# INFLUENCE OF DIFFERENT IMPACT MODIFIERS ON THE MECHANICAL PROPERTIES OF CELLULOSE FIBRE REINFORCED POLYPROPYLENE COMPOSITES

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**Keywords:** cellulose fibre, polypropylene, impact modification, mechanical properties, injection moulding

## Abstract

The aim of the present study was to improve the notched impact strength of a cellulose fibre reinforced PP composite, while at the same time limiting the loss of stiffness and strength. To this end, a variety of impact modifiers was tested, including several polyolefinic elastomers (POE; ethylene-octene and propylene-ethylene), styrene-butadiene based elastomers (SBS, SEBS), and ethylene vinyl acetate (EVA). The mechanical characterisation included testing for Charpy notched (NIS) and unnotched impact strength (IS), tensile properties and heat deflection temperature (HDT).

As expected, an increased amount of impact modifier improved the NIS considerably and reduced the tensile properties and HDT. The changes to the IS were negligible in most cases. Generally the ethylene-octene elastomers, as well as SBS and SEBS, appeared to be particularly suitable to achieve a good overall mechanical performance. Additionally, for elastomers of a given chemical composition, a higher viscosity of the elastomer increased its potential to improve the NIS of the composite.

While none of the tested formulations reached the impact performance of two commercial talc reinforced PP composites that served as a reference, their weight-related advantage over the PP-talc composites is worth mentioning. Moreover, several of the composites in this study surpassed the impact performance of commercial PP-cellulose composites by a considerable margin.

## 1. Introduction

Natural-fibre reinforced composites (NFC) based on polypropylene (PP) can be of special interest for the automotive industry as a substitute for conventional composites (e.g. talc or glass fibres), as they offer comparable stiffness and strength while at the same time exhibiting a higher potential for lightweight construction due to their lower density. In addition, they provide the ecological benefit of partially consisting of renewable raw materials. Kraft pulp fibres offer a good combination of perennial availability, good reinforcing potential, and price, and thus appear particularly suitable for the production of composites for the automotive industry.

Because of the low intrinsic impact toughness of PP, additives are often introduced to improve the impact strength. The most common approach to enhance the mechanical performance of PP-based NFCs is the addition of a coupling agent such as maleic acid (MA) grafted polymer [1-5] or silane [6], which serves to increase the fibre-matrix interactions; however, this has been shown to actually reduce the impact strength of the materials used in this study, since a stronger fibre-matrix interaction results in fibre breakage upon impact, which takes up less energy than the fibre pull-out which would happen otherwise. A different approach, especially in the case of PP-based composites, is to add an elastomeric phase to improve the impact performance of the matrix. Typical impact modifiers for PP and PP composites are ethylene-propylene based rubbers (EPR, EPDM) and styrenic elastomers (SBS, SEBS), which are often used in combination with coupling agents [7-10]. The use of EVA as impact modifier has also been reported [11]. In recent years, impact toughening of PP by poleolefinic elastomers has also gained importance [12].

For the present study, the selection of the materials was made according to recommendations from the manufacturers. The objective in all cases was to achieve the highest possible notched impact strength while at the same time retaining a tensile E-modulus of 1.8 GPa or higher.

## 2. Materials and Methods

### 2.1. Materials

The basic formulation for all composites that were investigated in this study consisted of an injection moulding grade, metallocene-catalysed polypropylene (PP) homopolymer with a melt flow rate (MFR) of 140 g/10 min (230 °C, 2.16 kg), and bleached Kraft cellulose fibres with an average fibre length of 800 µm (prior to processing) and a thickness of 15 µm. The fibre content in all composites was 20 % by weight. The reference formulation consisting of 80 wt% PP and 20 wt% fibres will subsequently be referred to as “PP20F”.

The impact modifiers that were tested included several grades of polyolefinic elastomers (POE; ethylene-octene and ethylene-propylene), styrene-butadiene based elastomers (SBS, SEBS), and ethylene vinyl acetate (EVA). A complete list of the impact modifiers is shown in Table 1.

