



The Use of Different Fractions of Waste Thermosets for Ther-moplastic Composites

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Abstract:

The use of recycled polymer materials with a low carbon footprint, which enable a circular economy, is becoming increasingly interesting for commercial use in the world. The main reasons for this are the accumulation of waste after the use of polymer products and the warming of the atmosphere as a result of the overloading of the environment with greenhouse gases produced during the extraction of these materials. We were interested in the differences in properties between differently modified samples of thermoset waste in a thermoplastic matrix. We paid the most attention to the influence of the modification of the interface on the stiffness and strength of the prepared material. The results of tensile and bending tests showed that with the right combination of compatibilizers, we could successfully increase the stiffness and strength of the composites. The simultaneous increase in stiffness and strength is a very good indicator that with the right combination of compatibilizers (PP-g-MA and modified TPU co-polymer) we were able to ensure a good surface interaction between the fibres and the polymer matrix in these samples with the right combination of compatibilizers (PP-g-MA and modified TPU copolymer). Waste paper was added to the thermoplastic matrix as a reference. The difference in the performance of the composites can be explained by the better wettability of the waste paper with the SEBS-g-MA compatibilizer and the better interfacial interactions of the modified TPU copolymer with the thermoset and the PP-g-MA with the glass fibres in the case of the waste thermoset composite.

Keywords: waste thermoset, compatibilizer, thermoplastics, different fraction of waste thermosets, composites



1. Introduction

The recycling of plastic waste, especially post-consumer recycling (PCR), has started and is still developing very slowly. Compared to other materials such as paper, glass and metals, recovery and recycling rates are generally low. Even in countries with advanced waste management systems and years of experience in recycling, the recycling rates of plastics are generally much lower than those of the other materials mentioned above. This is partly due to the wide variety of uses of plastics, types of additives and types of polymer composites. This diversity of uses is one of the main advantages of plastics, but it also poses a problem for recycling (Shen and Worrell, 2014).

Industrial waste is generally better separated than municipal waste, which is why municipal waste is sorted much more frequently than industrial waste. The same applies to washing, as municipal waste is usually much more contaminated. It is therefore easier to recycle industrial waste. As a result, we can produce recyclable materials of significantly higher quality and with better or, above all, more homogeneous properties (mechanical, chemical, optical) from this waste (Ragaert et al., 2017).

Polyolefins are a type of polymer that can be perfectly mechanically recycled, modified and reused in a variety of applications with suitable separation processes from industrial and municipal waste. The most common polyolefins include polypropylene (PP) and polyethylene (PE). Precisely because of their mechanical recycling properties and the possibility of modification, polyolefins are very often modified by adding various fillers in order to reduce the price and increase rigidity and strength. Due to their low cost, low density and high stiffness, natural fibres such as wood, cellulose, jute, bamboo, conifers and, last but not least, waste paper have attracted the attention of researchers. Thermoset waste and waste paper make up a large proportion of municipal waste which, if properly separated and processed, can be an excellent addition to the polymer matrix as a reinforcing agent, improving above all the mechanical properties of the biocomposite. The incorporation (dispersion) of fibres and the interfacial adhesion of thermoset waste and waste paper with the polyolefin matrix can be problematic. Therefore, improving the interfacial interactions between hydrophilic glass and natural fibres and hydrophobic polyolefins is an important research task, as the interfacial adhesion between glass and natural fibres and polyolefins plays an important role in determining the properties of composite materials. In this way, we can reduce the impact on the environment and realise the idea of transforming waste materials into valuable, commercially viable biocomposites (Bolka et al., 2020).

Thermoset composites are cured by a chemical reaction or by heat or radiation (Boquillon and Fringant, 2000; Hay and O'Gara, 2006; Walczyk and Kupperts, 2012). Their main advantage is excellent properties at the same time low production cost when combining the thermoset matrix with reinforcing fibers (Gore and Kandasubramanian, 2018; Blanco et al., 2021; Caydamli et al., 2021). At the end of the life of the composites, the waste stream can be divided into three main streams. Waste stream of smaller products that are not systematically collected at national or European level and end up as mixed waste. The second waste stream consists of large composite parts (parts of wind turbines, aeroplanes, ships). The third waste stream consists of industrial waste from the production of composite materials. For this waste stream, the composition and quantity of the waste are known (Colledani, 2022).

There are mechanical, thermal and chemical recycling technologies for composites. Regardless of the technology, the reinforcement fibres obtained cannot completely replace the virgin fibres, but can only be added to the virgin fibres to a certain percentage in order to maintain the properties of the composites. Mechanical recycling always takes place in several steps. Firstly, large composite parts are cut into smaller pieces that can be shredded. This is followed by shredding and, if necessary, screening, during which fractions of different sizes are obtained. The smallest fraction in the form of dust can be used as filler or reinforcement (Bernardeau et al., 2018), but is usually utilised for energy by incineration. Larger fractions are usually used in the manufacture of new composite products (Pickering, 2006; Oliveux et al., 2015).



