Effect of Grafting Methacrylate Monomers onto Jute Constituents with a Potassium Persulfate Initiator Catalyzed by Fe(II)

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ABSTRACT: The graft copolymerization of methyl methacrylate and ethyl methacrylate monomers onto jute fiber was carried out in an aqueous medium with potassium persulfate as an initiator under the catalytic influence of ferrous sulfate in the presence of air. The effects of parameter variables, such as the monomer, initiator, and catalyst concentrations, the reaction time, and the temperature, on grafting and the effect of grafting the monomers onto jute constituents were studied. The degree of grafting depended on the kinds of monomers and the parameter variables. The maximum graft yield percentages with methyl methacrylate and ethyl methacrylate under optimized conditions were 18.9 and 38.8%, respectively, and the grafting onto jute fiber was largely affected by one of its main constituents, such as hemicellulose. The graft copolymers were characterized, and their improved properties were also examined. © 2006 Wiley Periodicals, Inc. J Appl Polym Sci 103: 2369–2375, 2007

Key words: fibers; graft copolymers; monomers

INTRODUCTION

Chemical modifications of both natural and synthetic polymers can provide a potential route for significantly altering the physical and mechanical properties. The modification of polymers via graft copolymerization has been the subject of much interest and has made a paramount contribution toward improved industrial and biomedical applications. The graft copolymerization of vinyl monomers onto cellulose, lignocellulose, and their derivatives has been the subject of extensive studies since 1946.[1]

Jute, a lignocellulosic polymer, is the most abundant renewable and raw agricultural material and is transformed into multifarious products affecting every phase of our daily life because of its widespread applications for making coarse-woven fabrics, such as gunny sacks and bagging, for which cheapness is the prime factor. Recently, efforts have been made in Bangladesh and India for the commercial utilization of jute fiber to meet the total fabric requirements of the countries and to minimize the import of synthetic fibers. Although jute fiber possesses high dimensional stability, certain unfavorable textile properties such as high stiffness, very low elasticity, and susceptibility toward sunlight have tremendously limited their use. With a view toward minimizing the undesirable properties and enhancing the effectiveness for intensified textile use, graft copolymerization onto jute fiber has recently been attempted with chemical methods of initiation with higher valence transition-metal ions such as V(V), Mn(VII), and Ce(IV).[2,3] However, no report seems to be available on the grafting of jute fiber and its constituents with methyl methacrylate (MMA) and ethyl methacrylate (EMA) monomers with $K_2S_2O_8$/Fe(II) redox systems. It is therefore thought that chemical modification through the graft copolymerization of jute fiber by a combination of esterification and chain polymerization after the application of readily polymerizable monomers, such as MMA and EMA, under the influence of appropriate catalysts such as ferrous sulfate may turn out to have more potential with respect to all-around property improvement. Grafting was determined on the basis of the weight gained for the treated fiber, and the results of related studies were reported and characterized in this investigation.

EXPERIMENTAL

Materials

The tossa variety (*Corchorcus olitorious*) of jute fiber was procured from a local market in Rajshahi, Bangladesh; 60 cm of the fiber was discarded from the bottom, and 30 cm was taken for investigation. Vinyl

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monomers, such as MMA (BDH, London, England) and EMA (BDH, London, England), were purified via washing with 5% sodium hydroxide followed by distilled water and finally dried over anhydrous calcium chloride. All other chemicals were analytical-reagent-grade.

Preparation of the jute sample

Jute fiber was scoured with 6.5 g of detergent and 3.5 g of soda per liter at 70–75°C for 30 min.[4] This raw jute fiber was bleached with a 5 g/L sodium chlorite (NaClO$_2$) solution at pH 4 and at 85–90°C for 90 min in a liquor ratio of 1 : 50.[4,5] After being washed well with distilled water, this fiber was rinsed with a 0.2% sodium metabisulfite (Na$_2$S$_2$O$_5$) solution for 15 min. The bleached jute fiber was thoroughly washed with distilled water, dried at 105°C, and then stored in a desiccator.

Isolation and estimation of the main constituents of the jute fibers

The main constituents of jute fiber, such as α-cellulose, hemicellulose, lignin, pectic matter, fatty matter, and waxy matter, were determined and calculated according to a TAPPI standard[6] and Abou-Zeid et al.[7]

Estimation of the aqueous extracts

The raw jute fiber was treated with distilled water at boiling (100°C) for 2 h, filtered, and dried at 105°C to a constant weight. This was dewatered fiber.

