Residual stresses in thermoplastic composites—A study of the literature—Part I: Formation of residual stresses

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Abstract

Continuous fibre reinforced thermoplastic composites are increasingly applied in aircraft structures. These high-performance thermoplastic matrices need processing at high temperatures; therefore, thermal residual stresses arise due to the mismatch in coefficients of thermal expansion between the fibres and the thermoplastic matrix. Since residual stresses are inherently present in virtually all composite materials and influence the properties of the composite structures significantly, it is of utmost importance that the residual thermal stresses are taken into account in both design and analytic modelling of composite structures. In order to understand the effects of residual stresses and find ways to decrease their magnitude or use them to our advantage, the factors responsible for the residual stress build-up need to be understood. This first part of a literature review discusses these factors, focusing on thermoplastic composites, and in particular the material properties of their constituents and processing parameters governing stress build-up.

Keywords: A. Polymer–matrix composites (PMCs); A. Thermoplastic resin; B. Residual/internal stress

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1. Introduction

Currently more continuous fibre reinforced thermoplastic composites (TPCs) are applied in aircraft structures as is shown by the composite programs for the Airbus A380 and A350 [1]. Arguments for using thermoplastic matrices instead of thermoset matrices are mostly of economical nature: thermoplastic composites can be formed and consolidated in several minutes whereas the thermoset composites need a time-consuming cure cycle. Moreover, thermoplastics are known for their weldability, toughness, long (indefinite) storage life, recyclability and chemical resistance [2,3]. However, since this class of materials is relatively new for primary and secondary aircraft structures, some of the properties are still unfamiliar and therefore reluctance exists to apply TPCs. In order to provide TPCs with a wider applicability, more knowledge regarding these materials needs to be generated, summarised, and published.

One of the issues that need addressing is residual stress formation in high performance TPCs. After processing and subsequent cooling of composite laminates from the relatively high processing temperature to the service temperature, residual stresses arise due to the significantly higher shrinkage of the matrix compared with the fibres. This type of residual stress formation is present in all fibre-reinforced polymers, due to their inherent inhomogeneous nature. Residual stress\(^1\) can be defined as “a stress that persists in a material that is free of external forces or temperature gradients” [4]. In this paper we will refer to one type of residual stress: Thermal stress, which can be defined as a residual stress introduced within a body resulting from a change in temperature [4].

Residual stresses influence the properties of the composite structures significantly [5–8], and it is of most importance that the residual thermal stresses are taken into account in both design and analytic modelling of composite structures. It was already argued in the early nineties [3], that the effects of large residual stresses generated in the processing of high performance TPCs (most often accompanied by high processing temperatures) need to be understood, and influences on final properties, quality and durability of the manufactured products need to be studied [9]. To assess the effects of residual stresses on these parameters, a literature study on residual stresses in continuous fibre reinforced thermoplastics was carried out and an attempt was made to identify the knowledge gaps in this field. This review will consist of several parts of which this is the first, describing the available literature on sources and mechanisms of formation of residual stresses in TPCs, related to material properties and processing conditions. To find methods to lower the residual stress magnitude or use the stresses to our advantage, first the factors responsible for the residual stress build-up need to be understood, not in the least to incorporate them in both design and modelling of composite structures. This part will be followed by a literature study on experimental techniques developed for determination of residual stresses in polymer matrix composites, as well as a review on the effects of thermal residual stresses on the composite material properties.

1.1. Summary of previous literature reviews

From a historic point-of-view, Nairn and Zoller [10,11] were, to the best of the authors’ knowledge, the first to publish an experiment-based study on thermal residual stresses in fibre reinforced thermoplastic composites. They determined the stresses in a polysulfone matrix of a uni-directional graphite fibre reinforced tape by means of photoelasticity. In 1988, a review of the then available literature on residual thermal stresses in continuous fibre reinforced polymers (thermosets as well as thermoplastics) was published by Favre [5]. This review can be regarded as base literature when studying residual stresses in fibre reinforced polymers. Favre [5] covered the origins and magnitudes of the thermal residual stresses, temperature, moisture and time effects, and the effects of residual stresses themselves, such as geometrical distortions, microcracks and delaminations due to the free edge effects. Included were predictive models and experimental techniques available at the time and proposals for reduction of residual stresses in composites. In addition, the work by Eijpe [12] is introduced by a good summary of the available literature, and several publications [5,9,13] summarised most of the work done earlier on the sources and driving forces for internal stresses in composite materials, including composite intrinsic properties, processing conditions and environmental conditions.

