Research Article

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Modification of self-reinforced composites (SRCs) via film stacking process

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Abstract: This work presents the mechanical behavior of self-reinforced composites (SRCs) manufactured and modified via film stacking. For modification, interleaved films made of polypropylene (PP), a thermoplastic elastomer and a polyolefin engage were combined in different ways to induce the elastic modifier into the matrix material. The content of modifier was also varied in two ways. First, the films were produced out of a single material and second out of a compound. So, the same content of modifier was implemented in two different ways. It is shown that, in case of this research, only the kind of modifier and the content but not the way of implementation are responsible for the mechanical behavior of SRCs. It is shown that the modification can adjust the tensile strength, tensile stiffness and impact properties in a broad range. It is also shown that different mechanical properties of the composite can be predicted by a regression model that uses the Shore A hardness and the content of modifier.

Keywords: elastomer modification; film stacking; hot compaction; self-reinforced; single polymer composite.

1 Introduction

Contrary to the case for conventional fiber-reinforced materials, self-reinforced composites (SRCs) are made up to 100% of the same material. The basic principle involves producing fibers, increasing their tensile strength and stiffness through orientation of the macromolecules in the fiber direction and then embedding them in a matrix of the same polymer which has not undergone any orientation.

One of the first publications on SRC was authored by Hine et al. (1993). While studies had been conducted of the morphology in fibers or injection molded materials beforehand, Hine et al. were some of the first to discover the potential of the highly oriented thermoplastic fibers for manufacturing a composite. They succeeded in manufacturing the composite through hot compaction of polyethylene fibers. In 1995–1997, hot compaction tests were also carried out with other polymers (Rasburn et al. 1995), including polyethylene terephthalate (PET), polyethylene (PE) (El-Maaty et al. 1996) and Polymethylmethacrylate (PMMA) (Wright et al. 1997). Further tests followed for the mechanical characterization of SRCs (Ismail et al. 2001; Le Bozec et al. 2000). Hine et al. (2003) studied the influence of different fabrics on the composite and reported that an estimate of the mechanical properties can be achieved on the basis of simple mixing rules. This was followed, in 2005, by SRCs in cellulose (Gindl and Keckes 2005). In the following years, numerous studies were conducted on the process and the influence of the process parameters on the mechanical properties of SRC (Alcock et al. 2007, 2008; Banik et al. 2008, Bárány et al. 2006a,b, 2007; Hine and Ward 2006; Izer 2010). An overview of further research work up to 2010 is presented in Kmetty et al. (2010). After research in the period up to 2010 had concentrated primarily on polypropylene (PP), partly on account of its good impact properties, some researchers then turned to the processing of SRC based on PET (Andrzejewski et al. 2016; Chen 2011; Jerpdal and Åkermo 2014; Jerpdal et al. 2016, 2018; Poulikidou et al. 2016; Schneider et al. 2013; Zhang and Peijs 2010). Other focal points of current research include hybridization with carbon fibers of SRC based on PA (Hine et al. 2014, 2017) and PP (Mesquita et al. 2018; Selezneva et al. 2018; Tang et al. 2018) and hybridization with steel fibers (Swolfs et al. 2017).

A team of researchers investigated the possibility of grading SRC (Biermann et al. 2012; Bledzki et al. 2008, 2012; Heim et al. 2012; Paßmann 2009; Schöppner et al. 2013). This involves selectively adjusting the mechanical properties of

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certain areas of the SRC via the process sensitivity (Heim et al. 2013a,b, 2014; Rohde et al. 2014). In the case of SRC based on PP, in particular, the locally differing process parameters of pressure and temperature allow the material properties within a component to be varied over a wide strength, stiffness and impact range (Ries 2015). Grading opens up a broad spectrum of technical options, but this is always associated with an increased outlay on process control and additional costs for the tool technology.

In this paper, studies on the modification of the mechanical properties of PP-based SRCs are presented. The modification of the mechanical properties is induced by the implementation of some elastomer modifier into the matrix phase of the SRCs. The elastomer modifier is implemented via film stacking process. The film stacking process is a common method to produce SRCs (Bárány et al. 2006; Karger-Kocsis et al. 2010; Wang et al. 2015) and was used by several researches for modification of matrix materials in SRCs (Abraham et al. 2009; Bárány et al. 2009; Wang et al. 2014, 2015; Wu et al. 2012; Zhang et al. 2009). A more detailed description of the film stacking process is given in Kmetty et al. (2010). The modification of mechanical properties of SRCs via process parameter has already been investigated in the past (Heim et al. 2013a,b, 2014; Rohde et al. 2014). As the process conditions have to be adjusted very carefully to modify SRCs with process conditions, this research presents a new way of modification via film stacking with elastomer modifier.

The following theses were put forward at the start of the experiments:

– The mechanical properties can be selectively influenced and adjusted through use of the modifiers

– Since the fabrics determine the strength and the stiffness to a decisive extent, the impact properties can be adjusted largely independently of the strength and stiffness in the fiber direction.

2 Experimental studies

2.1 Materials

2.1.1 Fabric (PP)

The SG 30/30 fabric made by Low & Bonar NV, Zele, Belgium, is a fabric comprised of tapes woven in a 2/2 twill weave. The tensile strength of the fabric to EN ISO 10319 is specified as 30 kN/m in the warp and weft directions. The elongation is 20% in the warp direction and 11% in the weft direction. The thickness under 2 kPa is given as 0.9 mm and the weight per unit area is 124 g/m^2. The fabric is normally used as a geotextile and comprises 100% PP colored with carbon black. Carbon black is known as a beta nucleation agent with effects on the mechanical properties of PP, i.e., a lower stiffness, and a higher elongation. As the carbon black is inside the fabric and the amount of carbon black stays constant, there is no closer look to the effect of carbon black in this paper. As the fabric is treated the same for each modification and the fabric is not part of this investigation, there will also be no closer look on this in this paper. In what follows, the warp direction is designated the machine direction (MD) and the weft direction the transverse direction (TD).

2.1.2 Polypropylene (PP)

This PP is purchased in the form of plastic pellets (trade name PP575, Sabic Polyolefine GmbH, Gelsenkirchen, Germany) and is used both for producing the samples for compounding and for extrusion. The PP has an melt flow rate (MFR) of 10.5 g/10 min (at 230 °C and 2.16 kg), a density of 0.905 g/cm^3, a tensile strength of 36 MPa and an elastic modulus of 1800 MPa.

2.1.3 TPE7

The thermoplastic elastomer (TPE) used is a styrene block polymer based on a styrene-ethylene-butylene-styrene block copolymer (SEBS). This TPE (trade name TPE7MED, Kraiburg TPE GmbH & Co. KG, Waldkraiburg, Germany) is used for seals, flexible connections and valves among other things. It has a hardness of 68 Shore A and a density of 0.89 g/cm^3. Its tensile strength is specified as 13.0 MPa and its elongation at break as 800%. The TPE is suitable for injection molding and extrusion and was used for the production of films.

2.1.4 TPE4

The TPE used is a styrene block polymer based on a SEBS. This TPE (trade name TPE4MED, Kraiburg TPE GmbH & Co. KG, Waldkraiburg, Germany) belongs to the same product range as the TPE7MED and has fundamentally similar properties as far as its field of application is concerned. With a hardness of 36 Shore A and a tensile strength of 8 MPa, it does, however, differ clearly from TPE7MED. The elongation at break is also given with 800%. TPE4MED has a density of 0.88 g/cm^3 and was also used for the production of films.

2.1.5 POE

This polyolefin elastomer (POE) (trade name: ENGAGE POE 7270, Dow, Midland, USA) is used to increase the impact strength of polyolefins. It has a density of 0.88 g/cm^3 and an MFI of 0.8 g/10 min (at 190 °C and 2.16 kg). Its hardness is specified as 80 Shore A, its tensile strength as 13.9 MPa and its elongation at break as >600%. For the modification of polymer films in these investigations, it was compounded with PP in different proportions and extruded into films.

According to the manufacturers, all the materials used are compatible with PP. They can be compounded into PP and fundamentally adhere to PP. Adhesion between the fibers and the matrix will significantly influence the mechanical properties of the composites. As the adhesion properties are not only based on chemical composition, but also depend to a large extend on the process parameters, more detailed investigations are needed and will be subject for future research. Therefore, the adhesion properties are not investigated in this paper. Adhesion between TPE4, TPE7 and POE is not necessary, since these materials are not used in combination with each other.

