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Functional graded self-reinforced polypropylene sheets[**)]

Summary — Self-reinforced polypropylene composites (SR-PP) combine exceptional mechanical properties with an impressive light-weight construction potential and good recycling qualities. This paper focuses on the description of the structural composition and the potential of self-reinforced polypropylene sheets. First of all, the functionality of self-reinforcement is specifically illustrated. The performance of SR-PP sheets in comparison to conventionally reinforced fibre composites is highlighted. In order to gain basic knowledge of SR-PP composites, the processing technology is illustrated in detail. The consolidation process significantly influences the self-reinforcement and, consequently, the final property composition of the SR-PP sheets. Based on the requirements regarding component application, the challenges in transferring the material technology to application series are outlined. Thermo-mechanical gradation provides a good approach to solving the problem of depicting complex demand profiles in order to enable economic viability in broad fields of application. Functional gradation significantly influences the mechanical properties of SR-PP sheets locally, thus, making it possible to produce impact strengths and consistencies compatible adjusted to the component functions.
Keywords: self-reinforcement, polypropylene, hot-compaction, thermoforming, functional gradation.

FUNKCJONALNIE GRADIENTOWANE SAMOWZMOCNIONE KOMPOZYTY POLIPROPYLENOWE

Streszczenie — Artykuł stanowi przegląd literaturowy dotyczący samowzmocnionych kompozytów polipropylenowych (SR-PP) wykazujących wyjątkowe właściwości mechaniczne (tabela 1, rys. 2, 7) w połączeniu z niezwykłą lekkością konstrukcji oraz łatwością recyklingu. Opisano budowę strukturalną i potencjalne możliwości aplikacji samowzmocnionych płyt polipropylenowych. Szczegółowo omówiono proces funkcjonalnego, gradientowego samowzmacniania polipropylenowych płyt (tabela 2, rys. 3—6), wskazując na zasadniczy jego wpływ na właściwości końcowego produktu i porównano metody wytwarzania SR-PP z metodami otrzymywania tradycyjnych kompozytów wzmacnianych włóknami. Scharakteryzowano możliwości zastosowania samowzmocnionych płyt PP wykorzystujące specyficzne cechy tak uzyskiwanego materiału.
Słowa kluczowe: samowzmocnienie, polipropylen, gradientowe termoformowanie, kompaktowanie na gorąco, gradientowa funkcjonalizacja.

FUNCTIONALITY AND PERFORMANCE OF SELF-REINFORCEMENT

Self-reinforcement characteristics

Self-reinforced polypropylene (SR-PP) sheets consist of continuously merged polypropylene fibres or tapes and do not require extrinsic fibre reinforcement. The self-reinforcement of polymeric materials is based on the orientation of the macromolecules and a characteristic shish-kebab structure. The shish-kebab structure is achieved utilising melt and solid phase deformation and suitably conducted processing, which both facilitate the preservation of the structure in the component [1—4]. Self-reinforced thermoplastic composite materials essentially differ from classically foreign fibre reinforced composites due to the method of reinforcement applied [5].

If organic and inorganic based extrinsic fibres — such as carbon, glass, aramid or natural fibres — are bedded in the polymer matrix, the self-reinforced thermoplastic composites consist of a single-component system. The self-reinforced fibre phases are made up of identical basis polymers as the matrix [6, 7]. Due to this, a transferral of the self-reinforcement onto three-dimensional geometries is difficult, necessitating an intermediate step involving layered

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semi-finished products in form of foils, fleece or tape fabric systems [8]. The ensuing consolidation of the composites may be completed either by combining the hot compaction and the thermoforming processes or using the compression molding process [9—11].