**Table 1** List of impact modifiers (material data as shown on the technical data sheets)

| Code | Density [g/cm$^3$] | Hardness Shore A | MFR [g/10 min] | Comment |
|---|---|---|---|---|
| *Polyolefinic elastomers – Ethylene-propylene* | | | | |
| EP1 | 0.862 | A 67 | 3 (230 °C, 2.16 kg) | 16% Ethylene content |
| EP2 | 0.863 | A 64 | 20 (230 °C, 2.16 kg) | 15% Ethylene content |
| EP3 | 0.865 | A 71 | 48 (230 °C, 2.16 kg) | 13% Ethylene content |
| *Polyolefinic elastomers – Ethylene-octene* | | | | |
| EO1 | 0.870 | A 73 | 1 (190 °C, 2.16 kg) | |
| EO2 | 0.885 | A 84 | 30 (190 °C, 2.16 kg) | |
| EO3 | 0.870 | A 70 | 5 (190 °C, 2.16 kg) | |
| *Styrene-butadiene based elastomers* | | | | |
| SBS | 0.94 | A 71-81 | < 1 (200 °C, 5 kg) | 31% Styrene content |
| SEBS | 0.90 | A 47 | 22 (230 °C, 5 kg) | 13% Styrene content |
| *Ethylene vinyl acetate* | | | | |
| EVA | 0.952 | A 75 | 2.5 (230 °C, 2.16 kg) | 28% VA content |

The amount of impact modifier in the composites were typically 10 and 20 % by weight, and 30 % in a few selected formulations. Regarding the nomenclature, the formulations used for this study will subsequently be referred to according to the scheme *"ModifierCode_wt%"*; e.g. the designation "EP1_30" describes the formulation consisting of 30 wt% EP1, 50 wt% PP and 20 wt% fibres.

Two commercially available talc-reinforced polypropylene copolymers that are being used as high-impact grades for the automotive industry were tested as reference materials: Hostacom TRC 333N (20% talc filled) and Hostacom EKC 330N (16% talc filled), both from LyondellBasell. As a reference specifically for PP-cellulose composites, the mechanical properties of the commercially available high impact grade Formi EXP 20 (20 wt% cellulose filled PP) from UPM Biocomposites, according to the technical data sheet [13], were taken into account.

### 2.2. Sample preparation

The mixtures were prepared on a Brabender DSE20 twin screw extruder, with a screw diameter of 20 mm and a L/D ratio of 40. The screw speed was set to 600 rpm and the throughput to 5 kg/h. The polymeric components were added at the hopper and the fibres were introduced via a side feeder at position 11d. An ECON EUP50 underwater pelletising device was used to obtain pellets from the compounds. For the mechanical characterisation, standard shoulder bars according to ISO 527-2 were prepared on a Battenfeld HM1300/350 injection moulding machine. Prior to testing, the specimens were conditioned at 23 °C and 50% r.h. for at least 120 hours.

### 2.3. Mechanical testing

Tensile tests according to ISO 527 were carried out on a Messphysik BETA20-10 universal testing machine. The tensile modulus $E$ was determined in the range between 0.05 and 0.25% elongation at a rate of 1 mm/min, afterwards the elongation rate was gradually increased to 50 mm/min to determine the tensile strength $\sigma_M$. The Charpy impact strength (IS) and notched impact strength (NIS) according to DIN EN ISO 179-1eU (unnotched) or -1eA (notched) was tested on an Instron CEAST 9050 impact pendulum. Depending on the impact toughness of the materials, a pendulum with an energy of 0.5, 2 or 7.5 J was used. The heat deflection temperature *HDT-B* according to DIN EN ISO 75 was performed on an Instron CEAST HDT Vicat Series 3 Station Tester, using a load of 0.45 N/mm². This test was only carried out for selected samples. The melt flow rate (*MFR*) according to ISO 1133 was tested on a Zwick 4105 Extrusion Plastometer, at a temperature of 230 °C and using a load of 2.16 kg. As an additional criterion for the flowability of the compounds, the maximum injection moulding pressure ($P_{max}$) was measured, as the MFR values are, due to the much lower shear rates, not necessarily representative for the flow behaviour of the compounds under process conditions.