This paper presents a two-pronged review of thermoset composite waste. In the first part, the use of the smallest fraction of waste thermoset composites in combination with a thermoplastic polyamide matrix (Bernardeau et al., 2018) is presented. In the following, the use of a larger fraction as reinforcement for a thermoplastic recycled polypropylene matrix (Bream and Hornsby, 2000; 2001) is presented, using composites with waste paper for comparison (Bourmaud and Baley, 2007; Zhidan et al., 2011; Xiaolin, Xiangfeng and Rumin, 2013; Akbulut et al., 2016; Scholten and Meiners, 2019).

2. Methods & materials

Commercially available PA 6 (Badamid B70) was purchased from Bada, Germany (PA6 in Table 1). Commercially available recycled polypropylene was donated by TAB-IPM, Slovenia in ground form (rPP in Table 2). Commercially available antioxidant (AT 10 – AO in Tables 1 and 2) was purchased from AMIK ITALIA, Italy. Commercially available lubricant (Crodamide ER – SA in Table 1) was purchased from Croda, Italy. Commercially available ethylene elastomer modified with compatibiliser (Fusabond N416) was purchased from DuPont, Switzerland (C₁ in Table 1). The commercially available compatibilizer PP-g-MA (Exxelor PO 1020) was purchased from Exxon Mobil, Netherlands (C₂ in Table 2). The commercially available compatibilizer SEBS-g-MA (Taipol 7126) was purchased from TSRC Corporation, Taiwan (C₃ in Table 2). Commercially available compatibilizer modified TPU copolymer (Kuramiron U TU-S5265) was purchased from Kuraray Europe, Germany (C₄ in Table 2). The waste paper was donated by Papirnica Vevče, Slovenia in ground form (WP in Table 2). The thermoset composite waste in dust form was donated by the company Iskra ISD plast, Slovenia (rTC₁ in Table 1).

Table 1. Sample composition with PA6 matrix.

Sample	PA6 (wt.%)	AO (wt.%)	SA (wt.%)	C ₁ (wt.%)	rTC ₁ (wt.%)
PA6	95.5	0.5	0	0	0.0
PA6 AO rTC ₁ 0.1	99.4	0.5	0	0	0.1
PA6 AO rTC ₁ 0.5	99.0	0.5	0	0	0.5
PA6 AO rTC ₁ 1.0	98.5	0.5	0	0	1.0
PA6 AO SA1 C ₁ 5 rTC ₁ 30	63.5	0.5	1	5	30.0

Thermoset composite waste as a fraction sieved through a 4 mm sieve was donated by Technol, Slovenia (rTC₂ in Table 2). The composition of the samples is listed in Tables 1 and 2. For all tests, the materials were mixed separately and extruded on the Labtech LTE 20-44 twin-screw extruder. The screws had a diameter of 20 mm, an L/D ratio of 44:1, a screw speed of 600 rpm and an increasing temperature profile for PA6 and rPP from the hopper (165 °C and 220 °C, respectively) to the die (190 °C and 250 °C, respectively) (Pracella et al., 2010).

The injection moulding was carried out on a Krauss Maffei 50-180 CX with a screw diameter of 30 mm. The temperature profile for PA6 and rPP increased from the hopper (175 °C and 220 °C) to the nozzle (190 °C and 250 °C, respectively). The mould temperature for PA6 and rPP was set to 80 °C and 45 °C, respectively and the cooling time to 10 s (Jazani et al., 2011). The tensile tests were carried out with the Shimadzu AG-X plus in accordance with ISO 527-1 (Huang et al., 2013). Tensile stiffness (Et), tensile strength (om), elongation at yield (em) and elongation at break (etb) were evaluated in the tensile tests. Five measurements were carried out for each sample. The thermomechanical properties were analysed using a Perkin Elmer DMA 8000 (Wunderlich, 2005).

**Table 2.** Sample composition with recycled PP matrix.

Sample	rPP (wt.%)	AO (wt.%)	C ₂ (wt.%)	C ₃ (wt.%)	C ₄ (wt.%)	WP (wt.%)	rTC ₂ (wt.%)
A	100.00	0.00	0.0	0.0	0.0	0	0
B	69.62	0.38	0.0	0.0	0.0	30	0
C	64.62	0.38	0.0	0.0	5.0	30	0
D	64.62	0.38	2.5	0.0	2.5	30	0
E	64.62	0.38	0.0	2.5	2.5	30	0
F	69.62	0.38	0.0	0.0	0.0	0	30
G	64.62	0.38	0.0	0.0	5.0	0	30
H	64.62	0.38	2.5	0.0	2.5	0	30
I	64.62	0.38	0.0	2.5	2.5	0	30