In terms of their structure, self-reinforced fibre and tape based layered composites can still be classified as classic composite materials, but morphology-wises, the responsible for the exceptionally high performance potential of this group of materials: in comparison to non-reinforced compact materials, the composite mechanical properties can be increased several times over by using implemented self-reinforcement thus enabling them to compete even with foreign fibre reinforced composites [16]. In order to illustrate the power spectrum of self-reinforcement, the theoretical and practice-oriented potential of selected thermoplasts is compared with glass, steel and aluminium in Table 1. When comparing self-reinforced fibres to compact materials, the ability of self-reinforced polyolefine fibres to compete with metallic materials is evident when the modulus of elasticity and strength are taken into consideration.

T a b l e 1. Comparison of theoretical and experimental evaluated figures for modulus of elasticity and strength [17—20]

Material	Modulus of elasticity			Strength		
	theoretical GPa	experimental		theoretical GPa	experimental	
		fiber, GPa	compact, GPa		fiber, GPa	compact, GPa
Polyethylene	300	200 (67 %)	1.0 (0.3 %)	27	5.5 (20 %)	0.030 (1.0 %)
Polypropylene	50	20 (40 %)	1.6 (3 %)	16	1.3 (8 %)	0.038 (0.2 %)
Polyamide 66	160	5 (3 %)	2.0 (1 %)	27	1.7 (6 %)	0.050 (0.2 %)
Glass	80	80 (100 %)	80 (100 %)	11	4.0 (36 %)	0.055 (0.5 %)
Steel	210	210 (100 %)	210 (100 %)	21	4.0 (19 %)	1.40 (7 %)
Aluminium	76	76 (100 %)	76 (100 %)	7.6	0.8 (10 %)	0.60 (8 %)

macromolecularly orientated self-reinforced fibres and tapes with the distinctive shish-kebab structure can be clearly separated from the matrix impregnation [12—14]. For example the macromolecularly oriented structures can be identified by confocal laser scanning microscopy, see Fig. 1. Therefore, the hot compaction process joins fibre and matrix areas microscopically. Macroscopically however, only a single-component system is obtained [15]. Furthermore, failure mechanisms such as crack growth and delamination which can be attributed to the conventional fibre composite systems are not a feature of homogeneous compact materials.

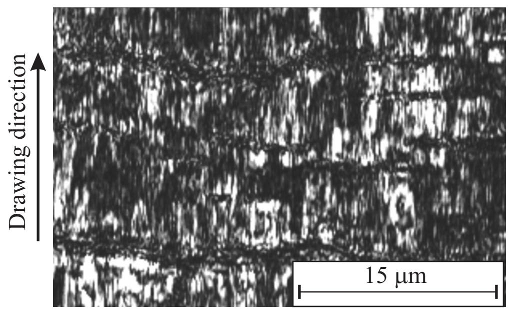

Fig. 1.Through drawing processes implemented oriented structures of a co-extrudered self-reinforced polypropylene tape (confocal laser scanning microscopy)

Performance of self-reinforcement

The paradox of the present self-reinforced composite materials with the single-component system is jointly

Self-reinforced polypropylene sheets

At present, predominantly self-reinforced layered composites are used for the manufacture of polypropylenes. They possess adequate crystallisation behaviour, which is obligatory for the implementation of higher drawing degrees [21]. In addition, sufficient PP-semi-finished products in form of tapes, fibres and foil systems are available [22].

The potential of the mechanical tensile properties of SR-PP composite materials makes them have an advantage over conventional foreign fibre reinforced PP-com-

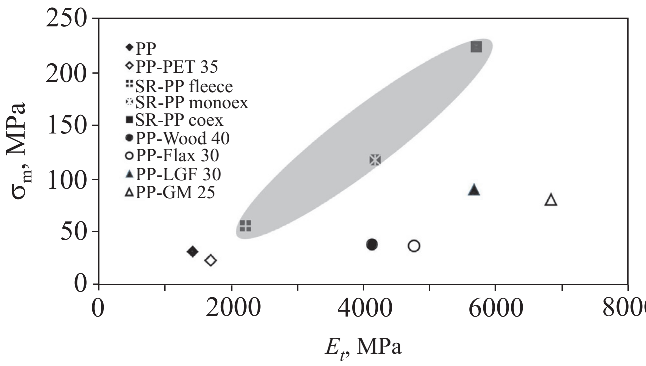

Fig. 2. Mechanical tensile properties of different reinforced polypropylene composites [23]

