



The influence of maleic anhydride-grafted polymers as compatibilizer on the properties of polypropylene and cyclic natural rubber blends

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Abstract

The enhancement of mechanical properties of immiscible blend, i.e. polypropylene/ cyclic natural rubber (PP/CNR), could be achieved by incorporating a compatibilizer into the blend system. The main challenge is how to compatibilize the two materials with distinct characteristic of saturated and apolar structures. As compatibilizers, copolymer of maleic anhydride-grafted polypropylene (POLYBOND 3002 and POLYBOND 3200) and maleic anhydride-grafted cyclic natural rubber (CNR-g-MA) were incorporated with PP/CNR (80/20 wt.%) blends in different weight ratios. The impact of those compatibilizers were analysed using Scanning Electron Microscope (SEM), which shows a homogenous phase of PP matrix indicating a homogenous dispersion of CNR into PP matrix. Thermogravimetry (TGA) analysis confirmed that the addition of compatibilizers improved the thermal stability of blends. The Differential Scanning Calorimetry (DSC) curve shows that the glass transition temperature (T_g) was significantly decreased due to the maleic anhydride-grafted polymers incorporation. The decrease of CNR's T_g value implies that the incorporation of maleic anhydride-grafted polymer into PP/CNR blends can improve blend compatibility. The compatibilization was further confirmed by an increase in tensile strength of compatibilized blends.

Keywords Polypropylene · Cyclic natural rubber · Maleic anhydride-grafted polymer · Compatibilizer · Blend

Highlights

1. The maleic anhydride-grafted polymers can enhance the compatibility of PP/CNR blend
2. The highest mechanical properties were shown by the addition of 1 wt.% of CNR-g-MA
3. Thermal stability was enhanced with the incorporation of maleic anhydride-grafted polymer

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Introduction

Polypropylene (PP) is a polyolefin material that is mostly utilized not only in plastic industry but also in automotive industry. This thermoplastic polymer has a linear structure and consists of repeated units of hydrogen and carbon atoms. PP is characterized by its versatility, and resistance to numerous chemical solvents either acids or alkaline, excellent barrier properties, relatively low-price/ low manufacturing cost, ease of formulation, and good flexibility [1, 2]. Despite its resistant nature, PP is still susceptible to oxidative stress which may influences its mechanical properties [3]. Blending process with other polymer materials is one method to improve the mechanical properties of PP blend, hence application of PP can be expanded through blending process with the incorporation of various materials.

In the previous work, we had blended PP and cyclic natural rubber (CNR) in the internal mixer and the morphological analysis resulted in two phase structure in which proved the immiscibility behaviour of the PP/CNR blends [4]. This fact

indicates, that there is a limitation in the interfacial adhesion in the PP/CNR blends that influences the mechanical properties of the blends. In addition, components which can improve excellent mechanical properties should be evenly distributed within both phases [5, 6].

Utilization of compatibilizer as an intermediate between two materials in the blend materials can enhance the compatibility properties of blends. In the immiscible blend, it can interact or even react with the blend components to improve the interfacial adhesion of the blend phases [2, 7–10]. In different studies, compatibilizer is effective to improve the compatibility of blend components through enhancement of interdiffusivity properties of each blend components [9, 11–13]. Compatibilizer which contains maleic anhydride (MA) was widely preferred to improve the dispersion of immiscible blend [14, 15]. In the previous study, Franco-Marques et al. [16] successfully improved the mechanical properties of polypropylene composite with recycled fiber by the addition of several commercial maleated polypropylene compatibilizers. The effectiveness of maleated polypropylene was also proved by Feng et al. [17], it can improve the compatibility between fiber and polypropylene as matrix. Other kinds of maleated polymers also can be used to improve the mechanical properties and compatibility of blend or composite, i.e. polyethylene [18, 19]. In the previous work of our group, maleated polymer based on cyclic natural rubber has been prepared, and its effect as compatibilizer will be evaluated in the present study.

In our perspective, there is still limited effort on the application of maleic anhydride-grafted polymer, i.e. PP-g-MA (POLYBOND 3002 and 3200) and CNR-g-MA as compatibilizers in the blend of PP/CNR. In this work, the effect of PP-g-MA and CNR-g-MA copolymers as compatibilizer in the PP/CNR blends were evaluated in terms of morphological, mechanical and thermal properties.

Materials and methods

Materials

Cyclic natural rubber ($\rho = 0.91$ g/mL, acid value ≤ 5 mg KOH/g) was obtained from PT. Industri Karet Nusantara, Deli Serdang – North Sumatera. Polypropylene EL-Pro™ P701J ($\rho = 0.91$ g/mL) was purchased from SCG Performance Chemicals, Co. LTD. POLYBOND 3002 ($\rho = 0.91$ g/mL, acid value = 2.8 mg/KOH g, MA content = 0.2 wt.%) and 3200 ($\rho = 0.91$ g/mL, acid value = 6.1 mg/KOH g, MA content = 1.0 wt.%) were purchased from CHEMTURA, USA. CNR-g-MA was obtained from the previous work [20].

