



KTH Engineering Sciences

Processing of self-reinforced poly(ethylene terephthalate) composites for automotive applications

LARS JERPDAL

Doctoral Thesis
Stockholm, Sweden 2017

TRITA AVE 2017:94
ISSN 1651-7660
ISBN 978-91-7729-651-5

KTH School of Engineering Sciences
SE-100 44 Stockholm
SWEDEN

Akademisk avhandling som med tillstånd av Kungl Tekniska högskolan framlägges till offentlig granskning för avläggande av teknologie doktorsxamen i farkostteknik FREDAG den 26 Januari 2018 klockan 9:00 i Kollegiesalen, Brinellvägen 8, Kungliga Tekniska Högskolan, Stockholm.

© Lars Jerpdal, 2017

Tryck: Universitetservice AB

Abstract

The vehicles of the future must have less negative environmental impact during their use phase than the vehicles of today in order to avoid major climate change on earth. Consequently electric vehicles are currently under development with the purpose of reducing CO₂ emissions when the vehicle is driven. There are also efforts put in to reducing the weight of vehicles in order to reduce the demand for energy to drive them. One important aspect of weight reduction is that new materials and technologies are developed.

Plastic materials have low a density and can therefore be used to reduce the weight of vehicle components and with composite materials there is further potential for weight reduction. Traditional composite materials are however difficult to recycle and therefore often end up as landfill at end-of-life, which is negative for the environment.

Self-reinforced thermoplastic composite materials are materials in which both reinforcement and matrix are thermoplastic materials and therefore theoretically easy to recycle. Thanks to their low density and relatively good mechanical properties, these materials may be used for weight reduction of vehicle components. A component made of self-reinforced poly(ethylene terephthalate) can be manufactured through pressing into a three-dimensional geometry. During this forming process, the material is exposed to different degrees of stretching. How this stretching and other parameters in the manufacturing process influences the final properties of the component is important knowledge that is required in order to reach a robust process for serial production.

The aim of this thesis is to study selected process parameters for component manufacturing with self-reinforced poly(ethylene terephthalate) (SrPET) in order to increase knowledge and thereby advance the field of self-reinforced PET composites.

This thesis shows that stretching the material in the manufacturing process increases the mechanical performance of the material due to increased orientation of the amorphous phase in the PET reinforcement. However, stretching introduces stresses in the material that give rise to negative shape distortions in the formed component. When the component is exposed to higher temperatures at later stages after the manufacturing process, these stresses can be released which reduces the tensile modulus and influences the dimensions. The degree of stretching during forming must therefore be controlled in order to achieve a robust serial production, and relaxation of residual stresses must be considered in the phases following manufacturing.

The concept of a SrPET component over-moulded for integration of stiffeners and attachments has been evaluated in a life-cycle-assessment. This evaluation shows that the component weight can be reduced compared to technology currently in use and thereby contribute to increased sustainability of transport.

Sammanfattning

Framtidens fordon måste ha mindre negativ påverkan på miljön under användningsfasen jämfört med dagens fordon för att förhindra stora klimatförändringar på jorden. Därför sker idag ett omfattande utvecklingsarbete dels med att elektrifiera fordon i syfte att minska CO₂ emissioner när fordonet körs, men även ett arbete med att minska fordonsvikten för att på så sätt minska energibehovet för att köra fordonet. En viktig del i denna utveckling är framtagning av nya material och tillverkningstekniker för fordonskomponenter.

Plast är ett material som tack vare låg densitet kan användas för att reducera vikten på fordonskomponenter och med plastkompositer finns ytterligare potential till viktreducering. Traditionella kompositmaterial har dock problem med återvinning eftersom det enda alternativet efter användning är att deponera hela eller delar av materialet, som därmed ger en negativ inverkan på miljön.

Självförstärkta kompositer är material där både förstärknings- och matrismaterial är av samma typ av plastmaterial och därmed teoretiskt sett enkla att återvinna. Tack vare låg densitet och relativt goda mekaniska egenskaper hos dessa material kan de möjliggöra viktreduktion för t.ex. fordonskomponenter. En komponent kan tillverkas genom pressning till en tre-dimensionell geometri. Denna pressning innebär att materialet sträcks under formningsförloppet. Kunskap om hur parameterar i tillverkningsprocessen påverkar materialtes slutliga egenskaper i en komponent är viktigt för att uppnå en robust serieproduktion.

Syftet med denna avhandling är att undersöka utvalda processparametrar för tillverkning av komponenter med självförstärkta poly(ethylene terephthalate) (SrPET) för att öka kunskapen om komponenttillverkning och därmed utveckla forskningsområdet med självförstärkta PET kompositer.

Avhandlingen visar att sträckning av material vid komponenttillverkning ökar den mekaniska prestandan hos materialet genom att det sker en ökning av orienteringen hos de amorfa delarna i materialet. Sträckning ger dock upphov till negativa formförändringar hos en formad komponent pga spänningar som byggs in i materialet. Vid exponering för höga temperaturer i senare skeden efter formningsprocessen kan de ökade mekaniska reduceras pga av den orienterade amorfa fasen inte är temperaturstabil. Komponentens form kan också påverkas till följd av att inbyggda spänningar relaxerar. För att uppnå en robust tillverkningsprocess måste därför sträckningsgraden vid formning kontrolleras samt relaxation av eventuella restspänningar beaktas i senare faserna efter formningsprocessen.

Avslutningsvis har SrPET analyserats ur ett livscykelperspektiv vid applicering på en fordonskomponent. Denna analys visar att ett koncept där SrPET formas till en komponent och övergjuts genom formsprutning kan ge en viktminskning i jämförelse med aktuell metod vilket ger en sänkning av miljöpåverkan och därmed bidrar till att öka hållbarheten för transporter.

Acknowledgment

The work presented in this thesis was carried out within the centre for ECO2 Vehicle design at the department of Aeronautical and vehicle engineering at KTH Royal Institute of Technology. The support from my employer during this project, Scania CV and Volkswagen AG, are gratefully acknowledged. It has been interesting, fun and challenging to work close to academy parallel with industry along this project.

I would like to give a special thanks to Christer who introduced me to the department of light weight structure at KTH and this project. Thanks also to Ola and Sara who supported me and made it possible for me to work on this project as a industrial PhD student. During the project a number of people have been involved and contributed to this work and to whom I am very grateful. To my supervisors Malin and Daniel, thank you for your guidance, support and fruitful discussion along the project, wherever I have been located. To Christof it has been great to get know you and sharing problems with you during this project. During my time in Germany, thanks to Jörg and Olaf and at KTH thanks to all employees at Lightweight structures and the centre of ECO2.

Last but not least a grateful thanks to my family giving a contrast to every day work. To my lovely wife Linda, thanks for your endless support and to my lovely children Gustav, Elias and Filip for supporting dad going to school. I love you!

Wolfsburg, December 2017

Dissertation

This doctoral thesis consists of an introduction to the area of research and the following appended papers:

Paper A

Jerpdal L, Åkermo M. Influence of fibre shrinkage and stretching on the mechanical properties of self-reinforced poly(ethylene terephthalate) composite. *Journal of Reinforced Plastics and Composites* Comp. 2014;33:1644-1655.

Paper B

Jerpdal L, Ståhlberg D, Åkermo M. Influence of fibre stretching on the microstructure of self-reinforced poly(ethylene terephthalate) composite. *Journal of Reinforced Plastics and Composites* 2016;35(22):1634-1641.

Paper C

Jerpdal L, Åkermo M, Ståhlberg D, Herzig A. Process induced shape distortions of Self-reinforced poly(ethylene terephthalate)composites. Submitted for publication

Paper D

Jerpdal L, Schuette P, Åkermo M, Ståhlberg D. Numerical and experimental investigation on over-moulding of self-reinforced poly(ethylene terephthalate). Manuscript.

Paper E

Poulikidou S, Jerpdal L, Björklund A, Åkermo M. Environmental performance of selfreinforced composites in automotive applications - Case study on a heavy truck component. *Materials and Design* 2016;103:321-329.

Conference proceeding not included in the thesis:

Jerpdal L, Schiwik J-M, Åkermo, M. Self-reinforced Poly(ethylene terephthalate) (PET) organo sheets; Properties and applications. In: Tagungsband 2. Niedersächsisches Symposium Materialtechnik. February 23-24, 2017: Clausthal-Zellerfeld, Germany.

Jerpdal L, Hain J, Täger O. Leichtbau für mehr Nutzlast - Neue Wege mit Kunststoff-Verbunden und Thermoplast-Schaum-Spritzguss. In: Proceedings of, Kunststoffe in Nutzfahrzeugen 2017. March 29-30, 2017: Mannheim, Germany.

Contents

Acknowledgment	v
Dissertation	vii
I Introduction	1
1 Background and context	3
2 Polymers	8
3 Self-reinforced polymer composites	19
4 Recycling of plastics	26
5 From materials research to product development	28
6 Objective and scope	30
7 Summary of appended papers	31
8 Conclusion	34
9 Contribution to the field	35
10 Future work	36
Bibliography	37
II Appended papers	45

Part I

Introduction

1 Background and context

Transportation is a significant aspect of life that makes it possible for people to go to places for work, for pleasure and to conduct business. Transports are also necessary in order to move cargo according our needs, and have become more important in our modern society based on consumption and economic growth.

The vast majority of all transports are currently undertaken by vehicles powered by hydrocarbon fuel driven combustion engines, which emits carbon dioxide (CO_2), nitrogen oxides (NO_x) and other substances that are hazardous to the environment and our health. CO_2 emissions have become a serious problem since it is the main source of global warming, or climate change [1]. CO_2 is a greenhouse gas (GHG) that absorbs and emits radiation in the thermal infrared range and is consequently a fundamental cause of the greenhouse effect, which increases the temperatures on Earth. Rockström et. Al. [2] have introduced an approach to global sustainability in which they define planetary boundaries for different environmental factors including climate change. It is proposed that the concentration of CO_2 in the atmosphere should not exceed 350 ppm in order to minimize the risk for an irreversible change of global warming and to avoid a collapse of major climate pattern. The current concentration of 387 ppm is, as illustrated in Figure 1, beyond the proposed lower limit but in the zone of uncertainty with an increasing risk, meaning that the CO_2 emissions from transports and other sectors must be reduced if we are to continue to operate safely on earth.

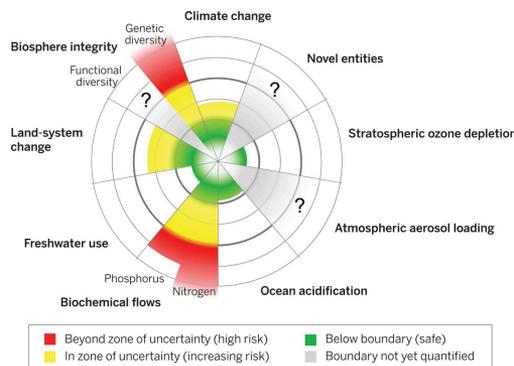


Figure 1: *Current status of control variables for seven of the planetary boundaries [3]*

For passenger cars CO₂ emissions are measured as grams per kilometre driven (g CO₂/km), and was in 2016 in average 118.1 g/km for a new car sold in Europe [4]. For freight vehicles CO₂ emissions are measured as grams per tonne transported goods and kilometre (g/tkm) and depend heavily on the goods filling rate. The CO₂ emissions for a heavy truck can therefore vary from 20 g/tkm up to several kilograms per tonne-kilometre [5]. The sum of all CO₂ emissions generated from ground transports in Europe was 835 million tonnes in 2014, which is about a quarter of the total CO₂ emissions in Europe [6]. Transportation is the only sector that has increased its amount of CO₂ emissions compared 1990 levels [6], and the expectation is that it will continue to grow. The European Commission have estimated that passenger transports will grow by more than 50% and freight transports by 80% until 2050 compared to 2013 levels [7]. Therefore the strategy from the European Union (EU) is to reduce CO₂ emissions generated by transports in Europe. One of the actions included in this strategy is stronger regulations for emissions that forces vehicle manufacturer to develop more energy-efficient vehicles. From 2020 the average CO₂ emission of the fleet from a manufacturer may not exceed 95 grams CO₂ per kilometre [8]. If this limit is exceeded, the manufacturer will pay an excess emissions premium for each car registered. Similar regulations for freight transport vehicles will most probably follow.