Estimation of the fatty and waxy matters

The raw jute fiber was immersed in a benzene/alcohol mixture (2 : 1 v/v) in a beaker in a liquor ratio of 1 : 100 and then allowed to stand for 10 h with occasional stirring. The fiber was then washed with a fresh benzene/alcohol mixture and finally with alcohol, and then the fiber was dried at 105°C to a constant weight.

Estimation of the pectic matter

The dewaxed jute fiber was heated with a 0.5% ammonium oxalate solution in a liquor ratio of 1 : 100 at 70–80°C for 3 days in a heating mantle. As evaporation continued, hot, distilled water was added to keep the level of the solution constant throughout the process. The fiber was filtered, washed thoroughly with hot, distilled water, and then dried at 105°C to a constant weight.

Estimation of lignin by the kappa number (K) method

The jute sample was thoroughly disintegrated and dissolved in exactly 800 mL of distilled water at 25°C. Exactly 100 mL of 0.1N KMnO$_4$ and 100 mL of a 4N H$_2$SO$_4$ solution were added quickly to the disintegrated specimen solution. At the end of 10 min, the reaction was stopped by the addition of 20 mL of a 1N KI solution. Immediately after mixing, but without the filtration of the fibers, the free iodine in the suspension was titrated with a 0.2N Na$_2$S$_2$O$_3$ solution, and a few drops of a starch indicator were added toward the end of the reaction.

A blank titration with the same procedure, but without the fiber, was performed. The bath-temperature-corrected K value was calculated as follows:[8]

$$K = \frac{p \times f}{w}[1 + 0.013(25 - t)]$$

where f is the factor for correction to 50% KMnO$_4$ consumption, depending on the value of p; w is the amount of moisture-free fiber (g); p is the volume of 0.1N permanganate actually consumed by the specimen (mL); and t is the actual temperature (°C).

With the K value, the lignin content in the jute sample was calculated as follows:

$$\text{Lignin}(\%) = K \times 0.15$$

Estimation of α-cellulose and hemicellulose

In this step, all noncellulosic matter in the jute fiber was removed by a treatment with bleaching agents, such as NaClO$_2$, and chlorite holocellulose, a combination of α-cellulose and hemicellulose, was obtained.

Preparation of chlorite holocellulose

A suitable amount of dewaxed and depectinized jute fiber was treated with a 0.7% NaClO$_2$ solution at pH 4 and at 90–95°C for 90 min in a liquor ratio of 1 : 80. After being washed with distilled water, chlorite holocellulose was treated with a 0.2% Na$_2$S$_2$O$_5$ solution for 15 min.

Separation of α-cellulose from hemicellulose

The dried chlorite holocellulose was treated with a 24% KOH solution for 4 h with occasional stirring in a liquor ratio of 1 : 100. α-Cellulose was separated by filtration, washed thoroughly with a 2% acetic acid solution and then with distilled water, and dried at 105°C to a constant weight. The amount of α-cellulose thus obtained was deducted from the weight of holocellulose and gave the amount of hemicellulose.

Graft copolymerization of the jute fiber with the methacrylate monomers

Purified MMA or EMA was dissolved in warm water, and then a 1 g/L soap solution (surface-active agent) was added to make a suspension of the monomer.

The required amounts of the monomer (0.188 mol/L MMA and 0.159 mol/L EMA), initiator (7.40×10^{-4} mol/L), and catalyst (7.19×10^{-4} mol/L) were placed in a 100-mL, stoppered Erlenmeyer flask in a liquor ratio of 1:50. The polymerization was started at 30°C by the addition of a jute sample in a nitrogen atmosphere and the stoppering of the flask. The temperature was slowly increased almost to the boiling point within 30 min; the reaction continued for 3 h with occasional stirring and then was allowed 30 min more as the bath cooled down. Homopolymers from the grafted fiber were removed by thorough washing with distilled water and by repeated refluxing in warm acetone.[9,10]

The graft yield and grafting efficiency were calculated according to the following formulas:[10]

$$\text{Graft yield } (\%) = (B - A)/A \times 100$$

$$\text{Grafting efficiency } (\%) = (B - A)/C \times 100$$

where A is the weight of the bleached jute fiber before grafting, B is the weight of the grafted jute fiber after extraction with acetone, and C is the weight of the total monomer used for grafting.

