Effect of Additives on Reinforcement of Radiation-Induced Jute–Urethane Polymer Composites

K. M. IDRISS ALI,[1] MUBARAK A. KHAN,[1] SHYAMAL K. BALO,[2] M. U. AHMAD[2]

[1] Radiation and Polymer Chemistry Laboratory, Institute of Nuclear Science and Technology, Bangladesh Atomic Energy Commission, P.O. Box 3787, Dhaka, Bangladesh

[2] Department of Chemistry, Jahangirnagar University, Savar, Dhaka, Bangladesh

Received 24 January 1997; accepted 6 June 1997

ABSTRACT: Thick polymer film was prepared under gamma irradiation using urethane acrylate in the presence of *N*-vinylpyrrolidone, ethyl hexyl acrylate, and trimethylol propane triacrylate. Both jute dust and hessian cloth (jute fabric) were used to constitute composites based on the prepared resin matrix. Some of their physical and mechanical properties were studied. Some additives such as acetic acid, acrylamide, urea, talc, and titanium oxide were incorporated into the formulation to investigate their effect on the physical and mechanical properties. Water absorption and weathering resistance of the resin and composites were also investigated. © 1998 John Wiley & Sons, Inc. J Appl Polym Sci **67:** 79–85, 1998

INTRODUCTION

The importance of natural or synthetic polymers is growing fast because of their high demand in modern technology. In fact, various types of polymers are progressively being used to replace iron and steel, brass, and other alloys, wood, etc., for making many engineering items such as bodies of electronic appliances, transport vehicles, and parts of aircrafts. Biodegradable polymers are important to keep the environment free of pollution. Most of the synthetic polymers are not biodegradable and, as such, they cause environment pollution. Natural polymers, being biodegradable, and their combination with synthetic polymers is an option to prepare biodegradable polymeric materials. There has already been some successes in preparing such polymer-based products by radiation methods.[1–3] Recently, preparation of jute-reinforced urethane polymer-based composites[4] under gamma irradiation was reported. Enhanced tensile properties with reduced elastic character result, due to reinforcement of the urethane polymer with jute. The present investigation was an extension of this work. Different additives incorporated into the formulation import some property advantages as reported in a previous investigation.[4] The effect of these additives on various properties of the composites was studied and is reported here.

EXPERIMENTAL

Materials

Ebecryl 264, a triacrylated urethane oligomer, a Radcure product, was procured from UCB, Belgium. *N*-Vinylpyrrolidone (NVP), for increasing tensile strength, ethyl hexyl acrylate (EHA), for enhanced elongation and bending strength, and trimethylol propane triacrylate (TMPTA), as a crosslinking agent, were used as obtained from Merck (Germany). The additives acetic acid (Ac),

Correspondence to: K. M. Idriss Ali.

Journal of Applied Polymer Science, Vol. 67, 79–85 (1998)
 CCC 0021-8995/98/010079-07

acrylamide (AM), urea (U), talc (Ta), and titanium oxide (Ti) were procured from BDH Co. Jute fabrics (hessian cloth) and fibers were collected from the local market of Bangladesh.

Methods

Thick (2 mm) polymer film was prepared under Co-60 gamma irradiation using a formulated solution composed of urethane triacrylate oligomer (Ebecryl 264) and reactive monomer diluents in the following proportions, viz., the oligomer: NVP : EHA : TMPTA = 50 : 40 : 5 : 5, w/w. When a minute amount (1%) of an additive selected from Ac, AM, U, Ta, and Ti was incorporated into this formulated solution, the proportions of the different constituents were changed as the oligomer: NVP : EHA : TMPTA : additive = 50 : 39 : 5 : 5 : 1, w/w. The formulated solution was poured into a mold ($8 \times 6 \times 0.2$ cm) having a glass plate pressed against a suitable steel plate with a screw-tight arrangement so that the mount could be descried after the preparation of the film in the mold under gamma irradiation obtained from a Co-60 gamma source (20 kCi). The total dose used for the preparation of the film as well as the composite was 300 krad at a dose rate of 600 krad/h. This dose was found to produce the highest physicomechanical properties with the film and composite prepared in the previous work,[4] where it was observed to fully cure the polymer at this dose. Two types of composites were prepared by mixing jute particles (40 mesh) or layers of hessian cloth at different proportions with the above formulated solutions.

