TENSILE AND FLEXURAL CREEP BEHAVIOUR OF SELF-REINFORCED POLYPROPYLENE COMPOSITES PREPARED BY COMPRESSION AND INJECTION MOLDING

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Abstract
In this work the creep behavior of self-reinforced polymer composites (SRPC) were investigated. As matrix material olefin based thermoplastic elastomeric (o-TPE), as reinforcement highly oriented polypropylenes were used. The tested self-reinforced composites were prepared by filament winding with subsequent compression molding and injection molding (from new pre-impregnated pre-product material) technique. On the SRPC creep tests were carried out. The tensile and flexural creep tests were performed by universal tensile machine and dynamic mechanical analyzer (DMA) test. The results revealed that the creep behavior of the self-reinforced materials depended on the matrix material and the reinforcement structure. The highest strain was achieved using woven fabric while the unidirectional structure showed the lowest strain.

1 Introduction
The long-term behavior of structural materials is one of the most important properties. Under steady loading the material can slowly deform, which phenomenon is called creep. When a plastic material is loaded with a constant load, it deforms continuously with time. This time-dependent behavior of materials (viscoelasticity) is an important characteristic of polymers. To know this long-term behavior of polymers is essential to estimate their life-time under load [1]. Although SRPC as an anisotropic, multi-phase system, like composites, is very complex, the analysis of creep properties is important for the use of composites in long-term applications. This may be more significant in case of self-reinforced polymer composites than in classic composite materials reinforced by glass or basalt fiber, since not only the matrix but the reinforcement is also made of polymer. Self-reinforced polymer composites have some advantages beyond the good mechanical properties; such as easy and fully recyclability and low density (the same as that of the matrix material) [2]. There are some studies on the creep behavior of (mainly polypropylene based) self-reinforced composites reinforced by continuous fibers or tapes (aligned in unidirectional or cross-ply or woven structure). These studies have pointed out that the creep behavior of self-reinforced PP composites depends strongly on stress, temperature, void content (consolidation), and fiber content [3-4]. Furthermore it was presented, that creep resistance decreases if temperature or stress rises and increases with increasing consolidation quality (lower void content).
The aim of this study is to compare the creep behavior of self-reinforced polypropylene composites (SRPPC) prepared by compression (reinforced by continuous fibers in unidirectional, cross-ply, and woven structure) and injection molding (reinforced by special pre-impregnated pre-product). The short term creep behavior is tested at both tensile (using universal tensile testing machine) and flexural load (in dynamic mechanical analyzer, DMA).

2 Materials and their processing

As matrix material two kinds of materials were used (1: random polypropylene copolymer (rPP) (Tipplen R959A, TVK, Tiszaújváros, Hungary); 2: olefin based thermoplastic elastomeric (Dow Versify 4200 (TPE)). As reinforcement materials woven polypropylene fabric and highly oriented homo-polypropylene (hPP) multifilament (Stradom Sa., Czestochowa, Poland) were applied. From the matrix granulates foil with thickness of 50 µm was prepared by extrusion film blowing (R959A) and flat film extrusion (TPE) technique.

Three kinds of reinforcement structure were used. The first was the woven fabric (WF), the second was the unidirectional reinforcement (UD), and the third was the cross-ply (CP) (0-90°) structure. The matrix film and the reinforcing hPP multifilament were laminated under pre-tension onto an aluminum core according to the film-stacking method by filament winding process, which aligned the fiber directional (Figure 1). The woven fabric was laid layer by layer between the matrix foils. The package was consolidated by compression molding (Schwabentan Polystat 300S) according to the following steps. The package was inserted into a preheated hot press at 180°C (rPP based SRPPC) and 140°C (TPE based SRPPC) and hold for 4 min without pressure and then was compression molded for 4 min (rPP) and 8 min (TPE) with 5.3 MPa pressure. The composite was cooled (cooling rate: 30°C/min) under pressure. The thicknesses of the resulting composite plates were between 0.9...2.3 mm. The nominal fiber content was set to 75 wt%.

![Figure 1. Preparation of unidirectional and cross-ply hPP based self-reinforced composite with filament winding](image)

From the consolidated TPE based self-reinforced (UD) composite plate, pre-impregnated granulates (5x5 mm) were prepared by a granulator machine. Thereafter this pre-product was used for injection molding.

