Jute Composite with MMA by Gamma and UV Radiations in the Presence of Additives

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ABSTRACT: Jute yarns treated with MMA + MeOH solutions were irradiated either with Co-60 gamma source or with UV radiation. In gamma radiation, polymer loading of MMA (methyl methacrylate) onto jute increased quite substantially, but the strength of the composite decreases sharply after 15% polymer loading. The gamma-treated jute samples were very brittle. On the other hand, jute yarns irradiated *in situ* under UV radiation was found to be grafted with MMA. The tensile strength of the UV-cured jute yarn composite increases with an increase of grafting level, in contrast to the behavior observed with the gamma-irradiated jute composite samples. The tensile properties of the composites can be further enhanced by the incorporation of certain additives and coadditives into MMA + MeOH solutions. This opens diverse applications for jute materials. © 1999 John Wiley & Sons, Inc. J Appl Polym Sci 74: 900–906, 1999

Key words: jute; composite; jute composite; gamma radiation; UV radiation; MMA–jute composite

INTRODUCTION

Jute is a natural polymer composed mainly of cellulose, hemicellulose, and lignin. Applications of natural polymers including jute materials faded away at the advent of synthetic polymers that are quite cheap and durable compared to natural polymer materials. This situation is now changing and people are more interested in using natural polymers in place of synthetic ones. The main reason is that the synthetic polymers are not easily decomposable and degradable and thus, cause environmental pollution. The natural polymers, on the other hand, are environmentally friendly. Nevertheless, attempts have been continually taking place to improve natural polymers both genetically and chemically. Some have prepared thermoplastics by laminating synthetic polymer on natural polymer.[1] Others have blended different natural polymers together to improve quality and property of the blended threads (e.g., cotton is mixed with jute cellulose to prepare an improved variety of material). Khan et al.[2,3] have improved jute materials by treating jute with some formulated solutions of urethane-based polymeric materials under UV radiation. The improved jute materials behave like jute–plastic composites. Some formulations were developed in such a way that the jute products treated with the developed formulations under UV radiation attained high durability like that of synthetic polymers; however, these jute products could be decomposed and degraded in the same way as natural polymer after the end-use.[4] The present investigation deals with the modification of jute yarns with methyl methacrylate (MMA) in the presence of various additives and coadditives under Co-60 gamma and ultraviolet radiations.

EXPERIMENTAL

Materials

Jute yarns (tossa, 40 counts) were collected from the Islam Jute Mills (Bangladesh). Bulk mono-

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mer MMA and additives such as NVP (*N*-vinyl pyrrolidone), MA (methyl acrylate), EHA (ethyl hexyl acrylate), TP (tripropylene glycol diacrylate), and TM (trimethylol propane triacrylate) were used as procured from Merck, Germany. Coadditives such as lithium nitrate ($LiNO_3$), urea (U), and diallylphthalate (DP) were obtained from Aldrich Chemical Co (Australia). The swelling solvent methanol (MeOH) was purchased from the local market.

Methods

Bulk monomer MMA was mixed with swelling solvent methanol at different proportions of MMA (10–90%) and MeOH (90–10%, v/v). To these solutions, additives (1%) and/or coadditives (1%) were incorporated. Jute yarns (each 40-cm long and predried at 105°C) were dipped for 1 h into these solutions contained in test tubes and irradiated with either Co-60 gamma or UV radiation at NTP, whereas the yarns were still in the solutions exposed to normal air. In the case of gamma radiation, a Co-60 source (25 kCi) was used. The samples were irradiated at different dose rates (2.5–7.50 kGy/h) for different total doses (2–10 kGy). In the case of UV radiation, a global UV lamp (model B-100 A, 100 W, 365 nm, manufactured by UVP, Inc., USA) was used. The test tubes were placed in a rotatory rack that moved around the lamp. Each test tube spins on its own axis during the rotation of the tube surrounding the lamp that is hung at the top in the middle of the entire setup. This process of radiation at rotatory axes ensures uniform UV radiation to the samples. The samples were irradiated for different periods (15–180 min) to achieve the maximum grafting of MMA with yarn.