The samples were heated at 2 °C/min from 25 °C to 210 °C and 170 °C under air atmosphere for PA6 and rPP matrix based composites, respectively. A frequency of 1 Hz and an amplitude of 20 μm were used in dual cantilever mode (Jazani et al., 2011). Thermogravimetric analyses (TGA) were performed on a Mettler Toledo TGA/DSC 3+ thermal analysis instrument. The analyses were carried out in a nitrogen atmosphere (20 mL/min) from 40 to 550 °C with a heating rate of 10 °C/min, followed by an isothermal segment in an oxygen atmosphere (20 mL/min) at 550 °C for 30 min. using an Al₂O₃ crucible. Thermal measurements were performed using a differential scanning calorimeter (DSC 2, Mettler Toledo) under a nitrogen atmosphere (20 mL/min). The temperature of the samples was raised from 0 to 260 °C for PA6 and 0 to 180 °C for rPP samples at a heating rate of 10 °C/min and kept in a molten state for 5 minutes to extinguish the thermal history. After cooling at 10 °C/min, the samples were reheated to 260 °C for PA6 and to 180 °C for rPP at 10 °C/min. The crystallisation temperature (T_c), the crystallisation enthalpy (ΔH_c), the melting temperature (T_m) and the melting enthalpy (ΔH_m), the glass transition temperature (T_g) and change in the specific heat capacity (Δc_p) were determined from the cooling and the second heating scan. (Wunderlich, 2005). The impact tests were performed with the Dongguan Liyi Test Equipment pendulum, type LY-XJJD5 (Zhidan et al., 2011), in accordance with ISO 179. The distance between the supports was 60 mm and a 1 J pendulum was used.

3. Results and Discussion

3.1. Tensile tests

A comparison of E_t in **Table 3** shows that the addition of waste thermoset composite powder as nucleating agent greatly increases E_t and that 0.5 wt.% of added waste thermoset composite powder is the optimum amount. When 30 wt.% of waste thermoset composite is added and in combination with a compatibilizer, E_t even decreases slightly. The tensile strength is also increased when waste thermoset powder is added as a nucleating agent, and the highest increase in σ_m is also achieved with 0.5 wt.% waste thermoset powder added. At an addition of 30 wt.% and in combination with a compatibilizer, the tensile strength decreases drastically, as this is a consequence of the shape of the particles, which only act as a filler and not as a reinforcing agent. In terms of elongation, both ε_m and ε_{th} , the trend is that elongation decreases with increasing addition of thermoset composite powder. However, they decrease drastically when 30 wt.% of waste thermoset composite powder is added and in combination with a compatibilizer.



Table 3. Tensile tests results with PA6 matrix.

Sample	E_t (GPa)	σ_m (MPa)	ϵ_m (%)	ϵ_{tb} (%)
PA6	2.31 ± 0.20	66.3 ± 0.4	4.3 ± 0.1	158.6 ± 64.8
PA6 AO rTC ₁ 0.1	2.78 ± 0.25	68.1 ± 0.2	4.4 ± 0.1	15.3 ± 7.2
PA6 AO rTC ₁ 0.5	2.97 ± 0.20	69.9 ± 1.5	4.2 ± 0.3	6.2 ± 3.0
PA6 AO rTC ₁ 1.0	2.90 ± 0.17	65.9 ± 6.5	3.9 ± 0.7	5.1 ± 2.2
PA6 AO SA1 C ₁₅ rTC ₁ 30	2.25 ± 0.27	47.0 ± 0.2	3.5 ± 0.2	3.8 ± 0.4

We can conclude that the waste thermoset composite powder acts as a nucleation agent and that the optimum amount to add is 0.5 wt.%. The waste powder of the thermoset composite could also be used as a filler, but the σ_m and strains are drastically reduced.

The E_t comparison shown in **Table 4** clearly shows that the rPP samples with added fibres achieved significantly higher E_t values than the pure rPP polymer matrix samples. This is of course the expected result, as E_t increases when fibres are added to the polymer, regardless of whether they are synthetic or natural fibres. The E_t value of the pure rPP matrix was 0.9 GPa. Among the rPP samples with added waste paper fibres, the highest E_t value was achieved by the D sample and the lowest by the C sample. The E_t values of the rPP and waste paper samples with various additives (modifiers) were between 1.26 GPa and 1.69 GPa. Among the samples containing waste thermoset composite with glass fibres and various additives, the highest E_t value was achieved by the F sample and the lowest by the G sample. The values of the tensile modulus of the samples containing the thermoset composite with glass fibres were between 1.52 GPa and 1.98 GPa. We can conclude that the addition of fibres to the polymer matrix improves the E_t of composite. Even better properties can be achieved by modifying the interface between matrix and fibres, namely by adding antioxidants, modifiers and compatibilizers. Improving the interface between fibres and matrix means better interfacial interactions, as the modifiers act as binders both on the side of the polymer matrix and on the side of the fibres.

