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Sustainable approach to river buoy production using recyclable composites based on thermoplastic polymer wastes

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ABSTRACT

This paper presents a novel approach to the production of river buoys, focusing on the use of new composite materials with high recyclability at the end of the buoy's life cycle. The composites were produced from post-production thermoplastic waste - polyethylene and polyamide 6 - reinforced with glass fibre, and modified using block copolymer polyethylene maleic-grafted-anhydride (PE-g-MAH) to improve the heterogeneity of the waste structure. The composites were prepared by melt compounding with a corotating twin-screw extruder. The melting behaviour, rheology, degree of crystallinity, and moisture sorption characteristics of the rPE/rPA6-30GF/CoM compositions were investigated. Rheological behaviour was analysed using an oscillatory rheometer. Given the operating conditions of the new composites, key functional properties such as specific density and water absorption of the rPE/rPA6-30GF composites were also assessed. The structure of the composites was further examined using scanning electron microscopy (SEM). The results showed that modifying the rPE/rPA6-30GF composite with varying amounts of compatibilizer led to an increase in specific density and a decrease in water absorption, compared to unmodified polyethylene-polyamide 6 composites containing 30% glass fibre. These improvements were attributed to increased homogeneity of the composite structure as a result of the PE-g-MAH modification. The findings indicate that polyethylene–polyamide-based waste polymer composites can be effectively used as potential materials for constructing navigation buoys.

Keywords: modification, recycling, polyethylene, physico-chemical properties, polymer composites, circular economy, plastics waste, river buoys.

INTRODUCTION

The selection of materials for structural components is based on an assessment of their properties suitable for the intended environmental conditions. Nowadays, it is also crucial that material selection align with the principles of the circular economy (1–3), especially in light of the growing volume of post-use and post-production polymer waste. Examples of such materials include polyethylene and polypropylene, which are widely used in the construction of waterway markers, commonly known as buoys. A buoy is a floating structure anchored to the seabed, used to indicate navigational hazards such as reefs and shallow waters and to guide ships along safe sea routes (4). Traditionally, buoys are manufactured from steel. However, as demonstrated by Park (4) in a 2016 study, using polyethylene significantly reduces the weight of buoys compared to steel and offers other benefits that have supported the growing adoption of plastic materials for navigational markers.

Despite these advantages, such markers are continuously exposed to degradation due to harsh weather conditions and marine environments (e.g., saltwater). Additionally, the quality of buoy products supplied by manufacturers is a concern, with observed issues including deformation, structural flexibility, and discoloration (see Figure 1). Buoys are susceptible to mechanical and structural damage (creep, stress relaxation, excessively thin walls) as well as operational damage (inappropriate storage conditions). In addition, they are subject to post-operational discolouration on the surface of the laminate, which is associated with the accumulation of plankton on the fin, the effects of water and UV radiation.

Therefore, there was a need to identify high-quality, environmentally friendly materials, particularly to enable their reuse in the cases where a product is defective. The issue of plastic waste contributing to the pollution of marine and terrestrial ecosystems is widely acknowledged. The research by Sainte-Rose et al. (5) has shown that the accumulation of plastic waste on land, along shorelines, and within aquatic environments represents one of the most urgent environmental challenges of the 21st century. The ongoing efforts aim to develop effective strategies for managing plastic waste, including the implementation of efficient disposal methods through mechanical processing. For instance, Lamba P (6) provided a comprehensive review of physical modification technologies for plastic waste, with a particular focus on their application in the production of construction materials.

However, even with the appropriate choice of technology (7) and the definition of downstream processing parameters (8), physicochemical challenges may still arise when combining polymers in the molten state, particularly those derived from waste. Therefore, it is essential to evaluate the feasibility of combining such polymers through the addition of so-called compatibilisers, which act as bridging agents in the formation of mixtures and composites. The authors of the present study conducted research aimed at supporting the sustainable development of polymer material management (9). They investigated the potential impact of compatibilisers on the interactions between polymer recyclates (10) in order to determine the properties of the recovered materials. Similar studies were carried out by Martikka et al. (11), who explored the influence of various compatibilisers on the properties of polymer composites made from plastic waste combined with wood. The goal was to promote plastic recycling and evaluate the potential use of plastic waste originating from wood residues in composite materials. The researchers found that the addition of selected compatibilisers significantly improved the properties of polymer composites containing wood waste—with all tested mechanical properties and moisture resistance showing enhancements of 50% or more. Similarly, the research conducted by Volokhova (12) and collaborators focused on blends of cellulose triacetate (CTA) and poly(l-lactic acid) (PLA), which were prepared using grafted polymer compatibilisers. These grafted polymers were synthesised via a ring-opening polymerisation of *l*- and