Preparation of CNR-g-MA

The free-radical melt-grafting process of maleic anhydride into cyclic natural rubber was prepared in a 60-cc internal mixer (Brabender Plasticoder PLE 331 with a mixer head (W50H), Duisburg, Germany). The value of torque and chamber temperature of processes in internal mixer were recorded. The processing conditions were set at 150 °C with 8 min of mixing time and 80 rpm of rotor speed. The reaction of MA with CNR was conducted by loading CNR granules into the chamber. After 4 min, the CNR was molten and 0.85 wt.% of benzoyl peroxide was added for another 1 min. Finally, 13.68 wt.% of MA was added into the chamber and the mixing process was stopped until the torque value showed the constant value.

Preparation of blends

The blends of PP and the maleic anhydride-grafted polymer were prepared using a Brabender® internal mixer at 50 rpm, with maximum loading capacity of 35 g. The processing condition was set at 170 °C with 10 min of blending time. The blending of CNR and PP was carried out by loading PP pellets into the chamber (Table 1). After 4 min, the PP was molten, then CNR and compatibilizer granules were added for another 3 min. The maleic anhydride-grafted polymer was then added into the chamber and the blending process was stopped when the torque value showed a constant value.

Characterization method

Mechanical testing

The dumbbell-shaped specimens for mechanical testing were prepared by compression moulding using hot press at 190 °C for 10 min. The mechanical properties of the PP/CNR blends were measured according to ASTM D638–08 and the measurement of tensile strength, Young's modulus, and elongation at break was conducted using a universal testing machine DTC-10 supplied by IDMtest (San Sebastián, Spain) equipped with a 5-kN load cell at speed 10 mm/min. The Young's modulus was measured using the same machine and equipped with MF, MFA 2 extensometer (Velbert, Germany).

Attenuated Total reflectance – Fourier transform infra-red (ATR-FTIR) analysis

Film specimens of blended material of PP, CNR and the maleic anhydride-grafted polymer were prepared by compression moulding using hot press at 190 °C for 5 min to be analysed by ATR-FTIR. The surface composition of the film specimen was conducted on ALPHA ATR (Bruker). The measurement

was performed in the wavenumber range between 4000 and 500 cm⁻¹ with 32 running scans at a resolution of 4 cm⁻¹.

Morphology study

The microscale study of fractured and cracked surfaces was observed using *Scanning Electron Microscope (SEM)* HITACHI *TM4000*. The samples with 1 mm × 1 mm were coated with gold to ensure the good conductivity for the analysis.

Wettability

The wettability was obtained by measuring the contact angle of the surface of PP/CNR blends using the sessile drop method with a KRUSS DSA25B analyser (Kruss GmbH-Germany). All measurements were performed with drops of distilled water after about 10 s. The reported values were the average of eight measurements at different places on the same film specimens of blended materials. The contact angles were measured by video, with the contact angle >90° and <90° are called as non-wetting and wetting, respectively. The obtained contact angle can be used for calculating the surface tension of blend surface according to Wenzel equation [21], as:

$$r(\sigma_S - \sigma_{SL}) = \sigma_L \cos\theta \tag{1}$$

Where, σ_S : surface energy of solid, σ_L : surface energy of liquid, σ_{SL} : surface energy between solid and liquid.

Thermogravimetric analysis

The thermal stability of samples was examined by thermogravimetric analysis (TGA) using DTG-60 Shimadzu with the temperature ranging from 30 to 600 °C at a heating rate of 10 °C/min. Heating of samples was carried out under nitrogen atmosphere to prevent any thermoxidative degradation.

Differential scanning calorimetry analysis

A TA instrument DSC 2960 SDT was used to evaluate the thermal properties of blends under nitrogen flow. The samples (5–10 mg) were sealed in DSC sample pan. Samples were heated from 20 to 200 °C at a heating rate of 10 °C/min.

Results and discussions

The influence of the maleic anhydride-grafted polymer, CNR-g-MA, POLYBOND 3002 and POLYBOND 3200 on the properties of PP/CNR blends was evaluated. The tensile strength, Young’s modulus and elongation at break of the PP/CNR blend that compatibilized with CNR-g-MA,

POLYBOND 3002 and POLYBOND 3200 are shown in Fig. 1 and Table 2.

For PP/CNR blends, the addition of 1 wt% of maleic anhydride-grafted polymer increased the mechanical properties (tensile strength and elongation at break), however the addition of higher concentration of compatibilizer (2 and 3 wt%) led to a significant decrease in comparison to neat blend of PP/CNR. The addition of the maleic anhydride-grafted polymer significantly modified the fracture behaviour that was observed from a significant increase of mechanical properties. This result can be associated to the better interaction of PP and CNR in the presence of maleic anhydride-grafted polymer [22]. Similar behaviour was also observed and reported by Salleh et al. [23] and Gabriel et al. [13] where the incorporation of maleic anhydride-grafted polymer improved the adhesion interface and mechanical properties of PP/CNR blends.

