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***Tamarindus indica* mucilage and its acrylamide-grafted copolymer as flocculants for removal of dyes**

Received: 12 September 2005
Accepted: 23 May 2006
Published online: 25 August 2006
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Abstract The application of *Tamarindus indica* seed mucilage (Tam), a food grade polysaccharide, and its acrylamide grafted copolymer as flocculants was assessed for the first time for removal of various types of dyes from model textile wastewater containing azo, basic, and reactive dyes. Acrylamide grafted *T. indica* mucilage (Tam-g-PAM) was obtained by ceric ion initiated polymerization technique. A series of contact time experiments were conducted to assess the system variables such as concentrations of mucilage and dyes and pH.

These flocculants reduce the dye concentration by flocculation and settling. The grafted copolymer, Tam-g-PAM, showed better results for dye removal. The optimal flocculant concentration, which was required to effect flocculation, was independent of dye concentration within the range examined. Both the flocculants performed better for removal of azo dyes than for reactive and basic dyes.

Keywords *Tamarindus indica* mucilage · Tam-g-PAM · Dyes · Flocculation · Jar test method

Introduction

The textile industry is one of those industries that consume considerable amounts of water in the manufacturing process. Water is primarily employed in the dyeing and finishing operations in which the clothes are dyed and processed to finished products. These activities generate highly colored effluents and should be treated before discharge to avoid negative environmental impacts.

Color is one of the most obvious indicators of water pollution. The discharge of colored wastewater is not only damaging to the aesthetic nature of the receiving streams, but the colored compounds may also be toxic to aquatic life. There are more than 100,000 dyes available commercially, and most of these compounds are very stable and difficult to decolorize due to their complex structures. Reactive and acidic dyes are generally used in textile industries due to their color fastness. Azo dyes are a ubiquitous class of synthetic organic pigments that represent an increasing environmental danger [9]. The toxicity and potential carcinogenic nature of these dyes pose a specific threat to the environment. As tougher environ-

mental legislation is enforced in most countries, there is a greater need for cost-effective treatment systems. The conventional treatments of wastewater containing dyestuff include adsorption [2, 16, 19], nanofiltration by membranes [8], chemical coagulation, and oxidation [7].

Many flocculants/coagulants are widely used in conventional wastewater treatment processes. For this purpose, organic polymers are usually favored over inorganic salts. Although organic polymeric flocculants are more costly, they tend to produce less sludge as compared to their inorganic counterparts. The use of natural polymers, mainly polysaccharides, as flocculants is gaining importance in water treatment by virtue of being biodegradable, easily available from reproducible resources, and usually nontoxic [15]. These natural polymers suffer from various drawbacks such as a shear instability, uncontrolled biodegradability, and varying inefficiency. These shortcomings may be overcome by developing copolymers based on efficient but shear-degradable acrylic polymer and stable polysaccharides. It is observed that grafting of shear degradable polymers onto rigid polysaccharide backbone provides fairly shear stable systems. Recently,

we have reported the use of some natural and grafted polymers for the treatment of various types of wastewater [1, 10, 12–14]. In the present study, *Tamarindus* mucilage was chosen as flocculant for the dye solutions as tamarind trees are found in abundance in this region and using its seeds for various applications is very cost-effective. Mucilage extracted from the seeds is called “aging free starch” because its property is similar to starch but is more stable. The various applications of Tam include thickening sauce, ice cream, dressing, and processed vegetables. It is expected to find new applications. A brief evaluation of the effectiveness of pure *Tamarindus* mucilage and its polyacrylamide (PAM) grafted copolymer as flocculants for color removal is reported in this paper. The variables studied are the flocculant dose, contact time, and pH.

Materials and methods

Tamarind mucilage (Tam) was extracted from the milled seeds of *Tamarindus indica* with hot water. For deproteinization, it was treated with 0.3 N Ba(OH)₂–5% aqueous ZnSO₄·7H₂O [18]. All the chemicals were of analytical grade and were used as received without any purification. Isopropanol and buffer tablets were purchased from BDH (India), and acetone, acrylamide, ceric ammonium nitrate, hydroquinone, buffer tablets, and nitric acid from were purchased from S.D.Fine-Chem (India). The azo dyes [Congo red (CR) and naphthalene black (NB)], basic dyes [methylene blue (MB) and rhodamine B (RB)], and reactive dyes [reactive black (Rb) and reactive violet (RV)] used were of commercial grade. Their structures are given in Table 1.