posites due to a manifold higher strength and stiffness increase, see Fig. 2. Depending on the material structure of the employed self-reinforced semi-finished textile products, a nearly six-fold increase in strength and an increase in stiffness of nearly three-fold can be achieved [24—27]. Thus, a foundation for the substitution of glass fibre reinforced composite materials is provided. A decisive factor regarding the application of self-reinforced thermoplastic composites in semi-structural and structural components is the convincing light-weight construction potential with a low density of σ_{PP} = 0.91 g/cm^3 combined with a superb recycling qualities. By varying the material system, the self-reinforced fibre and tape ratios or the layer configuration and using precisely conducting processing the product properties can be adjusted for almost every designated application.

HOT COMPACTION OF SELF-REINFORCED POLYPROPYLENE SHEETS

Depending on the component thickness and demand profile, PP-semi-finished products are layered and consolidated to sheet composites according to the processing parameters such as pressure, temperature, and time after a completed pre-tempering, see Fig. 3. The formative process can either be carried out via thermoforming after completing hot compaction or by compression molding.

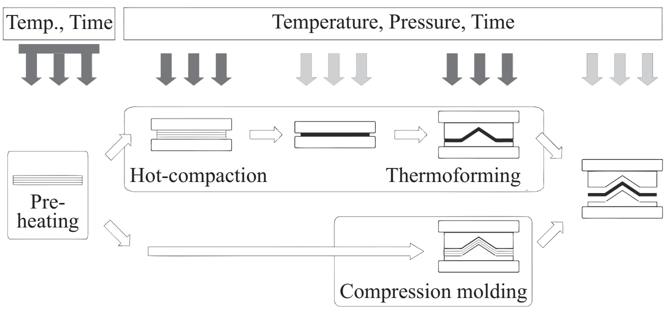

Fig. 3. Processing strategies of self-reinforced layered composites

The thermo-mechanical, sensitive self-reinforcement properties of the semi-finished textile products should be maintained optimally during the whole consolidation process. In order to obtain exceptional layered composite qualities it is necessary to utilise a very precise pressing technology with accurate temperature and pressure conductions. The element of contrast regarding consolidation lies in the necessity to generate enough matrix phase to produce the obligatory interlaminated adhesion without reducing the self-reinforcement ratio significantly [1, 24], see Fig. 4. The latter would result in a loss of the outstanding basic mechanical property. Alcock *et. al.* [25] confirmed that even a matrix content of approx. 10 % is enough to produce very well consolidated layered composites. Moreover, the compaction time, which is de-

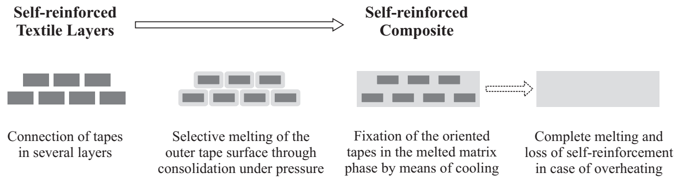

Fig. 4. Schematic diagram of matrix generation via melting of the self-reinforced fibre surfaces during consolidation

pendent on temperature-pressure-combination must be chosen. Its practice, the production of the extremely processing-sensitive self-reinforced composites embodies a big challenge in overcoming equipment and technological problems. Depending on the matrix system, the kind of semi-finished products and the layer configuration, the temperature setting should be chosen so that at least a selective melting of the outer fibre surface for the generation of a matrix is possible. The temperature cannot be too high so as to cause a complete melting of the self-reinforced polymer phase [26]. The interacting pressure setting must be able to resist the temperature-induced relaxation process and the setting in usually established so as to enable at least one sufficient composite consolidation. This should lie below the limit, in which damaging melt-flow-processes occur [26].