Physical Properties

The hardness of the film/composite was measured by the pendulum method using a pendulum hardness tester (Model 5458, BYKE, Labotron). The tensile properties such as tensile strength (TS), bending strength (BS), and elongation at break (Eb) of the films and the jute composites were determined directly with the help of an Instron machine (Model 1011, UK) using gauge length of 1.2 cm and crosshead speed 0.003 m/min. The sample size was $8 \times 1 \times 0.2$ cm.

Water Uptake

The water-absorption ability of these films and composites was measured by soaking the substrates (films and composites) in water contained in a static water bath at 25°C for different periods (up to 70 h). The weight gained by the immersed samples was used to determine the water uptake by the samples.

Weathering Test

Both films and composites were subjected to a severe simulated weathering test using an accelerated weathering tester (Q-Panel Co., Model Q.U.V., U.S.A.) by simulating alternating cycles of sunshine (4 h) and dews and condensation (2 h). The temperature was varied between 45 ± 2°C (sunshine) and 25 ± 2°C (condensation).

RESULTS AND DISCUSSION

The composites were prepared with jute dust as well as with hessian cloth (jute fabric). Thus, two sets of composites were obtained; various properties of these composites are discussed below.

Composites with Jute Dust

Pendulum Hardness

The pendulum hardness (PH) measured for the films and the jute-reinforced polymer composites is represented in Figure 1 by plotting in bars the pendulum hardness against formulations using different additives. It is observed that the polymer film has the highest (about 40%) PH. When 5% jute dust is mixed with the formulation to prepare the composites, the hardness decreases to 36%. Incorporation of 1% additives into the formulation reduces the hardness further. This decrease is dependent on the type of additives incorporated into the formulation. Incorporation of AM drastically reduced the film hardness to about 24%. Addition of jute dust (5%) into the AM-containing film increased the hardness of the composite to about 33% (from 24%). Similarly, addition of jute dust (5%) to the formulations containing U or Ta enhanced the hardness from that of the respective films free from jute. On the other hand, the jute dust (5%) addition into the films containing Ac or Ti slightly decreased the hardness over that of the corresponding polymer.

The reason for the decrease of the PH in the presence of Ac is not known at this state; however, Ti may have formed some organometallic com-

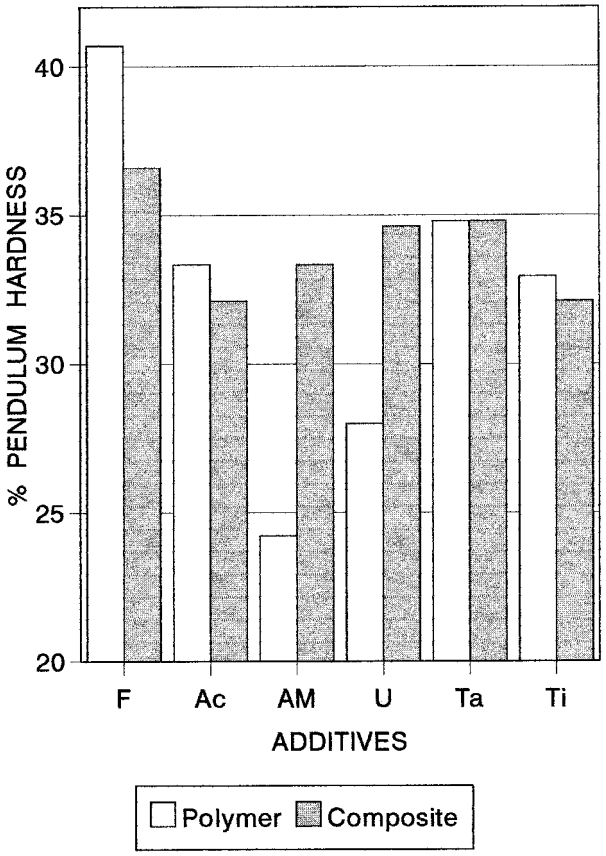

Figure 1 Effect of additives (1%) on PH of polymer and composites containing jute dust (5%).

plexes which may not have diffused easily in the reaction zone during the overall polymerization process. The enhancement in hardness of the jute composites containing AM or U can be explained by the fact that both AM and U containing the carboamide group (=N—CO—) become chemically anchored on the cellulosic chain of jute as already established in wood–plastic composites[5,6] and other jute–plastic composites.[7,8] The enhanced hardness of the composites containing Ta is the result of the inherent hardness of talc being used as the filler.

