Study of a New Route by Which to Make Fully Cured Thermoplastic Elastomers with Plastics and Ultrafine Powdered Rubber*

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SUMMARY: A series of fully cured thermoplastic elastomers (TPEs) have been developed by mixing plastics and fully cured, ultra-fine powdered rubbers (UFPRs). The fully cured UFPRs were prepared by the irradiation of rubber latexes and drying of the irradiated rubber latexes. The average particle size of the UFPRs ranged from 50 to 500nm. The influences of the dose rate, absorbed dose and content of cross-linking agent on the cross-linking of rubber latexes were studied. It was found that a new type of TPEs with excellent properties can be easily made by blending UFPRs with plastics.

Introduction

The rapid development of fully cured thermoplastic elastomers (TPEs) has led to significant interest in both industry and academia[1-10]. The TPEs, made by a dynamic vulcanization process, were invented by Coran et al.[11-13] and are currently produced by the Advanced Elastomer System Company. TPEs have been widely used in automobile, electrical and medical applications [14]. However, special mixing equipment is needed for producing TPEs, which leads to higher cost, because continuous strong shear stress is required during vulcanization of the rubber phase. It has been found that the particle

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size of rubber strongly affects the properties of TPEs. Coran et al. found for TPEs that the smaller the rubber particles sizes, the higher the ultimate tensile strength, the greater TPEs' ultimate elongation and the better the processing properties. However, it is difficult to obtain a TPE with particle size smaller than 1 μm, since particle size and its distribution are determined by the balance between the shear rate of the mixing process and the chemical interaction rate during dynamic vulcanization[7]. In addition, the particle size of the TPEs made by the dynamic vulcanization method is not stable. This is because many factors, including the content and type of cross-linking agent, temperature and time of the melt mixing, the shear rate of the mixing process and the interface energies of the given polymer pairs, can affect particle size and distribution. In order to overcome this disadvantage of the dynamic vulcanization technology and to reduce the rubber particle sizes, a new technology has been invented by the authors of this paper. TPEs could be made by mixing plastics and fully cured, ultrafine powdered rubbers (UFPRs) using standard plastic-mixing machinery. The fully cured UFPRs were prepared by irradiation of rubber latexes and drying of the irradiated rubber latexes. The fully cured UFPRs have small particles sizes, freely flow without the need of a partitioning agent and perfectly disperse in a resin matrix. Since UFPRs are fully cured before they are used to prepare fully cured TPEs, the process only involves the simple blending of fully cured UFPRs with plastics to obtain the required fully cured TPEs. Special processing machines and conditions are also not needed. Various factors influencing the properties of the fully cured TPEs obtained by our new technology will be studied and described in this paper.

Experimental

Materials

1. Carboxylic styrene butadiene rubber latex, Designation: XSBRL-54B1, Solid content:50% by weight, Yanshan Petrochemical Company.
2. Styrene butadiene rubber latex, Designation: Dingben-50, Content: 45% by weight, Lanzhou Petrochemical Company.
3. Carboxylic acrylonitrile butadiene rubber latex, Designation: XNBRL, Solid

content: 45% by weight, Lanzhou Petrochemical Company.

4. Natural rubber latex, Solid content: 60% by weight, Beijing Latex Manufacturing factory.
5. Trimethylol propane triacrylate (TMPTA), Beijing Dongfang Chemicals Co.
6. Iso-octyl acrylate (OMA), Beijing Dongfang Chemicals Co.
7. Medium viscosity nylon-6 (mv-Nylon-6), Designation1013b, η_r:2.7, UBE Industries,Ltd., Japan.
8. High viscosity nylon-6 (hv-Nylon-6),η_r:3.5, SINOPEC Yueyang Petrochemicals Co.
9. Polystyrene (PS), Designation: 666d, Yanshan Petrochemical Company.
10. High-impact polystyrene (HIPS), Yanshan Petrochemical Company.
11. Homo-polypropylene (PP-1), Designation: III, M_w:$3.0*10^5$, Tianjin Petrochemical Company.
12. Homo-polypropylene (PP-2), Designation:1300, Mw:$3.5*10^5$, Yanshan Petrochemical Company.
13. Co-polypropylene (PP-3), Designation:1740, Content of ethylene units is 7% by weight ,Yanshan Petrochemical Company.
14. Polypropylene-g-glycidylmethacrylate (PP-4), synthesized by the current authors.