The irradiated samples were washed with normal acetone several times before extraction in hot acetone for 20 h in a soxhlet. The different weights of the samples before irradiation and after extraction determined the extent of loading (grafting). The extracted samples were used to measure tensile properties (strength, *TS*, and elongation at break, *Eb*) with the help of an Instron machine (Model 1011) at a crosshead speed of 3 cm/min and gauge length of 1.2 cm. The samples were also used to determine water uptake by soaking the samples with suitable means in water contained in a static water bath at 25°C. The amount of water uptake was monitored periodically for 2.5 h; before weighing, the wet sample was dabbed between two filter papers (Whatmann No. 4) for 30 s. The increase in the weight of the wet sample determines the extent of water uptake.

RESULTS AND DISCUSSION

Jute yarns were treated with MMA using two different radiation sources (e.g., Co-60 gamma radiation and UV radiation). The bulk monomer (MMA) was mixed with methanol to swell the cellulose backbone for better impregnation of MMA solution. Some organic and inorganic additives were incorporated into MMA + MeOH solutions to improve grafting treatment process and mechanical strength of the treated yarn.

Gamma Radiation Technique

Optimum Dose Rate Determination

Jute yarns soaked in MMA + MeOH solutions contained in test tubes were irradiated at NTP in air with Co-60 gamma source (25 kCi) at different dose rates (2.5–7.5 kGy/h) for a total dose of 10 kGy. The irradiated jute yarns, taken from the test tubes and washed with acetone to remove adhered monomer solutions, were used to determine polymer loading (*PL*). The *PL* values thus obtained are plotted in Figure 1 versus concentration of MMA as a function of different dose rate. It is observed that *PL* increases with an increase in concentration of MMA up to 70% MMA and then decreases. At higher MMA concentration, the radical–radical recombination reaction among growing MMA molecules leading to poly(methyl methacrylate) (PMMA) may be dominant. Since the PMMA is formed through the dominant reactions of MMA + MMA, the reaction of MMA + jute is less prominent. Thus, the reaction between MMA and jute cellulose is diminished. It is also observed that the *PL* increases with dose rates up to 5 kGy/h and then decreases. The highest *PL* value is obtained with a dose rate of 5 kGy/h at 70% MMA. The lowest *PL* value is achieved with 7.5 kGy/h. The subsequent gamma radiation was, therefore, carried out with a dose rate of 5 kGy/h.

Total Dose Determination for Optimum Grafting

Jute yarns were irradiated at a dose rate of 5 kGy/h for different total doses, ranging from 2 to 10 kGy. The *PL* values determined as before are plotted in Figure 2 against MMA concentration as

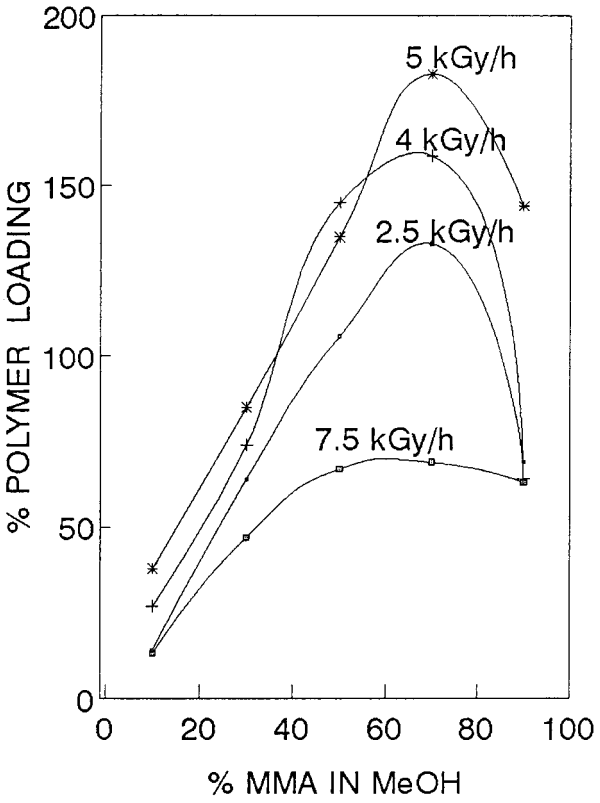

Figure 1 Polymer loading versus MMA concentration as a function of dose rate.

a function of total dose. It is observed that the maximum *PL* values are obtained at 70% MMA and at 10 kGy total dose. The *PL* values increase with total dose. The bulk solution became very viscous when the irradiation was >10 kGy and it was difficult to take out the jute yarn samples from the irradiated solutions.