The decrease of mechanical properties during incorporation of compatibilizer has been explained by Tjong et al. [24], Rek et al. [25] and Dobrovsky [26] with following explanations, the ductility of the blends will increase with the increase of blends homogeneity, thus it will lead to require a lower stress of value in order to make crack on the particle blends. The other perspective of the decreased of blend’s mechanical properties was expected by lubricant effect of compatibilizer which contains maleic anhydride [27], in this case CNR-g-MA, POLYBOND 3002 and 3200. Other assumption of this phenomenon is the chain scission effect to mechanical properties, which occurred during melt reactive processing polymer chain with the peroxy-initiated, PP and CNR [28–30].

Among compatibilizers, the cyclized natural rubber grafted maleic anhydride (CNR-g-MA) was the most effective one to compatibilize the blends than POLYBOND 3002 and 3200. This can be explained as a result of the reduction of the surface tension in melted phase of two materials during processing

Table 1 Composition of PP/CNR Blends

Sample code	PP (wt.%)	CNR (wt.%)	Compatibilizer (wt.%)		
			MA	3002	3200
PP/CNR	80.00	20.00	0.00	0.00	0.00
PP/CNR/MA1.0	79.20	19.80	1.00	–	–
PP/CNR/MA2.0	78.40	19.60	2.00	–	–
PP/CNR/MA3.0	77.60	19.40	3.00	–	–
PP/CNR/3002–1	79.20	19.80	–	1.00	–
PP/CNR/3002–2	78.40	19.60	–	2.00	–
PP/CNR/3002–3	77.60	19.40	–	3.00	–
PP/CNR/3200–1	79.20	19.80	–	–	1.00
PP/CNR/3200–2	78.40	19.60	–	–	2.00
PP/CNR/3200–3	77.60	19.40	–	–	3.00

[31]. CNR-g-MA in this blends system does not only act as compatibilizer, but also as a reinforcement agent. The presence of CNR-g-MA reduces the brittleness of CNR and act as bridge between PP and CNR in the blend system, with some CNR fractions being adhered to the surface of PP. Also, the molecular weight of POLYBOND 3002 (based on the MFI data, Table 3.) gave a significant effect in the reduction value of tensile properties of PP/CNR blends. Franco-Marques et al. [16] explained this phenomenon is attributed to the poor dispersion of POLYBOND 3002 into the blends system, which has lower MFI than PP, CNR, POLYBOND 3200 and CNR-g-MA. The high molecular weight of POLYBOND 3002 will render its complete dispersion therefore a prolonged time is needed. The presence of POLYBOND 3002 in the blends system was confirmed by SEM images where white phase assumed as POLYBOND was found on the surface of blends (Fig. 2d).

The morphological fracture surface of compatibilized PP/CNR in the presence of maleic anhydride-grafted polymer is shown in Fig. 2. Blend of incompatible PP/CNR (Fig. 2a) showed separated phases between PP and CNR, indicating a poor interfacial adhesion of CNR in the PP matrix [32]. The presence of maleic anhydride-compatibilizer significantly changed the morphological phase of PP/CNR blends. Based on the morphological images, it is possible to observe the maleic anhydride-grafted polymer's effect that gives a contribution for a better dispersion of CNR in the PP matrix. The previous work by Gabriel et al. [13] and Das et al. [33] reported that the incorporation of LLDPE-g-MA and PE-g-MA successfully reduced the presence of the second phase on the PA6/LLDPE blends. The result was attributed to the presence of copolymer, in this case maleic anhydride-grafted polymer, on the interface regions, which improved the dispersion of CNR into PP matrix. In the report by Jurkowisk et al. [32],

