Journal of Macromolecular Science, Part A
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713597274

Study on Mechanical Properties of Photocured Films of Chitosan/PVA and PEO/PVA Blend with Acrylic Monomers

Shaheda Ferdous[a]; A. I. Mustafa[a]; Mubarak A. Khan[b]
[a] Department of Applied Chemistry and Chemical Technology, University of Dhaka, Dhaka, Bangladesh [b] Radiation and Polymer Chemistry Laboratory, Institute of Nuclear Science and Technology, Bangladesh Atomic Energy Commission, Dhaka, Bangladesh

To cite this Article Ferdous, Shaheda , Mustafa, A. I. and Khan, Mubarak A.(2003) 'Study on Mechanical Properties of Photocured Films of Chitosan/PVA and PEO/PVA Blend with Acrylic Monomers', Journal of Macromolecular Science, Part A, 40: 8, 817 — 832

To link to this Article: DOI: 10.1081/MA-120022273

URL: http://dx.doi.org/10.1081/MA-120022273

JOURNAL OF MACROMOLECULAR SCIENCE®
Part A—Pure and Applied Chemistry
Vol. A40, No. 8, pp. 817–832, 2003

Study on Mechanical Properties of Photocured Films of Chitosan/PVA and PEO/PVA Blend with Acrylic Monomers

Shaheda Ferdous,[1] A. I. Mustafa,[1] and Mubarak A. Khan[2,*]

[1]Department of Applied Chemistry and Chemical Technology, University of Dhaka, Dhaka, Bangladesh
[2]Radiation and Polymer Chemistry Laboratory, Institute of Nuclear Science and Technology, Bangladesh Atomic Energy Commission, Dhaka, Bangladesh

ABSTRACT

Natural polymer chitosan acts as a bioblending material, which was obtained from dried prawn shell waste through the preparation of chitin and was characterized. Bioblend films of polyvinyl alcohol (PVA)/chitosan and PVA/polyethylene oxide (PEO) were prepared. Mechanical properties like tensile strength (TS), elongation at break (Eb) of chitosan film were studied. Five formulations were developed with 2-ethyl 2-hydroxy methyl 1,3-propandiol tri-methacrylate (EHMPTMA), a tri-functional acrylic monomer and 2-ethylhexyl acrylate (EHA), a mono-functional acrylic monomer in the presence of photoinitiator Darocur-1664 (2%). The films were soaked in those monomer formulations in dissimilar soaking times and irradiated under UV-radiation at different radiation intensities for the improvement of the properties of chitosan film. The cured films were then subjected to various characterization tests like TS, Eb, polymer loading (PL), water uptake, gel content etc. The formulation, containing 25% EHMPTMA and 73% EHA showed the best performance at 10th UV passes of UV radiation for 4 min soaking time. The bioblend film PVA/chitosan performed better physico-mechanical properties rather than PVA/PEO.

---

*Correspondence: Mubarak A. Khan, Radiation and Polymer Chemistry Laboratory, Institute of Nuclear Science and Technology, Bangladesh Atomic Energy Commission, P.O. Box 3787, Dhaka, Bangladesh; E-mail: makhan@bangla.net.

DOI: 10.1081/MA-120022273 1060-1325 (Print); 1520-5738 (Online)

www.dekker.com

Marcel Dekker, Inc.
270 Madison Avenue, New York, New York 10016

Key Words: Chitosan; Photocuring; Acrylic monomer; UV radiation.

INTRODUCTION

The field of photocuring has been developed to an important branch of photocrosslinking of polymers.[1] Photocrosslinkable polymers possess functional groups, which can undergo light-induced reactions to form directly a crosslinked polymer. This means that light irradiation polymers carrying more than two reactive groups per chain (or, of blend with a photoinitiator or photostabilizer) initiates crosslinking to a three-dimensional network structure.[2] The photoinitiator performs widespread application in the case of this photoinduced polymerization. Excitation of the photoinitiator under UV radiation yields reactive radicals that initiate subsequent reactions in the polymer bioblend and produce crosslinks between two macromolecular chains.[2] Chitin is the second most abundant polymer in nature, is a linear polysaccharide composed of 2-acetamido-2-deoxy-*D*-glucosidic bonds. The principal derivative of chitin is chitosan, generally produced by alkaline deacetylation of chitin. Its primary structure corresponds to a linear chain of β-1-4 linked 2-amino-2-deoxy-*D*-glucopyranose residue.[3,4] Chitin and chitosan have a different field of application and are biodegradable in nature. Chitin is prepared from waste prawn shell. Bangladesh, the largest delta country in the world, has a 724 km long coastal border out of a 4719 km border of Bangladesh. It has 367 km (200 nautical miles) and about a 20 km (12 nautical miles) economical and political coastal belt. Moreover, Bangladesh is a riverine country, prawn (*Penaeus monodon*) is the most abundant, containing highly natural polymer. In Bangladesh, the Mangrove area, near the Sundarban forest is suitable for the prawn cultivation and most of the prawn hatcheries are found in these localities. There are 23,423 prawn farms in 25 districts of Bangladesh. Generally, every year 758,000 MT of prawn is found from these firms.[5] Blends of biopolymer with common polymers are of great significance with expanding application to biochemical and biodegradable materials.[6] Polyvinyl alcohol (PVA) is a water-soluble polymer, which produces a crosslinked polymer under radiation curing in the molten and aqueous state.[6] But, in the solid phase at about 25°C, it is crosslinked. On the other hand, chitosan and polyethylene oxide (PEO) produces a crosslinked polymer under UV radiation in every state.[6] When PVA is mixed with chitosan (bioblend of PVA and chitosan), it becomes crosslinked under UV radiation treatment[6] and chitosan/PLA blend also crosslinked under UV radiation.[7] Multifunctional vinyl unsaturated monomer was used in the treatment of the polymer film to reduce the UV radiation intensities to obtain optimum properties or to achieve an increased cure state at the same UV radiation intensities.[8] In this respect, 2-ethyl 2-hydroxy methyl 1,3-propandiol tri-methacrylate (EHMPTMA) was used as a tri-functional acrylic monomer which makes the film character harder and more brittle. To avoid these difficulties, a mono-functional acrylic monomer 2-ethylhexyl acrylate (EHA) was mixed with EHMPTMA at a different proportion, which leads to the softer polymer and creates some flexibility. 2-Ethyl 2-hydroxy methyl 1,3-propandiol tri-methacrylate is more effective in producing a denser network than EHA.[9] Several works on the polymerization of acrylic monomers in polymer matrix have appeared recently.[10–14] The present report deals with the preparation of chitin and chitosan, as well as PVA/chitosan and PVA/PEO blends. Thin films were prepared using these blends and treated with different monomer (EHMPTMA and EHA) formulations, in different soaking times and cured under UV radiation at different intensities. Films prepared by a PVA/chitosan blend found better results as compared to that of a PVA/PEO blend through the enhanced physical and mechanical properties of the films.

