

# Enhanced Toughening of Poly(propylene) with Reclaimed-Tire Rubber

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**ABSTRACT:** The dynamic vulcanization of reclaimed-tire rubber (RTR) and homopolypropylene (PP) was performed by melt-mixing using either a sulfur crosslinking agent, maleic anhydride (MA), dicumyl peroxide (DCP), or the combination of MA and DCP, in two consecutive machines, first a two-roll mill and then a counterrotating twin-screw extruder. In the case of applying a sulfur crosslinking agent, it was demonstrated that the RTR/PP blend at the ratio of 30/70 had the highest impact strength. This could be attributed to the limitation of carbon black in the blend. When the combination of MA and DCP was applied, the result was higher impact strength of the blend at the same ratio. This could be attributed to not only the cohesion between the

polymer chains in each phase, PP phase and rubber phase, but also the interfacial adhesion between PP and RTR chains in these two phases. For comparison, the GRT/PP blends with and without sulfur crosslinking agent were prepared as well. All these blends showed low impact strength, which was nearly the same as that of PP. The effects of different crosslinking agents on dispersion and distribution of rubber domain size, viscosity, and percentage crystallinity were also studied. © 2003 Wiley Periodicals, Inc. *J Appl Polym Sci* 91: 510–515, 2004

**Key words:** tire rubber; recycling; poly(propylene) (PP); toughness; vulcanization

## INTRODUCTION

The waste generated from rubber tires is becoming a major environmental problem throughout the world. This problem is attributed to the huge volume of used tires generated each year and the fact that tires are thermoset. In addition, tires are virtually resistant to biological degradation.<sup>1</sup> Several approaches have been proposed to deal with the problem of used tires such as converting to tire-derived fuel in solid-fuel burners, using pyrolysis to recover valuable chemical components,<sup>2</sup> incorporation in various nonrubber tire applications, and their use as fillers/tougheners in plastics.<sup>3</sup>

Generally, used tires have been recycled by cutting into small pieces, called ground rubber tire (GRT), for blending with different thermoplastics. However, this process usually causes a significant deterioration of the mechanical properties of the blends even though a compatibilizer has been added. It has been shown, however, that the compatibilizer played an important role in improving the impact strength of the blends, given that higher impact strength—although still lower than that of the original thermoplastics—could be obtained when the compatibilizer was added into the system.<sup>4–7</sup>

In addition, much attention has recently been focused on toughening thermoplastics using ethylene–vinyl acetate copolymer (EVA),<sup>8,9</sup> ethylene–propylene rubber (EPR),<sup>10,11</sup> natural rubber (NR),<sup>12</sup> and other rubber materials.<sup>13–23</sup> Among thermoplastics, poly(propylene) (PP) has a most remarkable combination of various physical properties and processability but poor impact strength, particularly at low temperature, because of its high glass-transition temperature and high crystallinity. Therefore, blending PP with rubber has been reported by several research groups<sup>24</sup> to improve its properties and extend its applications. However, no report on blending of PP with reclaimed-tire rubber (RTR), the other form of recycled-tire rubber, has been published. Practically, RTR has been used commercially for cost savings by blending with virgin natural rubber. This study thus focuses on the toughening of PP with RTR compared with GRT. Because of the chemically dissimilar structure of rubber and PP, phase separation is generally greater than the optimum, and interfacial bonding is poor in their physical blends. Therefore, dynamic crosslinking using a sulfur crosslinking agent, maleic anhydride, and dicumyl peroxide has been introduced to the system to form an interpolymer.

## EXPERIMENTAL

Poly(propylene) (PP; Profax 6531, MFI = 4 g/10 min) was supplied by HMC Polymers Co., Ltd. (Bangkok,

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**TABLE I**  
PP/GRT Blends Using Sulfur Crosslinking Agent<sup>a</sup>

Blend <sup>b</sup>	PP (pbw)	Ground rubber tire (pbw)		
		GRT40	GRT16	GRT8
S1, S2	70	30	—	—
S3, S4	70	—	30	—
S5, S6	70	—	—	30

<sup>a</sup> TMTD (1.5 phr), MBT (0.75 phr), ZnO (5 phr), stearic acid (2 phr), and sulfur (3 phr).