Method of dyeing

Two direct dyes, Direct Red 81 and Direct Orange 31, were used. The dye bath was prepared with 1% dye and 5% cobalt sulfate as the electrolyte, on the basis of the fiber, in a liquor ratio of 1 : 40. Dyeing was performed at 80°C for 60 min with occasional stirring.[5] After dyeing, the fiber was washed well with water and dried. The absorption of the dye was determined colorimetrically as follows:

$$\text{Absorption of dye } (\%) = (D_o - D_e)/D_o \times 100$$

where D_o and D_e are the original and exhausted dye-bath concentrations, respectively.

IR spectroscopy

IR spectra of unmodified and selectively modified jute fibers were recorded with a Shimadzu (Kyoto, Japan) IR-470 spectrophotometer with the KBr pellet technique. The dried fiber samples were crushed to a size finer than 20 mesh before being pelletized with KBr. The test KBr pellet contained about 1% powdered fiber.

RESULTS AND DISCUSSION

Optimization of graft copolymerization onto the bleached jute fiber

The effect of the monomer concentration (e.g., MMA and EMA) on the polymerization with the bleached jute fiber with the $K_2S_2O_8/FeSO_4$ redox system is shown in Figure 1. The graft yield percentage increased with an increase in the monomer concentration for both MMA and EMA, and after a certain value, it decreased with a further increase in the monomer concentration. Cellulose macroradicals generated by redox initiation systems are immobile, and for grafting, the monomer should be in the vicinity of the cellulose. The higher monomer molecule near the immobile cellulose macroradical in the polymerization medium acts in favor of molecular collision, enhancing polymerization in general or homopolymer formation in particular.[10,11] The reduced graft yield percentage beyond a certain level suggested that the rate of homopolymer formation prevailed over graft formation at higher monomer concentrations. The maximum graft yield was obtained when bleached jute fibers were grafted with 0.188 mol/L MMA and 0.160 mol/L EMA.

Figure 1 shows that the graft yield percentage of MMA was less than that of EMA. This was possibly caused by the following: (1) the molecular weight of EMA is higher than that of MMA, (2) the reactivity of the functional groups of EMA is higher than that of MMA (as the activation energy of EMA is lower than that of MMA, EMA is readily esterified with bleached jute fiber), and (3) the reactive thermodynamic feasibility of the monomers varies according to the substituents present in it. Substituents increase the reactivity of a monomer toward radical attack. The order of monomer reactivity corresponds to the order of increased resonance stabilization by the particular substituent of the radical formed from the monomer. Substituents composed of unsaturated linkages are most effective in stabilizing the radicals because of the loosely held π electrons, which are available for resonance stabilization. The structures of MMA [$CH_2=C(CH_3)-COOCH_3$] and EMA [$CH_2=C(CH_3)-COOC_2H_5$] show that a $-COOCH_3$ (methyl ester) group of MMA is replaced by a $-COOC_2H_5$ group from EMA. Therefore, EMA is more reactive than MMA. As a result, the maximum graft yield percentage was in favor of EMA. The effective graft yields were 15.5 and 30% for MMA and EMA, respectively, and were determined on the basis of the maximum grafting efficiency.

Figure 2 shows that no graft yield was obtained when bleached jute fiber was grafted with 0.00–4.44×10^{-4} mol/L initiator ($K_2S_2O_8$). In the absence of the initiator, cellulose could not be converted to cellulose macroradicals and, at low initiator concen-
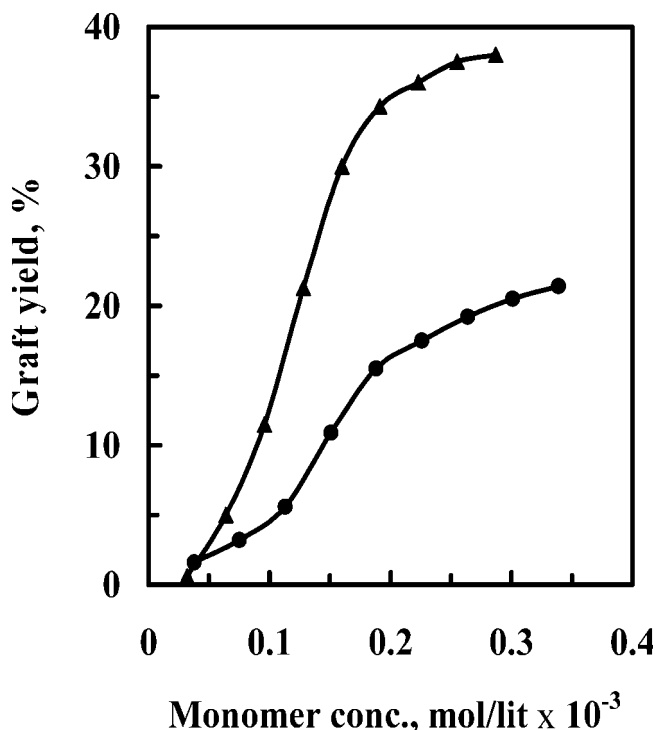
Figure 1 Effect of the monomer concentration on the grafting of (●) MMA and (▲) EMA onto bleached jute fiber ([$K_2S_2O_8$] = 7.40 × 10^{-4} mol/L, [$FeSO_4$] = 7.19 × 10^{-4} mol/L, temperature = 100°C, time = 180 min, liquor ratio = 1 : 50).