## 3. Results and discussion

### 3.1. Influence of the viscosity of the impact modifier

In the first test series, three grades of ethylene-propylene POE from the same manufacturer, which mainly differed in their viscosity (the differences in ethylene content and Shore hardness were considered negligible in comparison) were added to the PP-fibre composite in amounts of 10, 20 and 30 wt% respectively.

As expected, increasing the concentration of the modifiers resulted in a strong increase of the NIS and a reduction of the tensile modulus and strength. Additionally the flowability and the HDT were reduced as well. The unnotched impact strength, however, remained largely unaffected by the modifier.

With an increasing viscosity of the elastomer, the effect of the impact modification was strengthened. The highest NIS value was thus obtained for EP1_30, which was 184% higher than the NIS of the reference material. In comparison, with EP3_30 the NIS was only increased by 115%. Moreover, a higher viscosity of the modifier slightly reduced the detrimental effect on the HDT. The downside of a high modifier viscosity was obviously an increase in the total viscosity of the compound; however, despite the strong decrease in MFR / increase in $P_{max}$, the flowability even of EP1_30 was still considered acceptable. Regarding the tensile properties and the unnotched impact strength, the viscosity of the modifier did not appear to have a significant influence. All results are summarised in Table 2. A further illustration of the E-modulus and the NIS is given in Figure 1, as the focus regarding the mechanical performance lies mostly on these two properties.

Despite the very high NIS increase that was achieved with 30% elastomer content, the resulting stiffness was far below the defined limit of 1.8 GPa; therefore, in the subsequent experiments only formulations with 10% and 20% modifier content were produced (with very few exceptions).

**Table 2** Mechanical properties of the compounds containing ethylene-propylene elastomer

| Code | $\rho$ (g/cm³) | *MFR* (g/10 min) | $P_{max}$ (bar) | *E* (GPa) | $\sigma_M$ (MPa) | *IS* (kJ/m²) | *NIS* (kJ/m²) | *HDT-B* (°C) |
|---|---|---|---|---|---|---|---|---|
| PP20F | 0.981 | 9.2 | 450 | 2.76 | 40.1 | 41.0 | 4.5 | 134.6 |
| EP1_10 | 0.981 | 5.1 | 535 | 2.19 | 36.6 | 39.3 | 6.2 | 136.1 |
| EP1_20 | 0.976 | 2.6 | 615 | 1.83 | 31.0 | 39.1 | 9.2 | 127.7 |
| EP1_30 | 0.972 | 2.2 | 710 | 1.33 | 24.3 | 36.9 | 12.8 | 114.3 |
| EP2_10 | 0.978 | 7.1 | 500 | 2.05 | 35.3 | 39.0 | 5.6 | 132.6 |
| EP2_20 | 0.978 | 4.6 | 570 | 1.72 | 30.4 | 41.0 | 8.1 | 122.9 |
| EP2_30 | 0.974 | 3.6 | 625 | 1.33 | 24.2 | 36.9 | 11.1 | 108.0 |
| EP3_10 | 0.978 | 7.3 | 485 | 2.07 | 35.6 | 39.4 | 5.3 | 129.4 |
| EP3_20 | 0.978 | 6.3 | 535 | 1.74 | 30.8 | 40.1 | 7.5 | 119.1 |
| EP3_30 | 0.973 | 6.0 | 565 | 1.32 | 24.7 | 38.6 | 9.7 | 104.0 |

