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Study on the Properties of Radiation Induced Acrylamide Grafted Natural Rubber Latex Film

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Grafted rubber films were prepared by mixing various proportions of acrylamide with natural rubber latex (NRL) with varying proportions by gamma radiation from Co-60 source at room temperature. Tensile strength, tear strength, elongation at break, permanent set, and swelling ratio, gel content and degree of grafting were measured. Tensile strength, tear strength, gel content and degree of grafting were found increasing with absorbed dose, as well as the proportion of monomer concentration and the optimum dose was found 12 kGy from the results. Elongation at break, permanent set, and swelling ratio of grafted rubber decreases with increase in absorbed dose, as well as proportion of grafted rubber.

Keywords: Irradiation, grafted rubber, tensile properties, swelling ratio

1 Introduction

Natural rubber is gummy, plastic in nature, highly sensitive to oxidation, and possesses poor tensile properties. The properties of natural rubber can be improved by vulcanization, which increases retractile force and reduces the amount of permanent deformation after removal of deforming force. Ionizing radiation can be used for vulcanization of natural rubber latex.

Grafting and blending are techniques to improve some mechanical properties of polymeric materials (1). Researchers showed that aerated natural rubber latex reacted readily with acryl amide, in the presence of a polymerization initiator, to form graft polymers (2). The molecular weight of the grafted side chains was found large and the percentage of polymerized acryl amide present as free polymer was small. Graft copolymer from natural rubber latex was prepared (3) using visible, ultraviolet and gamma-ray initiation. The influence of monomer concentration, temperature, radiation intensity and in the case of photoini-

tiation, photosensitizer concentration, on polymerization rates and molecular weights was studied. The irradiation dose in radiation emulsion polymerization of acryl amide monomer was lower compared to the irradiation dose for grafting of acryl amide monomer on natural rubber latex, in order to obtain the same degree of conversion. The tensile properties, swelling ration and permanent set are improved by incorporating hard segments like poly (methyl methacrylate) (PMMA) into natural rubber by grafting (4). The mechanical properties of thermoplastic (5) elastomer (TPE) prepared by blending methyl methacrylate (MMA) grafted radiation vulcanized natural rubber latex (RVNRL-g-PMMA) and MMA grafted natural rubber latex (NRL-g-PMMA) are comparable with commercially available olefin and styrene-type thermoplastic elastomers. George *et al.* (6) prepared methylmethacrylate graft natural rubber using gamma rays. The combined effect of radiation and chemical initiation was also studied. It was found that the properties of methylmethacrylate graft natural rubber prepared by radiation initiation are almost comparable to that prepared by chemical initiation. The slightly better properties were obtained for methylmethacrylate graft rubber prepared from latex by combination of initiation and chemical initiation. Earlier workers (7) studied morphology, micromechanical, and thermal properties of undeformed and mechanical deformed PMMA rubber

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blend and found higher orientation of PMMA molecules near the periphery of the stretched rubber particles. The properties (tensile strength, thermal stability, and gel fraction) of copolymer films (8) obtained from natural rubber latex (NRL) and styrene butadiene rubber latex (SBRL) increase with increasing SBRL in NRL and absorbed dose. In this study, the tensile and swelling properties of grafted rubber film prepared by mixing acrylamide and natural rubber latex with varying proportions are evaluated. The effect of absorbed dose on the properties is also reported.

2 Experimental

2.1 Natural Rubber Latex (NRL)

The NRL obtained from the Atomic Energy Research Establishment (AERE) rubber garden, Savar, Bangladesh was used throughout this work. The total solids content (TSC) of NRL was approximately 60%.

2.2 Preparation of Acrylamide-Grafted Rubber Latex by Radiation

The NRL was diluted to 35% Total Solid Content (TSC) with 1.5% dilute ammonia solution. 2, 5, 10 and 15 parts per hundred rubber (phr) of acrylamide were added to the dilute NRL and stirred by a magnetic stirrer for an hour. 5 phr CCl_4 was used as radiation sensitizer. The acrylamide-mixed rubber latex was irradiated (9) with gamma rays at the absorbed dose of 2, 5, 7, 10, 12 and 15 kGy with a dose rate of 313 Krad/hour.