Preparation of Tam-g-PAM copolymer

Tam-g-PAM was synthesized by grafting acrylamide onto tamarind mucilage by a radical polymerization method in aqueous system, using ceric ion/nitric acid redox initiator, and the percent grafting of acrylamide (PG) was calculated by the standard equation [6, 11].

The structure of the flocculants was confirmed by Fourier transform (FT) IR spectroscopy (Brucker Vector 22 spectrophotometer). The flocculants Tam and Tam-g-PAM were soluble in cold water. The viscosity of their aqueous solutions was measured by an Ostwald viscometer. The intrinsic viscosity measured from the point of intersection was obtained after extrapolation of two plots, i.e., η_{sp}/C vs C and $\ln \eta_{rel}/C$ vs C to zero concentration, where C is the concentration of polymer in grams per deciliter, and $\eta_{sp}/C = \eta_{rel} - 1/C$, where $\eta_{rel} = \eta/\eta_0 = t/t_0$ and t is the time of flow of the solvent at the time of measurement.

Flocculation experiments

The jar test is the most widely used method for evaluating and optimizing the flocculation/coagulation processes [15]. This study consists of batch experiments involving rapid mixing, slow mixing, and sedimentation. The apparatus allowed six beakers to be agitated simultaneously. In each case, 300 ml flocculant-dye solution was agitated in a flocculator at 100 rpm for 1 min, and then 30 rpm was quickly established for 10 min. After slow mixing, the beakers were carefully removed from the flocculator, and the solution was allowed to settle for approximately 10 min. Portions of the settled solutions are then removed and tested at definite intervals. The portions of the supernatant solution taken out were analyzed spectrophotometrically (Perkin Elmer, Lambda 40, UV–Vis spectrophotometer). Several contact time experiments were undertaken to assess the effect of system variables. The different dye concentrations chosen for jar experiments were in the range of 1 to 50 mg/l.

The known concentrations of dyes solutions were prepared to get optimal time and flocculant dose for the treatment of simulated wastewater samples. The concentrations of dyes were analyzed at different wavelengths so as to obtain maximum absorbance. All tests were done at room temperature (32±2 °C) to eliminate any temperature effects. The pH values for the test solutions were measured by Microprocessor pH meter CP931.

The percent dye removal was calculated from initial (C_o) and final equilibrium (C_e) concentrations of test solutions as follows [17]:

$$\% \text{ Dye Removal} = \frac{C_o - C_e}{C_o} \times 100. \quad (1)$$

Statistical analyses

The data obtained for percent dyes removal by *Tamarindus* mucilage were analyzed statistically using Matlab software (The Mathwork, USA). Triplicates were done in each case and the experimental design was a randomized design in a controlled experimental setup.

Result and discussion

Characterization of Tam and Tam-g-PAM

The polysaccharide Tam consists of D-glucose, D-xylose, and D-galactose in 3:2:1 ratio and is amorphous in nature. The FTIR spectra of Tam and Tam-g-PAM (PG=66.24) are shown in Fig. 1. The FTIR spectrum of Tam-g-PAM is different from that of Tamarind mucilage by showing characteristic peaks at 1,665.4 cm^{−1} of C=O of amide, at

Table 1 Structures of the dyes used

Dyes	Structure
Congo Red $\lambda_{\text{max}} = 650\text{nm}$	
Naphthalene Black $\lambda_{\text{max}} = 510\text{nm}$	
Methylene Blue $\lambda_{\text{max}} = 480\text{nm}$	
Rhodamine B $\lambda_{\text{max}} = 610\text{nm}$	
Reactive Black $\lambda_{\text{max}} = 530\text{nm}$	
Reactive Violet $\lambda_{\text{max}} = 430\text{nm}$	

1,527.3 cm^{-1} of $-\text{NH}$ bending, at 1,383.1 cm^{-1} of $-\text{CN}$ stretching, and at 1,283.5 cm^{-1} of $-\text{C}-\text{C}-\text{N}$ asymmetric. Moreover, the broadening and shifting of the band coming after 3,000 cm^{-1} towards slightly higher wave number in

Tam-g-PAM as compared to that of pure mucilage was also expected due to the overlapping of $-\text{NH}$ (amide) and $-\text{OH}$ (mucilage) bands.