Depending on the component geometry which is to be depicted, a wrinkle-free molding must be guaranteed [27]. In order to produce visually attractive components, suitable cut designs and draping techniques should be considered. Moreover, if applicable, conducting devices for semi-finished textiles or tool adapted clamping frames should be employed [28—30].

COMPONENT APPLICATION OF SELF-REINFORCED PP-SHEETS

Up until now economic and prementioned processing-technological criteria have prevented a widely established employment in open fields of application. Nevertheless, complete prototype series have been performed, which are seldom transferred to low or intermediate series applications. The self-reinforced underbody shield of the Mercedes A-Class can be mentioned as an example in this context [31].

Although the exceptional material properties and the corresponding, competition-free recyclability are convincing factors, very few components are transferred into commercial application series. The foundation was laid by Legacy Paddlesports with Ultimate™ Kajak in 2007 [32]. Two years later, Samsonite® introduced the luggage system Cosmolite, which is made of Curv® [33]. Since their introduction into the market, both products have proven that the combination of low component weight, exceptional impact strength and stiffness behaviour are economically compatible with this new material technology.

In order to fulfill the high technological standards of competition, extensive, economic solutions for the further integration of this material technology must be found. It is necessary to acquire additional knowledge for the implementation of extra functions in structural components, so as to be able to exploit new series applications.

PROCESS-INDUCED PROPERTY GRADATION

Process-induced gradation makes it possible to implement various final material properties in the completed component without having to perform additional processing steps. During the hot compaction and thermoforming or compression molding, the self-reinforcement is extensirely modified by thermo-mechanically coupled consolidation processes so that differential component properties are obtained [34]. Without having to employ extrinsic fibre reinforcement or indigenous modifications in the layering concept to utilise an armouring process, an additional component function can be achieved with merely a pure process conduction. The self-reinforced polymer phase is influenced so significantly in this way that later component properties can be illustrated according to their functionalities. Moreover, the low material weight and the exceptional recycling qualities remain preserved.

Mechanical gradation

Mechanical gradation is achieved using a local pressure increase [35, 36]. For the sake of convenience, stamp and hole matrices are used to realise the necessary pressure variation. By applying cavity matrices, structural areas can be achieved alongside hot compacted composite areas. In turn, for example, energy absorbing crash pads can be obtained alongside impact-resistant links, see Fig. 5. Locally differentiated pressure conditions or set gaps can be regulated directly without having to use manual inlay matrices if pressure and distance regulated pressing units with specially modified tools are employed. The pressure variation, which is put into effect on the textile layers, influences the selective melting of the tapes and fibres. Furthermore, it effects the melt-flow--processes, the matrix percolation, the interlaminate adhesion and the crystallisation processes [23]. The desired component properties can therefore be mechanically-induced according to their functionalities.

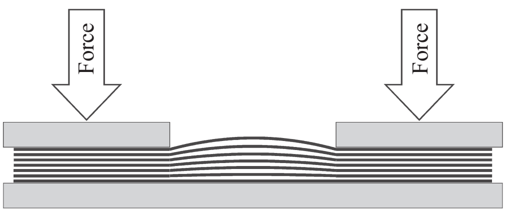

Fig. 5. Operating principle of mechanical gradation based on a hole matrice for the implementation of energy-absorbing crash-pads

Thermal gradation

Principally, thermal gradation can be applied during all heating processes. This concerns the partially masked pre-heating sequence and either an adapted hot compaction process in combination with thermoforming or a compression molding process [35, 36]. It is also important to achieve a temperature difference across the component surface in this case, in order to implement a property gradient.

