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Thermal, Mechanical and Swelling Studies of Semi-Interpenetrating Polymer Network Composed of Natural Rubber and Polyacrylamide

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Semi-interpenetrating polymer network (IPN) composed of two polymers, namely natural rubber (NR) and polyacrylamide (PAAm) were prepared by crosslinking with glutaraldehyde. The NR/PAAm semi-IPN films were developed by varying the concentration of crosslinking agents and the polymer blend ratios. The impact of preparation parameters on the mechanical properties was studied. The semi-IPN was characterized by FTIR, DTG, TG and SEM. Swelling studies were done by using toluene as solvent. The kinetics and mechanism of swelling were investigated and the initial rate of swelling, swelling constant, swelling exponent, equilibrium swelling and diffusion coefficient were also determined.

Keywords: Semi-IPN systems, Mechanical properties, Swelling studies, Diffusion coefficient.

INTRODUCTION

A relatively new group of polymeric network systems containing two or more polymers in which at least one of the polymers gets crosslinked in the immediate presence of other are called interpenetrating polymer networks (IPNs) [1,2]. Separation of the network is not possible without breaking the natural bonds. Even though there are no chemical bonds holding these networks together, they are found to be linked in such a way that they will not become separated. Synthesis of IPNs can be performed by blending methods and polymerization processes such as sequential, simultaneous, *etc.* [3-5].

Interpenetrating polymer networks (IPNs) based polymer systems display various characteristics varying from hardened elastomers to high-strength plastics [6]. The final properties of a newly designed IPNs are based on the composition of the polymers under study, the concentration of crosslinking agent used, the physical nature of the constituent polymers, *etc.* [7,8]. IPNs are divided into two categories, semi and full, based on the crosslinking behaviour of the component polymers [9] and can be prepared in different manners. One method is by mixing two simultaneously polymerized and cross-linked monomers or by dissolving a monomer in a polymer network [10]. Another method for IPNs is by mixing two thermodynamically miscible polymers and crosslinked subsequently. In full IPN, the two polymers form a network of crosslinked structures, even though bonding between the two polymers is negligible [11].

Natural rubber (NR) and polyacrylamide (PAAm) are two familiar polymers and have long been used for many industrial applications, especially as an elastomer for many applications in the engineering field [12]. Its poor thermal stability, easy diffusion of organic solvents and non-compatible with the human body are considered to be its drawbacks [13-15]. Due to these drawbacks, it has been merged with several polymers. For example, chitosan [6,16], cellulose [17], polypropylene [18], polyacrylic acid [19], polyethylene [20] and polystyrene [21]. Polyacrylamides are the water-absorbent polymers forming a soft gel when hydrated [22], since they contain aquaphobic polyethene chief framework and a water loving $CONH_2$ lateral group. In this work, a semi-IPN composed of natural rubber and polyacrylamide in the presence of glutaraldehyde as crosslinker, through a sequential technique is prepared and its properties were compared with the natural rubber alone. The effect of polymer ratio and the concentration of glutaraldehyde was studied in the thermal, mechanical and swelling behaviour of the semi-IPNs synthesized.

EXPERIMENTAL

Natural rubber latex which contains 60% DRC and 0.47% conc. NH_3 were procured from Indraje Latex Centrifugal factory, Sullia, India. Glutaraldehyde (25%), toluene, acrylamide (AAm) and potassium peroxodisulphate (KPS) were provided by Merck, India and were used as such.

Preparation of Semi-IPN: The NR/PAAm semi-IPN systems were prepared as follows. The polymerization of AAm was achieved by using KPS initiator dissolved in deionized water. The IPNs from latex and PAAm were synthesized by blending them for 0.5 h to form a homogeneous mixture. Then, a 10 mL of 2% crosslinker was added and stirred uniformly for approximately 1 h. Subsequently, the solutions were cast on a glass apparatus at 45 ºC for 2 days and then the casted films were separated. Films having a total weight of 20 g and thickness of 20 mm were developed by varying polymer compositions up to 40% PAAm. Also, by fixing the polymer composition, variation in glutaraldehyde has been studied. The prepared films were then subjected to different characterization techniques. The blend formulations and sample codes are given in Table-1.

TABLE-1
PREPARATION CONDITIONS OF NR/PAAm SEMI IPN

Code	NR (%)	AAm (g)	KPS (g)	GA (%)
NR	100	0	0	2
NR/PAAm1	90	10	0.1	2
NR/PAAm2	80	20	0.1	2
NR/PAAm3	70	30	0.1	2
NR/PAAm4	60	40	0.1	2
NR/PAAm5	90	10	0.1	1
NR/PAAm6	90	10	0.1	3

Characterization: Fourier transform infrared spectrometry (FTIR) analysis were carried out using Bruker Alpha Spectrometer in the range of 4000-400 cm^{-1} at a scanning rate of 2 mm/s. The thermogravimetric analysis of the samples was done using Perkin-Elmer STA 6000 TG/DTG. Samples were heated at a temperature varying from 40 ºC to 830 ºC at 20 ºC/min in a N_2 atmosphere. NETZSCH DSC 204F1 Phoenix was used for DSC analysis. The films were subjected to heating from -80 ºC/10 (K/min)/350 ºC in a N_2 atmosphere. The morphological characterization of the prepared samples were performed. It was done at room temperature using JEOL-JSM 5800LV Scanning Electron Microscope.

Mechanical testing: Tensile and tear strength of the prepared samples were conducted as per ASTM D 412 method by taking dumb-bell shaped and angle type tear test samplings at a crosshead speed rate of 500 mm/min. The experiment was performed using a universal testing machine (Hounsfield, H10KS, UK) at room temperature.

Swelling measurements: Swelling studies were conducted using square specimens of thickness 20 mm. Samples of recorded weight were submerged in toluene as solvent in test bottles and placed at room temperature. Samples were taken out at regular intervals and the wet surfaces were dried rapidly using filter paper. The weights of the specimens were recorded immediately using an electronic balance. Samples were again kept back in the test bottle and the swelling process continued till equilibrium swelling has attained. The percentage of swelling (S%) was calculated using eqn. 1:

$$\text{Swelling ratio (\%)} = \frac{W_t - W_o}{W_o} \times 100 \qquad (1)$$

where W_t = weight of swollen sample at time t and W_o = weight of dry sample.

RESULTS AND DISCUSSION

SEM: The morphology of pure vulcanized natural rubber and semi-IPNs are shown in Fig. 1. A formation of well-developed interlocked structure, however high porosity has been developed due to the evaporation of the excess water. Although the NR surface has a homogeneous appearance, the surface of prepared sample of NR/PAAm semi- IPN has a heterogeneous texture, which indicates that both NR and PAAm are present in the IPN form.