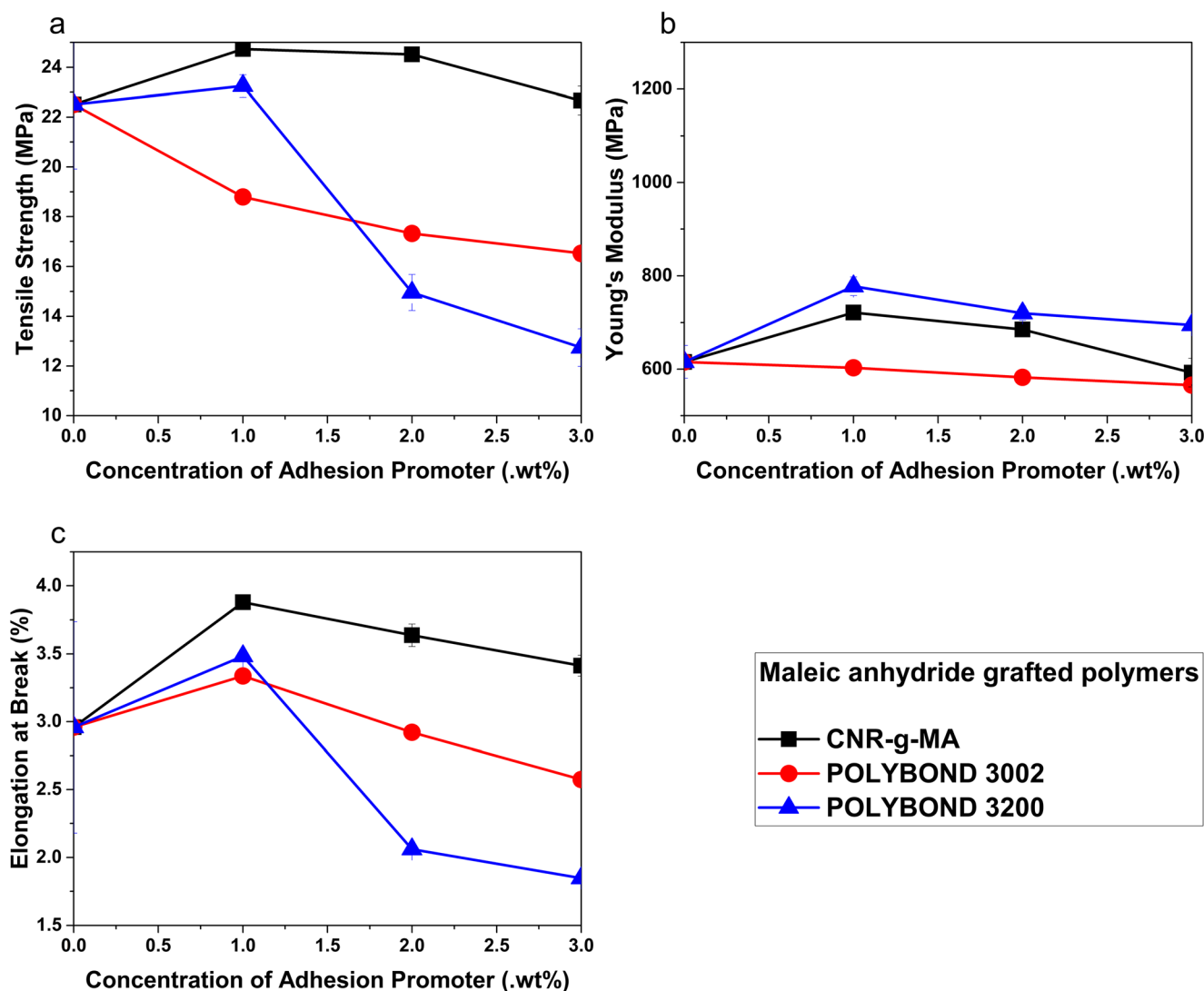


Fig. 1 The plot of maleic anhydride-grafted polymer concentration vs tensile strength (a), maleic anhydride-grafted polymer concentration vs Young's modulus (b), and maleic anhydride-grafted polymer concentration vs elongation at break (c)

Table 2 Mechanical properties of PP/CNR blends

Sample	Tensile strength (MPa)	Young's modulus (MPa)	Elongation at break (%)
PP/CNR	22.51 ± 2.60	615.40 ± 34.99	2.96 ± 0.78
PP/CNR/MA1.0	24.73 ± 0.15	721.39 ± 12.19	3.88 ± 0.02
PP/CNR/MA2.0	24.51 ± 0.20	684.96 ± 11.90	3.63 ± 0.08
PP/CNR/MA3.0	22.66 ± 0.58	592.14 ± 30.71	3.41 ± 0.07
PP/CNR/3002-1	18.79 ± 0.02	602.95 ± 6.56	3.33 ± 0.01
PP/CNR/3002-2	17.33 ± 0.02	581.97 ± 0.45	2.92 ± 0.00
PP/CNR/3002-3	16.53 ± 0.10	565.68 ± 2.41	2.57 ± 0.04
PP/CNR/3200-1	23.25 ± 0.45	777.81 ± 20.05	3.48 ± 0.02
PP/CNR/3200-2	14.95 ± 0.72	719.90 ± 15.36	2.06 ± 0.01
PP/CNR/3200-3	12.73 ± 0.76	694.70 ± 2.14	1.84 ± 0.01

the improvement of PA6/LDPE blend dispersion is associated with the mechanical compatibilization in the presence of LDPE-g-MA.

The infrared spectra of PP/CNR blends is shown in Fig. 3. All spectra were dominated with the spectrum from PP because the film that measured in ATR was composed with ~80 wt.% of PP. In specific to PP/CNR blend, three main regions can be observed, such as (i) the maleic anhydride group, (ii) the specific band of CNR and (iii) the common band of hydrocarbon.