Tensile Properties

The results of additive effects on tensile properties of both plain resin films and jute-based composites are shown in Figures 2–4. The incorporation of a minute amount (1%) of an additive into the formulation greatly enhanced (unlike pendulum hardness, Fig. 1) the tensile strength (TS) (Fig. 2) of the polymer films and slightly of the composites (except titanium-containing composites) irrespective of the additives used. The highest TS value of 8.8 kPSi was obtained with urea, followed by AM and Ac. It is generally known that both hardness and tensile strength enhancements follow a gain in the crosslinking density; the hardness is the index of the crosslinking density at the surface (of the film substrate) while the TS is the index of the crosslinking density through the bulk of the entire film (composite). Generally, strong acids (sulfuric acid, nitric acid, etc.) reduce the TS of the composites based on cellulosic substrates[9] (e.g., wood–plastic composites) because the strong acids readily break the cellulose backbone and some polymer chains having condensed interunit linkages. However, weak acids such as Ac and adipic acid were found to enhance TS values in a wood–plastic composite.[10] The titanium complex also enhances the tensile strengths of jute- and cotton-reinforced composites.[11,12]

The present studies of the enhancement or otherwise of TS values as a result of the incorporation

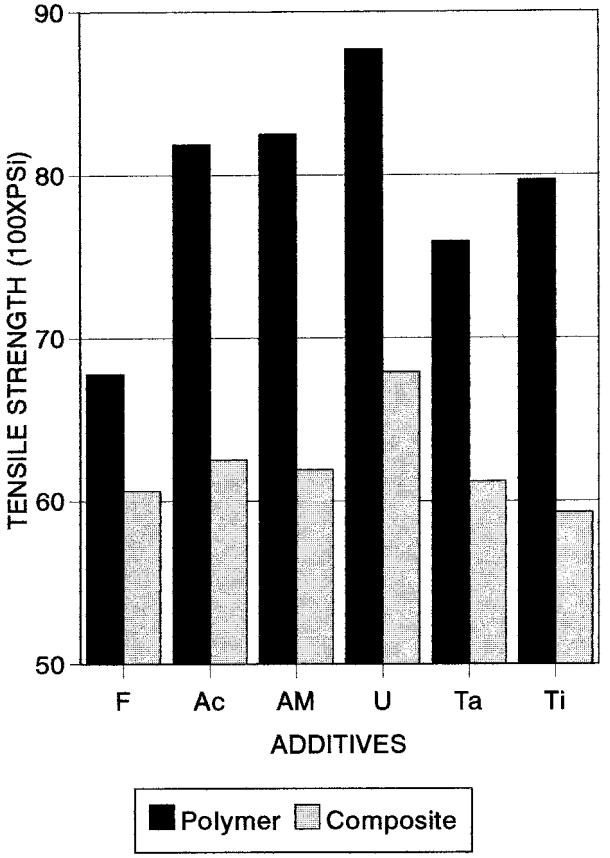

Figure 2 Effect of additives (1%) on TS of polymer and composites containing jute dust (5%).

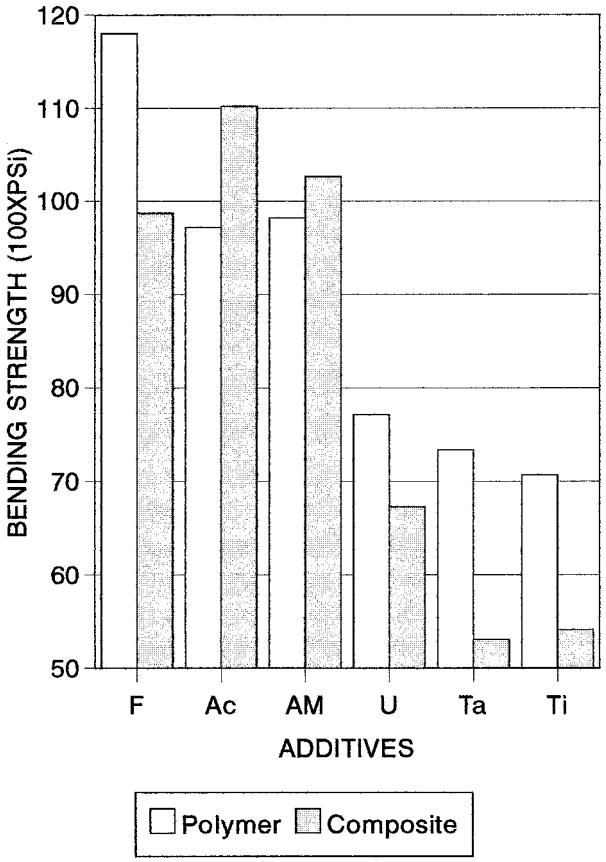

Figure 3 Effect of additives (1%) on BS of polymer and composites containing jute dust (5%).

of different additives in the resin films and related composites are important for the assessment of diverse applications of such polymer films and composites.