The CO₂ emission from a vehicle depends on powertrain efficiency and driving resistances. As shown in Figure 2, the main parameters for driving resistance are aerodynamic drag, rolling resistance and acceleration, the latter depending on the mass of the vehicle. Powertrain efficiency can be improved in order to reduce fuel consumption; however physical laws limit the improvement of these parameters. Consequently, a technology shift towards electrified powertrains has been chosen by the automotive industry in order to reach the new more stringent CO₂ regulations.

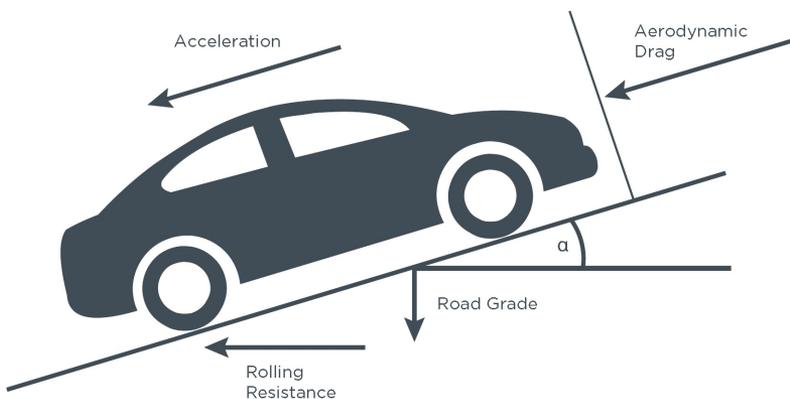


Figure 2: *Resistance forces affecting a moving vehicle.* [9]

The different types of electric powertrains currently developed are, as illustrated in Figure 3, hybrid Electric Vehicles (HEV) including Plug-in Hybrid Electric Vehicles (PHEV), Battery Electric Vehicle (BEV) and Fuel cells (FCV). HEV and PHEV combine an internal combustion engine (ICE) with batteries. The batteries are charged by the ICE and through energy recovery at braking. The batteries in PHEVs can additionally be charged at charging stations or at home. A BEV has batteries as its single power source and is also charged at stations or at home while an FCV produces its own electricity from N_2 . Even though the CO_2 emissions from these vehicles are reduced during use, there can be CO_2 emission related to the production of electricity for example if electricity is produced by burning fossil fuel. In addition to CO_2 , toxic rare materials are used for production of batteries and electric components, which also gives negative environmental impact. If the CO_2 emissions are merely moved from one source or life cycle phase to another, this would just be a way of fooling ourselves that electrical vehicles are more environmental friendly as compared vehicles with conventional ICE powertrain. In a technology shift it is therefore vital to assess the environmental impact of the complete life cycle from manufacturing to End-Of-Life (EOL), and this is why Life Cycle Assessments (LCA) are used.

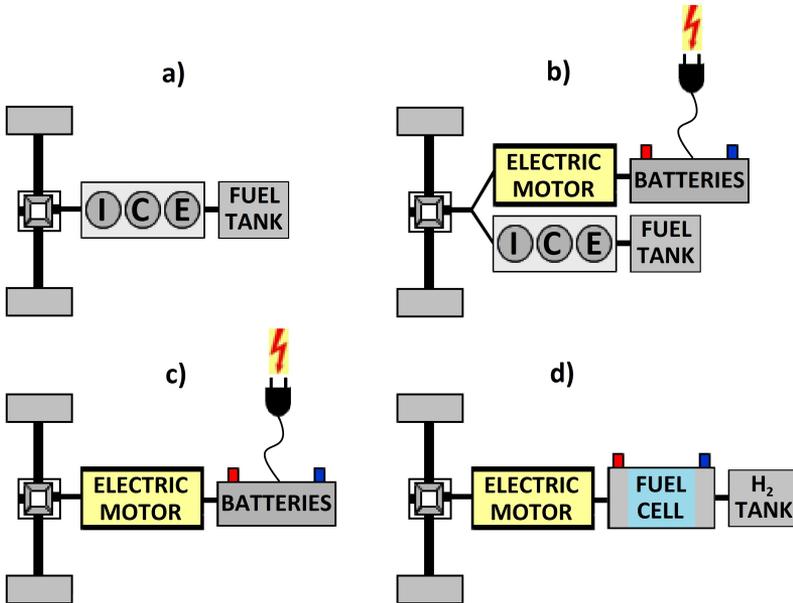


Figure 3: Classification of vehicle power sources. a) Conventional vehicle with internal combustion engine (ICE), b) Hybrid Electric Vehicle (HEV and PHEV), c) Battery Electric Vehicle (BEV), d) Fuel-Cell Electric Vehicle

The increase of energy efficiency through weight reduction applies to all vehicles regardless of power source. Lightweight design using polymer composites have been successful for aircrafts in order to reduce weight, mainly through the use of carbon fibre reinforced polymer composites (CFRP). CFRP possess excellent stiffness and strength to weight ratio and therefore are very attractive for weight critical structures. The most modern aircrafts are designed with 50% of the structural weight made of CFRP which significantly reduces fuel consumption [10]. A similar approach is used for high performance sport cars made in small series to improve performance, and recently also for larger serial production of standard cars to improve energy efficiency [11]. The body in white (BIW) can be made with a monocoque design of CFRP as shown in Figure 4, and other metal parts can be replaced with components of composite materials in order to reduce weight. However, one general disadvantage for polymer composites is that raw material production requires considerable amount of energy, especially the production of carbon fibres, and recycling opportunities for most composite materials are limited and thereby having a negative environmental impact [12]. In order to assess total environmental impact for a new material or technology for lightweight design it is therefore essential to examine the complete life-cycle.

To date there has been no break thorough with extensive use of polymer composites in ground vehicles as there has been for aircrafts, partly due to cost and the lack of opportunities for high volume production. One current trend for lightweight design within the automotive industry is instead to develop components combining composite materials and metals into multi material parts [13]. Figure 5 shows an example of how composite parts can be combined with traditional metal parts. This is one approach to keep the efficient steel processes and combining materials for optimized tailored performance and lightweight design for high volume production.

Multi-material design is only one example of ongoing research and development into new materials and technologies within the field of material science for the automotive industry. Metals will in the future maintain a dominating position as design material for vehicles, but polymers will be continuously developed to challenge existing technologies and will be important if vehicle design is to contribute to more sustainable transports.



Figure 4: *Passenger car monocoque Body-In-White (BIW) designed with composite materials for weight reduction [14]*

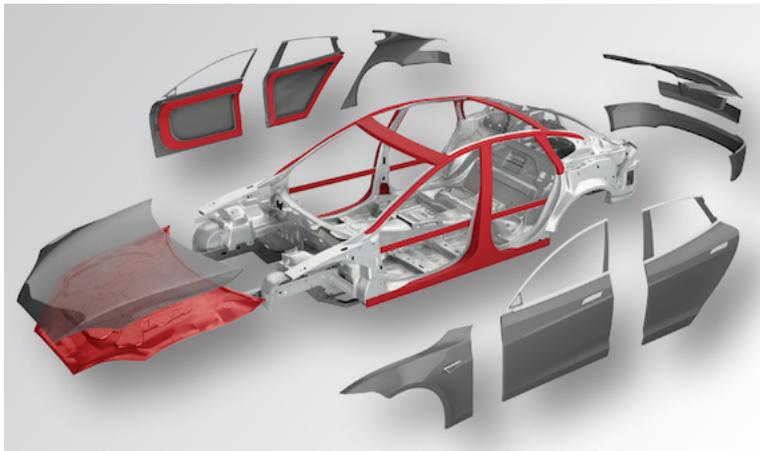


Figure 5: *Multi material hybrid design. Sheet metal (grey parts) in combination with composite materials (red parts) [15]*

2 Polymers

Polymers belong to the modern materials that have been intensively developed over the last century and are important for many product sectors thanks to their wide range of properties and different opportunities for component manufacturing. There are polymers that exist in nature, e.g. natural rubber and cellulose, and there are many different synthetic polymers developed in laboratories and produced industrially.

Structure and properties

Polymers are materials that consists of molecules with many equal repeating units, monomers, bonded to each other in a long molecule chain called macromolecule [16]. The thermal, mechanical and physical properties of polymers are dominated by the chemical structure i.e. which atoms the macromolecules consist of and how they are arranged and bonded to each other within the molecule chain, as well as how the chains are arranged in relation to each other. The atoms within a molecule are bonded to each other with primary covalent bonds while the bonds between molecule chains are weaker secondary van der Waals, Dipole or hydrogen bonds acting as cohesive forces.

The molecule chains can be arranged to each other either in an ordered crystalline structure or randomly in an amorphous structure as shown in Figure 6. The molecules in a crystalline structure are symmetrically arranged and thereby more closely-packed, which results in a stronger bonding force between the chains. The crystalline structure is formed from an unordered structure; however no polymer can crystallize completely, i.e. polymers are either fully amorphous or semi-crystalline with a portion of crystalline structure as also illustrated in Figure 6. The melting of the crystalline structure occurs within a certain temperature range; however a precise melting temperature is defined which can be considered as the temperature at which a perfect polymer crystal would melt. The amorphous structure behaves differently and does not have a defined melting temperature. Polymers in an amorphous state are stiff, hard and can carry load at lower temperatures, in the so called glassy state when the molecule chains are constrained. The mobility of the molecule chains gradually increases with increased temperature and a transition take place from the glassy state into a rubbery state at the glass transition temperature (T_g). When the molecule chains become looser, the material softens and the modulus drops. With further increased temperature the material becomes less viscous and can be considered as a melt.

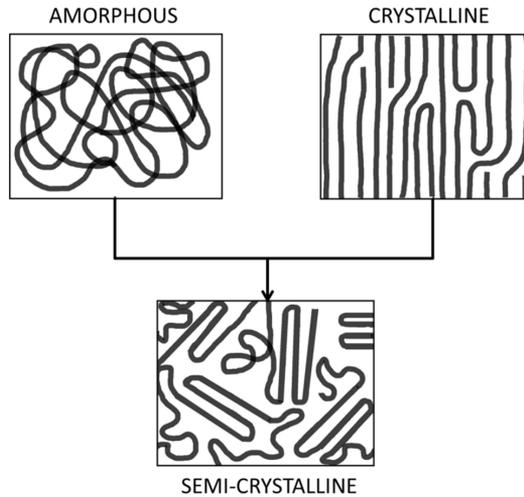


Figure 6: a) *Amorphous structure*, b) *Crystalline structure*, c) *Semi-crystalline structure*

In addition to the amorphous and crystalline arrangement of the molecule chains, there is a possibility for the chains to be aligned and oriented in one specific direction as shown in Figure 7. As the molecules are aligned they can be even more closely-packed and bonded to each other with greater force resulting in a increased performance in that orientation. The ability to be closely-packed is further enhanced by conformational changes of the molecule chains. A conformal change means that the atoms in the molecule are rearranged though rotation in the covalent bonds and make the chain straighter, which promotes chains' ability to move closer to each other.