Effect of Additives

A minute amount of additives (1%, v/v) was incorporated into MMA + MeOH solutions. Polymer-loading values of the jute yarns irradiated for 10 kGy at 5 kGy/h are plotted in Figure 3 versus MMA concentration. It is observed that solutions containing additives TM, TP, U, and NVP yielded maximum *PL* values at 70% MMA (30% MeOH), whereas additives DP, Li, EHA, and MA contributed maximum *PL* values with 50% MMA. This reduces the amount of bulk monomer MMA required for making the jute composite and the cost of the process. The polymer-loading values have undoubtedly increased by the incorporation of these additives into the solutions. The highest *PL* value is obtained by TM, a trifunctional acrylated monomer that has three distinct acrylated double-bond groups that can act like branches for enhanced and increased crosslinking activities with jute cellulose and MMA moieties. The second highest *PL* value is obtained by TP, a difunctional monomer. Lithium molecule has possibly entered into a ligand formation with jute cellulose and MMA molecule. However, this needs further investigation with ESR and other suitable techniques.

Tensile Properties

Most of the gamma-irradiated jute samples were quite brittle and it was difficult to measure their tensile properties. The change in tensile properties of the treated jute samples is expressed as factors T_f and E_f. The tensile strength factor or tenacity factor (T_f) is the ratio of the tensile strength or tenacity (TS_t) of the treated jute sample to that (TS_0) of the untreated sample. This means $T_f = TS_t/TS_0$; similarly, $E_f = Eb_t/Eb_0$, where Eb_t is the elongation at break of the treated sample and Eb_0 is that of the untreated virgin sample. The results plotted in Figure 4

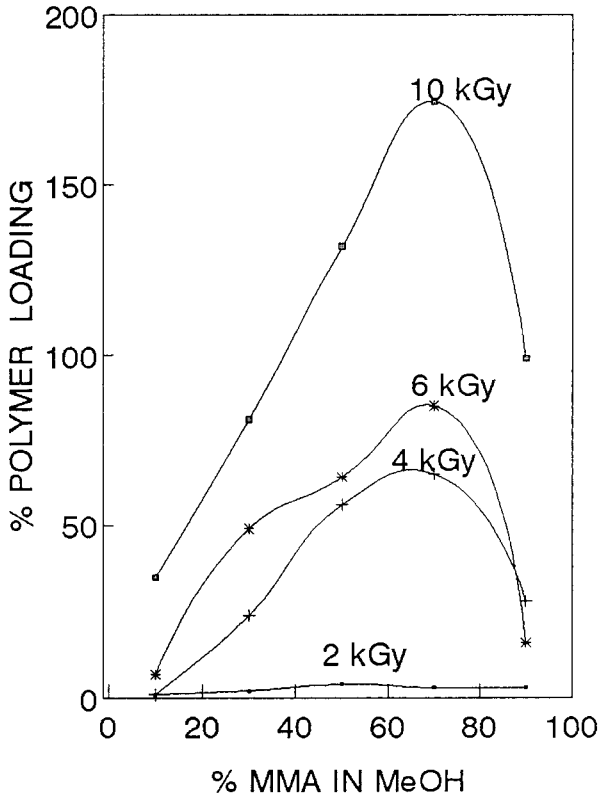

Figure 2 Polymer loading versus MMA concentration as a function of total dose.

