Plastics are recycled using different technologies, classified into 'primary', 'secondary', 'tertiary' or 'quaternary' methods, as shown in Figure 2.7.



Figure 2.7 Polymer Recycling Techniques

Primary (closed loop) recycling involves the reprocessing of uncontaminated, single type of polymer to obtain a product with the same function as the original one. Typically, the feedstock is processing waste reintroduced into the heating cycle of the operating line to increase production [40]. Secondary (mechanical) recycling provides secondary raw materials whose use is different from the one of the original products, using physical transformations. The process is often called "downgrading" or "downcycling" because materials of lower value and with less mechanical strength than virgin materials are obtained. Primary and secondary recycling both involve mechanical processes in which polymers are sorted, grounded, washed, and extruded. This treatment lead to polymer degradation, so applications of mechanical recycling being limited by the number of reprocessing cycles that a given polymer can endure [41]. Tertiary (chemical) recycling implies a chemical alteration in the polymer structure by chemical or thermochemical procedures, in order to recover monomer or petrochemical components in plastics. However, chemical recycling is not commonly implemented on an industrial scale because present methods require sizable energy inputs [41]. Quaternary (energy recovery) recycling is incineration, which involves the recovery of steam, heat, and electricity from the waste through combustion. Generally, complex mixtures of even hazardous materials are burned, so greenhouse gases and toxins can be released. In addition to the environmental

problems, it represents an energy loss, because the energy generated by mass burning of plastics is less than the energy conserved by recycling. For example, the heating value for plastics is ~  $36,000 \text{ kJ kg}^{-1}$ , whereas mechanical recycling conserves ~  $60,000-90,000 \text{ kJ kg}^{-1}$ [41].

Mechanical recycling (primary and secondary methods) is sensitive to the type and age of the feedstock and works best with separately collected plastics, while it cannot efficiently recycle mixed plastic waste [6]. A thorough sorting and purification process is required to remove contaminants and identify individual polymers that may react differently to reprocessing. Numerous additives, plasticizers and dyes that can affect the tendency of materials to be easily transformed into other products may be present. Moreover, due to performance losses and downcycling, products ultimately have to be incinerated or end up in landfills after shorter use cycles. Given the technological limitations, it cannot solve the problem of plastic waste accumulation on its own [39].

Over the past few years, chemical recycling has been advocated as a potential solution to fully utilize plastic waste, recycling larger quantities of polymers and meeting the increased energy demand. Plastics are made up of petroleum-based products, so they can be chemically transformed into liquid oil and gas with a high calorific value, comparable to commercial fuels [5]. Tertiary recycling generates not only petrochemicals but also monomers, which can be used to produce new plastic products. This would reduce the demand for fossil resources to produce plastics, as well as greenhouse gas emissions associated with their production and use, which in 2015 were 1.78 Gt of CO2-eq and are expected to rise to 6.5 Gt. CO2-eq in 2050 [42].

Chemical recycling reduces the amount of plastic waste while generating the raw materials from which polymers are originally made, by recycling the monomer or the feedstock [40]. Thus, it plays an important role in the transition towards circular economy and a more sustainable society, enabling the plastic waste material cycle to be closed. In fact, the main goal of the circular economy is to keep as many products as possible in a loop, allowing them to be reused and reducing raw materials and energy consumption, waste production and GHGs emissions, promoting an all-round effective use of materials [43].

In comparison with mechanical recycling, chemical one allows the treatment of heterogeneous and contaminated waste streams, if sorting and pre-treatment processes are not economically or technically possible [44]. It would therefore be useful to consider chemical recycling as a complement to mechanical recycling in a holistic perspective. When plastic-to-fuel (PTF) technologies deal with end-of-life plastics that are not easily or economically recyclable, they are considered complementary to classical recycling [45].

### 2.7 Challenges and opportunities for pyrolysis of plastic waste

Pyrolysis is one of the most promising processes among all the different chemical recycling technologies, increasing the waste management method, reducing reliance on fossil fuels, generating additional energy sources and preventing environmental pollution [46]. The following sections present the process and the factors that influence it most, while the final chapter of the literature review focuses on the pyrolysis process of plastics used in cables.

#### 2.7.1 Pyrolysis process

Pyrolysis is defined as thermal degradation in inert atmosphere free from oxygen of long chain organic materials with the presence of a catalyst (catalytic pyrolysis) or without (thermal process). The organic macromolecules degrade in smaller and less complex molecules by controlling the heat flow and the operating pressure [39]. Thermal pyrolysis is an endothermic process involving the breaking of bonds, so it requires high levels of energy and it is generally carried out at temperatures between 350 and 900 °C [38]. Pyrolysis of plastic waste produces non condensable gases, liquids and solid carbonaceous residue (char) namely "pyrolysis gases," "pyrolysis oils," "pyrolysis coke," respectively [47]. From the oil fraction hydrocarbons in the gasoline range (C4-C12), diesel (C12-C23), kerosene (C10-C18) and motor oil (C23-C40) can be recovered [38].

The relative yield and the quality of the different products depend on the composition of the feedstock, the type of reactor and the operating parameters, such as temperature, pressure and residence time [6]. Pyrolysis is very flexible since process parameters can be optimized for the product yield based on the product requirement and this is highly advantageous in terms of the economy of the whole process. The main goal of plastic pyrolysis is to obtain high-value

pyrolytic oil at relatively medium temperature ranges between 500 and 650 °C, with yields up to 75-80 wt%, but the process can be adapted to optimize the production of wax, monomers, aromatics [39].

In addition to the liquid fraction, the process produces carbonaceous residue, the unburnt plastic left over in the reactor, which could be used as a fuel or a feedstock for other petrochemical processes. Pyrolysis char production is very low in quantity (1-1.3 g from 1 kg plastic) in comparison to other process products such as liquid oil and gases [48], [49], even if it could have a higher heating value (HHV) in the range of 23.04-36.29 MJ kg<sup>-1</sup> [39]. The process generates also hydrocarbon rich gas, which has, depending on feed and conditions, a heating value of 25-45 MJ kg<sup>-1</sup> making it ideal for energy recovery [6]. Thus, the pyrolysis gas can be circulated back into the process for heating purposes, greatly reducing dependence on external heating sources.

#### 2.7.2 Feedstock

Pyrolysis can use different types of plastics as feedstock, from those contaminated with toxic substances to composite materials, consisting of different polymeric layers, because utilizes an inert atmosphere free from oxygen, so it does not form dioxins by reaction of products with oxygen. Moreover, even if there is a limit, it can tolerate higher levels of contaminants than mechanical recycling, reducing the number of pre-treatments required [6], [39].

