

Microwave-assisted synthesis and characterization of polyacrylamide grafted co-polymers of *Mimosa* mucilage

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Abstract Response surface methodology was applied to optimize the microwave-assisted graft co-polymerization of acrylamide on *Mimosa pudica* seed mucilage. The effect of variables, microwave power and time of exposure, concentrations of acrylamide, mucilage and ammonium persulfate on grafting efficiency of graft co-polymerization was screened using Plackett–Burman experimental design. The results revealed that the concentration of acrylamide and mucilage are the most significant variables, which were further optimized using, a central composite design. A second-order polynomial equation fitted to the data was used to predict the response in the optimal region. The optimal grafting parameters provided graft co-polymer with grafting efficiency close to the predicted values. The proposed mathematical model is found to be robust and accurate for graft co-polymerization of acrylamide and *Mimosa* mucilage consistent with goals of maximizing grafting efficiency. The results of FT-IR, DSC, XRD, and SEM studies confirmed the formation of graft co-polymer of acrylamide and *Mimosa* mucilage.

Keywords Graft copolymers · Modeling · Microwave · Polyacrylamide

Introduction

Natural gums, mucilages and their derivatives are widely employed in pharmaceutical and food industry, as these are generally considered as non-toxic and safe for human and animal consumption [1]. These natural polysaccharides are obtained from plant exudates and seeds of land and marine plants sources. Mucilages are produced by normal metabolic processes and are usually formed from the cell wall

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or deposited as layers on it. Chemically, mucilages are polyuronides comprised of sugar and uronic acid units [2]. Mucilages are partially soluble in water in which they swell and form gel. Mucilages of okra, dika nut, *Ocimum gratissimum*, *Plantago ovata*, *Eulophia campestris*, and *Mimosa pudica* have earlier been evaluated for their pharmaceutical applications as binding agent [3–5], suspending agent [6], disintegrating agent [7] and as sustained release matrices [8, 9]. *Mimosa pudica* Linn. (fam: mimosaceae) is an annual creeping or perennial herb, native to Brazil. The seeds of *Mimosa* yield mucilage, which is composed of D-xylose and D-glucuronic acid.

Non-toxicity, low cost, and easy availability of natural polysaccharides makes them a preferred choice over synthetic polymers. However, they also possess certain drawbacks like uncontrolled hydration, microbial contamination, and drop in viscosity during storage, etc. The property of these natural polymers can be modified by hybridization with synthetic polymers. The chemical combination of natural and synthetic polymers yields new materials, which could have desirable properties. The properties of natural polymer have earlier been easily modified by graft co-polymerization and formation of interpenetrating network hydrogels with vinyl monomers [10]. Mucilages obtained from fenugreek seeds [11], okra seeds [12], *Plantago psyllium* husk [13, 14], *Coccinia indica* fruits [15], and *Tamarindus indica* seeds [16] have earlier been modified by ceric-induced graft co-polymerization of vinyl monomers.

During an earlier study in our laboratory microwave-assisted graft co-polymerization was employed to prepare graft co-polymer of xanthan gum and acrylamide [17]. The microwave irradiation is an efficient method, which results in rapid transfer of energy in the bulk of the reaction mixture. The microwave-assisted graft co-polymerization requires a very short reaction time and proceeds even in the absence of any redox initiator [18]. In this study, the graft co-polymerization of acrylamide onto *Mimosa pudica* seed mucilage was carried out using microwave-assisted graft co-polymerization. Various variables affecting the microwave-assisted graft co-polymerization of acrylamide on mucilage were first screened using Plackett–Burman screening design. The significant variables so screened were further optimized using central composite design (CCD) for preparing the graft co-polymer of acrylamide on *Mimosa* mucilage. The graft co-polymer of acrylamide and *Mimosa* mucilage, so prepared, was characterized by FT-IR, DSC, XRD, and SEM studies.