The small band of maleic anhydride group can be seen at 1740 cm^{-1} as a consequence of its low concentration in the blend. That band was corresponding to the vibration of anhydride group. The specific band of CNR can be observed at $740\text{--}840\text{ cm}^{-1}$, which assigned to the cyclic carbon's vibration of CNR. In the different article it can be attributed to the C-H bending's vibration of the polyisoprene trialkyl substitution [34–36]. The last region of the hydrocarbon's common band can be seen at 1470 cm^{-1} , which generally can be found in aliphatic and cyclic hydrocarbon, including PP and CNR in this case. The band corresponds to the CH_2 of methyl and methylene group's vibration. The IR spectra of PP/CNR blends with compatibilizer of CNR-g-MA, POLYBOND 3002 and 3200 confirmed that there is no chemical interaction between the maleic group of compatibilizer to the main chain of PP or CNR. The interaction that occurred in the blends system of PP/CNR is only physical interaction between CNR with the compatibilizer, also with PP. Although no chemical interaction or compatibilization occurs it does not always show that the as-prepared blend in this study is not compatible. As reported by Jurkowski et al. [32] and Gabriel et al. [13], compatibilized blend can be achieved not only by chemical interaction or compatibilization, but also by mechanical compatibilization. The presence of chemical compatibilization in the surface of blend is difficult to determine based only from the FT-IR spectra, due to the low concentration of the covalent bonding that formed between the compatibilizer and matrix [37]. Still, it is possible to see the compatibilization effect of maleic anhydride-

grafted polymer in PP/CNR blend through the improvement in the blend's mechanical properties.

There are direct and indirect methods to explore and measure the adhesion strength of the material. This work only shows the indirect measurement to measure the adhesion strength using optical contact angle. Optical contact angle measurement can give a good estimation of the mobility and polarity of the polymer chains in the outer layer of the hydrophobic surface. The wettability of PP/CNR blends with and without maleic anhydride-grafted polymer are shown in Fig. 4.

The compatibilizer, CNR-g-MA, POLYBOND 3002 and 3200, are amphiphilic possessing both hydrophobic and hydrophilic components. The hydrophobic component interacts with the hydrophobic surface, in this case, C-H group of PP while the hydrophilic component interacts with the hydrophilic coat, in this case C=C that can be found in the structure of CNR. With those properties of the maleic anhydride-grafted polymer, the smallest contact angle was obtained. Figure 4 shows optical contact angle and surface tension data of PP/CNR blends, the data showed PP/CNR blends with 1% compatibilizer of POLYBOND 3200 has the lowest value of water contact angle. The correlation of optical contact angle to the adhesion strength was described as surface tension. The smaller contact angles indicate that the interaction of the coating to the hydrophobic surface has a high energy surface, which means that the surface is more wetting and have greater adhesion. The better interaction of POLYBOND 3200 as compatibilizer than POLYBOND 3002 and CNR-g-MA can

Table 3 MFI data of material at $190\text{ }^\circ\text{C}$ (2.16 kg_f)

Material	MFI (g/10 min)
PP	8.95 ± 0.29
POLYBOND 3002	6.02 ± 0.69
POLYBOND 3200	122.05 ± 1.5
CNR	36.37 ± 0.80
CNR-g-MA	73.17 ± 0.99

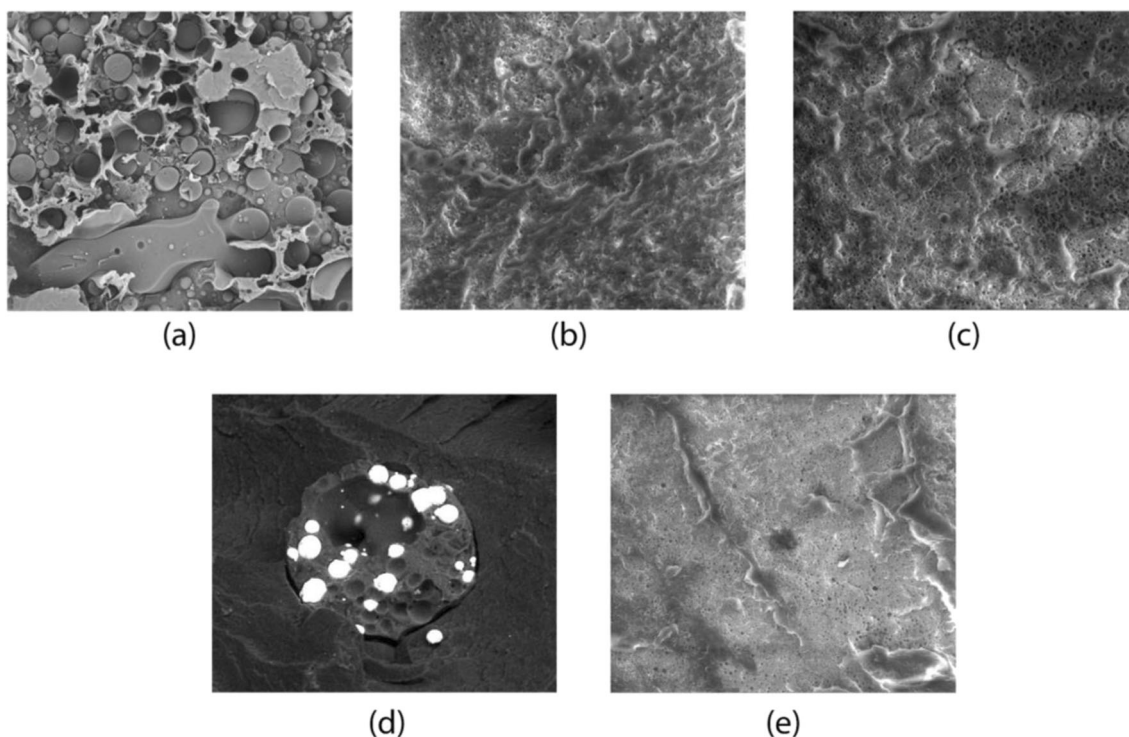


Fig. 2 Microphotograph SEM of PP/CNR (80/20) (a), in the presence MA 1% (b) POLYBOND 3002 1% (c-d), and POLYBOND 3200 1% (e)