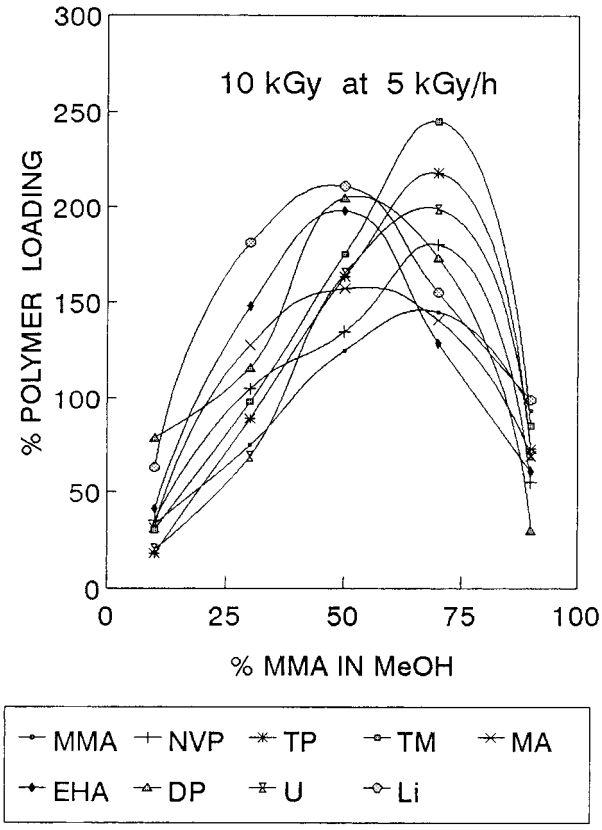

Figure 3 Polymer loading with MMA in the presence of different additives.

show that tensile strength (TS) of the gamma-treated jute yarns reduces sharply with increased polymer loading. Contrary to this, higher tensile properties (strength and elongation) have been achieved with hessian cloth (jute products) under the UV radiation system.[5] Thus, it is quite advisable to treat the jute yarns under UV radiation instead of gamma radiation to improve their properties. In the case of the hessian cloth,[5] it was coated with urethane acrylate solutions and then irradiated (cured) under UV lamp, but in the present case, jute yarns (not hessian cloth) are soaked in MMA (instead of urethane acrylate) solutions and irradiated under UV lamp while the yarns are still soaking inside the solutions.

Ultraviolet Radiation Technique

Jute yarns immersed in MMA + MeOH solutions contained in test tubes were irradiated under global UV lamp (100 W) for different periods (15–180 min). After the irradiation, samples were taken out and washed several times in cold and hot acetone to ensure removal of all the adhering materials from the jute yarns. Grafting of jute yarns with MMA under the UV irradiation was determined by using the formula, % grafting = $100(W_e - W_0)/W_0$, where W_0 is the weight of the untreated jute sample and W_e is the weight after the extraction; the results are plotted in Figure 5 against MMA concentration as a function of UV irradiation time. The extent of grafting increases with MMA concentration as well as with time of UV irradiation. The highest grafting (11%) is achieved for 1 h irradiation and the grafting decreases with irradiation time after 1 h. The maximum grafting is obtained mainly at 50% MMA concentration; however, 9 and 8% graftings were also achieved at 70% MMA concentration for 90 and 150 min irradiation, respectively. Thus, subsequent UV irradiations were carried out for a 1-h period.

Effect of Additives

Jute yarns immersed in MMA + MeOH solutions containing an additive (1%) were irradiated un-

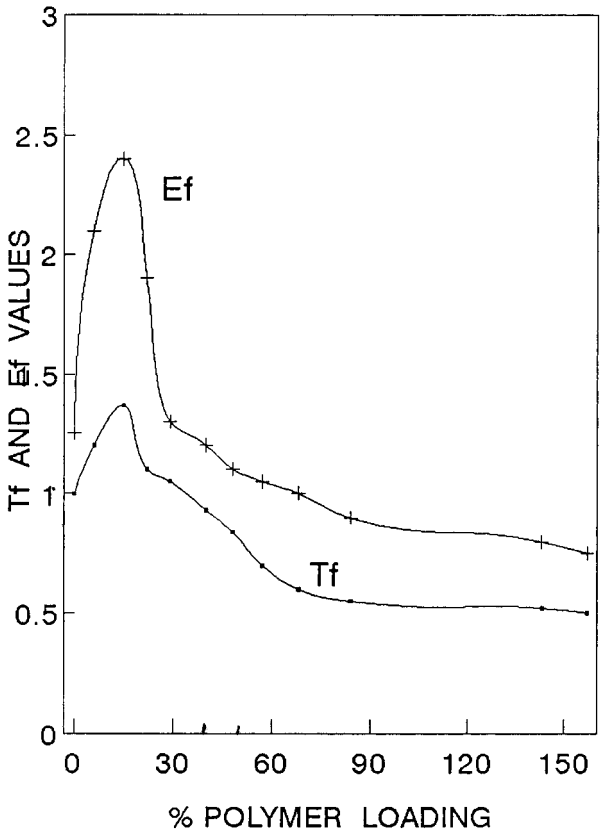

Figure 4 Tensile strength of gamma-treated jute yarns against polymer loading with MMA.

