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Single-Polymer Composites (SPCs): Status and Future Trends

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<u>Outline</u>

1	INTROD	UCTION, DEFINITIONS	5
2	"TOOLB	OX" FOR CREATING SPCS	6
	2.1 Mat	rix/reinforcement6	5
	2.1.1	Molecular weight (MW)	ō
	2.1.2	Nucleation	6
	2.1.3	Tacticity	7
	2.1.4	Polymorphism	3
	2.1.5	Melting, crystallization	3
	2.2 Inte	rface/-phase	L
3	PRODUC	CTION, CHARACTERISTICS	3
	3.1 One	-constituent SPCs	3
	3.1.1	Reinforcements13	
	3.1.2	Preforms 14	4
	3.1.3	Parts	5
	3.2 Two	p-constituent SPCs	5
	3.2.1	Reinforcements	5
	3.2.2	Preforms	5
	3.2.3	Parts	Э
4	REPROC	CESSING	Ð
5	OUTLO	DK AND FUTURE TRENDS 40	0
6	LIST OF	SYMBOLS AND ABBREVIATIONS 40	0
7	REFERE	NCES	2

Abstract

Preparation, properties and applications of single-polymer composites (SPCs), representing an emerging family within the polymeric composite materials, have been surveyed. SPCs were classified in respect to their composition (one- and two-constituents), and preforms (non-consolidated and consolidated). SPCs composed of amorphous or semicrystalline matrices and semicrystalline reinforcements were considered. Methods to widen the temperature difference between the matrix- and reinforcement-giving materials of the same polymer (one-constituent) or same polymer type (two-constituent approach) have been introduced and discussed. Special attention was paid to the unsolved questions related to the interface/interphase in SPCs. It was emphasized that the development of SPCs is fuelled by the need of engineering parts in different applications which have low density and "ultimate" recyclability (i.e. reprocessing *via* remelting). Recent development of SPCs is supported by novel preform preparation, consolidation and production possibilities.

Key words: A. Polymer-matrix composites (PMCs); A. Recycling; A. Textile composites; Single polymer composites

1 Introduction, definitions

Nowadays considerable research efforts are undertaken to produce lightweight, easy reprocessable all-polymeric composites and especially single-polymer composites (SPCs). In all-polymeric versions the reinforcement and matrix are given by different polymers whereas in SPCs they are composed of the same polymer or of polymers belonging to the same type. For the latter sometimes "the same polymer family" is mentioned. SPC is at odds with the traditional definition of composite materials referring to a combination of chemically different materials in which one component serves for reinforcing (load bearing component) while the other for the matrix (for embedding, protection and stress transfer). On the other hand, similar to traditional composites, characteristics (stiffness, strength) of the reinforcing component differ from those of the matrix in SPCs. The former are highly anisotropic (by whatever method produced), whereas the latter is mostly isotropic or far less anisotropic compared to the reinforcement. A further similarity is that the reinforcements in SPCs are also fibers, tapes and related woven or non-woven textile architectures.

SPCs are also referred to self-reinforced, single-phase, homogeneous, mono-material, homogeneity or homocomposites. Next we prefer the term single-polymer composite (SPC) and make two subgroups: i) SPCs from the same polymer (one-constituent SPC) and ii) SPCs from the same polymer type (two-constituent SPC). This straightforward classification will be followed in this paper. Albeit SPCs and related materials have been already surveyed in reviews [1-5], recent developments in this field necessitate their up-date, regrouping and reinterpretation of the results. Our aim was to give a well structured, exhaustive overview on this field. On the other hand, SPCs composed of amorphous matrix and amorphous reinforcement, being less relevant than those with semicrystalline reinforcement, are not included in this review.

The concept of SPC (referred to "one polymer composite") has been introduced by Capiati and Porter in 1975 [6]. The working principle of SPC is identical with that of traditional composites: to transfer the stress from the "weaker" matrix to the "stronger" reinforcement. The latter may show one-, two- or three-dimensional architecture. Because semicrystalline polymers outperform amorphous companions with respect to Young's modulus and strength, they form almost exclusively the reinforcement. The matrix materials may be either amorphous or semicrystalline polymers. Similar to traditional composites the stress transfer occurs via an interface/interphase in SPCs. In traditional composites weak van der Waals forces act across the interfacial region. This does not yield acceptable bonding and thus the reinforcement is generally surface treated (sizing, coating...) and/or the matrix is modified (coupling agent). By contrast, molecular entanglements, favorable amorphous/crystalline superstructures and even H-bonding may serve for improved adhesion and thus for stress transfer between the matrix and reinforcement via the interphase of SPCs. This is one of the great advantages of SPCs over traditional composites containing glass (GF), carbon (CF), aramid (AF) or natural fiber (NF) reinforcements. The other beneficial properties are linked with their low density (the density of many polymers is less than that of the above mentioned fibers) and preferred recycling through remelting. SPCs may compete with traditional

composites in various application fields based on their favored recycling and beneficial performance/cost balance. This is the main reason for the industrial and commercial interests behind the SPCs' development.

2 "Toolbox" for creating SPCs

It is intuitive that the major task when producing SPCs is to widen the temperature range between the matrix and reinforcement with respect to their softening and melting. This is imperative to avoid, or to minimize, the temperature-induced deterioration in the mechanical properties of the reinforcing entities. Loss in stiffness and strength of the reinforcement happens *via* thermally induced shrinkage (relaxation), partial melting which are accompanied with substantial changes in the morphology [7, 8].

On the other hand, the key processing parameters (heating/cooling rates, pressure regime applied) are all time-dependent. The dwelling, holding time during processing strongly influence the property deterioration of the reinforcement, and thus also the quality of the consolidated SPC. Accordingly, the basic task can be specified by emphasizing that the "processing window" should be widened. Widening of the latter may occur by exploiting some intrinsic features of the corresponding polymers and by choosing proper processing methods and conditions. The related aspects will be surveyed next.

2.1 Matrix/reinforcement

2.1.1 Molecular weight (MW)

Unlike amorphous polymers the softening/fusing temperature of which increases with increasing MW, semicrystalline polymers show a more complex behavior. Decreasing MW is usually associated with enhanced crystallinity. On the other hand, the melt viscosity may be dramatically reduced with decreasing MW. Note that the related effect depends also on the polydispersity of the matrix resin. Low melt viscosity is beneficial for the matrix giving polymer in two-constituent SPCs. Jordan *et al.* [9] found that low MW polypropylene (PP) exudes more material and produces a weaker interface by recrystallization than high MW in hot compaction (see later). MW may also affect the properties of the reinforcing fibers as recently concluded on example of polyvinyl alcohol (PVOH) based SPCs [10].

Unfortunately, MW data are seldom reported for SPC constituents in the literature. Differences in the MW characteristics are usually exploited in SPCs with amorphous matrix and amorphous reinforcement [11], which are not covered in this paper.

2.1.2 Nucleation

Several polymers, especially thermoplastic polyesters, exhibit slow crystallization due to delayed self-nucleation. Such materials can be even produced in fully amorphous form through fast cooling operation (quenching from the melt). Above their glass transition (T_g) and below their melting temperature (T_m) these materials undergo cold crystallization. Their softening and crystallization are running simultaneously above a given temperature between T_g and T_m when heating from ambient temperature. The T_g - T_m interval can be considered to select the processing temperature. Two-constituent SPC are produced by "sandwiching"

crystalline reinforcements (film, fabric) in between amorphous films followed by hot pressing. This process is usually termed to film stacking. The concept has been proved on examples of polyethylene terephthalate (PET) [12] and polylactic acid (PLA) [13].

Here we have to mention another method which is based on the same principle but being more general. Practically all semicrystalline polymers show the undercooling (supercooling) phenomenon. Undercooling means that the crystallization temperature (T_c) is markedly below that of T_m due to the fact that the length of the molecular chains is different. Crystallization, preceded by nucleation, occurs in the temperature range between T_m and T_g . Accordingly, the reinforcement can be introduced into an undercooled melt in a two-step process, as recently demonstrated on example of polypropylene [14].

In some systems polymerization is associated with crystallization from an undercooled melt. This is the case in the *in situ* polymerization of cyclic butylene terephthalate (CBT) oligomers. Their ring-opening polymerization, associated with crystallization, may be finished below the melting temperature $(T_m \sim 225^{\circ}C)$ of the resulting polybutylene terephthalate (PBT, but for this system the accepted abbreviation is pCBT) [15, 16]. Irrespective of the scientific debate whether polymerization and crystallization occur simultaneously or consecutively [17], the nucleation is the key issue from the undercooled pCBT [18]. Accordingly SPCs composed of pCBT (matrix) and PBT (reinforcement) can be easily produced by liquid composite molding (LCM) techniques - cf. Figure 1.