1.2. Residual stress formation on three mechanical levels

Residual stresses in continuous fibre reinforced thermoplastics are present in the laminate or composite structure immediately after processing and subsequent cooling to the service temperature. They can be regarded at three different levels based on their origins [5,9,13,14]:

\(^1\) In the literature, the terms residual stress and residual strains are often used interchangeably. In some studies, the residual matrix or fibre strains are explicitly measured or calculated, and not the residual stresses. It is the effect of such strains on the developed residual stresses in a composite laminate, which may affect the composite properties.
On the micromechanical level or constituent level, the mismatch in coefficient of thermal expansion between the fibres and the matrix is the governing parameter. Unlike a thermoset matrix, the thermoplastic matrix is heated to a processing temperature above its glass or melting temperature and subsequently solidified upon cooling to the service temperature (often ambient conditions), where no chemical reaction should take place. The cooling involves volumetric shrinkage of the thermoplastic matrix, which is significantly higher than the fibre shrinkage. This represents an important driving force for the development of residual strains in the fibres.

Fig. 1. Schematic view of the effect of cooling on the matrix around a fibre. The bonded (constrained) case represents the situation after processing, where △ and ▽ represent tensile and compressive residual stresses, respectively, after [5].

Fig. 2. Schematic view of residual stress formation in unbalanced cross-ply laminate (a), (b) residual thermal stresses when laminate is constrained, (c) front view of out-of-plane deformation when unconstrained.
and the surrounding matrix [10,15,16]. Assuming that fibre-matrix bonding is present during cooling, Fig. 1 shows that the result is a residual compressive stress in the fibre along the longitudinal axis as well as in the radial direction [10,13], and a residual tensile stress in the matrix in the longitudinal and radial direction [8].

- **Macro-mechanical or lamination** residual stresses are present on a ply-to-ply scale due to lamina anisotropy. The residual stresses arise due to a difference in the transverse and longitudinal ply coefficients of thermal expansion [8]. For example with cross-ply composites, the 90° fibres impose a mechanical constraint on the 0° fibres during cooling and vice versa, because of differences in thermal shrinkage directions [17], see Fig. 2 (where \( \sigma \) represents the residual stresses). The anisotropic shrinkage resembles the behaviour of a bi-metallic strip and, if unbalanced and unconstrained, deformation (curvature) of the laminate may be the result [11].

- **On a ‘global’** [13] level, a gradient in cooling rate, temperature or moisture conditions throughout the thickness of the composite laminate or structure may result in a residual stress distribution through the thickness of the laminate. In general, a thick laminate will experience a slower cooling rate in the centre of the laminate than at the surface plies. At a certain temperature, the centre plies may still need to solidify, whereas the surface plies have already become solid. Upon further cooling, the surface plies impose a constraint on the shrinkage of the centre plies. This will often result in a parabolic distribution of compressive residual stresses in the surface plies and tensile stresses in the centre plies (Fig. 3) [9,15]. In literature, these stresses are referred to as thermal skin-core stresses [9], which in, for example, semi-crystalline composites, may result in morphological skin-core stresses.

Considering the above, these three levels would result in a complex three-dimensional residual stress state within a composite structure. In order to assess the responsible parameters for residual stresses formation, these levels will be dealt with separately. In a composite structure however, the distinction between the three is not so clear-cut.