Experimental

Materials

Mimosa pudica seeds were procured from the local market of Hisar (India) and authenticated by the taxonomists of Forest Research Institute (Dehradun, India). A voucher specimen was deposited with the Department of Pharmaceutical Sciences (authentication voucher no: Pcog/2007/65). Acrylamide and ammonium persulfate were procured from Sisco Research Laboratory (Mumbai, India), and acrylamide used in the study was recrystallized from methanol twice, followed by drying at

40 °C in vacuum oven. All other chemicals used were of reagent grade and were used as such.

Extraction of mucilage

Mimosa pudica seeds (100 g) were soaked in de-ionized water (1000 mL) for 10 h; the hydrated mucilage along with seeds was spread in thin layer on the stainless steel tray and dried in an oven at 50 °C for 4–5 h. The dried mucilage was scraped from the tray by blade and separated from the seeds by passing through No. 18 mesh. The mucilage was further, purified by winnowing to separate seed husk.

Preparation of graft co-polymer of acrylamide and mimosa mucilage

Microwave-assisted grafting of acrylamide on *Mimosa* mucilage was done using the method reported earlier [19]. Briefly, *Mimosa* mucilage was added to the aqueous solution of acrylamide and dispersed by stirring. For grafting with redox initiator, ammonium persulfate was added to this above solution. The solution so obtained was irradiated by microwave in domestic microwave oven (2300 ET-B, Bajaj Electricals Ltd., Mumbai, India) for different times and different power to prepare various batches of grafted mucilage. The grafted mucilage was treated with acetone, and washed with mixture of methanol:water (80:20) mixture to remove the unreacted monomer and reagent followed by drying in vacuum oven at 40 °C to a constant weight. The % grafting efficiency was calculated using the following equation [20].

$$\% \text{ Grafting efficiency (\%GE)} = \frac{(W_1 - W_0)}{W_2} \quad (1)$$

where, W_0 weight of *Mimosa* mucilage, W_1 weight of graft copolymer, and W_2 weight of acrylamide.

Plackett–Burman design

Plackett–Burman design is a useful and efficient mathematical approach employed to screen the effect of large number of factors on particular response by conducting minimum number of experimental runs [21]. In this study, microwave power, time of microwave exposure, concentration of mucilage, concentration of acrylamide, and ammonium persulfate were selected as the independent variables. Each variable was investigated at two levels, high level (+1) and low level (−1). The factor ranges were selected based on the prior knowledge about the system under study. The experimental design is presented in Table 1. The experimental design and analysis of the data were done using Design Expert software (Version 7.1.6, Stat-Ease Inc., Minneapolis, USA).

CCD

A CCD was applied to determine the optimum concentration of two significant variables screened from Plackett–Burman screening design. The effect of these variables on %GE was studied at three experimental levels −1, 0, +1. A face

Table 1 Plackett–Burman screening design

Exp. no.	X_1 (%)	X_2 (s)	X_3 (%)	X_4 (mmol)	X_5 (mmol)	X_6	X_7	X_8	X_9	X_{10}	X_{11}	Y_1 (%)
1	-1 (60)	-1 (60)	-1 (1)	-1 (2)	-1 (0)	1	1	-1	-1	-1	-1	0
2	-1 (60)	-1 (60)	-1 (1)	1 (8)	-1 (0)	-1	1	1	-1	-1	1	5.68 ± 1.78
3	-1 (60)	1 (120)	1 (2)	1 (8)	-1 (0)	-1	1	1	1	-1	-1	33.68 ± 4.3
4	1 (100)	-1 (60)	1 (2)	1 (8)	-1 (0)	-1	-1	1	1	1	-1	24.93 ± 4.02
5	1 (100)	-1 (60)	1 (2)	1 (8)	1 (10)	1	1	-1	1	1	-1	32.39 ± 2.98
6	1 (100)	1 (120)	1 (2)	-1 (2)	-1 (0)	1	1	1	-1	1	1	25.58 ± 3.91
7	-1 (60)	1 (120)	-1 (1)	1 (8)	1 (10)	1	-1	1	1	-1	1	4.39 ± 0.26
8	-1 (60)	-1 (60)	1 (2)	-1 (2)	1 (10)	-1	1	-1	1	1	1	11.97 ± 1.31
9	1 (100)	-1 (60)	-1 (1)	-1 (2)	1 (10)	1	-1	1	-1	1	-1	5.63 ± 0.86
10	-1 (60)	1 (120)	1 (2)	-1 (2)	1 (10)	1	-1	-1	1	-1	1	7.74 ± 1.20
11	1 (100)	1 (120)	-1 (1)	-1 (2)	-1 (0)	-1	-1	-1	-1	1	1	4.92 ± 0.64
12	1 (100)	1 (120)	-1 (1)	1 (8)	1 (10)	-1	-1	-1	-1	-1	-1	27.28 ± 1.90