Synthesis

Preparation of fully cured UFPRs

Rubber particles in the rubber latex were fully cured by high-energy irradiation (γ-rays from a ^{60}Co source were used) in the presence of a cross-linking agent (TMPTA or OMA). The rubber latex was dried after radiation and thus fully cured UFPRs were obtained. The particles sizes of the fully cured UFPRs were the same as the sizes of the rubber particles in the latex.

Preparation of fully cured TPEs

The above-obtained fully cured UFPRs and plastics were blended using a ZSK-25 twin screw extruder (WP Company, Germany). The extruder's temperature was determined according to the processing temperature of the plastics used. After drying, the specimens for testing were obtained after injection molding.

Measurement of properties

1. Measurement of the gel fraction: The gel fraction was measured by immersing about 0.1g samples covered with 120-mesh copper gauge in boiling xylene for 8 hours. The gel fraction was calculated by the following formula: gel fraction (%) = w_2/w_1 (w_1 is the original sample weight; w_2 is the gel weight after extraction in xylene).
2. Measurement of the swelling index: A small box weighing w_0 and made of 120-mesh copper gauge was immersed into toluene for a moment, then taken out and immediately weighed (w_1). The box and 0.1 gram of UFPR were immersed in toluene for 24 hours at room temperature. Thereafter, the box and the swelled UFPR were weighed (w_2). Then the box and the gel were dried to a constant weight (w_3). The swelling index was calculated according to the following formula: swelling index = $(w_2 - w_1) / (w_3 - w_0)$.
3. Tensile strength, elongation at break, tension set, and tensile stress at 100% elongation were determined according to the ASTM D412. Shore hardness was obtained by ASTM D2240-95.
4. Weight-average molecular weight (M_w) was measured by gel permeation chromatography (GPC) at 135°C, using ortho-dichlorobenzene as solvent.
5. Determination of rubber particle sizes in the rubber latex was done using a transmission electron microscope (TEM).
6. Determination of rubber particle sizes in the powdered rubber was done using TEM for blends and SEM for rubber powders.

Results and Discussion

1. **Preparation of Fully Cured UFPRs:** In order to obtain TPEs with optimum

properties, the UFPRs must be fully cured before blending them with the thermoplastics. The rubber particles in the rubber latexes, with which the mentioned UFPRs were prepared, were unvulcanized or slightly vulcanized and had therefore to be further cured by high-energy irradiation. This process is easily controllable and ideally suited to industry[17]. The fully cured UFPRs can be obtained by drying the irradiated rubber latexes. The particle sizes of the UFPR are as same as the particle sizes of the rubber particles in the latexes. A cross-linking agent, e.g. multi-functional acrylate, has to be used in the radiation cure of the rubber latex in order to increase the efficiency of the radiation cure and reduce the irradiation dose. The mechanism of the irradiation cure of the rubber latex is a free radical reaction. During the irradiation with high-energy rays, C-H and C-C bonds in the rubber's molecular chains are broken to produce free radicals. The free radicals can react with the free radicals in other molecular chains, or initiate a chain reaction of the double bonds in the unsaturated rubber's molecular chains or unsaturated bonds in the cross-linking agent [17]. The irradiation dose, dose rate, the type and amount of the cross-linking agent used all strongly affect the degree of cross-linking of the rubber in the latex during the irradiation cure of the rubber latexes.

2. **Preparation of Fully Cured Natural UFPR:** As natural rubber latex is unvulcanized, high irradiation doses and crosslinking agents are required to prepare fully cured natural UFPR. Trimethylol propane triacrylate (3% by wt.) was added dropwise into the natural rubber latex, while stirring. The rubber latex was then irradiated with an irradiation dose of 150 kGy, at a dose rate of 50 Gy/min. After drying with a spray dryer, a free-flowing powder of the fully cured natural rubber was obtained[15]. The particle size of the powdered rubber was determined to be about 500 nm, the gel content was 94.1 %, and the swelling index was 12.8. The effect of the absorbed dose on the gel content of the natural powdered rubber is shown in Figure 1. The gel content of the rubber increased as the absorbed dose increases. The gel content increased only slowly after the absorbed dose reached to 100 kGy. Figure 2 shows the effect of the irradiation

dose rate on the gel content of the powdered natural rubber at the same absorbed dose. When the irradiation dose rate was less than 90 Gy/min the gel content of the powdered natural rubber increased as the dose rate decreased, which shows that a low dose rate is beneficial for the irradiation cure of rubber under the same irradiation dose. The possible reason is that longer irradiation time leads to a decrease in the concentration of the free radicals during unit time when the dose rate is low, therefore, the free radicals in the rubber chains have sufficient time to react with each other to generate a network structure. When the dose rate is higher, the concentration of the free radicals is also higher. The free radicals in the same molecular chain are able to react easily, which leads to the linking probability of free radicals from different molecular chains decreasing; and the probability of forming network structure therefore decreases. A lower irradiation dose rate and higher irradiation dose are required to prepare fully cured UFPR.