The compositions of the different polymers are often reported in terms of their approximate analysis, which represents the chemical properties of the plastics based on four elements: moisture content, fixed carbon, volatile matter, and ash content. Among these, volatile matter and ash content have the greatest influence on the yield of liquid oil in the pyrolysis process. High volatile matter favors the liquid oil production while high ash content decreases it, consequently increasing the gaseous yield and char formation [5]. Table 2.3 summarizes the proximate analysis of different plastics.

| Type of plastics          | Moisture | Fixed carbon | Volatile | Ash   |
|---------------------------|----------|--------------|----------|-------|
|                           | (wt%)    | (wt%)        | (wt%)    | (wt%) |
| High-density polyethylene | 0.00     | 0.01         | 99.81    | 0.18  |
| Polyvinyl chloride        | 0.80     | 6.30         | 93.70    | 0.00  |
| Low-density polyethylene  | 0.30     | 0.00         | 99.70    | 0.00  |
| Polyethylene              | 0.10     | 0.04         | 98.87    | 0.99  |

Table 2.3 Proximate analysis of different plastics [5]

Based on Table 2.3, it can be noted that the volatile matter for all plastics is very high while the ash content is low. As a consequence, plastics have high potential to produce large amount of liquid oil through pyrolysis process [5]. Moreover, the high carbon content and the absence of significant amounts of oxygen confirm the high potential of polymers as a chemical feedstock [44]. However, when commercial polymers contain additives like fillers, plasticizers, colorants, and flame retardants, such as polymers in cables, the yield of char increases due to an intrinsic increase in the fixed carbon content [50].

#### 2.7.3 Temperature

In addition to the feedstock, temperature is one of the most important parameters in pyrolysis process, as it controls the cracking reactions of the polymer chains. It has been shown that temperature has the greatest impact on the reaction rate which can influence the product composition of liquid, gaseous and char for all the different types of plastics [5]. At low temperatures, long-chain hydrocarbons are produced, while an increase in temperature leads to shorter carbon chains due to the breaking of C-C bonds [51]. In fact, the high temperature favors increased gas formation as the molecules breakdown and form a wide range of smaller, more thermally stable organic molecules. Moreover, with the higher amount of energy there is an increase in secondary reactions, so the amount of oil and wax decreases with an increase in temperature and aromatic compounds can be produced [39], [51]. Regarding the properties of the oil fraction, lower pyrolysis temperature favors the formation of light hydrocarbons, whereas oils obtained at higher pyrolysis temperature results in a high amount of heavier hydrocarbons due to the conversion of volatiles into light hydrocarbons as non-condensable

gases. Increase in process temperature results in increased density, which in turn increases the viscosity of oil [46].

#### 2.7.4 Type of reactors

The choice of the reactor type is another key parameter in defining the yield of the process. Reactors with excellent heat and mass transfer properties should be preferred. Batch reactors, closed systems with no inflow or outflow of reactants or products, allow high conversions with high residence time, but the variability of product from batch to batch, high labor costs per batch and the difficulty of large-scale production represent important disadvantages. A semibatch reactor allows reactant addition and product removal at the same time and the flexibility of adding reactants over time implies a better reaction selectivity. The disadvantages of semibatch reactor are similar with the batch reactor, so they are more suitable for small scale production and are more appropriate for laboratories, even if the operating parameters can be easily controlled [5]. Fixed-bed reactors are easy to design and are very economical due to the manageable maintenance and operation of the unit. However, temperature control and heat transfer limitations are of great importance, and constraints such as the irregular particle size and shape of plastics as feedstock would cause problem during feeding process [5], [39]. In contrast, in fluidized bed reactors the operating parameters such as temperature can be controlled quite flexibly, allowing for adequate product distribution. They are characterized by excellent heat and mass transfer due to the presence of a heated fluidizing media, typically sand, but however there is a possibility of bed defluidization when melted plastics stick [39]. One of the major advantages of the fluidized bed reactor is its scalability. Moreover, thanks to the continuous feeding, it is more flexible than the batch reactor since frequent feedstock charging can be avoided and the process does not need to resume often. For this reason, fluidized bed reactor would be the most suitable reactor for large scale operation in terms of economic point of view [6].

Other reactor configurations have been described in the literature, such as kiln reactors, stirred tank reactors, conical spouted bed reactors and microwave-assisted reactors, but generally in laboratories batch reactors allow a higher oil recovery, while fluidized pyrolysis is suited on a large scale, for continuous process [52].

#### 2.7.5 Pressure and residence time

Pressure could affect the yield of the process, but most experiments were conducted at atmospheric pressure (in the reactor with open vent), so the effect of pressure is not well described in literature. It was shown that an increase in pressure increases the gas production and the pressure has the greatest effect at lower temperatures [39], [53]. There is not much research on the effect of pressure on the pyrolysis process because it has less effect at higher temperatures, which are generally the ones of greatest interest. In any case, the effect of pressure on the product distribution should be taken into account when running at temperature below 450  $^{\circ}$ C [5]. In addition, costs of compressors and pressure transmitters would have to be added if pressure was also considered.

Even residence time, the average time that the particle spends in the reactor, may influence the product distribution. A long residence time allows secondary reactions to occur and crack oil, increasing the conversion of primary product towards more thermal stable product as light molecular weight hydrocarbons and non-condensable gases. However, there could be a temperature limitation in the process that influence the product distribution where until that instant, the residence time has not much effect [5], [54]. Therefore, also residence time is a temperature dependence factor that may have potential influence on product distribution of the plastic pyrolysis at lower temperature. In addition to the main operating conditions, the pyrolysis process can be influenced in terms of yield and product quality by many other parameters such as particle size, plastic type, catalyst, loading ratio [55].

#### 2.7.6 Heating technology

Another important element that influences the process is the heating technology used. In addition to the conventional pyrolysis, microwave-assisted technology has recently been used to pyrolyze plastic material and have attracted much attention because of its advantages over conventional electric heating processes.



Figure 2.8 Temperature distribution, heat and mass transfer in the conventional electrical heating and microwave-assisted heating [56]

For the conventional electrical heating, heat is transferred from high-temperature gas to the sample particle surface through convection and it is then further transferred from the outside surface to the inside core through conduction. A temperature gradient from the outside to inside of the feedstock particle is formed, and the released volatile diffuses from the inside core to the outside surface through a higher temperature region, as shown in Figure 2.8. For the microwave-assisted heating, microwave penetrates the feedstock particle and then microwave energy is transformed into thermal energy inside the particle. Due to the heat loss effect of particle surface, thermal energy constantly accumulates inside the plastic particle and is transferred outwards. A temperature gradient from inside to outside of the particle is formed, and the released volatile diffuses from the inside core to the outside surface through a lower temperature region, as shown in Figure 2.8. This effectively avoids the secondary cracking of the vapors, resulting in a higher oil yield than the conventional electrical heating pyrolysis. Due to different heating mechanisms, microwave technology has different characteristics compared to conventional electric heating [56]. A comparison between microwave assisted and electrical heating is presented in Table 2.4.

| Microwave assisted heating                | Electrical heating                         |
|---|--|
| Conversion of energy                      | Transfer of energy                         |
| In-core volumetric and uniform heating at | Superficial heating via conduction,        |
| molecular level                           | convection, and radiation                  |
| Hot spot                                  | No hot spot                                |
| Rapid heating                             | Slow heating                               |
| Higher electricity conversion efficiency  | Lower electricity conversion efficiency    |
| Lower energy consumption                  | Higher energy consumption                  |
| Selective                                 | Non-selective                              |
| Dependent on material's properties        | Less dependent                             |
| Higher level of control                   | Lower level of control                     |
| Lower thermal inertia and faster response | Higher thermal inertia and slower response |
| Improved product yields                   | Lower product yields                       |