2.3 Preparation of Grafted Rubber Film

The irradiated grafted rubber latex was poured on a raised-rimmed glass plate to make rubber film with a thickness of 0.8 to 1.0 mm. The film was dried in air till transparent. The air-dried film was leached in water for 24 h at room temperature and again dried in air till transparent (10). Then the film was heated in an oven at 70°C for 1 h.

2.4 Measurement of Properties of Grafted Rubber Film

Tensile properties (tensile strength, tear strength, and elongation at break) of grafted rubber film were measured by using INSTRON testing machine, model 1101, England,

connected to a personal computer with the testing speed of 5mm/min. Swelling ratio was measured by British Standard (BS 1673: part 4, 1953) using toluene as swelling solvent for 24 h at room temperature. Permanent set was measured by ISO 2285-1 981 (E) method.

The dumbbell shaped tensile test piece was used to determine permanent set of rubber latex film. The gauge length 1 cm of sample was marked by a pen and was extended to three times and fixed. In this condition the sample was allowed to stand for 24 h at room temperature, then the tension of the sample was released and the sample was left to stabilize for at least one hour. Then the length of the sample between the marks was measured. Permanent set was measured by the following expression:

$$\text{Permanent Set (\%)} = (L_0 - L_i)/L_i \times 100$$

Where, L_0 = Final length of the film after release the tension and L_i = Initial length of the film between marks (cm) = 1 cm.

To determine the gel content the Rubber samples were dried to constant weight in a vacuum oven at 60°C. The dried samples were immersed in Toluene at room temperature for 24 h to remove (soluble parts). The undissolved parts remaining in the solvent were again dried to a constant weight in vacuum in an oven at 60°C. The gel content was calculated as follows:

$$\text{Gel content (\%)} = W_1/W'_0 \times 100$$

Where W_1 is the weight of dry rubber after extraction in toluene and W_0 is the initial weight of dried rubber sample.

The degree of grafting was measured as a percentage of weight of grafted polymer to the weight of rubber.

3 Results and Discussion

The physical properties of NRL film and Acrylamide-grafted natural rubber film at 12 kGy absorbed dose are shown in Table 1. The tensile strength and tear strength of NRL film are lower than that of acrylamide grafted rubber film. Elongation at break, permanent set and swelling ratio of NRL films are higher than those of grafted rubber film. The cause of higher tensile strength and modulus and lower elongation at break, permanent set and swelling ratio of acrylamide grafted rubber may be due to the presence of crosslinked rubber, grafted rubber, and homopolymer of acrylamide in grafted rubber.

Table 1. Physical properties of NRL film and acrylamide grafted rubber film at 12 kGy absorbed dose

Sample	Tensile Strength, MPa	Tear Strength, N/mm	Elongation at Break, %	Permanent Set, %	Swelling Ratio	Gel Content, %	Degree of Grafting
NRL Film	5.92	6.977	987	180	31.42	91.58	4.46
Grafted Film	32.56	33.71	677	40	8.04	96.89	25.48

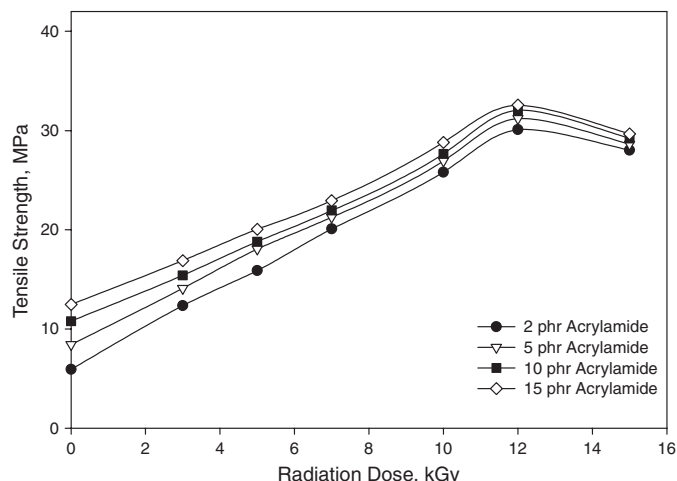


Fig. 1. Tensile strength of acrylamide grafted NRL film with various concentration of acrylamide monomer at various absorbed doses.

