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RESEARCH ON UREA CONCENTRATION EFFECT IN THE RADIATION VULCANIZATION OF NATURAL RUBBER LATEX

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Abstract- Radiations vulcanization of natural rubber latex (RVNRL) has been developed extensively through various research and development programme. This holds important benefits for rubber industries in Southeast Asia and the Pacific Region. Myanmar also develops more and more rubber plantation and many rubber raw materials are processed to manufacture medical products, household items and industrial mechanisms. In this paper, advanced and effective radiation vulcanization technique is utilized to introduce the technology in rubber industries in Myanmar. It uses gamma radiation to initiate vulcanization, a process that chemically bonds molecules to promote rubber elasticity and strength. In the present study, the radiation effect on the vulcanization of natural rubber latex and the synergistic effect of urea addition along with n-butyl acrylate (n-BA) sensitiser on the crosslinking density and physical properties of RVNRL film have been investigated. Acrylate monomer was used in order to reduce the radiation dose required to vulcanize natural rubber latex, originally 300 kGy to 15 kGy. However, there is still a need to further reduce the dose requirement without increasing n-BA concentration. The way to fulfil the requirement is the using of urea. Therefore the various concentration of urea (0.1, 0.2, 0.3, 0.4 and 0.5) (phr) was used. Tensile strength, elongation at break, modulus at 500%, swelling ratio, gel content and crosslinking density were determined. Tensile strength and modulus at 500%, gel content and crosslinking density were found increasing with absorbed dose, as well as the proportion of monomer concentration in the rubber phase in presence of urea. The radiation dose for better cross-linking of natural rubber latex with six parts per hundred rubber (phr) n-BA was 15 kGy absorbed dose from the results. The results show that by addition of 0.3 parts per hundred rubber (phr) urea to the latex before irradiation, the crosslinking density and tensile strength increases respectively.

Keywords- Natural rubber latex, N-butyl acrylate, Urea, Gamma radiation, Vulcanization, Tensile strength, Crosslinking density

I. INTRODUCTION

Significant progress has been made in the last 30 years in developing new rubber material using radiation technology. Radiation vulcanization of natural rubber latex offers several advantages over conventional vulcanization with sulfur. RVNRL is suitable for production of gloves, condoms, balloons, baby teats and many other dipped products readily exploited by the users. Rubber products for medical and hygienic uses are obviously the most promising applications of RVNRL technology because of the absence of carcinogenic and toxic products [1].
By using newer sensitizers, the radiation vulcanization offers many advantages such as Better latex stability (longer shelf life possible), Lower modulus (suit requirement of certain applications), Less or absence of toxicity problem (free from nitroamines and accelerators induce allergies), Better clarity of products (better colouration), Cleaner industrial effluents (less environmental pollution), Lower ash content (less environmental pollution) and Absence or lower acid combustion gases (safer disposal of used products) [1].
However for this technology to become commercially viable, it is essential that (i) the dose required to bring about vulcanization be brought down to the about 10-15 kGy and (ii) the concentration of the sensitizer required for adequate crosslinking should be minimized [2]. This has prompted mechanistic comparisons to be made between vulcanization and related processes. The use of n-butyl acrylate (nBA) as sensitizer has already resulted in reducing the radiation dose required to vulcanize rubber from 300 kGy to 15 kGy [3]. However, there is still a need to further reduce the dose requirement without increasing n-BA concentration. The way to fulfil the requirement is the using of urea. The purpose of the present work is to study the effect of addition of urea on the crosslinking density and mechanical properties of the radiation vulcanized natural rubber latex films to see whether similar improvement can also be obtained for the radiation vulcanization process.
Natural rubber latex (NRL) is a dispersion of natural rubber particles in water. It comes from Hevea Brasiliensis plantation. Rubber hydrocarbon is a polymer of isoprene. The average molecular weight of natural rubber varies from 1.6×10^5 to 7.0×10^5 while the weight average molecular weight varies from 5.0×10^5 to 10.0×10^5 [4].The properties of natural rubber can be improved by vulcanization. Vulcanization is an important chemical reaction of rubber. NRL can be vulcanized by irradiation without and with radiation vulcanization accelerator. Latex being a natural product, the physic-mechanical characteristics of radiation products is dependent upon the origin and the micronutrients present in the latex [5, 6].
The process parameters are sensitizer concentration, radiation dose, concentration of rubber particles in the latex, effect of various additives and ambient conditions of irradiation. The product depends upon these parameters. Oxygen may effect on the

vulcanization of rubber latex in the presence of sensitizers. Irradiation at various doses in the presence and absence of oxygen has to be carried out in order to clarify the role of oxygen in the vulcanization process [7].