Figure 1 Stress-strain behavior of pCBT reinforced with PBT fiber (50 wt%) in unidirectional (UD) and cross-ply (CP) arrangements. Note: Tensile strength of the matrix is given for comparison.

2.1.3 Tacticity

Many polymers contain an asymmetric or stereogenic carbon atom in their chains. This means that a carbon atom is bonded to four different entities. In PP for example the four entities, attached to the tertiary carbon atom, are: hydrogen, methyl group and two side chains of different molecular length. PP exists in two steroregular, viz. isotactic (iPP) and syndiotactic (sPP), and one stereoirregular form. The latter is the atactic PP (aPP) the methyl pendant groups in which are sterically random oriented. In iPP the methyl groups are all at the same side, whereas in sPP alternatively at either side of the planar zigzag-form molecular chain.

aPP is amorphous though its commercial grades often contain some crystalline (mostly iPP) fractions [19, 20]. iPP and sPP are prone to crystallize owing to their highly regular chain conformations. The melting temperature of sPP (T_m~135°C) and iPP (T_m~165°C) are different. This is due to the fact that the overall helical conformations in their elementary crystalline cells, and thus their related packing, differ from one another, as well. It is a straightforward approach to use atactic polymer for the matrix and its stereoregular crystalline versions as reinforcements. Recall that the atactic or amorphous polymer softens/fuse far below the melting temperature of the crystalline form. Surprisingly, this approach has not been followed to produce SPCs though several feasible matrix/reinforcement combinations exist, such as aPP/iPP; atactic polystyrene (aPS)/sPS, atactic polymethyl methacrylate (aPMMA)/iPMMA, atactic polyvinyl chloride (aPVC)/iPVC. For iPP-reinforced SPCs elastomeric PP (stereoblock copolymer composed of aPP and iPP) and sPP may also be used as matrices. On the other hand, the melt compatibility of the corresponding combinations should also be considered. Phase separation that occurs for example in sPP/iPP, has a strong effect on the mechanical performance of the corresponding blends [21, 22]. Nonetheless, exploitation of tacticity-related phenomena may give a new impetus to the SPCs' production.

2.1.4 Polymorphism

Polymorphism means that a material may exist in more than one crystalline forms. Polymers have a strong tendency for polymorphism because their chains can adapt different molecular conformations including both helical and planar arrangements. iPP for example exists in four different crystalline modifications (α , β , γ , δ) with different crystal unit (monoclinic, hexagonal, triclinic and trigonal lattices, respectively) cell parameters owing to various packing of the chains [19, 20, 23]. The usual crystalline modifications of polyamide 6 (PA6) are α and γ , which frequently coexist in molded parts. Usually not all of the crystalline forms are stable. Some of them, considered as metastable, undergo a phase transition toward the more stable version. This transition may be triggered by heat, solvents, mechanical loading or other means. The most important feature of polymorphism is that these crystalline modifications possess various T_m data. Their difference may be very useful to expand the processing window of SPC preparation. The question is whether the "metastable" versions, having lower melting temperatures than the stable ones, can be produced with acceptably high selectivity. In the latter case, they could fulfill the role of the matrix in SPCs in which the reinforcement structure is given by the stable polymorph. This concept has been introduced by Karger-Kocsis who filed a patent on SPC composed of β -PP (matrix)/ α -PP (reinforcement) [24]. Feasibility of this concept was experimentally confirmed in later studies [25, 26]. This was facilitated by the fact that highly selective β -nucleants became available for iPP in the meantime [27]. Other options, like γ -PA6 (matrix)/ α -PA6 (reinforcement), should be explored. The first attempt in this direction has been done by Bhattacharyya et al. [28].

2.1.5 Melting, crystallization

Crystallinity and crystallization-related features have been widely used to enlarge the temperature range of SPCs' processing. In general, all semicrystalline polymers contain crystallites with different perfections. As a consequence, the crystallites have different

melting temperatures and the corresponding polymer has a quite broad melting range. Like crystallization, the melting is also characterized by temperatures linked to the onset, maximum and final fusion. Temperatures in the vicinity of melting onset are well suited to produce SPCs. This is the principle of hot compaction during which the surface of the reinforcing structure is melted and transferred into the matrix [29, 30]. As mentioned earlier, they are different tools to influence the melting/crystallization phenomena which are not only polymer related being strongly influenced by processing variables.

2.1.5.1 Chain branching

Regular chain branching can be generated in the polymerization process *via* various conditions (catalysts, pressure, addition of comonomer etc.). The extent and density of the branches have a strong impact on the crystallization, crystallinity and also on the melting of the related products. Note that the melting range of polyethylenes (PEs) is increasing according to the ranking: low density (LDPE, $T_m \sim 110^{\circ}$ C) < high density (HDPE; $T_m \sim 130^{\circ}$ C) < ultra high molecular weight (UHMWPE; $T_m \sim 135^{\circ}$ C). LDPE contains long side chains in high density which are practically absent in UHMWPE. The temperature difference, that can be guaranteed based on the above T_m data, is sufficient enough for the production of two-constituent SPCs, such as LDPE (matrix)/HDPE (reinforcement), HDPE (matrix)/UHMWPE (reinforcement). Note that in the latter system the MW is also of importance. Such PE combinations were explored in the early stage of the history of SPCs [31-34] as listed later.

2.1.5.2 Copolymers

Through copolymerization the macromolecular chain becomes less regular. As a consequence, the crystals formed are less perfect and they melt at lower temperature compared to the corresponding homopolymer. The related temperature difference of min. 15°C for PP-based systems is large enough for SPC preparation. Note that some copolymers, such as given thermoplastic copolyesters, do not crystallize. This yields an even larger processing window. The copolymer is always the matrix giving constituent. It can be incorporated separately (in forms of fibers, films *via* hot pressing, film stacking) or combined with the reinforcement through a suitable preform (coextruded tape, core-shell type bicomponent fiber, cocarding, comingling etc). The techniques based on shell type bicomponent fiber are especially useful because the reinforcement content of this preform may be very high, up to ca. 90 wt%. Highly stretched copolymer-coated iPP tapes have been marketed by Lankhorst Indutech, the Netherlands [35]. Recall that according to our terminology all copolymer using production methods yield two-constituent SPC versions.

2.1.5.3 Stretching-induced crystallization

Reinforcing components for SPCs are produced by various spinning and stretching processes from semicrystalline polymers. Though there are basic differences between spinning (from the melt and drawing in a temperature gradient) and stretching (cold drawing at different temperatures between T_g and T_m in several steps at increasing temperatures), the common feature is that the resulting product is highly anisotropic and strongly crystalline. Their tendency to thermal relaxation is often suppressed by additional heat treatment usually

performed under load (annealing, heat setting). Spun or stretched semicrystalline polymers exhibit a higher melting temperature than the same polymer crystallized from (quasi) quiescent melt [36]. This is due to the morphology involving aligned and perfect crystal entities. A further benefit of this morphology for SPC application is a pronounced memory effect. This means that the stretching-induced superstructure is preserved longer when heated and restores in higher extent when cooled compared to the isotropic counterpart. As a consequence, one-constituent SPCs can also be produced from preforms, like fiber mats, which are composed from the same polymer however with different draw ratios. This was demonstrated by Izer and Bárány [37].

2.1.5.4 External load/pressure - overheating phenomenon

It was early recognized that highly oriented, crystalline polymer fibers, tapes show a higher melting temperature when heated in constrained (under tension or compression) than unconstrained form [36, 38]. This phenomenon, termed to overheating, impedes the relaxation of fibers and tapes and yields a T_m increase of around 10°C. So, this method is very useful to enlarge the processing window from the viewpoint of the reinforcement. This method is preferentially used for the production of one-constituent SPCs. The broad crystalline melting range along with this kind of overheating form the base of hot compaction of fibers and tapes that has been invented by the group of Prof. Ward (GB 2 253 420). As mentioned before, polymorphic transition may occur upon heating which is influenced also by pressure. The related phase diagram, i.e. polymorphic phases in function of temperature and pressure, is of paramount importance for hot compaction. It was reported that during melting of UHMWPE fiber a metastable hexagonal phase appears in a given temperature and pressure range which contributes to the compaction owing to the enhanced segmental mobility of the macromolecular chain [39].