According to Favre [5], if the long-term and environmental parameters are omitted, the magnitude of residual stresses in composite structures is dependent on four parameters: *temperature difference*, coefficients of thermal expansion/shrinkage upon cooling of the composite constituents (or plies), *elastic coefficients* of these constituents (or plies), and *fibre volume fraction*. These parameters are, in turn, largely dependent on the thermoplastic matrix morphology (semi-crystalline or amorphous), type of fibres, fibre-matrix interface properties, fibre morphology (woven or unidirectional prepreg), and processing conditions. These factors will be discussed separately, starting with the micromechanical level and the fibre-dominated properties, followed by the matrix-dominated properties. Most of the governing parameters for residual stress formation apply to all three levels.

2. Micromechanical residual stresses

2.1. Reinforcing fibres

Studies on residual stresses in thermoplastic composites most often involved carbon fibres (CF), with a few exceptional studies on glass fibres (GF). Aramid fibres were investigated in only one study [10]. Both aramid fibres (AF) and carbon fibres show highly anisotropic thermal expansion behaviour, with small shrinkage in the fibre longitudinal direction when heated [8,18,19]. The thermal expansion behaviour of these fibres can be described by separate coefficients of thermal expansion (CTEs) in the directions parallel (often negative) and perpendicular (positive) to the fibre axis [10,19]. For the temperature range during processing and service life of thermoplastic composites, the expansion behaviour of the fibres is linear. The CTE for the reinforcing fibres is in general much lower than for the thermoplastic matrices, which results in a large difference in expansion behaviour between the matrix and the reinforcing fibres. However, the perpendicular (radial) fibre shrinkage upon cooling was shown to be a relatively insignificant contribution to the formation of residual stresses [13].

2.2. Thermoplastic matrix dominated properties

2.2.1. Shrinkage

The shrinkage in thermoplastic polymers depends on the matrix morphology, i.e. amorphous or semi-crystalline. In semi-crystalline thermoplastics, the volumetric shrinkage results from the densification upon crystallisation (with crystals being of higher density than the amorphous phase) in addition to the shrinkage due to the temperature drop. In amorphous polymers, the shrinkage is only due to the latter. The total shrinkage for semi-crystalline matrices was found to be ten times higher than for amorphous matrices, as can be seen in Fig. 4, where \( T_g \) is the glass tran-
sition temperature, $T_m$ is the melting temperature, and $T_c$ is the crystallisation temperature on slow cooling from the melt [11]. The portion of the curve that includes $T_c$ is a representative plot during cooling. For the temperature range experienced by the thermoplastic matrix, the expansion and shrinkage behaviour is not linear, i.e. the CTE is subject to change at different temperatures [20,21].

2.2.2. Temperature difference

Upon cooling amorphous thermoplastic composites between the processing temperature and the glass transition temperature ($T_g$), the amorphous matrix is in the visco-elastic state. This means that possible stress build-up due to thermal shrinkage can still be relaxed, because the molecular chains possess enough energy for freedom of movement at high temperatures, especially at low cooling rates. When the $T_g$ is reached, the amorphous polymer becomes glassy (thermo-elastic state) and residual stresses are formed and “frozen in” upon subsequent cooling of the composite. Therefore, for amorphous matrix composites such as polyetherimide (PEI), thermoplastic polyimide (TPI), polyethersulfone (PES), and polysulfone (PSU) reinforced with carbon fibres, the temperature at which thermal stresses start building-up (also defined as the “stress-free” temperature) was found to be around the glass transition temperature [13,22–25]. For semi-crystalline matrices such as polyetheretherketone (PEEK), the stress-free temperature (SFT) can be found near the peak crystallisation temperature [10,26,27], because of the load-bearing capability of the crystalline phases below that temperature [15]. The cooling rate affects the peak crystallisation temperature, which in turn influences the stress-free temperature [13,15,23,26,28]. It can therefore be concluded that the thermal transition properties of the polymer matrix are the governing parameters for the SFT. For all polymer matrix composites applies that the further the cool-down or service temperature from the SFT, the more residual stresses reside in the composite due to the mismatch in coefficient of thermal expansion (CTE) between the polymer matrix and the fibres. The SFT was often determined by heating the composite until the residual stresses were found to be zero [10,13].