The properties of polymers can be tailored in the polymerization process by bringing two types of repeating units (monomers) together during polymerization to create a copolymer. Figure 8 illustrates a polymer blend from two polymers A and B (a), and an example of a co-polymer from two polymers A and B (b).

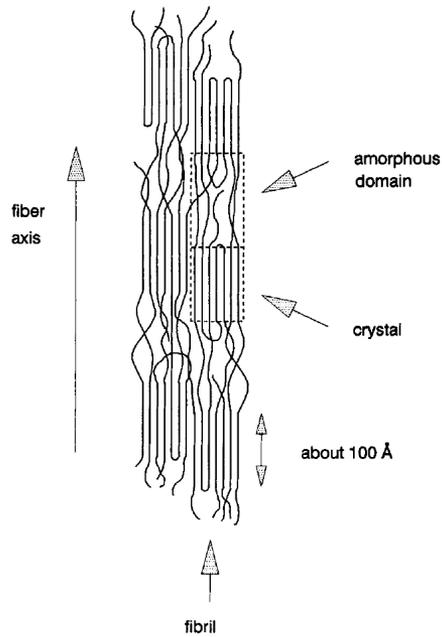


Figure 7: *Oriented molecule structure of a polymer fibre with indicated amorphous and crystal domains [17]*

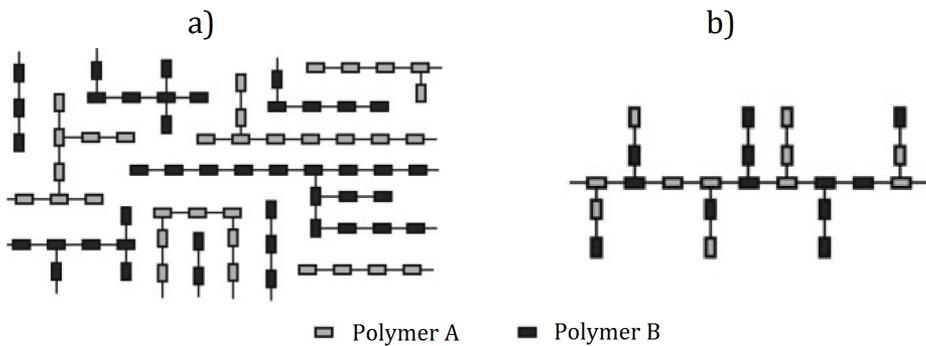


Figure 8: *a) A polymer blend of two base polymers A and B b) Example of a copolymer consisting of two different monomers A and B (Adapted from [18])*

Plastics and composites

Plastic materials are utilized in an increasing number of applications and have become essential to our modern economy thanks to their unique properties that can make the impossible possible. In 2015, a total of 49 million tons of plastic material was produced in Europe and Figure 9a shows the distribution of this amount per sector. The dominating application for plastics is packaging followed by building and automotive. Plastics are important for automotive thanks to low density and high design freedom that enable functional and aesthetical components for efficient high-volume production. This has resulted in that an average passenger car consists of about 10% plastic material by weight [19]. The most basic types of plastic materials are Polyethylene (PE), polypropylene (PP) and polyvinyl chloride (PVC) which has a simple molecule structure and thereby are easy to produce and process for manufacturing of products. This is also why these materials are used in large amounts for many types of applications. Figure 9 b shows the shares for the most common plastic materials used in Europe in all sectors.

Plastics is the denomination for polymers that are modified with additives with additives in order to tailor their properties. An additive may be a substance that increases the stability of the polymer by reducing the degradation caused by high temperatures, UV-radiation or chemical substances. Additives may also be pigments that provide a certain colour, softener to reduce the glass transition temperature or fillers that improve processability. Certain type of fillers may also act as reinforcement in order to increase stiffness and strength. It is common to use fibres from e.g. glass or carbon as reinforcement to create fibre reinforced plastics (FRP). These reinforcement fibres may be of different types and lengths, either as short fibres (1-2 mm) long fibres (up to 40 mm) or as continuous fibres (endless). In a FRP, or composite, the plastic material is referred to as matrix material and the adhesion between the reinforcement and matrix is very important for the performance of the composite material. This is because the reinforcing component must carry the load and the matrix material must distribute the load evenly between the reinforcements.

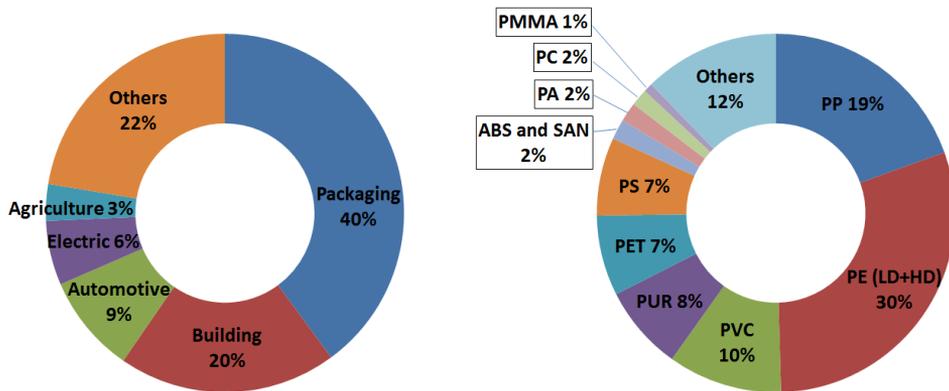


Figure 9: a) The use of plastics per sector. b) Volume share for the ten most used plastic materials. Both statistics for European market in 2015 [20]

Poly(ethylene terephthalate) PET

PET is, as shown in Figure 9 b, by volume the fifth most common polymer and is, in addition to packaging, used for textile fibres and surgical suture thread. PET is a thermoplastic polyester and a semi-crystalline polymer which, due to its slow crystallization, can be quenched from the melt to give a completely amorphous structure at room temperature. PET has a glass transition temperature of 65-80 °C, depending on degree of crystallinity, and a melting temperature of 240-270 °C. A fully amorphous PET has a Tg of 65 °C and Tg increases with increasing degree of crystallinity. PET crystallizes in the temperature range of 10 °C above its glass transition temperature up to 10 °C below its melting temperature, with maximum crystallization rate at 178 °C. PET normally reaches a crystallinity of 40-50%, but can also be polymerized to a co-polymer that cannot crystallize.

Chemistry of PET

PET is synthesized through stepwise growth which is industrially accomplished either by using terephthalic acid (TPA) and monoethylene glycol (MEG) or by using dimethyl terephthalate (DMT) and MEG as raw materials. The PET material formed is a macromolecule with repeating units of an aromatic group (benzene ring, C₆H₆) and six aliphatic groups (-CO-O-CH₂-CH₂-O-CO-) as shown in Figure 10.

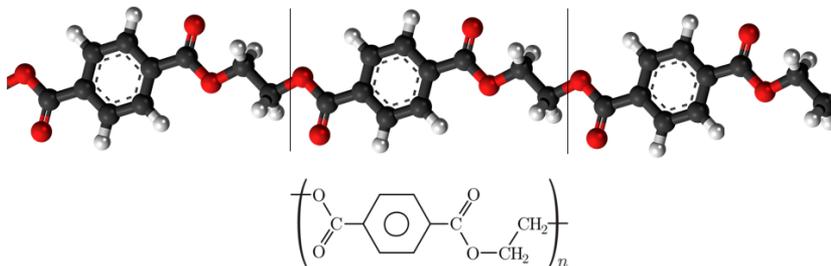


Figure 10: *PET* repeating unit

PET configuration

The configuration of PET, shown in Figure 11, is nearly planar and zig-zag with the benzene rings in the plane of the zig-zag. The repeat unit of PET is 1.075 nm which is slightly shorter than the length of a fully extended chain that is 1.09 nm. This allows chains to be closely packed with cohesion among the chains from the interaction of hydrogen bonds and van der Waals interactions. The aromatic character together with packing and cohesion of PET chains leads to increased modulus, strength and resistance to moisture and solvents.

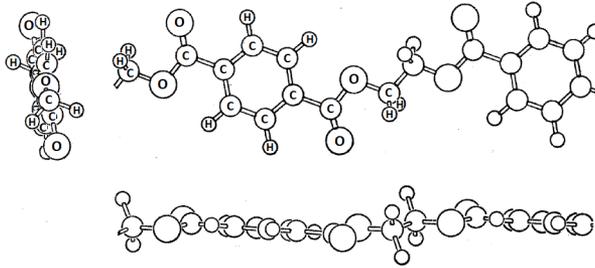


Figure 11: *PET configuration* [21]

The ethylene glycol linkages in PET i.e the linkages to the CH₂ groups, can exist in two rotational conformations; as *trans* (extended form) or as *gauche* (relaxed form) [17]. The transformation from one conformation to the other take place by partial rotation about C-C bonds as illustrated in Figure 12. The *trans* conformation which is related to straight parts of the molecules is mainly present in the crystalline phase of PET while the *gauche* conformation related to disorganized parts only exists in the amorphous phase.

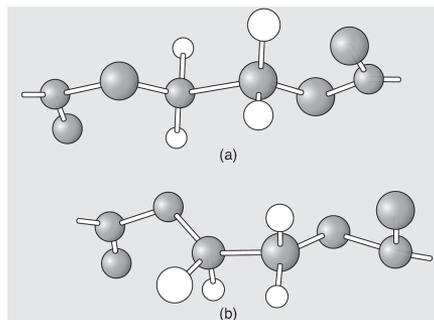


Figure 12: *Rotational conformations of PET, a)trans and b)gauche.* [22]

Polymer fibres

The inspiration and knowledge to create synthetic fibres came from spiders and silkworms. These creatures showed that fibres can be produced from a liquid by forming a jet that is hardened to a solid fibre. The first fully synthetic fibre was produced commercially from Polyamide 6 (PA6) in the 1930s and based on this technology PET fibres were made in the 1940s.

Melt spun PET fibres

There are different methods of making synthetic polymer fibres, melt-spinning being the simplest, most economical and therefore the preferred and most common process for manufacturing fibres from PET and other thermoplastics. In this technique, molten polymer is extruded through narrow channels to form fibres. The fibres that come out of the channels are quenched to solidify and then rapidly pulled and wound onto tube rolls at a speed of 1000 to 7000 meters per minute. The resulting fibre dimensions and properties are determined by several process variables in the spinning process. The two characteristics that influence the properties most are molecular orientation along the fibre and crystallinity [23]. Extrusion of the polymer through the narrow channels contributes very little to molecular orientation because the molecules become disoriented when exiting the channel. Instead the main contribution of molecular orientation derives from the elongation of the fibre during spinning, where the degree of orientation is related to spinning speed. Figure 13 illustrates the development of orientation in a fibre through the spinning process.

Melt spun PET fibres are classified after molecular orientation according to Table 1. Theoretically, the maximum orientation is obtained at spinning speed of 10000 m/min, however surface defects will appear on the fibre somewhere above 7000 m/min which exert a negative influence on its mechanical properties. Higher spinning speed imposes higher level of demand on the melt and spinning equipment in order to achieve higher quality fibres. At higher spinning speeds it also becomes more difficult to control the fibre tension in the wind-up.