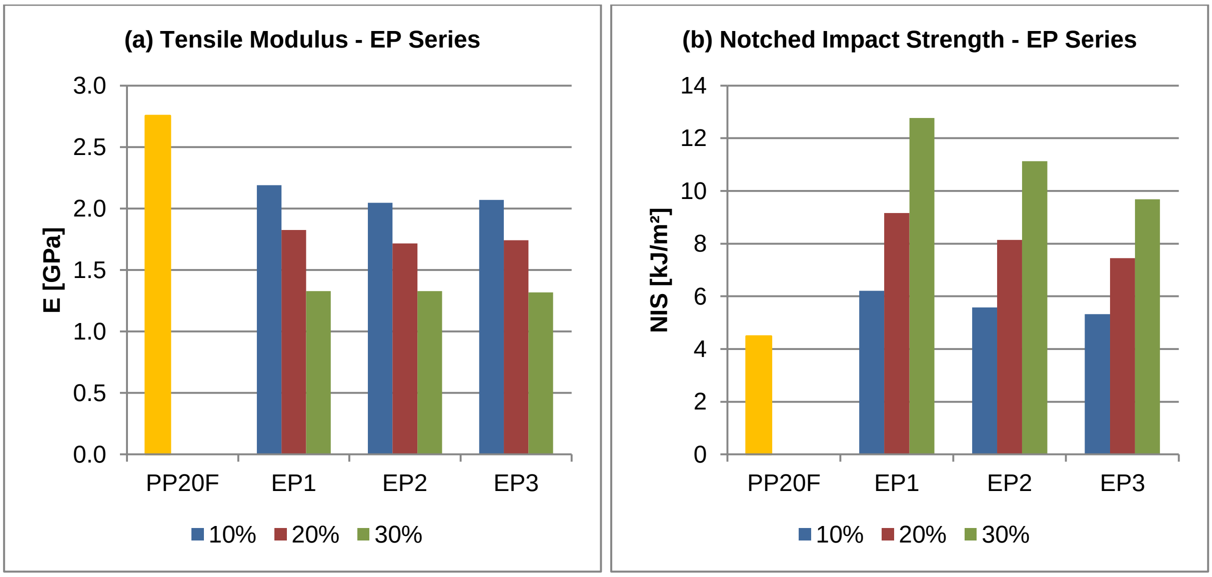

**Figure 1.** (a) Tensile modulus and (b) NIS of the compounds containing the EP elastomers

### 3.2. Comparison of different types of impact modifiers

For the second series, three different ethylene-octene elastomers, one SBS grade, one SEBS grade and one EVA grade were used in amounts of 10 wt% and 20 wt% respectively. In case of the SBS, a formulation with 30 wt% modifier content was produced as well.

In comparison with the EP elastomers, most of the other modifiers led to a further improvement of the NIS, the notable exceptions being the EO2 and the EVA. The IS was slightly improved with all EO types, but not with any other modifier; however, generally the IS was only marginally affected. The reduction of tensile strength was in a similar range for all modifiers, whereas for the stiffness slightly larger differences were observed, with the best values being reached with EO1, EO3 and SBS. Generally all observed (positive and negative) effects were stronger with a higher modifier concentration.

Among the POEs, the best overall performance was achieved with 20 wt% of EO3, with an increase of the NIS by 132 % and a comparatively moderate loss of stiffness (- 26 %). Similar tensile properties were achieved with EO1, but at a slightly poorer impact performance and flowability. With EO2 the best IS of all formulations was achieved, but the NIS and stiffness were noticeably worse compared to the other EO grades.

Among all compounds containing 20 wt% of impact modifier, the highest NIS was achieved with SEBS (+ 153 %), which however came at the cost of a comparatively low stiffness, even though it was still above the acceptable limit of 1.8 GPa. The formulations containing SBS showed promising mechanical properties, with the impact performance being similar to that of EO1, but the E-modulus being slightly higher. Therefore an additional formulation with 30 wt% SBS was produced; with this material, by far the best NIS of all tested samples was achieved (+ 213 % compared to PP20F). The stiffness was significantly higher compared to the compounds with 30 wt% EP elastomer, despite being slightly below 1.8 GPa. The drawbacks of this formulation were a comparatively poor tensile strength and flowability, and a slightly higher density than most other materials.
The samples containing EVA showed comparable tensile properties and IS to the other formulations, but a significantly worse NIS.

All measured mechanical data are summarised in Table 3, and the tensile modulus as well as the NIS are illustrated in Figure 2.