2.2.3. Processing conditions

The most important processing condition that affects residual stress formation is the cooling rate. The effects of this parameter will be explained later in this section. Few studies mention pressure [6,10,29] and processing environment [5,30] in conjunction with residual stress formation in thermoplastic composites. Pressure may affect the polymer thermal properties [31], which in turn influence the formation of residual stresses. However, it was concluded that pressure levels in composites processing are relatively low [10], therefore limited effects can be expected on the matrix properties. The fibres remain unaffected by pressure. In addition, processing environment may prove to have a significant effect on formation of residual stresses. Oxidation during heating may cause chemical changes to the polymer matrix or fibre–matrix interface. This will influence the thermal and mechanical properties of the polymer and hence, the residual stresses [5]. An extensive study by Augh et al. [30] showed that longer dwell times and higher temperatures during processing affect the glass transition temperature and hence the stress free temperature. This was found to be different for every processing atmosphere.

2.2.3.1. Cooling rate effects in amorphous matrix composites.

As stated in the previous section, due to the pronounced visco-elastic behaviour of thermoplastic matrices [32], matrix relaxation accounts for significant relief of residual stresses, especially at low cooling rates, since the polymer simply has more time to relax at high temperatures. The concept of “free volume” was formulated to explain the materials thermal expansion and shrinkage response [31,33,34]. This concept describes how the total volume in an amorphous polymer consists of the volume occupied by the polymer molecules and the free volume between the molecules. When the polymer is cooled at a high rate, more free volume is “trapped” or “frozen-in” below $T_g$, hence the total volume of the polymer will be higher after cooling down, see Fig. 5. However, the free volume theory is being disputed and new theories are proposed in terms of “excess configurational entropy” [33]. Nonetheless, graphs similar to Fig. 5 can still be used for illustrative purposes. As can be seen in this figure, the glass transition temperature is lower with lower cooling rates (CR), leaving a
The free volume is represented by the shaded area.

smaller temperature range at which stresses can build up [35]. It was calculated [23,35] that the cooling rate in the glass transition region mainly controls the residual stress formation and upon lower temperatures, the residual stress variation with temperature is mainly independent of cooling rate. Therefore, for amorphous polymer matrices, the faster the cooling rate in the glass transition region, the higher the residual stresses will be. Studies have shown that for amorphous matrix (CF/PEI) composites, the (radial) residual stresses indeed increase upon increasing cooling rate [23,35].

2.2.3.2. Cooling rate effects in semi-crystalline polymer matrices. For semi-crystalline thermoplastics, a higher cooling rate will give a lower peak crystallisation temperature and lower crystallinity levels [31,36,37]. This leads to a lower stress-free temperature and less shrinkage [14]. Therefore, it was concluded that residual stresses due to crystallisation could be reduced by increasing the cooling rate [36,38]. However, when taking the relaxation behaviour of the (increased) amorphous content into account, two competing mechanisms will be the result: higher residual stresses due to fast cooling of the amorphous phases, and lower residual stresses because of lower stress-build-up temperature and less crystallisation shrinkage. In this respect, it was stated [39] that a relation needs to be established between visco-elastic behaviour and crystallinity kinetics in order to fully understand the development of residual stresses. In addition, low crystallinity content is often unwanted, due to poor solvent resistance [2,38].

The crystallisation kinetics are different for every semi-crystalline polymer. Some semi-crystalline polymers crystallise so readily upon cooling, that for every practical cooling rate a constant level in crystallinity will be found (polyethylene), whereas other polymers can even be amorphous when quenched or cooled rapidly enough [40]. For PEEK/CF composites subjected to low cooling rates, it was shown that the crystallisation shrinkage is fairly constant [41] and thus only had a small effect on the total residual strain [35,38,41–44]. In effect, it seems that there is a limiting cooling rate for PEEK composites, below which no significant change in residual stresses with different cooling rates can be found [43]. When exposed to higher cooling rates, the residual stresses were found to decrease with increasing cooling rate [42–44], although most practical cooling rates lie below this level (±600 °C/min).