trations below 4.44 × 10^{-4} mol/L, failed to induce the graft polymerization of the monomers onto the jute fiber.[7] The graft yield percentage increased with an increase in the initiator concentration up to 8.88 × 10^{-4} mol/L for both MMA and EMA and thereafter decreased. The increasing trend of the graft yield percentage might be due to the formation of a great number of active sites on the backbone of the jute fiber. The retarding effects of the graft yield percentage above 8.88 × 10^{-4} mol/L $K_2S_2O_8$ might be due to the formation of the homopolymer by an excess of primary radicals $SO_4^{-•}$ and $^•OH$ formed from the initiator and/or premature termination of the growing grafted chains by an excess of primary radicals. Thus, the effective initiator concentration was 8.88 × 10^{-4} mol/L for MMA and EMA, and the corresponding graft yield percentages were 16.6 and 36%.

Figure 3 shows that the graft yield percentage increased with an increase in the ferrous sulfate concentration up to 4.32 × 10^{-4} mol/L for MMA and 5.76 × 10^{-4} mol/L for EMA and thereafter decreased. With an increase in the Fe(II) ion in the polymerization medium, the Fe(II)–$K_2S_2O_8$ transient adduct increased; it could be decomposed to $SO_4^{-•}$ and $^•OH$ radicals, which resulted in the production of jute macroradicals at a faster rate. The decrease in the

graft yield percentage beyond 4.32 × 10^{-4} mol/L Fe(II) for MMA and 5.76 × 10^{-4} mol/L Fe(II) for EMA by a detrimental factor arising from excess Fe(III) ions produced the oxidation of excess Fe(II) ions, leading to the retardation of the rate through premature termination of the growing grafting chains. The effective concentrations of ferrous sulfate were 4.32 × 10^{-4} mol/L for MMA and 5.76 × 10^{-4} mol/L for EMA, corresponding to 18 and 38% grafting, respectively.

The graft copolymerization of MMA and EMA onto jute fibers was studied at different temperatures ranging from 30 to 90°C. Figure 4 shows that the graft yield percentage increased initially with the rise in the temperature from 30 to 60°C for both MMA and EMA and then fell with a further increase in the temperature. The increase in the temperature increased the rate of diffusion of the monomers into the fiber matrix.[12] Hence, a rise in the temperature up to 60°C increased the rate of production of active free radicals, and this increased the number of grafting sites at a higher rate. An increase in the rate of homopolymer formation and premature termination of the growing grafted chains might have caused a decrease in the graft yield percentage beyond 60°C. The effective modification temperature for both MMA and EMA was 60°C. At an effective temperature, the graft yield percentages of MMA and EMA were 18 and 38.5%, respectively.
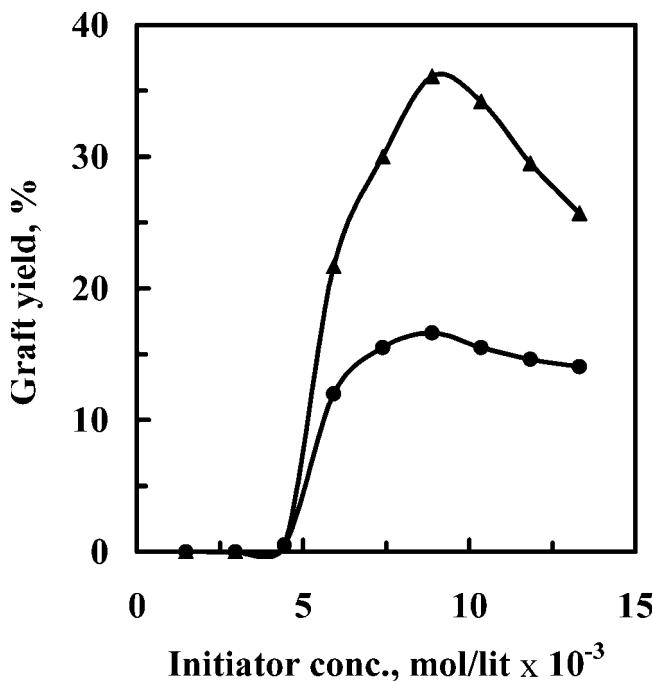