Table 2.4 Comparison between microwave assisted and electrical heating [56]

In contrast to electrical heating, microwave heating heats the material at the molecular level, showing a higher energy efficiency, but relies on the interactions between microwave irradiation and the feedstock involved, therefore the yield and quality of oil obtained strongly depends on the properties of the feedstock. Moreover, polymers in general have poor dielectric properties, and they are not very susceptible to microwave heating. Thus, in addition to many advantages, this technology suffers from the poor thermal conductivity of plastics, whereby some conductive materials have to be included in the shredded plastic waste, e.g., graphitic carbon or inorganic oxides. Carbonaceous material exposed to microwaves can reach temperatures of up to 1000 °C in a few minutes, depending on the intensity of the microwaves. The heat is absorbed by the carbon and is conducted to the waste plastic [6]. In addition, there is a problem with the generation of hot spots because molten plastic has a higher dielectric loss factor than solid plastic. This difference in dielectric loss factor leads to small spots with a higher temperature than the rest of the mass. In fact, since plastics have a very low microwave susceptibility, a huge amount of microwave energy is needed to melt the plastic, but when the plastic becomes molten, it has a higher susceptibility and can trigger a thermal runaway reaction. This thermal runaway can lead to a pyrolytic process with poor temperature control.

For this reason, researchers have countered this problem by using microwave susceptors mixed with the plastic [57].

Other studies have analyzed the possibility of a third technology: induction heating. Although there are only a few examples of induction heating in the pyrolysis processes, induction heating has been shown to reach high temperatures in less time than conventional pyrolysis, with better temperature control. In contrast to these advantages, induction heating has a major disadvantage: it cannot heat all materials and requires a specific magnetic material with high resistivity, so it is essential to build a reactor with iron or aluminium to implement induction heating in the reactor [58].

### 2.8 Pyrolysis of common plastics used in cable industry

As mentioned in chapter 2.5, the main polymers used in cable industry are PVC, PE and XLPE. It should be noted, that additives are very often added to the polymers used in cables, reducing the net energy content and increasing the yield of char in the pyrolysis process [46], [50]. However, this aspect is not studied in this work.

Thermal pyrolysis of different types of plastic has been carried out in the last years and polyolefins are considered an ideal feedstock for pyrolysis, degrading into valuable hydrocarbon products, while PVC shows many challenges and is limited in the feedstock mixtures to less than 5% and typically around 1 to 2% [6]. This because during thermal degradation it produces chlorinated hydrocarbons and HCl, which lead to corrosion problems in the reactor and generates halogenated oil.

#### 2.8.1 Polyethylene pyrolysis

Several studies have been conducted on the pyrolysis of PE under different operating conditions and in different reactors. A high calorific value was found for pyrolytic oil obtained from HDPE (42.9 MJ kg<sup>-1</sup>) compared with conventional fuels, where gasoline, kerosene and diesel have a reported heating value in the range of 43.4-46.5 MJ kg<sup>-1</sup>, 43.0-46.2 MJ kg<sup>-1</sup> and 42.8-45.8 MJ kg<sup>-1</sup>, respectively [39]. Table 2.5 shows a comparison of heating value between HDPE and LDPE oils and conventional fuels. Pyrolysis of HDPE in a batch reactor has been

shown a liquid oil yield of up to 84.7 wt% and a gas yield around 16.3 wt% at 550 °C [59], but a rise in temperature increased the gas due to further oil cracking. This shows that too high temperature would reduce the liquid oil yield and increase the gaseous product since the process had passed the maximum thermal degradation point [59], [60]. It has also been found that in a semi-batch reactor at temperature range of 400-550 °C, an increase in temperature reduced char yield, and the sulfur content in the HDPE pyrolytic oil was very low (0.019%), which made it cleaner to the environment [61]. Pyrolysis of LDPE has been also studied and a high CV pyrolysis oil (39.5 MJ kg<sup>-1</sup>) compared to conventional fuels has also been reported [62], while high liquid yields have been obtained, reaching 93.1 to 95 wt% with low gas production between 500 and 550 °C [59], [63].

| Fuel          | HDPE                | LDPE                | Gasoline            | Kerosene            | Diesel              |
|---------------|---------------------|---------------------|---------------------|---------------------|---------------------|
| Units         | MJ kg <sup>-1</sup> |
| Heating value | 42.9                | 39.5                | 43.4 - 46.5         | 43.0 - 46.2         | 42.8 - 45.8         |

A single-stage degradation has been proposed for PE, with similar characteristics between HDPE and LDPE reaction [52], [64]. HDPE undergoes thermal depolymerization at temperatures above 400 °C, considering that the average thermal decomposition temperature for PE is 423 °C [47], but only small quantities of low-molecular-weight products are produced [41]. In fact, at low temperature waxes are produced due to an incomplete conversion, as the feed need high activation energy (170-230 kJ mol<sup>-1</sup> for LDPE and 140-230 kJ mol<sup>-1</sup> for HDPE), [64], while at higher temperature there is also an important gas fraction production. Increase in process temperature increases the oil density, reducing the light hydrocarbon fraction due to the conversion of volatiles into light hydrocarbons and non-condensable gases [46], [64]. Overall, in a semi-batch reactor the maximum oil yield has been obtained at 550 °C while maximum low-density oil volume has been recovered at 500 °C with no presence of waxes [65]. It has been found that in HDPE pyrolysis oil, the lower hydrocarbons fraction (C5-C12) reduced from 45.06% to 28.75% while the heavier one (>C19) increased from 5.09% to 30.02% with an increase in temperature from 450 to 550 °C [46], as shown in Table 2.6.

| Temperature HDPE | 450 °C | 500 °C | 550 °C |
|------------------|--------|--------|--------|
| C5 - C12 (%)     | 45.06  | 78.90  | 28.95  |
| C13 - C19 (%)    | 49.84  | 41.22  | 41.22  |
| >C19 (%)         | 5.09   | 13.89  | 30.02  |

Table 2.6 Hydrocarbon fractions in HDPE pyrolysis oil based on carbon number [46]

The same increase in temperature showed a slight increase in the oil density while the volume obtained was profoundly raised; a further increase in process temperature resulted in a reduction in oil volume [46]. This confirms that during pyrolysis of individual plastics the mass fraction and the volume of liquid increase with an increase in the operating temperature, reaches a peak and then decrease [65].

HDPE produced a low amount of carbon residue and a high oil amount of 117 ml (for 100 g of feed sample) at 500 °C, with an average production of aromatics in oils of 13.67%, so not a significant fraction. Moreover, the max amount of light fraction (C5-C12) produced is 78.90% at 500 °C [46]. This confirms that pyrolysis oil from HDPE is mainly constituted of paraffinic and olefinic hydrocarbons while the presence of aromatics is nearly negligible [64].