Figure 1. Damaged river buoys made of composite material after the production process

d,d-lactide, initiated by the free hydroxyl groups present along the CTA backbone. Evaluative studies using energy-dispersive X-ray spectroscopy (EDS) demonstrated that the crystallisation behaviour of PLA was influenced by the addition of both the compatibiliser and CTA (12). Furthermore, Ahmad Shamsuri and Siti Nurul Ain Md. Jamil (13) provided a concise overview of the use of quaternary ammonium compounds (QACs) as compatibilisers in polymer blends and composites, aimed at enhancing their thermomechanical properties. They found that QACs used in various polymer composite systems can be broadly categorised into two main groups: quaternary ammonium surfactants (QAS) and quaternary ammonium ionic liquids (QAIL). Appropriate compatibilisation of polymer composites using QAS and QAIL can effectively induce intermolecular interactions between the components of the blend. In addition, polymer composites compatibilised with QAS and QAIL can promote strong interactions between the polymer matrices and the fillers. The presence of such interactions enhances interfacial adhesion and overall compatibility among the components of the polymer composite. This improved compatibility, in turn, leads to enhanced thermomechanical properties. Similar challenges were addressed by Boutevin B. et al. and Laguna O. et al. (14,15). The preparation of blends incorporating glass fibre-reinforced polyamide 6 (16,17) facilitates the development of novel materials with improved strength, stiffness, thermal resistance, and impact performance. Polyamide 6 contains numerous amide bonds that tend to interact with the modified, active surface of glass fibres (17).

This study aimed to demonstrate the applicability of new composite materials produced by modifying polyethylene with polyamide 6 and glass fibre for the manufacture of floating buoys that are lighter, more environmentally friendly, and safer for vessels.

RESEARCH MATERIAL AND METHODS

Materials composition

The research material consisted of recycled polyethylene (rPE) and waste polyamide 6 containing 30 wt.% glass fibre (rPA6-30GF), as well as a composite obtained by melt blending these polymers with the addition of a compatibiliser polyethylene-graft-maleic anhydride (PE-g-MAH, hereafter referred to as CoM) in a co-rotating twin-screw extruder. The CoM was used at concentrations of 1 wt.% and 3 wt.%. CoM functions by stabilising the dispersed polymer phases, reducing interfacial stress between the recycled polyethylene and polyamide 6 phases. The resulting composites and their corresponding sample codes were as follows:

- 100% recycled polyamide 6 with 30 wt.% GF (abbrev. rPA6-30GF),
- 100% recycled polyethylene (abbrev. rPE),
- unmodified composite recycled (abbrev. rPE/rPA6-30GF),
- modified composite by 1 wt. % PE-g-MAH (abbrev. rPE/rPA6-30GF/1CoM),
- modified composite by 3 wt. % PE-*g*-MAH (abbrev. rPE/rPA6-30GF/3CoM).

Prior to composite preparation, the components, particularly polyamide 6 with glass fibre and the compatibiliser were dried in a vacuum dryer at 80 °C for 4 hours.

Characterised techniques

Scanning electron microscope (SEM)

The fractured surfaces of the rPE/rPA6-GF/CoM composites were examined using a Mira 3 scanning electron microscope (SEM) (Tescan, Czech Republic), employing both backscattered electron (BSE) and secondary electron (SE) imaging for high-resolution analysis. BSE signals originate from deeper regions within the composite sample, whereas SE signals are generated from near-surface areas. Prior to imaging, the samples were coated with a thin (20 nm) layer of carbon powder to improve conductivity and image quality.