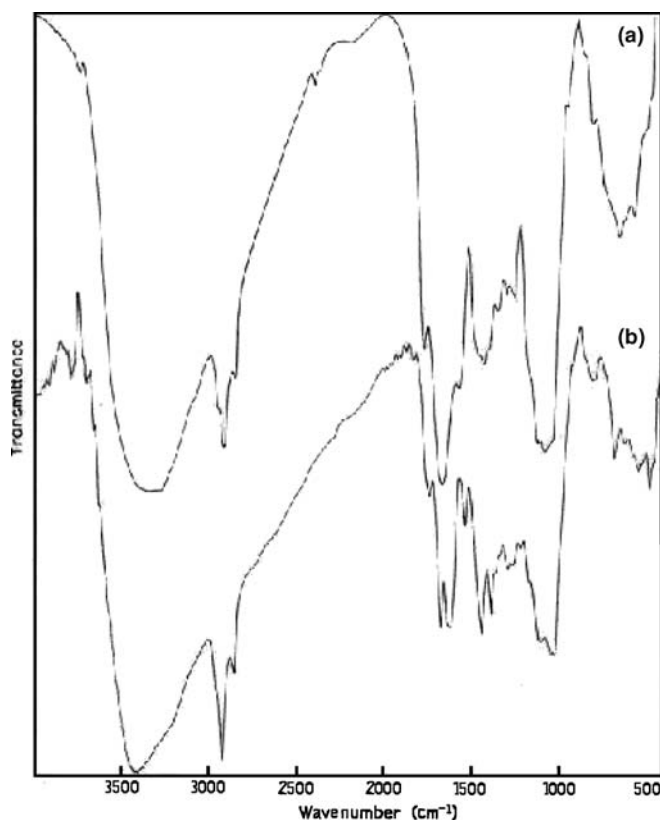


Fig. 1 IR spectra of a Tam and b Tam-g-PAM

The aqueous solutions of Tam and Tam-g-PAM are neutral in nature at pH 7, but at acidic pH, Tam-g-PAM has positive surface charge due to lone pair of electrons on nitrogen of amide groups. The possible binding sites for dye molecules in Tam are the free hydroxyl groups, whereas in grafted copolymer, in addition to hydroxyl groups there are amide groups as well. The intrinsic viscosity of *Tamarindus* mucilage was found to be 3.20 dl/g, whereas intrinsic viscosity of Tam-g-PAM was 6.58 dl/g.

Effect of flocculant dose on percent dye removal

The data showing the effect of variation in the flocculant dose, i.e., Tam and Tam-g-PAM, on the percent removal of azo (CR, NB), basic dyes (MB, RB), and reactive dyes (Rb,

RV) are given in Tables 2 and 3, respectively. It was observed with each flocculant that the percent removal increased with an increase in flocculant dose only up to a certain level and then a decreasing trend in dye removal was observed with a further increase in dose level. The optimum flocculant dose was 10 mg/l for azo and basic dyes and 15 mg/l for reactive dyes when Tam was used as flocculant, whereas with Tam-g-PAM, it was 5 mg/l was for all the dyes.

In the present experimental conditions, it was very likely that the polymer bridging played a large part in the flocculation process and the higher the dosage of flocculant, aggregation between colliding particles was more likely to occur. This trend (increasing and then decreasing) in percent removal was because the optimum amount of flocculant in the suspension caused a larger amount of dye particle to aggregate and settle. However, an over optimum amount of mucilage in the dye solution would cause the aggregated particle to redisperse and would also disturb particle settling [3].

Effect of dye concentration on percent dye removal

The effect of variation of dye concentration on the percent removal of dyes by Tam and Tam-g-PAM is given in Tables 4, 5, and 6. It is apparent from the data given in tables that the percent removal of the dyes decreased with increase in their initial concentrations except in the case of reactive dyes where the percent removal increased with an increase in initial dye concentration up to a certain level and then started to decrease. The optimum concentration of azo and basic dyes was 1 mg/l with both the flocculants used. For reactive dyes, it was 2 and 5 mg/l with Tam-g-Pam and Tam as flocculants, respectively.