2.2 Processing

Different methods were employed in this work for modifying the matrix in an SRC. All composites were made by the film stacking method. For each SRC an amount of seven fabric layers and eight film layers were used. First, a composite with eight films of PP between each layer was made. For the modification, individual films of the eight layers were replaced by films made of one of the elastomer materials POE, TPE7 and TPE4 in different variations. The process is shown in Figure 1. The exact layering will be presented later.

Additionally, the way in which the elastomer component is introduced was varied. The ratio was kept the same, but a compound was created beforehand from the PP used and the POE, which was then extruded into films (Figure 2). The films were similarly produced with three different

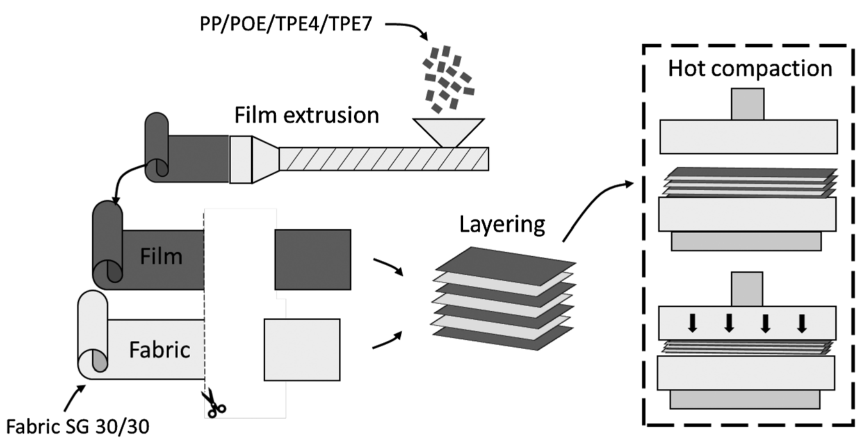


Figure 1: Process used to produce self-reinforced composites (SRCs) with film extrusion, layering and hot compaction process.

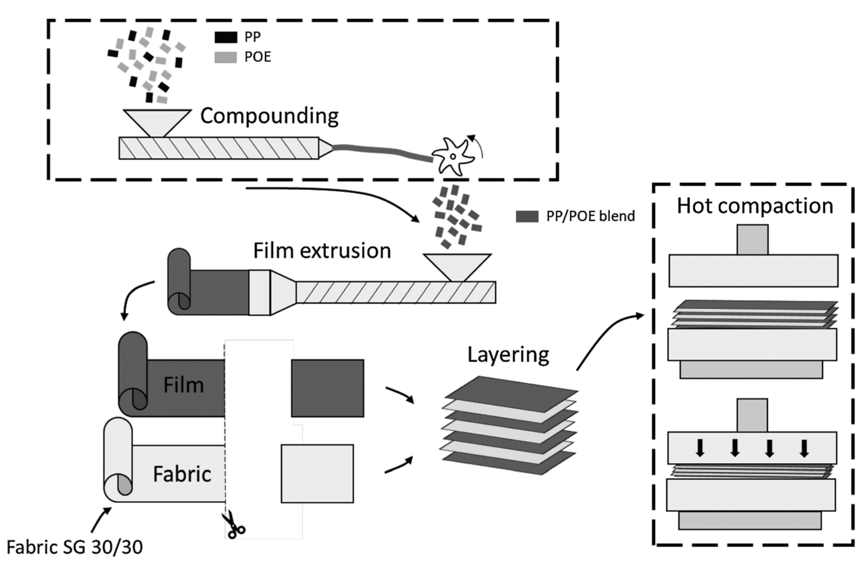


Figure 2: Process used to produce self-reinforced composites (SRCs) with compounding process, film extrusion, layering and hot compaction process.

contents of modifier and they replaced all the PP films that had been previously used.

2.2.1 Compounding

Three different mixing ratios of PP and POE were produced. To ensure homogeneity, the compounds were blended proportionately by weight and homogenized on a twin-screw extruder. Processing was performed with a barrel temperature of 195 °C on a co-rotating twin-screw extruder (L/D = 32:1, D = 25 mm), model Haake Rheomax, from Thermo Fisher Scientific Inc., Waltham, USA. The screw configuration is shown in Figure 3.

The screw speed was 200 min^{-1} with a throughput of approximately 1.65 kg/h. The extrudate is then granulated so that it can be further processed. The composition of the compounds is listed in Table 1.

The PP used without modifier was similarly processed on the twin-screw extruder so as to exclude any influences of the compounding. To control the results of compounding, mechanical testing of the material was made. The results are shown in Section 3.

2.2.2 Extrusion

The films were produced on a single-screw extruder (L/D = 24:1) with a screw diameter of 14 mm and a slot die in combination with a downstream laboratory calender from Collin Lab Pilot Solutions GmbH, Maitenbeth, Germany. The single-screw extruder and the slot die were heated to 210 °C to produce the film. The extruder temperature was adjusted for PP and kept constant for each material. The calender rolls were heated to 60 °C. The take-off speed was adjusted individually for each material to keep the thickness of films constant and was varied between 1.6 and 1.9 m/min. The cooling conditions were kept constant. The films will be melted again after extrusion in the following hot compaction process. It can be assumed that the prior thermal history of the films is of no relevance.

Table 1: Overview of the different compounds used to produce the films.

No.	PP content (%)	POE content (%)	Name
1	25	75	75%POE
2	50	50	50%POE
3	75	25	25%POE
4	100	0	PP

2.2.3 Layering

As described in Section 2.1, different concepts are pursued for the modification. On the one hand, use is made of films in POE, TPE7 and TPE4. These films are placed between the layers of fabric in different combinations with the PP film. On the other hand, films made of PP with different POE contents are used. Figure 4 shows the layering of the fabrics in the film stacking process and the elastomer modifier content which is given as the total weight of the interleaved films. The fabric layers were not taken into account, thus the content of modifier only refers for the matrix of the SRC.

As the individual layers were stacked, they were rotated through 90° compared with the previous layer of the same material. This is essential in the case of the fabrics used, because although they are specified with the same tensile strengths by the manufacturer, they have a different number of tapes in the warp and weft direction and a differing degree of undulation, which leads to very different stress–strain behavior in the MD and TD in the hot compacted composite. Figure 5 shows, by way of example, the stress–strain curve for composites made of SG 30/30 produced without rotation of the individual layers through 90°.

2.2.4 Hot compaction

The hot compaction is performed in a hydraulic press made by OTT (Lambach, Austria). Use is made of an electrically heated tool with an area of 150 × 150 mm^2. The cooling medium is water that flows through the cooling channels of the mold. The mold attains a mean heating rate of 10 K/min (80–180 °C) and a mean cooling rate of 20 K/min (180–80 °C).

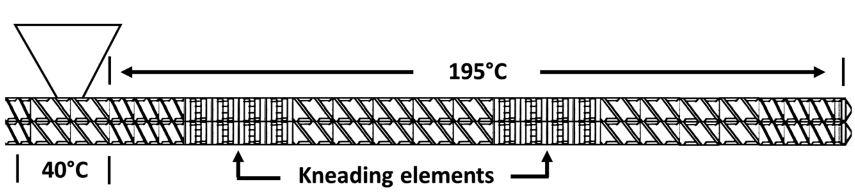


Figure 3: Screw configuration of the twin-screw extruder used for compounding.