FTIR: Fig. 2 shows the infrared spectra of NR and NR/PAAm semi IPN. The spectra of NR as well as IPNs exhibit an intense peak at 3400 cm^{-1} indicating the NH stretching vibr-

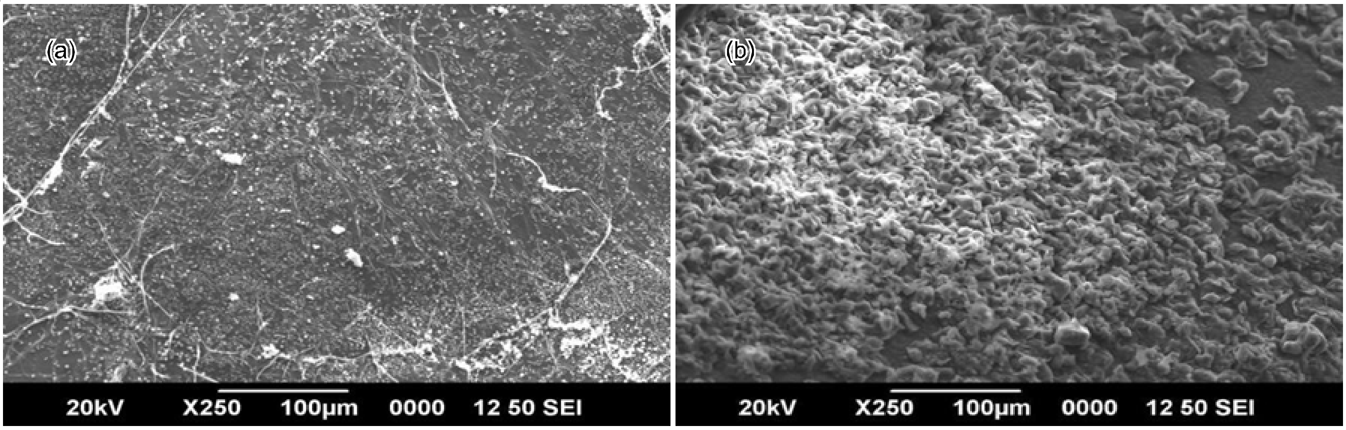

Fig. 1. SEM micrograph of (a) Ga vulcanized NR and (b) NR/PAAm4

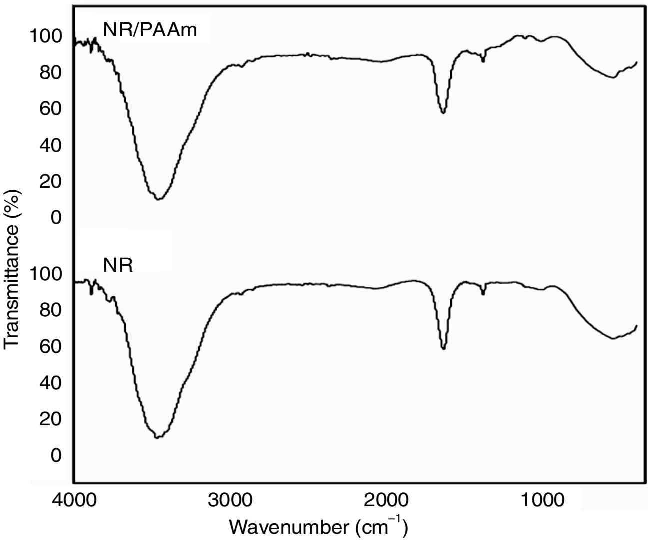

Fig. 2. FTIR images of NR and NR/PAAm semi IPN systems

ation. The peak at 2960 cm^{-1} shows the C-H stretching of methylene groups, while the peak at 1634 cm^{-1} indicates the stretching frequency of the non-conjugated double bond. The absorption band at 1384 cm^{-1} specifies the C-H asymmetric stretching [23] and the carbonyl stretching peak is observed at 1636 cm^{-1} [24]. The band at 1438 cm^{-1} assigns the features of $CONH_2$ group, which correlates C-N stretching together with N-H bending. From this, it is confirmed that both natural rubber and polyacrylamide are present in the prepared semi-IPN system. It also indicates the possibility of an ether linkage, by condensing with glutaraldehyde which is confirmed from the appearance of absorption peak at 1110 cm^{-1}.

Thermal studies: The thermogravimetric curves of pure NR as well as NR/PAAm4 semi-IPNs are shown in Fig. 3. Thermal stabilities of individual polymers and semi-IPNs can be compared from the TG curves. In the temperature range of 287-400 ºC, NR undergoes thermal degradation and results in the formation of 39% isoprene, 13.2% dipentene and some traces of *p*-menthene [14]. It was observed that when crosslinked with glutaraldehyde, the thermal stability of NR and NR/PAAm were enhanced. It was observed that in case of NR, an apparent weight loss appears only after 300 ºC and at 354.04 ºC, the weight percentage is about 97.439%.

In the semi-IPN thermogram, a small weight loss near 200 ºC is observed. It may be due to the presence of free PAAm. For the blend at 347.94 ºC, the weight percentage is about 85.497%. Also NR has a sharp weight loss, but for the blend system the weight loss is not so sharp indicating the presence of some strong interaction existing between NR and PAAm. The NR curve shows that the system reaches a saturation point by 550 ºC whereas in the blend system degradation continues again from 400-700 ºC and becomes saturated after that. From the TG studies, it can be concluded that the thermal stability of semi-IPN prepared is better than the thermal stability of constituent members.

DTG: It was reported that the DTG curve of pure PAAm shows that the decomposition rate attains its peak level at about 370 ºC. On further increasing temperature, at about 500 ºC the mass of the specimen becomes constant and remains as char-like substance [25]. However, the DTG curve (Fig. 4) of the prepared semi-IPN indicates that when PAAm mixed with NR, the decomposition rate has increased. The decomposition rate attains its peak level at 397.66 ºC, whereas it is 394.96 ºC for pure NR. Thus, the decomposition temperature of blend system is found to be on the higher side when compared to pure NR. This shows that the blend system has got more thermal stability than individual components. Also, the NR/PAAm curve represents that the sample becomes constant at 700 ºC, which indicate the better thermal stability of the prepared blend compared to pure NR. A weight loss near 200 ºC was observed, which may be due to the presence of free polyacrylamide in system.