Thermal differential scanning calorimetry (DSC)

The melting behaviour of rPE/rPA6-30GF composites modified with CoM was analysed using a DSC 204 F1 Phoenix (Netzsch, Germany), operating under a protective nitrogen atmosphere (flow rate: 30 mL/min). Approximately 10 mg of each sample was heated from room temperature to 270 °C at a rate of 20 °C/min, followed by cooling to 20 °C at a rate of 10 °C/min to eliminate the thermomechanical history of the materials. The degree of crystallinity (X_c) for the rPE and rPA6-30 GF/CoM composites was determined using Equation 1.

$$X_c = \frac{\Delta H_m}{\Delta H_m^0} \cdot 100\% \quad (1)$$

where: ΔH_m is the melting enthalpy (J/g), and ΔH^0_m is the calculated melting enthalpy of purely crystalline PE, being 293 J/g (18).

Density and water absorption

The solid mass of recycled PA6-30GF, polyethylene, and their composites was measured using an AXIS AD50 balance (Poland). Density was determined in 96% pure ethyl alcohol (ρ = 0.786 g/cm^3) using the hydrostatic method, in accordance with PN-EN ISO 1183-1:2025 (19). To evaluate the hydrophobicity of the recycled polymers and their composites, water absorption tests were conducted over periods of 24, 48, and 72 hours. These tests were carried out on sets of five samples for each material: rPE, rPA6-30GF, and the rPE/rPA6-30GF/CoM composites, following the ISO 62:2008 standard (20).

Dynamic thermo-mechanical analysis (DMTA)

The rheological properties of the composites were evaluated using an Anton Paar MCR 301 rheometer (Anton Paar GmbH, Austria) in a plate–plate configuration, with a plate diameter of 25 mm and a gap of 1 mm, at a strain of 0.1%. Prior to testing, the virgin polymers and their composites were dried at 80 °C for 24 hours. Measurements were carried out over an angular frequency range of 0.1–500 rad/s, using a strain of 1%, which was within the linear viscoelastic (LVE) range, as confirmed by a preliminary amplitude sweep test.

RESULTS AND DISCUSSION

As a preliminary step, the effect of CoM modifier content on the specific density of the resulting composites was evaluated. The results are presented in Table 1.

Table 1. Results of the specific density of rPE and recycled PA6-30 GF, and the modified composites

Material	Density [g/cm^3]
rPA6-30GF	1.2990 ± 0.0090
rPE	0.9405 ± 0.0140
rPE/RPA6-30GF	0.9589 ± 0.0390
rPE/rPA6-30GF/1CoM	1.0033 ± 0.0170
rPE/rPA6-30GF/3CoM	1.1002 ± 0.0020

As it was shown in Table 1, the density of the rPE/rPA6-30 GF/3CoM composite increased compared to the unmodified rPE/rPA6-30GF composite. This is attributed to the improved packing of polyamide particles within the polyethylene matrix, resulting from better dispersion of the components facilitated by the compatibiliser. At a concentration of 3 wt.%, CoM enhanced adhesion between the rPA6 and rPE phases, reducing discontinuities and voids at the phase was confirmed by phase microstructure analysis using scanning electron microscopy (SEM). Figures 2 and 3 present the structure of the rPE/PA6-30GF composite before and after modification with the CoM compatibiliser.

As an example, Figure 2 presents microfractures observed typical on the impact-fractured surface of the rPE/rPA6 composite with glass fibre. Moreover, Figure 3 presents SEM images reveal that the rPE/rPA6-GF/CoM composite exhibits a compatible, heterogeneous structure, with the dispersed phase visible in the form of uniformly distributed glass fibre particles.

To evaluate the water absorption capacity of the samples, the rPE/rPA6-30GF composites modified with CoM were immersed in water for 24, 48, and 72 hours at 25 °C. The water absorption results for the rPE/rPA6-30GF/CoM composites at 25 °C are presented in Figure 5.