Modification with different films

Modifier →		none	POE / TPE7 / TPE4		
7	Film	PP	PP	PP	PP
6	Fabric	SG 30/30	SG 30/30	SG 30/30	SG 30/30
5	Film	PP	PP	PP	POE/TPE
4	Fabric	SG 30/30	SG 30/30	SG 30/30	SG 30/30
3	Film	PP	PP	POE/TPE	POE/TPE
2	Fabric	SG 30/30	SG 30/30	SG 30/30	SG 30/30
1	Film	PP	POE/TPE	POE/TPE	POE/TPE
0	Fabric	SG 30/30	SG 30/30	SG 30/30	SG 30/30
1	Film	PP	POE/TPE	POE/TPE	POE/TPE
2	Fabric	SG 30/30	SG 30/30	SG 30/30	SG 30/30
3	Film	PP	PP	POE/TPE	POE/TPE
4	Fabric	SG 30/30	SG 30/30	SG 30/30	SG 30/30
5	Film	PP	PP	PP	POE/TPE
6	Fabric	SG 30/30	SG 30/30	SG 30/30	SG 30/30
7	Film	PP	PP	PP	PP
Content →		0 %	25 %	50 %	75 %

Modification with films made with different content of POE

Modifier →		none	POE Compound		
7	Film	PP	25%POE	50%POE	75%POE
6	Fabric	SG 30/30	SG 30/30	SG 30/30	SG 30/30
5	Film	PP	25%POE	50%POE	75%POE
4	Fabric	SG 30/30	SG 30/30	SG 30/30	SG 30/30
3	Film	PP	25%POE	50%POE	75%POE
2	Fabric	SG 30/30	SG 30/30	SG 30/30	SG 30/30
1	Film	PP	25%POE	50%POE	75%POE
0	Fabric	SG 30/30	SG 30/30	SG 30/30	SG 30/30
1	Film	PP	25%POE	50%POE	75%POE
2	Fabric	SG 30/30	SG 30/30	SG 30/30	SG 30/30
3	Film	PP	25%POE	50%POE	75%POE
4	Fabric	SG 30/30	SG 30/30	SG 30/30	SG 30/30
5	Film	PP	25%POE	50%POE	75%POE
6	Fabric	SG 30/30	SG 30/30	SG 30/30	SG 30/30
7	Film	PP	25%POE	50%POE	75%POE
Content →		0 %	25 %	50 %	75 %

Figure 4: Layering of the produced self-reinforced composites (SRCs) with different content of modifier given as weight in percentage of the total weight of all interleaved films (matrix).

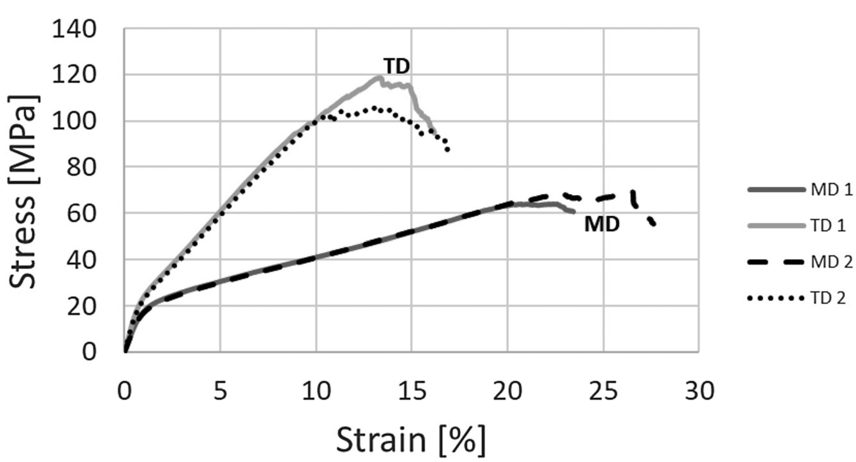


Figure 5: Stress–strain curves for self-reinforced composite (SRC) based on SG 30/30 with non-rotated fabric layers (180 °C|2.5 MPa).

All the SRCs are produced with identical process parameters. The stacked fabric and film are placed in the cold mold and a pressure of 7 N/mm^2 is applied. They are heated up to 180 °C under pressure. The air is evacuated through the fabrics before melting of the films takes place. So, no air is left between the fibers in this process. Once the compaction temperature has been attained, a holding time of 2 min is observed to ensure uniform heat distribution in the thickness direction before initiating mold cooling (Paßmann 2009; Ries 2015). The composites are removed from the mold when the core temperature is below 80 °C. To ensure a uniform heat distribution across the compacted area, temperature was monitored with thermocouples in different positions (center and at the edges). An offset of up to −5 °C was detected in 10 mm wide edge areas and is considered during sample preparation.

3 Sample preparation, measuring and results

3.1 Tensile tests of films

To allow the individual films to be evaluated, and especially those films that were produced from the compounds, they were characterized in a tensile test. Five specimens used were cut out of the middle of the film in the MD and TD, as shown in Figure 6.

The prepared film strips are 100 × 20 mm in size. Special film clamping units are used for the tensile tests.

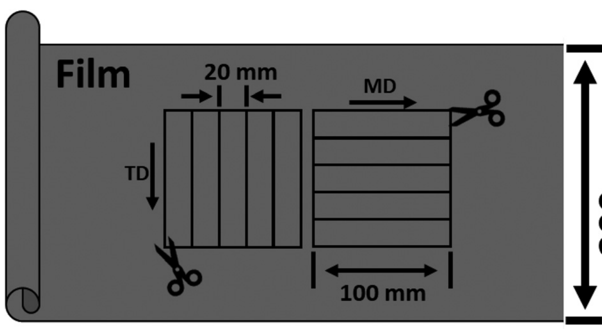


Figure 6: Different stress–strain curves for self-reinforced composite (SRC) based on SG 30/30 with non-rotated fabric layers (180 °C|2.5 MPa).

The free clamping length for the measurements conducted is 60 mm. The elastic modulus is determined at a speed of 1 mm/min. Once the measurement of the elastic modulus has been completed, the testing machine switches to a speed of 10 mm/min. This procedure corresponds to DIN EN ISO 527 for tensile testing. Figure 7 shows the characteristic values determined and a number of sample curves for the films that were tested.

The films used for modification have a measured elastic modulus of 17.6 MPa (POE), 16.7 MPa (TPE7) and 1.4 MPa (TPE4). These characteristic values were determined from three MD and three TD measurements. The characteristic values determined in the tensile test for the films from the previously compounded material show that the stiffness decreases with increasing modifier content. The elastic modulus falls from 1414.4 MPa (PP) to 176.43 MPa (75%POE).

As expected, the results show that the extrusion causes direction-dependent behavior in the films. As mentioned above, the films are molten again in the hot compacting process, so the thermal history of the films does not influence the properties of the SRCs and can thus be neglected.

3.2 Tensile tests of SRCs

The tensile tests are made on specimens prepared from the hot compacted composites with the aid of a computerized

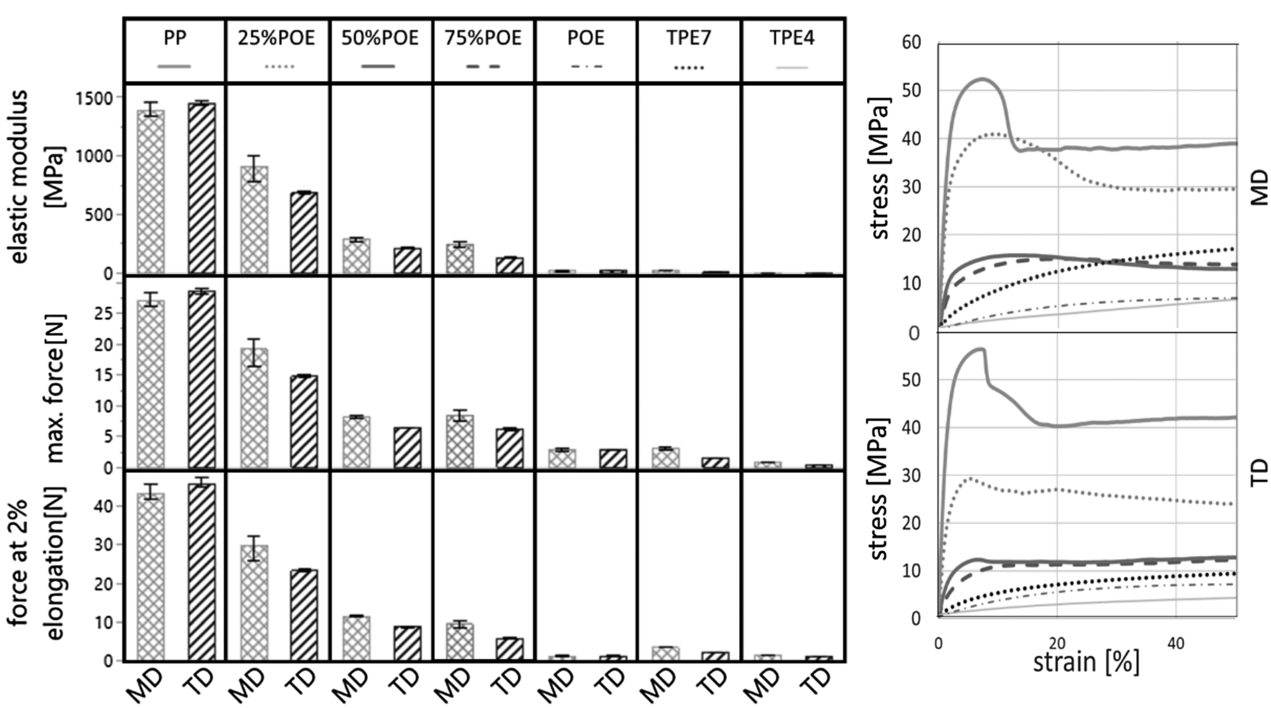


Figure 7: Results of mean tensile test on the films with a different modifier content.