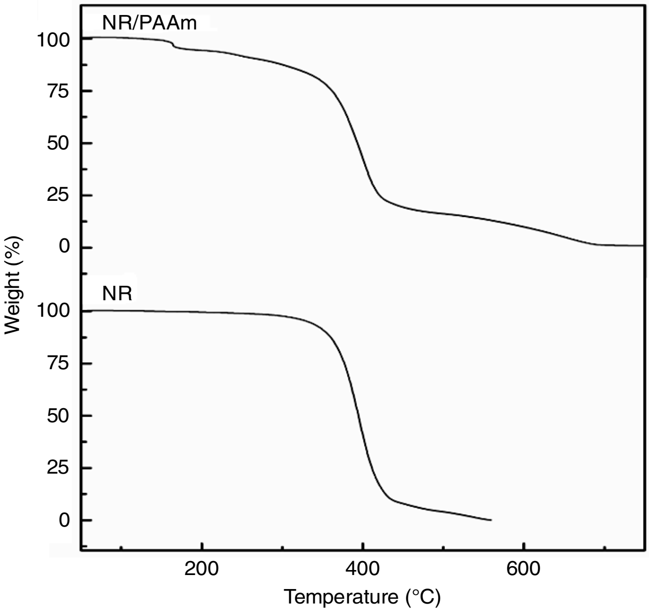

Fig. 3. TG images of NR and NR/PAAm semi IPN systems

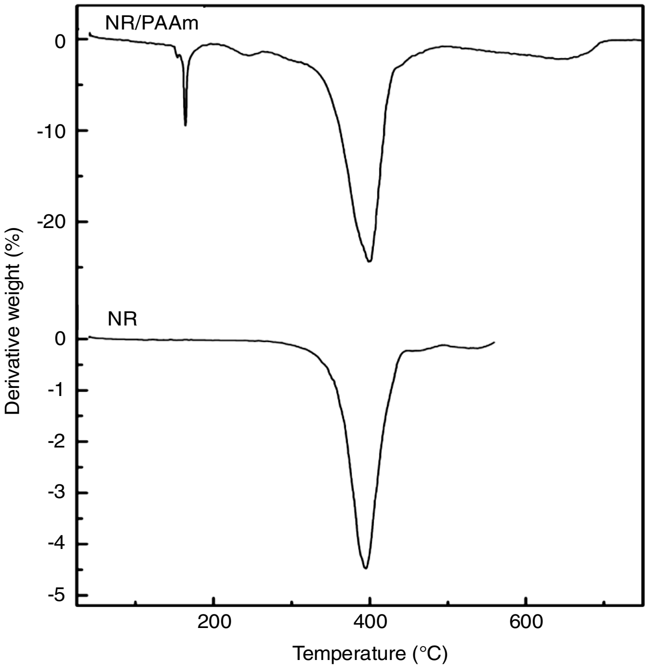

Fig. 4. DTG images of NR and NR/PAAm semi IPN systems

DSC: The glass transition temperature of pure NR is found to be -63 ºC (Fig. 5). A complex peak at 316.4 ºC may be indicating the degradation occurring in pure NR, which is confirmed by the TG curve. But no such peaks are observed in the blend DSC curve, which may be due to the formation of IPN that enhances its thermal stability. It is found that the prepared blend has a glass transition temperature at -62.7 ºC, while the T_g value of PAAm is 162.85 ºC. This may vary according to its hydrophilic nature [26]. DSC studies of the blend revealed only one glass transition and hence NR/PAAm blends are thermodynamically compatible.

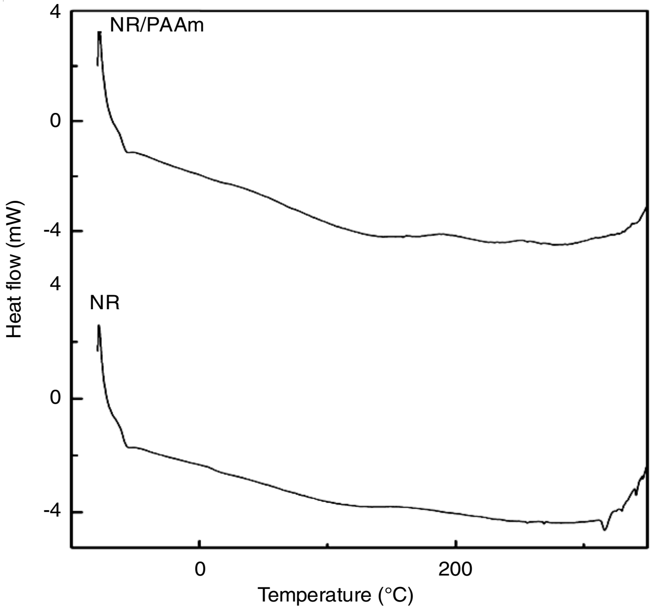

Fig. 5. DSC images of NR and NR/PAAm Semi IPN systems

Mechanical properties: The impact of polymer ratios on the tensile strength of semi-IPN systems is shown in Fig. 6a. A decrease in the tensile value is observed with the incorporation of PAAm. The compressive strength of pure NR is highest, while that of 60% NR/40% PAAm mixture is the lowest. During the crosslinking with glutaraldehyde, the NR chains are held firmly and make the maximum tensile strength. In the semi-IPN, PAAm chains were entangled with the NR chains. This makes a reduction in tensile value. Fig. 6b represents the tensile strength of NR/PAAm semi-IPNs based on the variation in crosslinking agent concentration. A significant improvement in tensile property was observed when glutaraldehyde concentration increased. As the concentration of crosslinking agents increases, the count of crosslinks in the network also increases. Thus, the tensile behaviour of semi-IPN is in the expected order.

Fig. 7a-b represents the percentage of elongation in terms of different polymer composition and glutaraldehyde variation [27,28]. A substantial reduction in the elongation property is observed with the addition of PAAm into latex. This makes the system less flexible, diminishing the polymer chain elasticity and reducing the elongation at break. But as the percentage of glutaraldehyde increased, elongation at break has also increased. As the amount of crosslinking agent increases, the extent of crosslinking also increases. This is the reason for the enhanced value for elongation at break.

Young's modulus: Young's Modulus is one of the basic properties of all materials that cannot be altered [29,30]. This property depends on stress, strain and elongation. From Fig. 8a, an increase in acrylamide has resulted in the increased modulus value up to the NR/PAAm3 composition. This may be due to an increase in the stiffness of blend to its peak level. Further increase in the acrylamide concentration has reduced the modulus value. When the percentage of glutaraldehyde increases from 1 to 2, an increase in modulus value has been observed. But at 3% glutaraldehyde modulus value decreased again as shown in Fig. 8b.

Tear strength: In the prepared semi-IPN, it was observed that the tear strength reduces with an increase in the amount of polyacrylamide (Fig. 9a). Addition of polyacrylamide can reduce the level of tear resistance by weakening the crosslink bonding within the rubber matrix. The amount of glutaraldehyde strengthens the bonding between the polymer chains and