A comparison of σ_m and ϵ_m is shown in **Table 4**. **Table 4** for σ_m shows that σ_m of the pure rPP sample is 25.7 MPa. In the following, we can observe a slightly higher σ_m of the rPP materials to which paper fibres and additives were added, while at the same time a decrease in ϵ_m can be observed for all samples, which is due to the added fibres, as their elongation is much more limited than that of the rPP matrix. It can be observed that the B sample, which contained rPP and paper fibres but no additives, reached the same σ_m value (27.7 MPa) as the samples made of pure rPP and that at the same time its ϵ_m value decreased significantly, namely to an elongation of 5.4 %. For all other rPP samples with added paper and additives, we can observe a concrete increase in the σ_m value and at the same time a decrease in the ϵ_m value compared to the samples made of pure rPP. The highest σ_m value was achieved by the sample D, which reached 33.3 MPa at an elongation of 5.7 %. The worst tensile properties of the rPP samples with added paper and additives were exhibited by the sample C, which achieved a value of σ_m of 25.6 MPa at an elongation of 5.3 %. It can be concluded that all combinations of additives in the samples proved to be effective, as all combinations achieved better results than the rPP sample with added paper without additives. The exception was the addition of modified TPU copolymer to the C sample, which achieved comparable results to the B sample. The combination of the compatibilizers PP-g-MA and modified TPU copolymer had the greatest influence on the improvement in tensile strength. The σ_m values of the rPP samples with the addition of a waste thermoset composite with glass fibres and additives decreased and were between 19.8 MPa and 21.7 MPa. The only exception was the sample H, which reached σ_m of 28.0 MPa. In addition, ϵ_m decreased for all samples. The elongations of the samples varied between 3.1 % and 4.7 %. The lower tensile strengths can be attributed to poorer interfacial interactions between the matrix and the waste thermoset composite with glass fibres, the lower elongations are the result of significantly lower elongations of the glass fibres and the proportion of waste thermoset composites compared to cellulose fibres. Similar to the rPP samples with added waste paper, the combination of the compatibilizers PP-g-MA and modified TPU copolymer proved to be the best additive.



The comparison of ϵ_{tb} shown in **Table 4** is extremely revealing as we can see how the addition of fibres to the matrix of pure rPP significantly affects the ability of the composite to elongate before it collapses. The elongation at break decreased from 289.5 % for the pure rPP sample to a minimum of 6.4 % to a maximum of 7.4 % for the rPP samples with added waste paper and additives and to a minimum of 3.8 % to a maximum of 11.2 % for the samples with added waste thermoset composites with glass fibres and additives. The decrease in elongation at break can be attributed to the limited tensile properties of the added waste paper fibres and glass fibres, which allowed less plastic deformation (more brittle fracture) due to their presence in the matrix.

Table 4. Tensile tests results with recycled PP matrix.

Sample	E_t (GPa)	σ_m (MPa)	ϵ_m (%)	ϵ_{tb} (%)
A	0.90 ± 0.12	25.7 ± 0.7	7.8 ± 0.4	289.5 ± 128.4
B	1.64 ± 0.24	24.7 ± 0.5	5.4 ± 0.3	7.3 ± 0.4
C	1.26 ± 0.17	25.6 ± 0.3	5.3 ± 0.2	6.5 ± 0.7
D	1.69 ± 0.37	33.3 ± 0.3	5.7 ± 0.1	6.4 ± 0.3
E	1.64 ± 0.20	29.5 ± 0.3	5.9 ± 0.2	7.0 ± 0.5
F	1.98 ± 0.18	21.6 ± 0.2	3.2 ± 0.4	7.6 ± 2.4
G	1.52 ± 0.14	19.8 ± 0.2	4.7 ± 0.2	11.2 ± 1.8
H	1.83 ± 0.19	28.0 ± 0.2	3.1 ± 0.2	3.8 ± 0.3
I	1.54 ± 0.21	21.7 ± 0.1	3.2 ± 0.1	8.9 ± 1.4

With the right combination of compatibilizers, we were able to successfully increase the stiffness and strength of the composites, and in all cases the elongation at break was drastically reduced. We were able to increase the stiffness of the composites with waste paper by 82 % and the strength by 33 %, and the stiffness of the waste thermoset composites by 120 % and the strength by 9 %. The simultaneous increase in stiffness and strength is a very good indicator that we have succeeded in ensuring good surface interactions between the surface of the fibres and the thermoplastic matrix in these composites with the right combination of compatibilizers. By using different combinations of compatibilizers, we can produce a composite material with precisely defined properties. The lower strength increase in composites with a waste thermoset composite is to be expected because the waste thermoset composite contains approx. 60 % cured thermoset, which is very brittle, and only 40 % glass fibres. For composites with added waste paper, the partial volumes containing the waste paper particles are much smaller than in the waste thermoset composites, so the partial strains at the interface between the waste paper and the rPP matrix are also much smaller compared to the waste thermoset composite. The size of the partial volumes is reflected in the differences in the tensile strength of these composites.

3.2. DMA tests

The storage modulus (**Figure 1**) in the range from 40 to 90 °C for samples with PA6 matrix is consistent with the results of the tensile test. The more waste thermoset composite powder is added, the higher the storage modulus. The highest storage modulus is found in the entire temperature range for the composite PA6 AO SA1 C₁₅ rTC₁₃₀, to which 30 wt.% of waste thermoset composite powder was added. All samples show drastic drop in the storage modulus (around 50 °C) in the area of the glass transition of PA6, although this depends on the amount of waste thermoset powder added.