II. MATERIALS AND METHODS

A. Materials

The rubber latex used in this work was a high ammonia centrifuged 55 % DRC natural rubber latex obtained from RTTCRP (Research, Technology & Training Centre For Rubber Products), Yangon, Myanmar. N-butyl acrylate (n-BA), high ammonia solution (NH_4OH), potassium hydroxide (KOH) and urea were from Able chemical store, Mandalay. All of reagents are Analar grade.

B. Irradiation of NRL

The concentrated NRL was diluted to 50% DRC by adding 1.5% dilute ammonia solution. The 50% DRC latex was prepared by adding 10% KOH solution (0.5 phr) while stirring. And then various concentration of n-Butyl acrylate (5.0 phr and 6.0 phr) was added into the latex solution while stirring as shown in Fig.1. Samples were prepared by addition of urea with various concentrations of (0.1phr, 0.2phr, 0.3phr, 0.4phr and 0.5phr).

Fig.1. Preparation of NRL with n-BA, KOH and Urea on the Magnetic Stirrer

The stirring was continued for two hours and the solution was left overnight at room temperature to mature before irradiating. The various radiation doses (15.0 kGy and 20.0 kGy) were carried out in a gamma chamber at a dose rate of 1.24 kGy hr^{-1}. The irradiated rubber latex in the gamma chamber is shown in Fig.2.

Fig.2. Irradiation of Natural Rubber Latex in the Gamma Chamber

C. Preparation of Rubber Films

Irradiated rubber films were prepared by casting the latex on glass plate (14 cm x 14 cm) and drying at room temperature until the films become clear. The films were then leached in hot water for one hour and then drying at room temperature and then dried at 70°C in an oven until the films became clear again. The casting of rubber latex on the glass plate at room temperature is shown in Fig.3.

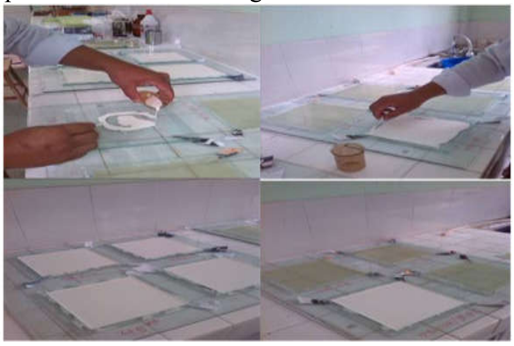

Fig.3. Casting of the Rubber Latex Film on the Glass Plate

III. MEASUREMENT OF MECHANICAL PROPERTIES OF FILMS

The mechanical properties of irradiated rubber films (tensile strength, modulus at 500% elongation and elongation at break) were conducted using dumbbell-shaped test specimens. The tensile strength of irradiated rubber films were measured by using an H-5000E tensile testing machine according to BS 903 Pt A2.

IV. SWELLING AND GEL CONTENT MEASUREMENT

About 1 gram of radiation vulcanized rubber film was soaked in benzene for 36 hours to measure swelling. Based on the equilibrium swelling data measurement, the crosslink density was estimated by Flory-Rehner equation as follow [8]:

$$V_O = K \times Q^{-5/3} \tag{1}$$

Where, V_O = crosslink density c.l/ml,
$K = 4.71 \times 10^{20}$ for benzene/NR system,
Q = volume swelling ratio.

The swelling ratio (Q) of the rubber films was calculated by measuring the mass of the sample before and after immersing in benzene [9]:

$$Q = \frac{W_s - W_d}{W_d} \tag{2}$$

Where Wd and Ws are the weight of dry and swollen sample in benzene, respectively.

The swollen samples were then dried in an oven at 50°C for 12 hours to remove the residual solvent. The gel content of specimens was calculated the following equation [10]:

$$\text{Gel content (\%)} = \frac{W_{ds}}{W_d} \times 100 \tag{3}$$

Where Wd and Wds are the weight of dry RVNRL films before and after being swollen in benzene, respectively.

V. RESULTS AND DISCUSSION

The mechanical properties of RVNRL film and the physical determination of network structure through swelling measurements have been investigated.