2.1.5.5 In situ (co)polymerization

Several cyclic monomers and oligomers, such as cyclic amides (ε -caprolactam, lauryl lactam), cyclic esters (ε -caprolactone, CBT) can be polymerized *in situ*. *In situ* means that the related ring opening polymerization is carried out in the mold giving the final shape of the product. In case of SPCs the mold contains the reinforcing structure which is wet out by the low viscosity monomer/oligomer melt. This technique, called liquid composite molding (LCM), shows a clear analogy to the resin transfer molding, widely used to produce thermoset composites with suitable resins. *In situ* polymerization molding features several advantages. The melt viscosity of the polymerizing system is very low, sometimes orders of magnitude lower than the usually accepted threshold for LCM operations, given by 1 Pas [16]. This is very helpful to achieve a complete wet-out of the reinforcing structure. A further benefit is that the polymerization may occur well below the melting temperature of the final polymer. This has been mentioned for CBT already, but holds also for other systems, like PA6, polymerized *via* activated anionic polymerization of ε -caprolactam [40, 41].

Last but not least, other comonomers may be introduced in the polymerizing monomer or oligomer yielding copolymers [42]. Even polymers may be added which generate copolymers *via* various transesterification reaction [43]. The polymerization/crystallization behavior along

with the final thermomechanical properties of the resulting copolymers can be tailored upon request. This has been shown for example with CBT-containing systems [42-44], and also well explored with the earlier Nyrim[®] technology for polyamides. It should be born in mind that the polymerization below the T_m of the final polymer is accompanied with crystallization. The latter, being exotherm, is superimposed to polymerization which should be taken into account [40, 41]. Nowadays there is a vivid interest to produce PA6-based SPCs by this technique [45, 46], fuelled by the revival of the "old" activated anionic polymerization of ε -caprolactam owing to more robust catalyst/activator systems [46, 47].

2.1.5.6 Other methods

Extended polymer chains with strong tendency for oriented crystallization can also be created by special solvent-assisted host/guest inclusion complexation. As host compounds cyclodextrine and urea, whereas as guest macromolecules PA6 and PCL were already used. The guest macromolecules coalesced and appeared in aligned form after appropriate removal of the host. They acted subsequently as strong self-nucleants and generated a reinforcing crystalline superstructure during the bulk melt crystallization of the same polymer [48, 49]. Sintering is another option of SPC production. High-temperature, high-pressure sintering, showing some similarity to hot compaction, has been adapted for UHMWPE textiles [50]. This technique is, however, inevitable in case of SPCs from polytetrafluoro ethylene (PTFE) [51, 52]. Note that PTFE cannot be melted because of superimposed thermal degradation.

Major influencing factors to shift the melting and crystallization temperatures are summarized in Figure 2 schematically.





2.2 Interface/-phase

Creation of strong, durable interface (when two-dimensional, 2D) or –phase (when threedimensional, 3D, owing to a given thickness) between the matrix and reinforcement is a classical problem of the composites' science and technology. This is due to the fact that in all other composites than SPCs the constituents are chemically different and possess various surfaces energies. The latter also governs the wetting of the reinforcement by the matrix. Moreover, direct bonding between the composite's components can hardly be achieved. Such problems do not appear in SPCs. This, however, does not mean that the interfacial effects are negligible. In the pioneering work of Capiati and Porter [6] it has been reported that a transcrystalline layer (TCL) of gradient structure was formed between the reinforcing fiber and matrix. TCL appeared as a strong interphase. TCL occurs by heterogeneous nucleation provided that the nucleating surface possesses active nuclei in high density. In this case the lateral growth of spherulites is impeded. The spherulites grow in one direction, viz. transverse to the nucleating surface. Though the controlling factors of TCL and its effects on the mechanical properties of the related composites are topic of scientific discussions [19, 53-56], it is usually accepted that TCL is caused by epitaxial overgrowth [55, 57]. This implies that the nucleating surface is crystalline and its structure is matched with that of the growing crystals from the melt. This prerequisite is always met in SPCs when the matrix is not amorphous. The interfacial phenomena in SPCs were scarcely studied in depth. This is very surprising based on the aspects listed below. The presence of TCL was frequently demonstrated in SPCs and its positive effect on the interfacial bonding also surmised and emphasized. Note that the appearance of TCL does not necessarily yield improvement in the interfacial transverse and shear stresses. Very few papers delivered further insight in the TCL morphology and related effects on the SPCs' mechanical performance. This is, however, a crucial issue that deserves further attention in this field. Jordan et al. [9] concluded that the formation of opposed TCLs in hot compacted SPCs is detrimental. This is intuitive, especially at transverse loading of the TCL, because the lamellae of the TCs involved do penetrate in each other. The type (meaning the actual morphology) and the thickness of the TCL strongly depend on the crystallization conditions [56, 58] and even on the MW difference between the fiber and crystallizing melt [38, 59]. The mother lamellae, developed first, may lay edge- or flat-on at the reinforcement surface [60]. Reports appeared usually claim about edge-on lamellae structuring with c-axis orientation along the fiber length [61, 62]. The subsequent development of daughter lamellae makes the resulting TCL morphology very complex. Its load bearing capacity, being anisotropic, is direction dependent [19, 54]. Accordingly, interfacial shear strength data, usually determined in pull-out test, may differ from the transverse ones. A further implication arises from the relaxing tape or fiber used as reinforcements in SPCs. Their relaxation through shrinkage generates a local shear/elongational flow in the crystallizing melt. This is the cause of a special type superstructure (cylindrite) developed via homogenous nucleation [63]. Unfortunately, it is tough task to differentiate between cylindritic and TCL on optical level when only the final products are investigated. The difference between transcrystallization and cylindritic crystallization is highlighted in Figure 3.



Figure 3 Columnar crystallization caused by heterogeneous (transcrystallization, a) and homogeneous, row-induced nucleation (cylindrite formation, b), schematically. Note: structure of the cylindrite may be far more complex when the nuclei generate the spherulitic growth of different polymorphs [63].

Last but not least, we have to consider the wetting of the reinforcement by the matrix. Wetting is facilitated by amorphous polymers in traditional composites as shown on the example of aPP/GF [64]. The role of the amorphous fraction is supported also by the fact that lower critical fiber length data were measured for SPCs composed of aPP/iPP than for iPP/iPP [65]. Based on the above aspects Karger-Kocsis [66] recommended a TCL model with lamellar interlocking and amorphous adherent that should enhance the interfacial bonding irrespective of the loading direction.

Apart of TCL, researchers considered other possibilities to improve the interfacial strength in SPCs. Ratner *et al.* [67, 68] combined the hot compaction of UHMWPE fibers with *in situ* crosslinking. The fibers were solution coated by peroxide prior to compaction. This treatment yielded substantial enhancements in both modulus and strength compared to the uncrosslinked reference.

Linear polyamides and polyesters are prone to transreactions [69]. First attempt undertaken by Bhattacharyya *et al.* [28] showed that triggering transamidation is suitable strategy to improve the interfacial properties of PA6-based SPCs. The cited author used antimony trioxide for powder coating of the PA6 yarn before film stacking with quenched PA6 films. Exploitation of transesterification and transamidation reactions may be very promising for two-constituent SPCs.

3 Production, characteristics

3.1 One-constituent SPCs

3.1.1 Reinforcements

Fibers and tapes, also in different assemblies and textile structures, are exclusively used as reinforcements of SPCs. They have to exhibit high Young's modulus and strength. These properties are set by spinning (melt, wet, dry, gel), solid-state drawing (continuous, discontinuous; one-step, multi-step; zone drawing etc.) or by their combination (e.g. melt extrusion followed by in line drawing in several steps at different temperatures). The final aim is to get highly oriented fibers and tapes with high degree of crystallinity. Their morphology is characterized by lamellae, connected with tie molecules, which are embedded in an amorphous phase. The lamellar structure itself may be different, such as shish-kebab type,

extended chain fibrils, oriented folded chain lamellae etc. Needless to say that the desired structure and properties can be guaranteed by selecting suitable polymers (MW, polydispersity, melt flow rate etc.) and proper production conditions (e.g. drawing temperature, rate, ratio etc.). For their SPC applications it is straightforward to stabilize them, i.e. to reduce their temperature-induced shrinkage. This may happen by heat setting, crosslinking and also by incorporation of nanofillers (silica [70], layered silicate, carbon nanotubes [71]). Fibers and tapes may be assembled in different forms and ways as shown later.

Highly oriented nanoscaled fibers, usually in mat form, can be produced by solution or melt electrospinning. Micro- and nanofibrils can be created also by making use of the microfibrillar composite concept, credited to Prof. Fakirov [72]. These fibrils derive from the minor component of properly drawn binary blends after removal of the matrix (major component) by dissolution ("lost matrix").