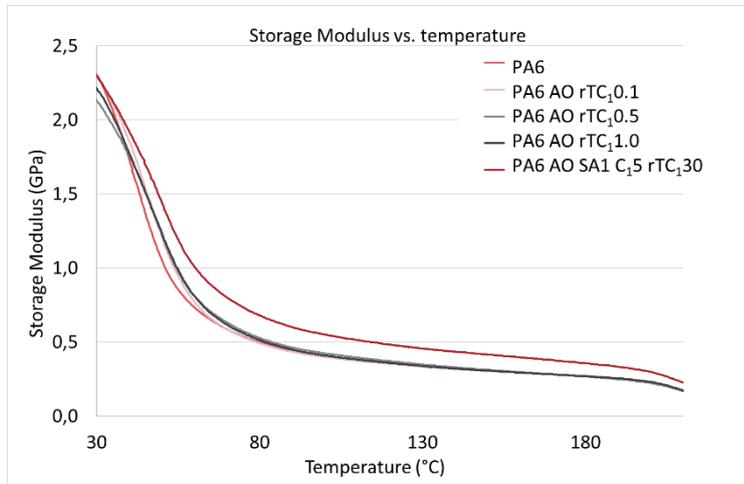


Figure 1. Storage modulus vs. temperature for the composites with PA6 matrix.

The glass transition increases with the amount of waste thermoset powder added. This can also be seen in **Figure 2**, which shows the loss factor. The level of the loss factor illustrates the elastic response of the material and is lowest at 0.5 wt.% for the versions to which the waste thermoset composite powder is added. The result is consistent with the tensile tests, in which the PA6 AO rTC_i0.5 sample showed the highest stiffness and strength. The PA6 AO SA1 C_i5 rTC_i30 sample has the peak of loss factor at the highest temperature and the lowest peak height. This means that the sample has the highest glass transition and reacts most elastically of all the samples measured. This is due to the waste thermoset composite powder added at 30 wt.% which acts as a filler but still influences the stiffness and dynamic behaviour of the composite.

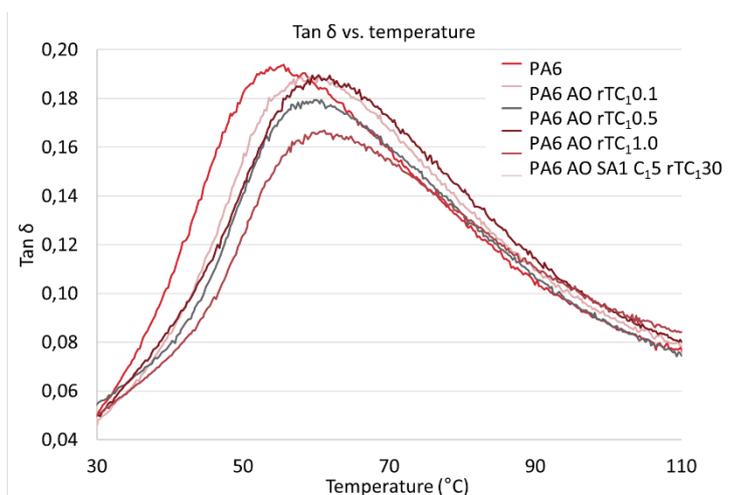


Figure 2. Loss factor vs. temperature for the composites with PA6 matrix.

The storage modulus for the samples with rPP matrix (**Figures 3 and 4**) is significantly higher for composites with waste paper and waste thermoset composites in the entire temperature range than for the pure rPP matrix. The highest storage modulus is exhibited



by the composite samples to which a combination of PP-g-MA compatibilizers and modified TPU copolymer was added. The storage modulus is lower for composites with waste paper than for composites with waste thermoset composites. This result was expected, as glass fibres have a much higher stiffness than cellulose fibres in waste paper, despite their low proportion in the waste thermoset composite. Regardless of the composition, the temperature resistance of the composites is also drastically improved compared to the pure rPP matrix. For composites with waste thermoset composite, the F sample, to which no compatibilizer has been added, is comparable to the H sample. This means that the waste thermoset composite is homogeneously mixed with the rPP matrix. At a temperature of 70 °C, the storage modulus of the F sample starts to decrease compared to the H sample, as there are no good interfacial interactions between rTC₂ and the rPP matrix.