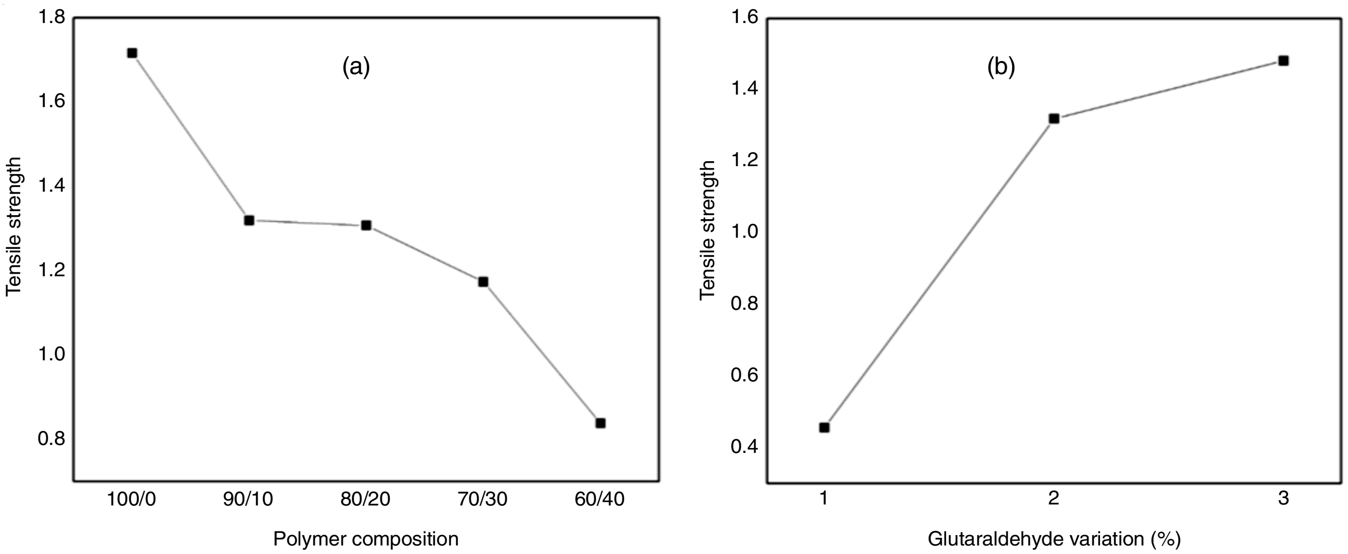

Fig. 6. Tensile strength against (a) various polymer composition and (b) different glutaraldehyde concentration

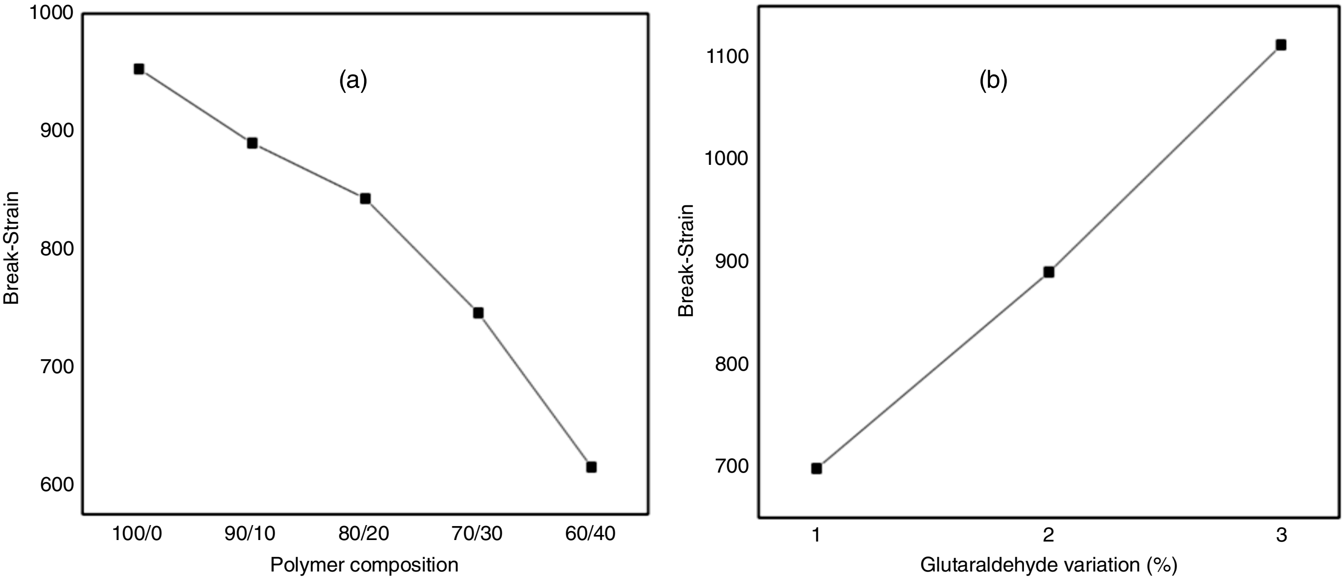

Fig. 7. Elongation at the break against (a) various polymer composition and (b) different glutaraldehyde concentration

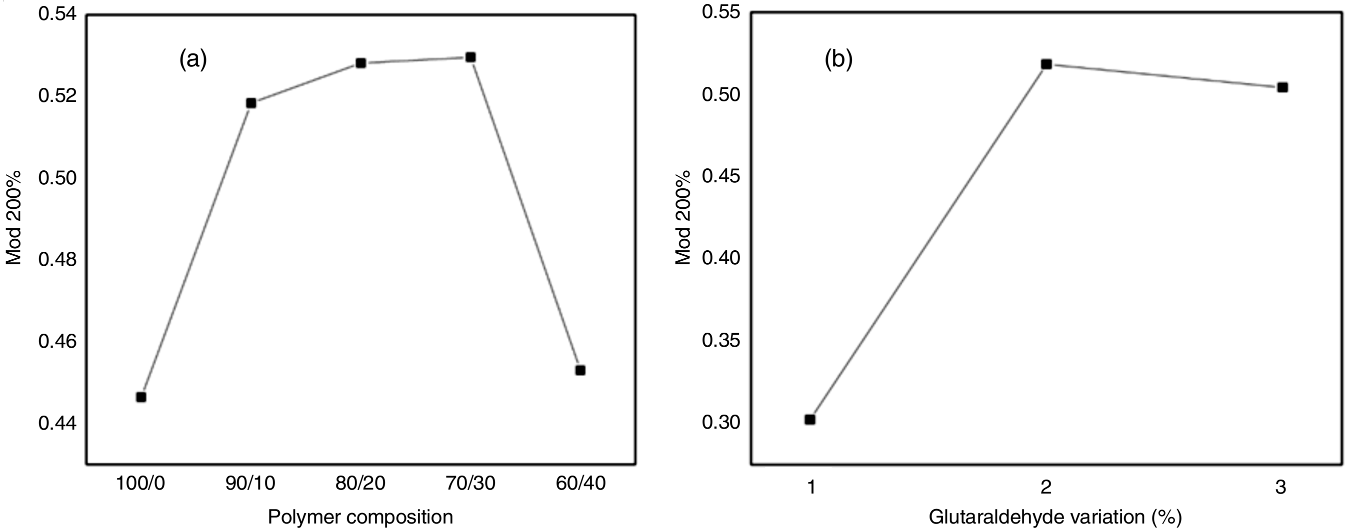

Fig. 8. Modulus value against (a) various polymer composition and (b) different glutaraldehyde concentration

