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Modification of Okra mucilage with acrylamide: Synthesis, characterization and swelling behavior

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Abstract

In the present communication, the synthesis and characterization of Okra mucilage, a food grade and water-soluble polysaccharide, based-materials are described. Okra mucilage has been modified by grafting acrylamide (AAm) for developing the new green polymeric materials of specialty applications. Grafting has been done under N_2 atmosphere using redox initiator and hydrogels were prepared by using *N*,*N*-methylenebisacrylamide (NN-MBAAm) as crosslinker. The effect of monomer concentration, initiator concentration, reaction time and temperature in terms of grafting efficiency (%GE), percent grafting (PG) and percent gel (%G) has been investigated. The grafted polymers and hydrogels were characterized by SEM, XRD and FTIR techniques to study various structural aspects. The swelling behavior of the crosslinked polymeric material has also been studied as a function of time, temperature and pH. The application area of these polymers is varied from biomaterials to the wastewater treatment.

Keywords: Polysaccharide; Crosslinked polymers; FTIR; SEM; Swelling behavior

1. Introduction

In recent years, chemical modification of natural macromolecules, especially polysaccharides, has received considerable interest. Polysaccharides are unique raw materials as they are from renewable resources, widely available in many countries, inexpensive, stable, hydrophilic and modifiable biopolymers. They offer tremendous potential for development of alternate materials (Chauhan, Dhiman, Guleria, & Kaur, 2002; Okieimen, 2003; Singh, Tiwari, Tripathi, & Sanghi, 2004; Soppimath, Kulkarni, & Aminabhavi, 2001). The biodegradability of natural polysaccharides reduces their shelf life and needs to be suitably controlled. The improvement in the properties of natural and synthetic polymers (Adhikary, Tiwari, & Singh, 2007) can be performed in different route; one of these routes is graft copolymerization. In graft copolymerization, natural polysaccharides are chemically modified by inclusion of synthetic monomers onto their backbone; the host polymer gains some of the desired properties of the guest monomers used for grafting ending up with novel polymer and properties. Various methods for graft copolymerization such as radiation induced, chemical initiation etc., are reported in the literature (Bhattacharya & Mishra, 2004).

The Ce(IV) induced graft copolymerization of vinyl monomers onto polysaccharide substrates has been extensively used for their property modification (Adhikary et al., 2007; Mishra & Bajpai, 2006). We have also reported the synthesis of polyacrylamide and polyacrylonitrile-grafted copolymers based on mucilage obtained from *Plantago psyllium* husk, *Coccinia indica* fruits, *Tamarindus indica* and Fenugreek seeds by this method (Mishra & Bajpai, 2005, 2006; Mishra, Rajani, Agarwal, & Dubey, 2002; Mishra, Rajani, & Gupta, 2003; Mishra, Yadav, Pal, & Singh, 2006). Hydrogels based on copolymers of vinyl monomers and biopolymer such as pectin, starch, gelatin and hydroxy-