The flocculating capacity of the flocculants probably became exhausted beyond optimum concentration of the dye. The explanation for this observation is based on a particle-polymer-particle complex formation in which polymer serves as a bridge. To be effective in destabilization, a polymer molecule must contain chemical groups, which can interact with sites on the surface of the colloidal particle. When a polymer molecule comes into contact with a colloidal particle, some of these groups adsorb at the particle surface, leaving the remainder of the molecule extending out into the solution. If a second particle with

Table 2 Percent removal of dyes on varying Tam concentration (dye concentration= 1 mg/l, contact time=180 min)

Serial no.	Flocculant dose (mg/l)	Percent removal					
		CR	NB	MB	RB	Rb	RV
1	1	11.65	9.48	11.68	9.25	14.87	12.67
2	5	15.32**	12.66**	14.53**	10.62*	17.13**	15.80**
3	10	36.89**	31.82**	27.82**	19.25**	23.43**	20.53**
4	15	32.21**	29.35*	16.91**	12.38**	26.50**	24.40**
5	20	28.83**	24.41**	07.32**	04.57**	19.05**	21.60**

* $P < 0.01$

** $P < 0.001$

Table 3 Percent removal of dyes on varying Tam-g-PAM concentration

Serial no.	Flocculant dose (mg/l)	Percent removal					
		CR	NB	MB	RB	Rb	RV
1	1	38.43	32.62	22.48	19.36	25.37	19.53
2	5	43.62**	43.18**	29.56**	27.12**	32.25**	26.78**
3	10	26.52**	24.41**	15.63**	14.43**	27.81**	24.38**
4	15	—	—	—	—	15.30**	14.32**
5	20	11.89**	12.62**	8.95**	7.89**	10.86**	11.67**
6	25	10.35*	11.93*	4.73**	3.62**	—	—

Dye concentration=1 mg/l

Contact time: CR and

NB=60 min, MB

and RB=120 min,

Rb and RV=30 min

* $P<0.01$ ** $P<0.001$

some vacant adsorption sites contacts these extended segments, attachment can occur. A particle-polymer-particle complex is thus formed in which the polymer serves as a bridge. If a second particle is not available, in time the extended segments may eventually adsorb on other sites on the original particle so that the polymer is no longer capable of serving as a bridge. There is a direct stoichiometric relationship between optimum polymer dosage and colloid concentration, and restabilization due to overdosing can occur.

Table 4 Percent removal of azo dyes on varying the dye concentration

Serial no.	Flocculant	Dye concentration (mg/l)	Percent removal	
			CR	NB
1	Tam	1	36.89	31.82
2	Tam	10	26.82**	22.62**
3	Tam	20	14.39**	11.98**
4	Tam	50	09.61**	9.43*
5	Tam	100	08.35**	8.61*
6	Tam-g-PAM	1	43.62	43.18
7	Tam-g-PAM	5	32.62**	28.81**
8	Tam-g-PAM	10	20.00**	15.63**
9	Tam-g-PAM	15	04.43**	7.28**
10	Tam-g-PAM	50	08.62**	7.01*

* $P<0.05$ ** $P<0.001$ **Table 5** Percent removal of basic dyes on varying the dye concentration

Serial no.	Dye concentration (mg/l)	Percent removal			
		MB		RB	
		Tam	Tam-g-PAM	Tam	Tam-g-PAM
1	1	27.82	29.56	19.25	27.12
2	10	12.67**	11.68**	9.67**	11.83**
3	20	7.23**	8.25**	8.43*	4.68**
4	50	4.56**	7.33*	4.21*	3.95*
5	100	3.21*	6.58*	2.10*	2.11*

* $P<0.01$ ** $P<0.001$

Effect of contact time on percent dye removal

The effect of percent removal of the dye with contact time is shown in Fig. 2. The removal of the dye in each case was rapid in the initial stages of contact time and gradually decreased with lapse of time and finally became constant. The maximum removal with pure mucilage was obtained in 3 h for all the dyes, whereas with Tam-g-PAM, maximum removal was obtained within 30 min for reactive dyes, in 1 h for azo dyes, and in 2 h for basic dyes.