be influenced by the similar structure of POLYBOND 3200's backbone with the polypropylene surface and the lower value of molecular weight. That factor may enhance the probability of entanglement process of adhesive mechanism on polypropylene surface. That statement was based on the fact that the surface of blend film was dominated by PP backbone from the

formation of PP with POLYBOND 3200. The dominant presence of PP backbone was caused by the repulsion of PP crystal. PP is categorized as a (semi) crystalline polymer and as consequence the crystal and amorphous region was formed when the film cooled. As the impact of the PP backbone's domination in the surface of blend film, it is obvious that some

Fig. 3 ATR-FTIR spectra of PP/CNR blends

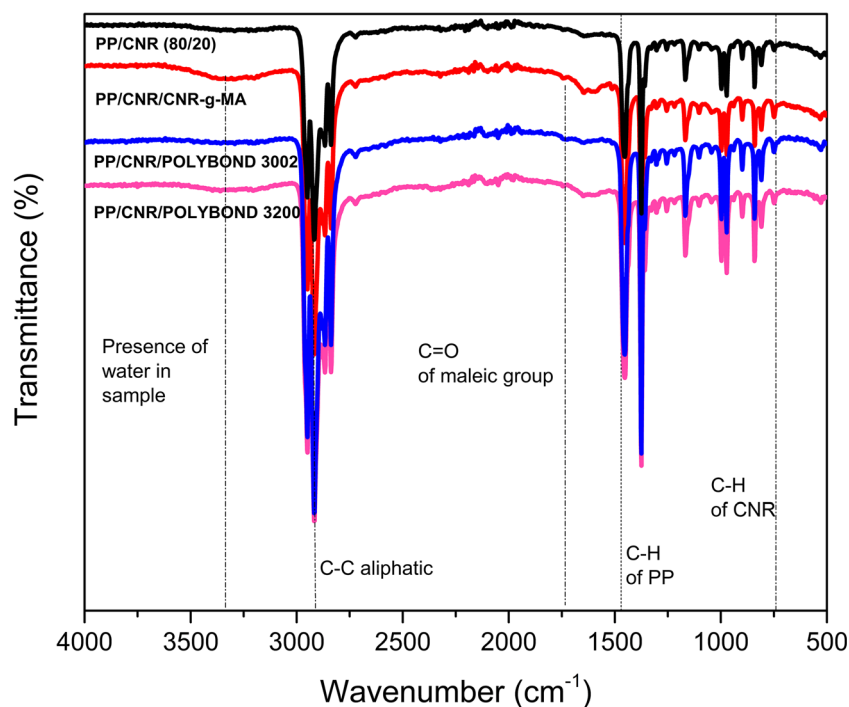
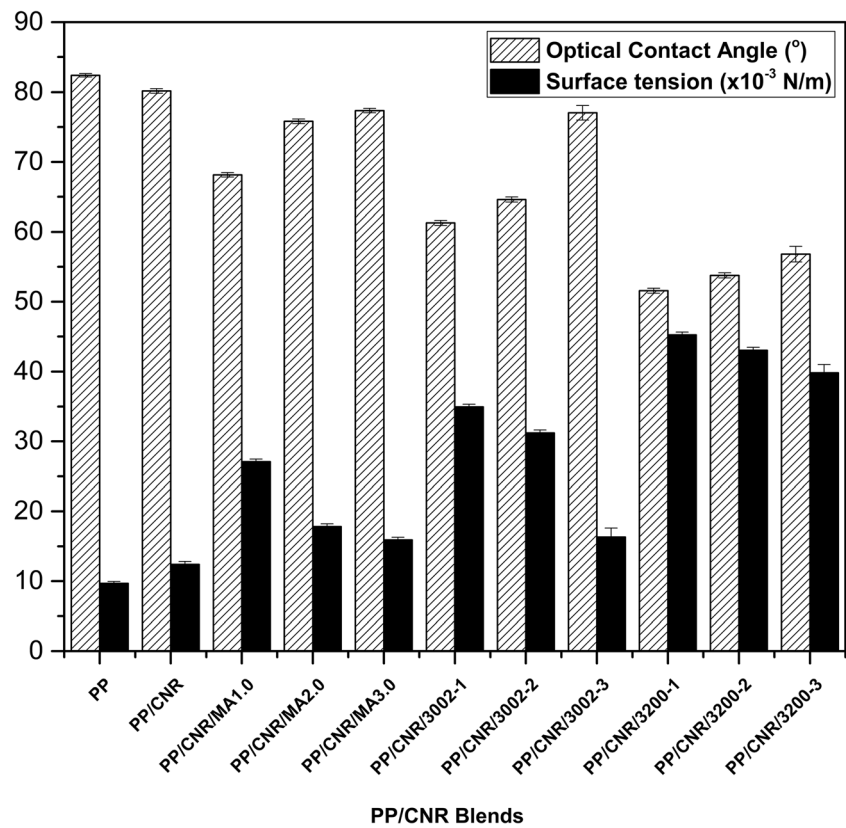