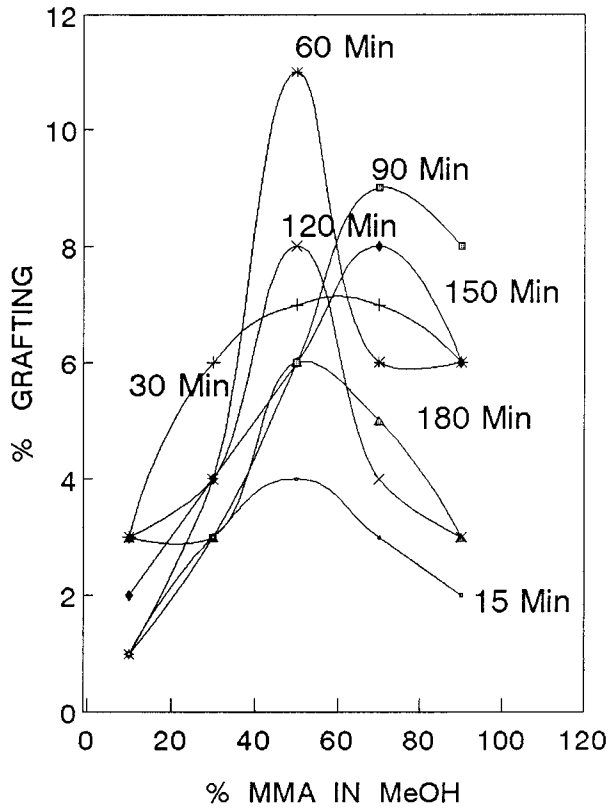

Figure 5 Grafting of jute yarns with MMA under UV radiation at different irradiation periods as a function of MMA concentration.

der the UV lamp for 1 h. Grafting of jute yarns with MMA in the presence of additives is shown in Figure 6. It is observed that the maximum grafting is obtained mainly at 50% MMA for all the additives except NVP and Li, which exhibit maxima at 70% MMA concentration. It is also observed that most of the additives enhanced the grafting except EHA and $LiNO_3$. The highest grafting is obtained in the presence of DP, a plasticizer. A similar observation has also been made by other workers[6] with hessian cloth treated under UV radiation in the presence of DP and urethane acrylate, and it was found that the plasticizer increased both grafting and tensile properties of the hessian cloth. NVP is a monomer with carboamide group (>N—CO—) that is known to create better augmentation with cellulose through the carboamide group.[7] The additive MA has also yielded the second highest grafting at 50% MMA.

Tensile Properties

Tensile properties, particularly strength (TS) and elongation at break (Eb) of the grafted yarns were determined and compared with those of the untreated yarns. The results are expressed as factors ($T_f = TS_{\text{treated}}/TS_{\text{untreated}}$ and $E_f = Eb_{\text{treated}}/Eb_{\text{untreated}}$). It is expected that the yarns with the highest grafting would yield the highest tensile strength (because the untreated jute sample is not brittle like the sample treated under gamma radiation). The tensile strength factor, T_f, and elongation factor, E_f, for MA and DP systems (without coadditives) were determined. The results are shown in Figure 7 along with the corresponding grafting values against MMA concentration. In the case of the MA system, the tensile strength of MMA + MeOH grafted jute yarns has been increased by about 31% [T_f (MA) = 1.31] and the TS of the DP system by about 50% [T_f (DP) = 1.50] over those of the virgin jute. In the case of gamma irradiation, the treated jute yarns were brittle and there was virtually no elongation, but in the UV radiation system, there is substantial enhancement of elongation of about 45% [E_f (MA) = 1.45] more for the MMA + MA-treated yarns than that of the untreated yarn. It