EXPERIMENTAL

Materials

Polyvinyl alcohol, PVA-15000 was obtained from Fluka, Switzerland. Polyethylene oxide, PEO was obtained from BDH, UK. Prawn shell, from which chitosan was extracted, was collected from Mongla (near the Sundarban forest) in Bangladesh. Sodium hydroxide (NaOH) was supplied by Merck, Germany and hydrochloric acid (HCl) was supplied by BDH, United Kingdom. Ethanoic acid (CH_3COOH), purity 99.9%, EHMPTMA and EHA (2-ethylhexyl acrylate) and Photoinitiator, Darocur-1664 were used and obtained from Merck, Germany.

Method

Chitosan was extracted from prawn shell waste. The collected waste prawn shell was washed with hot water and dried in an oven at 105°C for 72 h. Dried prawn shell (about 100 g) was ground (40–60 mesh) using a blender. Ground prawn shell was deprotenized with 3% NaOH and demineralized with 3% HCl. The mixture was washed with distilled water by decantation and filtration to neutralize and dried in an oven at 105°C for 24 h. Thus, produced chitin (77 g) is an intermediate product of chitosan. Chitosan was obtained by deacetylation of chitin with 50% NaOH where chitin: NaOH = 1 : 20 (w/w) at 100°C for 3 h. After this process, the solid separated from the alkali layer was extensively washed with distilled water to remove traces of alkali. The resultant solid was dried in vacuum oven at 50°C for 24 h. Chitosan (27 g) was extracted in this way from prawn shell waste. The chitosan solution (2%) was prepared in a 2% ethanoic acid (CH_3COOH) aqueous solution. The PVA/chitosan blend, containing PVA and chitosan in the film form named Z1, Z2, and Z3 were prepared by mixing an aqueous solution of PVA with a chitosan solution in different ratios. Polyvinyl alcohol/PEO blend, containing PVA and PEO in the film form named Z4, Z5, and Z6 were prepared by mixing an aqueous solution of PVA with PEO in different ratios, which are given in the Table 1. The films were prepared by a casting method on silicon cloth. The mechanical properties of the films were investigated. Tensile properties (TS and Eb) of the film were measured by Universal Testing Machine (INSTRON, model 1011, UK). The load capacity was 500 N, efficiency was within ±1%, the cross-head speed was $2\,\mathrm{mm\,min^{-1}}$ and gauze length was 20 mm. Five formulations named F1 to F5 were developed with two monomers, EHMPTMA and EHA in the presence of the photoinitiator, Darocur-1664 (2%). Their compositions are given in Table 2. These films were soaked in these monomer formulations for different soaking times and irradiated under UV radiation, using UV mincure (IST Technik, Germany). The intensity of the lamp was 2 kW at 9.5 amp current and the wavelength was 254–313 nm with a conveyor speed of $4\,\mathrm{m\,min^{-1}}$. The efficiency of the machine was within ±1%. The films were washed with acetone in the case of separation of homopolymers from the blends and this may be the convenient method for the separation

Table 1. Different blending composition (% w/w).

	Compositions (%)			Properties	
Films	Chitosan	PEO	PVA	TS (MPa)	Eb (%)
Z1 (=A)	14.29	—	85.71	3.5	23
Z2	25.0	—	75.0	3.1	20.2
Z3	50.0	—	50.0	2.8	18
Z4 (=B)	—	14.29	85.71	3.0	11.5
Z5	—	25.0	75.0	1.8	10.0
Z6	—	50.0	50.0	1.0	12.1

of homopolymers. After 24 h, the cured films were then subjected to various characterization tests. Polymer loading (PL) of the monomer formulation with the film were determined on the basis of weight gained by the film after the entire treatment process.