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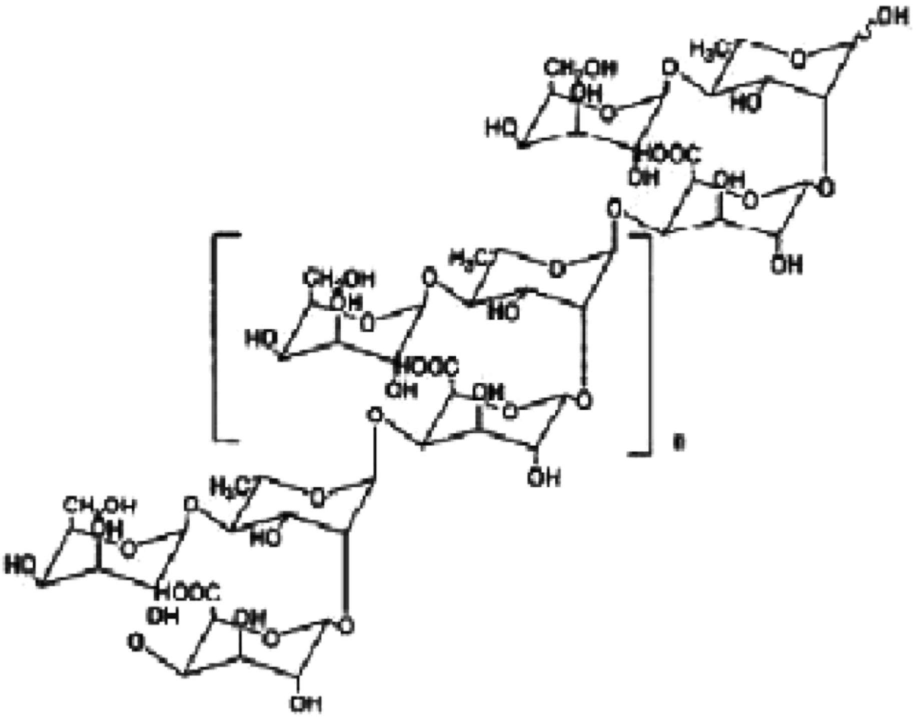

Chemical structure of polysaccharide of Okra mucilage

propyl cellulose have been developed and utilized in various applications (Chauhan, Kumar, Kumari, & Sharma, 2003; Chauhan, Sharma, & Lal, 2004; Liu, Cooke, Coffin, Fishman, & Hicks, 2004; Wu, Wei, Lin, & Lin, 2003).

Okra (*Hibiscus esculentus*) is a bulky annual plant cultivated throughout the tropical and subtropical areas of the world. Traditionally, Okra is either used fresh or is dried and ground to thicken and flavour soups. The immature fruit is also used in folk medicine as a diuretic and for the treatment of dental disease. It has been evaluated for a number of food and non-food applications. Chemical analyses of Okra mucilage have yielded widely differing results (Ndjouenkeu, Goycoolea, Morris, & Akingbala, 1996; Tanaka et al., 2002; Woolfe, Chaplin, & Otchere, 1997). The polysaccharide obtained from it is composed of galactose, rhamnose and galacturonic acid (Hirose, Endo, & Hasegawa, 2004).

In the present communication, the synthesis of graft copolymers of acrylamide with Okra mucilage, using ceric ion initiated solution polymerization technique, has been reported. The influence of reaction time, temperature and concentrations of AAm, NN-MBAAm and ceric ammonium nitrate (CAN) in the reaction mixture on %grafting and %gel is studied. The prepared samples were characterized by FTIR, XRD, SEM and biodegradation studies.

2. Experimental

Okra fruits were purchased locally. They were thoroughly washed with water, cut into pieces, deseeded and soaked in distilled water overnight. The mucilage was extracted by filtering through muslin cloth. For deproteinization, it was treated with 0.3 N $Ba(OH)_2$ – 5% aqueous $ZnSO_4 \cdot 7H_2O$ (Singh et al., 2004). Acrylamide (AAm) (Merck-Schuchardit, Germany), ceric ammonium nitrate (CAN), hydroquinone and *N*,*N*,-methylene bis acrylamide (NN-MBAAm) from S. D. Fine Chemicals, and iso-propanol (Ranbaxy) were used as received.

2.1. Synthesis of O-g-poly(AAm) copolymers

Okra-g-poly(AAm) was synthesized by grafting acrylamide (AAm) onto Okra mucilage by radical polymerization method in aqueous system using ceric ion/nitric acid redox initiator (Mishra & Bajpai, 2005). The following procedure has been adopted in carrying out the reactions to prepare copolymers. One gram of Okra mucilage was dissolved in distilled water (200 mL) in an Erlenmeyer flask. The flask was then sealed with septum stopper and flushed with nitrogen for 20 min. Then the required amount of AAm solution prepared in 100 mL distilled water was added into the solution through the stopper by hypodermic syringe with constant stirring. The solution was stirred for 30 min while being bubbled with nitrogen. The required amount (Table 1) of ceric ion solution (in 1 N HNO_3) was injected through the stopper by hypodermic syringe. The nitrogen flushing was continued for another 20 min; then the needles were taken out, and the flask was further sealed with Teflon tape. The reaction temperature was maintained by immersing the flask in constant temperature bath. The reaction was continued for the time required with occasional stirring unless stated otherwise, and then terminated by injecting 0.5 mL of saturated aqueous