3.1.2 Preforms

The preforms of SPCs can be grouped into non-consolidated and consolidated ones. The latter is a matrix impregnated preform (or prefabricate) for final shaping. The non-consolidated variants are usually transferred to consolidated preforms, prefabricates through hot consolidation. Consolidation and shaping may occur simultaneously in one step, as well.

3.1.2.1 Non-consolidated

Matrix- and reinforcement-giving fibers and tapes, in both continuous and discontinuous forms, can be combined in many ways. Co- or intermingling to produce various rovings and yarns is well established. Recall that this is necessary if differences in the MW, draw ratio, melt flow rate etc. are exploited for SPC production using the same polymer. Hot compaction is a peculiar technique, in which the matrix is formed after partly and controlled melting of the initial fiber the residue of which fulfills the role of the reinforcement.

Rovings and yarns, comingled or not, are often combined in textile structures, such as nonwoven, woven, UD aligned, knitted, and braided fabrics. Majority of the hot pressing studies used UD and non-woven fabrics. Note that hot pressing related to preforms composed of two constituents but from the same polymer.

For the film-stacking method the films required for sandwiching the reinforcing fabrics are usually available in quenched, less crystalline state compared to the reinforcement.

Discontinuous fibers are generally combined in non-wovens though other options are also possible. Mat is also the usual form of the reinforcements which are produced by electrospinning and by the "lost matrix" method. SPCs produced of different non-consolidated preforms of various polymers are listed in Table 1.

Based on Table 1 the following conclusions can be drawn:

- film-stacking remains in focus of R&D works. As matrices fully amorphous or less crystalline polymer than the reinforcement will be used,
- for compression molding tools with fast heating/cooling options (e.g. Rocktool®) will be used,

 LCM techniques, and especially those which are based on the fast polymerization of ε-CL, will be preferentially followed.

Matrix	Reinforcement type,	Production			
material and type	arrangement, content	Method/concept	Parameters	Comments	Reference
UHMWPE powder	UHMWPE gel-spun fiber, unidirectional (UD) and cross- ply (CP), up to 10 wt%	comp. molding	T~180°C; p=7 MPa	Tensile mechanical, creep and impact tests performed and found superior to plain UHMWPE. Theoretical prediction checked.	[73]
UHMWPE powder	UHMWPE gel-spun fiber, chopped fiber and plain woven fabric, 25-75 wt%	comp. molding	T=152, 162°C	 γ-irradiation of the fabric did not yield improvement. The temperature window was 10°C. Mechanical and wear characteristic determined 	[74]
PP fiber	PP fiber, in-laid fibers in knitted fabric (24-38 wt%) and carded&neddle punched mat of long chopped fibers (30-70 wt%)	comp. molding	T=160-170°C; T reached dwelling for 30 s, followed by p=6 MPa for 90 s and cooling under p=6 MPa	Tensile properties enhanced, transverse perforation reduced with increasing T. Strong effect of the reinforcing fiber content.	[37]
PP sheet	PP multifilament, plain woven fabric, 50 wt%	two-station compression molding	PP sheets melted at T=200°C with p=1 MPa for 10 min; sheets quickly transferred in the second compression station, sandwiched with the fabric and pressed at T=125-150°C	Undercooling concept followed. Tensile strength increased with increasing T and reached 220 MPa.	[14]

Table 1 One-constituent SPCs from non-consolidated preforms

			and p=9 MPa for 10 min		
β-iPP α-iPP	α-iPP tape, UD and CP, 50 wt%	tape winding with film stacking + compression molding	T=160°C for 10 min at p=0 MPa followed by p=7 MPa for 5-8 min prior to cooling under pressure	DMA and perforation impact properties determined. β-iPP matrix outperformed the α-iPP in the corresponding SPCs. α-TCL found.	[75, 76]
β-iPP	α-iPP, carded mat of chopped fibers, 50 wt%	film stacking followed by compression molding	T=155-170°C, p=0 MPa for 1 min followed by p=7 MPa for 2-20 min before cooling under pressure	Tensile strength, E-modulus, density and peel strength increased with increasing T. Perforation energy followed adverse trend. Energy adsorption is governed by delamination.	[77]
β-iPP	α-iPP split tapes, plain woven fabric, 50 wt%	film stacking followed by compression molding	T=136-186°C, p=0 MPa for 30 s followed by p=7 MPa for 2 min before cooling under pressure	Tensile characteristics went through a maximum as a function of T. Perforation energy is reduced with increasing T.	[25, 26]
PA6 film	PA6 yarn, UD aligned	film stacking followed by compression molding	T=200°C; p= 15 MPa for 5 min	Excellent mechanical properties (E-modulus, UTS) reported. Effect of polymorphism suggested.	[28]
ε-CL	PA6, plain woven fabric, 65 wt%	LCM-version, <i>in</i> <i>situ</i> anionic polymerization	T=140-200°C; After fabric impregnation by -CL molding at p=10 MPa for 1 h	Optimum T=160°C found based in flexural and tensile results. Final monomer content < 7 wt%, low void content (2.5%).	[45]
ε-CL	PA6 continous monofilament, UD, 10-20 wt%	LCM variant	T=160-170°C polymerizing melt injected at T=110°C in	UTS and elongation increased by 70-80 and 150-190%, respectively for SPC with 15-20 wt%	[46]

			the mold containing the fibers, reaction time 15 s	reinforcement. Polymorphism studied and presence of TCL demonstrated. TCL thickness depended on the fiber surface treatment.	
PLA sheet	PLA woven fabric (CP multifilament), 25, 50 wt%	film stacking followed by compression molding	T=130, 140°C; holding time (dwelling): 20, 50 s; other parameters varied	Crystallization of the matrix hindered by fast heating. SPCs produced by fast cooling. Temperature window ca. 30°C.	[13]
PET (amorphous)	PET fibers (crystalline) in granules, 30 wt%	extrusion plastification followed by compression flow molding	Extrusion temperature 220-260°C; compression molding at p=20-50 MPa for 120 s	Improved mechanical properties compared neat PET. Mold filling of complex shaped parts problematic.	[78]
PET sheets (amorphous)	PET (crystalline), plain woven fabric	film stacking followed by compression molding	T=180°C; holding time: 0-90 s; p~0.7 MPa	Optimum interfacial adhesion received with 10 s holding time. Rapid heating to avoid cold crystallization of the matrix was crucial.	[12]
LCP yarn (lower T _m)	LCP yarn (higher T _m), UD 50 vol%	compression molding after UD winding	T=260-285°C; p=1.8 MPa until T reached then p=4.48.8 MPa for 30 s followed by pressure reduction to p=1.8 MPa and cooling	Tensile properties, Charpy impact and ILSS determined in function of T. With increasing T tensile strength and ILSS strongly improved; Charpy impact was less influenced.	[79, 80]
LCP fiber	LCP fiber (higher T _m), UD 50 vol%	Winding followed by	p=1.8 MPa until T=260- 285°C reached; p=4	Crystal structure and its change studied. Transition to nematic	[81]

		compression molding	MPa set for few seconds prior to cooling at p=1.8 MPa; compaction pressure also varied (1.8-8.8 MPa) at T=275°C	phase determined. It was delayed by processing at high p.	
LCP fiber chopped	LCP fiber chopped (heat treated) 10, 20, 30 wt%	Dry mixing of the fibers followed by compression molding	T=295°C; p= 4.4 MPa for 30 s	Tensile modulus markedly improved by discontinuous fiber reinforcement. Even higher improvement when using plasma treated annealed fibers. UTS and elongation decreased compared to those of matrix.	[82]
PVOH (plastified)	PVOH chopped fiber 10-30 wt%	compression molding	T=180°C for 5 min at p= 1 MPa after melt compounding the fiber	E-modulus, yield stress and Vicat softening temperature strongly increased with the reinforcement. Ductility reduced.	[10]
PTFE powder	PTFE chopped fiber (30, 40 and 50 μm diameter), 5-20 wt%	cold compaction followed by sintering	T=27°C, p= 15 MPa; for cold compaction; sintering at T=380°C for 30-90 min	SPC outperformed the plain PTFE based on flexural and tensile properties. Optimum composition, condition determined.	[51]
PTFE powder (in aqueous dispersion)	PTFE fiber (chopped), plain woven fabric, 20-80 wt%	after PTFE sheet production, film attacking (compression molding)	Multistep temperature cycle at p=15 MPa	Tensile strength markedly enhanced compared to PTFE at expense of ductility. Friction coefficient slightly, wear rate greatly reduced.	[52]

3.1.2.2 Consolidated

Under consolidated preforms those are considered which also contain the future matrix. This criterion is met "latently" for fibers and tapes when hot compacted. During hot compaction different assemblies of fibers or tapes are compacted at a temperature within the melting range, more exactly in the vicinity of the melting onset of the fibers. A certain proportion of surface of each fiber melts which upon cooling, solidifies and binds the structure together. The melting temperature of the latter "binder" is below that of reinforcing fiber – see Figure 4 [83]. So, the molten portion of the fiber volume forms the matrix of the composite [29, 30, 83]. To select the suitable temperature for hot compaction it is recommended to consider the normalized modulus *vs.* normalized temperature chart, recommended by Ward and Hine – cf. Figure 5 [83]. It is worth noting that hot compaction may result in final products, as well [84].