Regarding the gas fraction, as shown in Figure 2.9, an increase in temperature causes an increase in mass and volumetric gas yield, a trend that reflects the behavior of all plastics, as demonstrated in the literature [47].

High levels of hydrocarbons, mainly alkanes and alkenes ranging from C1 to C5, are present in the gas resulting from PE pyrolysis, with an average hydrocarbon contents of about 87% [46], [47]. Thus methane, ethane, ethylene, propane, propylene, butane and 1-butene can be found in significant quantities in different percentages depending on the operating conditions, together with hydrogen [46], [64]. Figure 2.12 shows the volumetric proportions of main hydrocarbons in gases from PE and PVC. Actually at 700 °C there is a decrease in the amount of hydrogen and at the same temperature it has been found the highest hydrocarbon content, but this observation requires further analysis because secondary reactions, which promote the production of hydrocarbons and need to be further investigated, may be present [47]. At temperatures above 700 °C, the amount of hydrocarbons decreases, thanks to the further splitting of heavier compounds due to the increase in supplied thermal energy.



Figure 2.9 Gas yields from PE and PVC by (a) volume and (b) mass. Adapted from [47]

Relatively high levels of higher hydrocarbons generate gas with a high density (1.54 kg  $m_N^{-3}$ ), which decreases with increasing the operating temperature, owing to the further splitting of higher hydrocarbons to gases with lower densities [47]. Energetically, high quality gases can be obtained from pyrolysis of PE whose gross calorific value at 500 °C is of 86.58 MJ  $m_N^{-3}$  thanks to the relatively high contents of energy-rich higher alkanes and alkenes [47].

#### 2.8.2 Crosslinked-polyethylene pyrolysis

Waste cross-linked polyethylene (PEX or XLPE) generated in electric cable industry shows a good potential as energy recovery source because of its high energy density of around 47 MJ kg<sup>-1</sup> that can be converted to syngas using pyrolysis [36]. However, while numerous studies have been carried out on recycling and thermochemical conversion of non-crosslinked polyethylene, little information is available on the pyrolysis of XLPE, often limited to TGA studies, concluding that the main degradation stage should be in the temperature range of 395-503 °C [66]. This temperature range has been derived for irradiation cross-linked polyethylene and probably depends on the type of PEX investigated, but there are no studies about this influence.

In a pyrolysis study carried out in a semi-batch reactor at high temperatures in the range of 973-1173 K, it was shown that increasing the temperature increased the gaseous hydrocarbon yield, in particular the production of methane and C2 gases (acetylene, ethylene, ethane) due to enhanced thermal cracking of volatile XLPE oligomers. Figure 2.10 shows the effect of temperature on the composition of the XLPE pyrolysis gas.



Figure 2.10 Effect of temperature on product gas composition from XLPE during pyrolysis [36]

At higher temperatures, the amount of  $H_2$  in the syngas decreased due to the dilution of syngas with gaseous hydrocarbons which reduced the  $H_2$  mole fraction. In particular,  $H_2$  gas yield

increased until 1073 K and then decreased with further increase in the temperature beyond 1073 K, as shown in Figure 2.10. Syngas yield during pyrolysis increased with temperature until 1100 K before reaching a plateau of conversion values around 60%, possibly due to enhanced aromatization and coking at higher temperatures, as shown in Figure 2.11. This shows that below 1100 K, an increase in temperature increased hydrocarbon gas and  $H_2$  production, while above 1100 K, coking and aromatization begin to affect syngas yield negatively. The syngas produced is comparable to natural gas in terms of mass-specific heating value and can therefore be an efficient way of recovering clean energy from XLPE compared to the incineration technology, which is widely used today. However, there is no information regarding the actual heating value of the produced gas. Knowing that the starting plastic has a heating value of around 47 MJ kg<sup>-1</sup>, through the apparent thermal efficiency, which is a measure of the increase in heating value from sample to syngas, it was shown that very high energy could be extracted via conversion to syngas [36].



Figure 2.11 Effect of temperature on syngas yield from pyrolysis of XLPE [36]

#### 2.8.3 Polyvinyl chloride pyrolysis

Polyvinyl chloride is a major polymeric component in low voltage cables, but it is also one of the most problematic plastics for the environment. In fact, upon degradation releases chlorine-containing organics and phthalate plasticizers due to the additives used, contaminating products

of the recycling process [41]. During pyrolysis it releases hydrochloric acid (HCl) which leads to equipment corrosion, even in small quantities. Therefore, because of the corrosive and toxic chlorine component, only few studies were carried out by different researchers who, in most of the cases, decided to exclude PVC from the pyrolysis feedstock components [65].

It was found that pyrolysis of PVC in a batch reactor at temperatures varying between 225 and 520 °C, under vacuum and total pressure of 2 kPa produced a very low yield of liquid oil, which did not significantly increase with temperature, and the main compound obtained was HCl, with the highest yield of 58.2% [67]. Pyrolysis of PVC in a fixed-bed reactor at 500 °C only produced 12.3 wt% of liquid oil and the low liquid yield indicated the high production of gases. In fact, it has been reported that gas production during the pyrolysis of PVC is up to 87.7%, much higher than the liquid yield at 500 °C. Concerning the oil composition, PVC was decomposed to naphthalene and its derivatives around 33.55% [68]. During the pyrolysis of PVC in a Fixed bed reactor at temperature range of 500-700 °C it was observed that an increase in temperature leads to a decrease in the amount of HCl, because the increase of temperature increases the number of radicals available to react with the HCI molecules. The chlorine content in PVC is about 57% and up to 90% of the chlorine is evolved as HCl when pyrolyzed [69].

A second-stage degradation has been proposed for PVC in literature, with two temperature ranges showing different weight losses: the 280-385 °C with the weight loss of 62.25% and 385-520 °C where about 21.74% weight loss occurred [70]. These steps are known as dehydrochlorination of PVC and decomposition of dechlorinated-PVC. As the temperature was raised up till 800 °C, the weight loss of the sample became insignificant (1.62%). Thus, the degradation temperature was assumed to be in the range of 220-520 °C and the activation energy for PVC was reported to be in the range of 136-267 kJ mol<sup>-1</sup>. It also has been reported that in the first stage the release of HCl and other chlorinated hydrocarbons takes place, producing mainly HCl and alkanes, while in the second stage structure-reforming reactions of PVC, such as isomerization, cross-linking, aromatic cyclization of paraffin, are predominant, resulting in alkenes and aromatic compounds [71], [72].

It can be concluded that PVC is not preferable for the pyrolysis process, due to the low liquid yield and HCl production which is corrosive to the equipment. Moreover, the release of HCl and the presence of chlorinated compounds such as chlorobenzene in the pyrolysis liquid may be toxic to the environment and human health as well as depreciate the fuel quality. As a consequence, an additional dechlorination step has to be employed to prevent the generation of chlorinated compounds when PVC samples are pyrolyzed. This pretreatment could be accomplished through different methods such as step pyrolysis and pyrolysis with adsorbents, which are the methods most often reported in the literature. In stepwise pyrolysis, a first low-temperature phase is performed to remove chlorine as HCl, followed by pyrolysis of the sample as in a conventional one-step run. On the other hand, the addition of adsorbents to the sample traps the HCl released by physical and/or chemical adsorption [73].