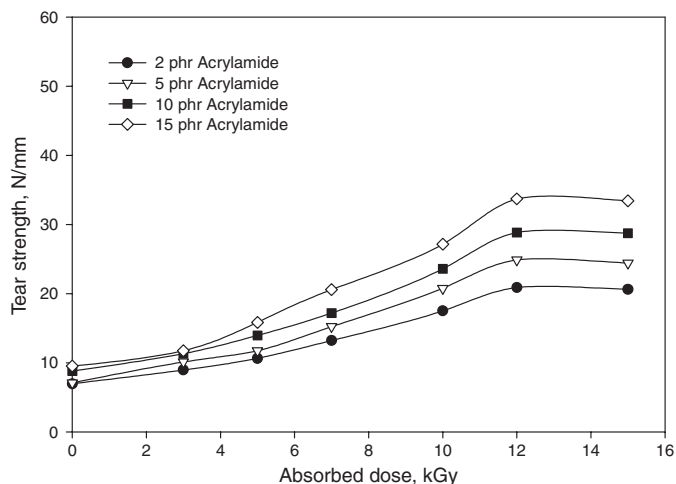


Fig. 2. Tear strength of grafted rubber film prepared from various concentration of acrylamide monomer in NRL at various absorbed doses.

It is reported (9) that tensile properties of grafted rubber is improved by crosslinked rubber and graft copolymer.

The tensile strength of grafted rubber films obtained by mixing various concentration of acrylamide monomer with NRL at various absorbed doses is shown in the Figure 1. The initial value of the tensile strength of grafted rubber films increases with an increased amount of acrylamide monomer concentration in the mixed. The increase in tensile strength may be due to increased cross-linked rubber and the monomer concentration in the mixed. The tensile strength of the grafted rubber films increased with increase in absorbed doses and attains maximum at the absorbed dose of 12 kGy. A decreasing trend of tensile strength for grafted rubber is found after attaining maximum values, due to higher crosslinking and brittleness. It is known that with increased crosslink density, tensile strength increased. The maximum tensile strength is obtained at optimum crosslinking of a polymer (11), but grafted polymer in addition with the crosslinking polymer is found to be better for the improvement of the tensile properties of natural rubber film. The maximum tensile strength of irradiated rubber film with 15 phr acrylamide was obtained 32.56 MPa at the absorbed dose of 12 kGy, where both the crosslinking and grafted polymers are present.

The effect of acrylamide monomer in the mixed and absorbed doses on tear strength of rubber film is also studied. The results are depicted in Figure 2. The value of tear resistance attains maximum at the absorbed dose of 12 kGy for the grafted rubber films. From this figure, it is found that, tear strength depends on the proportion of acrylamide concentration in the rubber mixed. Tear strength of the rubber film increases with increased amount of acrylamide monomer. It increases from 20.89 to 33.71 N/mm with varying composition of grafted rubber at the absorbed dose of 12 kGy. An increase in proportion to

the acrylamide monomer concentration in the mixed increases the hard segment due to the acrylamide grafted rubber chains and homopolymer of acrylamide. This may be the cause of improved tear strength of the grafted rubber film.

Figure 3 shows the elongation at break of grafted rubber films with various composition of acrylamide monomer at various absorbed doses. Elongation at break decreases with increased absorbed doses. This may be due to the presence of increased cross-linked rubber with increase in absorbed doses. It is also found that elongation at break depends on the proportion of acrylamide concentration in the grafted rubber film. It reduces with increased proportion of acrylamide concentration in the mixed.