A sloped curve indicates the flocculation capacity of the mucilage. The plot visualized three distinct phases. The first phase (initial steep slope) indicated the interaction of dye molecules with flocculant, which caused destabilization of the particles in suspensions, and they began to flocculate. The second phase of the plot showed a slight decrease in percent removal of the color; this might be due to the destabilization of the aggregated particles [5]. The third phase of the plot indicated attainment of stability by the flocs.

Effect of pH on percent dye removal

The data related to the removal of dyes as a function of pH are given in Table 7 and 8. The maximum percent removal of azo dyes CR and NB was 36.43 and 31.89%, respectively, using Tam as flocculant, whereas with Tam-g-PAM as flocculant, it was 45.32 and 43.68% for CR and

Table 6 Percent removal of reactive dyes on varying the dye concentration

Serial no.	Dye concentration (mg/l)	Percent removal			
		Rb		RV	
		Tam	Tam-g-PAM	Tam	Tam-g-PAM
1	1	26.50	32.25	24.40	26.78
2	2	29.32**	43.51**	26.68**	39.68**
3	5	31.65**	26.85**	28.95*	27.81**
4	10	25.01**	19.34**	23.36**	15.66**
5	15	16.93**	11.07**	14.31**	10.77**

* $P<0.01$ ** $P<0.001$

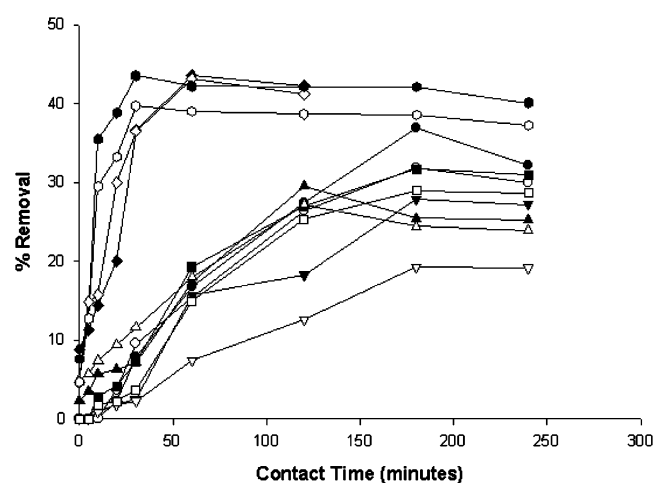


Fig. 2 Effect of percent removal with contact time. With Tam: ○ CR, ● NB, ▼ MB, ▽ RB, ■ Rb, □ RV. With Tam-g-PAM: ◆ CR, ◇ NB, ▲ MB, △ RB, ● Rb, ◊ RV

NB, respectively, at pH 6. It was observed that at pH below 6, both the azo dyes decolorized even without the addition of the flocculants. Therefore, at pH 4, the percent removal was measured with an increased dose of azo dyes (5 mg/l), and these values were not considered in the statistical analysis. The maximum removal observed for acidic dyes at acidic pH was justified on the fact that the acidic dyes would give colored anions when dissolved in water and these anionic species would adsorb better at low pH since negatively charged particles would start getting neutralized at low pH [4]. The better results obtained with Tam-g-PAM was attributed to the protonation of the acrylamide chains at low pH, thus increasing the capacity of the flocculant to absorb and retain more anions by forming an electrovalent bond between the dye and the copolymer.

The maximum percent removal of basic dyes was observed at alkaline pH with both the flocculants, since basic dyes give cations in solution and positively charged particles would start getting neutralized as pH of the medium increases beyond neutral pH [4]. At acidic pH,

Table 7 Percent removal of dyes on varying the pH with Tam as flocculant

Serial no.	pH	Percent removal					
		CR	NB	MB	RB	Rb	RV
1	1	—	—	—	—	52.13	41.65
2	2	—	—	—	—	51.19*	38.68**
3	4	38.89	34.45	12.68	9.65	46.85**	34.85**
4	6	36.43*	31.89**	27.31***	15.42***	32.69***	28.10***
5	7	24.82***	21.92***	35.42***	23.68***	25.41***	19.53***
6	9.2	16.36***	14.55***	38.96*	25.44*	17.38***	15.40**

Flocculant dose=10 mg/l (azo and basic dyes), 15 mg/l (reactive dyes); contact time=180 min (azo, basic, and reactive dyes); dye concentration=1 mg/l (azo and basic dyes), 5 mg/l (reactive dyes)

At pH 4, initial dye concentration of CR and NB was 5 mg/l.