Table 1
Effect of concentrations of crosslinker, monomer and initiator on PG, %GE and %gel

Sample No.	$[\text{AAm}]\times 10^2$	$[\text{NN-M BAAm}]\times 10^3$	$[\text{Ce(IV)}]\times 10^3$	Grafting/gel (%)	Grafting efficiency (%)	Intrinsic viscosity (η_{int})
1	1.40	–	0.164	27.90	84.43	5.87
2	1.40	–	0.255	42.90	89.59	6.48
3	1.40	–	0.346	34.60	91.57	5.93
4	2.11	–	0.164	34.00	89.51	6.22
5	2.11	–	0.255	56.12	91.59	7.92
6	2.11	–	0.346	49.28	92.83	6.95
7	2.81	–	0.164	42.45	91.25	6.84
8	2.81	–	0.255	61.54	94.02	8.38
9	2.81	–	0.346	51.66	95.29	7.22
10	4.22	–	0.164	30.60	76.45	6.14
11	4.22	–	0.255	39.98	81.51	6.68
12	4.22	–	0.346	38.42	82.98	6.47
13	2.81	0.129	0.255	33.62	82.97	–
14	2.81	0.259	0.255	41.00	86.01	–
15	2.81	0.389	0.255	69.11	97.18	–

Temperature = 30 °C; mucilage = 1.0 g; time = 2 h.

hydroquinone solution. The reaction product was precipitated in excess of isopropanol and filtered through sintered glass filter. A mixture of *N*,*N*-dimethyl formamide and acetic acid (v/v, 1:1) was used to separate homopolymer. After separation of homopolymer, the graft copolymer precipitate was again slurried in acetone followed by filtration and finally the precipitate was dried in vacuum oven at 40 °C.

The %grafting was calculated by the equation

$$\%\text{Grafting} = \frac{\text{Weight of polymer grafted}}{\text{Weight of pure mucilage}} \times 100$$

The %efficiency was calculated by the equation

$$\%\text{Efficiency} = \frac{\text{Weight of polymer grafted}}{\text{Weight of polymer grafted} + \text{weight of homopolymer formed}} \times 100$$

2.2. Synthesis of Okra-cl-poly (AAm)

Same procedure as described for the synthesis of Okra-g-poly(AAm) was followed for the synthesis of Okra-cl-poly(AAm). The only variation in the reaction contents was the addition of NN-MBAAm. Three crosslinked samples were prepared with varying concentrations of NN-MBAAm and rest of the reaction parameters were the same as taken in the synthesis of the Okra-g-poly(AAm). Okra-*cl*-poly(AAm), in each case, copolymer was separated from poly(AAm) by stirring the reaction mixtures in hot distilled water and finally dried in air oven at 40 °C. The %gel was calculated by equation

$$\%\text{Gel} = \frac{\text{Weight of dry hydrogel}}{\text{Weight of gelled polymer}} \times 100$$

2.3. Characterization and analysis

The structure of Okra mucilage and Okra-*cl*-poly(AAm) was determined by Fourier Transform (FT) IR spectrum (Brucker Vector 22 spectrophotometer) using KBr pellets. Scanning electron micrographs (SEM) of pure and the crosslinked polymer were obtained on JEOL, JSM-840 SEM to investigate and compare surface morphology. The sample in the form of films were mounted on the specimen stabs and coated with gold ion by sputtering method. The micrographs were taken at a magnification of 2000 (Okra and Okra-g-poly(AAm) and 10,000 (Okra-*cl*-poly (AAm)). X-ray diffraction patterns of powder samples of pure mucilage and grafted copolymer were obtained at ambient conditions on an X-ray diffractometer, model Iso-Debyflux-2002 (Rich and Scifert), using Cu Kα radiation. The viscosity of the mucilage and Okra-g-AAm samples in distilled water was measured by Ostwald's viscometer.