<sup>b</sup> Odd and even blend numbers refer to the unvulcanized and vulcanized blends, respectively.

Thailand). Ground rubber tire (GRT) and reclaimed-tire rubber (RTR) were supplied by Union Commercial Development Co., Ltd. (Bangkok, Thailand). The crosslinking agents, either 2-mercaptobenzothiazole (MBT), tetramethyl thiuram disulfide (TMTD), sulfur, zinc oxide (ZnO), and stearic acid or maleic anhydride (MA) and dicumyl peroxide (DCP), were all reagent grade and used without further purification. Ultranox 626 (supplied by Nagase Thailand, Bangkok) was added as an antioxidant for blends.

GRT and RTR obtained from the same lot of used tires were used in this research. Whereas GRT contains 5 wt % of sulfur, only 0.01 wt % of sulfur was present in RTR. In the case of GRT, three different mesh sizes were studied: 8 mesh ( $\cong$  2.38 mm: GRT8), 16 mesh ( $\cong$  1.19 mm: GRT16), and 40 mesh ( $\cong$  0.42 mm: GRT40). The recycled-tire rubber, either GRT or RTR, was melt blended with homopolypropylene (PP) at various ratios, in the presence of either sulfur crosslinking agent, TMTD, MBT, ZnO, and sulfur or MA/DCP, according to formulations given in Tables I–III. Each mixture was first melt-mixed in a two-roll mill at 170°C (front roll) and 165°C (back roll). The composites were normally cut diagonally from time to time and folded over several times during mixing. After 10 min of mixing, one type of vulcanizing agents and ultranox 626 were added and were allowed to mix for another 5 min before being removed from the mill. The blend was chopped and fed into a counterrotating

**TABLE II**  
PP/RTR Blends Using Sulfur Crosslinking Agent<sup>a</sup>

Blend <sup>b</sup>	PP (pbw)	Reclaimed tire Rubber,
		RTR (pbw)
S7, S8	80	20
S9, S10	75	25
S11, S12	70	30
S13, S14	65	35
S15, S16	60	40

<sup>a</sup> TMTD (1.5 phr), MBT (0.75 phr), ZnO (5 phr), stearic acid (2 phr), and sulfur (3 phr).

<sup>b</sup> Odd and even blend numbers refer to the unvulcanized and vulcanized blends, respectively.

**TABLE III**  
PP/RTR Blends Using MA/DCP

Blend	PP (pbw)	RTR (pbw)	MA (pbw)	DCP (pbw)
S17	70	30	2	—
S18	70	30	—	0.4
S19	70	30	2	0.4

twin-screw extruder at 190, 195, 200, and 205°C (from hopper to nozzle), with a rotating speed of 80 rpm, and then palletized. Test specimens were molded on a Toshiba IS 100G injection-molding machine (Toshiba, Japan) at 215, 220, 225, and 220°C (from hopper to nozzle). Impact-shaped specimens (6.35 × 1.27 × 0.64 cm and 0.0254-cm notched radius) were prepared and the individual impact strength was measured at room temperature at the impact pendulum swing 2.7 J according to ASTM D 256.

Melt flow index (MFI) data were obtained on a Keyness 7053 melt flow indexer according to ASTM D 1238. Thermal properties were measured by a differential scanning calorimeter (Perkin–Elmer DSC7; Perkin Elmer Cetus Instruments, Norwalk, CT) at room temperature to 185°C at heating rate of 20°C/min. The blend morphology was examined by a JEOL model JSM 5800LV scanning electron microscope (SEM; JEOL, Tokyo, Japan). Samples were prepared by fracturing the blends under liquid nitrogen, followed by staining in osmium tetroxide (OsO<sub>4</sub>) at room temperature for 17 h; stubs were bonded with double-sided adhesive tape and sputter coated with gold.