Values in parenthesis show the actual values

X_1 microwave power, X_2 microwave time, X_3 concentration of nucleage, X_4 concentration of acrylamide, X_5 concentration of ammonium persulfate

Table 2 Central composite design

Exp. No.	X_3 (mg)	X_4 (mmol)	Y_1 (%)
1	-1 (1)	-1 (2)	1.37
2	-1 (1)	0 (5)	5.16
3	-1 (1)	1 (8)	16.33
4	0 (1.5)	-1 (2)	10.35
5	0 (1.5)	0 (5)	32.54
6	0 (1.5)	1 (8)	34.83
7	1 (2)	-1 (2)	26.50
8	1 (2)	0 (5)	37.17
9	1 (2)	1 (8)	33.88
10	0 (1.5)	0 (5)	39.27
11	0 (1.5)	0 (5)	37.32
12	0 (1.5)	0 (5)	25.32
13	0 (1.5)	0 (5)	26.03

Values in parenthesis show the actual values

centered full factorial central composite experimental design with five replicates at the central points and resulting in a total of 13 experiments, was used to evaluate the two chosen variables. The experiment was designed using Design Expert Software (Version 7.1.6, Stat-Ease Inc., Minneapolis, USA). The levels of CCD used in the study are presented in Table 2. The response generated was subjected to natural logarithmic transformation and analyzed using a second-order polynomial equation and the data were fitted into the equation by multiple regression procedure. The model equation used for analysis is as follows:

$$Y = \beta_0 + \beta_1 X_1 + \beta_2 X_2 + \beta_3 X_1 X_2 + \beta_4 X_1^2 + \beta_5 X_2^2 \quad (2)$$

where, β_0 is the intercept representing the arithmetic average of all quantitative outcomes of 20 runs; β_1 to β_5 are the coefficients computed from the observed experimental values of Y ; and X_1 and X_2 are the coded levels of the independent variable(s). The terms $X_1 X_2$ and X_i^2 ($i = 1-2$) represent the interaction and quadratic terms, respectively [22]. The interaction term show how the response changes when two factors were simultaneously changed. The second-degree term (X_i^2) is included to investigate non-linearity.

Characterization of mucilage and grafted mucilage

The graft co-polymer of acrylamide and *Mimosa* mucilage was characterized by FT-IR spectroscopy, differential scanning calorimetry, X-ray diffractometry, and scanning electron microscopy.

FT-IR spectroscopy

The samples were subjected to FT-IR spectroscopy in a Fourier-transform infrared spectrophotometer (Perkin Elmer, USA) in range of ($4000-500 \text{ cm}^{-1}$) as KBr pellet.

Differential scanning calorimetry

Differential scanning calorimetric thermogram of *Mimosa* mucilage, acrylamide, and graft co-polymer of acrylamide and *Mimosa* mucilage was recorded using differential scanning calorimeter (Q10, TA Systems, USA) in the temperature range of (40–250 °C) at a heating rate of 10 °C per minute in nitrogen atmosphere.

X-ray diffractometry

X-ray diffractogram of *Mimosa* mucilage, acrylamide, and graft co-polymer of *Mimosa* mucilage and acrylamide samples were recorded employing X-ray diffractometer (XpertPRO, Panalytical, Germany) using copper K α -radiation generated at 40 kV and 35 mA in the differential angle range of 10–80° (2 θ) using an X-ray diffractometer.