* $P < 0.05$

** $P < 0.01$

*** $P < 0.001$

Table 8 Percent removal of dyes on varying the pH with Tam-g-PAM as flocculant

Serial no.	pH	Percent removal					
		CR	NB	MB	RB	Rb	RV
1	1	—	—	—	—	69.03	52.39
2	2	—	—	—	—	68.12*	52.27*
3	4	48.96	45.48	19.62	11.68	52.56***	41.45**
4	6	45.32*	43.68*	25.41***	21.10***	48.90**	37.76*
5	7	38.65**	35.50***	38.66***	28.53**	25.07***	21.49***
6	9.2	24.43***	19.38***	41.42*	30.68*	19.53***	15.36**

Flocculant dose=5 mg/l (azo, basic dyes, and reactive dyes); contact time=60 min (azo), 120 min (basic), and 30 min (reactive dyes); dye concentration=1 mg/l (azo and basic dyes), 2 mg/l (reactive dyes)

At pH 4, initial dye concentration of CR and NB was 5 mg/l.

* $P < 0.05$

** $P < 0.01$

*** $P < 0.001$

poor removal was seen that could be explained on the basis of much increase in the repulsive energy between the positively charged flocculant and the dye cations, which causes hindrance in floc formation. The maximum removal of MB and RB was 38.96 and 25.44%, respectively, when Tam was used as flocculant, while in the case of Tam-g-PAM, it was 41.42 and 30.68% for MB and RB, respectively. With Tam-g-PAM, the percent removal was not any better than that observed with Tam as flocculant. At basic pH, the amide groups were not protonated and the dye cations might adsorb on the copolymer surface to form a floc. With Tam-g-PAM, the maximum removal of Rb and RV was 69.03 and 52.39%, respectively, and with Tam as flocculant, the maximum removal was 52.13 and 41.65% for Rb and RV, respectively. The best results at acidic pH were expected since these dyes give anions in aqueous solution [4].

A statistical analysis using two-way analysis of variance shows that the results are significant. As the data indicates, the interaction of basic dyes with mucilage and its graft copolymer is quite different from the azo and reactive dyes. For all the dyes, initial dye concentration, flocculant dose, and treatment time influence the percent removal significantly. The change in pH is the major factor governing the percent removal of all the dyes. In case of the two azo dyes, the percent removal is highly significant on changing the pH value from acidic to alkaline. In the case of the two basic dyes, the percent removal is highly significant on changing the pH from acidic to neutral, whereas the change observed is moderately significant when pH changes from neutral to alkaline. In the case of the two reactive dyes, the

percent removal is highly significant on changing the pH value from acidic to alkaline.

Conclusions

A food grade polysaccharide, Tam, from a renewable source and its acrylamide-grafted copolymer were used as flocculants for removal of various types of dyes from simulated wastewater samples. From the present set of experiments, flocculation, using biodegradable materials, was shown to be a simple eco-friendly and economic treatment. Grafting of polyacrylamide did not affect the biodegradability of Tam although the shelf life was improved. The grafted copolymer, Tam-g-PAM, showed better flocculation efficiency for dye removal than the pure mucilage. These flocculants performed better for removal of azo and reactive dyes than that of basic dyes. The results found in this study do not suggest that Tam and Tam-g-PAM are very effective flocculants for the removal of dyes used. The possibility of their industrial application as flocculants for the removal of various types of dyes, especially anionic, may be explored for its much lower cost and biodegradability as compared to that of other flocculants.

Acknowledgement The authors are grateful to University Grants Commission, New Delhi for financial support of this study [Project No. F. 12-133 / 2001 (SR -1)].

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