In carbon fibre/polypropylene (CF/PP) microcomposites higher compressive fibre strains were found for slower cooling rates [39]. At high temperatures and slow cooling rates, relaxation of the amorphous phases was observed to take place, which was counteracted by the high crystallisation rate causing the residual strains to be higher. In other studies [36,37] it was observed that in glass fibre reinforced polypropylene (GF/PP) a higher level of residual stresses was present for higher cooling rates. For this material system, the relaxation rate of the matrix amorphous phases prevailed over the crystallisation rate upon slow cooling. The difference in observation may be explained by the difference in reinforcing fibres and the effect of these fibres on the crystallisation rate (trans-crystallinity). This effect will be discussed in Section 2.3.

2.2.4. Elastic properties

One physical polymer property that influences the formation of residual stresses in composites, is the matrix Young’s modulus [5]. The modulus is temperature-dependent and for semi-crystalline materials, cooling rate dependent. A higher cooling rate generates a lower level of highly elastic crystals, hence the resulting Young’s modulus will be lower [31,43]. Barnes and Byerly [13] ranked their calculated stress levels for different composite material systems and found a certain order, linked to the difference in transverse stiffness of the laminates. This transverse stiffness depends on the matrix modulus among other things. In this respect, it may be said that if the matrix Young’s modulus is higher, the residual stresses will increase [45].

2.3. Fibre–matrix interface

As was shown in Section 1.2, the significant difference in shrinkage behaviour between the fibre and the matrix will result in radial as well as longitudinal residual compressive strains in the fibre (Fig. 1). The anisotropic aramid and carbon fibres will sustain higher residual strains in the longitudinal fibre direction [5]. The residual strains in the radial fibre direction offer a significant fraction of the fibre–matrix interfacial (shear) strength for thermoplastic composites, since it “provides one of the mechanisms whereby the matrix grips the fibres and allows stress transfer via the fibre–matrix interface” [17,46].

Since the fibres are loaded in compression along their length due to thermal residual strains, the tendency for
fibre buckling increases and interfacial shear stresses are produced. This may lead to interface debonding and eventually microcracking [8]. A strong bond between the fibre and the matrix is necessary to prevent this from occurring. Treatments to improve the fibre–matrix bond strength are based on chemical bonding (on a molecular level), with e.g. a fibre coating (sizing), and mechanical interlocking [46], where the fibre surface may be treated to become “rougher”. Arguments for the latter treatment are based on the idea that stress transfer is governed by friction, and a linear relation can be assumed between the radial stress and the friction shear stress at the interface [35]. These treatments are synergistically enhanced by the residual radial stresses.

A higher fibre–matrix interfacial bond strength by chemical treatment will increase the residual stresses [35,37], because the residual stresses can be little relieved by interfacial debonding. A weak chemical interface would allow for fibre–matrix slippage in the longitudinal fibre direction relieving partially the stress build-up. This reduces the residual fibre strains [37], preventing the fibre from buckling and interfacial debonding. However, this is not very desirable, as in the end this will yield a composite with lower mechanical properties [37,47]. Nonetheless, experiments with AS4/PEEK composites showed [21] that if no chemical type of fibre–matrix interfacial bonding was present, the shrinkage stresses in AS4/PEEK are sufficiently large to overcome the interfacial shear stresses. This shows that for every fibre–matrix system an optimum in fibre–matrix adhesion strength can be achieved in relation to residual stresses.