**Table 3** Mechanical properties of the compounds containing various impact modifiers

| Code | $\rho$ (g/cm$^3$) | $MFR$ (g/10 min) | $P_{max}$ (bar) | $E$ (GPa) | $\sigma_M$ (MPa) | $IS$ (kJ/m$^2$) | $NIS$ (kJ/m$^2$) |
|---|---|---|---|---|---|---|---|
| PP20F | 0.981 | 9.2 | 450 | 2.76 | 40.1 | 41.0 | 4.5 |
| EO1_10 | 0.978 | 6.5 | 540 | 2.36 | 34.2 | 42.4 | 7.1 |
| EO1_20 | 0.978 | 3.5 | 620 | 2.02 | 29.9 | 40.9 | 9.6 |
| EO2_10 | 0.984 | 6.4 | 520 | 2.30 | 36.7 | 44.3 | 6.7 |
| EO2_20 | 0.980 | 5.2 | 560 | 1.83 | 31.9 | 44.5 | 8.4 |
| EO3_10 | 0.981 | 6.7 | 525 | 2.36 | 34.2 | 42.2 | 7.6 |
| EO3_20 | 0.977 | 4.7 | 580 | 2.05 | 29.4 | 43.0 | 10.4 |
| SBS_10 | 0.982 | 4.3 | 520 | 2.29 | 32.7 | 41.9 | 6.8 |
| SBS_20 | 0.993 | 2.4 | 600 | 2.12 | 28.5 | 40.9 | 9.6 |
| SBS_30 | 0.997 | 1.5 | 525 | 1.72 | 22.3 | 37.8 | 14.2 |
| SEBS_10 | 0.982 | 4.5 | 540 | 2.27 | 34.8 | 41.3 | 7.5 |
| SEBS_20 | 0.980 | 3.1 | 630 | 1.86 | 28.6 | 40.4 | 11.4 |
| EVA_10 | 0.990 | 5.8 | 520 | 2.30 | 35.1 | 39.3 | 6.2 |
| EVA_20 | 0.996 | 5.3 | 570 | 1.93 | 31.3 | 38.2 | 7.3 |