Table 1: *Classification of PET fibres with respect to structure/spinning speed [24]*

Denomination	Abbreviation	Spinning speed
Low oriented yarn	LOY	<1800
Medium oriented yarn	MOY	1800-2800
Partially oriented yarn	POY	2800-4000
High oriented yarn	HOY	4000-6000
Fully oriented yarn	FOY	>6000

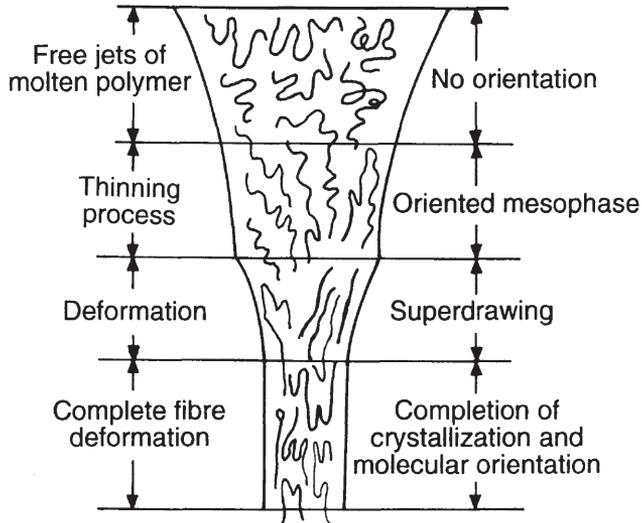


Figure 13: *Development of molecular orientation in a fibre during spinning and drawing [24]*

The molecular orientation of fibres spun at speeds lower than 6000 m/min can be increased through drawing subsequent to the spinning process. Through the drawing operation the molecules are aligned, at the same time as the level of crystallinity can increase and conformational changes can take place [25]. Figure 14 illustrates how the amorphous region of a PET fibre with crumpled molecule segments in gauche conformation changes conformation through drawing into straighter segments with trans conformation. The straighter segments are better aligned to other segments whereof the coherence within the fibre increases and contributes to higher modulus and strength compared to the folded chain segments. The oriented segments that are created through drawing have a more even length distribution compared to the segments created by spinning which has a broader length distribution.

PET can be drawn at room temperature, but in order to obtain a uniform material it is necessary to perform drawing at a temperature close to or above the glass transition temperature of the spun fibre. To make high strength PET-fibres, drawing in two or more stages is recommended with the first stage at around 90 °C and the following draw stage at a higher temperature [25]. Higher drawing temperature requires a higher drawing rate as relaxation will occur.

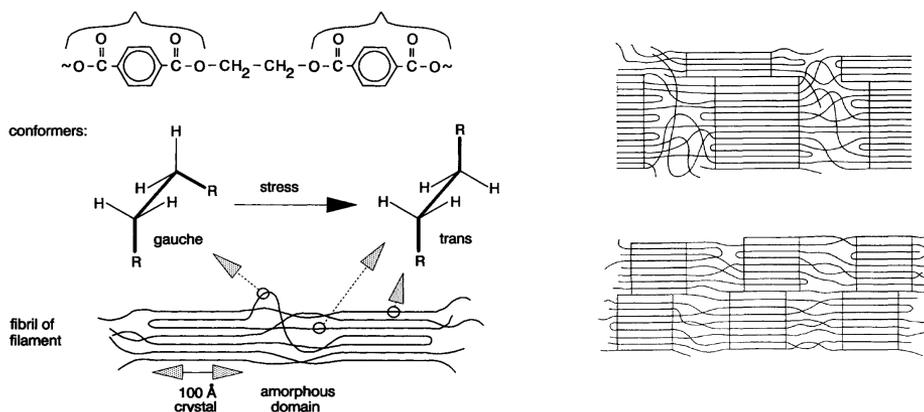


Figure 14: *Orientation of the amorphous domain with transformation from gauche to trans conformation* [17, 23]

The oriented macromolecules in spun and drawn fibres are rarely in their equilibrium state and therefore exhibit thermal shrinkage at temperature above T_g as the molecules tend to coil to reach a more favourable energy state. This shrinkage also leads to a reduction of the mechanical properties [26,27]. Therefore it is important that the fibre is stable at temperatures to which it can be exposed to during operation in the use phase. Fibres can be thermally stabilized through heat setting, which is a process normally performed in or subsequent to the final drawing stage in which the fibre is heated under tension to a temperature that is higher than any temperature to which the material will be exposed to during operation [28]. The optimum setting temperature for PET fibres is 210 °C where strains from the manufacturing process are relieved as the least stable crystals melt and recrystallize into a more stable structure. The level of shrinkage is characterized by measuring the shrinkage after 2 minutes at 177 °C and industrial fibres are classified according to different degrees of shrinkage, as presented in Table 2, where it can be seen that the initial tensile modulus also decreases with the level of shrinkage. This is due to the fact that the fibres with lower shrinkage have a lower fraction of oriented amorphous phase [29].

Tensile properties of polymer fibres

Polymer fibres show an impressive range of mechanical properties with tensile modulus between 5 and 330 GPa and tensile strength up to 7 GPa [30]. Very high tensile strength is achieved with high molecular weight and degree of orientation and crystallinity. The tensile modulus of a PET fibre is stated in the literature to be primarily related to average molecular orientation [23], amorphous orientation and crystal size [29,31], and the trans content in the crystalline and amorphous

Table 2: *Shrinkage and initial modulus of different industrial PET fibres [29]*

	Shrinkage after two minutes at 177 °C [%]	Initial Tensile Modulus [GPa]
High modulus low shrinkage (HMLS)	3.3	14.9
High tenacity (HT)	5.2	14.0
Low shrinkage (LS)	1.6	11.6
Super low shrinkage (SLS)	0.7	12.2

phases [32]. The theoretic modulus for a PET crystal along the chain is about 110 GPa [33]. However as shown in Table 2 the modulus for an industrial PET is between 11-15 GPa since the degree of crystallinity is 40% and all the crystals are not perfectly aligned in the fibre direction. Figure 15 a and b shows how spinning speed and drawing respectively influences the response to tensile loading due to changes in degree of molecular orientation. The initial response to tensile load for a PET fibre is elastic up to a yield that occurs between 0.5-2% strain. Thereafter, extension is composed of an elastic and plastic deformation. The latter involves chain slippage and does not contribute to the increased tensile stress where of the slope of the stress/strain curve decreases. At further extension the slope increases again due to strain hardening when the amorphous regions are straightened out.

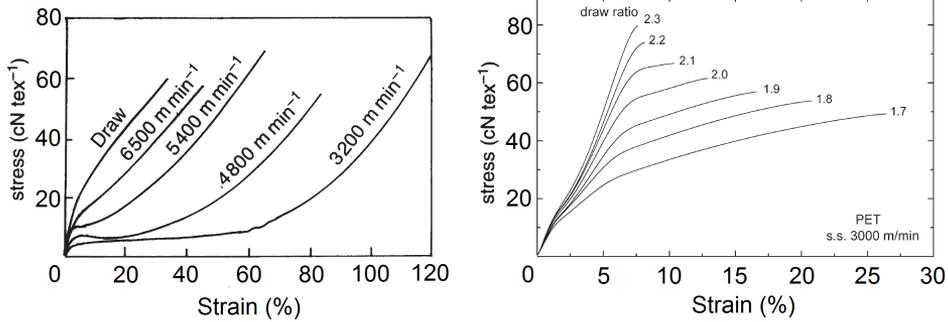


Figure 15: a) *Illustrates how tensile properties are influenced by spinning speed [24], while b) illustrates how drawing influences the tensile properties of a fibre spun at a speed of 3000 m/min [30]*

3 Self-reinforced polymer composites

Utilizing the increased mechanical properties of an oriented molecular structure, these polymers can be used as reinforcement of polymeric material creating what is termed all-polymer composites. The concept of using polymers with oriented molecular structure as reinforcement was introduced by Capiati and Porter more than 40 years ago when they created a one polymer composite out of high density polyethylene [34]. Since then different material systems and manufacturing technologies using reinforcement and matrix from same type of polymer have been developed, in the literature often referred to as self-reinforced polymer composites (SrPC).

Hot compaction

Hot compaction is one of the technologies developed for manufacturing of self-reinforced composites. Here the fibres or tapes, with an oriented molecular structure, are welded together into a solid material by melting only a thin skin of material on the surface of each fibre or tape, as illustrated in Figure 16. In this way the oriented structure is kept in the core of the reinforcement while the molten material perfectly join all reinforcements together into one solid material. This technology has been studied for several different materials including PE [35–37], PA [38] and PET [39–41] investigating optimum process conditions and their resulting mechanical properties. One of the advantages of using identical chemical composition in reinforcement and matrix is that it gives very good bonding between the two components. A small process window is, however, a challenge with this technology since precise control of the process parameters temperature, pressure and time is required in the manufacturing process in order to achieve a composite material with optimized properties. Furthermore this method is limited to very thin laminates.

Bi-component materials

A different approach to create SrPC materials is to use two different components for reinforcement and matrix respectively, but both from the same type of material. The second matrix component in this case can be a co-polymer that has a lower melting temperature compared to the reinforcement so when the two components are heated together, only the matrix material melts and forms the matrix around the reinforcement which remain in solid phase. Figure 17 shows two examples of how this technique can be used, the first with film stacking where a polymer film of

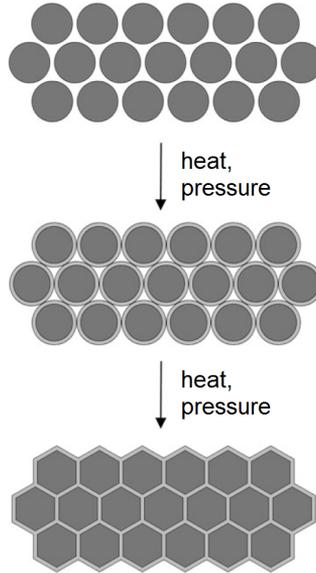


Figure 16: *Illustration of the hot compaction technology [42]*

a low melting matrix material is put between layers of reinforcement material and the second where fibres of the two components are commingled. The advantage of bi-component materials is that the process window is larger compared to single-component materials. On the other hand, using bi-component materials might introduce challenges to adhesion between the reinforcement and the matrix as these two components are not identical. No or poor bonding between the fibre and the matrix leads to reduced performance of the composite material.

Several studies have been performed on bi-component SrPC materials based on PP and PET where the influence from process parameters on the mechanical properties is studied aiming to identify optimum process parameters [26, 43–52]. Other studies have shown how mechanical properties changes with temperature [53–55] as well as establishing the creep [56, 57] and fatigue [58, 59] properties.

The material studied throughout this thesis is a SrPET-material, in the form of a woven fabric, made from yarns with commingled reinforcement and matrix fibres. The reinforcement fibres are high tenacity PET fibres that has a glass transition temperature of 128°C and the melting temperature 265°C. The matrix material is a essentially amorphous PET co-polymer that has a glass transition temperature of 67°C. When this material is heated the matrix material softens and can at a temperature of 215°C wet the reinforcement fibres so that a self-reinforced PET composite can be created.