numerical controled (CNC) milling machine. To do this, the 150 × 150 mm plates are first cut into strips of 23 mm and then milled into the shape of a 1 A tensile test rod (see DIN EN ISO 527) using a CNC milling machine. To ensure that each specimen has undergone the same temperature treatment, 15 mm broad edge areas in parallel direction to specimen were removed. Even with regular 90° rotation of the fabric layers, the uneven number of layers and the resulting presence of a central layer mean that an influence on the stress–strain behavior can be expected. The influence of the orientation of the central layer was not studied in this paper. In order to exclude any influence for the tests, the test specimens are always prepared and tested with the central layer in the MD. For the reference test specimens that were produced with the PP films, an elastic modulus of 2723 MPa and a tensile strength of 86 MPa were determined. When observing the measured characteristic values for stiffness and strength using the individual modifiers, it becomes clear that all the modifiers have the effect of reducing stiffness. They also affect the tensile strength, which declines as the modifier content increases. The results shown in Figure 8 for the specimens produced by film stacking show that it was possible to vary the elastic modulus of the SRCs by 88%, while the tensile strength of the composites is modified by up to 74%. The composites with a 75% content of TPE4 had the lowest stiffness. The elastic modulus in this case was measured at 315 MPa and the tensile strength at 23 MPa. The results shown do not yet include any composites made with “POE compound”.

The modification achieved takes different forms when the three different modifiers are used. With TPE7 and TPE4, an increasing modifier content results in a lower stiffness in the overall composite, while this behavior is less pronounced with the use of POE. The effect on tensile strength is also clearer when TPE4 and TPE7 are used rather than POE.

By varying the way in which the modifier POE is introduced, a check was also conducted to see whether a significant change results in the mechanical properties. A direct comparison of the methods of introduction is shown in Figure 9.

The comparison shows that, for the determined stiffness, both the height and range in which the values were measured are comparable. The biggest difference between the variants of “POE film” and “POE compound” is seen with a modifier content of 75%. The absolute range within which the measured elastic modulus is located is 228 MPa, corresponding to a deviation from the mean value of ±6.2%. A similar picture emerges for the tensile strength, where the maximum difference between the measured characteristic values with a modifier content of 75% shows an absolute range of 13.2 MPa. This corresponds to a

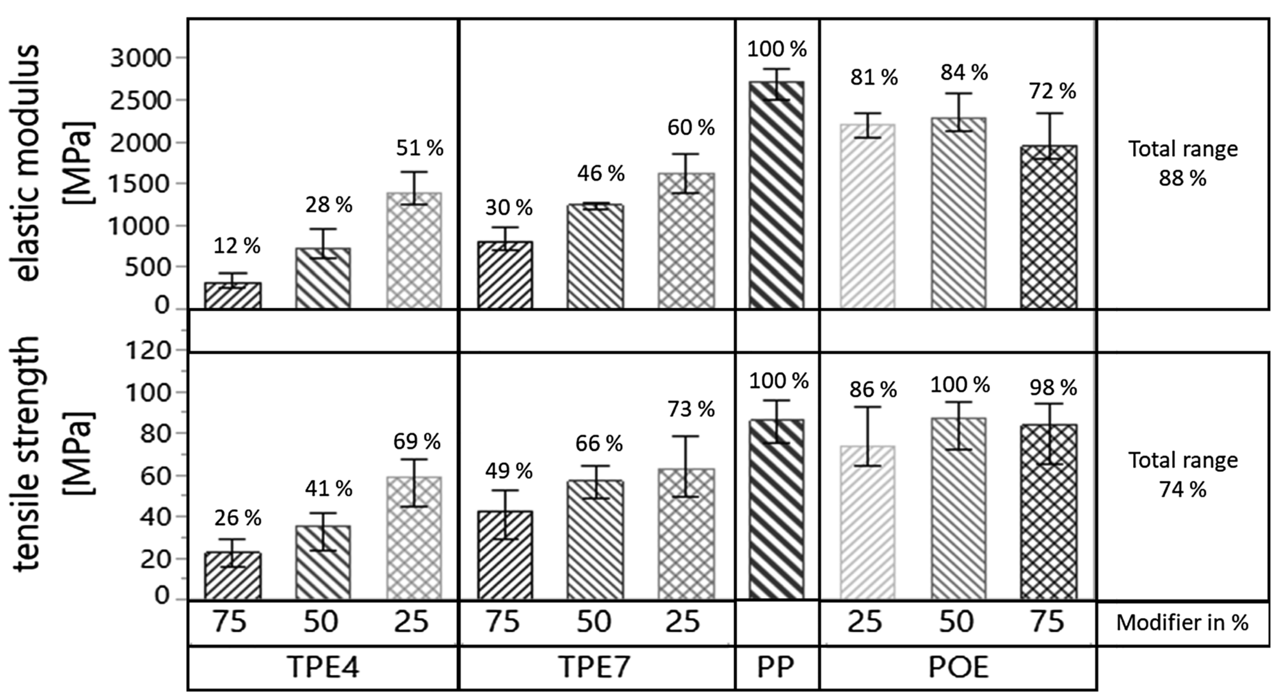


Figure 8: Results of tensile tests on self-reinforced composites (SRCs) made by the film stacking method with different modifier contents given in MPa and in percentage (in relation to polypropylene).

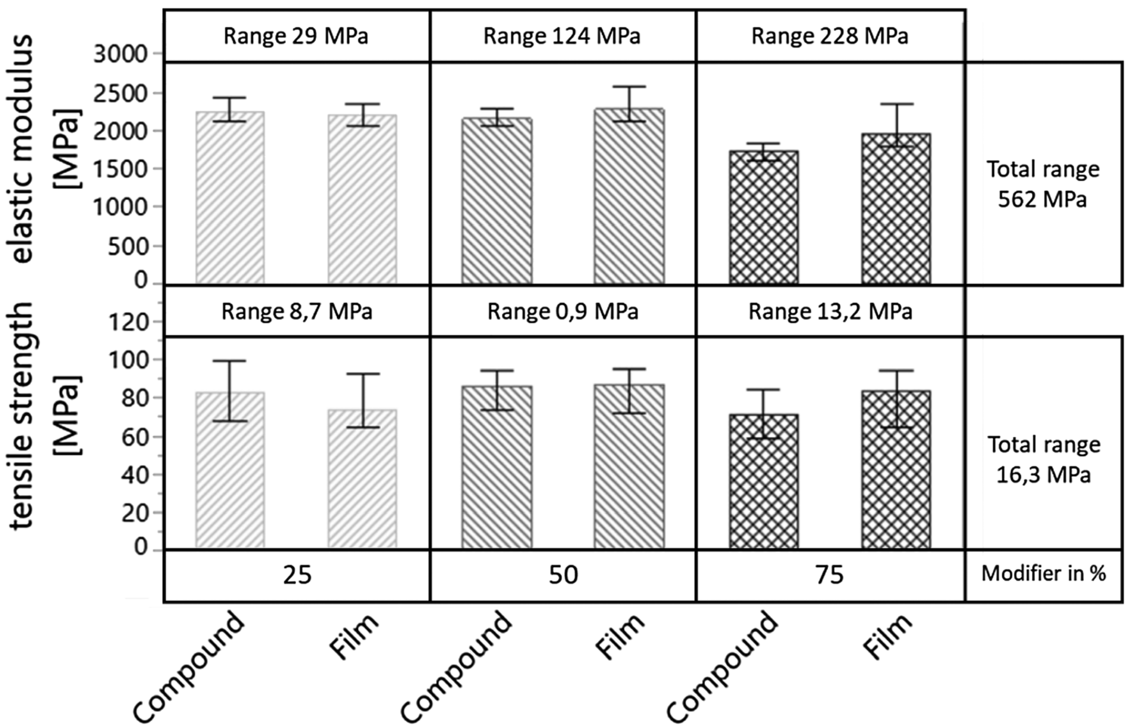


Figure 9: Comparison of the different ways of modifier implementation – results of tensile tests on self-reinforced composites (SRCs) made by the film stacking method.

deviation of ±8.5% from the mean value. Despite the apparently higher strength and stiffness that result with the “POE film” variant, no general statement can be made on this, since this tendency cannot be observed with either a 25% or 50% modifier content. It can thus initially be assumed that the method of introduction has no significant influence on the mechanical parameters of stiffness and strength in the SRCs.