Regarding the gas fraction, an increase in temperature causes an increase in mass and volumetric gas yield, and compared to other polymers, PVC produces significant amounts of gas, even at a process temperature of 500°C, as shown in Figure 2.9. In particular, in [47] the volume of gas produced by PVC was on average 1.56 times higher than that of other polymers and the mass yield was 1.68 more. The gas produced by PVC contains high amounts of H<sub>2</sub>: its average volume content was 53.59% across all temperatures, rising to 67.87% at a process temperature of 900°C. Thus, a progressive increase of H<sub>2</sub> obtained can be observed with increasing temperature. PVC also produced gases with high levels of alkanes, with an average volumetric concentration of 34.99%, where methane was the main component with average values of 25.86%. With increasing temperature, however, the concentration of alkanes decreased in favor of H<sub>2</sub>. In addition to methane, ethene, ethane, and traces of propane, propylene and butane can be found. Figure 2.12 shows the volumetric proportions of main hydrocarbons in gases from PVC compared with PE. The gas produced by PVC had a low density of 0.58 kg  $m_N^{-3}$  due to its high H<sub>2</sub> content, which has a density of only 0.09 kg  $m_N^{-3}$ under normal conditions; the gross calorific value of the gas produced by PVC is 40.68 MJ  $m_N^{-1}$ <sup>3</sup>, very close to that of natural gas which is 40.52 MJ  $m_N^{-3}$ .





Figure 2.12 Volumetric proportions of main hydrocarbons in gases from PE and PVC. Adapted from [47]

Experiments on the pyrolysis of plastics at different temperatures using a laboratory-scale batch reactor should have been carried out, in order to assess how the relative yields of the different fractions and the main components of the gas phase vary with temperature. Unfortunately, for technical reasons this was not possible, but in this chapter the materials, the experimental setup, and the procedure to be used for future work are described.

# 3.1 Materials

A black high-voltage cable and a green low-voltage cable provided by the company Skagerak Energi were used as experimental materials. The black high-voltage TSFL cable can be used outdoors directly in the ground or in underground ducts, or indoors if covered with fireproof paint.



Figure 3.1 Black high-voltage TSFL cable [74]

As shown in Figure 3.1, regarding its structure, from the inside to the outside there are different layers and materials:

- Conductor: multi-stranded compressed aluminum conductor filled with swelling powder.
- Inner semiconductor: extruded cross-linked semiconductor layer.
- Insulation: extruded, dry-cured polyethylene (PEX).
- **Outer semiconductor**: extruded semiconductor layer.
- Screen: one layer of round, annealed copper wires.
- Swelling strip: a layer of swelling strip to prevent long-term penetration of water.
- **Diffusion barrier**: a layer of aluminum laminate with overlap bonded to the outer sheath.
- **Outer sheath**: polyethylene outer sheath. The outer sheath consists of two layers: the inner layer, the usual outer sheath, is UV-resistant and white to make an optical distinction between this and the outer semiconductor layer, which is black.

The green low-voltage TFXP cable can be used in housing and distribution networks or in open air without additional protection.



Figure 3.2 Green low-voltage TFXP cable [75]

As shown in Figure 3.2, regarding its structure, from the inside to the outside there are different layers and materials:

- Conductor: annealed aluminum multi-wire conductor
- Insulation: PEX
- Twist: four phases are twisted together.
- Inner sheath: polyethylene
- **Outer sheath**: PVC, a microtube is extruded under the outer sheath.

Stripping wire under the inner sheath simplifies stripping. The cable is double insulated.

PE and PEX can therefore be recovered from high voltage cable, PEX and PVC from low voltage cable. The cable information was provided by Nexans, a leader in cable production [76].

### 3.2 Laboratory scale pyrolysis reactor and procedure

A laboratory-scale reactor was fabricated to conduct batch pyrolysis experiments and evaluate some product properties, such as char and tar amount, gas composition and heating value of the different fractions, by varying temperatures. The system consists of a muffle furnace (Nabertherm L 15-11 muffle furnace) used to heat the batch reactor inside, represented by an aluminum capsule. The furnace is equipped with a thermocouple to control the operating temperature, which can be set to the desired value by means of an electronic control system. To ensure an inert oxygen-free atmosphere in the reaction chamber during the experiment, a nitrogen flow is continuously supplied to the system and it can be controlled by a flow measurement device installed on the nitrogen stream. Impingement bottles were initially placed downstream to the furnace to collect tar and condensable gases, but due to the high pressure drop generated, they were replaced by a water-cooled condenser. Figure 3.3 shows the Nabertherm L 15-11 muffle furnace used in USN laboratories.



Figure 3.3 Nabertherm L 15-11 muffle furnace used in USN laboratories [77]

The system has been continuously developed, but due to some technical problems it was not possible to carry out laboratory tests. There were leakages from the reactor flange due to the increase in pressure as a result of gas production from the samples. Adjustment was made by replacing the impingement bottles with a water-cooled condenser. However, a sampling system should be adopted to trap the HCl released from the PVC by allowing the gas to bubble through

a NaOH solution in, for example, the trapping bottles placed on the reactor at the beginning. However, this factor was not considered, and further adjustments and precautions are necessary before analysis with PVC. In any case, the laboratory activity would involve pyrolysis of different polymers obtained from cables, with similar sizes and weights at different temperatures, 500 °C, 600°C and 700°C. The aim was to understand how the relative yields of the different fractions and the main gases components vary in relation to temperature, which has been described in many articles as the most influential parameter in the pyrolysis process [5], [6]. The gas fraction would be taken with syringes to analyze its composition with a gas chromatography and the solid fraction, which was expected to be in small quantities, would be analyzed with a calorimetric bomb to assess its heating value. Based on other experiments done with the same reactor but on biomass samples, the main steps can be deduced. First of all, the oven must be heated to 20 °C above the required temperature so that opening the door when inserting the sample does not cause the temperature to fall below the desired level and compromise the experiment. After ensuring that all the reactor connections to the condenser are correctly tightened, the cooling water flow is opened, ensuring that there are no water leaks. The closing flange of the reactor is gently tightened, the oven is switched on and the nitrogen flow is opened and adjusted it to approximately 0.1 L min<sup>-1</sup>. In the meantime, about 50 grams of sample are weighed on a precision balance and placed in the aluminum capsule that serves as the reactor. Once the oven has reached 20°C above the target temperature, the closing flange is removed and the aluminum capsule with the sample inside is inserted; the door is then closed tightly. At this point, the timer is started, and the nitrogen flow is increased for 3 minutes to remove the oxygen inside the reactor. When smoke formation is observed in the condenser, gaseous samples can begin to be extracted at 5-10 minute intervals with syringes and then analyzed by gas chromatography. Once the pyrolysis experiment is completed, the furnace is switched off and allowed to cool to room temperature, while the nitrogen flow is maintained during the cooling process until 200°C, to avoid possible oxygen ingress and subsequent secondary reactions. Gaseous samples are analyzed with the GC, the tar stuck in the flask is washed with IPA (isopropyl alcohol) and then the collected tar is heated to 105°C for weighing. Once the aluminum capsule is removed, the remaining char can be weighed and analyzed with a calorimetry bomb to obtain the calorific value. Figure 3.4 shows the experimental setup realized in the USN laboratories.