RESULTS AND DISCUSSION

Most of the data presented in the report are of average values of at least five samples and the results obtained are within the accuracy of ±1%.

Optimization of Chitosan and Polymer Ratio

The film prepared from the polymer blend Z1, containing chitosan (14.29%) and PVA (85.71%) in the ratio of 1 : 6 showed the best performance in case of chitosan/PVA film (Table 1) as compared to blends Z2 and Z3. The films Z2 and Z3 possess brittle character, even though they have almost similar properties. The film prepared from the polymer blend Z4, containing PEO (14.29%) and PVA (85.71%) in the ratio of 1 : 6 showed the best performance in case of PEO/PVA film (Table 1) as compared to Z5 and Z6. The films Z5 and Z6 possess brittle character as compared to Z4. That is why for later investigation of the work, 1 : 6 ratio of chitosan and PEO/PVA is optimized. For better understanding, Z1 and Z4 are named A and B, respectively.

Table 2. Composition of different monomer formulation (w/w).

	Monomer compositions (%)		
Formulations	EHMPTME	EHA	Photoinitiator Darocur-1664 (%)
F1	—	98	2
F2	25	73	2
F3	50	48	2
F4	75	23	2
F5	98	—	2

Characterizations of Chitin and Chitosan by FTIR

The prepared chitin and chitosan were characterized by infrared spectrum in a range of 3500–500 cm^{-1} and absorbency at 2870 cm^{-1} (the C—H) and 1550 cm^{-1} (the amide bond) were evaluated. In chitin, both N—H (1550 cm^{-1}) and C=O (1650 cm^{-1}) bonds are present, whereas in chitosan C=O bond was found to be absent.

Characterizations of Polyvinyl Alcohol/Chitosan and Polyvinyl Alcohol/Polyethylene Oxide Bioblend Films

Various mechanical properties, like TS, Eb were investigated. Tensile strength of the PVA/chitosan bioblend film was 3.5 MPa and Eb was 23%, while TS of the PVA/PEO bioblend film was 4 MPa and Eb was 11.5%.

Optimization of Monomer Formulation

Monomer formulation plays an important role because it affects the polymerization rate and overall conversion, as well as the properties of the crosslinked polymerization.[15] Triacrylate monomer possesses more cure speed than a monoacrylate monomer.[16]

The hardness of the UV-cured polymer film depends on the functionality of the acrylic monomer; but the polymer becomes brittle and twisted. On the other hand, a monoacrylate monomer posseses lower density, which leads to a softer polymer and imparts some flexibility. Only PVA film that exists in a solid phase is not capable of being crosslinked with various monomers. Polyethylene oxide and chitosan are capable of producing crosslinked polymers with various monomers.

The films A and B were soaked in different formulations (F1–F5) for 4 min soaking time to optimize a definite formulation for curing of the bioblend films. After soaking, the films were cured under UV radiation at different intensities (2, 4, 6, 8, and 10 passes). After 24 h of radiation, the samples were subjected to various physical and mechanical properties were determined.

Polymer Loading

The results of PL values of the cured bioblend films are represented in Figs. 1 and 2, respectively, against the number of UV radiation passes as a function of monomer formulation for 4 min soaking time. It is observed from both figures that, the highest PL is found to be 25% for A at the 6th passes and 19% for B at the 8th passes of UV radiation with the treatment of monomer formulation M2, containing 25% EHMPTMA, 73% EHA, and 2% photoinitiator, Darocur-1664 at the 10th UV passes for 4 min soaking time. The percentage of PL increases with EHMPTMA, a denser tri-functional monomer, up to 25%, but more than 25% EHMPTMA, reduce the curing of bioblend films. At low concentration, a multifunctional vinyl monomer like EHMPTMA promotes a rapid free radical induced propagation reaction with the help of a photoinitiator leading to a network polymer structure through curing via their double bonds.[17] At about 25%, EHMPTMA concentration, the amount of residual unsaturation increases with the consequence of a faster rate of formation of three dimensional network structure, causing restricted

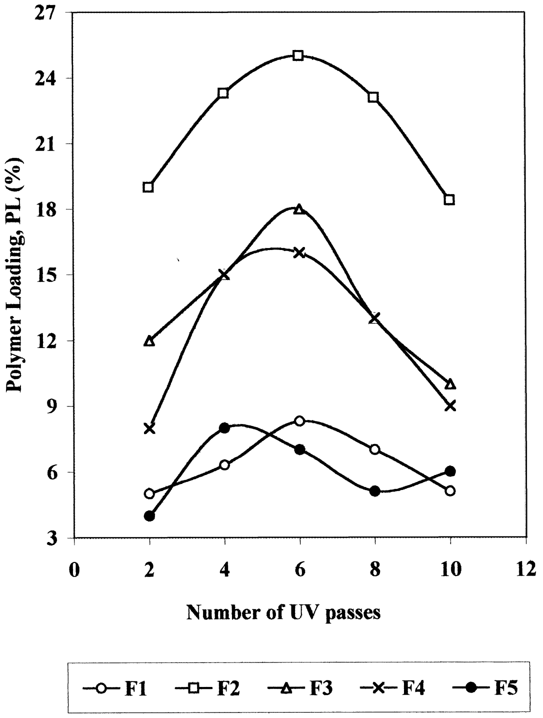

Figure 1. Polymer loading of the treated PVA/chitosan film against the number of passes (UV doses) with respect to monomer formulation for 4 min soaking time.