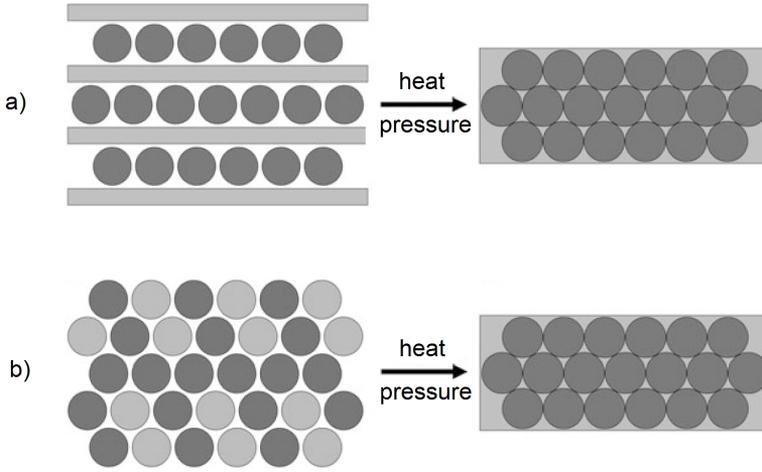


Figure 17: *Bi-component self-reinforced composites, principal for film stacking (a) and commingled material (b) [42].*

Component manufacturing with bi-component materials

The most common method of processing composite materials with continuous reinforcement fibres is to manufacture a woven fabric that can be used for further processing. Components with three-dimensional shape can be manufactured either through direct forming of fabric or by forming pre-consolidated sheets made from fabric material [42]. Desired part thickness is achieved by stacking required number of layer fabric. Consolidation of fabric into sheets or preforms is done at a temperature at which the matrix material can wet the reinforcement fibre. At this high temperature the reinforcement fibres with oriented molecular structure will shrink, and therefore the material shall be constrained during this process in order to avoid reduction of the mechanical properties in the consolidated material. If the constrained and heated material is formed this will lead to stretching of the reinforcement fibres, depending on how much the material is formed. The possibility to stretch the fibres increases the formability [60–62]. However, stretching can influence the properties of the reinforcement fibres. Therefore it is necessary to know how stretching influences the material properties of a formed and consolidated component. This has not been addressed in previous studies on forming of SrPC materials.

When a composite material is processed, the matrix material is in its molten phase and undergoes a phase transition into solid phase while the reinforcement remains in solid phase throughout the entire process. The reinforcement and matrix

may exhibit different thermal behaviour which, in combination with the phase transition of the matrix material, may give rise to internal stresses in the component during the manufacturing process. These stresses can remain in the component as residual stresses and induce shape distortions [63], e.g. warpage or spring-in as illustrated in Figure 18. Knowledge about process induced distortions is important in order to achieve a robust manufacturing process and fulfil requirements for dimensions and dimensional tolerances for the manufactured component.

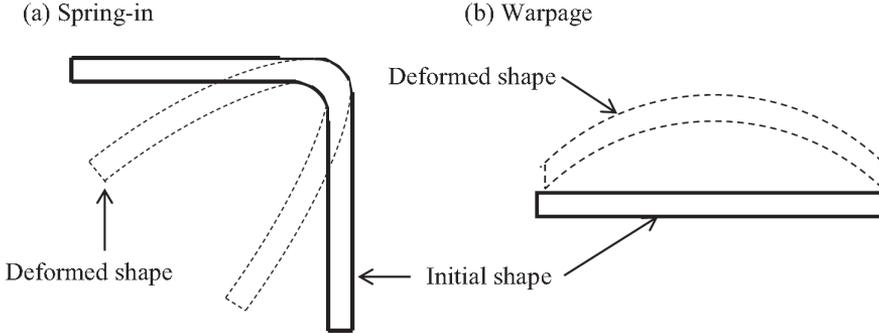


Figure 18: *Two common process induced shape distortions Spring-in (a) and warpage (b) [64]*

Composite sheets possess, in similarity to other sheet materials, a very low degree of design freedom and the opportunities to integrate stiffeners, interface to surrounding part or other functionality are limited. In order to overcome this disadvantage secondary operations can be used where a second component is joined to a preformed component. Adhesive bonding is a simple and flexible method of joining more components to each other and therefore often used. When a thermoplastic based sheet is used, the technology of over-moulding can be applied where a sheet or preformed sheet can be placed in a cavity for injection moulding and over-moulded with a thermoplastic material for functionalization. This is an efficient process used for different applications based on GFRP and has been proposed for use with SrPC as local reinforcement. Another opportunity is to use this technology for integration of stiffeners or other function on SrPC components. However this combination of technologies has so far not been investigated.

Properties and characterization

Table 3 provides an overview of the properties of different SrPC materials. As can be seen, the modulus of the reinforcement with oriented molecular structure is significantly higher in comparison to the corresponding bulk modulus. The composite material can be created with high volume fraction reinforcement with retained low density, which is one of the key factors for light weight performance of SrPC materials. Besides the low weight and high ductility, energy absorption and recyclability are properties often mentioned as benefits for SrPC materials compared to GFRP.

Table 3: *Properties of different SrPC materials in comparison to a GFRP material.*

	SrPE	SrPP	SrPET	PET/GF
Modulus of matrix material [GPa]	1 (HDPE)	1.4	3.0	3.0
Modulus of reinforcement [GPa]	172 (UHMWPE)	15	13	72 (E-Glass)
Volume fraction [%]	50-55	89	70	43
Structure type	UD	UD	UD	UD
Composite density [g/cm ³]	0.97	0.77	1.38	1.88
Composite modulus, longitudinal direction [GPa]	24.5	13	10	28.5
Composite strength, longitudinal direction [MPa]	990	385	350	571
Composite strain to failure		8		1-2
References	[65, 66]	[43, 66]	[26, 66]	[66, 67]

UD=Unidirectional

Characterization of material properties can be made with different methods. There are some fundamental methods that are often sufficient to provide a good basis for the understanding of the properties and behaviour of polymeric materials. For composite materials it is necessary to perform characterization of the different constituents, reinforcement and matrix, as complement to characterization of the composite in order to get complete knowledge about the properties.

According to standardized procedures [68–71] mechanical properties are characterized at room temperature; however the mechanical properties for a composite with thermoplastic reinforcement are influenced more by temperature than a thermoplastic composite with e.g. glass fibres. This since the load carrying performance of a thermoplastic reinforcement is reduced as the temperature increases. The influence of temperature on the mechanical properties depends on the type of polymer and its thermal transitions, especially the glass transition temperature. Analysis of how the mechanical properties are influenced by temperature can be performed using Dynamic Mechanical Analysis (DMA) which is a technique where material properties as function of temperature, time, frequency and stress or a combination of these parameters is characterized [72]. Figure 19 illustrates the ability for a PET and PP tape reinforcement respectively to carry load at elevated temperatures. Both PP and PET are semi-crystalline polymers but PET has a significantly higher glass transition temperature that enhances the ability to retain the stiffness at higher temperatures [73].

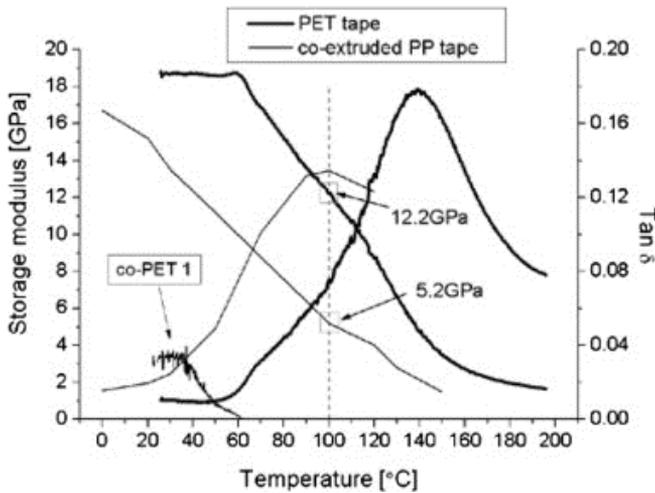


Figure 19: *Dynamic thermal mechanical analysis of PET tape and PP tape [26]*

Microstructural analysis at the molecular level, with determination of molecular weight or characterization of the physical structure (density, molecular orientation, crystallinity or crystal size) are also important to perform in order to study and explain the mechanisms behind the mechanical performance.

Differential scanning calorimetry (DSC) is a thermoanalytic technique used to study the response of polymers to heating [74]. The thermal transitions of glass transition, crystallization and melting can be characterized by measuring the amount of heat required to increase the temperature of a sample. DSC measurements can be done on the composite, however it is necessary to analyze the matrix and reinforcement materials separate in order to get more precise information about the transitions in these two components.

The crystalline structure of a semi-crystalline polymer can further be analyzed with X-Ray Diffraction (XRD), which is a technique to provide information about inter atomic distances, crystal unit cell structure, degree of crystallinity and molecular orientation for a sample [75].

Characterization using infrared (IR) spectroscopy can be carried out in order to analyse each molecule group in a polymer by utilizing the fact that single chemical bonds absorbs IR radiation of wavelength specific for each bond [76, 77]. By projecting IR-radiation and then measure the transmitted or reflected radiation over an interval of wavelengths, a spectrum of IR-absorption can be generated. PET is frequently studied with IR and therefore the spectrum with absorption peaks in the interval from 700 to 1800 cm^{-1} is well understood [78–85]. The bands near 1473, 1343, 973 and 848 cm^{-1} have been referred in literature as the vibrational modes of the trans ethylene glycol of the PET-chain, and the bands near 1453, 1372, 1042 and 898 cm^{-1} refer to the vibrations of the gauche ethylene glycol segments [86,87]. Analysis of these bands can therefore provide information about trans and gauche conformer content.

Applications

SrPC materials are currently used in products such as sports equipment and travel suitcases where their low density and ductile performance at low temperature is utilized [88]. These products are made from very thin SrPP sheets whereof the structural stiffness is also very low. SrPP has also been proposed as concept and studied for automotive applications such as underbody panels and exterior panels with the advantage to lower weight and increase recyclability [89–93]. However may SrPET be more suitable than SrPP for automotive applications since the standard requirement for usage temperature for an automotive component is -40°C to $+80^{\circ}\text{C}$, which is difficult to fulfil with SrPP for semi-structural components like body panels.

4 Recycling of plastics

All products have a limited service life and at End-Of-Life (EOL) the product must be dealt with. Thermoplastic materials possess the advantage of recyclability, which is important for applications such as packaging, which is not only the largest sector for plastics but is also a product that on average has very short service life time. Packaging waste is collected in many countries with the purpose of recycling. One example is beverage bottles made of PET which are collected in a separate flow to be recycled into new PET bottles. Other packaging material such as PE and PP are also recycled but then are often used in other products such as flower pots or pallets for goods. Even though recycling is the preferred options for plastic waste, it difficult to recycle everything that is collected so plastic waste is incinerated for energy recovery. Since plastic materials contains valuable energy and can be burned, incineration is an option that complements recycling to exploit the full potential of plastic waste. Waste collected that is neither recycled nor incinerated goes to landfill, which is the least beneficial option.

In 2014, 25.8 million tonnes of post-consumer plastic waste ended up in the official waste streams [20]. As illustrated in Figure 20, 69% of the plastic waste collected was recovered through recycling and energy recovery processes. Within the different plastic applications, plastic packaging reached the highest recycling rate of 40% and represented more than 80% of total recycled quantities. Amounts of recycled and recovered plastic waste has increased steadily over the last 8 years and in order to continue this positive trend it is necessary to develop and improve waste collection and treatment, as well as to improve human behaviour and attitude to plastic waste. The worst-case scenario is when plastic waste does not end up in any waste stream and instead is spread in nature, since plastic materials are not so easily broken down by natural processes. This has been highlighted as a current growing problem with plastic waste in the oceans that harms ecosystems and wildlife [94].

Recycling of plastics from End-Of-Life vehicles (ELV) is different to recycling of packaging waste because some automotive parts are reused instead of being recycled or recovered while only a small portion goes to landfill [95]. Recycling of plastics from automotive components can be difficult due to the additives used for these materials which increases the variety of materials and also due to the fact that the material has been degraded during the long use-phase which makes it difficult to control the properties of a recycled material. Nevertheless, it is important to use plastic materials that can be recycled or recovered to minimize the environmental impact. Thanks to the potential good recyclability for SrPC materials

these materials can be valuable for automotive applications in order to reduce the environmental impact for ELV.