3. **Preparation of Fully Cured Carboxylic Acrylonitrile Butadiene UFPR:** Carboxylic acrylonitrile butadiene rubber latex is a synthetic rubber latex which has been crosslinked to a degree before irradiation; therefore, a lower irradiation dose is required to prepare fully cured carboxylic acrylonitrile butadiene UFPR. Trimethylol propane triacrylate (3 phr, added dropwise) was added to the carboxylic acrylonitrile butadiene rubber latex while stirring. The rubber latex was then radiated with an irradiation dose of 25 kGy. A free-flowing powder of the fully cured carboxylic acrylonitrile butadiene was formed.

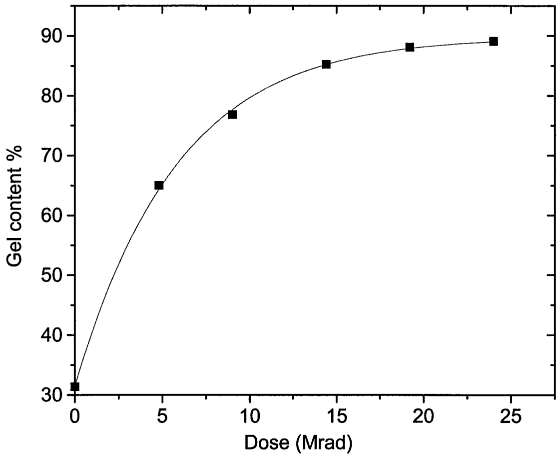

Fig.1 The effect of absorbed dose on gel content of natural UFPR (Dose rate: 50 Gy/min;Crosslinking Agent: OMA, Amount : 5 phr based on rubber).

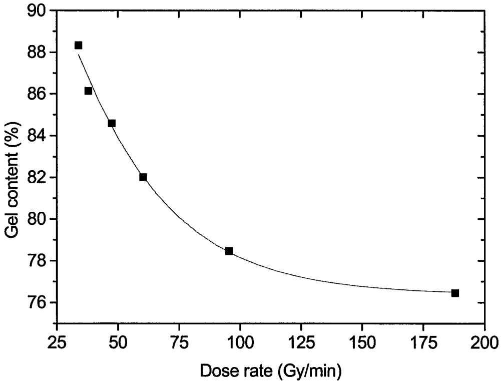

Fig. 2 The effect of dose rate on gel content of natural UFPR (Absorbed dose: 114 kGy; Crosslinking Agent: OMA Amount : 5 phr based on rubber).

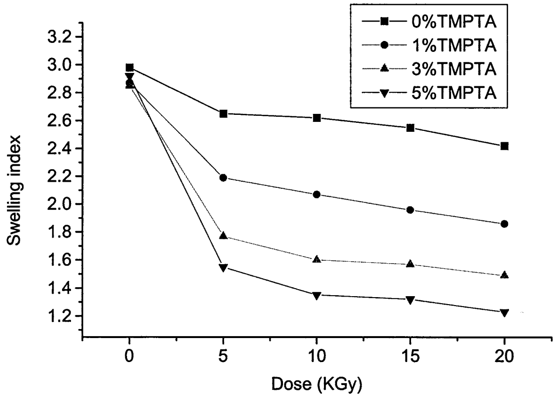

Fig.3 The relationship between the swelling index of carboxylic acrylonitrile butadiene UFPR with different amount of TMPTA and absorbed dose.

UFPR was obtained after drying the irradiated latex with a spray dryer [15]. The particle size of the powdered rubber was measured to be about 90 nm, the gel content was 97.1 %, and the swelling index was 6.7. Figure 3 shows the effect of the absorbed dose on the swelling index of the powdered carboxylic acrylonitrile butadiene UFPR with different amounts of TMPTA. The swelling index gradually decreases with an increase in the absorbed dose. It can also be seen that the higher the content of the cross-linking agent, the smaller the swelling index. This indicates that the cross-linking agent promotes the cure of carboxylic acrylonitrile butadiene rubber.