mobility.[18] The decrease in PL values at higher EHMPTMA concentration may be the reason that the radical–radical recombination process may be dominating, thus creating a homopolymer rather than monomer-chitosan backbone reaction, where the hydroxyl and/or amino group of chitosan may react with vinyl radicals in EHMPTMA in the case of bioblend A and monomer-PEO backbone reaction in the case of B.[19] In most of the cases, the PL value increases with initial UV radiation doses, attains a maximum at the 6th pass for A and 8th pass for B and then decreases. This may be due to the radiation degradation of the bioblend film at higher UV passes.[20]

Tensile Properties

Tensile strength is very important in selecting diverse application of the polymer. The results of the TS values are shown in Figs. 3 and 4 for bioblends A and B, respectively, where TS values are plotted against a number of UV passes as a function of monomer formulation for a 4 min soaking time. In these figures, it is observed that the highest TS value for A was19.7 MPa at the 6th UV pass and for B 14.0 MPa at the 8th pass with the monomer formulation F2, containing 25% EHMPTMA, 73% EHA, and 2% photoinitiator, Darocur-1664. The TS values increases with the number of UV passes up to the 6th pass for A and, the 8th pass for B beyond which they decrease as the radiation passes are

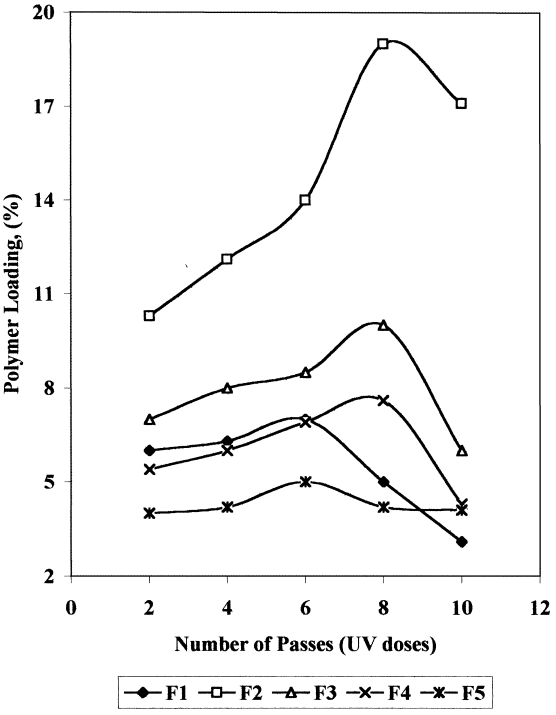

Figure 2. Polymer loading of the treated PVA/PEO film against the number of passes (UV doses) with respect to monomer formulation for 4 min soaking time.

increased. It is obvious that after reaching the maximum, the value TS falls. This result implies that the maximum TS is achieved at particular radiation doses, further radiation demolished the TS of the bioblend films due to the radiation degradation of the films. The mechanism of the degradation is not clearly known, however, it could be the cleavage of the chain due to the overdose of the UV radiation.

With the vinyl monomer treatment, the TS value of the treated bioblend films increase may be due to the crosslinking of the hydroxyl and/or amino group in chitosan with vinyl group forming three dimensional network structures causing restricted mobility in the case of A and PEO-monomer crosslinked reaction in the case of B. The bioblend films become brittle, twisted, and shrinkage occurs after 25% EHMPTMA concentration and the TS decreases. This may be due to the fact that the homo-polymerization reaction between the EHMPTMA and EHMPTMA radical is dominant and the reaction of chitosan with EHMPTMA for A and the reaction of PEO with EHMPTMA for B is less prominent.

Elongation at Break

Flexibility and elasticity of the bioblend films are essential parameters that are very actively considered during the application of the polymer. These two phenomena are related to the elongation at break of the bioblend film. The results of Eb are plotted in Fig. 5 for A

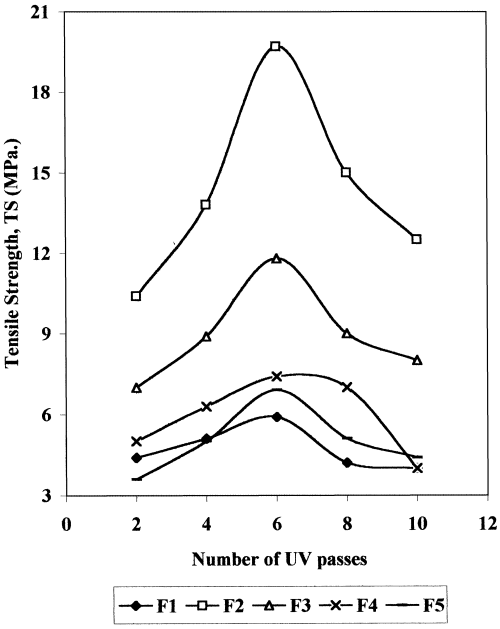

Figure 3. Tensile strength of the treated PVA/chitosan film against the number of passes (UV doses) with respect to monomer formulation for 4 min soaking time.