Figure 4 DSC melting traces of a semicrystalline polymer fiber (broken line) and its hot compacted version (continuous line), schematically (based on [83])



Figure 5 Normalized modulus *vs.* normalized temperature chart for hot compacted semicrystalline polymer fibers, schematically. Notes: tensile moduli of the compacted fibers are normalized as a proportion of the initial fiber moduli, and the compaction temperatures as a fraction of the total melting temperature interval of the fibers, i.e. $=(T_{compaction}-T_{onset})/(T_{end of melting}-T_{onset})$. These temperatures are indicated in Figure 4 [83]

Hot compaction has been well explored for many different polymers and corresponding fabric systems. Moreover, hot compacted prefabricates are produced commercially (e.g. Curv[®]).Works, dealt with hot compaction, are summarized in Table 2.

According to the authors' opinion only technological developments can be expected in this field in the near future.

Table 2 One-constituent SPCs from consolidated preforms

Material Preform	Reinforcement arrangement, content (if given)	Production parameters	Comments	Reference
PE tape	UD, woven fabric 33-91 wt%	T=151-154°C; p=2.8 MPa when T reached; dwell time 10 min set before cooling	30% loss in the original structure associated with best mechanical performance. Morphology development studied.	[85, 86]
PE high modulus fiber	UD	T=138°C	Transverse and longitudinal cross section analyzed after chemical etching.	[62]
HDPE melt spun fiber	UD	T=134-142°C; p=0.7 MPa increased for 21 MPa when T reached for 10 s	Differential melting phenomena during hot compacting discussed	[87]
HDPE fiber (melt spun)	plain woven multifilament 15-55 wt%	T=135-140°C; p=0.7 MPa when T reached; dwell time 2 min set and simultaneously p increased for 2.8 MPa	Good mechanical properties along with interfiber bonding (peel test) observed at ~25 wt% matrix content. Lamellar build up in the interphase studied.	[88]
HDPE (melt spun) UHMWPE (gel spun)	plain woven fabric ~70 vol%	T=optimized; p=2.8 MPa; dwell time: 10 min	Calculated and measured tensile moduli agreed. Anisotropy followed by microhardness measurements.	[89]
UHMWPE gel spun fiber	plain woven fabric, CP, non- woven from chopped fiber	T=144°C; p=1.1 MPa increased for 21 MPa when T reached for 10 s before cooling	Ballistic properties determined and compared with commercial prepregs containing 80-92 wt% fibers. Energy absorption	[90]

			mechanism clarified.	
UHMWPE fiber (gel spun)	wound composite, lay-up:±50°	fiber winding on flat mandrel followed by hot compaction at T=145°C and p=31.0 MPa for 30 min followed by fast cooling	Interface "strengthening" by peroxide crosslinking. DMA, tensile and fatigue properties determined.	[67, 91]
UHMWPE fiber (gel spun)	UD by winding	holding at p=2.8 MPa until compaction temperature (T=142- 155°C) reached; dwell time 10 min; p increased for 12.1 MPa for 1 min prior to cooling	No evidence of surface melting and recrystallization. Fibers hold together by localized welding. Large change in the properties above 154°C.	[92]
PP fiber	UD	T=164-174°C; p=1.1 MPa when T reached; dwell time 20 min used; compaction by p=14 MPa for 10 s prior to cooling	Longitudinal flexural modulus and transverse strength increased with increasing T. Required selective surface melting achieved likely due to high shrinkage forces.	[93]
PP fiber, tape	woven cloths (multifilament, fibrillated tapes, flat tape)	T=180-187°C; p=2.8 MPa when T reached; dwell time 10 min set before cooling	Consolidation quality checked in peel tests. MW effects reported. Peel strength matrix dependent, however, with some effect of the weave style. Matrix ductility should be higher than that of the reinforcement. This can be achieved by high MW and low crystallinity.	[9, 94]
PP tape fibrillated	plain woven fabric	T=166-194°C; p=2.8 MPa when T reached; dwell time 10 min set followed by p=7 MPa for 10 s before cooling	For optimum compaction T=182°C deduced. TCL guaranteed the good mechanical behavior.	[95]

PP tape	plain woven fabric	T=166-190°C; p=2.8 MPa when T reached; dwell time 10 min set before p=7 MPa applied for 10 s followed by cooling	With increasing T larger volume of the molten/recrystallized matrix formed. Morphology studied and shish-kebab structure and flow- induced orientation in form of row structures reported.	[96]
PP tape	plain woven and twill 2/2 fabric	T=188°C; p=4.0 MPa during the compaction; dwell time 5 min followed by cooling to 40°C in 5 min	Effect of feeding/crimping (standard and overfed) of the fabrics studied. Interleaving also with the PP film. Compaction quality better with standard feeding, low crimping – associated with better mechanical properties. No effect of the weave pattern on the penetration impact.	[97]
PP solid state drawn tape containing 20 wt% CNF	UD, woven fabric	T=187-191°C; p=4.9 MPa for 5 min when T reached	CNF incorporation resulted in improved mechanical performance, especially the peel strength was enhanced. PP-g-MA compatibilizer acted properly.	[71]
PA6.6 multifilaments	plain woven fabric	T=260-264°C; p=2.8 MPa for 2 min when T reached	Overheating determined. Effect of water content on the mechanical properties studied. Properties compared with PP and PET compacts.	[98]
PET yarn	UD by winding	T=249-261°C; p=20-60 MPa for 15-30 min when T reached	Pressure regime was crucial. Optical properties influenced by T. Transverse strength enhanced with increasing T at expense of	[99]

			the longitudinal one.	
PET yarn	UD by winding	T=249-256°C; p=32.4 MPa for 15 min when T reached	Transverse modulus and strength improved with increasing T.	[100]
PET fiber	plain woven multifilament	T=253-259°C; p=2.8 MPa when T reached; dwell time 2-15 min set followed by rapid cooling	Increasing dwell time caused deterioration in the mechanical properties and it was associated with pronounced decrease. Optimum temperature window: T=257±2°C. Fast cooling was beneficial.	[101]
PEN multifilaments	СР	holding at p=2.4 MPa until T (268-276°C) reached; dwell time: 2 min before cooling	Excellent tensile mechanical properties (modulus, UTS) when compared to those of PET.	[102]

3.1.3 Parts

As mentioned before, final parts can be fabricated from non-consolidated preform with simultaneous shaping, from consolidated preforms by thermoforming and by in situ LCM techniques. Non-consolidated preforms are seldom converted into shaped products. The possible reason behind may be that the number of parts is less than a critical value substantiating a costly tooling. Majority of the works used consolidated preforms for shaping via thermoforming. [50, 84, 103]. SPCs proved to be more suited for thermoforming than traditional thermoplastic composite sheets ("organosheets"). This is due to followings: the deformability of polymeric fibers and fabrics is better than that of traditional reinforcements, and the traditional fiber reinforcements are far less "compliant" than polymeric ones. This strongly reduces the detrimental effects ("lifting" type deconsolidation, spring back and wrinkling phenomena owing to stored elastic energy and limited deformability) experienced during thermoforming of traditional thermoplastic composites. The melting temperature of the matrix formed is below that of the reinforcing fiber. This along with the temperature dependent strain hardening make the corresponding SPC well suited for thermoforming. Note that this processing requires balanced in plane tensile and shear, and out of plane shear properties [84]. Accordingly, hot compacted sheets are converted into large surface parts showing, however, low complexity in shape. LCM techniques, making use of the undercooling effect, may guarantee the desired design freedom. By contrast, so far composites with simple structured reinforcements (UD) and shapes (plates) have been prepared by LCM technique [46], which may be the next winner in the SPC parts' production.