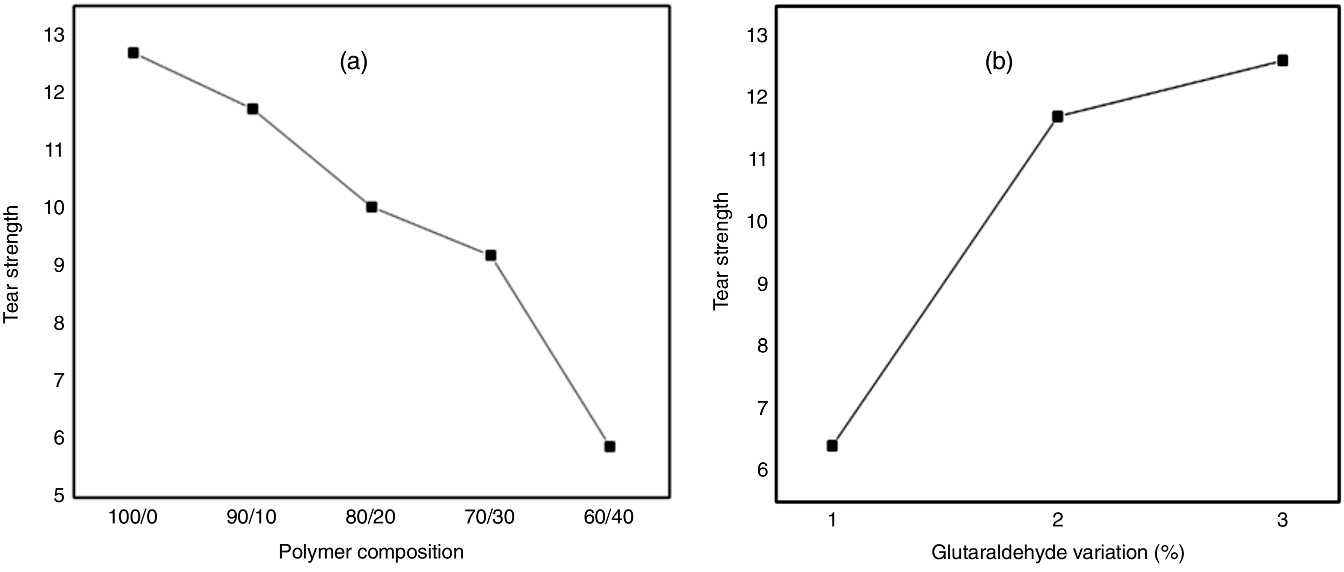

Fig. 9. Tear strength against (a) various polymer composition and (b) different glutaraldehyde concentration

thereby increases the tear strength with an increase in the glutaraldehyde content from 1% to 3% as shown in Fig. 9b.

Swelling studies: Swelling studies were carried out in toluene as solvent for pure NR and NR/PAAm IPNs prepared. It was observed that pure NR shows the maximum swelling compared to the semi-IPN. Fig. 10a shows that %swelling follows the order NR > NR/PAAm1 > NR/PAAm2 > NR/PAAm3 > NR/PAAm4. In semi-IPN, the maximum swelling (959%) was observed for NR/PAAm1 where the blend ratio was at 90:10. This indicates the less solvophilicity of semi-IPN, which restricts the polymer chains to get rearranged easily due to solvent pressure. The percentage of equilibrium solvent content (ESC%) also confirm that as the number of polyacrylamides increases, the solvent holding capacity of the semi-IPN decreases. Hence, the NR/PAAm semi-IPN systems prevent the substances from getting penetrated.

By considering NR/PAAm1 as representative sample, the swelling percentage (S%) was studied by varying glutaraldehyde concentrations from 1 to 3% using toluene as solvent. The rubber chain mobility gets reduced due to crosslinking, which subsequently diminishes the solvent transport through it. In the semi-IPN system, as glutaraldehyde vulcanizes natural rubber with higher concentration, S% decreased from 1294% to 498%. In this work, the system has maximum crosslinked and thereby prevented the solvent uptake. The ESC% from Table-2 also confirmed that as crosslinking increases, it prevents solvent holding property and it is represented in Fig. 10b.

Kinetics of swelling: Fig. 11 represents the 't/S' value plotted against 't' for swelling analysis. The swelling rate 'Ri' is computed from the intercept of the plot shown in Fig. 11. Similarly, S_{max} (maximum equilibrium swelling) was found from the slope. All these values are presented in Table-2. The linear nature of the plot shows that the swelling procedure has second-order kinetics. It can be observed that the S_{max} values obtained theoretically from the slope are in accordance with the equilibrium swelling percentage calculated experimentally. When the number of polyacrylamides increases, the 'S_{max}' and 'Ri' were found to be decreased. This may be due to the formation of IPN, which resist the solvent uptake indicating a good interaction existing between natural rubber and polyacrylamide. Considering the glutaraldehyde variation, a decrease in S_{max} value is observed with an increase in the glutaraldehyde concentration, which can be attributed to the intense vulcanization taking place in natural rubber due to the increased concentration of glutaraldehyde.

As crosslinking increases in a structure, it will resist the uptake of solvent through the vulcanized polymer. The S% and percentage equilibrium swelling values were found to be comparable to each other in all systems, which clearly represents that by increasing the amount of PAAm and concentration of glutaraldehyde uptake capacity is decreased. This is due to the formation of more network and highly crosslinked structural patterns in the system, which prevents the approach of more solvent particles. But in case of NR/PAAm5, swelling

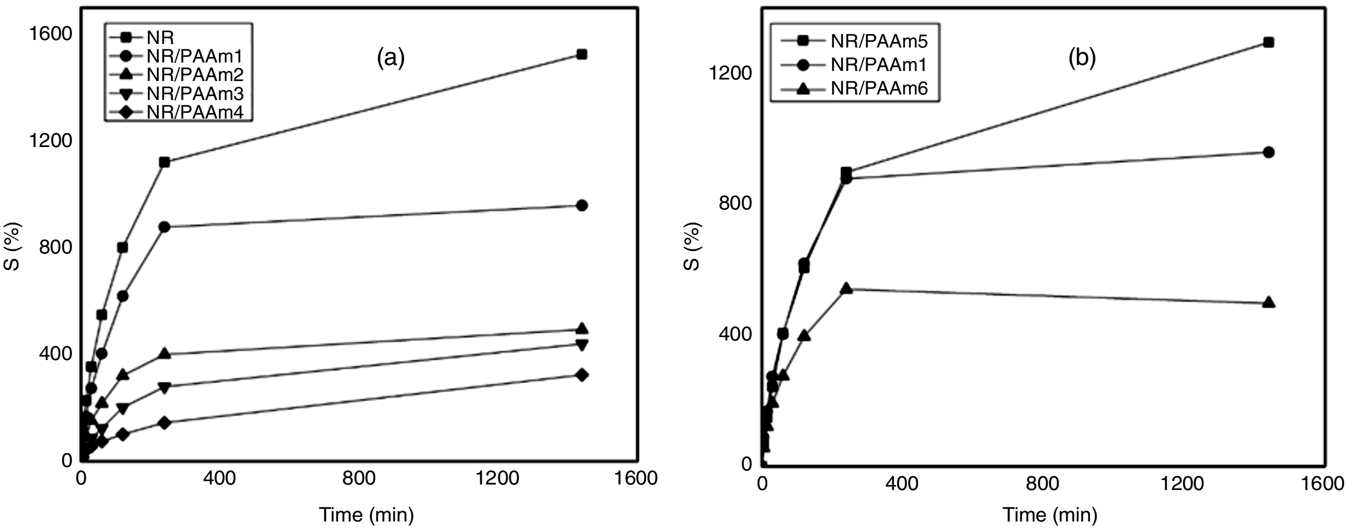

Fig. 10. Swelling percentage as a function of (a) polymer composition and (b) glutaraldehyde concentration