D. Tensile Test of Latex Films

The tensile strength of irradiated rubber films obtained by mixing various concentration of urea and n-butyl acrylate sensitiser with NRL at various absorbed doses were shown in the Fig.4, Fig.5, Fig.6 and Fig.7. The value of the tensile strength of rubber films increased with increase amount of n-butyl acrylate concentration and urea concentrations in the mixed at 15 kGy dose. However the tensile strength of 6 n-BA concentration of film slightly decreased than 5 n-BA concentration of film at 20 kGy radiation doses. These result shows that the tensile strength was optimum at the 0.3 phr urea concentration of 6 phr normal butyl-acrylate and 15 kGy dose. The increase in tensile strength may be due to increased cross-linked rubber and the sensitiser and urea concentration in the mixed. This can occur that increase the homogeneous cross-link density. It was known that with increase crosslink density, tensile strength increased [11].

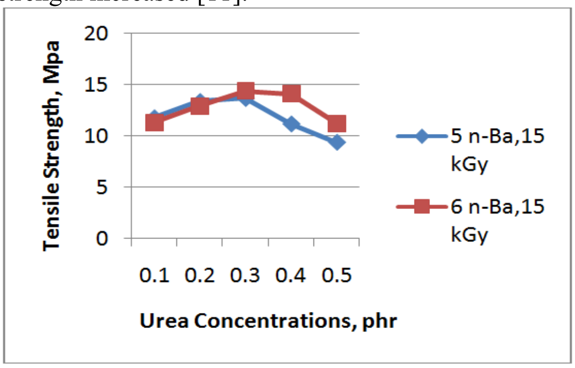

Fig.4. Tensile Strength of Rubber Film at the Various Urea Cocentrations of 5 n-BA,15 kGy and 6 n-BA,15 kGy

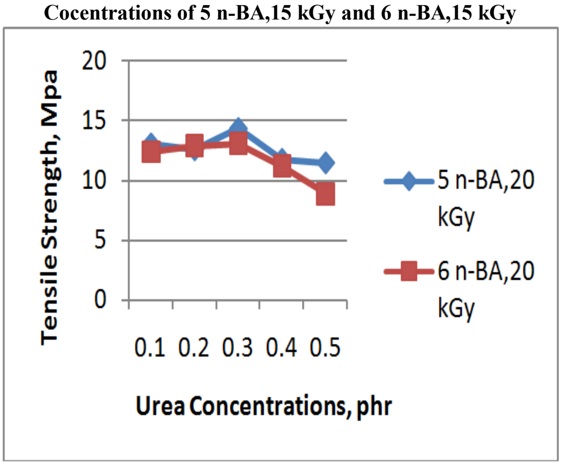

Fig.5. Tensile Strength of Film at the Various Urea Concentrations of 5 n-BA, 20 kGy and 6 n-BA, 20 kGy

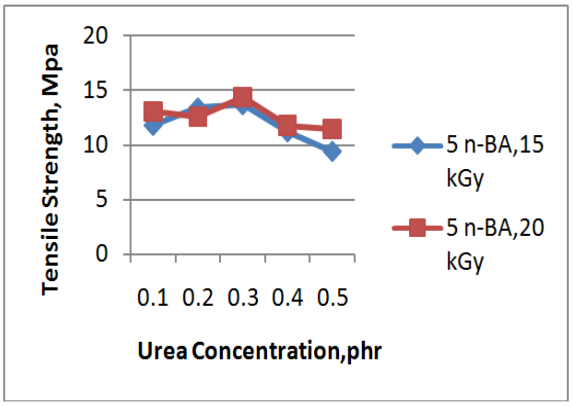

Fig.6. Tensile Strength of Rubber Film at the Various Urea Cocentrations of 5 n-BA,15 kGy and 5 n-BA,20 kGy

The tensile strength of films of modulus at 500% elongation increased with increase in concentration of urea as shown in Fig.8.The maximum tensile strength of irradiated rubber film with 6 phr n-butyl acrylate was obtained 14.5 Mpa at the absorbed dose of 15 kGy.

The elongation at break of irradiated rubber films with various composition of n-butyl acrylate at various absorbed doses is shown in Fig.9. It was found that the elongation at break decreased with increased in urea concentration. This may be due to the presence of higher rubber with increase in urea concentrations.