**Injection molding**

From the pre-product granulate (TPE) 80x80 mm flat specimens with a thickness of 2 mm (Figure 2) (MD) were injection molded by an Arburg Allrounder 370S 700-290 machine. Table 1 shows the injection molding parameters.
<table>
<thead>
<tr>
<th>Injection molding parameters</th>
<th>Value</th>
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<tbody>
<tr>
<td>Injection volume [cm³]</td>
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</tr>
<tr>
<td>Injection rate [cm³/s]</td>
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</tr>
<tr>
<td>Injection pressure [bar]</td>
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<tr>
<td>Switch over point [cm³]</td>
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</tr>
<tr>
<td>Holding pressure [bar]</td>
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<tr>
<td>Holding time [s]</td>
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<tr>
<td>Residual cooling time [s]</td>
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<tr>
<td>Screw rotational speed [m/min]</td>
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<tr>
<td>Back pressure [bar]</td>
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<tr>
<td>Back pressure time [s]</td>
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<td>5</td>
</tr>
<tr>
<td>Decompression rate [cm³/s]</td>
<td>5</td>
</tr>
</tbody>
</table>

Table 1. Injection molding parameters for sample preparation

The temperature zones were set to 140°C, 135°C, 130°C, 125°C, 120°C from the feeder to the nozzle and the mould was tempered to 20°C.

3 Testing methods

3.1 Tensile creep test
Short-time tensile creep tests were performed on a Zwick Z005 universal testing machine at room temperature (22±1°C) using dumbbell shaped specimens (EN ISO 8256 Shape 3) which were cut out of the consolidated plate by a water jet cutter. Firstly constant load rate tests were carried out on the specimens (3-3 samples were tested from each reinforcement structure). From these tensile tests average yield stress was calculated. This value means the 100% load level on the tensile creep tests. The creep tests were done on 10, 20, 30…80, 90% of the average yield stress. The test time was 30 min and the preloading speed was 50 N/s.

3.2 Flexural creep tests
The short-time flexural creep tests were performed on DMA Q800 machine with using three-point bending mode at different temperatures, ranging from -20°C to 80°C. In this temperature range, isothermal creep tests were carried out on the specimens. Prior to the creep measurement, each specimen was equilibrated for 5 min at the test temperature and then the flexural creep behavior was tested for 30 min, under a constant load of 5 MPa. Specimens with the dimensions of 60x10x2 mm (length x width x thickness) were used for creep tests cut out of the compression and injection molded composites. Span length was 50 mm.

4 Results and discussion

**Tensile creep tests**
Figure 3 shows the different strain-time curves of the two materials with different reinforcement structures.
One can note that increasing load level increased strain and in case of the Versify matrix creep fractures occurred at the highest loads. The highest strain was achieved in cross-ply and woven structure.

Figure 4 shows the strain-time curves of the different reinforcement structure strain at 1000 s.
Figure 4. Strain at (1000 s)-time curves of the self-reinforced polymer composites with different reinforcement structures (a) TPE-based and (b) rPP-based composites

One can conclude that the curves of the R959A (rPP) are similar and close to each other (Figure 4 (b)) which can be attributed to the deformation behavior of the rPP matrix. The matrix material limited the moving of the reinforcement. It seems the TPE curves are very different from rPP curves. The thermoplastic elastomeric material has higher deformation compared to the rPP. The reinforcements may move which cause the differences between the curves. The highest strain is shown for woven structure.

The constant force rate tensile curves of the different reinforcement structures are shown in the Figure 5.

Figure 5. Strain-time curves of the self-reinforced polymer composites with different reinforcement structures: (a) TPE-based and (b) rPP-based composites

These results show that using different reinforcement structures makes the strain variation versus time different. Test time on the horizontal axis was proportional to the loading force because of the constant 50 N/s loading speed. It can be observed that the tensile curves for the two kinds of matrix material are different as well, the deformability of the Versify is remarkably larger. The curves families of TPE composites reinforced with different fibrous structures are open separated from each other. The woven fabric reinforced composites showed the highest and the unidirectional composites showed the lowest strain.
Flexural creep tests

Figure 6 shows the creep compliance curves of the TPE based composites. It can be concluded that increasing the temperature the creep compliance increase.

![Creep compliance-time curves of the TPE composites](image)

**Figure 6.** Creep compliance-time curves of the TPE composites

Figure 7 shows the creep-compliance-time curves of the composites at different temperatures.

![Creep compliance-time curves of the composites at 30°C and 40°C](image)

**Figure 7.** Creep compliance-time curves of the composites at 30°C (a) and 40°C (b)

It can be stated that the creep compliance of the rPP based self-reinforced composites is lower than the TPE based composite. The highest creep compliance was showed by the injection molded TPE based composites which is probably the effect of the filling type (fiber distribution in the mold) and the chopped fibers.

5 Conclusions

The goal of this paper was to study the creep behavior of the thermoplastic elastomeric and random polypropylene based self-reinforced polymer composites with different reinforcement structure. Based on the tensile creep results it can be concluded, that the increasing load level increased the strain and the highest strain showed in cross-ply and woven structure. The tensile curves of the composites which have different reinforcement structure were different. The flexural creep results revealed that the rPP based self-reinforced composite has lower creep compliance compare to the TPE based systems. The injection moulded specimens showed the highest creep compliance which is due to the filling mode and the chopped fibres.
6 Acknowledgements

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7 References