## RESULTS AND DISCUSSION

### PP/GRT blends using sulfur crosslinking agent

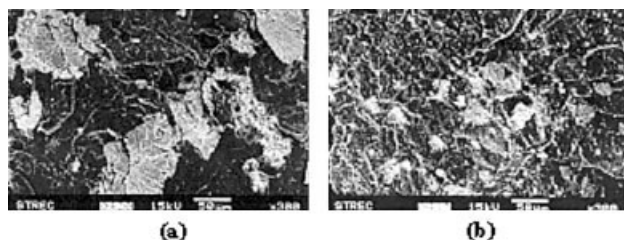
Even though much attention has focused on blending elastomer with thermoplastics, very few studies on blending GRT with thermoplastics such as PP and PE, particularly semirigid blends, have been reported.<sup>25–27</sup> In addition, it has already been shown that vulcanization would improve the mechanical property of the blend<sup>25</sup> and the most common vulcanizing agent is sulfur. Thus, a PP/GRT blend using sulfur crosslinking agent was prepared in this study. Table IV shows the notch-Izod impact strength of a 70/30 PP/GRT blend with various GRT particle sizes as well as a comparison between vulcanized and unvulcanized blends. It can be seen that all PP/GRT blends with and without vulcanization have slightly higher impact strength than that of PP because of the impact energy absorption of GRT, which is a softer material than PP. However, the impact strength of vulcanized PP/GRT blend is only slightly higher than that of the unvulcanized blend. This is attributed to the fact that GRT is a mixture of SBR, NR, and BR, which has already been

**TABLE IV**  
Notch-Izod Impact Strength of Unvulcanized and vulcanized 70/30 PP/GRT Blends

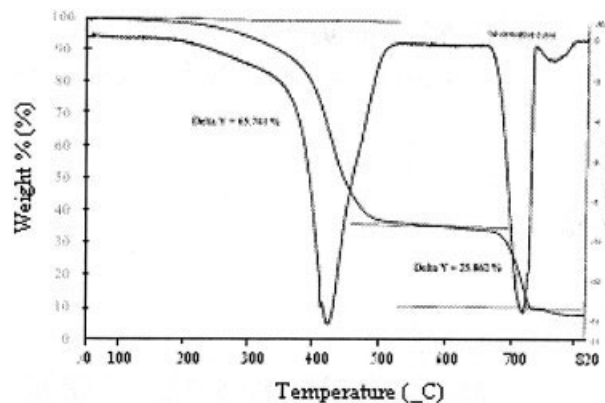
Blend <sup>a</sup>	Notch-Izod impact strength (kJ/m <sup>2</sup> )
PP	3.07
Unvulcanized PP/GRT	
S1 (40, 0.42 mm)	3.30
S3 (16, 1.19 mm)	3.27
S5 (8, 2.38 mm)	3.22
Vulcanized PP/GRT	
S2 (40, 0.42 mm)	3.75
S4 (16, 1.19 mm)	3.28
S6 (8, 2.38 mm)	3.25

<sup>a</sup> Numbers in parenthesis refer to mesh size and particle size of GRT.

vulcanized in the tire manufacturing process, and thus only a slight degree of crosslinking can occur. When tensile specimens of both vulcanized and unvulcanized PP/GRT blends were tested for elongation at break, the result was the sudden break of all testing specimens. This showed that all PP/GRT blends, although having strength, had no appreciable toughness. It was also observed that GRT particle size insignificantly influenced the impact strength of the blends. However, the vulcanized 70/30 PP/GRT blend using GRT mesh size of 40 (Sample S2) had 20% higher impact strength than that of PP, whereas other blends showed only 6% improvement. This can be explained by the fact that GRT with smaller particle size could be vulcanized more easily. It is clearly exhibited that the vulcanized PP/GRT blend had better dispersion and distribution of GRT into the PP matrix than the unvulcanized blend (Fig. 1). It can thus be said that when sulfur is combined in the vulcanization network the coalescence of rubber is prevented. Therefore, the vulcanized PP/GRT blend has higher impact strength than that of the unvulcanized blend. This result is similar to the work of Rajalinggam and coworkers,<sup>28</sup> who studied LLDPE/GRT blend using ethylene glycidyl methacrylate copolymer as the compatibilizer. They concluded that higher impact strength of the composites was obtained when smaller GRT particle size was used.