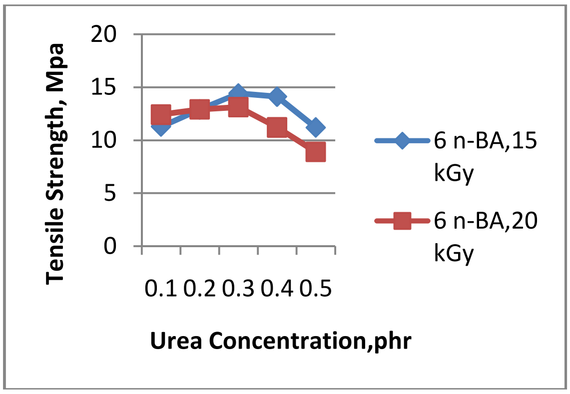

Fig.7. Tensile Strength of Rubber Film at the Various Urea Cocentrations of 6 n-BA,15 kGy and 6 n-BA,20 kGy

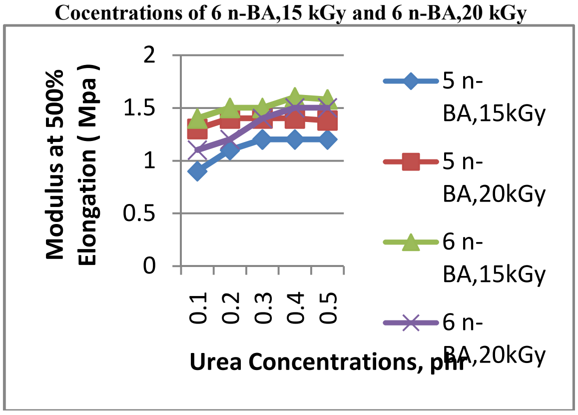

Fig.8. Modulus at 500% Elongation of the Rubber Film

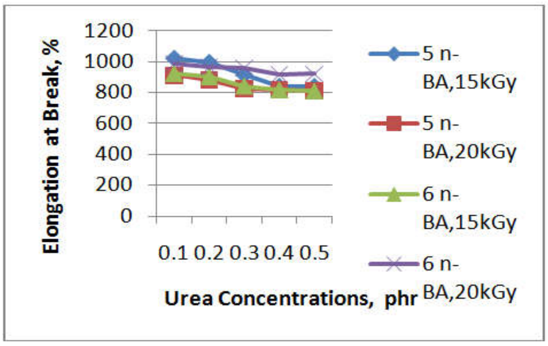

Fig.9. Elongation at Break (%) of the Rubber Latex Film

E. Physical Determination of Network Structure through Swelling Measurements

The results of gel contents of the rubber latex films were shown in Fig.10. It was observed that the optimum value of gel content was found at 0.3 phr of urea concentration. And then there was no change remarkably. The increase of gel content indicated the formation of three dimensional networks during radiation vulcanization. The swelling ratios of the rubber latex films prepared under different irradiation doses in the presence of various sensitizers and urea concentrations after leaching with benzene solvents were investigated and the results were mentioned in Fig.11. From this figure, it was observed that the swelling ratio of 5 n-BA, 20 kGy slightly decreased than of the 6 n-BA, 15 kGy. And the swelling ratio decreased that of with increasing urea concentrations. In the radiation vulcanization process, it is essential to know the response of network structure to change in the process parameters such as radiation dose, sensitizer concentration and ambient conditions. The natural rubber latex is subjected to radiation vulcanization to gain induced crosslinking. The cross-linking density of the rubber film on the various concentration of urea was shown in Table I. From this table, it was observed that the cross-linking density increased with increasing the concentration of urea. The optimum cross-linking density was found at the 0.3 phr urea concentration of 6 phr n-BA, 15 kGy. Beyond that concentration, the cross-linking density of 5 n-BA, 20 kGy of latex film slightly decreased which indicates depolymerization or chain scissors of the polymers.

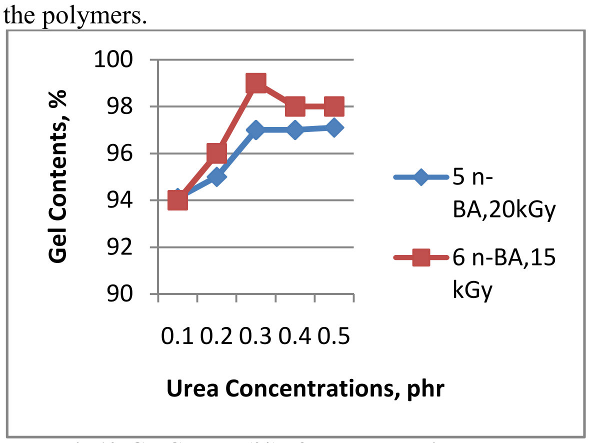

Fig.10. Gel Content (%) of the Rubber Film on the

Various Concentration of Urea

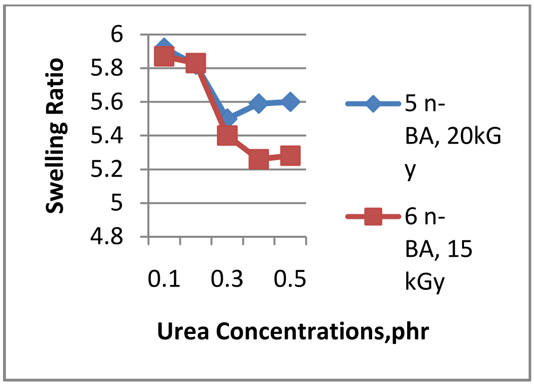

Fig.11. Swelling Ratio of the Rubber Film on the Various Concentrations of Urea

TABLE I. Crosslinking Density of the Rubber Film on the Various Concentration of Urea

