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### Radiation Graft Copolymerization of n-Butyl Acrylate on Natural Rubber Latex

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NOTE

## Radiation Graft Copolymerization of n-Butyl Acrylate on Natural Rubber Latex

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### ABSTRACT

A method of radiation graft copolymerization of n-butyl acrylate (NBA) on natural rubber (NR) latex has been studied. The rate of conversion increases with the increase of NBA in latex. An irradiation dose of about 12 kGy is needed to obtain 90% conversion with 40 phr of NBA in latex. Tensile strength, tear strength, and elongation at break of grafted NR are found to decrease with increasing degree of grafting. The physical strength of a vulcanizate prepared from a mixture of NR and poly-NBA was found to be better than that of NBA-NR graft copolymer vulcanizate. The graft copolymerization reaction takes place in the outer layer of NR particles, and because the secondary bonds between poly-NBA molecules may be weaker than those between NR molecules, the existence of a poly-NBA layer in NR particles will decrease its physical strength.

### INTRODUCTION

Radiation is a useful tool for the grafting of monomers on natural rubber (NR) latex. Research on radiation grafting of MMA, St, AN, and EA monomers on NR latex has been carried out by a number of

investigators. An increase in hardness, tensile strength, and modulus have been found [1-6]. The main purpose for radiation graft copolymerization of NBA on NR latex is to obtain an oil-resistant rubber. This paper presents the experimental results on the method of grafting NBA monomer on NR latex. The physical properties of the graft copolymer vulcanizates are also presented.

Radiation graft copolymerization of NBA monomer on NR latex can be regarded as a seeded radiation emulsion polymerization of NBA monomer in which the NR particles act as the seed particles. The mechanism of seeded emulsion polymerization has been presented by a number of investigators [7, 8]. According to this theory, in seeded emulsion polymerization there is no particle formation stage. The polymerization takes place inside the NR particles, directly to the second or third stage of emulsion polymerization. A constant rate of conversion is found in the second stage, but in the third stage the rate of conversion decreases with time.

## EXPERIMENTAL

Concentrated NR latex was obtained from Pasir Waringin Estate, West Java, Indonesia. The dry rubber content of the concentrated latex was about 60 wt% and the viscosity was about 50 cP. The NBA monomer was purified by distillation under reduced pressure. Other chemicals, such as oleic acid, ZnO, ZDC, and sulfur, were of technical grade and used without further purification.

Irradiation was carried out using a Panoramic Co-60 irradiator, 55 kCi activity, and the irradiation dose rate about 2 kGy/h. The irradiation dose rate was calibrated using a Fricker dosimeter. A Toyo Seiki rubber mill was used for mixing of the dry rubber with the chemicals. An Instron testing machine, Model 1122, was used to determine the physical properties of the vulcanizate.

A given amount of NBA emulsion was mixed with a concentrated NR latex and then irradiated. The NBA emulsion was prepared by stirring a mixture of 70 wt%  $\text{NH}_3$  solution (1.5%), 30 wt% NBA, and 0.5 wt% oleic acid. Irradiation doses were 3, 6, 9, and 12 kGy. The percent conversion  $C$  of NBA monomer to polymer or graft copolymer was determined gravimetrically just after irradiation by the following equation:

$$C = 100(W_1 - W_0)/M_0,$$

where  $W_0$  is the weight of solid material before irradiation,  $W_1$  is the weight of solid material after irradiation, and  $M_0$  is the weight of monomer.

The NR-NBA graft copolymer latex was then poured on a glass plate and dried. The dried film was mixed with the compounding ingredients on a rubber mill. The mixture was cured for 60 min at 100°C in a laboratory press. The physical properties of the vulcanizates were determined according to ASTM methods.

## RESULTS AND DISCUSSION

The rate of conversion increased with increasing monomer content in the latex (Fig. 1). Increasing NBA in latex gives a higher possibility for the monomer to form new polymer particles, increasing the rate of conversion. Previous experimental results showed that radiation emulsion polymerization of NBA can proceed quickly. A dose of less than 0.6 kGy is enough to obtain a conversion of about 90% [9]. It is seen that the rate of conversion is not constant as expected from the seeded emulsion polymerization theory developed by Hawket et al.