Although TS was enhanced in the presence of the selected additives, but both bending strength (BS) (Fig. 3) and elongation at break (Eb) (Fig. 4) decreased quite substantially on additive incorporation into the polymer or resin system. The lowest values of BS and Eb were given by the films that contained titanium. However, the jute–resin composites were somewhat different. Figure 3 shows that the bending strength of jute composites was enhanced slightly in the presence of Ac and AM but not with U, Ta, and Ti. The Eb (Fig. 4) of the jute-based composite was substantially enhanced in the presence of Ac but not so much in the presence of the other additives. Whatever may be the case, the breaking elongation (stretching ability) of the composites was generally more than that of the corresponding polymer films. This is possibly because the jute cellulose renders some assistance in the composites to gain stretching ability.

Effect of Jute Concentration

Figures 2–4 show that the tensile properties (TS, BS, and Eb) of the composite are commonly less than those of the corresponding polymer film and they are influenced by the presence of additives in the formulation. These values were obtained with 5% jute dust in the composite. It is worth studying if these properties are affected by the increase in the concentration of jute dust. Thus, the jute dust concentration was varied up to 20%. The results of PH, TS, BS, and Eb values of these composites indicate that the values of TS, BS, and Eb decrease with increase of jute dust in the composites. However, the PH values of the composites containing AM, U, and Ta initially increase with increase in the amount of jute dust and attain a maximum PH value at about 12% jute dust content in each case and then decrease again with

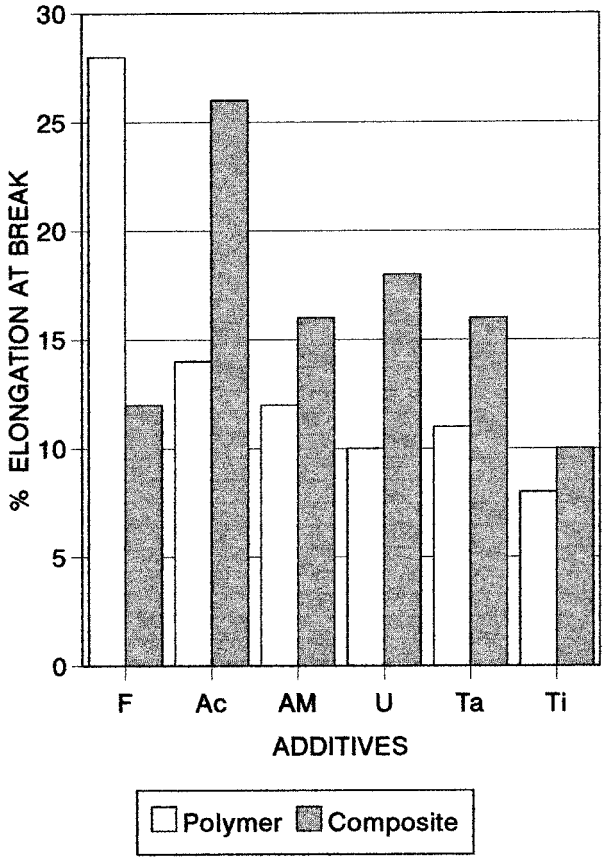

Figure 4 Effect of additives (1%) on Eb of polymer and composites containing jute dust (5%).