**Figure 1** SEM micrographs of (a) unvulcanized PP/GRT (70/30) and (b) vulcanized PP/GRT (70/30) blends.

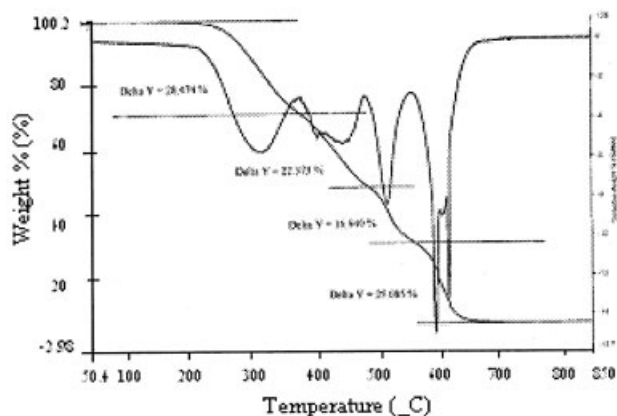


**Figure 2** TGA thermogram of reclaimed-tire rubber (RTR).

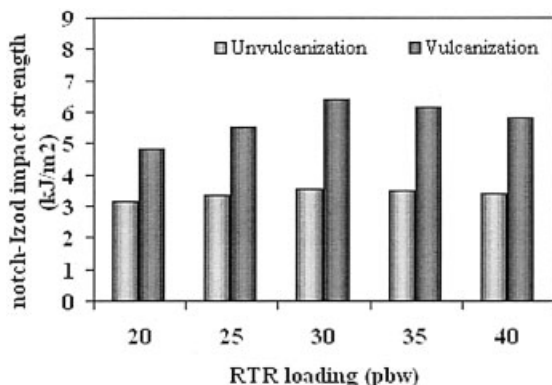
However, only a small increase in impact strength of the composites was observed.

#### PP/RTR blends using sulfur crosslinking agent

The RTR consists of 65.74% miscible blends of SBR, NR, and BR resulting from the devulcanization of recycled-tire rubber; 25.86% carbon black; and 8.40% residue as revealed by thermogravimetric analysis (TGA), heating from 50 to 850°C at a scanning rate of 20°C/min under oxygen atmosphere (Fig. 2). It is distinctly different from GRT whose major components were still the 67.79% vulcanized SBR, NR, and BR; 25.08% carbon black; and 7.12% residue (Fig. 3). At the same ratio of PP/elastomer at 70/30 with sulfur crosslinking agent, the PP/RTR blend exhibited nearly twice the impact strength of the PP/GRT blend. The explanation is that RTR can serve better than GRT to absorb impact energy, much the same as natural rubber or other elastomer does, given that RTR is a softer material than PP. In addition, blending of various amounts of RTR with PP was an additional focus of our investigation. The result indicates that the impact



**Figure 3** TGA thermogram of ground rubber tire (GRT).



**Figure 4** Notch-Izod impact strength of vulcanized and unvulcanized PP/RTR blends at various ratios.

strength of all blends is higher than that of PP ( $3.07 \text{ kJ/m}^2$ ) (Fig. 4).