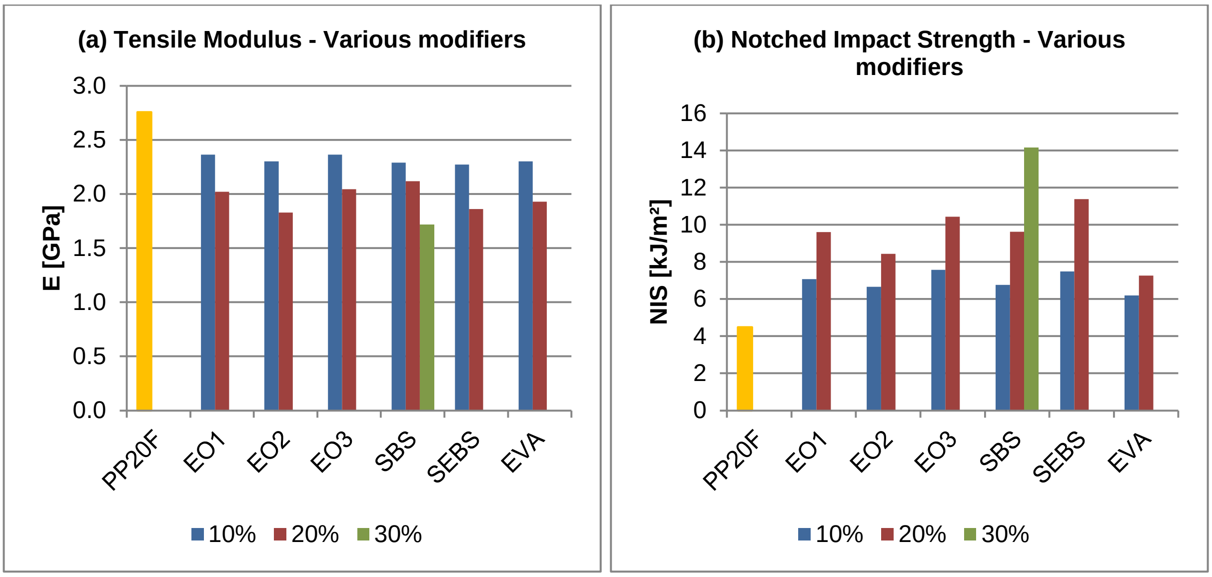

**Figure 2.** Influence of various impact modifiers on (a) the tensile properties and (b) the NIS.

### 3.2. Comparison with commercial materials

Regarding their overall mechanical performance, the formulations EO3_20 and SBS_30 were chosen as benchmark materials for the comparison with several commercially available, high-impact materials. Among those were two talc-filled PP composites and a 20 wt% cellulose reinforced PP composite.

In comparison with the talc reinforced Hostacom grades, none of the materials tested in this study came close to their impact performance. However, the formulation EO3_20 showed far better tensile properties, as well as a significantly reduced density. The weight advantage of SBS_30 over the talc filled grades was less prominent but still measurable; the tensile properties were in a similar range.
EO3_20 showed a significantly superior stiffness and NIS compared to the cellulose reinforced Formi EXP 20, and a slightly lower density. Tensile strength and IS values were slightly lower, but still comparable. SPS_30 exhibits a far higher NIS than Formi EXP 20, but lower IS, stiffness, and tensile strength.

**Table 4** Comparison of PP20F, EO3_20 and SBS_30 with several commercial materials. Data for Formi EXP 20 were obtained from the technical data sheet [13]. [1]NB = no break at impact.

| Code | $\rho$ (g/cm$^3$) | *MFR* (g/10 min) | $P_{max}$ (bar) | $E$ (GPa) | $\sigma_M$ (MPa) | *IS* (kJ/m²) | *NIS* (kJ/m²) |
|---|---|---|---|---|---|---|---|
| PP20F | 0.981 | 9.2 | 450 | 2.76 | 40.1 | 41.0 | 4.5 |
| EO3_20 | 0.977 | 4.7 | 580 | 2.05 | 29.4 | 43.0 | 10.4 |
| SBS_30 | 0.997 | 1.5 | 525 | 1.72 | 22.3 | 37.8 | 14.2 |
| Hostacom TRC333N | 1.062 | - | 550 | 1.90 | 21.4 | NB[1] | 31.7 |
| Hostacom EKC330N | 1.020 | - | 510 | 1.63 | 19.4 | NB[1] | 27.3 |
| Formi EXP 20 | 0.99 | - | - | 1.80 | 33 | 45 | 8.8 |

## 3. Conclusions

It could be shown that by adding a thermoplastic elastomer, the notched impact strength of cellulose reinforced PP can be considerably improved without significantly affecting the unnotched impact strength. However, the enhanced impact properties come at the cost of a reduction in stiffness and tensile strength; depending on the desired application, this is likely one of the most important limiting factors.

Generally a high viscosity and a low hardness of the modifier appear to be beneficial for the impact behaviour, but a soft material also leads to a more pronounced loss of stiffness. Among the materials tested in this study, ethylene-octene based POE and styrene-butadiene based elastomers led to the best overall mechanical performance. Within the given boundary conditions, for most of the modifiers a content of 20 wt% seemed to yield the best results; in the case of the used SBS grade, with a content of 30 wt% a very good notched impact strength was achieved while at the same time retaining acceptable tensile properties. However, overall the most well-balanced property profile was achieved with 20 wt% of EO3.

Some of the tested materials showed a clearly improved mechanical performance over the commercial cellulose reinforced reference material. While the impact properties of the commercial talc-filled reference materials were out of reach, their tensile properties could be surpassed. In addition, due to the lower density, a cellulose reinforced compound might find opportunities for applications where weight reduction plays a role.

## Acknowledgments

This work was funded within the scope of the Competence Centers for Excellent Technologies (COMET) programme of the Austrian Research Promotion Agency (FFG).

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