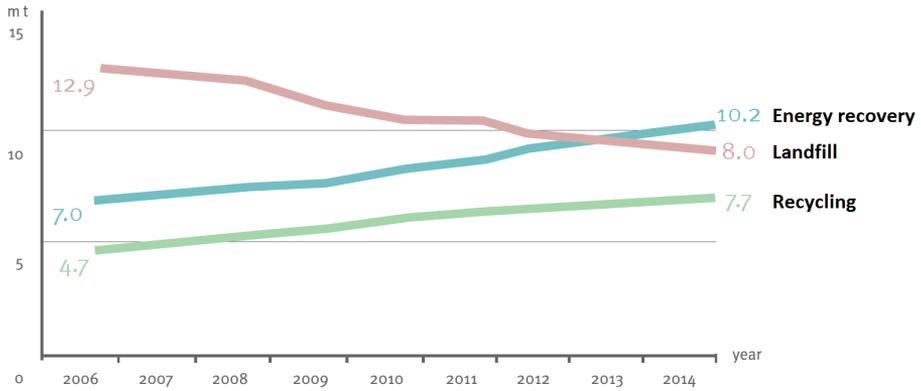


Figure 20: *Waste treatment evolution in Europe 2006-2014, for annual average post-consumer plastic waste of 25 million tonnes. Modified from [20]*

5 From materials research to product development

Materials and technologies can enable commercial and industrial success for both existing and not-yet existing products by becoming the key to new functionalities or improved properties, thus representing an invisible revolution. Vehicles are complex products composed of thousands of components made from a variety of materials, and new materials and technologies are often implemented to improve the properties and performance of these products.

The selection of material is a fundamental stage in the product development process, not only because the material partly defines the properties of the component but also because it is a choice of technology that defines the manufacturing process and influences product cost. The selection is made from certain criteria that have to respond to the functions, requirements and other targets for the intended component. Different materials can be compared or ranked using material indices or material efficiency coefficients which are parameters used for describing maximum of specific material performance [96]. However most often the material and technology chosen for a component is the same as already used in similar products in order to reduce risk of quality deviation and to shorten lead times for product development. This local parameter consisting of local knowledge and expertise from those involved in the development process is not to be underestimated when it comes to material selection. This is also one reason why it is, in general, difficult to introduce new materials and technologies into products. When a new material is introduced in a product, this has often been preceded by extensive pre-development and applied research by the product developer in order to build local knowledge and strengthen local parameters.

Knowledge and experience on new materials and technologies that is developed within academia and industry is very important, but must be transferred to those who are involved in the development of new products in order to be useful. Material science involves multiple disciplines including chemistry, physics and engineering. It is important to work in co-operation between the different disciplines, because basic materials research has to be followed by feasibility studies and technology development before an application can be identified for which benefits can be shown in order to motivate implementation in a product. Only when these steps are connected, as illustrated in Figure 21, is the development of new materials and technologies is successful. In all three steps it is necessary to investigate different aspects related to all phases of the life-cycle. In the technology development step, it is essential to perform thorough investigations related to manufacturing since material properties can be influenced by processing and changes in the material

on a micro level can influence macroscopic properties and engineering aspects of a manufactured component.

Improvement of sustainability for vehicles is not only reached by changing the powertrain, it can also be enhanced by implementation of new technologies and materials that either reduces weight or environmental impact in phases other than the use phase. Therefore applied research on new materials and technologies is important in order to increase the chances of new innovative solutions that can challenge existing technologies and be implemented in vehicles to increase the sustainability of transportation.

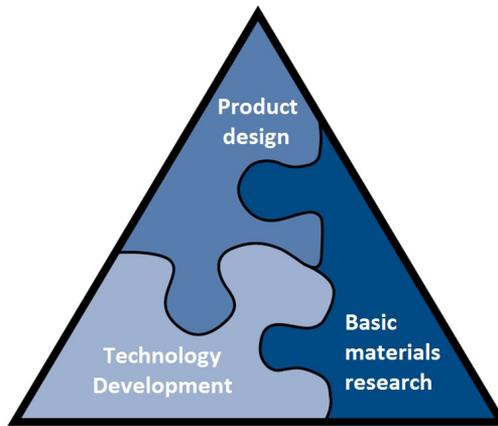


Figure 21: *For successful development and implementation of new materials the different steps in the process must be connected to each other*

6 Objective and scope

Reduction of the environmental impact is important in the development of future vehicles. By applying self-reinforced poly(ethylene terephthalate) (SrPET) composite material in the design of automotive components, weight may be reduced and the recyclability at EOL can be improved, in comparison to components made of glass reinforced thermoset material. However, the knowledge about processing of SrPET for automotive components is today not sufficient in order to achieve a robust high volume serial production. Therefore the aim of this thesis is to study processing of SrPET-materials and investigate certain parameters in the component manufacturing process in order to build important knowledge about processing of SrPET materials for automotive applications and to advance this field of research. Thus following topics are in focus of this thesis.

- Investigation on how stretching of the material, which may occur during the component forming, influence the properties of the manufactured component.
- Investigation on how a component is influenced by elevated temperatures that may occur during post-processes or in the use phase.
- Study of process induced distortions (PID)
- Investigation on over-moulding of SrPET components for efficient integration of stiffeners or other functions.
- Assessment of the environmental impact for a heavy truck component made from SrPET material.

7 Summary of appended papers

Paper A - Influence of fibre shrinkage and stretching on the mechanical properties of self-reinforced poly(ethylene terephthalate) composite

In Paper A, stretching of the SrPET-material prior to consolidation in a component manufacturing process is investigated in order to see how this parameter influences its tensile and compressive properties. Laminates with different degrees of stretched material are manufactured and characterized. It shows that stretching increases the tensile properties but has no significant influence on the crimp factor of the woven structure in the material, i.e. no correlation between these two parameters.

Paper B - Influence of fibre stretching on the microstructure of self-reinforced poly(ethylene terephthalate) composite

The results from Paper A lead to the studies in Paper B on influence from stretching on microstructure in the material. In this paper the crystallinity and molecular conformation were analysed for stretched and non-stretched SrPET-material. It shows that these two parameters are not influenced by stretching during the manufacturing process. The enhanced tensile properties are instead dominated by increased orientation of the amorphous domains in the PET reinforcement. Further it shows that a stretched material shrinks more at elevated temperature than a non-stretched material, i.e. that the oriented amorphous structure is unstable at elevated temperatures. This may be critical for a component if the mechanical or even dimensional properties are influenced in the following use phase. Therefore this influence of temperature is further investigated in Paper C.

Paper C - Process induced shape distortions of Self-reinforced poly(ethylene terephthalate)composites

In Paper C the influence of elevated temperatures on the tensile properties of SrPET laminates is investigated. It shows that the tensile stiffness for a stretched material is reduced after annealing at temperature above the glass transition temperature for the reinforcement fibre. This is due to the fact that high temperatures initiates relaxation and thereby disorientation of the amorphous phase in the reinforcement. Process induced shape distortions are also studied in this paper. Since the material

shrinks at elevated temperature, this indicates that there are residual stresses in the material after hot consolidation which may influence dimensional properties for a component after manufacturing or later in the life-cycle if these stresses are released e.g. if it is exposed to elevated temperatures. V-profiles are manufactured with and without stretching the material prior to consolidation in order to study the influence on spring-in and warpage. It shows that stretching increases warpage of flat sections and affects the profile angle due to stresses that are built up in the laminate during processing. Annealing further influences these two distortion parameters due to release of stresses between fibre and matrix as the material is heated to temperatures above the glass transition temperature for the matrix material. This study clearly shows that SrPET-materials are sensitive to elevated temperatures especially if the manufacturing process involves stretching of the material to form a component.

Paper D - Numerical and experimental investigation on over-moulding of self-reinforced poly(ethylene terephthalate)

Sheet materials allow a low degree of design freedom and so do SrPET-materials. In order to overcome this disadvantage, SrPET can be over-moulded with a thermoplastic material for efficient integration of stiffeners, interface to other components or other functionality. Over-moulding implies exposure of the SrPET-material to high temperature during the moulding cycle. Based on the knowledge gained from Paper C, there is a risk that temperature exposure during over-moulding may influence the properties of the SrPET component. Therefore the influence on tensile properties and dimensional properties from over-moulding is investigated. The temperature exposure during a moulding-cycle is simulated and experimentally validated and shows that the main part of the SrPET-insert is heated to a temperature above its glass transition temperature. At this temperature relaxation of stresses between fibre and matrix occurs. This stress relaxation is characterized as shrinkage and a reduction of the tensile modulus by 10% due to relaxation is measured. Even though over-moulding is an efficient technology for large volume serial production, the influence of temperature exposure on tensile and dimensional properties must be taken in consideration when applied to SrPET materials.

Paper E - Environmental performance of self-reinforced composites in automotive applications - Case study on a heavy truck component

In Paper E the design concept of an over-moulded SrPET-insert, as studied in Paper D, is evaluated with respect to its environmental impact from the phases manufacturing, use and end-of-life when applied on a body panel for a heavy truck. It shows that this concept has 25% lower environmental impact, measured in CO₂ equivalents, compared to currently used concept technology sheet moulding compound. This improvement is thanks to weight reduction for the component studied

and shows that SrPET is a materials that may contribute to more sustainable transports.

8 Conclusion

Self-reinforced poly(ethylene terephthalate) (SrPET) can be used in order to reduce weight and improve the recyclability for automotive components and thereby reducing the environmental impact compared to currently used design with glass reinforced thermoset composite material. However processing SrPET is challenging due to that component forming may lead to stretching of the reinforcement fibres which changes the mechanical properties of the material and introduces stresses in the component that will lead to shape distortions.

Residual stresses that are built in to a SrPET-laminate during the manufacturing process are released when the material is heated to a temperature above the glass transition temperature of the matrix material. This leads to further influence of the dimensional properties and reduction of the tensile properties. This can occur during post processes when the temperature is high or in an over-moulding process when the SrPET-insert is over moulded for integration of stiffeners, creation of interface for surrounding components or other functionality.

This study shows that the temperature-sensitivity of SrPET-materials must be taken in consideration when processing components, and if the component is formed with stretching of the material this has further influence on the dimensional properties and therefore degree of stretching must be controlled. Taking these parameters in consideration, components for automotive applications can be designed and manufactured to reduce weight and thereby contribute to more sustainable transports.

9 Contribution to the field

The contribution to the research field of self-reinforced composites from this thesis can be summarized in following points:

- A design concept with SrPET is for the first time evaluated in a Life cycle analysis, from material manufacturing to end-of-life, for a automotive application. This provides knowledge about how SrPET may be used in order to increase the sustainability for transports.
- For the first time forming of SrPET materials that includes stretching of the fibres is studied with the purpose to analyse the influence on the mechanical properties of the formed component. For the first time also the influence on the crimp of the woven structure due to stretching is studied.
- The microstructure of PET fibres is frequently studied, but this is the first time similar studies are made on composite materials with PET fibres.
- It has been shown that the fibre volume fraction in a SrPET-laminate can be evaluated by measuring the crystallinity with e.g. DSC. This method of evaluation is possible when the matrix material is essential amorphous and the crystallinity of the reinforcement fibres is constant during processing.
- The shrinkage of PET fibres at elevated temperatures belongs to one of the standard test for thermoplastic fibres. For the first time shrinkage has been measured for a SrPET composite and related to changes in mechanical properties. This is the first time that the influence from temperature in phases following consolidation is studied and the sensitivity to elevated temperatures for SrPET materials is shown.
- For the first time, process induced shape distortions are studied for SrPC materials.
- Over-moulding of SrPET is for the first time studied, and may provide a good base to create design guidelines for such concept.