As it was shown in Figure 4, the recycled PA6-30GF samples exhibited the highest water absorption capacity compared to the unmodified polyethylene/polyamide 6 composites containing 30 wt.% glass fibre throughout the entire test duration. However, it can be observed that the water absorption capacity of the modified composites decreases with increasing PE-g-MAH content. The rate of weight gain in the composites diminished over time as the materials approached saturation. The rPE/rPA6-30GF composites containing 1–3 wt.% CoM showed a significantly reduced water absorption compared to neat PA6-30GF. This improvement is attributed to the increased homogeneity of the composite structure, which limits the interaction of strongly bound water molecules with the polar amide groups in polyamide (21). For materials, especially secondary (recycled) ones intended for use in products such as river buoys, which operate in direct contact with water, high resistance to water absorption is essential. The results thus indicate that

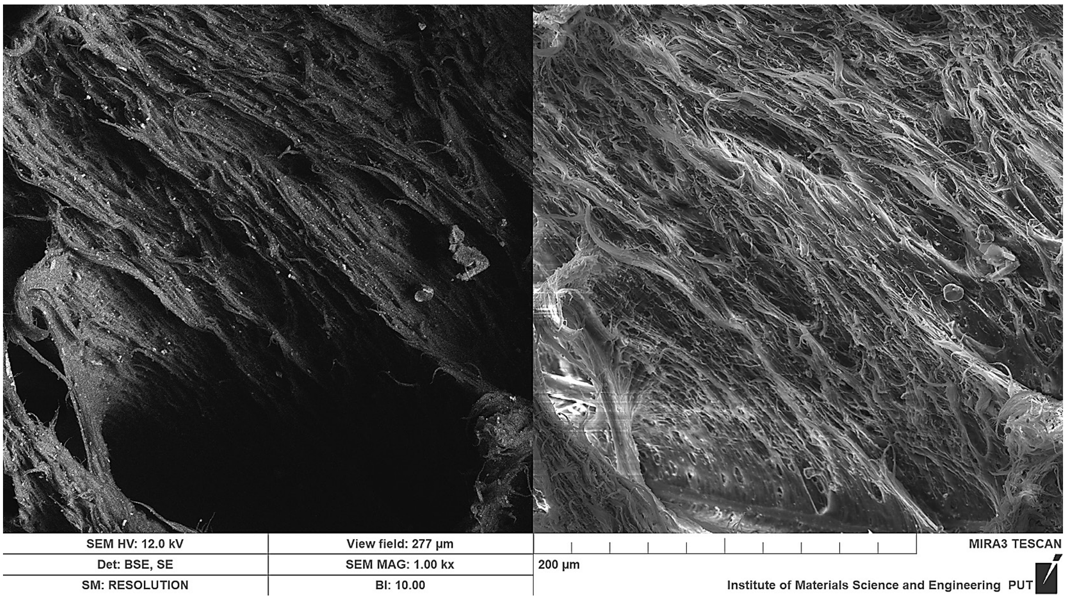

Figure 2. SEM BSE (left), SE (right) image of fractured surface of unmodified recycled rPE/PA6-30 GF composite sample