Scanning electron microscopy

Scanning electron micrographs of *Mimosa* mucilage, acrylamide, and graft copolymer of *Mimosa* and acrylamide samples were taken using a SEM (268 D, Fei-Philips Morgagni). These were coated with gold and mounted in a sample holder. The photomicrograph of sample was taken at an accelerating voltage at 15 kV at different magnifications.

Results and discussion

Screening phase

Conventionally graft co-polymerization of vinyl monomers is carried out by redox initiator induced graft co-polymerization method. Microwave-assisted graft co-polymerization has been found to be more efficient than the redox initiator induced graft co-polymerization [17]. Concurrent formation of homopolymer is the main constraint of graft co-polymerization resulting in low grafting yields. Concentrations of natural polymer, vinyl monomer, microwave power, and microwave exposure time were earlier reported to influence the grafting reaction [23]. Thus, these, variables were screened using Plackett–Burman design to study their influence on grafting. The averages of % grafting efficiency for graft co-polymerization of acrylamide on *Mimosa* mucilage are presented in Table 1. An ANOVA test was applied to the data corresponding to the design of Table 1, using the effect of dummy variables to obtain an estimate of standard errors in the coefficients. The results revealed that the factors X_3 (conc. of mucilage) and X_4 (conc. of acrylamide) with p -value of 0.0331 and 0.0386 are the most significant ($p < 0.05$) variables affecting the graft co-polymerization of acrylamide on *Mimosa* mucilage.

The polynomial model describing the correlation between the independent variables and % grafting efficiency of graft co-polymerization could be presented as:

$$Y_1\% = 15.35 + 4.77X_1 + 1.92X_2 + 7.36X_3 + 6.04X_4 - 0.45X_5 \quad (3)$$

The power of microwave exposure (X_1) and time of microwave exposure (X_2) showed a positive but insignificant influence on the %GE, while the concentration of ammonium persulfate (X_5) had a negative but insignificant influence on the %GE. There was a slight increase in the %GE, with increase in the microwave power and duration of exposure, which may be attributed to increase transfer of energy to reaction mixture. The slight decrease in the %GE in the presence of ammonium persulfate could be due to the formation of more homopolymer. Thus for the subsequent optimization phase, the power of microwave exposure and time of exposure were fixed at their maximum values, while ammonium persulfate was excluded from further trials.

CCD

CCD provides valuable information regarding the optimum level of each variable along with their interactions with other variables and their effects on the response. On the basis of Plackett–Burman screening, concentrations of mucilage (X_3) and acrylamide (X_4) were selected for CCD optimization. The experimental design for the two variables and the corresponding experimental data is shown in Table 2. The response generated in the experiments was subjected to natural logarithmic transformations, and the data was fitted into the second-order response surface model.

Table 3 shows the results of the ANOVA test on the quadratic regression model. The goodness of the model was checked by the determination coefficient R^2 (0.9775). The model F -value of 60.80 and very low probability value [$(P_{\text{model}} > F) = 0.0001$] implies that the model is significant. The “Lack of Fit F -value” of 0.83 implies that the “Lack of Fit” is not significant relative to the pure error. The Pred. R-Squared of 0.9173 is in reasonable agreement with the “Adj R-Squared” of 0.9614. “Adeq Precision”, measures the signal to noise ratio, a ratio greater than 4 is desirable. It was found to be 26.283, indicating adequate signal. The relatively lower coefficient of variation ($CV = 6.58\%$) indicates a better precision and reliability of the experiments carried out.