10 Future work

Hybrid materials and technologies

Ongoing research and development on hybrid materials and technologies could also include SrPET materials. Combination with over-moulding has already been investigated in this thesis, but injection moulding is versatile and there are many other possible applications to be investigated, e.g. in combination with physical foaming which can be used in order to reduce both weight and residual stresses in the injection moulded material.

Tools for simulation of fibre stretching during forming

Simulation of fibre stretching in the manufacturing process may be important in order to predict degree of stretching in the component. This information can be used as input for component design and process engineering in order to achieve a component with lower levels of residual stresses. Therefore it would be useful to develop this type of simulation-tools.

Optimization of fibre sizing for SrPET

PET fibres are mainly used for textiles and its fibre sizing therefore optimized for these processes and not for SrPET composites. Fibre sizing optimized for SrPET could improve performance by increasing the bonding strength between fibre and matrix for these materials.

Design and manufacturing of technology demonstrator

For the application of SrPET in automotive components it is necessary that components are designed and that a technology demonstrator is manufactured. This would show the advantages of using these materials for improved sustainability of vehicles.

Bibliography

- [1] T.F. Stocker, D. Qin, G-K. Plattner, M.M.B. Tignor, S.K. Allen, J. Boschung, A. Nauels, Y. Xia, V. Bex, and P.M. Midgley. Climate change 2013: The physical science basis. Technical report, Intergovernmental Panel on Climate Change (IPCC), 2013. Fifth Assessment Report (AR5), Cambridge University Press.
- [2] J. Rockström, W. Steffen, K. Noone, A. Persson, F. S. Chapin III, E. Lambin, T. M. Lenton, M. Scheffer, C. Folke, H. J. Schellnhuber, B. Nykvist, C. A. de Wit, T. Hughes, S. van der Leeuw, H. Rodhe, S. Sörlin, P. K. Snyder, R. Costanza, U. Svedin, M. Falkenmark, L. Karlberg, R. W. Corell, V. J. Fabry, J. Hansen, B. Walker, D. Liverman, K. Richardson, P. Crutzen, and J. Foley. Planetary boundaries: Exploring the safe operating space for humanity. *Ecology and Society*, 14(2), 2009.
- [3] W. Steffen, K. Richardson, J. Rockström, S. E. Cornell, I. Fetzer, E. M. Bennett, R. Biggs, S. R. Carpenter, W. De Vries, C. A. De Wit, C. Folke, D. Gerten, J. Heinke, G. M. Mace, L. M. Persson, V. Ramanathan, B. Reyers, and S. Sörlin. Planetary boundaries: Guiding human development on a changing planet. *Science*, 347(6223), 2015.
- [4] EU. Reducing co2 emissions from passenger cars. European Commission, September 2017. https://ec.europa.eu/clima/policies/transport/vehicles/cars_en.
- [5] VDA. The commercial vehicle - environmentally friendly and efficient. Verband der Automobilindustri (VDA), www.vda.de, Editorial German Association of the Automotive Industry, Technical and environmental division, 2017. Second edition.
- [6] EU. Eu transport in figures - statistical pocketbook 2016. Publications Office of the European Union, European Union, Luxembourg, 2016. ISBN 978-92-79-51528-6.
- [7] Marketa Pape. Towards low-emission eu mobility. Briefing March 2017, EPRS (European Parliamentary Research Service) , Members' Research Service PE 599.356, March 2017.
- [8] EU. *Regulation (EU) No 333/2014 of the European Parliament and of the Council of 11 March 2014 amending Regulation (EC) No 443/2009 to define the modalities for reaching the 2020 target to reduce CO₂ emissions from new passenger cars*. Official Journal of the European Union, 2014.
- [9] Jörg Kühlwein. *Driving resistances of lightduty vehicles in europe: Present situation, trends, and scenarios for 2025*. International Council on Clean Transportation Europe, Neue Promenade 6, 10178 Berlin (ICCT), 2016.
- [10] Justin Hale. Boeing 787 from the ground up. qtr 04 06 a quarterly publication, October 2006. boeing.com/commercial/aeromagazine.

-
- [11] Katrin Pudenz. "born electric": Bmw i3 weltpremiere in new york, london und peking. Onlineartikel springer Professional, Automobil + Motoren | Nachricht, July 2013. springer Professional.
- [12] Y. Yang, R. Boom, B. Irion, D. van Heerden, P. Kuiper, and H. de Wit. Recycling of composite materials. *Chemical Engineering and Processing: Process Intensification*, 51:53–68, 2012.
- [13] Ginger Gardiner. Leichtbau ist hybridbau. *Composites world*, September 2014. <http://www.compositesworld.com/blog/post/leichtbau-ist-hybridbau>.
- [14] Volkswagen. Bildnummer db2013au00181. Volkswagen AG Media Services, February 2013.
- [15] Plasticstoday. Dow automotive systems introduces new epoxy intermediates for structural composites to be commercialized on a front roof header in 2016. *Plastics today*, September 2017. <https://www.plasticstoday.com/automotive-and-mobility/dow-automotive-systems-introduces-new-epoxy-intermediates-structural-composites/52597078925161>.
- [16] H. Staudinger. über polymerisation. *Ber. dtsh. Chem. Ges. A/B*, 53(6):1073–1085, 1920.
- [17] C. J. M. Van Den Heuvel, H. M. Heuvel, W. A. Fassen, J. Veurink, and L. J. Lucas. Molecular changes of pet yarns during stretching measured with rheo-optical infrared spectroscopy and other techniques. *Materials Today*, 49(5):925–934, 1993.
- [18] Rolf Klein. *Laser welding of plastics*, chapter Material properties of plastics, pages 14–16. Wiley-VCH, Weinheim, 2nd ed. edition, 2012.
- [19] APME. *Plastics - a material of choiche for the automotive industry*. APME (Association of Plastics Manufacturers in Europe), 1999.
- [20] *Plastics - the facts 2016*. Plastics Europe, 2016. An analysis of European plastics production, demand and waste data.
- [21] R. de P. Daubeny and C. W. Bunn. The crystal structure of polyethylene terephthalate. In *Proceedings of the Royal Society of London. Series A, Mathematical and Physical Sciences*, number Vol. 226, No. 1167, pages 531–542. Published by: Royal Society, December 1954.
- [22] I. M. Ward and J. Sweeney. *Mechanical Properties of Solid Polymers, Third Edition*, chapter Structure of polymers, page 9. John Wiley & Sons, Ltd., 2013.
- [23] R. Huisman and H. M. Heuvel. Effect of spinning speed and drawing temperature on structure and properties of poly (ethylene terphthalate) yarns. 1989.
- [24] V.B. Gupta and V.K. Kothari. *Manufactured Fibre Technology*. Springer-Science+Business media, first edition, 1997.
- [25] A. Haji and R. S. Rahbar. Structure evolution and mechanical behavior of poly (ethylene terephthalate) fibers drawn at different number of drawing stages. *Chemical Industry and Chemical Engineering Quarterly*, 18(2):233–243, 2012.

- [26] J.M. Zhang, C.T. Reynolds, and T. Peijs. All-poly(ethylene terephthalate) composites by film stacking of oriented tapes. *Composites Part A: Applied Science and Manufacturing*, 40(11):1747 – 1755, 2009.
- [27] Jian Min Zhang and Ton Peijs. Self-reinforced poly(ethylene terephthalate) composites by hot consolidation of Bi-component PET yarns. *Composites Part A: Applied Science and Manufacturing*, 41(8):964 – 972, 2010.
- [28] V. B. Gupta. Heat setting. *Journal of Applied Polymer Science*, 83(3):586–609, 2002.
- [29] B. K. Samui, M. P. Prakasan, C. Ramesh, D. Chakrabarty, and R. Mukhopadhyay. Structure-property relationship of different types of polyester industrial yarns. *Journal of the Textile Institute*, 104(1):35–45, 2013.
- [30] M. G. Northolt, P. Den Decker, S. J. Picken, J. J. M. Baltussen, and R. Schlatmann. *The tensile strength of polymer fibres*, volume 178 of *Advances in Polymer Science*. 2005.
- [31] D. C. Prevorsek, Y. D. Kwon, and R. K. Sharma. Structure and properties of nylon 6 and pet fibres: Effects of crystallite dimensions. 1977.
- [32] S. R. Padibjo and I. M. Ward. A structural study of the tensile drawing behaviour of poly(ethylene terephthalate). *Polymer*, 24(9):1103–1112, 1983.
- [33] T. Thistlethwaite, R. Jakeways, and I. M. Ward. The crystal modulus and structure of oriented poly(ethylene terephthalate). *Polymer*, 29(1):61–69, 1988.
- [34] N. J. Capiati and R. S. Porter. The concept of one polymer composites modelled with high density polyethylene. *Journal of Materials Science*, 10(10):1671–1677, 1975.
- [35] P. J. Hine, I. M. Ward, R. H. Olley, and D. C. Bassett. The hot compaction of high modulus melt-spun polyethylene fibres. *Journal of Materials Science*, 28(2):316–324, 1993.
- [36] M. I. A. El-Maaty, D. C. Bassett, R. H. Olley, P. J. Hine, and I. M. Ward. The hot compaction of polypropylene fibres. *Journal of Materials Science*, 31(5):1157–1163, 1996.
- [37] P.J. Hine, M. Ward, and J. Teckoe. The hot compaction of woven polypropylene tapes. *Journal of Materials Science*, 33(11):2725–2733, 1998.
- [38] P. J. Hine and I. M. Ward. Hot compaction of woven nylon 6,6 multifilaments. *Journal of Applied Polymer Science*, 101(2):991–997, 2006.
- [39] J. Rasburn, P. J. Hine, I. M. Ward, R. H. Olley, D. C. Bassett, and M. A. Kabeel. The hot compaction of polyethylene terephthalate. *Journal of Materials Science*, 30(3):615–622, 1995.
- [40] P. J. Hine and I. M. Ward. Hot compaction of woven poly(ethylene terephthalate) multifilaments. *Journal of Applied Polymer Science*, 91(4):2223–2233, 2004.
- [41] I. M. Ward and P. J. Hine. Novel composites by hot compaction of fibers. *Polym Eng Sci*, 37(11):1809–1814, 1997.