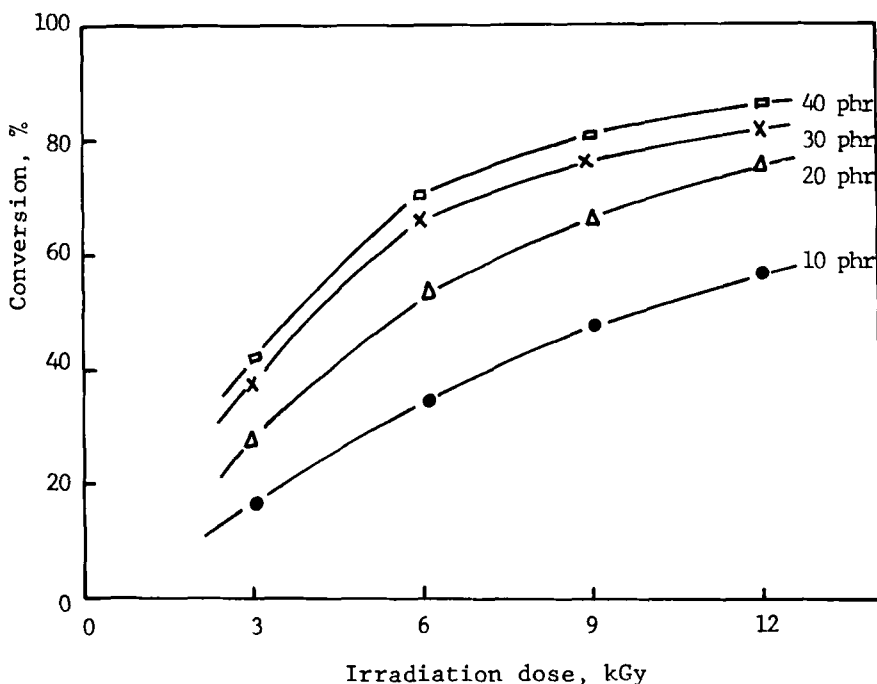


FIG. 1. Radiation graft copolymerization of n-butyl acrylate on natural rubber latex.

TABLE 1. Physical Property of NR-NBA Graft Copolymer Vulcanizate<sup>a</sup>

Dose, kGy	NBA monomer, phr	Poly-NBA, phr	Tensile strength, kg/cm <sup>2</sup>	Tear strength, kg/cm <sup>2</sup>	EB <sup>b</sup> , %	Hardness, Shore
0	40	0	212	8.1	685	48.5
3	40	16.9	24	5.2	238	50.0
6	40	28.5	12	1.2	96	54.3
9	40	32.5	11	0.9	51	55.0
12	40	34.7	8	0.8	21	55.0

<sup>a</sup>Compound composition: sulfur 2, ZnO 2, ZDC 2, NR-NBA 100.

<sup>b</sup>EB = elongation at break.

TABLE 2. Swelling Ratio of NR-NBA Graft Copolymer Vulcanizate in Benzene and the Fraction of Grafted NR Vulcanizate Extracted by Benzene<sup>a</sup>

Dose, kGy	Poly-NBA, phr	Swelling ratio, %	Weight fraction extracted, %
0	0	363	5.8
3	17	350	4.7
6	28	323	4.5
9	32	291	4.3
12	35	284	4.1

<sup>a</sup>Compound composition: ZDC 2, ZnO 2, sulfur 2, and NR-NBA 100.

[7]. It therefore appears that this grafting reaction is not a purely seeded emulsion polymerization, but may be a mixture of a pure NBA emulsion polymerization and a seeded emulsion polymerization.

Table 1 shows the physical properties of NR-NBA graft copolymer vulcanizate. It is found that tensile strength, tear strength, and elongation at break decrease with increasing degree of grafting. A very low tensile strength and elongation at break were found when the degree of grafting was around 30%. The graft copolymerization may take place only on the outer layer of NR particles due to the limited solubility of NBA in NR, and hence every NR particle will be covered by poly-NBA or NR-NBA graft copolymer, which has a poor bonding strength. A small increase in hardness was found with increasing degree of grafting. In this case NR-NBA graft copolymer acts as a filler. This is in contradiction to the property of a mixture of poly-NBA and NR latex. The tensile strength of the vulcanizate of such a mixture was found to be comparable with a NR vulcanizate.

Table 2 shows the swelling ratio of NR-NBA graft copolymer vulcanizate and the influence of the degree of grafting on the amount of materials extracted by benzene. It is seen that the swelling ratio decreases with increasing degree of grafting. This means that NR-NBA graft copolymer vulcanizate is more solvent resistant than NR vulcanizate. The amount of material extracted by benzene is higher from NR vulcanizate than from NR-NBA graft copolymer vulcanizate.

## CONCLUSION

Radiation graft copolymerization of NBA on NR latex is not a good method to improve the physical property of NR. Although the solvent resistance of NR-NBA graft copolymer vulcanizate was better than that of the ungrafted one, the tensile strength, tear strength, and elongation at break were found to be very poor.

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