Another effect regarding the fibre–matrix interface is that in semi-crystalline composites, the fibre surface may act as nucleation points for the crystalline spherulites. This can result in epitaxial spherulite growth from the fibre surface, which results in a trans-crystalline region around the fibre; see Fig. 6. This region was shown to cause additional radial residual stresses [43,48]. A thicker trans-crystalline layer was found to increase the stress transfer efficiency between the fibres and the matrix, probably due to increased radial compressive stresses owing to the anisotropic nature of the interlayer [48]. However, it was believed by others [49] that the trans-crystalline interface relieved the thermal residual stresses due to preferred orientation of the crystallites with respect to the fibre. This was based on findings of improved static strength and fatigue properties when a trans-crystalline interface was present. It seems that more research is necessary in order to understand the exact mechanisms behind the trans-crystalline layer and its effects on residual stress formation.

In accordance with the processing effects, the interfacial shear stress was higher for slow cooled semi-crystalline matrix composites owing to additional crystallisation shrinkage of the matrix and a thicker trans-crystalline region around the fibre. For amorphous thermoplastics, the interfacial shear stress increased for increasing cooling rate or lower end temperatures [35].

Fig. 6. Microstructure of polypropylene sample isothermally crystallised around a carbon fibre followed by quenching [48].

3. Macro-mechanical residual stress formation

Most of the factors described in Section 2 are also responsible for the macro-mechanical residual stress formation in angle-ply composites, which mechanism was explained in Section 1.2. Parameters that additionally influence the magnitude of residual stresses in TPCs during lamination are shortly listed below. These include the laminate lay-up and morphology of the fibres; such as unidirectional prepregs, fabrics, fibre volume fraction, etc. [5,8,17].

- For all cases, the fibre residual strains were found to be compressive and higher for the more-ply composites [39,50–52].
- As applicable to all levels of residual thermal stresses, the lamination residual stresses also increase as the end temperature is further away from the stress-free temperature [10,11,13,22,23,26,28,43,53].
- Regarding the fibre volume fraction, a few conclusions were drawn in literature:
  - The residual fibre stresses in PP/CF micro- and macro-composites were modelled [54] and it was found that for higher fibre volume fractions, the fibre residual strains were lower.
  - For some composites, an optimum in fibre volume may exist at which the tensile stresses in the matrix in between the fibres turn to compressive [8,55].
  - For cross-ply composites it was stated that the residual interlaminar stresses increase with increasing fibre volume fraction [56].
- A micromechanical analysis was performed into the effects of fibre waviness [57], and it was found that in general, the residual stresses can be affected by fibre waviness and hence the failure behaviour. Increased
fibre waviness lowers the difference in CTE in the transverse and longitudinal direction, consequently for unidirectional as well as angle-ply laminates the residual stresses are lower. The fibre waviness additionally induces longitudinal shear stresses.

- It was found that during consolidation, residual strains accumulate at different rates for each lay-up [58]. In addition, the optimum processing cycle, giving the lowest residual stresses and maximising the mechanical properties of the composite, was found to be different for each lay-up [16] or varying thickness ratios of 90–0° plies in PEEK/CF [26] and PEI/CF [53] cross-ply laminates.
- It was shown for cross-ply polyphenylenesulfide/carbon fibre (PPS/CF) laminates that a minimum processing temperature and pressure are necessary in order to prevent debonding of the plies due to interlaminar residual stresses [59,60].
- At fibre angles of $\omega \approx \pm 30^\circ$, interlaminar shear stresses are maximum [61,62].

Studies on thermoplastic composites and the effects of their fibre morphologies on the formation of residual stresses are not numerous. However, with respect to lamination residual stresses, the parameters governing residual stress build-up are little different between thermoset and thermoplastic matrix composites. Currently, more frequently fabric reinforced thermoplastic composites are applied [1], although few studies exist that compare the differences in residual stress formation in unidirectional prepreg laminates and fabric reinforced laminates.