3.2 Two-constituent SPCs

3.2.1 Reinforcements

Like the reinforcements of the one-constituent SPC, the reinforcing structure for the twoconstituent version is composed of highly oriented fibers and tapes, produced almost exclusively from semicrystalline polymers. Recall that their melting temperature is always higher than that of the melting or softening of the matrix giving polymer from the same polymer type. The latter may be available in various forms, such as fiber, tape, film.

3.2.2 Preforms

3.2.2.1 Non-consolidated

The preforms of two-constituent SPCs are similar in many cases to those of the oneconstituent counterpart. Comingling, coweaving of the components, available in fiber and tapes forms, are widely followed. Additional techniques for holding together the textile assemblies, such as stitching, may also be adapted [104]. For film stacking the films are produced form the polymer of lower melting range than the reinforcement. Other possibility is to avoid or hamper the crystallization through quenching or using suitable copolymers. Hot compaction can be combined with film stacking, as well. Hine et al. [105] introduced this interleaving method in order to reach a better balance in the mechanical properties and a wider temperature window for processing. A large body of works addressed the SPC production using non-consolidated preforms. They are listed in Table 3.

Matrix	Reinforcement type,	Pr	oduction		
material and type	arrangement, content	Method/concept	Parameters	Comments	Reference
LDPE	HDPE fiber, plain woven fabric, interleaved matrix film	film stacking followed by compression molding	T=126-136°C for 5 min dwell at p=4.9 MPa	Advantage of consolidation with interleaved film demonstrated. Concept widens the processing temperature window.	[105]
LDPE	UHMWPE fiber, UD, 55 vol%	the rovings in solution impregnated and UD prepregs produced at T=90°C	specimens (also rings) prepared by compression molding at T=120, 130, 140°C for 10 min	Tensile and compression tests done in fiber direction. Modulus decreased with increasing T, the UTS was less sensitive to T. Failure mode studied.	[106]
LDPE	UHMWPE fiber chopped (2-5 mm) random distribution, 5, 10, 20 wt%	melt compounding in batch mixer at T=125-130°C for 5 min	specimens prepared by compression molding at T=130°C	Tensile modulus and UTS increased with increasing fiber content. Similar effect at the same fiber content with increasing fiber length. TCL formation revealed.	[31]
LLDPE	UHMWPE fiber chopped, random distribution, 10-20 vol%	film-stacking and melt compounding	film-stacking at T=150°C for 10 min at p=1.3 MPa; compounding at T=138°C for 50 min followed by compression molding	Reinforcing effect lost after melt compounding. Cooling conditions affected the tensile mechanical properties of the SPCs produced by film-stacking.	[107]

LLDPE (different chain branching)	UHMWPE fiber, laminates with different lay-up, 65 wt%	filament winding on flat mandrel combined with film-stacking	T=100°C; p=15 MPa; holding time 30 min; laminates' lay-up: +28°/-28°, +32°/-32°, +42°/-42°	Effects of off-axis fiber alignment (winding angle) measured in static tension and in DMA. Values compared with theoretical ones.	[108]
LLDPE (different chain branching)	UHMWPE fiber, laminates with different lay-up, 65 wt%	filament winding on flat mandrel combined with film-stacking	T=100°C; p=22 MPa; holding time 30 min; laminates' lay-up: +28°/-28°, +32°/-32°, +42°/-42°	Wöhler-type fatigue curves elaborated and analyzed. Fatigue life affected by lay-up of the composites and short chain branching of the matrix.	[109]
LLDPE (different chain branching)	UHMWPE fiber, laminates with different lay-up, 65 wt%	filament winding on flat mandrel combined with film-stacking	T=100°C; p=5.5 MPa; holding time 30 min; laminates' lay-up: +35°/-35°, +50°/-50°	Wear and creep behaviors measured. High branching of LDPE resulted in inferior properties. Winding angle influenced the measured properties.	[110]
LDPE HDPE	UHMWPE fiber, CP, 65-70 wt%	filament winding on flat mandrel combined with film-stacking	LDPE: T=120-135°C, p=1.3-6.6 MPa for 40- 60 min followed by fast cooling HDPE: T=137°C, p=1.3-6.6 MPa for 30 min followed by slow cooling and isothermal (T=120-125°C) treatments for 60 min	Tensile mechanical, DMA and ballistic properties assessed. HDPE/UHMWPE SPC exhibited outstanding ballistic resistance. Viscoelastic damping contributed to the energy absorption.	[111]
LDPE HDPE	UHMWPE fiber, laminates with different lay-up,	filament winding on flat mandrel combined with	composites produced at T=120°C (LDPE) and T=137°C (HDPE) for	Effects of off-axis fiber alignment (winding angle) studied in DMA as a function of frequency (0.3-50	[112]

	~80 wt%	film-stacking	60 min and p=15 MPa; HDPE composites was also thermally treated; laminates' lay-up: +26°/-26°, +34°/-34°, +45°/-45°	Hz). Activation energy of α and γ relaxations determined. Effect of TCL could not be proven.	
HDPE	UHMWPE fiber, CP, 72 wt%	filament winding on flat mandrel combined with film-stacking	T=137°C, p=2.4 MPa for 60 min	Ballistic impact by gun bullets investigated. Energy absorbing events concluded.	[113]
HDPE	UHMWPE fiber (gel spun), UD, at various angles (30-45°)	fiber winding with film sandwiching	T=137°C; p=16.5 MPa for 1 h	Static and fatigue loading results sensitive to ply angle.	[114]
HDPE	UHMWPE fiber, laminates with lay up: 90°, 45°, ±45°, 50 vol%	film stacking followed by compression molding	T=132°C for 8 min	Tensile failure mode investigated using acoustic emission.	[115]
HDPE	UHMWPE fiber, single fiber composite	fiber embedding in molten matrix	Annealing at T=138°C for 5 min followed by cooling to crystallization temperature T=124°C and kept for 24 h	TCL morphology development studied.	[116]
HDPE	UHMWPE fiber chopped, random distribution, 10-20 vol%	film stacking version and compression molding	T=134-136°C for 15-20 min at p=0.75 MPa; then quenching or isothermal crystallization	Tensile E-modulus and UTS almost threefold of the neat HDPE. Isothermal crystallization improved the stiffness at cost of the ductility. Big difference between experimental and	[123]

				theoretical data.	
HDPE powder	UHMWPE fiber, UD, 20-50 vol%	wet powder impregnation of the fibers followed by winding on a plate SPC through compression molding	T=135°C for 10 min at p=0.5 MPa	Mechanical properties (tension, compression) determined in both parallel and perpendicular to the fiber alignment. Failure mode concluded.	[117]
HDPE	UHMWPE fiber, UD, 65 vol%	prepregs produced by filament winding around a drum followed by heat treatment (T=130°C, 30 min)	compression molding at T=132°C for 20 min at p=10 MPa	Tensile properties as a function of T and fiber orientation measured. Effect of electron irradiation dose studied, also for creep. Irradiation had negligible effect on both fracture and creep.	[32]
HDPE	UHMWPE fiber, UD, 45-55 vol%	filament winding on flat mandrel combined with film-stacking	compression molding at T=133-150°C for 30-60 min at p=0.32 or 0.48 MPa; cooling varied (slow, fast)	Thermal contraction of the UHMWPE fibers measured. Tensile modulus and UTS went through a maximum as a function of T. Effect of cooling rate was marginal. TCL detected.	[33]
HDPE	UHMWPE fiber, UD, 50-55 vol%	filament winding combined with film-stacking	composites produced at T=134°C for 60 min at p=0.32 MPa followed by various cooling method	Longitudinal and transverse tensile mechanical properties measured. Transverse strength theoretically predicted with the experimental results. Good	[34]

				bonding between fiber and matrix concluded. TCL observed at slow cooling. LLDPE did not show transcrystallization.	
iPP	iPP tape, twill 2/2 woven fabric, interleaved matrix film	film stacking followed by compression molding	T=175-190°C for 5 min dwell at p=4.9 MPa	Advantage of consolidation with interleaved film demonstrated. Concept widens the processing temperature window.	[105]
PP-C (random)	iPP chopped fiber, carded and needle punched mat, 50 wt%	film stacking followed by compression molding	T=150-175°C; p=5.5 MPa; holding time 90 s	Two matrix giving PP-C with different MFI used. ILSS and tensile E-modulus and strength went through a maximum, perforation impact energy monotonously decreased with increasing T.	[118]
PP-C (random) α and β forms	α-iPP tape	film stacking followed by compression molding	T=136-186°C; p=0 MPa for 1 min followed by p=7 MPa for 2 min before cooling under pressure	Tensile characteristics went through a maximum, while perforation impact energy reduced with increasing T. Slight effect of β nucleation.	[25, 26]
PP-C (random) α and β forms	α-iPP tape, UD and CP	tape winding with film stacking followed by compression molding	T=145, 155°C; p=7.0 MPa; holding time 8 min	DMA, flexural and perforation impact properties determined. β- PP-C outperformed the α matrix in respect to static flexural and perforation impact properties.	[119]
PP-C (random)	PP fiber chopped with different diameter, random oriented	film stacking with two-step compression molding	T=152-157°C; p=0 kPa for 5-7, after that p=11- 14 kPa for 8-10 min in both steps	Tensile and creep tests performed. Morphology and TCL development detected. Best performance found for 50 µm	[120-122]