2.4. Swelling behavior

An accurately weighed amount of the dried crosslinked polymer sample (%gel = 69.11) was placed in a previously dried and weighed glass crucible, which was then filled with 50 mL of solution of different pH, and after different time intervals, suction from a vacuum pump was applied. The glass crucible was then weighed to determine the amount of water absorbed by per gram of the dried material and taken as water absorption capacity. Water absorption was measured by the following equation:

$$C_{\text{WA}}\ (\text{g/g}) = \frac{W_1 + W_2}{W_1}$$

where C_{WA} is water adsorption capacity, W_1 and W_2 are masses of dried powdered sample and amount of water absorbed, respectively.

3. Results and discussion

Ceric ion initiated polymerization yields substantially pure graft copolymers since the free radicals are produced exclusively on the backbone, which in the presence of acrylamide, initiates polymerization to produce a graft copolymer. To obtain a better reproducibility in the results, the reaction was carried out under a constant light source because the oxidative capability of the Ce(IV) ions increases considerably under light (Vazquez, Goni, Guruchanga, Valero, & Guzman, 1992). The length of the grafted chains at a fixed monomer concentration is proportional to the concentration of ceric ions. With higher concentration of ceric ions, the number of free radical sites will be more and consequently the length of grafted chains will be larger. With lower ceric ion concentration, the grafted chain length should be shorter (Adhikary et al., 2007). The gel content of the crosslinked copolymer is dependent on the concentration of NN-MBAAm.

3.4. *Influence of reaction parameters on grafting*

3.4.1. *Effect of concentrations of AAm and NN-MBAAm*

The effect of monomer and crosslinker concentration on percent grafting/gel and grafting efficiency is shown in Table 1. As the AAm concentration increased from 0.0141 to 0.0281 moles, the PG and %GE increased but with further increase up to 0.0422 moles, the PG and %GE decreased. The increase of %GE and PG was expected with increase in AAm concentration due to the availability of AAm monomer with respect to polysaccharide macroradicals, leading to larger possibility of grafting, but the decrease in GE and PG might be due to the formation of more homopolymer. These homopolymers successfully hinder the rate of penetration of monomer molecules to the polysaccharide free radicals, resulting in decrease in GE (Fares, 2003). The gel content in Okra-cl-poly(AAm) samples increased from 33.62% to 69.11% with increase in concentration of NN-MBAAm from 0.129×10^{-3} to 0.389×10^{-3} moles.

3.4.2. *Effect of Ce(IV) concentration*

On increasing the concentration of initiator from 0.164×10^{-3} to 0.255×10^{-3} mol, both %GE and PG increased due to increase (Table 1) in the number of free radicals sites on polysaccharide chains. The falling off of %GE and PG at higher concentration of initiator (0.346×10^{-3}) is a well-known phenomenon and ascribed to the increasing participation of the ceric ion in the termination of the growing grafted chains (Mishra & Bajpai, 2005; Yao & Tang, 1992).

3.4.3. *Effect of reaction time*

The effect of time on %GE, PG and %gel is shown in Table 2. The PG and %gel increased with increasing the reaction time up to only 2 h and then a slight decline or constancy in both PG and %gel content was observed. The rapid increase of grafting between 1 and 2 h is due to an increase in rate of initiation and propagation and the decline or constancy of grafting after 2 h is a clear remark on the depletion of monomer from the solution This agrees with the earlier observation with free radical initiated polymerization (Fares, 2003; Sudhakar, Srinivasan, Joseph, & Santappa, 1981).