4. **Preparation of Fully Cured Carboxylic Styrene Butadiene UFPR:** Like the carboxylic acrylonitrile butadiene rubber latex, carboxylic styrene butadiene rubber is also vulcanized to a degree before irradiation cure. A lower irradiation dose is therefore required to prepare fully cured carboxylic styrene butadiene UFPR. Iso-octyl acrylate (3 % by wt.) was added to the carboxylic styrene butadiene rubber latex while stirring. The rubber latex was irradiated with an

irradiation dose of 25 kGy. The irradiated latex was dried with a spray dryer to yield a free-flowing powder of the fully cured carboxylic styrene butadiene UFPR [15]. The particle size of the powdered rubber was measured to be about 150 nm, the gel content was 92.6 %, and the swelling index was 7.6.

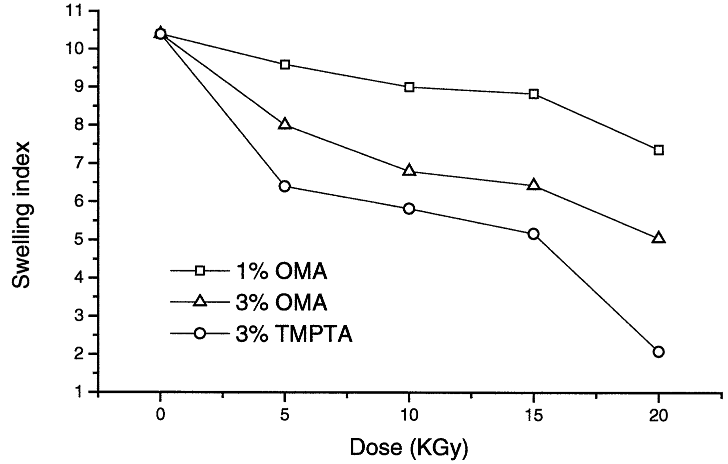

Fig. 4 Relationship between dose rate and swell index of carboxylic styrene butadiene UFPR prepared with various amounts of different crosslinking agents.

Figure 4 shows the relationship between the absorbed dose and the swelling index of the carboxylic styrene butadiene UFPR prepared with various amounts of different cross-linking agents. The swell index increases with the content of a given cross-linking agent. It can also be seen from Figure 4 that the higher functionality of a cross-linking agent, the greater is the promoting action on the cross-linking of the latex. For example, the functionality of OMA is 1 while that of TMPTA is 3. The swelling index of UFPR with TMPTA is evidently lower than that with OMA under the same absorbed dose and content.

5. **Preparation of Other Fully Cured UFPRs:** Fully cured styrene butadiene UFPR, fully cured acrylonitrile butadiene UFPR, fully cured chloroprene UFPR

and fully cured acrylic UFPR were also prepared by an irradiation cure and drying of the respective latexes.

6. **Preparation of Fully Cured Thermoplastic Elastomers:** Owing to phase inversion, normal rubbers and plastics cannot be used to prepare thermoplastic elastomers through simple blending. However, the above-mentioned fully cured UFPRs are fully cured, and they are always phase dispersed inside the thermoplastic matrix as long as their volume fractions are less than the critical rubber volume fractions. When the content of UFPR is greater than 50%, the blend is a thermoplastic elastomer. Compared to conventional dynamic vulcanization technology, the cost to produce fully cured thermoplastic elastomers with this technology is lower, the equipment used is simple and the products have stable properties, light colour and higher modulus. By using this process, fully cured thermoplastic elastomers can be simply developed not only from mixtures of the compatible elastomer-thermoplastic compositions, but also from immiscible blends, as long as UFPRs or the plastics are grafted with some monomers to improve their compatibility, as shown in Table 1.

A series of fully cured thermoplastic elastomers have been developed by simply mixing the above-mentioned fully cured UFPRs and various plastics, including the fully cured styrene butadiene UFPR/polypropylene thermoplastic elastomer, fully cured styrene butadiene UFPR/polyethylene thermoplastic elastomer, fully cured NR UFPR/polyethylene thermoplastic elastomer, fully cured carboxylic styrene butadiene UFPR/nylon thermoplastic elastomer and fully cured acrylonitrile butadiene UFPR/nylon thermoplastic elastomer, fully cured carboxylic acrylonitrile butadiene UFPR/nylon thermoplastic elastomer, and fully cured styrene butadiene UFPR/polystyrene thermoplastic elastomer.