Experimental study



Figure 3.4 Experimental setup carried out in the USN laboratories. Starting from the top left and proceeding clockwise, it is shown: 1. reactor inserted in the furnace with closed flange, 2. reactor inserted in the furnace with open flange, 3. aluminum capsule acting as reactor and 4. water-cooled condenser.

Some observations can be made about the experimental set up. Thanks to experiments on biomass samples, it was found that it takes about 50 minutes to heat the reactor to 500°C, and about ten minutes to pyrolyze the sample. In fact, since it is a flash pyrolysis, the sample is placed directly into the preheated reactor and the pyrolysis process is very fast. Because of this it is not advisable to insert more than 50 grams of sample, because the fast pyrolysis of the sample could lead to high amounts of gas released and higher pressure in the reactor, with the risk of leakage or explosion. The insufflated nitrogen is used to ensure an inert atmosphere throughout the experiment so that the sample is pyrolyzed and not burned. It therefore serves to avoid secondary reactions between the sample and oxygen, but also between the gas released by pyrolysis and the tar or carbonaceous residue produced, reducing the residence time of the gas in the reactor. In addition, enough nitrogen is needed to push out the liquid and avoid more condensation of vapors in the exit pipe from the reactor before the water-cooled condenser. Therefore, most of the liquid must be collected within the condenser and the flask at its base. Since some of the tar settles on the walls of the condenser, it is a good idea to wash the condenser walls with 10-15 mL of IPA immediately after the liquid is spilled, to prevent it from solidifying and collect it completely in the flask. The tar and isopropyl alcohol thus collected

are placed inside the reactor at a temperature above the boiling temperature of the solvent, which is about 82°C to evaporate the solvent and weigh the pure tar.

Regarding sample preparation, cables should be disassembled manually with a hand cutter and/or wire stripping tool in order to isolate plastic portions such as the outer plastic layer and metals. The materials should be separated by visual identification into three fractions such as plastic, metals and others. The resulting plastic material should be cut into small pieces of approximately the same size and washed well with a mild detergent solution and then with deionized water to remove dirt and oily materials attached.

### 3.3 Mass balances and energy balances

In the experiments conducted with the pilot-scale pyrolysis reactor, initial feedstock material has to be weighted prior to each run. The collected end-products, the char left in the aluminum capsule and the tar, have to be weighted separately after collections. Assuming negligible losses from condensation in the pipes, a mass balance can be used to deduce the amount of non-condensable gas produced by the pyrolysis process, as written in equation 3.1:

$$M_{non-condensable gases} = M_{feedstock} - M_{liquid} - M_{solid}$$
(3.1)

The percentage yield of the pyrolysis liquid and char collected can be calculated by the following equations:

$$Liquid yield (\%L) = \frac{M_{liquid}}{M_{feedstock}} \times 100$$
(3.2)

Solid yield (%S) = 
$$\frac{M_{solid}}{M_{feedstock}} \times 100$$
 (3.3)

Then the total percentage of gas produced during the experiment can be calculated by:

$$Gas yield (\%G) = 100 - (\%L + \%S)$$
(3.4)

The calorific value of plastics and solid char can be measured using an automatic calorimeter. With knowledge of the mass and composition of the gas (measured using GC), the fraction of energy transformed into gas can be calculated. In fact, the gross calorific value of a pyrolysis gas can be evaluated by the following equation [47]:

$$\hat{H}_{S} = \frac{1}{z_{G}} \sum_{i=1}^{i=n} \hat{H}_{S,i} \frac{\Omega_{i}}{100}$$
(3.5)

where n is the total number of compounds detected in the gaseous mixture and  $\Omega$  is the volumetric fraction of the i-th component [%].  $\hat{H}_{S,i}$ , the gross calorific value of the i-th component in the pyrolysis gas can be obtained from the National Institute of Standards and Technology (NIST) database [78]. Parameter  $z_G$  is the compressibility factor in the equation, which corrects for the behavior of real gases, specifically for the deviation in the volume of a real gas from that of an ideal gas. No pyrolysis gas behaves as an ideal gas; therefore, this factor has to be included in the equation for improved accuracy;  $z_G$  can be determined using the equation [47]:

$$z_G = 1 - \left(\sum_{i=1}^{i=n} \frac{\Omega_i}{100} \sqrt{b_i}\right)^2$$
(3.6)

where *bi* is the "sum factor" whose value for component i is available in, for example, standard ISO 6976 [47].

Then, by taking the difference between the calorific value of the plastic and the heat content of the gas fraction and the char, the amount of energy transferred to the tar can be approximately estimated.

### 3.4 Health and environmental safety

Safety is the prevention of accidents by using appropriate technology to identify hazards in a process and eradicate them before an accident occurs. There are common hazards, chemical or mechanical, that cause injuries from toxic substances, falls and explosion hazards [79]. Health and environmental safety (HSE) must always be a high priority. This laboratory system is a newly built unit and therefore there may be further modifications and problems with its operation. The main problem was the leakage from the closing flange. However, after replacing the impingement bottles with a water-cooled condenser, the problem was overcome. A safety job analysis should be performed before using the equipment, to consider every safety issue that can be of concern for the rig, and how to react if emergency. There are some precautions that must be taken when using the system.

- 1. The plastic sample is placed in a heated oven. Therefore, temperature resistant gloves must be always worn.
- 2. The unit has to be placed inside the chimney which will catch any leakage encountered by the user.
- 3. A maximum of 50 grams of sample has to be used for experiments so that will not generate a large amount of gas at once.
- 4. Nitrogen flow must be at least 0.1 L min<sup>-1</sup> (always check the nitrogen pressure).
- 5. All bolts must be fully tightened on the closing flange.
- 6. Any leakage can be observed from the gas coming out of the oven door or from two outlets at the back of the oven.
- 7. Even if a leak is observed, the oven door must not be opened, and the user has to wait for pyrolysis to complete (this can be decided when there is no smoke inside the condenser).
- 8. The furnace has to be cooled to below 200°C before leaving the laboratory.
- 9. The chimney fan must work (the sound can be heard).

### 3.5 Determination of heating values

Although it was not possible to carry out the experiments with the pyrolysis reactor, the heating value of the different plastics was assessed using an automatic calorimeter. The heating value of a solid, which is equal to the amount of energy released by the complete combustion of a specified quantity [80] can be determined experimentally by an automatic calorimeter. The high heating value - HHV is the amount of heat generated with the total and complete combustion of fuel, at assumption that the combustion products are cooled to the initial temperature of the components involved in the combustion process, and the water condenses [81]. The lower heating value - LHV is numerically equal to the difference between the high heating value and the amount of heat required to the separation of water contained in fuel, and also the water produced in the hydrogen combustion process [81]. So, it is determined when the heat of vaporization of water in the reaction products is excluded.