-
- [42] Ben Alcock and Ton Peijs. Technology and development of self-reinforced polymer composites. In *Advances in Polymer Science*, volume 251, pages 1–76. Springer Berlin Heidelberg, 2013.
- [43] B. Alcock, N. O. Cabrera, N.-M. Barkoula, J. Loos, and T. Peijs. The mechanical properties of unidirectional all-polypropylene composites. *Composites Part A: Applied Science and Manufacturing*, 37(5):716–726, May 2006.
- [44] B. Alcock, N.O. Cabrera, N.-M. Barkoula, A.B. Spoelstra, J. Loos, and T. Peijs. The mechanical properties of woven tape all-polypropylene composites. *Composites Part A: Applied Science and Manufacturing*, 38(1):147–161, 2007.
- [45] B. Alcock, N.O. Cabrera, N.M. Barkoula, and T. Peijs. The effect of processing conditions on the mechanical properties and thermal stability of highly oriented PP tapes. *European Polymer Journal*, 45(10):2878–2894, 2009.
- [46] K. J.a Kim, W.-R.a Yu, and P.b Harrison. Optimum consolidation of self-reinforced polypropylene composite and its time-dependent deformation behavior. *Composites Part A: Applied Science and Manufacturing*, 39(10):1597–1605, 2008.
- [47] B. Alcock, N. O. Cabrera, N.-M. Barkoula, J. Loos, and T. Peijs. Interfacial properties of highly oriented coextruded polypropylene tapes for the creation of recyclable all-polypropylene composites. *J. Appl. Polym. Sci.*, 104(1):118–129, 2007.
- [48] J. Andrzejewski, M. Szostak, T. Bak, and M. Trzeciak. The influence of processing conditions on the mechanical properties and structure of poly(ethylene terephthalate) self-reinforced composites. *Journal of Thermoplastic Composite Materials*, 29(9):1194–1209, 2016.
- [49] Chen J.C., Wu M.C, Pu F.C., and Chiu C.H. Fabrication and mechanical properties of self-reinforced poly(ethylene terephthalate) composites. *eXPRESS Polymer Letters*, 5(3):228–237, 2011.
- [50] C. M. Wu, C. Y. Chang, C. C. Wang, and C. Y. Lin. Optimum consolidation of all-polyester woven fabric-reinforced composite laminates by film stacking. *Polymer Composites*, 33(2):245–252, 2012.
- [51] P. Rojanapitayakorn, P. T. Mather, A. J. Goldberg, and R. A. Weiss. Optically transparent self-reinforced poly(ethylene terephthalate) composites: molecular orientation and mechanical properties. *Polymer*, 46(3):761–773, 2005.
- [52] C. M. Wu, P. C. Lin, and C. T. Tsai. Fabrication and mechanical properties of self-reinforced polyester composites by double covered uncommingled yarn. *Polymer Composites*, 37(12):3331–3340, 2016.
- [53] T. Abraham, K. Banik, and J. Karger-Kocsis. All-pp composites (pure) with unidirectional and cross-ply lay-ups: dynamic mechanical thermal analysis. *express Polymer Letters*, 1(8):519–526, 2007.
- [54] B. Alcock, N. O. Cabrera, N.-M. Barkoula, C. T. Reynolds, L. E. Govaert, and T. Peijs. The effect of temperature and strain rate on the mechanical properties of highly oriented polypropylene tapes and all-polypropylene composites. *Composites Science and Technology*, 67(10):2061–2070, 2007.

- [55] B. Alcock, N. O. Cabrera, N.-M. Barkoula, Z. Wang, and T. Peijs. The effect of temperature and strain rate on the impact performance of recyclable all-polypropylene composites. *Composites Part B: Engineering*, 39(3):537–547, 2008.
- [56] Kaushik Banik, Thomas N. Abraham, and Jozsef Karger-Kocsis. Flexural creep behavior of unidirectional and cross-ply all-poly(propylene) (pure textregisted) composites. *Macromol. Mater. Eng.*, 292(12):1280–1288, 2007.
- [57] C. M. Wu, P. C. Lin, and R. Murakami. Long-term creep behavior of self-reinforced pet composites. *Express Polymer Letters*, 11(10):820–831, 2017.
- [58] N. Barkoula, B. Alcock, N. O. Cabrera, and T. Peijs. Fatigue properties of highly oriented polypropylene tapes and all-polypropylene composites. *Polymers and Polymer Composites*, 16(2):101–113, 2008.
- [59] S. Valluri, S. Sankaran, and P. K. Mallick. Tensile and fatigue performance of a self-reinforced polypropylene. pages 130–137, 2010.
- [60] N. O. Cabrera, C. T. Reynolds, B. Alcock, and T. Peijs. Non-isothermal stamp forming of continuous tape reinforced all-polypropylene composite sheet. *Composites Part A: Applied Science and Manufacturing*, 39(9):1455–1466, 2008.
- [61] W. Prosser, P. J. Hine, and I. M. Ward. Investigation into thermoformability of hot compacted polypropylene sheet. *Plastics, Rubber and Composites Processing and Applications*, 29(8):401–410, 2000.
- [62] B. Alcock, N. O. Cabrera, N. M. Barkoula, and T. Peijs. Direct forming of all-polypropylene composites products from fabrics made of co-extruded tapes. *Applied Composite Materials*, 16(2):117–134, 2009.
- [63] M.R. Wisnom and K.D. Potter. 7 - understanding composite distortion during processing. In A.C. Long, editor, *Composites Forming Technologies*, Woodhead Publishing Series in Textiles, pages 177 – 196. Woodhead Publishing, 2007.
- [64] M. Fiorina, A. Seman, B. Castanie, K.M. Ali, C. Schwob, and L. Mezeix. Spring-in prediction for carbon/epoxy aerospace composite structure. *Composite Structures*, 168(Supplement C):739 – 745, 2017.
- [65] A. Teishev and G. Marom. The effect of transcrystallinity on the transverse mechanical properties of single polymer polyethylene composites. *Journal of Applied Polymer Science*, 56(8):959–966, 1995.
- [66] D.W. Van Krevelen. Chapter 13 - mechanical properties of solid polymers. In D.W. Van Krevelen, editor, *Properties of Polymers (Third, completely revised edition)*, pages 367 – 438. Elsevier, Amsterdam, third, completely revised edition edition, 1997.
- [67] Polyone. Technical data sheet unfilled poly(ethylene terephthalate) for injection moulding.
- [68] ISO527. Plastics - determination of tensile properties, iso 527, February 1996.
- [69] ASTM. Standard test method for compressive properties of polymer matrix composite materials with unsupported gage section by shear loading, astm d3410, September 1995.

- [70] ISO178. Plastics - determination of flexural properties, iso 178, December 2010.
- [71] ISO179. Plastics - determination of charpy impact properties, iso 179, 2010.
- [72] ISO6721. Plastics - determination of dynamic mechanical properties, iso 6721, May 2011.
- [73] H. H. Yang. *Aromatic high-strength fibres*. John Wiley & Sons, Inc., New York, 1989.
- [74] ISO11357. Plastics - differential scanning calorimetry (dsc), iso 11357, October 2016.
- [75] Mauro R. Sardela. *Practical Materials Characterization*, chapter X-Ray Diffraction and Reflectivity, pages 1–41. Springer New York, 2014.
- [76] Thierry Lefevre, Christian Pellerin, and Michel Pezolet. *Molecular Characterization and Analysis of Polymers*, volume 53 of *Comprehensive Analytical Chemistry*, chapter 8 Characterization of Molecular Orientation Review, pages 295–335. Elsevier B.V, October 2008.
- [77] I. M. Ward. *Determination of molecular orientation by spectroscopic techniques*, pages 81–115. Springer Berlin Heidelberg, Berlin, Heidelberg, 1985.
- [78] A. Cunningham, I. M. Ward, H. A. Willis, and V. Zichy. Infra-red spectroscopic study of molecular orientation and conformational changes in poly(ethylene terephthalate). 1974.
- [79] X. F. Lu and J. N. Hay. Crystallization orientation and relaxation in uniaxially drawn poly(ethylene terephthalate). *Polymer*, 42(19):8055–8067, 2001.
- [80] K. C. Cole, J. Guevremont, A. Ajji, and M. M. Dumoulin. Characterization of surface orientation in poly(ethylene terephthalate) by front-surface reflection infrared spectroscopy. *Applied Spectroscopy*, 48(12), 1994.
- [81] Neil Everall, Duncan MacKerron, and Derek Winter. Characterisation of biaxial orientation gradients in poly(ethylene terephthalate) films and bottles using polarised attenuated total reflection ftir spectroscopy. *Polymer*, 43(15):4217–4223, 2002.
- [82] D.J. Walls. Application of atr-ir to the analysis of surface structure and orientation in uniaxially drawn poly(ethyleneterephthalate). *Applied Spectroscopy*, 45(7):1193–1198, 1991.
- [83] D. J. Walls and J. C. Coburn. Influence of uniaxial draw on near surface structure in poly(ethyleneterephthalate). *Journal of Polymer Science Part B: Polymer Physics*, 30(8):887–897, 1992.
- [84] M. R. Smith, S. J. Cooper, D. J. Winter, and N. Everall. Detailed mapping of biaxial orientation in polyethylene terephthalate bottles using polarised attenuated total reflection ftir spectroscopy. *Polymer*, 47(15):5691–5700, 2006.
- [85] M. Momose and S. Ando. Quantitative analysis of near surfaces three-dimensional orientation of polymer chains in pet and pen films using polarized atr ftir spectroscopy. *Journal of Polymer Science, Part B: Polymer Physics*, 48(8):870–879, 2010.
- [86] Shaow-Burn Lin and Jack L. Koenig. The effect of temperature on the infrared spectra of poly(ethylene terephthalate). *Journal of Polymer Science: Polymer Physics Edition*, 21(10):2067–2083, 1983.

-
- [87] M. Yazdani, I. M. Ward, and H. Brody. An infra-red study of the structure of oriented poly(ethylene terephthalate) fibres. *Polymer*, 26(12):1779–1790, 1985.
- [88] Samsonite. Curv a fusion of lightness and durability in samsonite hard shell luggage. Samsonite, 2011. Samsonite IP Holdings S.ar.l.
- [89] R. S. Jones and D. E. Riley. A new self-reinforced polypropylene composite. In *SPE Automotive Composites Conference*, 2002.
- [90] Derek Riley, P. J. Hine, M. J. Bonner, and I. M. Ward. Curv a new lightweight, recyclable material for automotive applications. In *Reprinted From: Proceedings of the 2002 SAE International Body Engineering Conference and Automotive & Transportation Technology Conference on CD-ROM (IBAT2002CD)*, number SAE 2002-01-2039, pages -. Copyright 2002 Society of Automotive Engineers, Inc., 2002.
- [91] Derek E. Riley and Renita S. Jones. The development of lightweight, highly impact resistant components for automotive underbody applications. In *2003 SAE World Congress Detroit, Michigan March 3-6, 2003*. SAE International, 2003.
- [92] I. M. Ward, P. J. Hine, and D. E. Riley. Curv self reinforced polypropylene composite from invention to market - a story of collaboration. In *Composites Europe Conference Barcelona, Spain*, 2005.
- [93] B. M. Weager, G. R. Bishop, N.b Black, A.C.and Reynolds, M. W.b Pharaoh, G. F.b Smith, J.c Rowe, D. E. Riley, M.e Birrell, and P.f Donaldson. Development of recyclable self-reinforced polypropylene parts for automotive applications. *International Journal of Vehicle Design*, 44(3-4):293–310, 2007.
- [94] Marine Litter Solutions. What is marine litter? www.marinelittersolutions.eu, September 2017.
- [95] J-L Kanari, N. Pineau and S. Shallari. End-of-life vehicle recycling in the european union. *JOM - Metallurgy and processing (The member journal of the minerals, metals and materials society)*, pages 15–19, August 2003.
- [96] Michael F. Ashby, editor. *Materials Selection in Mechanical Design*. Butterworth-Heinemann, Oxford, fourth edition edition, 2011.

Division of work between authors

Paper A

Jerpdal performed manufacturing, testing and wrote the paper with support from Åkermo.

Paper B

Jerpdal performed manufacturing and testing and wrote the paper with support from Åkermo and Ståhlberg.

Paper C

Jerpdal designed the study with support from Åkermo and Ståhlberg, and performed the experiments together with Herzig. Jerpdal wrote the paper with support from Åkermo, Ståhlberg and Herzig.

Paper D

Jerpdal designed the study with support from Åkermo and Ståhlberg. Schütte performed the simulations with support from Jerpdal, Åkermo and Ståhlberg. Jerpdal and Schütte performed the manufacturing, testing and wrote the paper with support from Åkermo and Ståhlberg.

Paper E

Jerpdal and Poulidikou design the study and collected the input data with support from Åkermo and Björklund. The assessment model and calculation were performed by Poulidikou. Jerpdal and Poulidikou evaluated the results and wrote the paper with support from Åkermo and Björklund.

Part II

Appended papers