Table 2
Effect of reaction time and temperature on PG, %GE and %gel

Sample No.	PG	%GE	%Gel	Time (h)	Temperature (°C)	η_{int}
1	36.00	77.05	–	1	30	6.39
2	61.54	94.02	–	2	30	8.38
3	59.84	92.69	–	4	30	7.20
4	59.07	92.59	–	24	30	6.82
5	–	93.32	59.6	1	30	–
6	–	97.18	69.11	2	30	–
7	–	91.52	51.4	3	30	–
8	–	88.73	46.8	4	30	–
9	–	87.03	42.3	24	30	–
10	56.34	93.39	–	2	20	7.98
11	61.54	95.29	–	2	30	8.38
12	62.00	96.09	–	2	40	8.41
13	66.50	96.66	–	2	50	8.94
14	–	85.97	32.86	2	20	–
15	–	97.18	69.11	2	30	–
16	–	97.23	70.40	2	40	–
17	–	97.79	74.60	2	50	–

Mucilage = 1.0 g; $[\text{AAm}] \times 10^2$ mol = 2.81; $[\text{Ce(IV)}] \times 10^3$ mol = 0.255; $[\text{NN-MBAAm}] \times 10^3$ mol = 0.389.

3.4.4. *Effect of reaction temperature*

Reaction temperature is an important reaction condition in the graft copolymerization. The effect of temperature on percent grafting and grafting efficiency is shown in Table 2. PG/%gel and GE both increased on varying the reaction temperature from 20 to 50 °C. The increase in %GE and PG/%gel with increasing temperature may be due to the increased diffusion rate of monomer and initiator and raised rate of grafting (Shukla & Srivastava, 2003).

3.4.5. *Swelling behavior*

Swelling behavior of Okra-*cl*-poly(AAm) (%gel content = 69.11) in water was studied as the function of time, temperature and pH.

3.4.6. *Effect of time and temperature on swelling*

The swelling behavior of polymers was studied at different time intervals of 10 and 15 min and at 20, 30, 37 and 50 °C. It was seen that (Table 3) with increase in time and temperature, the extent of swelling of the polymer also increased (Ranby & Gadda, 1982).

3.4.7. *Effect of pH on swelling of hydrogel*

Table 3 summarizes the results obtained for swelling behavior of hydrogel at different pH. The water absorption capacity of hydrogel increased as the pH changed from acidic to alkaline. At neutral pH, the water absorption

Table 3
Effect of Time, pH and temperature on swelling of hydrogels

pH (temperature 30 °C)			Time (min)	Temperature (°C) pH 7		
4	7	9		20	37 ± 2	50
Water absorption capacity (g/g)						
9.4	12.1	15.7	10	8.2	13.1	16.1
13.8	21.2	24.2	20	14.4	22.4	24.1
21.1	29.2	32.8	30	23.3	30.1	31.0
29.1	34.8	37.1	40	29.8	35.1	38.7
30.5	39.9	42.2	50	32.3	40.3	40.9
37.4	42.6	46.9	60	41.8	42.7	49.7
–	48.3	–	70	44.2	49.3	51.8
44.0	51.2	54.4	75	–	–	–
–	52.3	–	80	52.1	53.4	56.9
52.1	59.2	63.3	90	58.0	59.4	60.4
–	61.3	–	100	60.3	63.6	68.1
58.4	62.3	67.6	105	–	–	–
–	63.7	–	110	62. 9	65.1	70.1
61.8	69.8	74.8	120	66.6	71.2	74.2

Okra-*cl*-poly(AAm) = 1 g.

capacity of hydrogel is lower than that at alkaline pH while it is higher than that at acidic pH. At alkaline pH, the –COOH groups of mucilage are ionized and —CONH— and —$CONH_2$ groups are deprotonated. These charged carboxyl and deprotonated amide groups cause mutual repulsion. Both the repulsion of the negatively charged polymer chains and the presence of free counter-ions in the gel, which cause a high osmotic swelling pressure, could contribute to increased swelling. At lower pH values, the functional groups are protonated and keep the network at its collapsed state which causes a decrease in water absorption values.