TABLE-2
SOME SWELLING PARAMETERS OF NR/PAAm SEMI-IPNs

Particulars	NR	NR/ PAAm1	NR/ PAAm2	NR/ PAAm3	NR/ PAAm4	NR/ PAAm5	NR/ PAAm6
S (%)	1525	959	494	440	324	1294	498
ESC (%)	93.84	90.56	83.17	81.48	76.43	92.83	83.26
Swelling rate (Ri)	0.1607	0.1469	0.0764	0.0347	0.0173	0.1044	0.1396
Maximum equilibrium swelling (S_{max}, %)	1628	1007	516	479	363	1412	513
Swelling exponent (n)	0.47	0.46	0.42	0.50	0.51	0.54	0.42
Swelling coefficient (K)	0.69	0.54	0.33	0.15	0.09	0.36	0.41
Diffusion coefficient (D) × 10^{-9}	5.85	3.61	4.01	4.22	4.79	5.45	3.71

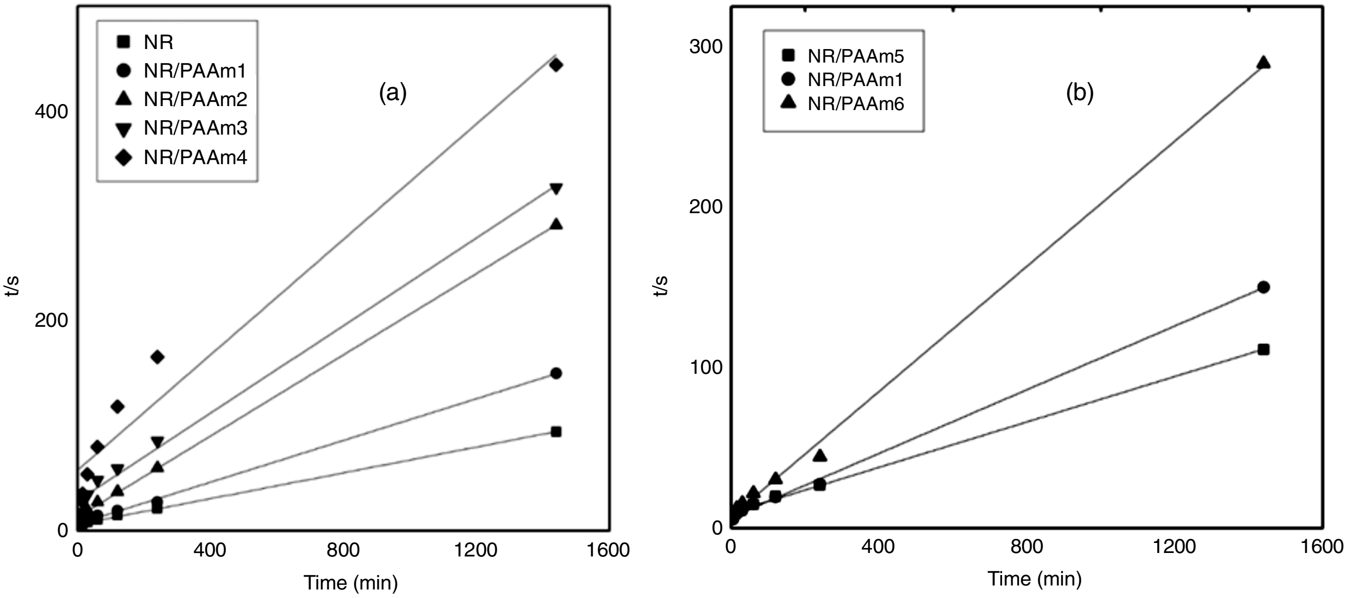

Fig. 11. Swelling rate curves as a function of (a) polymer composition and (b) glutaraldehyde concentration

rate is decreased compared to NR/PAAm1. By comparing NR/PAAm1 and NR/PAAm6, it is observed that as glutaraldehyde concentration increases, the swelling rate decreases, indicating that more crosslinking in the system and thus less solvent movement.

Diffusion studies: The capacity of penetrant to transport among the polymer chain segment is denoted by diffusion coefficient. Diffusion coefficient of pure NR and semi-IPN can be obtained by the following method, which is applicable only for first 60% of the swelling. In this method, the diffusion coefficient can be derived from the slope of the W_t/W_∞ plot based on eqn. 2 [31]:

$$\frac{W_t}{W_\infty} = \frac{4}{d}\left(\frac{Dt}{\pi}\right)^{1/2} \tag{2}$$

where 'W_t' is the solvent uptake of the sample at time t, 'W_∞' is the equilibrium solvent uptake, 'd' is the thickness of unswollen film; 'D' is the diffusion coefficient and 't' is the time. Fig. 12 showed a sudden decrease in the diffusion coefficient value from NR to NR/PAAm1. This is clear evidence of the strong interaction existing between two polymers, which prevents the transport of solvent molecules through the blend composition. This may be due to the formation of stable IPN in that composition. But a gradual increase in the diffusion coefficient value is observed from NR/PAAm1 to NR/PAAm4, but its

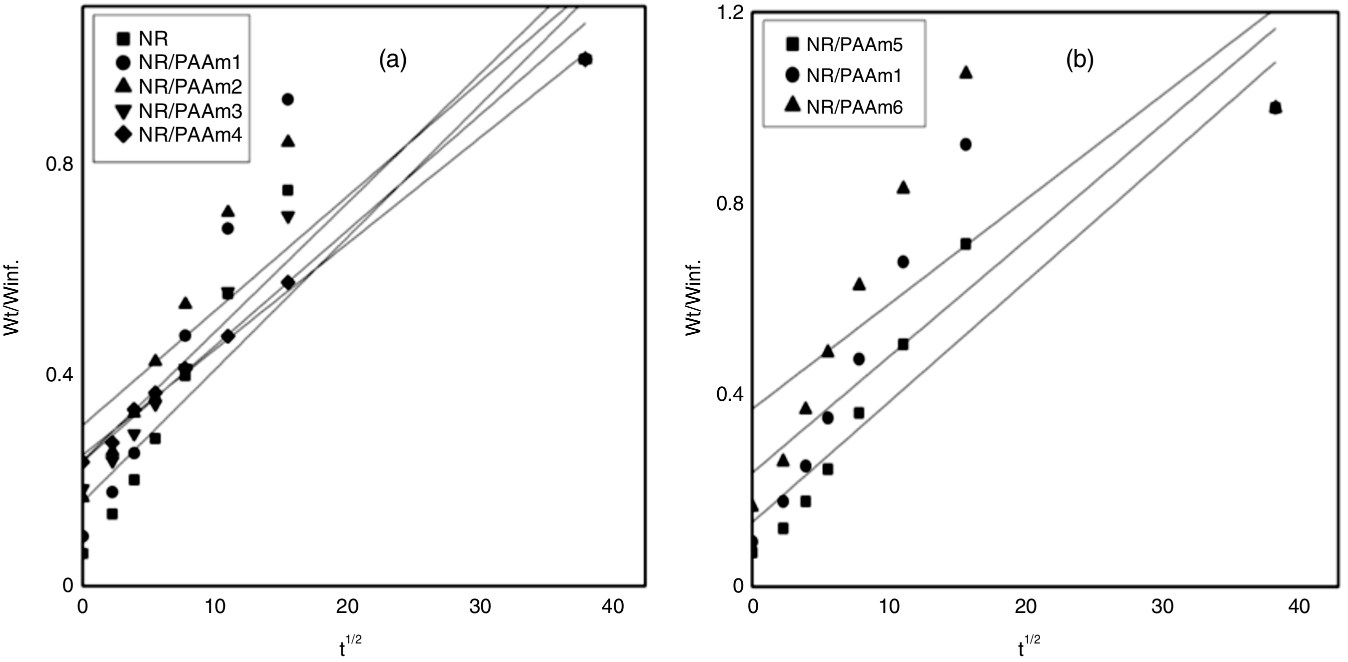

Fig. 12. Diffusion curves as a function of (a) polymer composition and (b) glutaraldehyde concentration