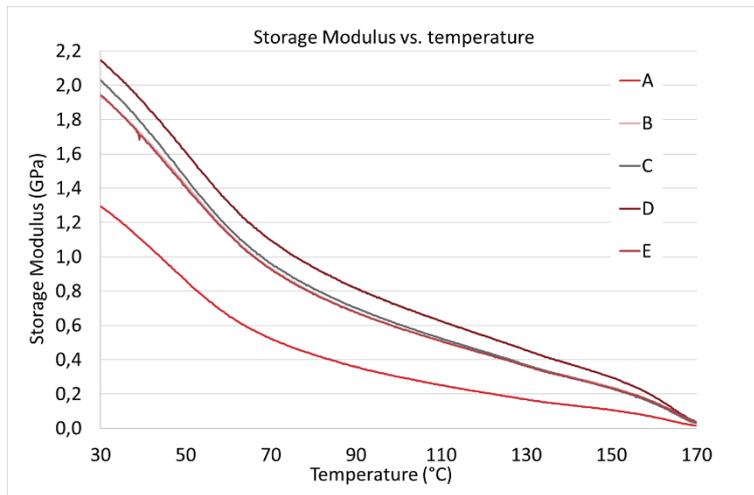


Figure 3. Storage modulus vs. temperature for the composites with rPP matrix and waste paper.

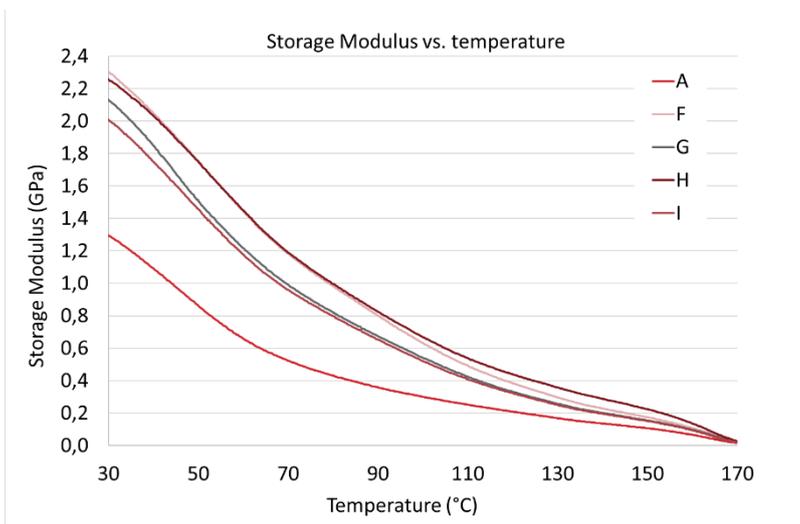


Figure 4. Storage modulus vs. temperature for the composites with rPP matrix and recycled thermoset composites.

The loss factor (Figure 5) in the rPP sample does not show a pronounced peak. The samples with added rTC₂ show pronounced peaks, which can be attributed to the glass transition of the thermoset resin in the source of the waste thermoset composite. The peak of the glass transition for the samples is between 100 °C and 110 °C, because this means that the compatibilizers in the samples are differently compatibilized for both the rPP matrix and the thermoset resin. The combination of the compatibilizers PP-g-MA and modified



TPU copolymer shows the best compatibility, as the glass transition for the thermoset resin has moved to lower values (closest to the glass transition for the rPP matrix).

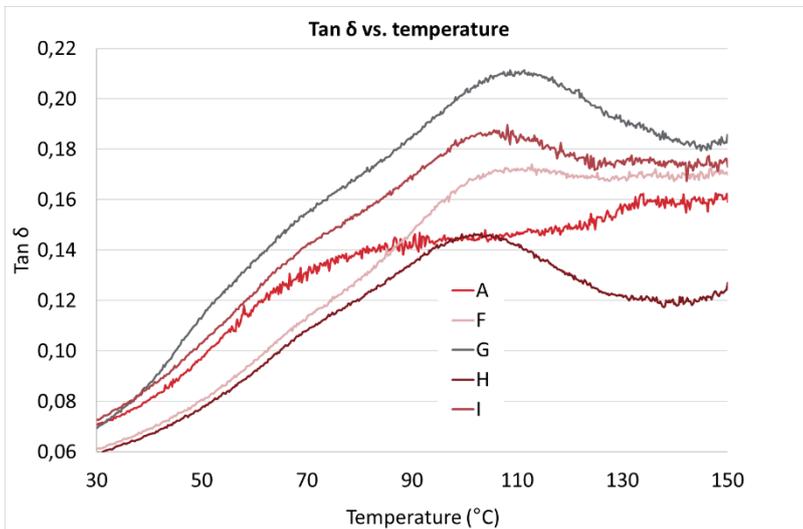


Figure 5. Loss factor vs. temperature for the composites with rPP matrix and recycled thermoset composites.

From the results, we can conclude that the compatibilizer PP-g-MA has good interactions between the glass fibers and the rPP matrix and has a great influence on the mechanical properties of the composite, while the modified TPU copolymer compatibilizer has good interactions between the thermoset resin and the rPP matrix and has great influence on the thermal properties of the composite, but has a weaker influence on the mechanical properties of the composite.

3.3. TGA tests

The characterization of the inorganic residues for the samples with PA6 matrix (Figure 6) after TGA annealing shows that there are quite a lot of inorganic additives in the pure PA6 matrix, which are probably processing additives. In the versions to which 0.1 to 1.0 wt.% rTC₁ was added, there were no differences in the inorganic residues after TGA annealing. In the PA6 AO SA1 C₁₅ rTC₁:30 sample, the inorganic residue was just under 4 wt.%, which means that only about 13 wt.% glass fibres are contained in the rTC₁.