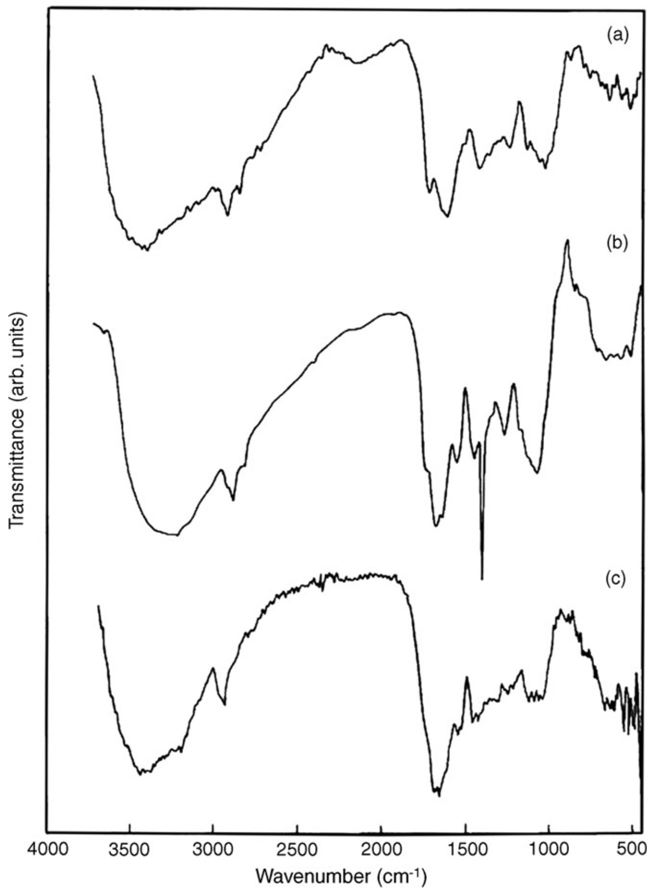

Fig. 1. IR spectra of: (a) Okra mucilage; (b) O-g-poly(AAm) and (c) O-cl-poly(AAm).

3.5. Characterization of graft copolymer

3.5.1. Infra red (IR) spectrum

The FTIR spectra of Okra mucilage, O-g-poly(AAm) (PG = 61.54) and Okra-*cl*-poly(AAm) (%gel = 69.11) are shown in Fig. 1(a, b and c), respectively. The FTIR spectrum of Okra-g-poly(AAm) is different from that of Okra mucilage by showing characteristic peak of —C=O of amide at