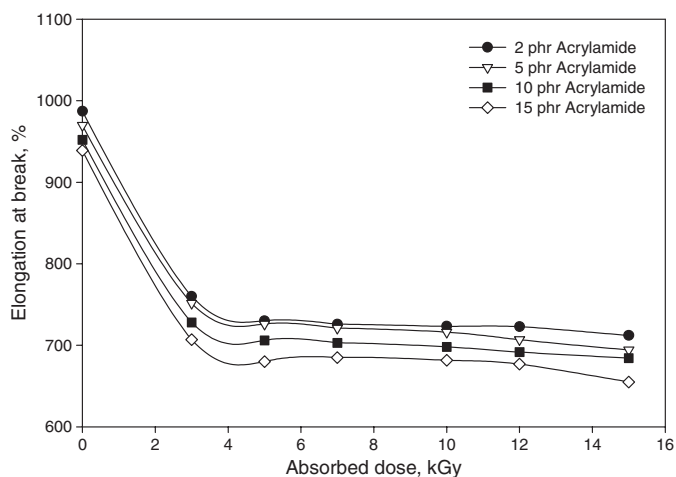


Fig. 3. Elongation at break of acrylamide grafted NRL film with various concentration of acrylamide monomer at various absorbed doses.

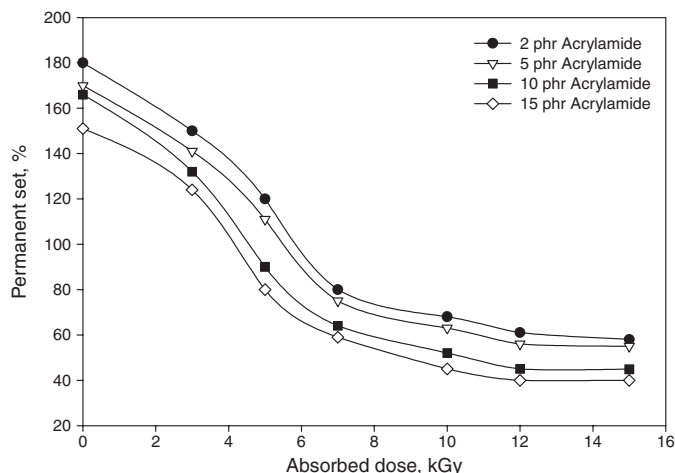


Fig. 4. Permanent set of grafted rubber film prepared from various concentration of acrylamide monomer in NRL at various absorbed doses.

Figure 4 shows a permanent set of rubber films obtained from grafting with a different composition of acrylamide in natural rubber latex at various absorbed doses. Permanent set also depends on composition of acrylamide in the grafted rubber film. It decreases with the increase in proportion of acrylamide monomer concentration in the mixed and as well as with increased absorbed doses. The decreasing trend of the permanent set is fast till 7 kGy absorbed dose is reached, and then it becomes slow.

The swelling ratio reflects the crosslinking of a polymer. It decreases with an increase in crosslinking density. The swelling ratio of rubber films obtained from grafted with different proportions of monomer concentration in natural rubber latex for different absorbed doses is shown in Figure

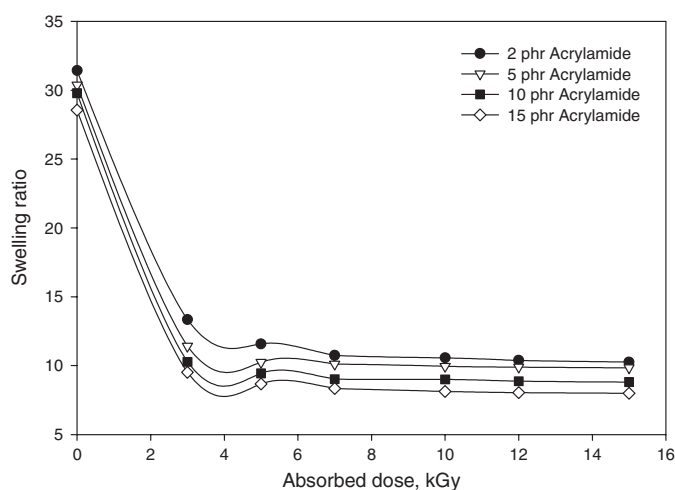


Fig. 5. Swelling ratio of grafted rubber film prepared from various concentration of acrylamide monomer in NRL at various absorbed doses.