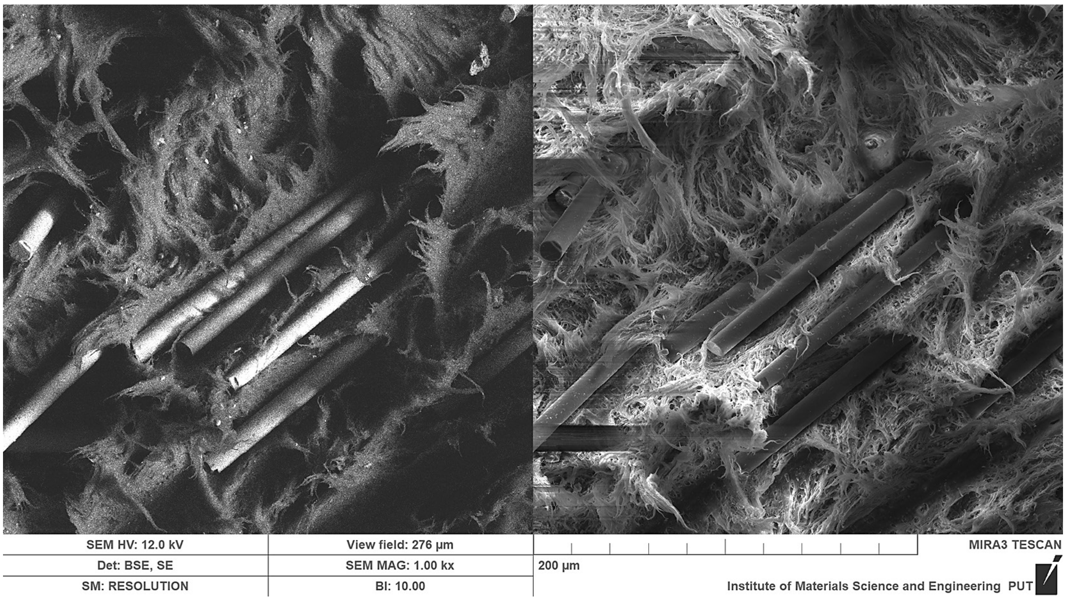

Figure 3. SEM BE (left) and BSE (right) images fractured surfaces of recycled rPE/PA6-30 GF composite sample modified with 3 wt.% CoM content

the developed rPE/rPA6-30GF/CoM composites meet this requirement, offering high dimensional stability along with adequate stiffness and mechanical strength.

The degree of dispersion of the components in the CoM-modified composites was assessed based on differential scanning calorimetry (DSC) analysis. The melting temperature, enthalpy, and degree of crystallinity of the polyethylene composites reinforced with polyamide 6 and glass fibre were compared with those of the unmodified composite. The DSC melting curves of the composites both before and after modification along with the reference PA6-30GF sample, are presented in Figure 5. A summary of the thermal analysis results obtained from the DSC tests is provided in Table 2.

T_M indicates the melting temperature, T_C the crystallisation temperature obtained from the maximum peak, and ΔH_M the melting enthalpy. An increasing amount of CoM led to a decrease in the crystallinity temperature of rPE /rPA6-30

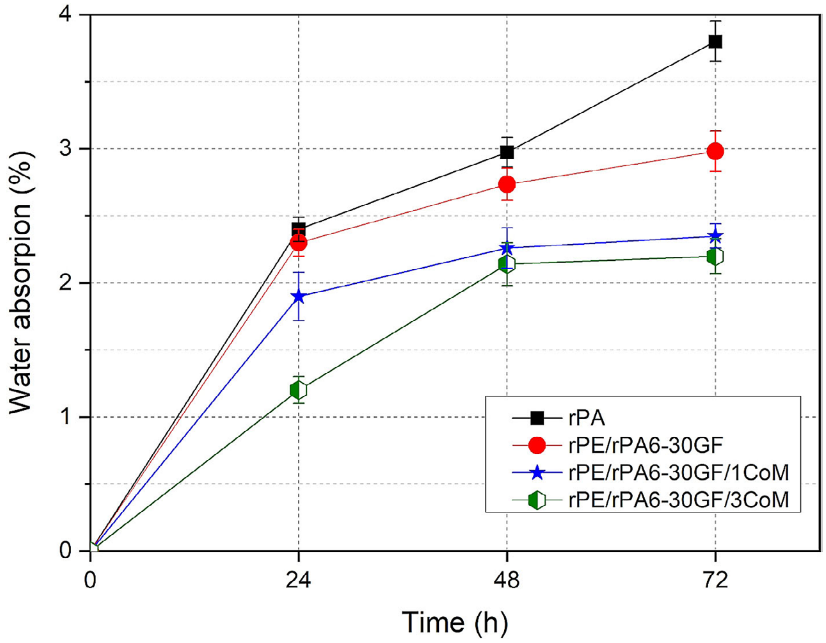

Figure 4. Water absorption at 25 °C of recycled rPA6-30 GF and rPE and their composite modified by different content PE-*g*-MAH (CoM)