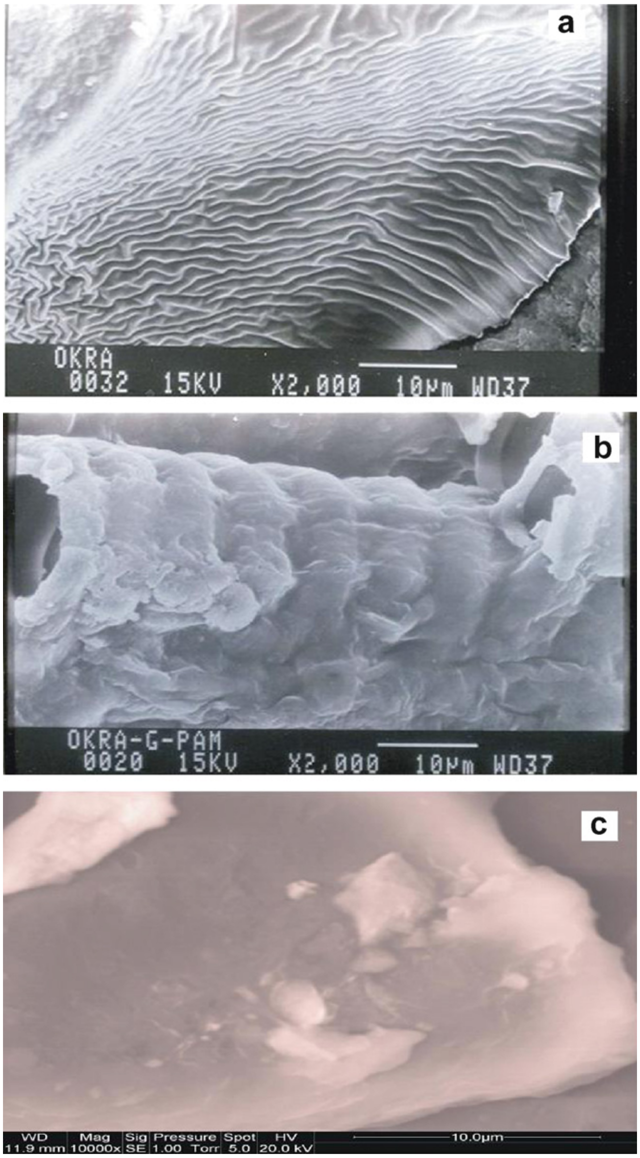

Fig. 2. Scanning electron micrographs of: (a) Okra mucilage; (b) O-g-poly(AAm) and (c) O-cl-poly(AAm).