Fig. 4 Influence of maleic anhydride-grafted polymer on optical contact angle and surface tension of PP/CNR blends



POLYBOND 3200 parts are located on the surface of blend film. This phenomenon may not work for the CNR-g-MA because it has different polymer backbone. The indirect effect of that phenomenon is the content of anhydride group of maleic acid on the surface is less than the blend film with POLYBOND 3002 and 3200, and it influences the way of the wetting process on the film surface, hence increasing the number of the anhydride group on the surface may also increase the chance of reacting with the polar component (e.g. water).

The thermal properties of PP/CNR blends are shown in Table 4 and Fig. 5. Table 4 shows the decomposition temperature at 1 and 95% of weight that has been lost ($T_{1\%}$ and $T_{95\%}$). Those data are obtained from Fig. 5 (some figures are not shown).

The TG curves of PP/CNR blends show a single-stage decomposition (Fig. 5). All samples prepared in this work started to decompose at different initial temperature ($T_{1\%}$) with no obvious patterns to explain the influence of the maleic anhydride-grafted polymer concentration. The lowest value of $T_{1\%}$ is shown by PP/CNR blends with the presence of MA of 2% at 294 °C. These results revealed that the initial decomposition of all PP/CNR blends in the presence of maleic anhydride-grafted polymer occurred at higher temperature than the neat PP (254 °C) and CNR (249 °C). This result indicates that the addition of maleic anhydride-grafted polymer can increase the thermal stability of PP/CNR blends.

Figure 5 also shows the derivative of TG curve (DTG) for the uncompatibilized PP/CNR with the additional of 1 wt% maleic anhydride-grafted polymers. These curves indicated the presence of one decomposition peak, and supported with $T_{1\%}$ value, the addition of compatibilizer improve the thermal stability. The addition of CNR-g-MA, POLYBOND 3002, and POLYBOND 3200 increased the onset temperature of blends. The onset temperature of uncompatibilized PP/CNR blend is 366 °C, but after the addition of 1 wt% of CNR-g-MA, POLYBOND 3002, and POLYBOND 3200, the onset

Table 4 $T_{1\%}$ and $T_{95\%}$ of PP, CNR and blends

Sample	$T_{1\%}$ (°C)	T_{max} (°C)	$T_{95\%}$ (°C)
PP	254	459	476
CNR	249	430	469
PP/CNR	206	437	478
PP/CNR/MA1.0	365	458	471
PP/CNR/MA2.0	294	457	472
PP/CNR/MA3.0	364	459	472
PP/CNR/3002-1	381	455	473
PP/CNR/3002-2	332	458	473
PP/CNR/3002-3	368	458	478
PP/CNR/3200-1	373	457	472
PP/CNR/3200-2	347	456	472
PP/CNR/3200-3	344	457	475

temperature of PP/CNR blends is 391, 402, and 376 °C. The increase of onset temperature of the compatibilized PP/CNR blends can be attributed to the improvement of the interfacial adhesion between both phases, PP and CNR. The presence of maleic anhydride-grafted polymer can enhance the compatibility of PP/CNR blends through the physical interaction between anhydride group from maleic anhydride and saturated/unsaturated of C-H from PP and CNR. The maleic anhydride-grafted compatibilizer is located at interface of blend and it is able to reduce the interfacial tension. As the result of this phenomenon, the interfacial between both phases is strong and the direct effect correspond to the improvement of thermal stability.

The glass transition temperature (T_g) measured by a differential scanning calorimetry for the blends is given in Table 5. The T_g of PP/CNR blend in the presence of maleic anhydride-grafted polymers led to significant changes compared to PP/CNR blend. The T_g of neat CNR is 95 °C, but after the blending process with PP the T_g value of CNR decrease to 89.11 °C. The T_g value of CNR in the blends after the addition of maleic anhydride-grafted polymers decrease to 65–67 °C. This suggests that maleic anhydride-grafted polymers acted like a

Table 5 $T_{1\%}$ and $T_{95\%}$ of PP, CNR and blends

Sample	PP/CNR blend			
	–	CNR-g-MA	POLYBOND 3002	POLYBOND 3200
T_g (°C)	89.11	65.90	66.98	67.65

compatibilizer between PP and CNR. There have been many reports which indicate the addition of maleic anhydride-grafted polymers into immiscible blends are able to enhance the blends compatibility [17, 27, 38, 39]. There are some evidences that could be used to identify the enhancing of blends compatibility, i.e. a decrease in domain size, a shift of T_g to backward direction and a single T_g in some blends [40, 41].