Figure 2 Effect of the initiator ($K_2S_2O_8$) concentration on the grafting of (●) MMA (0.188 mol/L) and (▲) EMA (0.159 mol/L) onto bleached jute fiber ([$FeSO_4$] = 7.19 × 10^{-4} mol/L, temperature = 100°C, time = 180 min, liquor ratio = 1 : 50).
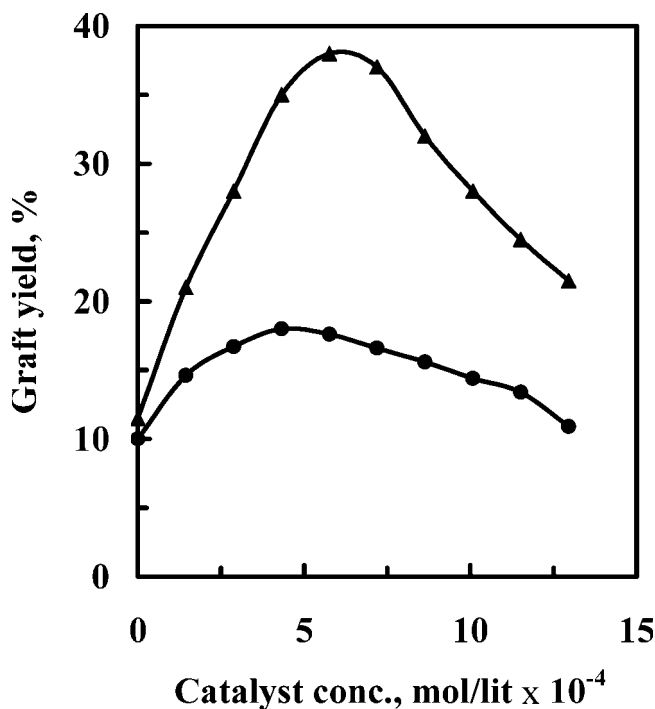
Figure 3 Effect of the catalyst (FeSO$_4$) concentration on the grafting of (●) MMA (0.188 mol/L) and (▲) EMA (0.159 mol/L) onto bleached jute fiber ([K$_2$S$_2$O$_8$] = 7.40 × 10^{-4} mol/L, temperature = 100°C, time = 180 min, liquor ratio = 1 : 50).

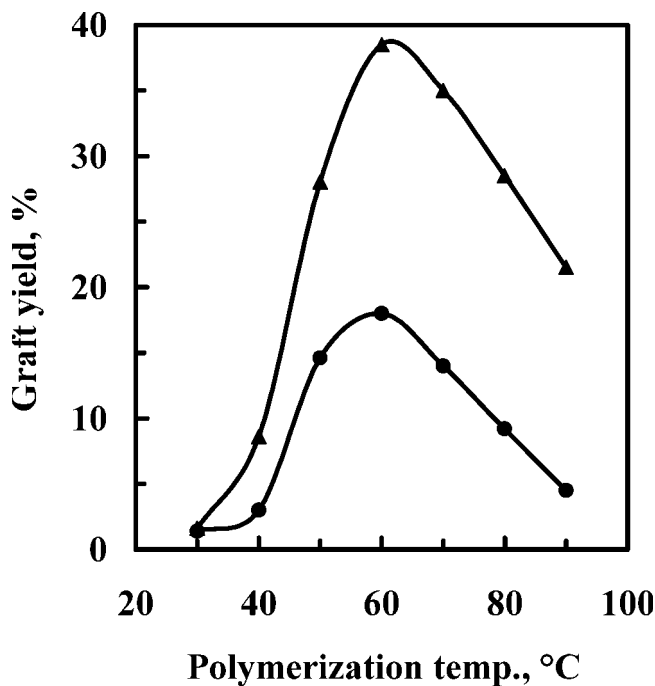**Figure 4** Effect of the polymerization temperature on the grafting of (●) MMA (0.188 mol/L) and (▲) EMA (0.159) onto bleached jute fiber ([K$_2$S$_2$O$_8$] = 7.40 × 10^{-4} mol/L, [FeSO$_4$] = 7.19 × 10^{-4} mol/L, time = 180 min, liquor ratio = 1 : 50).