3.2.1 Impact tests

The impact tests are conducted with test specimens measuring 65 × 65 mm that have been cut out of the compacted composite sheets with a thermoplastic circular saw. Four specimens (65 × 65 mm) were cut out of each composite sheet (150 × 150 mm) and tested on an impact test bench from Ceast, Martinsried, Germany. The impact energy applied was set at a weight of 3.65 kg over a drop height of 14 cm and amounts to 5 J. The impactor used has a semi-circular geometry and a diameter of 20 mm. The impact absorber used has an inside diameter of 40 mm. The impact test employed here is a non-standardized test method that is used specially to assess materials with regard to their behavior under impact point loading. Specific material characteristic values such as impact stiffness (measured between 4 mm and 8 mm deformation), stored energy and dissipated energy are determined. Following this, the damping behavior of the material can be calculated.

Figure 10 shows the determined damping, the maximum deformation and the maximum impact force, also given as percentage in relation to samples made with PP films, together with a number of force-deformation curves set out by way of example. Here again, the results are presented without the “POE compound” variant in the first instance.

The modification attained in the impact curve progression diagrams (right) is clear to see. The progression curves are given by way of example in each case. Each diagram shows a progression curve for the composites produced by film stacking, with pure PP film as a reference. The impact curves are seen to flatten off as the modifier content increases. In other words, the maximum impact force falls and the maximum deformation increases. This applies to all modifiers to differing extents. The maximum deformation is increased by 35% (compared with the composites with pure PP film). The maximum impact force is reduced by up to 26%. The modifiers are seen to have the biggest influence on the measured damping. This is increased by up to 218%. The impact stiffness falls by up to 58% with an increasing share of modifier. A comparison of the different implementation methods of “POE film” and “POE compound” reveals only a slight influence for the impact tests too. Figure 11 shows the measured parameters.

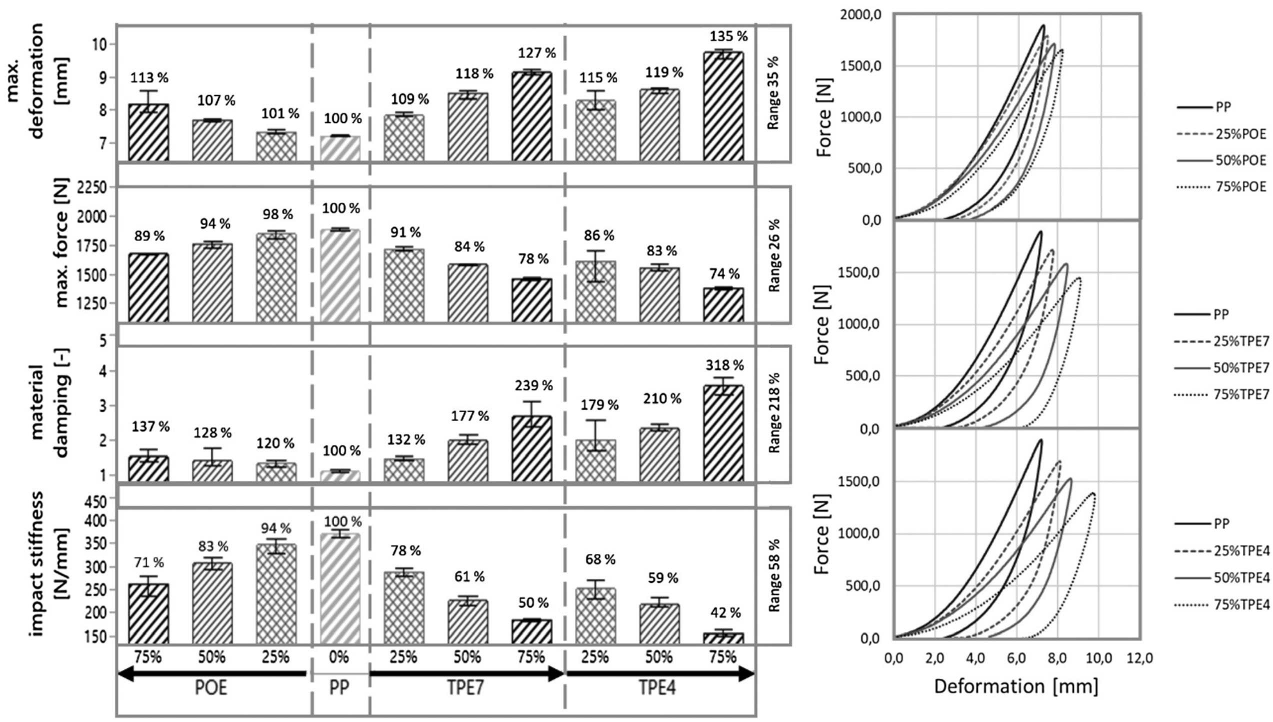


Figure 10: Results of the 5 J impact tests (left) and sample curves for the tested composites (right).

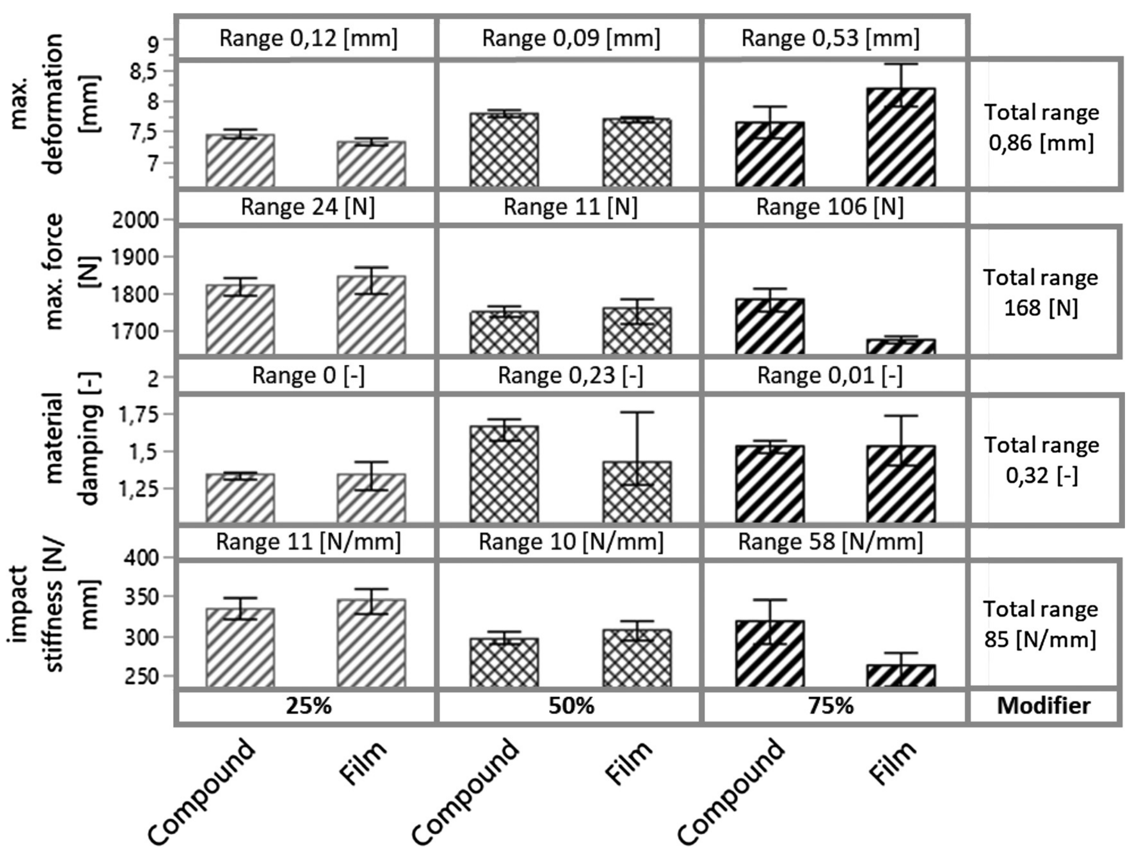


Figure 11: Comparison of the results of 5 J impact tests on the different implementation variants of “POE film” and “POE compound”.