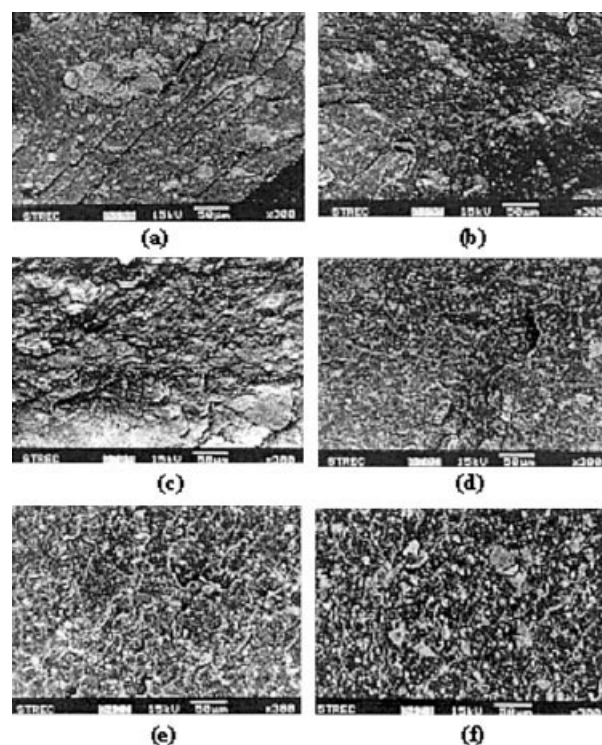
This study also compared the results between the vulcanized and unvulcanized PP/RTR blends. A marginal increase was observed in the impact strength of unvulcanized PP/RTR blend at the RTR loading up to 30 pbw. In the case of vulcanized PP/RTR blend the significant increase in impact strength was the result of not only the impact energy absorption of RTR but also the sulfur crosslinking between RTR particles. Crosslinking in the rubber phase actually increases the cohesive strength and allows the rubber particles to undergo greater deformation before cohesive failure. At RTR loading higher than 30 pbw, a decrease in the impact strength of PP/RTR blends with both vulcanization and unvulcanization was observed. This is probably attributable to the amount of carbon black that was already present in RTR. It can be reasoned that the incorporation of a more brittle and rigid material, like carbon black in the PP matrix, can form a layerlike structure depending on the amount of carbon black in the blend. Thus at the critical carbon black concentration, the PP/RTR blend can have a split in the layer structure providing a shorter path for fracture propagation, thereby causing the sudden decrease in impact strength. This phenomenon was previously discovered in some related works.<sup>27,29</sup> Phadke and De<sup>29</sup> blended 100 pbw PP, 20 pbw NR, and various GRT loadings at 20–60 pbw. They reported that the impact strength continued to increase to a GRT loading up to 40 pbw. At higher GRT loading, a decrease in impact strength was observed.

SEM micrographs of all unvulcanized blends [Fig. 5(a)–(c)] appear to have larger RTR aggregates than do all vulcanized blends [Fig. 5(d)–(f)] at the same loading, indicating a worse distribution of RTR into the PP matrix for the unvulcanized blends. This result indicates that better dispersion apparently occurs from the effective role of crosslinking in resisting rubber re-agglomeration. A similar observation was reported by

Dao.<sup>30</sup> He investigated the effect of degree of crosslinking on the dispersion of ethylene propylene diene monomer rubber (EPDM) in PP and concluded that high crosslink concentration resulted in the finest dispersion.

For PP/RTR blends at ratios of 80/20 and 70/30, it can be concluded that the RTR phase can distribute evenly into the PP matrix, whereas larger RTR aggregates are observed in the case of PP/RTR blend at a ratio of 60/40. This is probably attributable to the limitation of the amount of carbon black in the blend, given that carbon black can produce a partial immobilization of the chain segment of rubber. This line of reasoning is consistent with the sudden decrease of impact strength of the 60/40 PP/RTR blend.

The percentage crystallinity of PP decreased when RTR was present in the blends resulting from the inclusion of the more flexible phase of RTR, which could disturb the packing of PP chains (Table V). It should be noted that at 30 pbw RTR loading, the highest impact strength—but the lowest percentage crystallinity—was obtained for both vulcanized and unvulcanized PP/RTR blends. However, the vulcanized PP/RTR blend had lower percentage crystallinity than that of the unvulcanized blend. This can be attributed to the sulfur crosslinking of RTR, which acted as local defects, not allowing close packing of the



**Figure 5** SEM micrographs of unvulcanized PP/RTR blends at ratios of (a) 80/20, (b) 70/30, and (c) 60/40; and of vulcanized PP/RTR blends at ratios of (d) 80/20, (e) 70/30, and (f) 60/40.