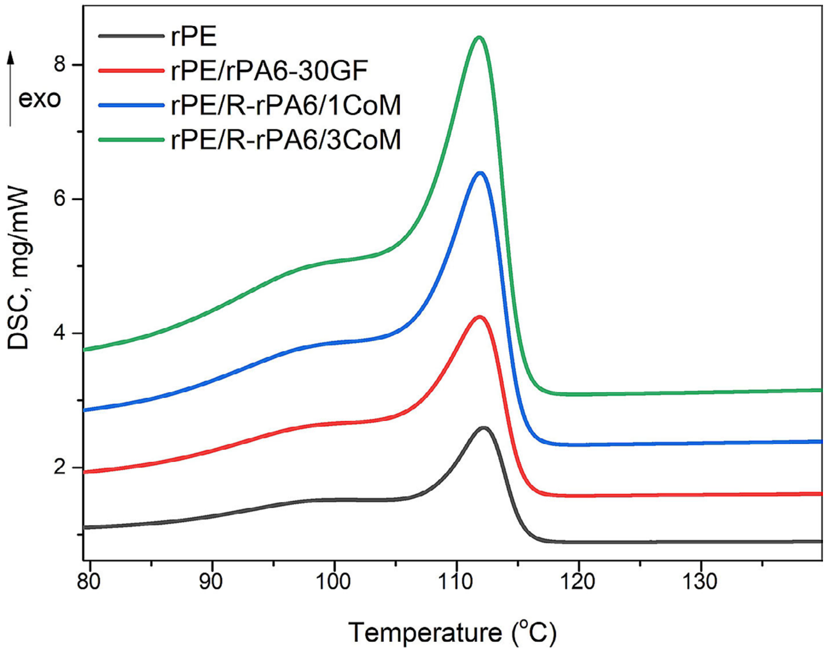

Figure 5. DSC melting curves of rPE, unmodified rPE/rPA6-30GF, and rPE/rPA6-30GF/3CoM

GF/CoM composites. The addition of CoM also resulted in significant changes in the crystallinity.

It can be seen that all composites have two melting peaks, T_{M1} and T_{M2} which are typical of two-phase polymer compositions (22, 23). The first peak in the composites is formed around the temperature equal to T_{m1} at 126 °C, and the second, $T_{m2,}$ around 224 °C, corresponds to the melting of polyamide 6 with 30 wt.% GF concerning recycled polyethylene. The presence of these two values indicates the inhomogeneous structure of the composites (21). The temperature of 184.6 °C corresponds to the crystallisation temperature of polyamide 6 with 30 wt. % GF, and the temperature of about 112 °C is the crystallisation of polyethylene. The results showed that the presence of two crystallisation peaks is also a confirmation of the heterogeneous structure of the waste

Table 2. DSC thermal properties of neat polymers and rPE/rPA6-30GF and rPE/rPA6-GF CoM composites (second heating at 10 °C/min)

Material	$T_{M1(PE)}$ [°C]	$\Delta H_{M2(PE)}$ [J/g]	$T_{M2(PA)}$ [°C]	$T_{C1(PE)}$ [°C]	$T_{C2(PA)}$ [°C]
rPA6-30GF	223.8	-29.8	-	-	184.6
rPE	125.6	-144.8	-	112.2	-
rPE/rPA6-30GF	127.0	-105.8	203.4	111.0	190.0
rPE/rPA6-30GF/1CoM	126.2	-110.7	218.9	112.0	189.6
rPE/rPA6-30GF/3CoM	128.0	-114.2	219.3	111.5	189.4

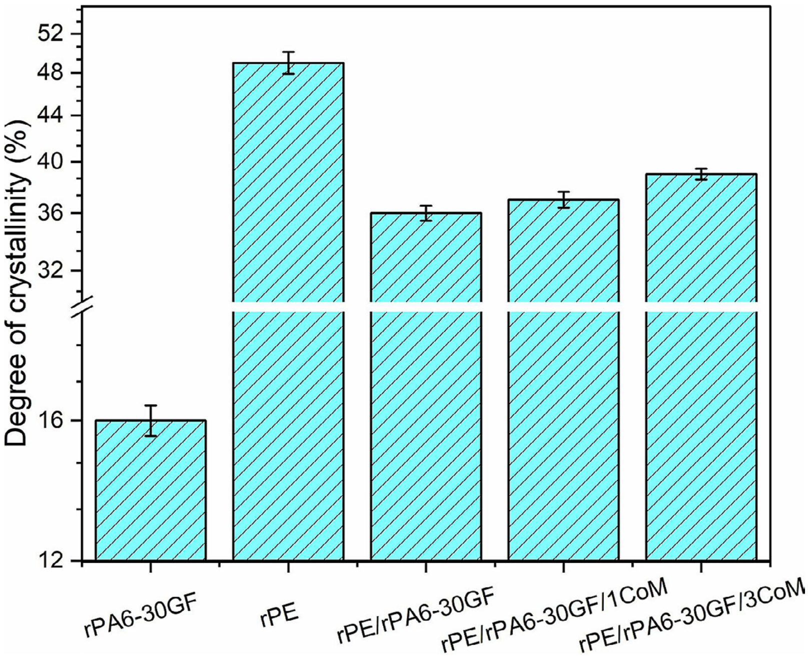

Figure 6. Compare the degree of crystallinity of unmodified rPE/rPA6-30 GF and composites modified by different content PE-g-MAH (CoM)