The maximum deformation measured on the "POE compound" composites is 0.11 mm lower on average. The measured maximum force for the "POE film" variant is 23.7 N lower on average. The parameters of material damping (0.08 [–]) and impact stiffness (12.33 [N/mm]) similarly only differ slightly from one another on average. Expressed in terms of the respective mean values, these are fluctuations that are less than ±3%. Observing the stiffness, it would appear that a higher impact stiffness is determined for the "POE film" variant with a modifier content of 25% and 50%. If the modifier content increases further, however, this effect is reversed. Consequently, this influence, if it does exist, cannot be captured in the measurement series performed here. In what follows, the results of the tests are used in summarized form as "POE film" and "POE compound".

3.2.2 Correlation between mechanical properties and modifier content

In order to be able to better assess the correlation between the mechanical properties and the content of the individual modifier employed, Figures 12 and 13 show the mechanical characteristic values plotted over the percentage of modifier. At least five specimens were measured in each case. The

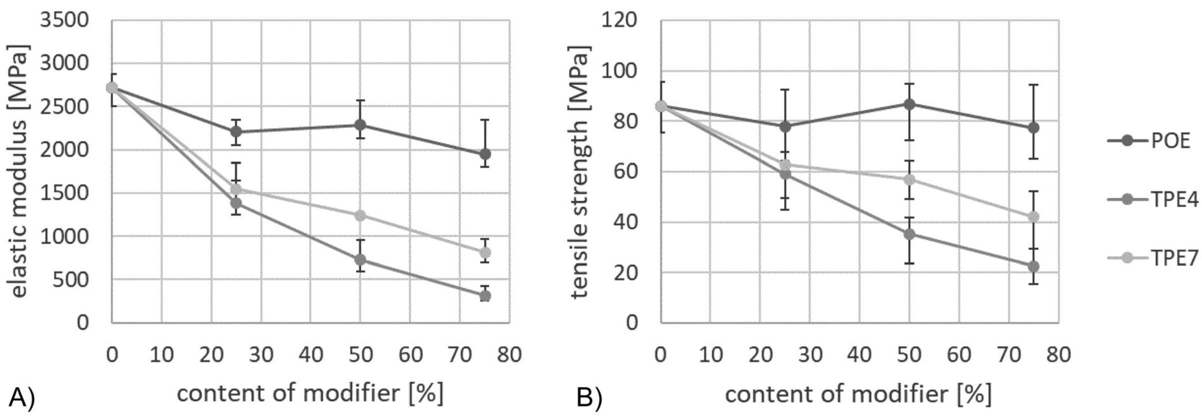


Figure 12: Tensile properties. Elastic modulus (A) and tensile strength (B) in relation to the modifier content for the different modifiers POE, TPE4 and TPE7.

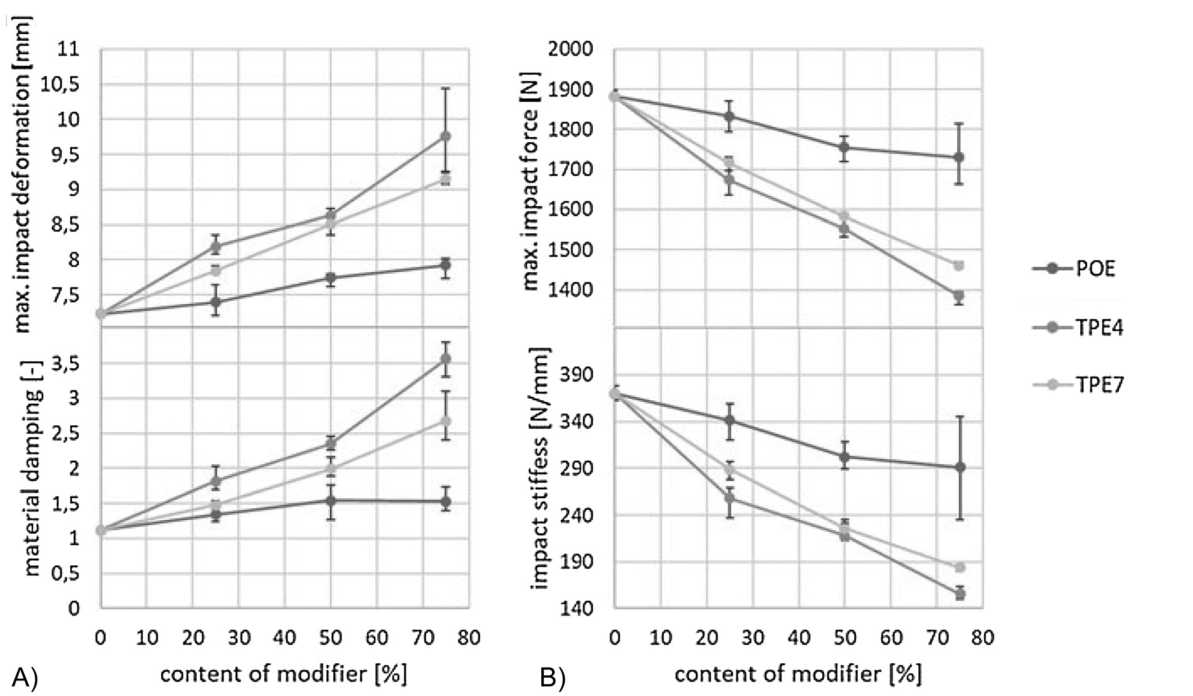


Figure 13: Impact properties. Max. impact deformation, material damping (A), max. impact force and impact stiffness (B) in relation to the modifier content, measured in an impact test (5 J) for the different modifiers POE, TPE4 and TPE7.

error bars drawn in show the minimum and maximum value of the individual test series with identical parameters.

The stiffness of the composites is seen to fall as the modifier content increases. It is also clear that using a modifier with a lower elastic modulus leads to a greater reduction in the stiffness of the composite, as had been expected. This correlation also applies to the tensile strength, with one restriction. By comparison, the composites produced with POE as the modifier display only a slight reduction in strength.

Comparing the composite properties with those of injection molded PP, it is striking that, with the addition of 25% TPE4 (1400 MPa) or TPE7 (1550 MPa), the elastic modulus of the composites falls below that of the injection molded material (1800 MPa). The tensile strength of the composites, by contrast, is only reduced to that of the injection molded component (36 MPa) when some 50% TPE4 is added. Using the POE modifier, the addition of 75% gives an elastic modulus of 1949 MPa and a tensile strength of 83 MPa. Both characteristic values are thus above the comparative values determined on injection molded material.

Figure 13 shows the characteristic values determined in the 5 J impact tests over the modifier content for the different modifiers. The impact stiffness and the maximum impact force determined here undergo a decline with an increasing modifier content. Contrary to the case for the tensile strength, the impact force falls further when use is made of the POE modifier. The deformation of the material and also the damping, by contrast, increase with the modifier content.