Table 3 ANOVA for the quadratic model

Source	SS	DF	MS	F-value	Prob > F
Model	11.45	5	2.29	60.80	<0.0001
Residual (error)	0.26	7	0.038	–	–
Lack of fit	0.10	3	0.034	0.83	0.5432
Pure error	0.16	4	0.041	–	–
Total	11.71	12			

$$r^2 = 0.9775, cv = 6.58, \text{adj. } r^2 = 0.9614$$

SS sum of squares, DF degrees of freedom, MS mean square

The polynomial model for Ln (%GE) (Y) can be expressed by the following equation:

$$Y = 3.40 + 0.94X_3 + 0.66X_4 - 0.56X_3X_4 - 0.65X_3^2 - 0.34X_4^2 \quad (4)$$

The results reveal that the concentration of mucilage (X_3) had the most pronounced effect on the grafting efficiency as it had the largest coefficient followed by concentration of acrylamide (X_4). The interaction terms (X_3X_4) and the quadratic terms of mucilage (X_3^2) and acrylamide (X_4^2) had a negative effect on the grafting efficiency. The linear effect of concentration of mucilage (X_3) and acrylamide (X_4) are highly significant indicating that they act as limiting factors and any subtle variation in their concentrations will alter the grafting efficiency.

Figure 1 displays the 3-D response surface plot showing the combined effect of concentration of acrylamide and mucilage on the %GE. These plots show the graphical representations of the regression equation and help in the identification of the type of interactions between the variables. The plots show a curvilinear relationship for %GE with a region of maxima lying between the mucilage concentrations of 1.5–2.0% and acrylamide concentration of 4.75–8.0 mmol. The increase in %GE with the increase in acrylamide concentration can be attributed to the greater availability of acrylamide molecules in the proximity of mucilage backbone, increasing the chances of molecular collision and hence grafting. The increase in %GE with increase in concentration of mucilage could be due to the availability of more sites for grafting. Further, a potential interaction between the concentration of mucilage and acrylamide, with one factor modifying the effect of another on grafting can be observed.

A numerical optimization tool of Design expert software, using the desirability approach was used to prepare graft co-polymer with desired grafting. In this study, optimization of independent variables was performed with a goal of maximizing

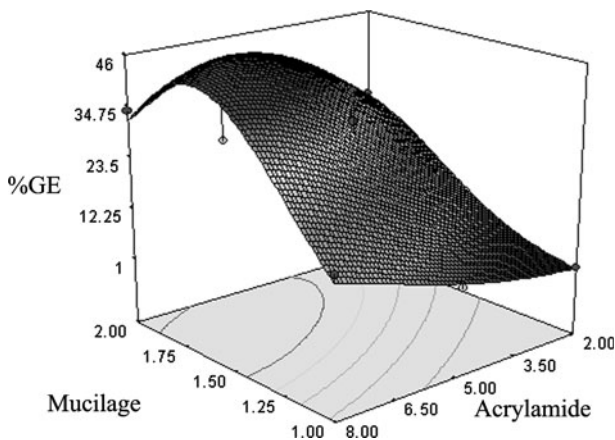


Fig. 1 Three dimensional response surface plot showing the combined effect of mucilage and acrylamide concentration on %GE

grafting efficiency. The optimal calculated parameters were concentration of mucilage (X_3) 1.92% and concentration of acrylamide (X_4) 4.91 mmol.

Validation of response surface methodology

To check the reliability of the developed mathematical model, grafting of the mucilage corresponding to the predicted mucilage and acrylamide concentrations, along with three additional random checkpoints covering the entire range of experimental domain was carried out. Table 4 lists the test conditions of the optimum and the random checkpoints, their experimental and predicted values for response variable, along with the calculated percentage prediction error. On linear correlation of the observed and predicted response variables, the value of correlation coefficient r^2 was found to be 0.994. Thus, the lower magnitudes of percentage prediction error (-4.97 to 5.17 for Y) as well as significant values of r^2 (>0.9) in the current study indicate the robustness of the mathematical model and high prognostic ability of response surface methodology. The optimized batch of grafted mucilage has maximum %GE of 41.59.