				reinforcing fiber. Good agreement between experimental and theoretical tensile E-moduli. Creep behavior modeled.	
PP-C (random)	PP fiber, UD, 7 vol%	film stacking followed by compression molding	T=163°C; p=1.9 MPa; holding time: 1 min followed by fast cooling	Effect of TCL in single fiber composites studied. Well- developed TCL at T=163°C detected that showed high ductility at transverse loading to the fiber.	[123]
PP-C (random)	PP fiber chopped, random oriented, 10-60 wt%	film stacking with two-step compression molding	T=152-157°C; p=0 kPa for 5-7 min, after that p=11-14 kPa for 8-10 min in both steps	Tensile characteristics peaked at 50 wt% reinforcement.	[124]
PP-C (random)	PP woven fabric (weave patterns: plain, basket, twill, satin), 50 wt%	film stacking followed by compression molding	T=155-160°C for 5-7 min, after that p=11-14 kPa for 8-10 min followed by cooling	Strong effect of the weave pattern on the mechanical properties, ascribed to differences in their 'unit cell'.	[125]
PP-C (random) fiber, chopped	PP fiber chopped, non-woven by comingling, 50 wt%	compression molding	Conditions varied, viz. holding time, pressure, cooling rate	DMA, static mechanical and creep properties studied and optimum processing parameters deduced.	[126]
PP-C (random) + EPR (EPR content up to 30 wt%)	PP fiber, non-woven mat, 50 vol%	film stacking with two-step compression molding	T=152±5°C for 5-7 min, after that p=11-14 kPa for 8-10 min	With increasing EPR content the E-modulus decreased but the ductility and impact resistance increased. Optimum EPR content of the matrix was at 20 wt%.	[127]
PA6	PA6.6 fiber, plain woven fabric,	film stacking followed by	T=225-245°C; p=5 MPa for 30 min, and slow	Almost threefold increase in UTS at optimum T=235°C. TCL	[128]

	~70 wt% also in single fiber composite	compression molding	cooling under pressure	development investigated.	
PA6	PA6.6 fiber, UD, 20 wt%	tape winding combined with film stacking followed by compression molding	T=250°C for 5 min at p=30 MPa	Polymorphism quoted but not fully confirmed.	[129]
PET-C	PET tape, UD, <70 wt%	tape winding on flat mandrel combined with film stacking followed by compression molding	T=120-180°C; p=0.12- 2.40 MPa	Effect of PET-C types and molding conditions studied. Results compared with hot compacted PET SPC and GF reinforced PET.	[130]
PET-C	PET fiber, plain woven fabric (multifilament) after heat setting, 46 vol%	film stacking with compression molding in two steps	First fabric and sheet molded at 200°C for 1 min at p=5 MPa. Second, several layers compression molded at T=215-235°C for 3-10 min at 10 MPa followed by slow cooling	Temperature window ~50°C. Effects of T and holding time on tensile and flexural properties measured. To avoid degradation low T and holding time should be selected.	[131]
PET-C (home synthetized)	PET fiber chopped (highly crystalline), random distribution	melt mixing followed by compression molding	Pressing at T=200°C for 30 s	Preliminary study to find suitable PET-C as matrix.	[132]

The content of Table 3 suggests that further considerable efforts will be made to produce twoconstituent SPCs from non-consolidated preforms. It can be prophesied that textile fabrics composed of commingled yarns, rovings will be developed for direct consolidation and shaping. Film stacking will also remain the preferred technique.

3.2.2.2 Consolidated

New options in this field represent bicomponent fibers (side/side, skin/core arrangements) and bicomponent tapes. By contrast to bicomponent tapes, the route of bicomponent fiber' production is not explored. In these bicomponent preforms the matrix giving polymer is *ab ovo* present by contrast to the one-constituent counterpart. Tapes with skin/core/skin cross section may be fabricated by coextrusion or lamination processes. The lower melting skin (also in case of bicomponent fibers) overtakes the role of the matrix during hot pressing, whereas the remaining core acts as reinforcement. The coextrusion techniques (spinning, tape fabrication) usually involve a stretching, drawing step to achieve the required stiffness and strength for the reinforcing constituent [133-136] – Figure 6.



Figure 6 Schematic illustration of bicomponent tapes and composite production based on [136]

Bicomponent tapes are available under the trade names Pure[®], Armordon[®], Kaypla[®]. For their processing in tape or fabric forms compression molding (discontinuous, continuous), consolidation by vacuum bagging, hot stamping, thermoforming, and tape winding may be used [103, 135, 137]. Bicomponent electrospun fibers were not yet used for SPC preparation – this is, however, just a question of time. A further new preform is the pellet. This is prepared by melt coating of the reinforcing fibers. The resulting pellets can be used for injection molding because the difference in the melting ranges of the matrix- and reinforcement-giving components may be large enough [138, 139] – cf. Figure 7.



Figure 7 DSC melting of a thermoplastic EPR matrix and reinforcing iPP fiber (a), and DMA traces of the corresponding injection-molded SPC as a function of the molding temperature (b). Note: the chopped iPP fiber content of the SPC was 70 wt% [138]

A process similar to the fabrication of long, discontinuous fiber reinforced thermoplastics for injection molding, seems to be also feasible. Other option is to consolidate and pelletize a previously commingled roving or yarn [78]. When the processing window for the pellet is narrow, instead of injection molding compression flow molding may be chosen [78]. In this case the masticated, molten pellets are transferred into the cavity of the mold where final shaping, along with substantial squeeze flow, occurs upon closing the mold. Note that one-constituent SPCs cannot be injection molded except adapting overmolding [140]. Works devoted to the SPC production using consolidated preforms are listed in Table 4.

Table 4 Two-constituent SPCs from consolidated preforms

Matrix	Reinforcement type,	Pr	oduction		
material and type	arrangement, content	Method/concept	Parameters	Comments	Reference
PP-C	PP tape (coextruded, Pure [®]), UD, >90 wt%	tape winding and compression molding	T=140, 160°C; p=2.4 MPa during heating (10 min) and compaction (10 min); cooling time to T=40°C 5 min	Tensile strength and modulus measured at different loading angles. Results compared with calculated ones using the laminate theory.	[133]
PP-C	PP tape (coextruded, Pure [®]), plain woven fabric, >90 wt%	vacuum bagging, compression molding	T=140, 160°C; p=0.1 MPa for vacuum bagging, and p=1-15 MPa for hot pressing; during heating (10 min) and compaction (10 min); cooling time to T=40°C 5 min	Temperature processing window >30°C. Tensile and compression behavior measured. With increasing p improved the mechanical properties. SPCs tested under different loading angles.	[134]
PP-C	PP tape (coextruded, Pure [®]), plain woven fabric, 90 wt%	continuous double belt press; vacuum bagging	Belt press: T=145°C; p=2.5 MPa Vacuum bagging: T=145°C; p=0.1 MPa	Fatigue (tension-tension) tests performed at various T and related damage assessed.	[141]
PP-C	PP tape (coextruded, Pure [®]), UD and CP, ~90 wt%	tape winding and vacuum bagging in autoclave	T=145°C; p=2.4 MPa when T reached for 90 min, consolidation for 30 min; cooling to 30°C	Static flexural and flexural creep properties determined and modeled.	[142, 143]