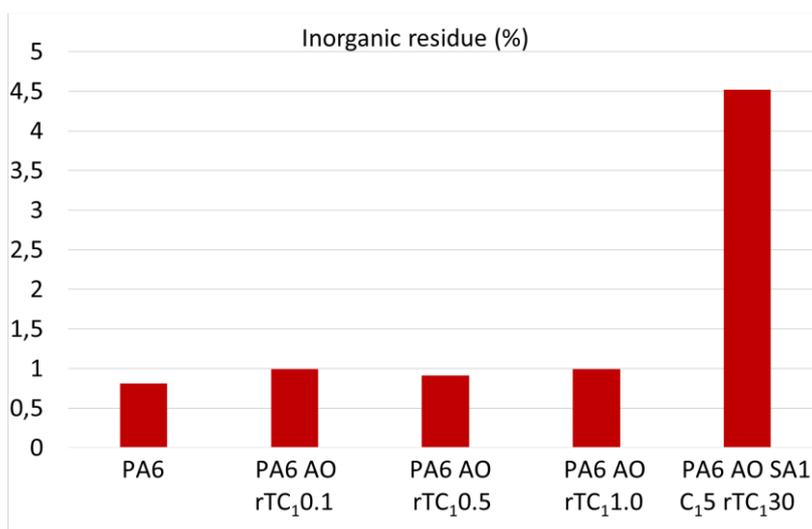




Figure 6. Inorganic residues after TGA measurements for the samples with PA6 matrix.

The characterization of the inorganic residues for the samples with rPP matrix (**Figure 7**) after tempering on the TGA shows that 0.3 wt.% of inorganic impurities are contained in the rPP matrix. In the samples of composites with waste paper, the inorganic residues are between 10.9 and 11.8 wt.%. In samples of composites with rTC₂, the inorganic residues are between 11.7 and 12.3 wt.%. In composites with recovered paper, there is an inorganic residue of CaCO₃, which is added to the paper as a filler and is present in the recovered paper between 36 and 39 wt.%. In rTC₂, the inorganic residue consists of glass fibres, which account from 39 to 41 wt.% in rTC₂.

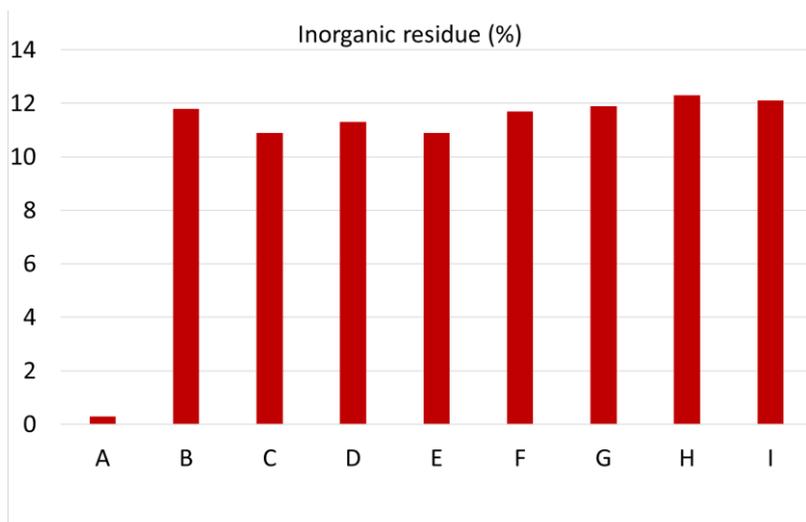


Figure 7. Inorganic residues after TGA measurements for the samples with rPP matrix.

3.4. DSC tests

For samples with a PA6 matrix (**Table 5**), the addition of small percentages of rTC₁ has practically no effect on the glass transition or crystallization on cooling. From this we can conclude that we cannot detect any change in the morphology of the composites with DSC measurements. In the PA6 AO SA₁ C₁₅ rTC₁30 sample, however, the glass transition is significantly higher, the crystallization temperature, the melting enthalpy and also the melting point decrease slightly.

Table 5. DSC tests results for samples with PA6 matrix.

Sample	T _g (°C)	C _p (J/gK)	T _c (°C)	ΔH _c (J/g)	T _m (°C)	ΔH _m (J/g)
PA6	55.2	0.05	192.2	31.1	220.9	38.5
PA6 AO rTC ₁ 0.1	55.6	0.06	191.9	32.4	221.1	36.8
PA6 AO rTC ₁ 0.5	53.6	0.03	191.8	30.0	221.1	37.4
PA6 AO rTC ₁ 1.0	55.0	0.05	192.1	32.6	220.7	38.5
PA6 AO SA ₁ C ₁₅ rTC ₁ 30	61.4	0.02	190.7	21.0	220.0	24.5

For the samples with rPP matrix (**Table 6**), the first melting point is the melting of the low PE content in the rPP matrix and thus the crystallization temperature for the PE content. The melting point and crystallization temperature for the rPP matrix are in the temperature range of 165.4 °C to 166.4 °C and 121.5 °C to 124.4 °C respectively. Composites with a combination of PP-g-MA compatibilizers and modified TPU copolymer exhibit the high-



est crystallization temperatures, which means that this combination triggers heterogeneous crystallization of the rPP matrix during cooling. These two composites also achieve the highest degree of crystallinity in the versions with added compatibilizers.