3.2.3 Regression models as a function of Shore A (modifier) and modifier content

To permit a holistic view of the correlations already presented and obtain a better overview of the mechanical properties and their values as a function of the materials used, regression models were compiled for the respective mechanical parameters measured. The regression analysis is performed using appropriate software for multiple regression analyses (partial least squares regression). The regression equation was compiled as a function of the Shore A hardness and the content of the modifier used in each case, so as to be able to estimate the mechanical properties in advance in future. The regression was performed for the target values of tensile strength, elastic modulus, maximum deformation, material damping, maximum force and impact stiffness.

Figure 14 shows the result of the regression equation for the target value of tensile strength. The measured reference values with pure PP film without modifier were not taken into account in the regression analysis for all the target values.

As expected, the tensile strength increases with the Shore A hardness and falls with increasing modifier content. It is also clear to see that, when using a modifier with a higher Shore A hardness, the modifier content has a considerably lower effect, or no effect at all, on the tensile strength compared to a modifier with a low Shore A hardness. Figure 15 shows the result of the regression calculation for the target value of elastic modulus. The quality of the model can be rated as good.

tensile strength[MPa]

$$= 31.9628 - 0.2876 * c_m + 20.5603 * ((1 - 5.3318 * 10^{-5} * S_A{}^2) + 5.7279 * 10^{-8} * (S_A{}^4)) + (c_m - 49.5370) * ((((1 - 5.3318 * 10^{-5} * S_A{}^2) + 5.7279 * 10^{-8} * (S_A{}^4)) - 2.245) * 0.3463)$$

c_m = content of modifier [%] S_A = Shore A (modifier) [-]

$r^2 = 0.787$	$r_{adj}{}^2 = 0.774$	$RMSE = 10.39$

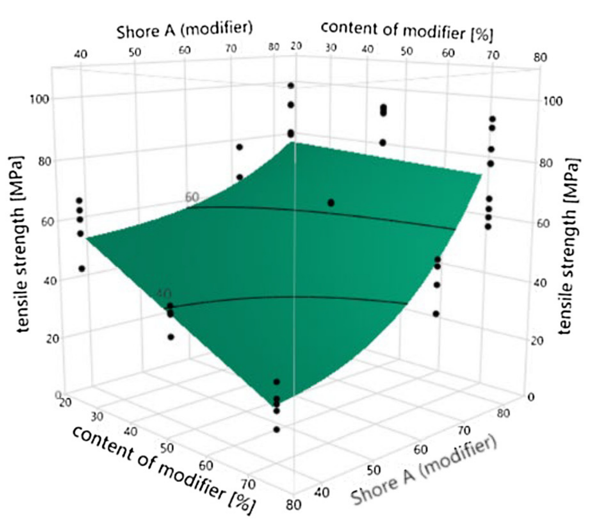
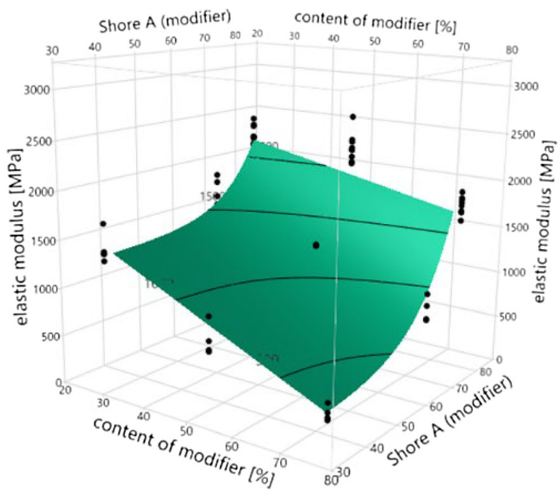


Figure 14: Modulation of the tensile strength as a function of “content” and “Shore A” of the modifier.



elastic modulus [MPa]

$$= 471.4848 - 13.8911 * c_m + 726.5716 * ((1 - 5.3318 * 10^{-5} * S_A{}^2) + 5.7279 * 10^{-8} * (S_A{}^4)) + (c_m - 49.5370) * ((((1 - 5.3318 * 10^{-5} * S_A{}^2) + 5.7279 * 10^{-8} * (S_A{}^4)) - 2.245) * 6.4071) + (((1 - 5.3318 * 10^{-5} * S_A{}^2) + 5.7279 * 10^{-8} * (S_A{}^4)) - 2.245) * ((((1 - 5.3318 * 10^{-5} * S_A{}^2) + 5.7279 * 10^{-8} * (S_A{}^4)) - 2.245) * 196.5366)$$

c_m = content of modifier [%] S_A = Shore A (modifier) [-]

$r^2 = 0.949$ $r_{adj}{}^2 = 0.945$ $RMSE = 152.65$

Figure 15: Modulation of the elastic modulus as a function of "content" and "Shore A" of the modifier.

It is seen that the elastic modulus increases with increasing Shore A hardness and decreasing modifier content. The elastic modulus attains its maximum value of approximately 2500 MPa with a Shore A hardness of 80 and a content of 25%. The minimum value for the elastic modulus is achieved with a Shore A hardness of 40 and a modifier content of 75%. With a coefficient of determination (R^2) of 0.949, the observations are depicted well by the regression equation. The specified root mean square error (RMSE) also indicates the mean deviation of the observations from the regression model. Expressed in terms of the mean value of the regression model, a deviation of 9.84% is seen here. It is not recommended to perform an approximation beyond the limits of the factor settings used, since this can lead to implausible or inaccurate results. Figure 16 shows the result of the regression model for the target value of maximum deformation.

The maximum deformation increases with modifier content and falls with increasing Shore A hardness. With a coefficient of determination of 0.941, the regression model is similarly suitable and depicts the observed characteristic values well. The RMSE is 0.184, corresponding to a fluctuation of 2.24% around the mean value.

Figures 17–19 show the models for material damping, maximum force and impact stiffness. While the material damping increases with a high modifier content and a low Shore A hardness, the situation for the maximum force and impact stiffness is precisely the opposite. The regression equations are fundamentally valid and depict the observed parameters well. This is borne out by the quality of the model and the RMSE in each case. It can consequently be assumed that the regression models compiled here constitute a suitable approach for estimating the modification of SRCs on the basis of the material used and the quantity of

max. impact deformation [mm]

$$= 8.8598 + 0.0205 * c_m - 0.6856 * ((1 - 5.3318 * 10^{-5} * S_A{}^2) + 5.7279 * 10^{-8} * (S_A{}^4)) + (c_m - 50.5556) * ((((1 - 5.3318 * 10^{-5} * S_A{}^2) + 5.7279 * 10^{-8} * (S_A{}^4)) - 2.2705) * -0.0105 + (((1 - 5.3318 * 10^{-5} * S_A{}^2) + 5.7279 * 10^{-8} * (S_A{}^4)) - 2.2705) * ((((1 - (5.3318 * 10^{-5} * S_A{}^2) + 5.7279 * 10^{-8} * (S_A{}^4)) - 2.2705) * -0.2120)$$

c_m = content of modifier [%] S_A = Shore A (modifier) [-]

$r^2 = 0.941$ $r_{adj}{}^2 = 0.935$ $RMSE = 0.184$

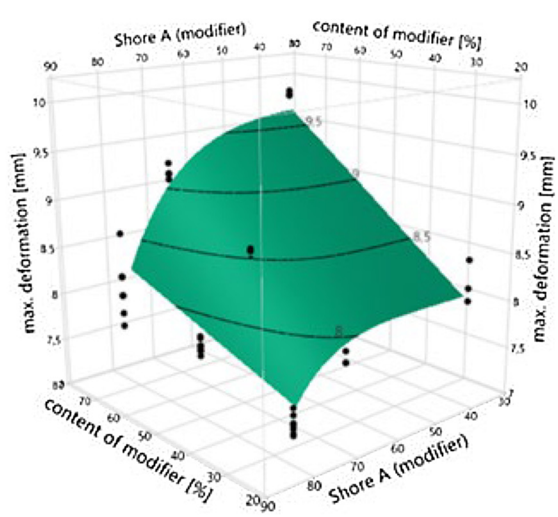
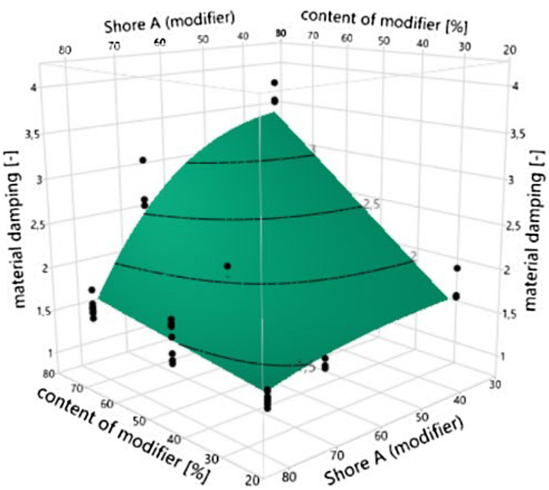