			in 60 min		
PP-C	PP tape (coextruded, Pure [®]), plain woven fabric, ~90 wt%	compression molding	T=130-170°C; p=0.1- 11.4 MPa	Transverse perforation impact of sheets determined and compared with those GMT and NMT. Interfacial strength assessed in T- peel tests. Impact performance increases with decreasing interfacial strength achieved at low consolidation temperature and pressure.	[144]
PP-C	PP tape (coextruded, Pure [®]), plain woven fabric, >90 wt%	compression molding	T=140, 160°C; p=1-15 MPa for 10 min consolidation	Transverse perforation and ballistic impact test performed. Interfacial strength concluded as key parameter.	[145]
PP-C	PP tape (coextruded, Pure [®]), plain woven fabric, >90 wt%	compression molding	T=140°C; p=4 MPa	DMA in flexure conducted, also on specimens at 45° loading angle. Static tensile modulus in function of T is comparable with that of GMT with 23 wt% GF.	[146]
PP-C	PP tape (coextruded, Pure [®]), UD, woven fabric (weave patterns: plain, twill, satin), ~90 wt%	compression molding	Different processing conditions	Effects of processing methods and conditions on the mechanical properties introduced.	[103]
PP-C	PP tape (coextruded), woven fabric	compression molding	T=140-153°C; p=10 MPa for 30 min	Creep also on off-axis specimens, static tensile and interfacial strength (T-peel) determined as a function of T. Small change in T resulted in significant changes in the mechanical behavior. This was	[147]

				attributed to partial melting of the reinforcement core of the tape.	
PP-C (random)	iPP fiber, chopped, 50-80 wt%	UD composite by compression molding followed by pelletizing then injection molding	SPC pellets: T=180°C; p=0 for 240 s, then p=5.26 MPa for 240 s followed by cooling to 50°C Injection molding: T=160°C; injection pressure: 80 MPa	Specimens prepared by injection molding with different gates and subjected to static tensile, DMA, perforation impact tests. Increasing fiber content enhanced the yield strength and perforation impact. Gate type affected the reinforcement distribution. Detected shrinkage ascribed to relaxation of PP fibers.	[139]
PP-C (TPE)	iPP fiber, chopped, 70 wt%	UD composite by compression molding followed by pelletizing then injection molding	SPC pellets: T=140°C; p=0 for 240 s, then p=5.26 MPa for 240 s followed by cooling to 50°C Injection molding: T=120-160°C; injection pressure: 80 MPa	Injection molded specimens tested (shrinkage, DMA, static tensile, perforation impact). Temperature window of ca. 90°C reached. Properties of SPC comparable with PP, however, with better low temperature impact performance. Fan gate reduced the melting of the reinforcing fibers.	[139]

R&D activities on two-constituent SPCs from consolidated performs will likely focus on the manufacturing and processing of injection moldable grades. Works may also address the development of bicomponent fibers which will be used in various textile assemblies.

3.2.3 Parts

Similar to one-constituent SPCs, final parts can be fabricated from non-consolidated preform with simultaneous shaping, film stacking. When first a panel is manufactured by hot pressing then final shaping occurs usually by thermoforming. Consolidated preforms, i.e. bicomponent tapes, can be combined in various textile structures for subsequent hot pressing and thermoforming. These preforms can be used in other processing methods. Tape winding and tape laying are also feasible. In these processes various heat sources (gas torch, laser) may be used because only the skin (surface) layer of the bicomponent tape should be melted to achieve the required "welding" [148]. Flat panels are produced also by tape winding with subsequent hot pressing. As underlined before, one of the major advantages of twoconstituent SPCs is that in their case injection molding may be practiced. The pellets' structure resembles to that of long discontinuous fiber reinforced thermoplastics. On the other hand, overmolding of reinforcing structures, placed in the mold cavity prior to the infiltrating with the matrix forming injected melt, is also viable [140]. This kind of injection molding may be considered as a peculiar version of the LCM techniques. Earlier this technique, termed film insertion injection molding was used to prepare "laminated" SPCs [149]. LCM techniques, focusing on in situ copolymerization possibilities, round up the processing methods of two-constituent SPCs. Though the latter has not been explored for SPC yet, its feasibility demonstrated on example of traditional composites [150].

The quality assurance of SPC prefabricates and parts is a crucial issue. Quality of the products was assessed by various destructive methods (different kind of mechanical loading) coupled with further *in situ* (such as acoustic emission [151, 152] or post mortem (light and scanning electron microscopy) failure inspections so far. Non-destructive techniques were rarely used for quality management though ultrasonic testing [153] and X-ray micro computed tomography seems (Figure 8) to be the right tools [154]. This is still an open issue, especially if in line quality inspection is the target.



Figure 8 Three-dimensional (3D) picture of an ultrasonic welding seam (overlapped) in a PPbased SPC revealed by X-ray micro computed tomography. Notes: the reinforcing iPP (ca. 40 wt%) in woven fabric form is well discernible. The seam thickness is about 1.5 mm.

4 Reprocessing

SPCs are referred to materials of "ultimate" recycling. Interestingly, this aspect was less studied. Recycling *via* remelting of SPCs was mostly studied in repeated injection molding. On the other hand, the works performed on this field [155] clearly show that the number of reprocessing/remelting cycles, which is not accompanied with property deterioration, agrees fairly with the related value derived for injection molded neat polymers. This result was received in works using PP-based systems.

5 Outlook and future trends

SPCs will remain under spot of interest further on. This prediction is based on the fact that they are lightweight (their density is lower than most of the traditional composites) and environmental benign (especially due to their straightforward recycling through melt reprocessing).

In respect to the matrix/reinforcement combinations amorphous/semicrystalline and semicrystalline/semicrystalline combinations will be the winners. Attempts will be further made to modify the reinforcement by nanofillers, especially by those which have high aspect ratios. Their incorporation should yield increased stiffness, strength and thermal stability. The fire resistance of SPCs should be improved. This is a high priority issue for further potential applications. Pioneering activity in this direction yielded some peculiar results [156, 157]. SPCs of more complex structure, such as panels containing honeycomb or foam cores, may help to acquire new applications fields [158]. A further hot topic is the development of (multi)functional SPCs. In this respect attention may be focused on the shape memory performance of SPCs [159]. Further works are needed to check the potential of electrospun reinforcements in SPCs. LCM production methods are very promising especially when exploiting possible transreactions between the reinforcement and the polymerizing melt. This aspect was covered in a recent patent to produce UD-CF reinforced composite core via LCM as load bearing cable of electric transmission lines [160]. In case of hot pressing of nonconsolidated preforms trials will be done to avoid problems with the "slow" heat conductivity. Incorporation of suitable (nano)particles in the matrix or matrix-giving component acting as hot spots (heat sources) by external triggering (electromagnetic field, microwave etc.) seems to be a sound strategy [161, 162]. To solve the quality assurance in line during production is also imperative. Joining of SPCs via various methods is a great challenge [154]. Describing the infiltration of the molten resin into the reinforcing structure [163]and modeling of the structure-property relationships in SPCs are further important tasks [164]

6 List of symbols and abbreviations

ILSS	[N/m]; [MPa]	interlaminar shear strength
MFI	[g/10 min]	melt flow index
р	[MPa]	processing pressure
		temperature

	glass transition temperature			
	melting temperature			
UTS [MPa]	ultimate tensile strength			
α-	alpha form of			
β-	beta nucleant or form of			
γ-	gamma form of			
δ-	delta form of			
AF	aramide fiber			
aPP	atactic polypropylene			
aPMMA	atactic polymethyl methacrylat			
aPS	atactic polystyrene			
aPVC	atactic polyvinyl chloride			
CBT	cyclic butylene terephthalate oligomer			
CF	carbon fiber			
CNF	carbon nanofiber			
СР	cross-ply structure			
DMA	dynamical mechanical analysis			
EPR	ethylene-propylene rubber			
GF	glass fiber			
GMT	glass (fiber) mat reinforced thermoplastic			
HDPE	high density polyethylene			
iPMMA	isotactic polymethyl methacrylat			
iPP	isotactic polypropylene			
iPVC	isotactic polyvinyl chloride			
LCM	liquid composite molding			
LCP	liquid crystalline polyester			
LDPE	low density polyethylene			
MW	molecular weight			
NF	natural fiber			
NMT	natural (fiber) mat reinforced thermoplastic			
PA6	polyamide 6			
PA6.6	polyamide 6.6			
PBT	polybutylene terephthalate			
рСВТ	polymerized cyclic butylene terephthalate			
PCL	polycaprolactone			
PE	polyethylene			
PEN	polyethylene napthalate			
PET	polyethylene terephthalate			

PET-C	polyethylene terephthalate copolymer
PLA	polylactic acid
РР	polypropylene
РР-С	polypropylene copolymer
PP-g-MA	maleic anhydride grafted polypropylene
PTFE	polytetrafluoro-ethylene
PVOH	polyvinyl alcohol
sPP	syndiotactic polypropylene
sPS	syndiotactic polystyrene
SPC	single polymer composites
TCL	transcrystalline layer
UD	unidirectional alignment, structure
UHMWPE	ultra high molecular weight polyethylene

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