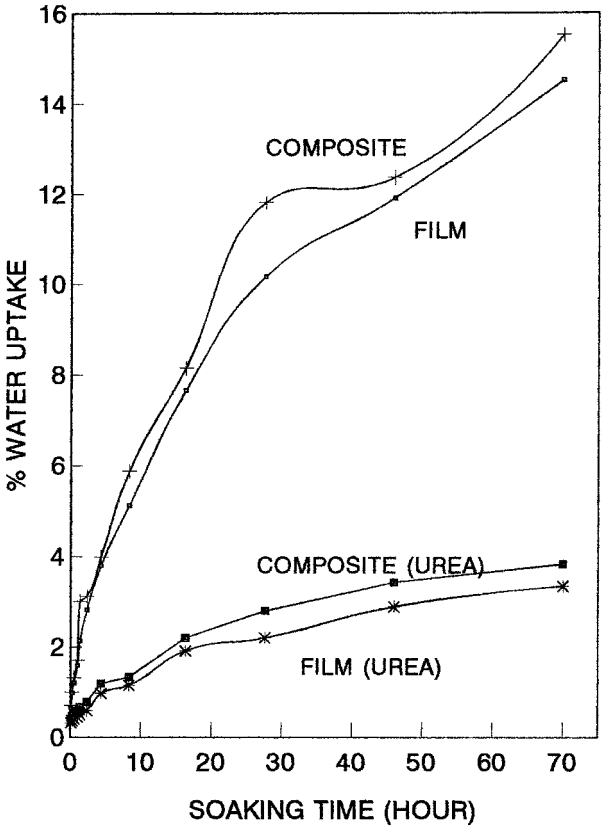

Figure 5 Water uptake of polymer and composite containing jute dust (5%) against soaking time.

further increase in jute dust content. (These are experimental observations that are not shown in Figs. 2–4.)

Water Uptake

Arbitrarily, four samples were selected for the assessment of the water absorption; two were simple resin films (without any jute substrate) and two were composites containing 5% jute dust. Urea was present in one of these composites and one film also contained urea (1%). The results are plotted in Figure 5 by water uptake vs. soaking time. Samples (film and composite) that do not contain urea show substantially higher water absorption than do the samples that contain urea, indicating that urea plays some important role to repel water molecules. The carboamide groups (=N—CO—) of the additive urea become chemically linked with segments of the matrix resin and cellulosic chains of jute, thereby forming a denser integrated network structure and, hence, more effectively restricting the penetration of water molecules. The high ultimate water uptake (14–15%) of the additive free resin or composite drops down to $< 5\%$ when urea is used as the additive in them.

Weathering Test

The weathering test was carried out for 1200 h with a composite sample that contained 5% jute dust and no additive at all. The change in tensile properties, particularly TS and BS, as a result of the weathering test was monitored and the results showing changes in TS and BS values with weathering time are given in Figure 6. Both the TS and BS values follow an increasing trend initially and pass through a maximum at 300 h of weathering for TS and 900 h of weathering for BS. This enhancement of the TS and BS values could be associated with the fact that some free radicals formed during exposure to gamma radiation remained trapped inside the samples; these radicals became activated during the weathering test (on exposure to the UV lamp). BS remains quite stable for a certain weathering period. Weathering resistance of the test composite is quite high.

Composite Containing Hessian Cloth

Composites were prepared by putting different layers of hessian cloth in between layers of polymer solutions that were irradiated by the gamma source to form polymer films. One layer of hessian

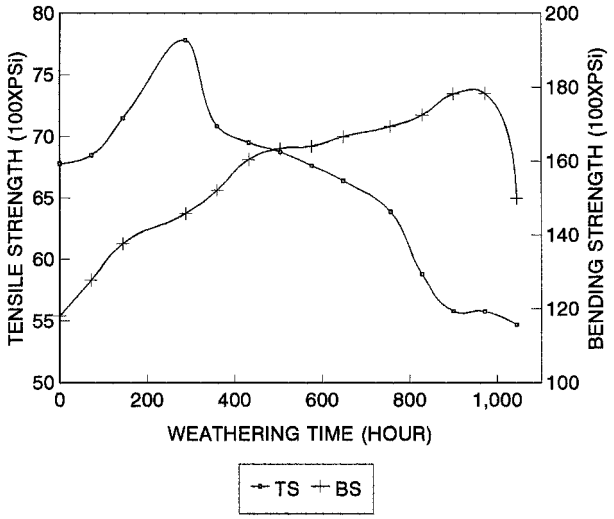

Figure 6 Effect of weathering on tensile properties of the composites containing jute dust (5%).