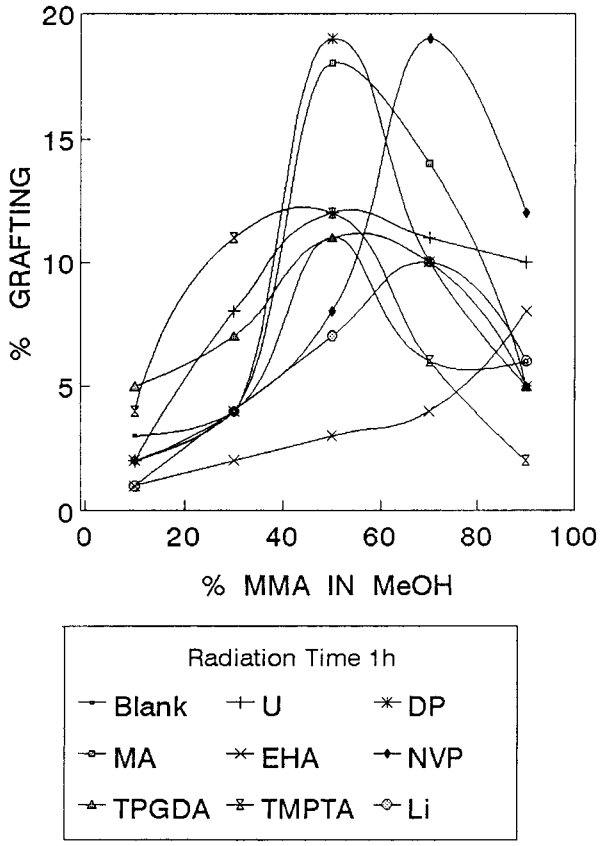

Figure 6 Grafting of jute yarns with MMA in the presence of different additives.

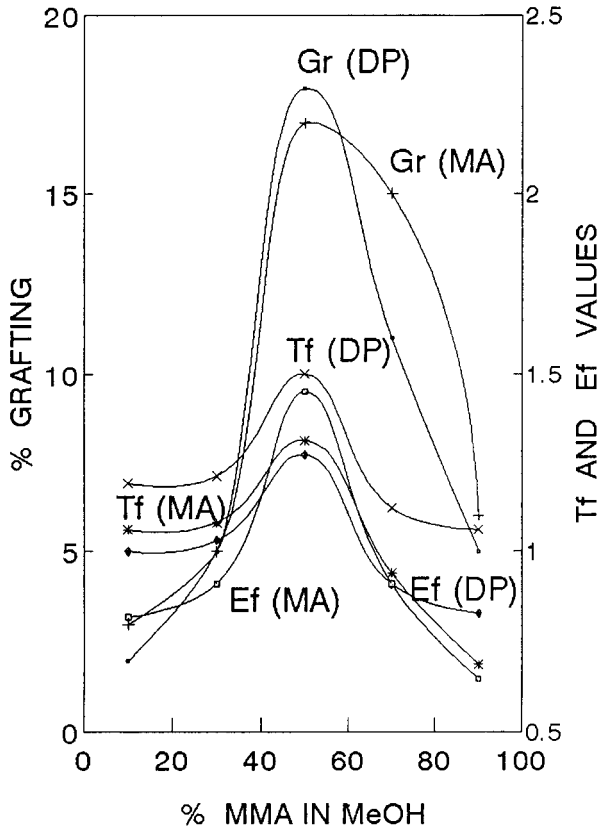

Figure 7 Grafting and tensile properties of jute yarns treated under UV radiation at different MMA concentrations in the presence of MA and DP additives only.

is really a significant achievement. In the case of DP system, the elongation has been increased by about 27% [E_f (DP) = 1.27] over the elongation of the untreated jute yarn.

Effect of Coadditives

In the case of jute plastic composite[6] and wood plastic composite,[8] it was observed that the tensile properties of the composites were enhanced when coadditives were incorporated into the bulk monomer solutions. Keeping this in mind, MA and DP, the two additives that rendered the highest grafting in the UV system, were used as additives to which a coadditive was incorporated to study the effect of coadditives on the tensile properties.

MA System

A coadditive was used to the solution of MMA + MeOH + MA; the extend of grafting of MMA onto the UV-radiated jute yarns is shown in Table I. It is observed that the level of grafting has been suppressed by the incorporation of a coadditive. The lowest grafting is obtained with the EHA coadditive system, but the tensile strength factor, T_f, for the EHA system is the highest (T_f = 1.65) among all other systems. It is interesting to note that although the EHA + MA system has rendered the lowest grafting (6%), it has yielded the highest tensile strength (65%, T_f = 1.65). The case of elongation for EHA + MA system is similar. Eb has been increased by about 109% (E_f = 2.09) for EHA + MA.