4. “Global” (skin-core) residual stresses

As explained in Section 1.2, cooling rate plays a significant role in the formation of global residual stresses. High performance thermoplastic composites are inherently manufactured at high temperatures. Therefore, the complex and high cooling rates within thick laminates are characteristically unavoidable [15]. As explained in Section 2.2.2, most of the stresses induced initially relax when temperatures are still high, whereas the stresses induced in the later stages of cooling remain when the matrix is partly crystallised or glassy and thus locks the material. Result is that the process leaves the surface regions of a plate in a state of permanent compression balanced by interior tension (thermal skin-core effects) [14], see Fig. 3. It was shown that with higher cooling rates, the residual stresses are higher and the distribution is more significant [9,14,15,38,63–65]. Most of the fast cooled or quenched composites such as PEEK/CF and PET/CF, show almost a parabolic residual stress profile [9,15,16,38,63,64,66,67], with the surface plies under compression of a higher (negative) magnitude than the tensile residual stresses in the centre plies. This residual stress gradient was found to be of a much higher magnitude in cross-ply laminates as in unidirectional laminates [15,16,63,64].

Crystallinity of semi-crystalline thermoplastic matrices plays a significant role in skin-core effects, when a non-uniform cooling rate through the thickness of the laminate exists. Therefore, many studies regarding residual stress distributions on this level focus on crystallinity distributions immediately after processing and subsequent annealing of the composite (morphological skin-core stresses) [15,63,68]. Annealing is typically executed on thick laminates to reduce the skin-core residual stresses through relaxation of the stresses. The effects of annealing on residual stresses will be considered later in this section. In some studies on semi-crystalline matrix composites, even though no gradient in crystallinity level was present after cooling, still thermal skin-core residual stresses exist, due to the temperature gradients during processing [15,38]. No skin-core residual stresses were measured in 8- and 20-ply composites, because of the low thickness [38], nor for very low cooling rates in 40-ply laminates, such as 0.2 °C/min [38,63]. It was shown that fast cooling rates can lead to significant temperature and crystallinity gradients in the vicinity of free edges and hence significant localised residual stress distributions [65].

4.1. Annealing

In isotropic materials, residual stresses may be removed by annealing since their elastic expansion behaviour with temperature is uniform [23]. However, for anisotropic composites this is not so straightforward. In order to relax a portion of the residual stresses, to lower the residual stress gradients through the thickness of the laminate, or simply to obtain optimal levels of crystallinity, annealing of the composite can be carried out, although this requires additional time and sources [69]. Annealing can be accomplished by raising the composites’ temperature above the glass transition temperature of the matrix and allowing relaxation processes to take place. In consequence, the stress-free temperature may effectively be altered by annealing [13,29].

For amorphous systems on a micromechanical ply scale, annealing does not significantly relax the residual stresses [10]. Moreover, on a macro-mechanical level it was concluded that in multi-axial laminates, the ability for self-relieving of residual stresses is very limited within timescales representative for processing time scales. The amount of relaxation itself is not very significant and even if significant stress relaxation could be achieved at these high temperatures, during cooling to the service temperature the stresses reappear. Therefore, most annealing studies were performed to lower the residual stresses on the global level, e.g. the residual stress gradient through the thickness [9,13,38,43,70].

Increasing the annealing time for semi-crystalline thermoplastics below the glass transition temperature ($T_g$), decreases the residual stresses by means of relaxation [38]. The crystallinity level will be similar before and after annealing, since annealing is carried out below $T_g$. For
annealing temperatures above the glass transition temperature and within the crystallisation range, annealing will increase the crystallinity level of (quenched) specimens (accompanied by an increase in matrix modulus) and hence the degree of residual stresses due to increased (crystallisation) shrinkage [29,43,70]. Mechanical properties and solvent resistance can be improved after annealing [43], although it was believed that relaxation during annealing was the main contributor to increased mechanical properties and not the increased crystallinity level [44]. Quenching of the semi-crystalline composite and subsequent annealing were shown to give similar results as slow cooling [9].

4.2. Influence of processing mould material

Another processing parameter that influences residual stress formation and which was investigated by few researchers, is the use of different mould materials during processing, such as a rubber mould on one half and a metal mould on the other as in rubber press forming. Two types of interaction between the tools and the composite part can be distinguished: thermal and mechanical, and both can influence the development of stresses in the composite.