and Fig. 6 for B against the number of passes as a function of monomer composition for 4 min soaking time. The maximum Eb for A was 74% at the 6th pass and for B was 48.5% at the 8th pass observed with the monomer formulation F1, containing 98% EHA. This is due to the fact that the low density mono functional monomer like EHA results in the film becoming flexible and softer. While the formulation F2, containing 25% EHMPTMA, 73% EHA, the Eb is not maximum and the value of Eb for A was 67% at the 6th pass and for B was 38% at the 8th pass. There is a rise in elongation at the initial stage of UV radiation like the TS value. The Eb value increases with the number of UV passes and reaches a maximum, after that, it decreases due to the radiation degradation of the film. Tensile strength is achieved in the overall crosslinking network within the cured film. 2-Ethyl 2-hydroxy methyl 1,3-propandiol tri-methacrylate gives the highest TS up to a certain limit, and after this, produces a brittle film that cracks easily during stretching. On the other hand, EHA increases high elasticity and creates some flexibility. Therefore, it is clear that TS and Eb are very much dependent on the nature of the formulation, where the monomer is mono functional or multifunctional. The combination of EHMPTMA and EHA at the ratio of 25 and 73 (F2), yields suitable conditions for better crosslinking phenomenon at the equilibrium condition that creates the chitosan film with the highest TS and moderate Eb.

Optimization of Soaking Time

On having the highest TS and PL at F2 formulation, the bioblend films (A and B) were soaked in F2 for different soaking times (1, 2, 3, 4, and 5 min), and then irradiated

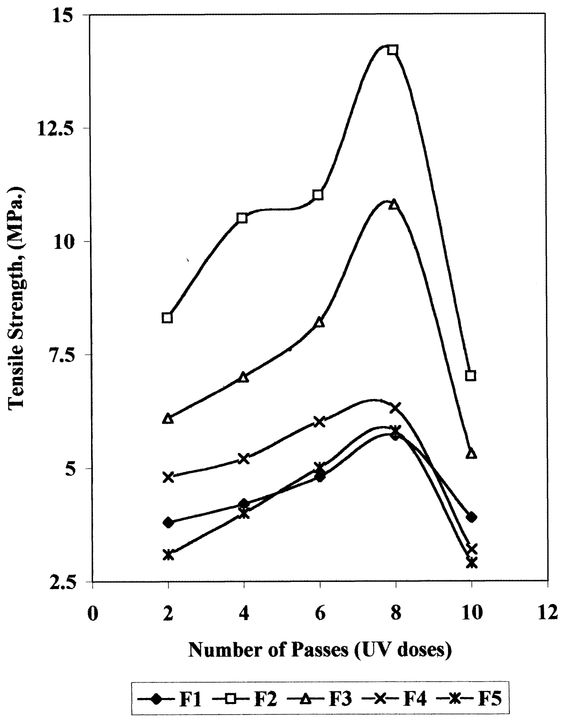

Figure 4. Tensile strength of the treated PVA/PEO film against number of passes (UV doses) with respect to monomer formulation for 4 min soaking time.

under UV radiation with a different number of passes (2, 4, 6, 8, and 10). After 24 h, of UV radiation, PL, TS, and Eb were investigated.

Polymer Loading

The results are graphically represented in the Figs. 7 and 8, where PL values are plotted against a number of UV radiation passes as a function of soaking time for optimized monomer formulation, F2. It is observed from these figures that the PL values increase with soaking time until it reaches 4 min, and gives maximum value of 25% at the 6th UV pass for A, and 19% at the 8th pass for B and above 4 min soaking time the PL value decreases with increasing the soaking time. For both cases, soaking increases the cross-section area of the film, at the same time the film surface becomes lustrous. As a result, the monomer can easily diffuse in the bioblend films and may react with hydroxyl and/or amino groups of chitosan and oxygen radical of PEO in low swelling tims. In a higher soaking time, the film becomes twisted, shrinkage occurs, and it looks pale. The PL value increases with initial UV radiation doses, attains the maximum value at the 6th pass for A and the 8th pass for B, and then decreases as the radiation doses increases. This may be caused by the radiation degradation at higher UV doses.

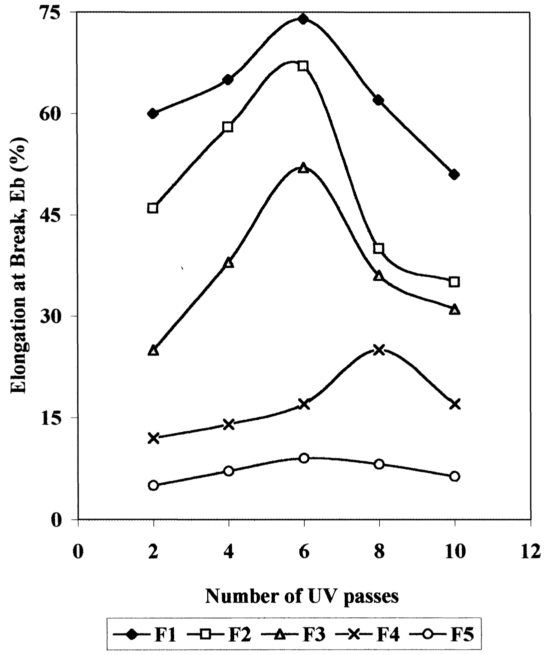

Figure 5. Elongation at break of the treated PVA/chitosan film against the number of passes (UV doses) with respect to monomer formulation for 4 min soaking time.