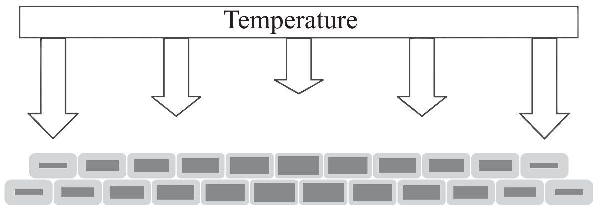

Fig. 6. Uneven melting of tape and fibre surfaces using thermal gradation during pre-heating or the consolidation process

The set temperature gradient significantly determines the selective melting behaviour of the tapes and fibre surfaces, thus also the ratio of self-reinforced polymer phases to the unreinforced matrix phase, see Fig. 6. The melting degree of the semi-finished textile products affects the quality of the matrix impregnation respective of the interlaminate stiffness. Moreover, it also effects the remaining part of the self-reinforced polymer phase. Thus, the basic mechanical properties of the layered composites are substantially influenced.

Verification of process-induced gradation

During thermo-mechanical gradation, both concepts are deployed, which, in turn, result in interacting temperature and pressure influences leading to a strong manifestation of the desired property gradient [35, 36].

By means of Young's modulus E_t the gradation ability can be shown exemplarily in Fig. 7. Eighteen layers of co-extruded tape fabric of the type Pure® from the company Lankhorst [37] were consolidated to graded, layered composites. Four divergent material zones were implemented using two different temperature ($\downarrow T$ = 165 °C, $\uparrow T$ = 195 °C) and pressure configurations ($\downarrow p$ = 2.1 MPa, $\uparrow p$ = 3 MPa), see Table 2. Thermal gradation in the half-masked IR-pre-heating station benefitted/promoted the tempering zones (cold-warm). Highly stiff (up to 6000 MPa at $\downarrow T$ with $\uparrow p$) as well as ductile composite properties (<3000 MPa at $\uparrow T$ with $\uparrow p$) can be reproduced along the surface of the component and verified.

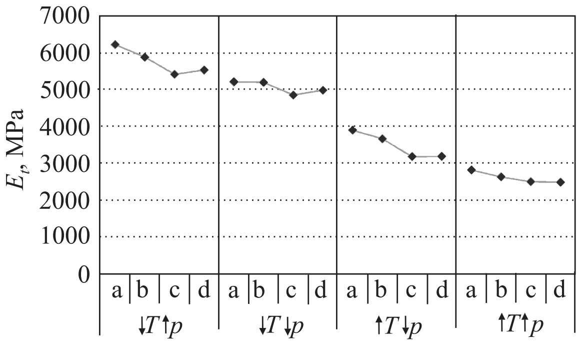

Fig. 7. Young's modulus — distribution across the component surface with four different thermo-mechanically graded property zones

T a b l e 2. Process parameters for realisation of divergent property gradients through variation of compression molding temperature and pressure

Infrared pre-heating			Compression molding				
temperature	time	pro-rate mask time	time	temperature	temperature	pressure	pressure
T_{IR}, °C	t_{IR}, s	t_{mask}, %	t_c, s	T_1, °C	T_2, °C	p_1, MPa	p_2, MPa
120	180	100	240	165	195	3	2.1

It has been verified, that apart from the stiffness properties, the stiffness, impact strengths and material damping can also be locally adjusted in order to illustrate further process-induced component functionalities [34, 38].

CONCLUSIONS

Self-reinforced PP-sheets are distinguished by their extraordinary impact strength and stiffness behaviour for utilisation in semi-structural and structural applications. Nevertheless, the transfer of this material technology to series applications is comparably slow, due to certain processing technology standards which must be fulfilled.

If these difficulties can be overcome a further key to establishing this material technology in conventional series applications in process-induced property gradation will be on hand. The targeted, locally dependent adaptation of the final material properties to the later component application makes it possible to achieve a higher component surplus value in regards to the processing technology. This surplus value may necessitate a demanding processing technology, but the functional gradation gained in this way fulfills a broader, more complex demand profile. Ultimately, economic establishment is enabled.

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