composites, including those with a compatibiliser. As it was shown in Figure 6, a clear increase in the degree of crystallisation of rPE/rPA6-30GF CoM blends can be seen. The increase in the heat of fusion implies the mobility of the rPA6-30GF CoM molecular chain, which is necessary for crystallisation, increasing the crystallinity of the polyethylene. Therefore, the addition of a compatibiliser (CoM) activates the mixing process of rPE/rPA6-30GF composites (14).

The viscoelastic properties of polyethylene/polyamide 6 with 30 wt.% glass fibre composites modified by CoM are characterised by viscoelastic functions, which include the complex modulus and the tangent of the phase angle (9). The tangent of the phase shift angle δ, also known as the tangent of the angle of mechanical loss, is used to analyse dynamic properties, and knowledge of the course of these changes allows the relationship between the molecular characteristics and mechanical properties of polymeric materials to be established (24). DMTA results of the storage modulus, G' (Figure 7) and the loss modulus, G" (Figure 8), for rPE/rPA6-30 GF at 230 °C and the composites modified by different content PE-g-MAH (CoM) were illustrated in Figures 7–8.

To evaluate the rheological behaviour primarily processability and structural properties of rPE and PA6-30GF with CoM in the melt state, a DMTA experiment was conducted. The results show changes in the storage modulus (G') and loss modulus (G") as a function of frequency. Figure 8 present the storage modulus (G') and loss modulus (G") of rPE, rPE/rPA6-30GF, and their composites with various CoM loadings as a function of frequency at 230 °C. Generally, compared to rPE samples, the rPE/rPA6-30GF composites exhibited increased values of G' and G". A higher

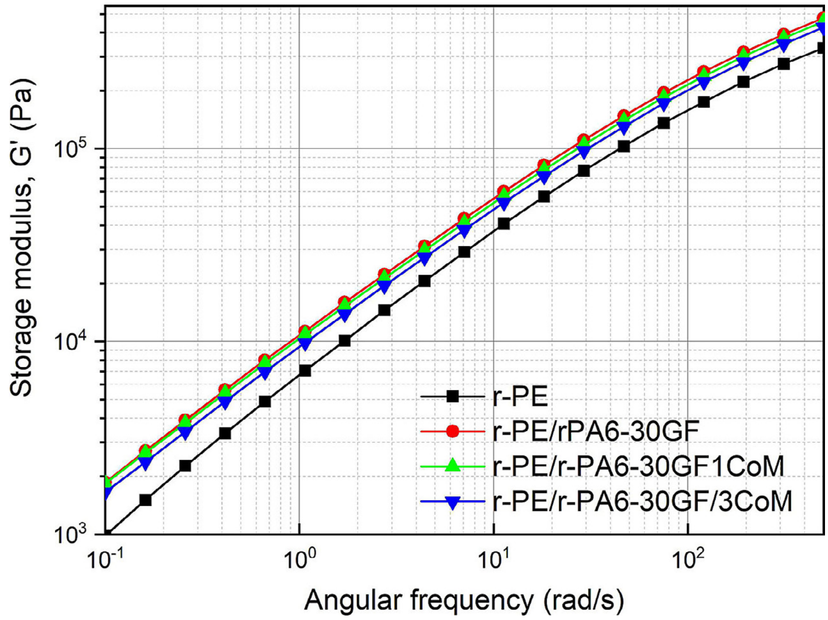

Figure 7. Storage modulus, G', for rPE/rPA6-30 GF at 230 °C and the composites modified by a different content PE-*g*-MAH (CoM)