Tensile Strength

The results of TS values are represented in Figs. 9 and 10, where the TS are plotted against a number of UV passes as a function of soaking time for the F2 formulation. The highest TS value 19.7 MPa for A at the 6th pass of UV radiation and 14.2 MPa for B at the 8th pass. The TS value increases with an increase in soaking time. This may be due to the increased swelling, and leads to an increased diffusion of the monomer into the reaction sites, and the amount of curing increase,[18] hence, the TS value increased. After attaining the maxima, the TS value decreases with an increase in soaking time. The TS value also increases with the increase of number of passes up to the 6th pass for A, and the 8th pass for B, beyond which they decrease as the radiation passes are increased.

Elongation at Break

The Eb values are plotted in Fig. 11 for A, and Fig. 12 for B, against the number of passes as a function of soaking time. For formulation F2, Eb increases with the increase in soaking time and it reaches maxima 67% for A and 38% for B of UV radiation, respectively for 4 min soaking time, the inherent character of the film appears to be superior as compared to other soaking times. So, optimization is established for 4 min soaking time and monomer formulation F2, containing 25% EHMPTMA, 73% EHA, and

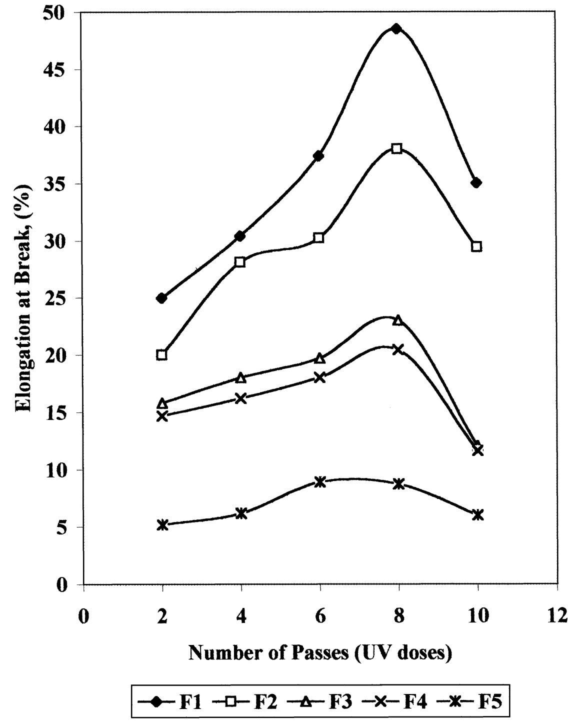

Figure 6. Elongation at break of the treated PVA/PEO film against the number of passes (UV doses) with respect to monomer formulation for 4 min soaking time.

2% Darocur-1664, comparing the physico-mechanical properties of bioblend films of PVA/chitosan (A) and PVA/PEO (B).

Comparative Studies of Bioblend Films Polyvinyl Alcohol/Chitosan and Polyvinyl Alcohol/Polyethylene Oxide

Having established the optimum monomer composition and soaking time for attaining the highest PL and TS values for both bioblend films, it is observed that PVA/chitosan bioblend film shows better physico-mechanical performance than that of the PVA/PEO film. The above mentioned physical and mechanical properties are summarized and represented in Fig. 13 where the highest value of each property is plotted against corresponding bioblend films.

CONCLUSION

Chitin, a natural polymer, was extracted from prawn shell waste. Chitosan was obtained from the extracted chitin. Besides, PVA (not crosslinked in solid phase) and PEO (crosslinked in all cases) are two water-soluble polymers. Polyvinyl alcohol/chitosan

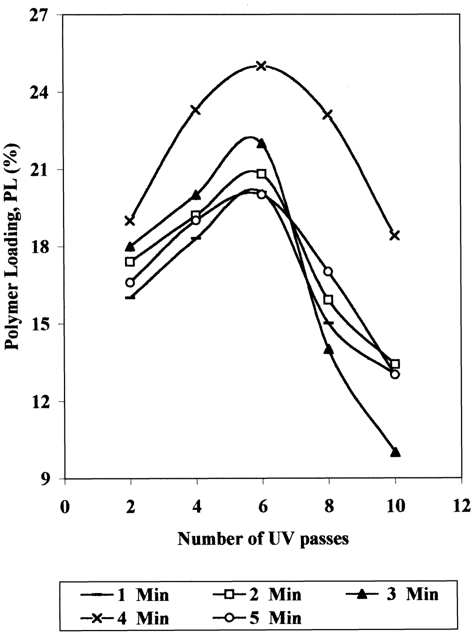

Figure 7. Polymer loading of the treated PVA/chitosan film against UV radiation (number of passes) with respect to soaking time for F2 formulation.