Figure 3.5 LECO AC -350 Automatic calorimeter used for measuring HHV [80]

The HHV was measured by using a LECO AC-350 Automatic Calorimeter, shown in Figure 3.5. By placing a bomb container, which is a high-pressure atmospheric environment, into a bucket containing exactly 2000 mL of distilled water, the calorimeter can measure the temperature difference before and after ignition to determine the calorific value of the sample. The temperature of the water is then measured using an electronic thermometer with a resolution of 1/10 000 of a degree. The temperature difference in prefire and postfire "modus" is processed by the computer where the result is corrected for the length of the fuse wire. The experiments were carried out according to [80].

As far as sample preparation is concerned, small pieces of the various plastics from the cables were initially cut up, but for analysis with the automatic calorimeter, compact pellets are needed, so larger pieces of around 0.5-0.6 grams were cut up and analyzed. The limitation on mass comes from the fact that they are compounds with a high hydrocarbon content and if too much high-energy mass is burnt, the calorimeter bomb may explode. The plastic sample was weighed directly into the crucible on a precision balance. Then the crucible with the sample inside was placed on the sample holder of the calorimeter bomb and a 10 cm fuse was placed between the electrodes and was used to ignite the sample. After inserting 5 ml of distilled water into the calorimeter bomb to obtain saturated steam, the sample holder was placed inside the

bomb which was then closed. Oxygen was then placed inside the bomb, which occupied the entire available volume and pressurized the bomb. After inserting 2000 ml of distilled water into the combustion chamber, the bomb was inserted. The calorimeter was closed, and the analysis started: the first three minutes were used to stabilize the temperature, after which the actual analysis took 5 minutes. Once the analysis is complete, the bomb is placed under a fume hood, depressurized and the length of the unburned wire is measured to adjust the calorific value actually obtained.

In this chapter, the results of experiments with the automatic calorimeter are presented, as well as some observations on the trends that can be expected from laboratory tests with the reactor.

# 4.1 Feedstock properties

Samples from each polymer layer of both cables were taken and analyzed with the automatic calorimeter, to better understand the pyrolysis process. From the high-voltage cable, a sample of the semi-transparent polymer layer, which was assumed to be cross-linked polyethylene (PEX - HV), and a sample of the black outer shell, which was assumed to be polyethylene (PE), were taken. From the low voltage cable, a sample of the grey polymer layer, which was assumed to be cross-linked polyethylene (PEX - LV), and a sample of the grey polymer layer, which was assumed to be polyethylene (PEX - LV), and a sample of the grey number of the grey polymer shell, which was assumed to be polyethylene voltage (PEX - LV), and a sample of the green outer shell, which was assumed to be polyethylene (PEX - LV), and a sample of the green outer shell, which was assumed to be polyethylene voltage assumed to be polyethylene (PEX - LV).

| Sample | Mass [g] | HHV [MJ kg <sup>-1</sup> ] |
|--------|----------|----------------------------|
|        | 0.5674   | 16.327                     |
| PVC    | 0.5486   | 16.159                     |
|        | 0.5797   | 16.258                     |
|        |          |                            |

Table 4.1 HHV experimentally obtained for PVC samples

Table 4.2 HHV experimentally obtained for PE samples

| Sample | Mass [g] | HHV [MJ kg <sup>-1</sup> ] |
|--------|----------|----------------------------|
|        | 0.6022   | 45.301                     |
| PE     | 0.5678   | 45.235                     |
|        | 0.5702   | 45.546                     |

| Sample   | Mass [g] | HHV [MJ kg <sup>-1</sup> ] |
|----------|----------|----------------------------|
| PEX - HV | 0.5942   | 44.924                     |
|          | 0.5590   | 45.319                     |
|          | 0.5597   | 45.241                     |

Table 4.3 HHV experimentally obtained for PEX - HV samples

Table 4.4 HHV experimentally obtained for PEX - LV samples

| Sample   | Mass [g] | HHV [MJ kg <sup>-1</sup> ] |
|----------|----------|----------------------------|
|          | 0.5752   | 45.731                     |
| PEX - LV | 0.5036   | 45.561                     |
|          | 0.5455   | 45.341                     |

A weighted average can be calculated for the high heating value of each material, considering the different sample weights, using the equation 4.1:

$$HHV average = \frac{\sum_{i=1}^{N} HHV_i w_i}{\sum_{i=1}^{N} w_i}$$
(4.1)

Where N is the total number of measurements for a given material,  $HHV_i$  the calorific values of each measurement and  $w_i$  the different weights in each measurement. Table 4.5 shows a comparison between the averaged HHV experimentally obtained and the values reported in literature for each material.

| Sample   | HHV average [MJ kg <sup>-1</sup> ] | Theoretical HHV [MJ | kg <sup>-1</sup> ] |
|----------|------------------------------------|---------------------|--------------------|
| PVC      | 16.249                             | 16 - 20             | [82]               |
| PE       | 45.360                             | 40 - 45             | [83]               |
| PEX - HV | 45.156                             | 47                  | [36]               |
| PEX - LV | 45.547                             | 47                  | [36]               |

Table 4.5 Comparison between the averaged HHV experimentally obtained and the values reported in literature for each sample

In the literature, the calorific value of PVC is reported to be in the range of 16-20 MJ/kg, depending on the content and type of additives present [82], while the calorific value of polyethylene is reported to be between 40 MJ kg<sup>-1</sup>, corresponding to LDPE, and 45 MJ kg<sup>-1</sup>, corresponding to HDPE [83]. The calorific value of PEX has been reported in an article to be around 47 MJ kg<sup>-1</sup> [36].

It should be noted that the PVC samples remained partially melted in the crucible: the material was not completely burnt, so a slightly higher HHV value can be expected than the one obtained. In any case, comparing the measured values with those reported in the literature, it can be assumed that the material is PVC, although it is not possible to determine the plasticizers present without further analysis of the material. In addition, PVC is corrosive to the calorimetry bomb even after the first analysis.

Regarding the sample taken from the outer shell of the high voltage cable, we were not sure if it was PE, it could also be a halogen-free fire-retardant compound that would give off smoke and annoying/acidic/corrosive exhaust gases as side effects in case the cable is involved in a fire. This is because in a first analysis we got a lower calorific value, around 24 MJ kg<sup>-1</sup> and we wondered if it was polybutylene terephthalate, whose calorific value is reported to be 28 MJ kg<sup>-1</sup> [84]. Further analysis resulted in a calorific value compatible with the one reported in the literature for polyethylene. The lower result was probably obtained after an analysis with PVC, which has a lower calorific value, and which affected the subsequent analysis.