**TABLE V**  
**Thermal Analysis and Notch-Izod Impact Strength of**  
**Vulcanized and Unvulcanized PP/RTR Blends at Various**  
**RTR Loading**

Blend <sup>a</sup>	$\Delta H_f$ (J/g <sub>composite</sub> )	$\Delta H_f$ (J/g <sub>PP</sub> )	Crystallinity (%)	Notch-Izod impact strength (kJ/m <sup>2</sup> )
PP	85.52	85.52	45.01	3.07
Unvulcanized				
S7 (20)	68.18	85.23	44.86	3.15
S9 (25)	63.55	84.74	44.60	3.35
S11 (30)	57.29	81.85	43.08	3.52
S13 (35)	49.50	82.52	43.42	3.47
S15 (40)	55.38	85.20	44.84	3.38
Vulcanized				
S8 (20)	63.24	79.05	41.60	4.82
S10 (25)	55.91	74.55	39.24	5.50
S12 (30)	49.60	70.86	37.29	6.38
S14 (35)	47.22	72.65	38.24	6.13
S16 (40)	44.14	73.56	38.72	5.82

<sup>a</sup> The number in parentheses refer to RTR loading (pbw).

polymer chains and leading to the higher decrease in percentage crystallinity of PP.

The vulcanized PP/RTR blends exhibited a relatively larger decrease in MFI than that of unvulcanized PP/RTR blends at the same ratio. This result may be explained by the cured RTR phase contributing to a higher viscosity.

It is clearly observed that PP/RTR blends have better impact strength than that of PP/GRT blends both with and without vulcanization. Of course, the inherent crosslink density of GRT makes it difficult for any interpenetration of GRT and PP phases at the interface. On the contrary, no crosslink in RTR allows the proficient dispersion of rubber chains in RTR into the PP matrix that is higher than GRT.

#### PP/RTR blends using MA/DCP

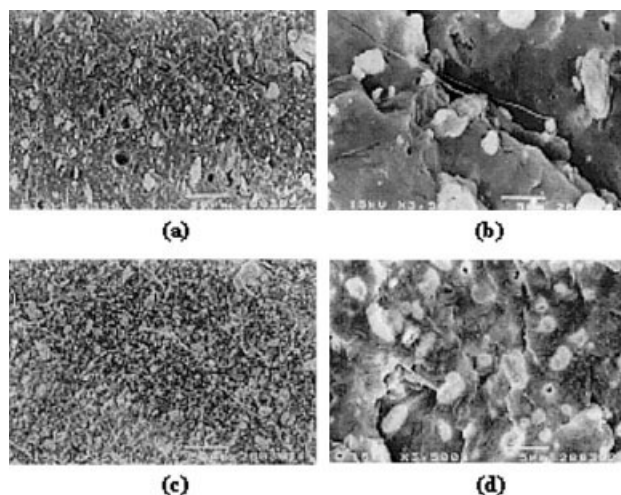
Generally, improvement in the toughening of a thermoplastic depends on the type of toughening agent and thermoplastic matrix. Even though a sulfur crosslinking agent is the most common, several others have also been used. MA is frequently used to compatibilize thermoplastic elastomer blends requiring the initiator, and organic peroxides such as DCP are known to be the initiators necessary for the vulcanization of saturated rubber<sup>6,31,32</sup>; thus a combination of MA and DCP was used. The results were then compared with the blends using the sulfur crosslinking agent.