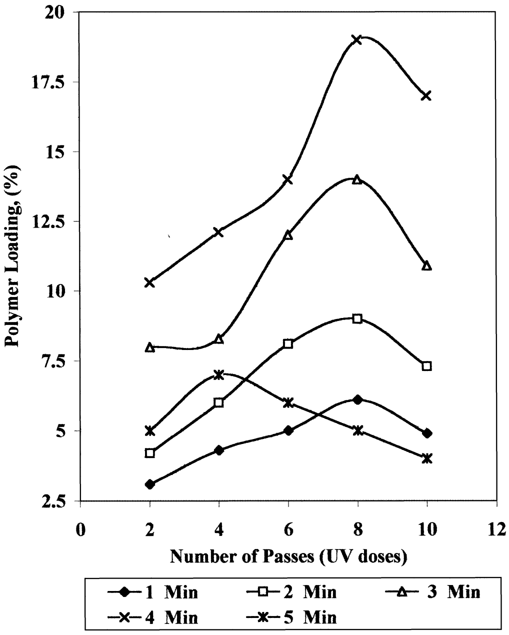

Figure 8. Polymer loading of the treated PVA/PEO film against UV radiation (number of passes) with respect to soaking time for F2 formulation.

Marcel Dekker, Inc.
270 Madison Avenue, New York, New York 10016

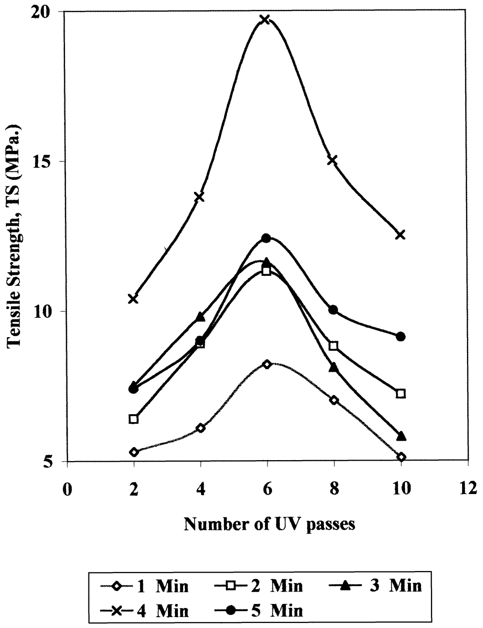

Figure 9. Tensile strength of the treated PVA/chitosan film against UV radiation (number of passes) with respect to soaking time for F2 formulation.

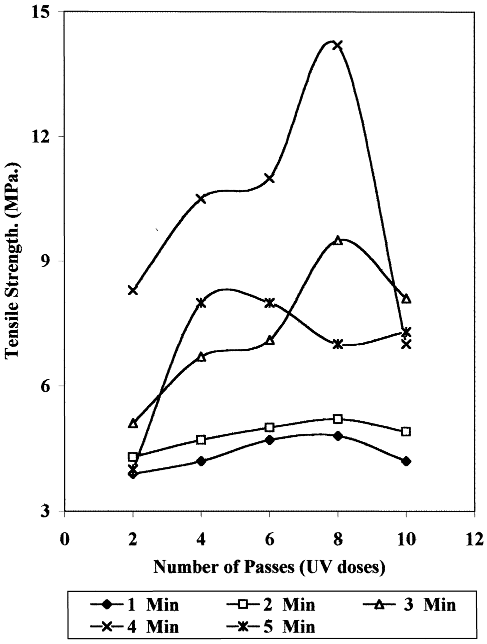

Figure 10. Tensile strength of the treated PVA/PEO film against UV radiation (number of passes) with respect to soaking time for F2 formulation.

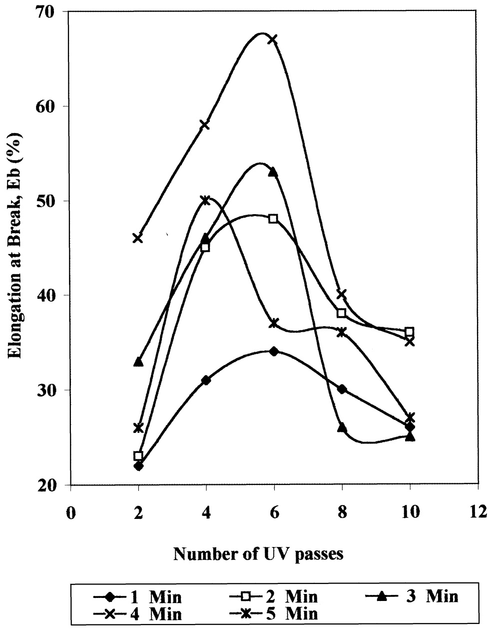

Figure 11. Elongation at break of the treated PVA/chitosan film against UV radiation (number of passes) with respect to soaking time for optimized F2 formulation.

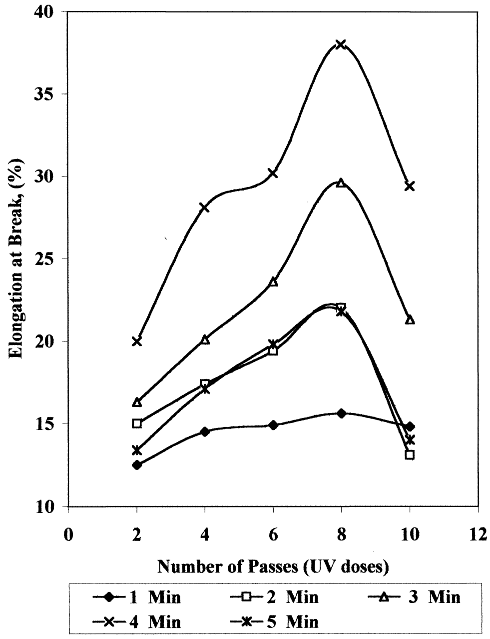

Figure 12. Elongation at break of the treated PVA/PEO film against UV radiation (number of passes) with respect to soaking time for optimized F2 formulation.