In one article, PEX was reported to have a calorific value of 47 MJ kg<sup>-1</sup>, although it was experimentally found to be slightly lower, at around 45 MJ kg<sup>-1</sup>. It should be noted that it was not possible to separate the semi-conductive layer from the PEX sample and this may have affected the measurement. In any case, being cross-linked polyethylene, one would expect a value around the highest value for the calorific value reported for polyethylene, which was actually obtained.

### 4.2 Effect of temperature on relative product yields

Unfortunately, it was not possible to conduct experiments on the pyrolysis reactor due to technical problems. The idea was to place the sample to be analyzed in an already heated environment, resulting in a fast pyrolysis of the material and a fast increase in pressure inside the reactor. As a result, there were leakages from the closing flange of the reactor due to the increase in pressure as a result of gas production from the samples. The system was continually developed and finally with the replacement of the impingement bottles by a water-cooled condenser, the problem was overcome. In any case, the aim was to perform heat treatment of PE, PEX and PVC in a batch reactor under atmospheric pressure at three different temperatures, 500 °C, 600°C and 700 °C, and based on the literature we can expect to achieve certain trends. An increase in temperature causes an increase in the density of the oil produced because of the reducing the light hydrocarbon fraction due to the conversion of volatiles into light hydrocarbons and non-condensable gases [47], [64]. Increasing the operating temperature also leads to a decrease in char production [61].

Polyethylene is the polymer with the highest oil production (up to 84.7%) and the highest liquid yield occurs around 550 °C, so we can expect a decrease in the oil production between 600 and 700 °C [59], [65]. Regarding the composition of the gas fraction, the main hydrocarbons are expected to be methane, ethane, propylene, butane and 1-butene, with an increase of methane, ethylene, and propylene in the range between 500 and 700 °C. There should then be a decrease in hydrogen produced at 700 °C, probably due to secondary reactions that facilitate the production of hydrocarbons [47].

There are no studies on the pyrolysis of PEX in the temperature range considered in this work, the only one reported in literature [36], to the knowledge of the author, shows pyrolysis of PEX between 700 and 900°C, so we cannot rely on the literature to analyses the trends of the various compounds. What we can expect is the presence of hydrogen, methane, acetylene, and ethylene in the gaseous fraction, as well as an increase in gas production by increasing the temperature. Further observations will require experimental analysis.

PVC would have been the most difficult material to study, due to the release of HCl that occurs during its degradation, corroding the reactor and not allowing the other components of the gas fraction to be easily identified. Therefore, it would have been absorbed downstream of the process with a 0.1 M NaOH solution in the impingement bottles. According to the literature, PVC produces a low oil yield and a large amount of gas, even at the relatively low operating temperature of  $500^{\circ}$ C [44], [68]. The main expected components are hydrogen and methane, followed by ethene and ethane. As the temperature increases, there should be an increase in the gas fraction and a slight increase in the tar produced, with a reduction in char. The amount of char, however, depends on the additives and plasticizers used, which generally increase the char yield. At the same time, with regard to the composition of the gas phase, there should be a progressive increase in H<sub>2</sub> at the expense of the hydrocarbon content.

Conclusion

# **5** Conclusion

The main objective of this study was to analyze the problems related to the recycling of waste cables, which are an increasingly important part of electronic waste. Structure and materials commonly used in cable industry were analyzed, with particular emphasis on polymeric materials. It is well known that after the separation of metals and plastics, the former are recycled for further use, while the latter are considered as residue and incinerated and/or landfilled. In order to reduce the economic and environmental impact of these operations, more and more studies are focusing on the pyrolysis of plastics to obtain alternative fuels.

An extensive literature search has therefore been carried out on the pyrolysis of the main polymeric materials used in cables, such as polyethylene, cross-linked polyethylene, and polyvinyl chloride. It was concluded that polyethylene is the preferred material as feedstock for the pyrolysis process, while PVC shows several challenges, such as the production of chlorinated compounds and HCl, which cause corrosion problems in the reactor and depreciate the oil obtained. Unfortunately, research on PEX is still in its infancy and little information is available. The original plan included a series of experiments on pyrolysis of the analyzed materials at different temperatures using a laboratory-scale batch reactor, in order to assess how the relative yields of the different fractions and the main components of the gas phase vary in relation to temperature. Unfortunately, for technical reasons this was not possible, but the procedure to be used for future work was described.

Further studies on this topic are necessary and could analyze by means of laboratory experiments how the relative yields of the different fractions and the main components of the gas phase vary as a function of temperature, residence time and particle size in the reactor. In addition, the effect of pressure could be considered, which is still poorly described in the literature. Improvements could be made to the reactor, for example modify the door of the furnace and insert a spout with a valve for sample feeding, without having to open the reactor every time with risks for the operator due to high temperatures.

A further development could be to focus on the effect on pyrolysis products of the numerous additives in polymers from the cable industry, a factor not investigated in this study.

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# Appendices

#### Appendix A Task description

University of South-Eastern Norway Faculty of Technology, Natural Sciences and Maritime Sciences, Campus Porsgrunn FMH606 Master's Thesis Title: Closing the plastic waste material cycle in the power industry Assoc. Prof. Marianne S. Eikeland (main supervisor) **USN supervisor:** PhD Student Janitha Bandara External partner: Skagerak Energi AS v/ Finn Werner Bekken Task background: The power industry produces plastic waste. Since the 70s, plastic has been used as insulation material in cables. As cable systems reach their lifetime, discarded cables must be handled in a responsible manner. The metal is stripped for insulation and recycled. The plastic insulation can be used as fuel, but should be recycled from an environmental perspective. The amount of waste will eventually become significant and can reach a few tens of thousands of tonnes per year in Europe. Polymers utilized in the power industry include polyethylene and PVC for cable insulation, glasfiber reinforced polyester and/or epoxy in windmill rotor blades, epoxy and silicium for high voltage apparatus such as generators and surge arrestors. Pyrolysis technology transforms plastics and other organic waste into light gases and oils which can be useful raw material for fuels and/or chemical/polymer production. Plastics pyrolysis by electric heating is an emerging technology. This project shall compare thermal pyrolysis technologies with electrified pyrolysis technology (e.g. microwave, electric resistance heating) for conversion of waste plastic to chemical raw material. Energy and material balances need to be estimated with special focus on energy consumption. Task description: The thesis includes the following tasks: A detailed literature review to establish a knowledge base for handling plastic waste from the power industry. Analysis thermal and electrified pyrolysis technologies for plastic waste. Comparison of the respective technologies with respect to energy consumption and environmental sustainability. Perform experimental analysis with pyrolysis of polymeric materials from the power industry

- to be studied: char, tar, gas composition, heating value and ash content by varying temperatures and retention times.
- Analysis of mass- and energy balances.

#### Student category:

PT and EET students.

#### Practical arrangements:

#### Supervision:

As a general rule, the student is entitled to 15-20 hours of supervision. This includes necessary time for the supervisor to prepare for supervision meetings (reading material to be discussed, etc).

Signatures:

Keland, 01.02 2021 Marianne

Supervisor (date and signature):

ALESSIA FOGLIA Student (write clearly in all capitalized letters):

Alessie Forfia Student (date and signature):

01-02-2021