Characterization of grafted mucilage

Acrylamide grafted *Mimosa* mucilage was characterized by FT-IR, DSC, XRD, and SEM study. The FT-IR spectra of *Mimosa* mucilage showed the characteristic peaks at 3401 cm^{-1} owing to $-\text{OH}$ stretching of alcohol, at 1165 cm^{-1} owing to $\text{C}-\text{O}$ stretching of alcohol, and at 2922 cm^{-1} due to $\text{C}-\text{H}$ stretching of alkyl group. The spectra of acrylamide presented absorption bands at 3350 and 3187 cm^{-1} due to asymmetric and symmetric NH stretching of NH group. The amide-I band (CO stretching) appeared at 1674 cm^{-1} and amide-II (NH bending) appeared at 1611 cm^{-1} . The spectra of acrylamide presented a band at 1433 cm^{-1} which can be attributed to CN stretching, while the CH stretching appeared at 2820 cm^{-1} and CH out of plane bending at 988 cm^{-1} . The NH out of plane wagging appeared at 815 cm^{-1} . The IR spectra of acrylamide grafted *Mimosa* mucilage showed a broad absorption band at 3362 cm^{-1} due to overlap of OH stretching band of *Mimosa* mucilage and NH stretching band of acrylamide. The amide-I (CO stretching) occurred at 1671 cm^{-1} and amide-II (NH bending) occurred at 1611 cm^{-1} . The CN stretching of acrylamide appeared at 1433 cm^{-1} .

Table 4 The experimental and predicted values for response Y along with percentage prediction error observed for the optimum test conditions and random checkpoints

Checkpoint conditions X_3/X_4	Y			
	Obser	Pred	Error (%)	
1.92/4.91 ^a	40.36	41.59	2.95	
1/2	1.26	1.28	1.56	
1.5/2	10.45	11.02	5.17	
1.5/5	31.45	29.96	-4.97	
^a Optimum combination of mucilage/acrylamide	2/8	33.05	31.50	-4.92

Fig. 2 DSC thermograms of *Mimosa* mucilage, acrylamide, and acrylamide grafted *Mimosa* mucilage

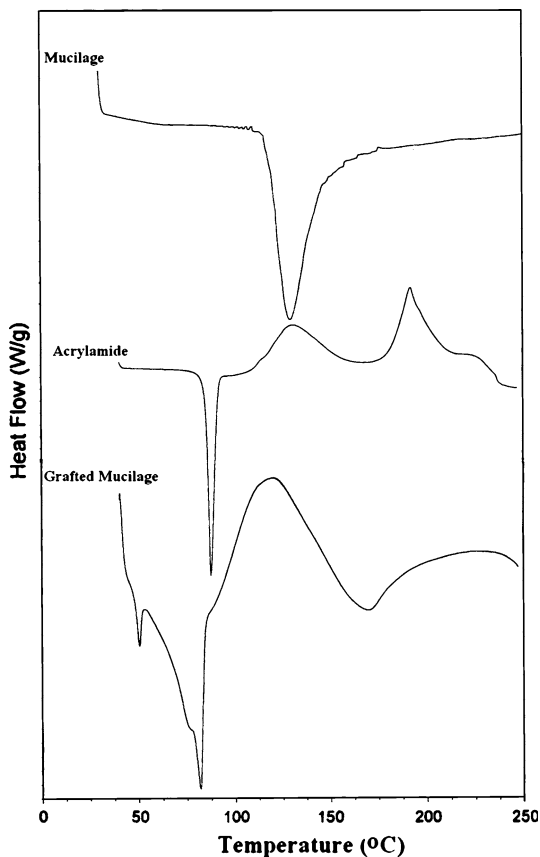


Figure 2 shows the DSC curve of *Mimosa* mucilage, acrylamide, and acrylamide grafted *Mimosa* mucilage. The thermogram of *Mimosa* mucilage showed a broad endotherm at 129 °C with the heat of fusion of 116.1 J/g, indicating the amorphous nature of mucilage. The thermogram of acrylamide showed the sharp endothermic peak at 87.5 °C with heat of fusion of 199.7 J/g. DSC curve of acrylamide also showed two exotherms at 131.4 °C with heat flow of 291.8 J/g and at 204.1 °C with heat flow of 50.28 J/g. The DSC curve of acrylamide grafted *Mimosa* mucilage showed the endothermic peaks at 50, 81.7, and 167.8 °C with heat of fusion of 2.44, 45.19, and 101.8 J/g, respectively.