Conclusions

The presence of maleic anhydride-grafted polymer on the blend of PP/CNR showed a positive influence on the tensile, morphology, wettability and thermal properties of blends. The

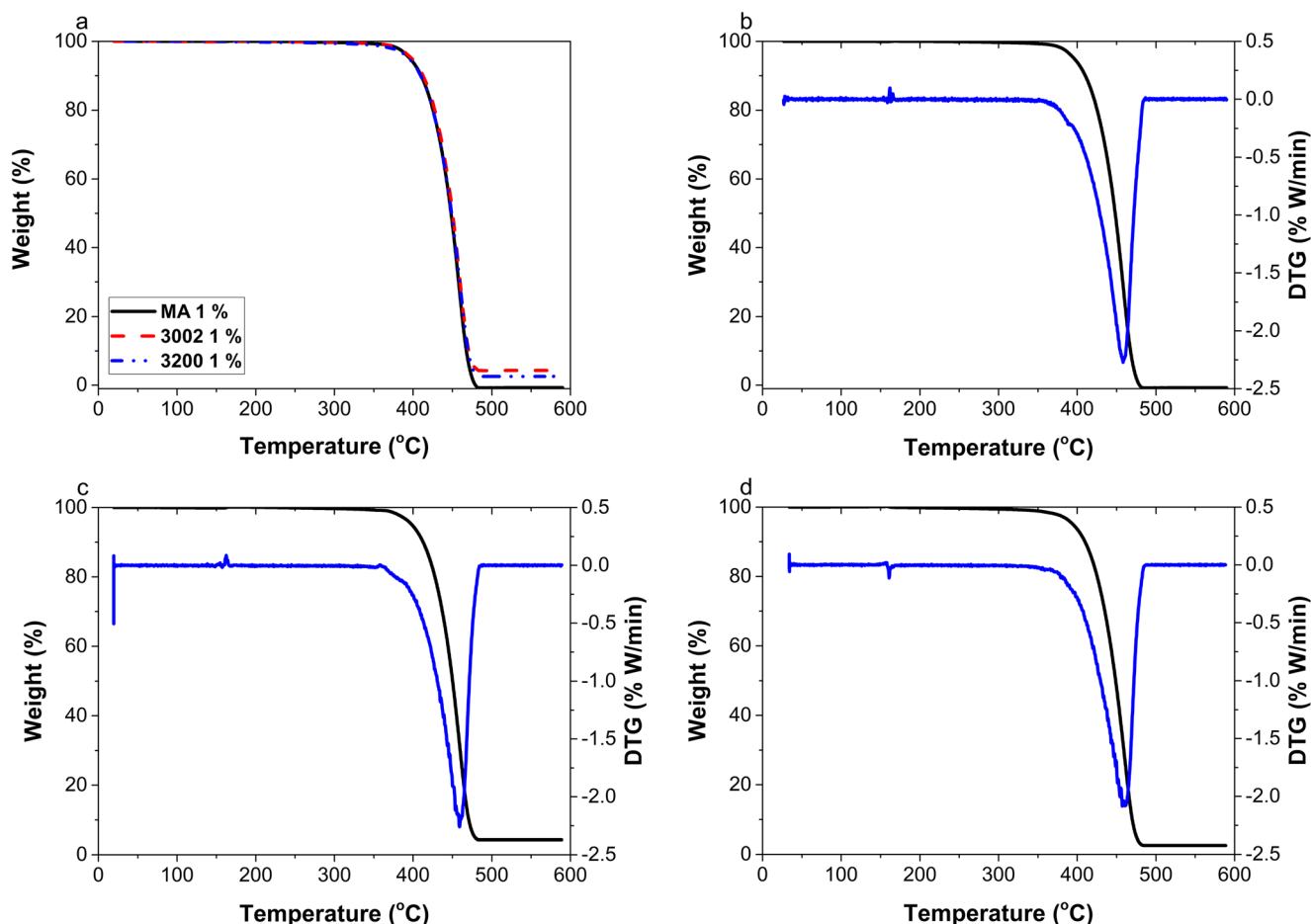


Fig. 5 a Thermogravimetric analysis of PP/CNR blends, TGA and DTG curves of PP/CNR blends in the presence b CNR-g-MA, c POLYBOND 3002, and d POLYBOND 3200

tensile properties showed a positive improvement in the addition of maleic anhydride-grafted polymer, especially in the presence of 1% MA. The fracture surface morphology also showed an improvement compared to the PP/CNR blend without maleic anhydride-grafted polymer. Increasing the content of compatibilizer, maleic anhydride-grafted polymer, resulted in a finer surface. The wettability is one of the important parameters in this research to show the hydrophilicity of the blend surface. In this study, we reported a unique wettability property of blends. The hydrophilicity of PP/CNR blends decrease by the increasing concentration of maleic anhydride-grafted polymer incorporated into the blends. The presence of maleic anhydride-grafted polymer enhanced the onset temperature of uncompatibilized PP/CNR blends and improved the thermal stability of blends. The maleic anhydride-grafted polymer acted as a bridge between the immiscible blend components, this can be seen from the T_g of blends that shifted backward with the incorporation of the maleic anhydride-grafted polymers.

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