It was found that the impact strength of the PP/RTR blend using a combination of MA and DCP is much higher than that of the PP/RTR blend using sulfur crosslinking agent, MA alone and DCP alone (Table VI). Besides the crosslinks, the significant increase in

**TABLE VI**  
**Notch-Izod Impact Strength of 70/30 PP/RTR Blends**  
**with Crosslinking Agents**

Blend	Notch-Izod impact strength (kJ/m <sup>2</sup> )
S12 (PP/RTR/sulfur crosslinking agent)	6.38
S17 (PP/RTR/MA)	5.70
S18 (PP/RTR/DCP)	6.62
S19 (PP/RTR/MA/DCP)	8.07

the impact strength is attributed to the grafting of MA onto PP<sup>32</sup> and rubber,<sup>33</sup> which induces the compatibilizing action through the dipolar interaction among the grafted materials, PP and rubber. This causes a reduction in interfacial tension and an increase in interfacial adhesion, which allow the interface to withstand a greater stress before the particle debonds, thus reducing the domain size of the dispersed phase. Apparently, the dispersion and distribution of RTR into the matrix of this blend is greater than in the blend with sulfur crosslinking agent, as shown in Figure 6. Because of the presence of carbon black in RTR, these blends are actually the ternary polymer composites in which carbon black is covered with rubber material and dispersed into the PP matrix. The impact toughness of PP/EPDM/glass bead, the other ternary polymer composite, was also studied by Liang et al.<sup>24</sup> They reported that the improvement of the impact toughness is attributable to the synergistic effect between the glass beads and EPDM, which are comparable to carbon black and rubber material, respectively, in this research work. The explanation is that such system is beneficial for improving the stress distribution in the matrix around the filler under action of external im-



**Figure 6** SEM micrographs of PP/RTR (70/30) blend crosslinked with sulfur crosslinking agent: (a)  $\times 350$ ; (b)  $\times 3500$ ; and PP/RTR (70/30) blend crosslinked with MA/DCP: (c)  $\times 350$ ; (d)  $\times 3500$ .

compact loading, in which the soft interlayer will yield first. This shear yielding will percolate through the matrix as soon as the stress exceeds some limit. In this case, a number of crazes and microvoids are induced and a great quantity of the impact energies are absorbed. On the other hand, these inclusions block the propagation of the cracks, either the original or those developed from the crazes. Consequently, the impact toughness of the composites is obviously improved. The toughening mechanism of PP-rubber blends, particularly a ternary polymer composite, is in fact complicated and involves several factors, as thoroughly reviewed and discussed by Liang and Li.<sup>34</sup>

Apart from the improvement of the impact resistance, which is significantly increased, the elongation at break of the 70/30 PP/RTR blend using MA and DCP is over 500%, which is much higher than that of PP alone. Therefore RTR can virtually toughen PP.

### CONCLUSIONS

This work reveals not only that recycled-tire rubber, RTR and GRT, can be used as a toughening agent of PP, but also that RTR is much better than GRT in enhancing the impact strength of PP. This is because RTR is the devulcanized used-tire rubber, in which the carbon-sulfur bonds that crosslink between the rubber chains have already been ruptured, whereas GRT is still the vulcanized one. Therefore the rubber chains of RTR can penetrate into the PP matrix much more easily than GRT loading to promote better adhesion and higher impact strength of the blends. In addition, all PP/RTR blends, either crosslinked by using sulfur crosslinking agents or MA/DCP, show higher impact strength than that of the noncrosslinked blends, whereas percentage crystallinity and MFI are significantly less than one. This research also demonstrates that PP/RTR blends using MA/DCP exhibit even greater impact strength than that of blends that use sulfur crosslinking agents. From the preceding results, it can be concluded that RTR plays an important role in toughening of PP by thorough dispersion into the PP matrix. Furthermore, during dynamic vulcanization, the crosslinked rubber phase becomes finer and is uniformly distributed in the PP matrix, thus attaining a stable morphology. Accordingly, the major increase in interfacial adhesion resulted when MA/DCP was used, which caused the crosslinking between two different phases, rubber chains and PP chains, in addition to the crosslinking inside each phase. Even though the impact strength of the PP/RTR blend increases with the amount of RTR in the blend, RTR loading is limited by the amount of carbon, originally

present in RTR. However, it is interesting that RTR, derived from the disposed rubber tire, has been discovered to be a useful material. Besides lowering the cost, the addition of RTR can obviously improve the toughness of PP and extend its use.

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