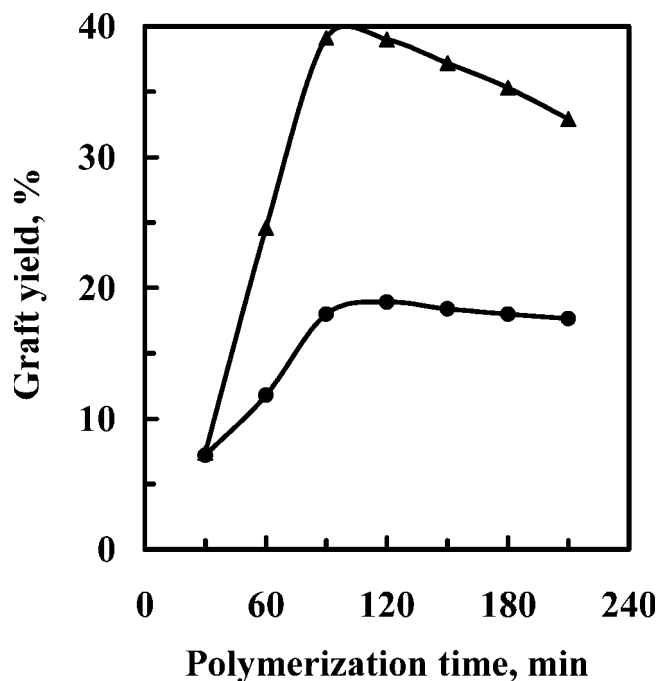**Figure 5** Effect of the polymerization time on the grafting of (●) MMA (0.188 mol/L) and (▲) EMA (0.159 mol/L) onto bleached jute fiber ([K$_2$S$_2$O$_8$] = 7.40 × 10^{-4} mol/L, [FeSO$_4$] = 7.19 × 10^{-4} mol/L, temperature = 100°C, liquor ratio = 1 : 50).

Figure 5 shows that the graft yield percentage increased with an increase in the reaction time up to 120 min for MMA and up to 90 min for EMA and thereafter decreased to some extent. The rate of the graft yield increased with an increase in the time because of the maximum polymerization reaction as sufficient time enhanced the swelling of the fiber and diffusion of both the monomer and initiators and a minimum homopolymerization reaction. The decrease in the graft yield percentage might be due to the partial dissolution of the grafted fiber upon prolonged exposure to heat.[13,14] The effective graft yield percentage of MMA at 120 min was 18.9%, and that of EMA at 90 min was 39%. The effective optimized conditions of grafting MMA and EMA onto jute fiber are listed in Table I.

Effect of grafting MMA and EMA onto the jute constituents

Jute, a multicellular fiber, has been analyzed, and the estimated average composition of the *C. olitorius* variety of raw jute is 58.59% α-cellulose, 23.01% hemicellulose, 12.97% lignin, 0.99% pectic matter, 0.94% fatty and waxy matter, and 3.50% aqueous extract. The main constituents of jute are α-cellulose, hemicellulose, and lignin, and the rest are very minor in proportion and therefore have little influence to

TABLE I
Effective Optimized Conditions for Modification

Monomer	Monomer concentration (mol/L)	Initiator concentration (mol/L)	Catalyst concentration (mol/L)	Reaction temperature (°C)	Reaction time (min)
MMA	0.188	8.88×10^{-4}	4.32×10^{-4}	60	120
EMA	0.159	8.88×10^{-4}	5.76×10^{-4}	60	90

the structure of jute fiber. Therefore, the structures of cellulose, hemicellulose, and lignin and the modes of combinations that exist between them dominate the structure of jute fiber.