The thermal interaction was found to have a significant influence on the formation and distribution of global residual stresses (thermal skin-core residual stresses) due to differences in cooling rate on both laminate surfaces [68]. This difference originates from the variation in heat transfer properties of the mould materials and hence the cooling rates [71]. The residual stress profiles for unbalanced cooling were modelled [64,72], by utilising two mould halves under different cooling conditions, which led to much higher residual stresses in the unidirectional laminate, when compared to balanced cooling.

The mechanical interaction comprises two ways: the first is the mismatch in CTE between the tool and the composite, and often the CTE of the tooling is higher than that of the composite [71]. Upon cooling, this may induce compressive residual stresses in the surface plies of the composite part at the tool interface [71], which can result in a through-the-thickness stress distribution in the composite part, depending on the interface conditions between the tool and the composite [72]. Ultimately, the CTE mismatch can lead to warpage of the laminate [72] or fibre waviness [71]. Interfacial stress transfer is determined by the friction behaviour between the tool and the composite material. It is conceivable that friction of the mould with the polymer matrix in its solidifying state [73], induces additional residual stresses in the surface ply, due to forced shrinkage.

In some processes such as in rubber forming, a deformable tool is used in combination with a rigid tool, which comprises the second mechanical interaction. The deformation of the deformable tool may be imposed on the composite part, which efficiency depends on the friction behaviour in the contact interface. The composite part hence experiences different loading on both surfaces, which may result in global residual stresses [72].

5. Discussion and conclusions

In order to increase the applicability of continuous fibre reinforced thermoplastic composites, it was argued already two decades ago that more knowledge regarding residual stresses and their effects needed to be generated. Up to now, still knowledge gaps exist, especially when it comes to studies on structural composites with other high-performance polymer matrices than polyethyetherketone (PEEK). For other semi-crystalline matrices than PEEK, no such extensive research has been performed on the crystallinity kinetics in composites and their effects on residual stress formation.

Some studies investigated other high performance thermoplastic matrices such as polyethersulfide (PES), polyphenylenesulfide (PPS) and polysulfone (PSU) reinforced with carbon fibres. Even fewer studies were published that concerned the residual stress formation in continuous fibre reinforced engineering plastics, such as polypropylene (PP) [36,37,39] and polyethyleneteraphthalate (PET) [11,38]. This can be explained by the fact that the processing temperatures of the high performance thermoplastic composites are, in general, very high and the magnitude of residual stresses will be accordingly. The effects will therefore be more significant and perhaps more disastrous, hence the need to investigate these high-performance materials is more stringent.

Few studies were published regarding residual stresses in fabric-reinforced thermoplastic composites. Since these materials will be applied more often in the future, it is of utmost importance to study residual stress formation in these materials and their differences with composite laminates based on unidirectional plies.

After identification of the most important parameters for residual stress formation with respect to the material properties, it may be concluded that the magnitude of the residual stresses is most dependent on the shrinkage behaviour of the thermoplastic matrix during processing, which varies for different processing conditions such as cooling rate, pressure and environment. Only few studies have been conducted on the effects of the latter two factors on residual stress formation. In order to predict the magnitude of residual stresses, the shrinkage coefficients of the composite constituents must be known, the quality of fibre–matrix adhesion must be taken into account, and the elastic moduli of the constituents must be identified as well as the fibre morphology.

The last few years, much effort is dedicated towards understanding the mechanisms of generation of additional residual stresses due to tool-part interaction in thermoset composites [74–78]. However, few studies are available that investigate this effect on residual stress formation in thermoplastic composites, therefore it may be concluded that the exact mechanisms remain to be investigated for thermoplastic composites.

As a last note, local residual stresses may arise during welding of thermoplastic composites. This may affect the
mechanical behaviour of welded structures significantly. To the best of the author’s knowledge, this issue was not addressed nor raised before. Hence, future research on welding of thermoplastic composites should incorporate the factor of residual stresses.

References