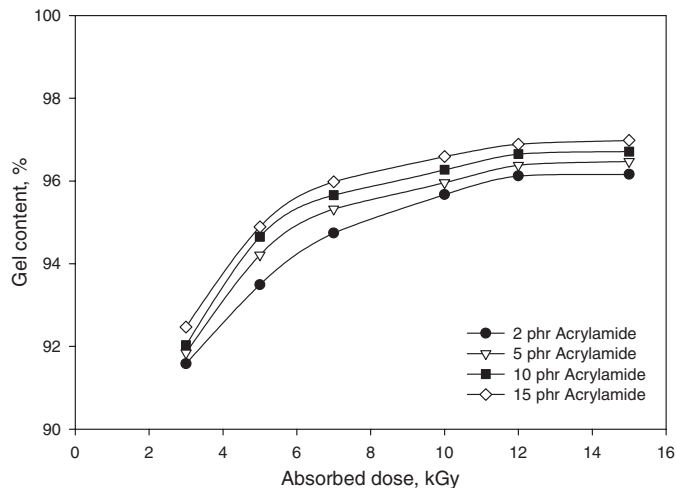


Fig. 6. Gel content of acrylamide grafted NRL film with various concentration of acrylamide monomer at various absorbed doses.

5. It is found that the decreasing trend of swelling ratio for blend rubber films is fast up to the 5 kGy absorbed dose, and then it reduces slowly.

Gel content of grafted rubber is the measure of the amount of the crosslinked rubber in to sample. Increasing crosslinking increases the gel content because of low solubility. It is found that for 15 phr monomer, grafted rubber has high gel content in comparison to other three monomer concentrations, which is shown in the Figure 6. From 3 kGy absorbed dose its gel content increases rapidly up to 10 kGy and after that the increment is not so significant. For 15 phr acrylamide monomer, the gel content at 3 kGy dose is 92.47% and the value is 96.89% at 12 kGy dose.

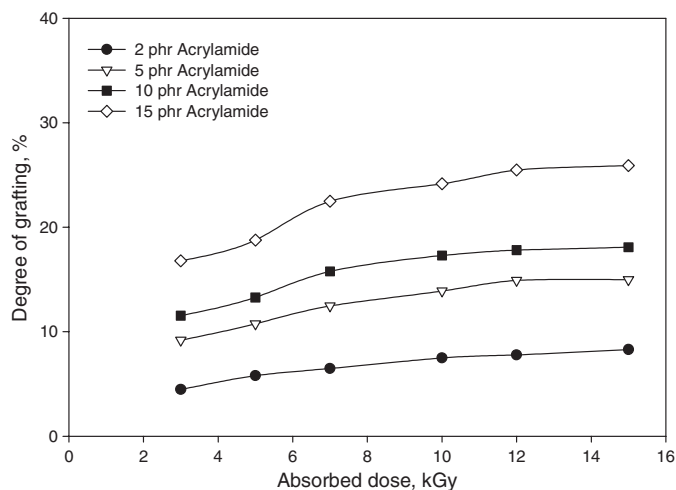


Fig. 7. Degree of grafting of acrylamide grafted NRL film with various concentration of acrylamide monomer at various absorbed doses.

The degree of grafting presents the amount of polymer actually linked with the rubber chains. The degree of grafting depends on the amount of monomer added to the rubber latex and the conversion ratio of the monomer to polymer. Figure 7 shows the results of degree of grafting of acrylamide grafted rubber film with varying composition at various absorbed doses. It is observed that the increasing amount of acrylamide monomer added with rubber latex along with increasing absorbed dose increases the degree of grafting in a large extent. 15 phr monomer with natural rubber latex shows a higher value of degree of grafting and the value is 16.79% at 3 kGy absorbed dose and the maximum value is found 25.89% at 15 kGy dose.

4 Conclusions

The properties of grafted rubber latex film prepared by mixing acrylamide and NRL in varying proportion have been studied. Tensile strength and tear strength of grafted rubber attain maximum at the absorbed dose of 12 kGy. Elongation at break, permanent set and the swelling ratio decrease with the increased amount of monomer added with rubber latex. Gel content and degree of grafting of the rubber films increases with increases monomer concentration as well as with increased absorbed doses. So, both the absorbed dose for crosslinking polymer and amount of acrylamide monomer for grafted polymer are responsible

for the improvement of the properties of natural rubber latex film.

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