Figure 3 displays the X-ray diffractogram of *Mimosa* mucilage, acrylamide, and acrylamide grafted *Mimosa* mucilage. The XRD spectra showed the amorphous nature of *Mimosa* mucilage, while the diffractogram of acrylamide showed the crystalline nature of acrylamide with the prominent characteristic peaks of acrylamide appearing at 12.2°, 19.6°, 24.2°, and 28.8° (2θ). The diffraction spectra of acrylamide grafted mucilage showed the characteristic peaks of acrylamide but with the decreased intensity, which confirms the formation of graft co-polymer.

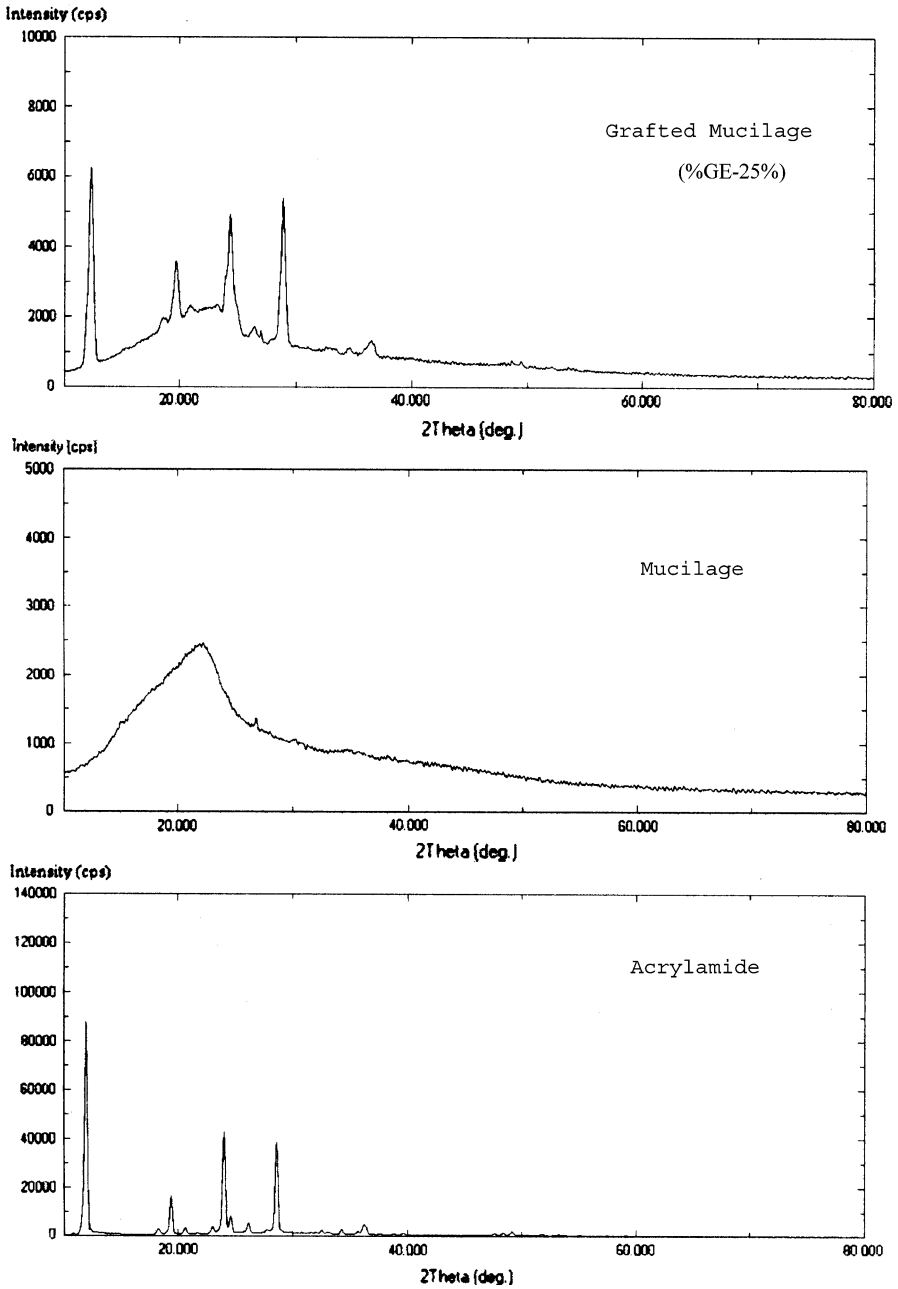


Fig. 3 X-ray diffractograms of acrylamide, *Mimosa* mucilage and acrylamide grafted mucilage

Figure 4 shows the scanning electron micrographs of *Mimosa* mucilage and acrylamide grafted *Mimosa* mucilage. The surface of mucilage shows the presence of honeycomb like structures. The SEM images of grafted copolymer show that the

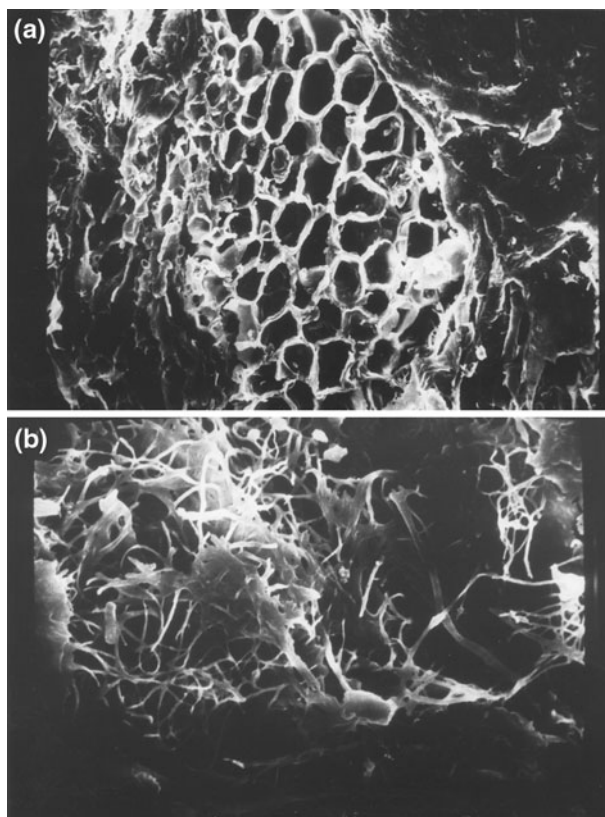


Fig. 4 Scanning electron micrographs of **a** *Mimosa* mucilage, and **b** acrylamide grafted *Mimosa* mucilage

grafting of acrylamide onto *Mimosa* mucilage which can be observed as threads of polyacrylamide entangling the mucilage particles.

Conclusion

Graft co-polymer of *Mimosa* mucilage and acrylamide was synthesized using microwave-assisted grafting. Screening of various factors for their influence on graft polymerization was carried out using Plackett–Burman screening design. The results of screening study revealed that the concentration of mucilage and acrylamide were the significant factors affecting grafting efficiency. Further, optimization of graft co-polymerization was done using CCD. The optimal conditions for synthesis of graft co-polymer were found to be, mucilage concentration between 1.5 and 2.0% and acrylamide concentration of 4.75 and 8.0 mmol. Further, the graft co-polymer was characterized by FT-IR, DSC, XRD, and SEM analysis which confirmed the formation of graft co-polymer of acrylamide and *Mimosa* mucilage. Thus, the

proposed mathematical model can be employed for synthesis of graft co-polymer of acrylamide and *Mimosa* mucilage of desired degree of grafting.

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