Figure 16: Modulation of the max. impact deformation as a function of "content" and "Shore A" of the modifier.



material damping [−]

$$= 2.3212 + 0.0166\ c_m - 0.56 * ((1 - 5.3318 * 10^{-5} * S_A{}^2) + 5.7279 * 10^{-8} * (S_A{}^4)) + (c_m - 50.5556) * ((((1 - 5.3318 * 10^{-5} * S_A{}^2) + 5.7279 * 10^{-8} * (S_A{}^4)) - 2.2705) * -0.0165)$$

c_m = content of modifier [%] S_A = Shore A (modifier) [-]

$r^2 = 0.933$ $r_{adj}{}^2 = 0.929$ $RMSE = 0.181$

Figure 17: Modulation of the material damping as a function of “content” and “Shore A” of the modifier.

max. impact force [N]

$$= 1474.9759 + -3.8588 * c_m + 150.3019 * ((1 - 5.3318 * 10^{-5} * S_A{}^2) + 5.7279 * 10^{-8} * (S_A{}^4)) + (c_m - 50.5556) * ((((1 - 5.3318 * 10^{-5} * S_A{}^2) + 5.7279 * 10^{-8} * (S_A{}^4)) - 2.2705) * 1.8855) + (((1 - 5.3318 * 10^{-5} * S_A{}^2) + 5.7279 * 10^{-8} * (S_A{}^4)) - 2.2705) * ((((1 - 5.3318 * 10^{-5} * S_A{}^2) + 5.7279 * 10^{-8} * (S_A{}^4)) - 2.2705) * 65.4783)$$

c_m = content of modifier [%] S_A = Shore A (modifier) [-]

$r^2 = 0.957$ $r_{adj}{}^2 = 0.952$ $RMSE = 31.177$

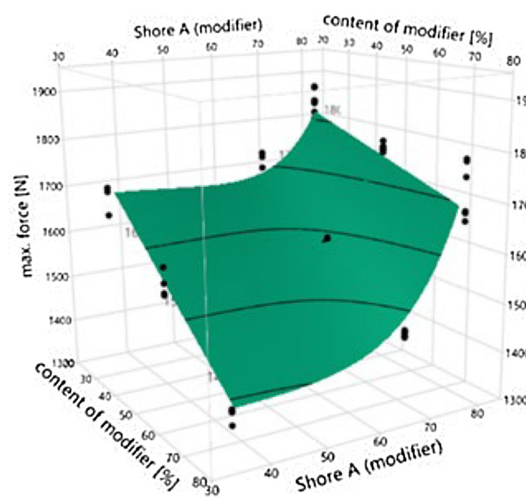
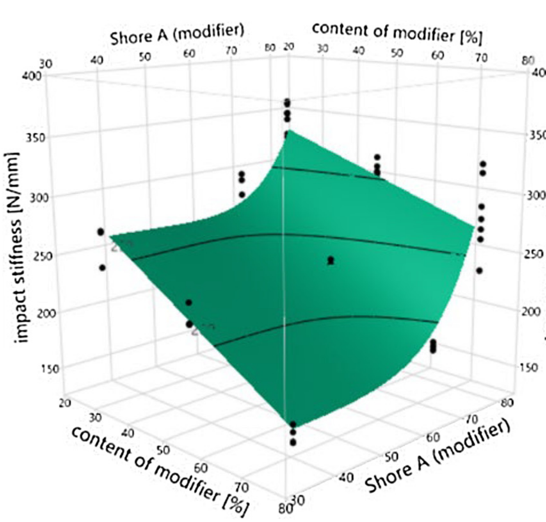


Figure 18: Modulation of the max. impact force as a function of “content” and “Shore A” of the modifier.



impact stiffness [N]

$$= 184.7165 - 1.6151 * c_m + 63.3425 * ((1 - 5.3318 * 10^{-5} * S_A{}^2) + 5.7279 * 10^{-8} * (S_A{}^4)) + (c_m - 50.5556) * ((((1 - 5.3318 * 10^{-5} * S_A{}^2) + 5.7279 * 10^{-8} * (S_A{}^4)) - 2.2705) * 0.5067) + (((1 - 5.3318 * 10^{-5} * S_A{}^2) + 5.7279 * 10^{-8} * (S_A{}^4)) - 2.2705) * ((((1 - 5.3318 * 10^{-5} * S_A{}^2) + 5.7279 * 10^{-8} * (S_A{}^4)) - 2.2705) * 27.2301)$$

c_m = content of modifier [%] S_A = Shore A (modifier) [-]

$r^2 = 0.929$ $r_{adj}{}^2 = 0.921$ $RMSE = 16.939$

Figure 19: Modulation of the impact stiffness as a function of “content” and “Shore A” of the modifier.

material in future. Additional validation experiments ought nevertheless to be carried out and the regression models presented here validated with the aid of the new data.

4 Conclusions

In this study, SRCs were produced by the film stacking process, either with the films used containing an elastomer modifier or with individual films made of TPE. The compounds and films used were produced beforehand by compounding and/or film extrusion under conditions that were kept as constant as possible. The proportion of modifier in the SRCs was specifically adjusted and the change in the mechanical properties then analyzed via impact and tensile tests.

It was assumed that the mechanical properties can be selectively influenced and adjusted through use of the modifiers. Modifications of up to 74% were achieved for tensile strength and up to 88% for the elastic modulus. In addition, the impact stiffness was modified by 58% and the damping behavior by 218%. The modifications achieved were influenced in different ways through the proportion and type of modifier. On the basis of the characteristic values determined, it is possible to describe the mechanical properties as a function of the modifier content, as shown in Figures 12–19. The properties can consequently be specifically influenced via the content and type of modifier, with the mechanical parameters being mutually dependent and this needing to be taken into consideration accordingly.

It was also shown that the modifier can be introduced both by interleaving layers of different combinations of films in homogeneous materials and by using films produced from a compound. The measurements performed additionally showed that, for the same proportion of modifier, the method of introduction under the conditions prevailing here did not have a significant influence on the mechanical properties. On this point, it is important to note that these results are most likely related to a large extent to the low thickness of the specimens used. With an increasing specimen thickness, the distribution of the modifier within the specimen will probably become more significant.

Finally, regression models were compiled for the target values of tensile strength, elastic modulus, maximum impact force, maximum impact deformation, material damping and impact stiffness, as a function of modifier content and Shore A hardness of the modifier. The regression equations drawn up deliver a good approximation of how the composites behave over the tested parameter range and can facilitate the adaptation of the material properties in future. Regression models for maximum strength in tensile test are less reliable compared to the other regression models presented. It can be assumed that the tensile strength is mainly dependent on the tapes. These are influenced by temperature, which was kept constant over the tests. Therefore, the difference can only be due to the matrix used, which may cause for this load case a smaller coefficient of determination. In order to make a clear statement on this, further investigations should be carried out in the future, but it can fundamentally be assumed that other systems with similar material configurations and similar objectives will display the same basic correlations as those presented here.

The modification of SRCs presented here illustrates the many different possibilities of these materials. Modification via films can essentially be performed using simple tool technology, thus resulting in a high level of process reliability. Comparable concepts for implementing locally different mechanical properties have so far always required a high level of process control (Biermann et al. 2015; Paßmann 2009; Ries 2015). The possibility of generating locally differing mechanical properties can thus be exploited more easily in future in order to guarantee specific occupant protection, for example. It would thus be conceivable to modify interior trim parts in such a way that they displayed particularly high damping in the areas that come into contact with the occupants in the event of a crash, without losing their fundamental stiffness and their function as a carrier panel for other components, for example. Since they can be manufactured in just a single process, it will be possible to save on production steps and hence also on tool costs and material costs in future.

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