Table I Effect of the Acetic Acid on the Tensile Properties of Composites

	% of Ac					
% of Hessain Cloth	F	1%	2%	3%	5%	10%
Tensile strength (PSi)						
12	7800	3781	4167	5079	5622	4869
24	8268	4806	4972	5892	3535	2718
38	6245	4182	4031	3866	3000	2500
Bending strength (Psi)						
12	13,898	4342	4433	8342	7369	3120
24	15,158	5488	5720	9488	6564	4928
38	12,724	7036	2551	7036	5800	3020
Elongation at break (%)						
12	14	18	25	22	20	20
24	12	17	21	20	18	18
38	10	16	20	18	16	15

F = no additive.

cloth yields 12% jute content in the composites; similarly, two and three layers of cloth amount to 24 and 38% jute contents, respectively, in the composites. Before putting the hessian cloth in the mold, it was soaked for 2 min in acetic acid. Different properties (TS, BS, Eb) of the composites reinforced with hessian cloth were compared with those of the polymer film prepared with the same formulated solution. These results are shown in Table I. Both TS and BS substantially increased on reinforcement of the resin using hessian cloth. The highest TS and BS values were obtained for the 24% hessian cloth composite. However, the elongation values decrease with increase in the hessian cloth (Table I). This is significant information: that hessian cloth-reinforced polymer composites possess enhanced physical properties (TS and BS) over those of the jute dust-containing composite. Hessian cloth has a network arrangement that assists augmentation and interaction processes among the polymer and cellulose units. This is not so with the jute dust that is spread throughout the composite and is less capable of augmentation and interaction with other polymer chains. This is possibly one of the reasons that hessian cloth contributes more toward enhancing physical parameters of the jute polymer composites than do the jute dusts. Jute content beyond 24% of hessian cloth in the composites decreases the TS, BS, and Eb values (Table I). Increased jute content (beyond 24%) may have caused brittleness in the composite, which reduced the physical properties (TS, BS, and Eb) of the composite.

Effect of Acetic Acid (Ac)

Figures 2–4 show that incorporation of 1% Ac into a jute dust polymer composite enhanced the TS, BS, and Eb values. The jute composites were prepared with hessian cloth at different proportions of the polymer solutions that also contained Ac at different concentrations (0–10%, w/w). The results of TS, BS, and Eb values of these composites are shown in Table I. Both TS and BS values decreased in the presence of Ac, but Eb values increased slightly with Ac; 2% Ac yields the highest increment in elongation after which the Eb values decrease with increase in Ac. Also, the 12% jute content yields the maximum elongation; with increase in the hessian cloth content in the composite, the elongation decreases. But in the case of TS and BS values, there are maximum values with the 24% hessian cloth, after which they (TS and BS) decrease.

From this study, it was observed that the jute polymer composite reinforced with a hessian cloth layer in between polymer layers possesses better rheological properties than those of the jute–polymer composite containing jute dust. Some of the additives, particularly those additives that contain the carboamide group, are better in enhancing certain physical properties.

REFERENCES

1. M. A. Khan, M. K. Uddin, M. N. Islam, and K. M. Idriss Ali, *J. Appl. Polym. Sci.,* **58,** 31 (1995).
2. M. K. Uddin, M. A. Khan, and K. M. Idriss Ali, *Polym. Degrad. Stab.,* **55,** 1 (1996).
3. M. K. Uddin, M. A. Khan, and K. M. Idriss Ali, *Radiat. Phys. Chem.,* **48**(4), 511 (1996).
4. M. A. Khan, S. Balo, and K. M. Idriss Ali, to appear.
5. M. M. Husain, M. A. Khan, K. M. Idriss Ali, and M. A. Hossain, *Radiat. Phys. Chem.,* **45**(4), 623 (1995).
6. K. M. Idriss Ali, M. A. Khan, and M. M. Husain, *Radiat. Phys. Chem.,* **44**(4), 427 (1994).
7. M. A. Khan, M. N. Islam, M. A. Hossain, and K. M. Idriss Ali, *Radiat. Phys. Chem.,* **48**(3), 337 (1996).
8. K. M. Idriss Ali, M. O. Ali, and M. A. Khan, to appear.
9. M. M. Husain, M. A. Khan, K. M. Idriss Ali, and A. I. Mustafa, *Radiat. Phys. Chem.,* **47**(1), 149 (1996).
10. M. M. Husain, M. A. Khan, and K. M. Idriss Ali, *Polym.-Plast. Technol. Eng.,* **35**(6), 959 (1996).
11. M. A. Khan and K. M. Idriss Ali, to appear.
12. K. M. Idriss Ali and M. A. Khan, to appear.