DP System

In the case of the DP system, grafting of MMA has also been suppressed similar to the MA system (Table I). The lowest grafting is obtained with EHA and the tensile strength has, in general, been slightly reduced by the incorporation of the coadditives, but the elongation has been increased with the addition of coadditives EHA and TP only.

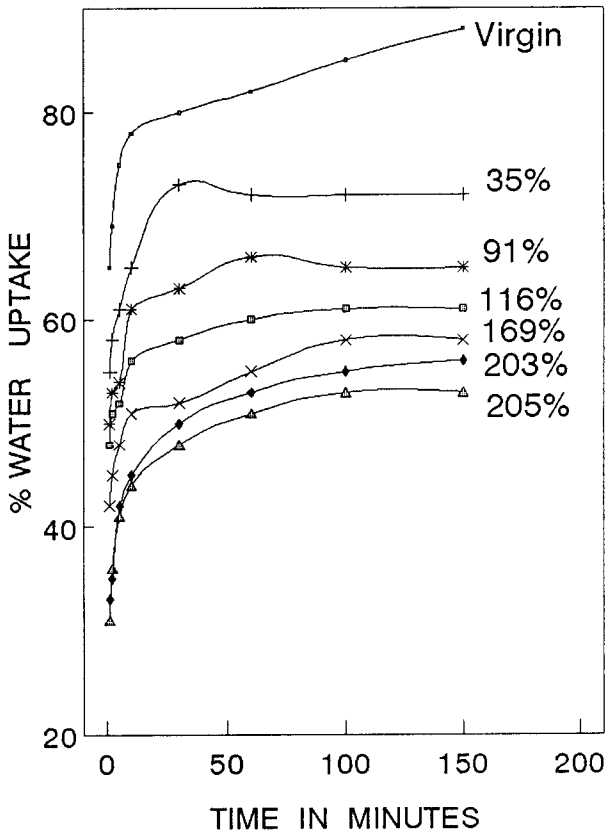

Figure 8 Water uptake by treated and untreated jute yarns against soaking time.

Table I T_f and E_f Values of Treated Jute Yarns in the Presence of Additives and Coadditives

	Additives					
	MA System			DP System		
Coadditives	Grafting	T_f	E_f	Grafting	T_f	E_f
Blank	17	1.31	1.45	18	1.5	1.27
NVP	16	1.45	1.36	17	1.45	1.18
EHA	6	1.65	2.09	6	1.44	1.36
TPGDA	9	1.38	1.18	9	1.44	1.36
TMPTA	11	1.23	1.36	10	1.45	1.27
Urea	10	1.31	1	9	1.31	1.18
Li^+	9	1.31	0.91	8	1.13	1.18

NVP, *N*-vinyl pyrrolidone; EHA, ethyl hexyl acrylate; TP, tripropylene glycol diacrylate; TM, trimethylol propane triacrylate; Li^+, lithium nitrate; MA, methyl acrylate; DP, diallyl phthalate.

Water Uptake

Water uptake measurements were carried out with the jute samples treated in MMA + MeOH and irradiated with Co-60 gamma irradiation only. The water absorption by both treated and untreated jute yarns is very fast within the initial few hours and highest with virgin yarns (Fig. 8). The gamma-irradiated jute yarns were very brittle and contained a high degree of polymer loading (150%); the water uptake by the treated samples seems to be quite high (55%) compared to that with the virgin yarns (80%) after 2.5 h of soaking in water. There may be a high density of polymer loading around the matrix of jute yarns without causing any substantial grafting of MMA with the jute yarns under the gamma radiation. This phenomenon may have caused a high degree of water absorption by the gamma-irradiated yarns, unlike UV-treated jute samples.[6–8]

CONCLUSION

Jute yarns treated with MMA + MeOH under Co-60 gamma source are brittle and yield a high degree of polymer loading. This treatment reduces the tensile strength of the yarns. In fact, as the polymer loading increases, tensile property decreases under the gamma radiation. On the other hand, grafting of MMA with jute yarns under UV radiation exists. Moreover, as the grafting increases, tensile properties increase, unlike in gamma radiation. In some cases, there is further enhancement of these properties under the UV system in the presence of some additives and coadditives with the bulk solution of MMA + MeOH. It is thus possible that jute yarns can be suitably grafted with MMA under UV radiation with the enhancement of tensile properties. This investigation tends to reveal diverse applications of jute yarns.

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