value is lower than the pure NR. Though IPN formation is taking place in such systems, it may not be as stable as NR/PAAm1. Acrylamide being hydrophilic in nature; as its concentration increases, in some places polymer may get degrade or get dissolved by moisture uptake allowing wider space for the solvent to transport through the polymer segments. As the amount of glutaraldehyde increases, a decrease in the D value is observed from NR/PAAm5 to NR/PAAm1 because of the vulcanization in the rubber chain. As chains become more crosslinked, the easy diffusion of penetrants through it will be highly constrained. But a slight increase in the D value was also observed for NR/PAAm6, which may be attributed due to the complete crosslinking didn't occur.

Swelling mechanism: In order to study the diffusion mechanism, the diffusion parameters were computed from the swelling values using eqn. 3 [32]:

$$F = \left(\frac{W_t - W_o}{W_o}\right) = Kt^n \tag{3}$$

where F is the swelling power of the system; K represents the swelling constant and 'n' is the swelling exponent, which explains the diffusion mechanism of penetrant into the IPN network. Ln F *versus* ln t graphs were plotted for pure NR and for NR/PAAm semi IPN systems and are represented in Fig. 13. The slope and intercept of the plotted line give 'n' and 'K' values respectively and are shown in Table-2.

In the semi-IPNs, the values of 'n' lie between 0.42 and 0.54, which suggests that there may be variation in the intertwined network structure that formed between the two polymeric chains and this alters the diffusion behaviour of the system. For NR/PAAm3, n = 0.5 indicates the Fickian nature of the diffusion process. Here, the diffusion of solvent molecules happens at a much slower rate compared to the speed at which polymer chain relaxation occurs. But for NR/PAAm4 'n' value is 0.51, indicating the anomalous behaviour. A similar trend is observed in NR/PAAm5 too. Both these cases show that the rate at which solvent diffusion occurs and the rate at which polymer relaxation happens are comparable. In NR/PAAm1 and in NR/PAAm6, the 'n' value is less than 0.5. From this, it can be inferred that due to vulcanization, the interaction between the solvent and the IPN is highly reduced.

The value of K indicates the structural features of the polymer as well as its interaction with the solvent molecules. From NR to NR/PAAm4, a decrease in the K value shows the interaction between natural rubber and polyacrylamide is diminishing as the amount of PAAm increases. These values are in accordance with the diffusion coefficient results attained earlier. While comparing the K values as a function of glutaraldehyde, it was observed that maximum interaction occurred in the semi-IPN systems as the concentration of crosslinking agents increased. But a sudden decrease in 'K' value is observed for NR/PAAm6, which matches with the increased 'D' value of that particular system. Also, when glutaraldehyde concentration becomes high, the system may get deformed and leads to the easy flow of the solvent.

Conclusion

In this work, a semi-interpenetrating polymer networks films were prepared by blending natural rubber and polyacrylamide using glutaraldehyde as crosslinking agent by varying the blend ratios and the concentration of glutaraldehyde. The mechanical properties of networks were found to depend on the concentration of component polymers and the extent of crosslinking. While the natural rubber surface shows a homogeneous texture, whereas the surface of NR/PAAm semi-IPN

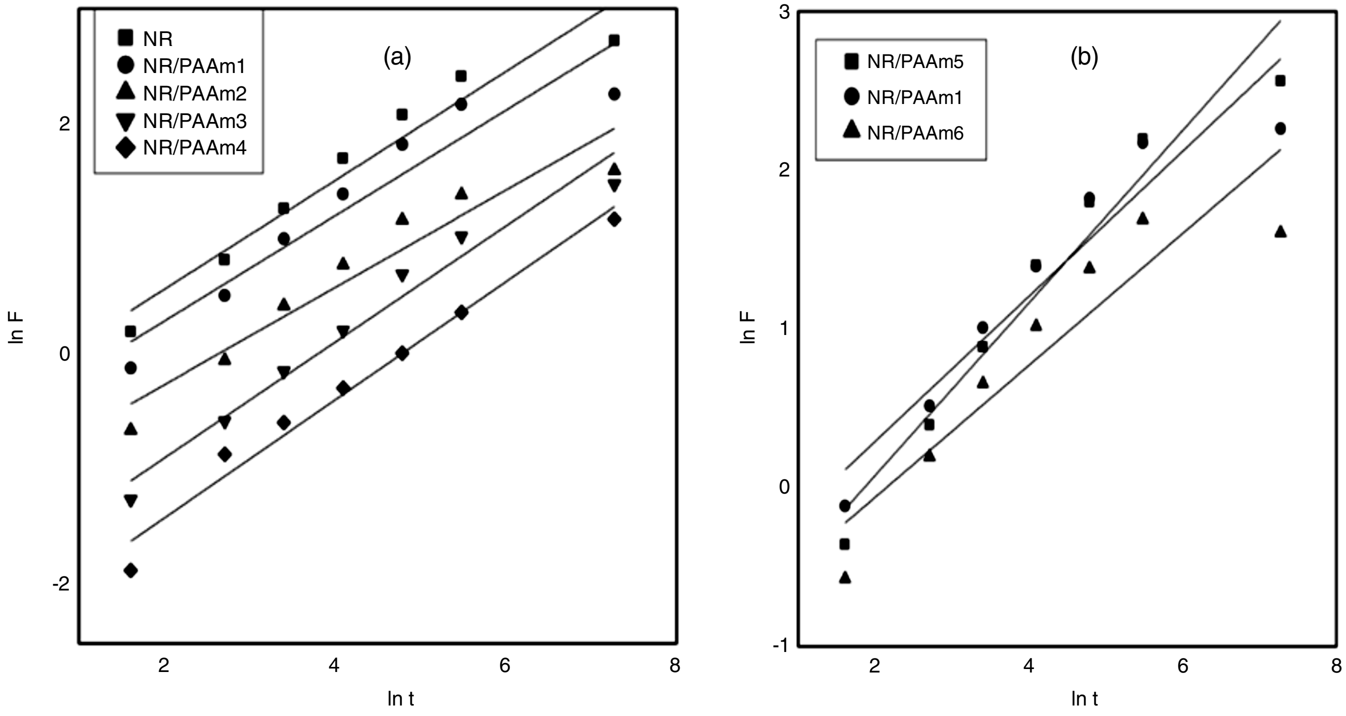

Fig. 13. Swelling kinetic curves as a function of (a) polymer composition and (b) glutaraldehyde concentration

has a heterogeneous appearance. It was observed that when the percentage of PAAm increases in the semi-IPN mechanical properties were found to be decreased. At the same time, all these properties vary in direct proportion with the increase in glutaraldehyde concentration from 1 to 3%. In both cases, the Young's modulus values increase to a maximum and then decrease. This shows the extent of rigidity of a material. The swelling studies were conducted by taking toluene as the solvent. It was observed that % swelling follows the order NR > NR/PAAm1 > NR/PAAm2 > NR/PAAm3 > NR/PAAm4 respectively. From kinetic studies, it was observed that the swelling process happens at second order kinetics. In diffusion studies, a sudden decrease in the diffusion coefficient value from NR to NR/PAAm1 can be observed. This is clear evidence of the strong interaction existing between two polymers which prevents the transport of solvent molecules through the blend composition.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