The optimized grafting conditions of bleached jute fiber were applied to investigate the effect of grafting onto jute constituents. Table II shows the effect of grafting MMA and EMA onto jute constituents. In this experiment, dewatered, dewaxed, depectinized, bleached, double-bleached, and triple-bleached fibers and α-cellulose were used. The dewatered, dewaxed, and depectinized fibers contained the original amounts of lignin (12.97%) and hemicellulose (23.01%). When these fibers were grafted with MMA and EMA, the graft yield percentages of the dewatered, dewaxed, and depectinized fibers were 12.0, 12.9, and 10.3% for MMA and 16.4, 19.2, and 18.3% for EMA, respectively. This result indicated that the aqueous extract, fatty and waxy matter, and pectic matter had little effect on the total amount of grafting. However, the graft yield percentages of bleached, double-bleached, and triple-bleached fibers and α-cellulose were 18.9, 20.8, 19.0, and 11.0% for MMA and 38.8, 39.1, 36.8, and 12.0% for EMA, respectively. The graft yield percentages of bleached, double-bleached, and triple-bleached fibers were higher than that of α-cellulose. Table III shows the effect of lignin and hemicellulose of jute on grafting. The bleached fiber, double-bleached fiber, 3, 6, and 12% KOH treated, double-bleached fiber, and α-cellulose contained 4.4, 1, 0, 0, 0, and 0% lignin and 18.9, 19.7, 9.6, 5.6, 1.4, and 0% hemicellulose, respectively. When these fibers were grafted with MMA and EMA, the graft yield percentages were 18.9, 20.8, 16.0, 13.8 12.4, and 11.0% for MMA and 38.8, 39.1, 21.8, 16.0, 14.0, and 12.0% for EMA, respectively. From these results, it is clear that the decrease in the hemicellulose content in jute reduced the graft yield percentage for both MMA and EMA. This means that hemicellulose in jute promoted the grafting reactions. Thus, the graft yield of jute fibers with MMA and EMA mainly depended on the amount of hemicellulose as well as other jute constituents.

TABLE II
Effect of Grafting MMA and EMA onto the Jute Constituents

Sample	Graft yield (%)	
	MMA	EMA
Dewatered fiber	12.0	16.4
Dewaxed fiber	12.9	19.2
Depectinized fiber	10.3	18.3
Single-bleached fiber	18.9	38.8
Double-bleached fiber	20.8	39.1
Triple-bleached fiber	19.0	36.8
α-Cellulose	11.0	12.0

Physical characteristics of the grafted jute fiber

IR spectra of the grafted jute fiber

The IR spectra of bleached, MMA-grafted, and EMA-grafted jute fibers are shown in Figure 6. The spectra of the grafted samples are more or less similar in nature to that of the bleached jute fiber, except at the peaks at 1730 and 1729–1732 cm^{-1}. The additional peaks at 1730 and 1729–1732 cm^{-1} for the MMA- and EMA-grafted fibers, respectively, were due to the ester carbonyl groups of MMA and EMA.

TABLE III
Effect of Grafting MMA and EMA onto α-Cellulose, Hemicellulose, and Lignin of Jute Fiber

Sample	α-Cellulose (%)	Hemicellulose (%)	Lignin (%)	Grafting (%)	
				MMA	EMA
Bleached fiber	76.7	18.9	4.4	18.9	38.8
Double-bleached fiber	79.3	19.7	1.0	20.8	39.1
3% KOH treated, double-bleached fiber	90.4	9.6	0.0	16.0	21.8
6% KOH treated, double-bleached fiber	94.4	5.6	0.0	13.8	16.0
12% KOH treated, double-bleached fiber	98.6	1.4	0.0	12.4	14.0
α-Cellulose	100.0	0.0	0.0	11.0	12.0

These additional peaks in the spectra of the grafted fibers suggested that the monomers were grafted through chemical bonds with the bleached fiber.