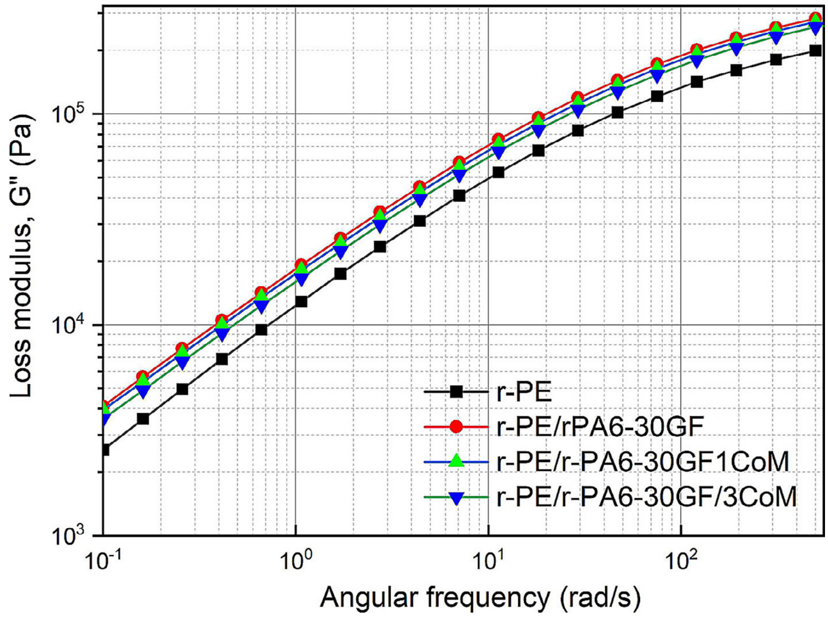

Figure 8. Loss modulus, G", for rPE/rPA6-30 GF at 230 °C and the composites modified by a different content PE-*g*-MAH (CoM)

G' indicates a more elastic structure, suggesting restricted polyethylene chain mobility due to the presence of CoM. A similar effect was observed by Bai in polyamide 12 modified with carbon nanotubes (25). Compared to pure polyethylene, the composites containing polyamide additives showed significantly higher G' and G" values at a strain rate of 0.1 rad/s. However, the composites modified with CoM exhibited a decrease in these moduli with increasing compatibiliser content. This behaviour indicates the compatibilising effect of maleic anhydride, which stabilises the heterogeneous structure at the micro-phase level. In addition, rPE/rPA6-30GF/3CoM composites exhibit significantly lower water absorption than rPA6-30GF, while essentially retaining the fundamental mechanical properties of pure polyamide, as demonstrated by the example of a composite containing 3 wt.% CoM in the rPE/rPA6-30GF system. The low moisture absorption contributes to improved resistance to stress corrosion cracking and enhanced dimensional stability of the products made from rPE/rPA6-30GF blends, which is particularly important for their service life. It is also worth noting that the mechanical properties of the rPE/rPA6-30GF/CoM blends are significantly higher compared to the corresponding unmodified PE/PA6-30GF blends.

CONCLUSIONS

In this study, the composites made from waste polyethylene reinforced with glass fibre and modified with grafted maleic anhydride (CoM), designed for river buoy applications, were successfully produced. The results demonstrated that the rPE/rPA-30GF/CoM composite exhibits a compatible heterogeneous structure, where the dispersed phase appears as uniformly distributed glass fibre particles. The mechanical, thermal, and water absorption properties of composites with varying compatibiliser content were investigated. It was observed that as the density of the produced materials increased, water absorption decreased. Additionally, the addition of the maleic anhydride compatibiliser improved the adhesion and dispersion of the waste polymers, as confirmed by the DSC analysis.

We can observe that water absorption increased with the addition of rPA6-30GF due to the polar groups in PA6, which have a high affinity for water, while rPE exhibits low water absorption. However, in the produced composites, water absorption decreased when both rPA6 and the compatibiliser were added, compared to pure polyamide. This reduction is attributed to the improved structural homogeneity resulting from the compatibiliser, which enhances adhesion and dispersion of the waste polymers. Maleic anhydride reduces the interaction between strongly bound water molecules in rPA and the polar amide groups. Therefore, the less polar the PA particles become, the lower the water absorption of the rPE/rPA6 composite. These results indicate that composites based on waste polyethylene and polyamide 6, combined with CoM compatibiliser, are suitable as construction materials for river buoys. For buoys made from rPE/rPA6-30GF/CoM blends, the production of rotationally moulded products with improved stiffness and water resistance is particularly advantageous.

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