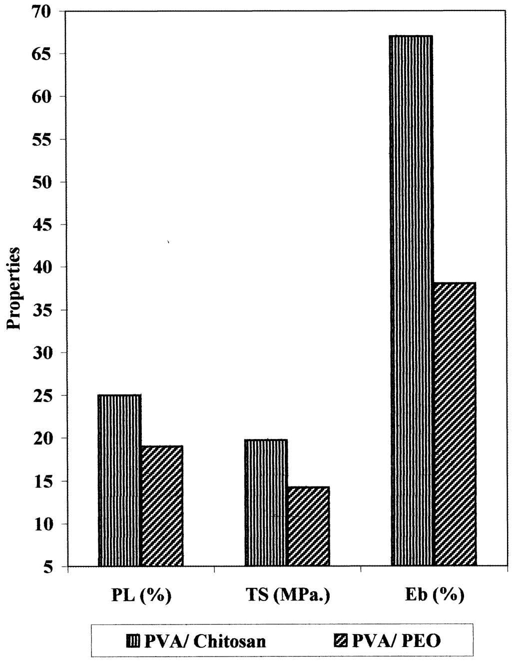

Figure 13. Comparative study of PVA/chitosan and PVA/PEO bioblend film on the basis of physico-mechanical properties.

and PVA/PEO bioblend films were prepared with these polymers. Five formulations were prepared with EHMPTMA, EHA, and the photoinitiator, Darocur-1664. Monomer soaking of these bioblend films was performed in various soaking times. The monomer treated bioblend films were cured under UV radiation at different radiation intensities. Various mechanical properties like TS (MPa) and Eb (%) and PL (%) of these bioblends were studied. It was observed from the above discussion that the PVA/chitosan bioblend film showed better physico-mechanical properties as compared to that of the PVA/PEO film. Chitosan and PEO were used as additives in this respect. But as an additive in the bioblend, chitosan performed better as it showed a higher percentage of strength.

REFERENCES

1. Fouassier, J.P.; Rabek, J.F. Eds. *Radiation Curing in Polymer Science and Technology*; Elsevier: London, England, 1993; Vol. 1.5.
2. Salamone, J.C. *Polymeric Materials Encyclopedia*; 1996; Vol. 7.
3. Roberts, G.A.F. *Chitin Chemistry*; The Macmillan Press Ltd.: London, 1992.
4. Mathur, N.K.; Narang, C.K. J. Chem. Educ. **1990**, *67* (11), 938.
5. Paul, S.K. Chingri: Adhunik Chash Projucti O Upokulyo Paribesh Babosthapona; Bangladesh, 1996, 1st Ed.

6. Mucha, M.; Marszulek, J.; Miskiewicz, D. *Proceeding, from the 3rd International Symposium on Natural Polymer and Composites*, 1SNa Pol/2000; May 14–17, 2000; 5a aPedro, SP-Brazil.
7. Zhux A.; Zhang, M.; Wu, J.; She, J. Biomaterials **2002**, *23* (23), 4657–4565.
8. McGininse, V.D. *Encyclopedia of Polymer Science and Technology*; Wiley: NY, 1986; 418 pp.
9. Banik I.; Bhowmick, A.K. J. Appl. Polym. Sci. **1998**, *69*, 2079–2087.
10. Kronggauz, V.V.; Yohannan, R.M. Polymers **1990**, *31*, 1131.
11. Kronggauz, V.V.; Yohannan, R.M. Mol. Cryst. Liq. Cryst. **1990**, *183*, 495.
12. Fitzgerald, S.; Landry, J. J. Appl. Polym. Sci. **1990**, *40*, 1727.
13. Timpe, H.J.; Strehmel, B. Angew. Macromol. Chem. **1990**, *178*, 131.
14. Nakamura, Y.; Yamaguchi, M.; Kitoyama, A.; Iko, K.; Okubo, M.; Matsumoto, T. J. Appl. Polym. Sci. **1990**, *39*, 1045.
15. Rahman, M.M.; Khan, M.A.; Ali, K.M.I.A.I. Mustafa. J. Appl. Polym Sci. **2001**, *81*, 1854–1867.
16. Cecker, C.; Moussa, K. Macromolecules **1989**, *22*, 4455.
17. Datta, S.K.; Chaki, T.K.; Bhowmik, A.K. Radiat. Phys. Chem. **1996**, *47*, 913.
18. Khan, M.A.; Shehrzade, S. J. Polym. Environ. *in press*.
19. Khan, M.A.; Ali, K.M.I.; Imam, M.R.; Mannan, R. J. Polym. Plast. Technol. Eng. **1997**, *36* (1), 89.
20. Khan, M.A.; Ali, K.M.I.; M.N. Islam. J. Polym. Plast. Technol. Eng. **1996**, *32* (2), 299.

Received November 2002
Accepted March 2003