REFERENCES

1. L.H. Sperling, *J. Polym. Sci. Macromol. Rev.*, **12**, 141 (1977); https://doi.org/10.1002/pol.1977.230120103
2. S. Goswami and J.K. Ranjan, *Fibers Polym.*, **21**, 1096 (2020); https://doi.org/10.1007/s12221-020-9338-5
3. Y. Nie, G. Huang, L. Qu, P. Zhang, G. Weng and J. Wu, *J. Appl. Polym. Sci.*, **115**, 99 (2010); https://doi.org/10.1002/app.31045
4. A. Mathew, *J. Membr. Sci.*, **201**, 213 (2002); https://doi.org/10.1016/S0376-7388(01)00738-4
5. N.C. Liu and W.E. Baker, *Adv. Polym. Technol.*, **11**, 249 (1992); https://doi.org/10.1002/adv.1992.060110403
6. J. Johns and V. Rao, *Int. J. Polym. Anal. Charact.*, **13**, 280 (2008); https://doi.org/10.1080/10236660802190104
7. C.-J. Tung and T.-C.J. Hsu, *J. Appl. Polym. Sci.*, **46**, 1759 (1992); https://doi.org/10.1002/app.1992.070461007
8. A.A. Donatelli, L.H. Sperling and D.A. Thomas, *Macromolecules*, **9**, 671 (1976); https://doi.org/10.1021/ma60052a029
9. S.S. Bird, D. Clary, K.C. Jajam, H.V. Tippur and M.L. Auad, *Polym. Eng. Sci.*, **53**, 716 (2013); https://doi.org/10.1002/pen.23305
10. V.J. Dave and H.S. Patel, *J. Saudi Chem. Soc.*, **21**, 18 (2017); https://doi.org/10.1016/j.jscs.2013.08.001
11. K. Dean, W.D. Cook, M.D. Zipper and P. Burchill, *Polymer*, **42**, 1345 (2001); https://doi.org/10.1016/S0032-3861(00)00486-9
12. W. Wichaita, D. Promlok, N. Sudjaipraparat, S. Sripraphot, T. Suteewong and P. Tangboriboonrat, *Eur. Polym. J.*, **159**, 110740 (2021); https://doi.org/10.1016/j.eurpolymj.2021.110740
13. K. Simma, G.L. Rempel and P. Prasassarakich, *Polym. Degrad. Stab.*, **94**, 1914 (2009); https://doi.org/10.1016/j.polymdegradstab.2009.08.005
14. A.P. Mathew, S. Packirisamy and S. Thomas, *Polym. Degrad. Stab.*, **72**, 423 (2001); https://doi.org/10.1016/S0141-3910(01)00042-8
15. J. Han, K. Lu, Y. Yue, C. Mei, C. Huang, Q. Wu and X. Xu, *Ind. Crops Prod.*, **128**, 94 (2019); https://doi.org/10.1016/j.indcrop.2018.11.004
16. J. Johns and V. Rao, *Int. J. Polym. Mater.*, **60**, 766 (2011); https://doi.org/10.1080/00914037.2010.551361
17. A. Andrio, V. Compañ, R.C. Reis-Nunes, M.L. López and E. Riande, *J. Membr. Sci.*, **178**, 65 (2000); https://doi.org/10.1016/S0376-7388(00)00476-2
18. H. Ismail and Suryadiansyah, *J. Polym. Test.*, **21**, 389 (2002); https://doi.org/10.1016/S0142-9418(01)00101-5
19. S. Mohanty, G.B. Nando, K. Vijayan and N.R. Neelakanthan, *Polymer*, **37**, 5387 (1996); https://doi.org/10.1016/S0032-3861(96)00391-6
20. N.R. Choudhury, T.K. Chaki, A. Dutta and A.K. Bhowmick, *Polymer*, **30**, 2047 (1989); https://doi.org/10.1016/0032-3861(89)90292-9
21. R. Asaletha, M.G. Kumaran and S. Thomas, *Polym. Degrad. Stab.*, **61**, 431 (1998); https://doi.org/10.1016/S0141-3910(97)00229-2
22. T. Tanaka, *J. Phys. Rev. Lett.*, **40**, 820 (1978); https://doi.org/10.1103/PhysRevLett.40.820
23. R. Promsung, Y. Nakaramontri, N. Uthaipan, C. Kummerloewe, J. Johns, N. Vennemann and E. Kalkornsurapranee, *Express Polym. Lett.*, **15**, 308 (2021); https://doi.org/10.3144/expresspolymlett.2021.27
24. H. Dweik, W. Sultan, M. Sowwan and S. Makharza, *Int. J. Polym. Mater.*, **57**, 228 (2008); https://doi.org/10.1080/00914030701413280
25. W.M. Leung, D.E. Axelson and J.D. Van Dyke, *J. Polym. Sci. A Polym. Chem.*, **25**, 1825 (1987); https://doi.org/10.1002/pola.1987.080250711
26. J.J. Maurer, D.N. Schulz, D.B. Siano and J. Bock, eds.: In: J.F. Johnson and P.S. Gill,, Thermal Analysis of Acrylamide-Based Polymers, Analytical Calorimetry, Springer: Boston, USA (1984).
27. F. Riahi, D. Benachour and A. Douibi, *Int. J. Polym. Mater.*, **53**, 143 (2004); https://doi.org/10.1080/00914030490267564
28. A. Thitithammawong, C. Nakason, K. Sahakaro and J. Noordermeer, *Polym. Test.*, **26**, 537 (2007); https://doi.org/10.1016/j.polymertesting.2007.02.002
29. J. Johns and C. Nakason, *Polym. Plast. Technol. Eng.*, **51**, 1046 (2012); https://doi.org/10.1080/03602559.2012.689053
30. M. Schneider, T. Pith and M. Lambla, *Polym. Adv. Technol.*, **6**, 326 (1995); https://doi.org/10.1002/pat.1995.220060511
31. M. Sen and O. Guven, *Eur. Polym. J.*, **38**, 751 (2002); https://doi.org/10.1016/S0014-3057(01)00240-3
32. E. Karadag, O.B. Uzum, D. Saraydin and O. Güven, *Int. J. Pharm.*, **301**, 102 (2005); https://doi.org/10.1016/j.ijpharm.2005.05.026