Table 1. Compatibility between the fully cured UFPRs and plastics [18]

Material and solubility parameters	PP	PE	PA	POM	PET	PBT	PVC	AS	PC	PS
	7.8-8.0	7.9	12.7-13.6	11.0	10.7	11.0	9.5	9.4-9.6	9.5	9.1
DB 8.6	Y	Y	GR	GR	GR	GR	GR	GR	GR	Y
*SDB 9.0	GP	GP	Y	Y	Y	Y	Y	Y	Y	GP
DJ 9.4	GP	GP	Y	Y	Y	Y	Y	Y	Y	GP
*SDJ 9.6	GP	GP	Y	Y	Y	Y	Y	Y	Y	GP
BS 8.8	GP	GP	Y	Y	Y	Y	Y	Y	Y	GP
NR 8.0	Y	Y	GR	GR	GR	GR	GR	GR	GR	GR
CR 9.0	GP	GP	GR	GR	GR	GR	Y	Y	Y	Y
GXJ 7.5	Y	Y	GR	GR	GR	GR	GR	GR	GR	GR
DP 9.35	GP	GP	GR	GR	GR	GR	Y	Y	Y	Y

*PP:polypropylene, PE:polyethylene, PA:nylon, POM:polyoxymethylene, PET:polyethylene terephthalate, PBT:polybutyleneterephthalate, PVC:polyvinylchloride, AS:acrylonitrile styrene butadiene copolymer, PC:polycarbonate, PS:polystyrene, PMMA:polymethylmethacrylate, DB:fully cured styrene-butadiene UFPR, SDB: fully cured carboxylic styrene-butadiene UFPR, DJ: fully cured acrylonitrile butadiene UFPR, SDJ: fully cured carboxylic acrylonitrile butadiene UFPR□BS: fully cured butyl acrylate UFPR, NR: fully cured NR UFPR, CR: fully cured chloroprene UFPR, GXJ: fully cured silicone UFPR□DP: fully cured butadiene-vinylpyridine UFPR.

Y stands for direct blending, GP stands for plastics needed to be modified□GR stands for UFPR needed to be modified.

**the solubility parameters of the polymers are estimated according to the SIMOR chart [19]

7. **Fully Cured UFPRs/Nylon Thermoplastic Elastomer:** Nylon can be directly mixed with fully cured acrylonitrile butadiene UFPR, fully cured carboxylic acrylonitrile butadiene UFPR and fully cured carboxylic styrene butadiene UFPR to prepare fully cured thermoplastic elastomers. Owing to the reaction of carboxyl groups with amine groups in nylon, the blending of fully cured UFPRs with nylon can ensure the dispersion of rubber particles uniformly and stably in a plastic

matrix. The mechanical properties of the resultant fully cured thermoplastic elastomers are shown in Table 2.

Table 2. Mechanical properties of fully cured UFPRs/nylon thermoplastic elastomers.

Rubber/Plastics	Tensile strength (MPa)	Elongation at break (%)	Tension set (%)	Tensile stress at 100% elongation (MPa)	Shore hardness (HD)
Carboxylic acrylonitrile butadiene UFPR / hv-Nylon-6 (50/50)	39.2	142	62	33.5	62
Carboxylic acrylonitrile butadiene UFPR / hv-Nylon-6 (60/40)	30.5	118	44	26.9	55
Carboxylic acrylonitrile butadiene UFPR / hv-Nylon-6 (70/30)	29.4	137	44	25.2	52
Carboxylic styrene-butadiene UFPR / mv-Nylon-6 (50/50)	29.4	188	96	23.2	56
Carboxylic styrene-butadiene UFPR / mv-Nylon-6 (60/40)	24.7	167	94	18.4	52
Carboxylic styrene-butadiene UFPR / mv-Nylon-6 (70/30)	18.7	186	73	12.8	44
Carboxylic styrene-butadiene UFPR / mv-Nylon-6 (75/25)	17.0	175	61	11.8	40
Carboxylic styrene-butadiene UFPRr / mv-Nylon-6 (80/20)	15.6	166	52	10.9	36

The tensile strength, tension set and hardness decrease as the content of UFPRs increases for both the carboxylic acrylonitrile butadiene UFPR/nylon thermoplastic elastomer and carboxylic styrene butadiene UFPR/nylon thermoplastic elastomer. Nevertheless, tensile strength of the former decreases slowly, when the ratio of the carboxylic styrene butadiene UFPR and nylon-6 is greater than 70/30. It can also be seen from Table 2 that the tensile strength of the carboxylic acrylonitrile butadiene UFPR/nylon thermoplastic elastomer can reach 30MPa, although the ratio of the rubber and nylon is 70/30.