	Cross-link density (c.l/ml)				
	0.1 phr of urea	0.2 phr of urea	0.3 phr of urea	0.4 phr of urea	0.5 phr of urea
5n-BA, 20 kGy	2.43×10^{19}	2.5×10^{19}	2.75×10^{19}	2.68×10^{19}	2.67×10^{19}
6n-BA, 15 kGy	2.46×10^{19}	2.49×10^{19}	2.83×10^{19}	2.85×10^{19}	2.74×10^{19}

CONCLUSIONS

The swelling ratios, cross-linking density, gel content, tensile strength and elongation at break of the NRL films were investigated variable radiation doses and various concentrations of n-butyl acrylate sensitiser and urea. Tensile strength of irradiated rubber film with 6 phr n-butyl acrylate attained maximum at the absorbed dose in the range of 15 kGy. Elongation at break and the swelling ratio decreased with the increase amount of urea added with rubber latex. Gel content and degree of cross-link density of the rubber films increased with increase urea concentration as well as with increase absorbed doses. The results indicated the increase of cross-linking during vulcanization. Recent results have also shown that in the radiation process, the sensitizing action of the n-butyl acrylate depends upon its ability to penetrate into the rubber phase. The presence of urea may further increase the solubility, and hence concentration of the n-BA in the rubber phase which results in the increased crosslinking density as well as improved tensile strength of the film. These results have shown that the addition of 0.3 phr urea to latex does not have any adverse effect on the properties of latex. Both amount of n-butyl acrylate sensitiser for crosslinking polymer and urea concentrations are responsible for the improvement of the properties of natural rubber latex film. So from this study, it has been proved that the cross-linking of the NR latex film with n-BA sensitiser and urea addition can be done by gamma radiation. From this study it can be concluded that the production of RVNRL (radiation vulcanization of natural rubber latex) and its rubber products such as condom and rubber gloves in laboratory scale can be carried out. By using these techniques local rubber production will be developed to export finished or semi-finished products, rather than raw materials only.

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REFERENCE

[1] Makuuchi, K.,Markovic, V. ,Radiation processing natural rubber latex, IAEA Bulletin, 25(1991).

[2] Makuuchi, K., Tsushima, K., Radiation vulcanization of natural rubber latex with monofunctional monomers. J. Soc. Rubber Ind., Japan, 61(1988) 478.

[3] Garnett, J.L., Dworjnyan, P.A., Bett, S.J., Dang, H.P., “Sensitisation in radiation grafting, curing and vulcanisation process”, Radiation Vulcanisation of Natural Rubber Latex, Proc. Int. Symp. Takasaki, (1990)(Machi, S., Ed), Japan Atomic Energy Research Institute (1990) 268-289.

[4] Yoshii, F., Kulatunga, S., Makuuchi, K., “Improvement of aging properties of rubber films prepared from radiation-vulcanized natural rubber latex”, Die Angewandte Makromolekulare Chemie 205 (1993) 107-115.

[5] Y. Minoura, and M. Asao, J. Appl. Polym. Sci., 1961, 5(14): p. 233.

[6] Y. Minoura, and M. Asao, J. Appl. Polym. Sci., 1961, 5(16): p. 401.

[7] ASTM-D-1672-66(1971)

[8] C. V. Chaudhari,Y. K. Bhardwaj, N.D. Patil, K.A. Dubey and S. Sabharwal, Radiat. Phys. Chem, 2005, 72(5): p. 613.

[9] Y. Minoura and M. Asao, Studies on gamma rays irradiation of natural rubber latex. The effect of organic halogen compound on crosslinking by gamma irradiation, J. Appi. Polym. Sci., 5, 401 (1961)

[10] S. W. Karunaratne, “Standardization of radiation vulcanised natural rubber latex,” in Proceedings of the International Symposium on Radiation Vulcanization of Natural Rubber Latex, JAERI-M 89-228, pp. 225–233, 1990.

[11] Haque, M.E., Dafader, N.C., Akhtar, F. andAhmad,M.U. (1996) Radiat.Phys.Chem.,48(4),505.

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