Dyeing behavior of the grafted fiber

Table IV shows the exhaustion of direct dyes by bleached and grafted jute fibers. The dye exhaustion of the bleached jute fiber was greater than that of the modified fiber, and the dye exhaustion decreased with an increase in the graft yield percentage. The dye absorption of a fiber depends on the availability of the total external surface of the fiber pores or cavities in a fixed amount of the fiber and the attractive forces between the fiber and dye ions. The bleached jute fiber had more available pores or cavities than the modified jute fiber, which on grafting were blocked. Hence, the dye exhaustion of the grafted fiber was less than that of the bleached jute fiber. It may also be explained that the hydrophobic nature
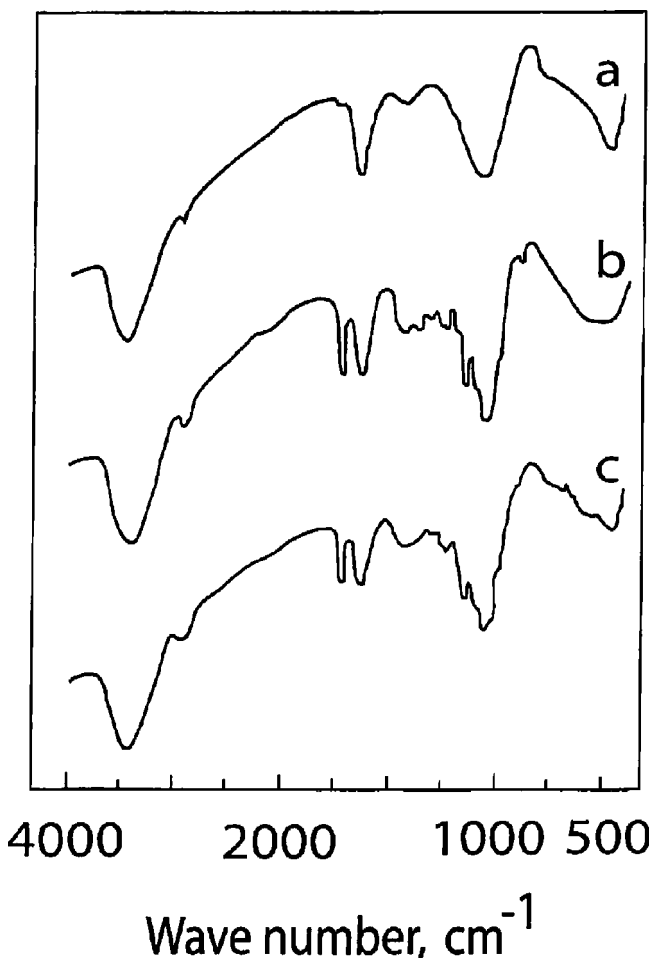
TABLE IV
Effect of Dyeing on Bleached and Grafted Jute Fiber Dyed with Direct Red 81 and Direct Orange 31

	Dye exhaustion (%)	
Sample	Direct Red 81	Direct Orange 31
Bleached fiber	90.0	83.8
MMA-grafted fiber (graft yield = 5%)	89.2	83.8
MMA-grafted fiber (graft yield = 15%)	87.8	83.2
MMA-grafted fiber (graft yield = 19%)	87.1	82.6
EMA-grafted fiber (graft yield = 5%)	89.0	83.6
EMA-grafted fiber (graft yield = 20%)	86.9	82.4
EMA-grafted fiber (graft yield = 30%)	85.6	81.4

of the grafted fiber increased with an increase in the grafting amount, and so the dye exhaustion of the bleached jute fiber was higher than that of the grafted fiber.

CONCLUSIONS

The effect of grafting methacrylate monomers onto jute constituents depends on the parameter variables and on the constituents of jute fiber. Among the main constituents of jute fiber, grafting depends mainly on the amount of hemicellulose present. Hemicellulose has many more free hydroxyl groups than α-cellulose, and these hydroxyl groups are much more responsible for forming jute macroradicals and graft copolymers than α-cellulose.

Figure 6 IR spectra of (a) bleached jute fiber, (b) MMA-grafted fiber, and (c) EMA-grafted jute fiber.

References

1. Ushakov, S. N. Fiz-Mat Nauk 1946, 1, 35.
2. Samal, R. K.; Sahoo, P. K.; Samantary, H. S. J Macromol Sci Rev Macromol Chem Phys 1986, 26, 81.
3. Habibuddowla, M. ACS Symp Series 1982, 187, 73.
4. Farouqui, F. I.; Mondal, M. I. H. Rajshahi Univ Stud Part B 1989, 17, 1.
5. Mondal, M. I. H.; Farouqui, F. I.; Enamul Kabir, F. M. Cell Chem Technol 2002, 36, 471.
6. TAPPI Standard Methods; TAPPI: New York, 1993.
7. Abou-Zeid, N. Y.; Higazy, A.; Hebeish, A. Angew Makromol Chem 1984, 121, 69.
8. TAPPI Standard T 236 m-60; TAPPI: New York, 1960.
9. Mondal, M. I. H. J Appl Polym Sci 2003, 87, 2262.
10. Sikder, B.; Basak, R. K.; Mitra, B. C. J Appl Polym Sci 1995, 55, 1673.
11. El-Rafie, M. H.; Abdel Hafiz, S. A.; Hassan, S. M.; Hebeish, A. J Polym Polym Compos 1994, 2, 2.
12. Samal, R. K.; Nanda, C. N.; Satrusallya, S. C.; Nayak, B. L. J Appl Polym Sci 1983, 28, 1311.
13. Misra, A.; Mohanty, A. K.; Singh, B. C. J Appl Polym Sci 1987, 33, 2809.
14. Mohanty, A. K.; Singh, B. C.; Mishra, P. G. M J Appl Polym Sci 1986, 31, 1964.