Tensile strength and elongation at break are inversely dependant on the particle diameter of the rubber, therefore, the smaller the rubber particle size, the better the mechanical properties. In dynamic vulcanization, smaller rubber particles (1-2 μm) can be prepared, and therefore, better properties obtained[7]. The average particle size of the UFPRs is even smaller, so it is understandable that the tensile strength of the thermoplastic elastomers obtained is even higher.

8. **Fully Cured UFPRs/Polystyrene Thermoplastic Elastomers:** Fully cured UFPRs/polystyrene thermoplastic elastomers are prepared from a simple blend of homo-polystyrene or high-impact polystyrene and fully cured butadiene-styrene UFPR. Their properties are shown in Table 3. The properties of these blends are not as good as those of the nylon type, since the entanglement density of polystyrene is very low. When the content of the rubber is greater than 70%, or high-impact polystyrene is used, thermoplastic elastomers with elongation at break of more than 100% can be obtained.

Table 3. Mechanical properties of fully cured styrene-butadiene UFPR/polystyrene thermoplastic elastomers.

Rubber/Plastics	Tensile strength (MPa)	Elongation at break (%)	Tension set (%)	Tensile stress at 100% elongation (MPa)	Shore hardness (HD)
Styrene-butadiene UFPR /PS (30/70)	33.2	58	37	---	70
Styrene-butadiene UFPR /PS (50/50)	18.8	93	56	18.5	58
Styrene-butadiene UFPR /PS (70/30)	11.4	113	29	10.7	40
Styrene-butadiene UFPR / HIPS(60/40)	11.7	107	41	11.6	46
Styrene-butadiene UFPR/ HIPS(70/30)	8.1	113	33	7.6	29

9. **Fully Cured UFPRs/polypropylene Thermoplastic Elastomers:** Fully cured UFPRs/polypropylene thermoplastic elastomers with various properties were prepared by simply blending different polypropylenes with UFPRs. Table 4 shows that the molecular weights of polypropylene and the homo- or co-polymer with ethylene units have a great effect on the properties of the TPEs. As the molecular weight of polypropylene increases, the elongation at break increases. The tension set, tensile strength and hardness do not change. Comparing these results with those of the fully cured UFPRs/homo-polypropylene thermoplastic elastomers, it is seen that the elongation at break, tension set and hardness of fully cured UFPRs/co-polypropylene thermoplastic elastomers are considerably improved. Further, fully cured thermoplastic elastomers with good oil resistance are obtained by blending acrylonitrile butadiene UFPR and polypropylene, with

improvement of compatibility.

Table 4. Mechanical properties of fully cured UFPR/polypropylene thermoplastic elastomers.

Rubber/Plastics	Tensile strength (MPa)	Elongation at break (%)	Tension set (%)	Tensile stress at 100% elongation (MPa)	Shore hardness (HD)
PP-1/ Styrene-butadiene UFPR (30/70)	13.1	134	25	12.9	27
PP-2/ Styrene-butadiene UFPR (30/70)	13.7	267	24	13.0	27
PP-3/ Styrene-butadiene UFPR (30/70)	8.7	273	17	8.1	21
PP-4/ Carboxylic acrylonitrile-butadiene UFPR (30/70)	14.1	127	20	12.5	35

Conclusions

In the preparation of fully cured UFPRs a high irradiation dose and low irradiation dose promoted an increase in the degree of crosslinking of the rubber. Addition of a cross-linking agent can decrease the irradiation dose needed and increase the degree of cross-linking of the rubber. Moreover, the higher the functionality of the cross-linking agent, the greater its promoting action on the cross-linking of the latex.

A series of fully cured thermoplastic elastomers have been developed by the mixing of plastics and fully cured UFPRs. By using this process, fully cured thermoplastic elastomers can be simply prepared, not only from mixtures of compatible elastomer-thermoplastic compositions but also from incompatible elastomer-thermoplastic compositions as long as the UFPRs or the plastics are grafted with selected monomers to improve their compatibility.

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