Table 6. DSC tests results for samples with recycled PP matrix.

Sample	2 nd Heating				Cooling			
	T _{m1} (°C)	ΔH _{m1} (J/g)	T _{m2} (°C)	ΔH _{m2} (J/g)	T _{c1} (°C)	ΔH _{c1} (J/g)	T _{c2} (°C)	ΔH _{c2} (J/g)
A	126.4	11.4	165.6	79.0	112.5	9.5	124.0	76.5
B	126.1	8.1	165.8	52.3	112.6	6.9	123.6	51.0
C	126.0	6.9	165.9	49.9	112.3	6.0	122.7	48.9
D	125.9	8.9	166.2	55.3	111.1	6.3	124.4	52.7
E	125.6	6.3	165.4	47.7	112.3	5.5	121.9	46.7
F	126.5	6.8	166.4	54.9	112.4	6.2	123.1	54.2
G	126.3	7.5	166.1	52.5	112.3	6.0	122.6	51.5
H	125.9	8.8	165.8	53.7	111.2	6.6	123.3	50.8
I	125.8	6.0	165.6	50.4	112.0	5.5	121.5	50.2

3.5. Notched impact tests

With regard to the notched impact strength (**Figure 8**), a drastic decrease in all composites samples can be seen, both with waste paper and with added rTC₂. The notched impact strength is slightly higher for composites with waste paper than for composites with rTC₂, which is due to the low glass fibre content of rTC₂. Compatibilizers increase the notched impact strength in all variants. This increase in notched impact strength is more pronounced in composites with rTC₂, as the polyester resin without compatibilizer does not have good interfacial interactions with the rPP matrix. The sample G has the highest notched impact strength, which means that the modified TPU copolymer with the compatibilizer improves the interactions between the rPP matrix and the polyester resin the most. The results are in very good agreement with the results of the tensile and DMA tests.

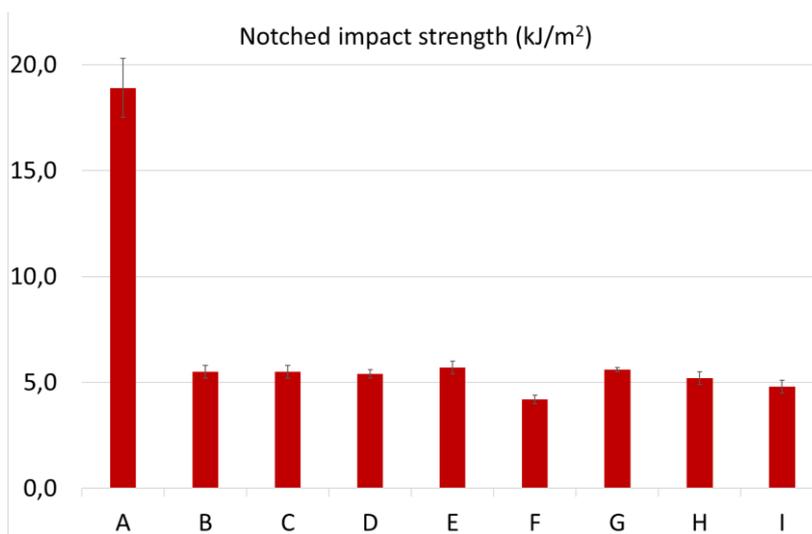


Figure 8. Notched impact strength for the composites with rPP matrix and recycled thermoset composites.



4. Conclusions

The tests carried out have shown that with the right choice of thermoplastic matrix, both fine powder from thermoset waste and larger fractions can be used to improve the properties of the newly created composite. The fine powder from thermoset waste works perfectly as a core in the PA6 matrix and the composite material improves stiffness, strength, temperature resistance and glass transition, but drastically reduces elongation at break and toughness. Larger proportions of thermoset waste, which in our case contained only about 40 wt.% of glass fibres, improve the stiffness, strength and temperature resistance of the composite in combination with the right compatibilizers and the rPP matrix, but drastically reduce the elongation at break and toughness. For the rPP matrix, PP-g-MA and modified TPU copolymer proved to be the best combination of compatibilizers, with PP-g-MA providing good interfacial interactions between glass fibers and rPP matrix and modified TPU copolymer between thermoset matrix particles and rPP matrix. The results provide a good basis for further research into the mechanical recycling of thermoset waste as nucleating agent or reinforcement for thermoplastic matrices. In this way, mechanical recycling could be used to utilise the entire amount of waste thermoset composite, as fine dust is a by-product or waste from grinding for larger fractions.

Conflicts of Interest: The authors declare no conflict of interest.

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