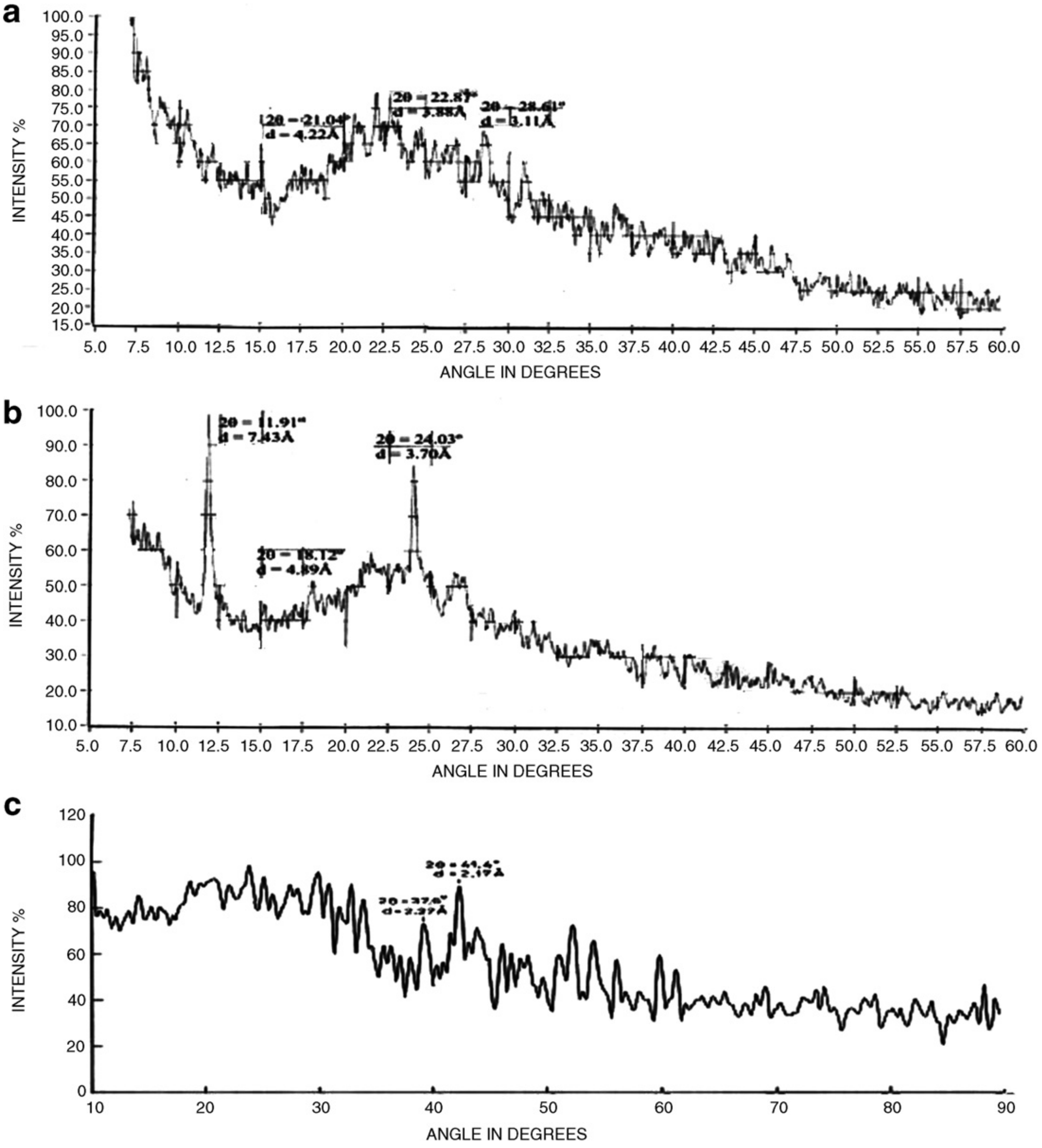

Fig. 3. XRD patterns of: (a) Okra mucilage; (b) O-g-poly(AAm) and (c) O-cl-poly(AAm).

1671.12 cm^{-1}; of —NH bending at 1542.17 cm^{-1}; of —CN stretching at 1384.53 cm^{-1}; of —C—C—N asymmetric at 1245.26 cm^{-1} and Moreover, the broadening and shifting of band coming after 3000 cm^{-1} towards slightly lower wave number in Okra-g-poly(AAm) as compared to that of pure mucilage is because of the overlapping of —NH of amide and —OH of mucilage and increased hydrogen bonding due to —CONH$_2$ groups respectively. The FTIR spectrum of Okra-*cl*-poly (AAm) shows characteristic peak of —C═O of amide at 1651.14 cm^{-1} and shifting of peak coming after 3000 cm^{-1} towards slightly higher wave number as compared to that in Okra-g-poly(AAm) is probably due to loss of crystallinity by crosslinking.

3.5.2. Scanning electron microscopy (SEM)

The SEM technique is considered to be one of the best techniques to study the surface morphology of different kinds of polymers. A comparative study of the scanning electron micrographs (Fig. 2a–c) of Okra mucilage, Okra-g-poly(AAm) and Okra-*cl*-poly(AAm) are used as a supportive evidence for grafting. The morphology of the surface of pure Okra mucilage is different than that of its grafted and crosslinked copolymers. A considerable amount of grafted polymer is deposited, which appears to have a different structure from the pure mucilage (Gao, Tian, Yu, & Duan, 1994). The lamellar structure is evident for Okra mucilage whereas the surface of Okra-g-poly(AAm) show well-defined layers of grafted polyacrylamide chains and Okra-*cl*-poly(AAm) show heterogeneity.

3.5.3. X-ray diffraction (XRD)

Okra mucilage is an amorphous polysaccharide. A comparative study of the XRD patterns (Fig. 3a and c) of Okra mucilage, and Okra-*cl*-poly(AAm) showed amorphous character of the polymers while XRD pattern (Fig. 3b) of

Okra-g-poly(AAm) showed crystalline peaks. These patterns are in accordance with the results obtained by FTIR spectra and SEM pictures.

3.5.4. Biodegradation

The viscosity of solution as a function of time is taken as the criterion for study of biodegradation of Okra mucilage and Okra-g-poly(AAm). The decrease in intrinsic viscosity value with time was taken as measure of the biodegradability (Owen and Shen, 1977). In each experiment, 0.1 g of polymer was dissolved in 100 mL distilled water and viscosity measurements were performed using Ostwald's viscometer. All measurements were carried out at room temperature. The values of the intrinsic viscosity of aqueous solutions of Okra mucilage and O-g-poly(AAm) were 5.31 dL/g and 8.38 dL/g, respectively on first day. A decrease in the viscosity values was observed over a week's time. In case of Okra mucilage, degradation started from the day one while O-g-poly(AAm) started degrading after 3 days. The intrinsic viscosity value of Okra mucilage and Okra-g-poly(AAm) was less than 1.0 dL/g after 5 and 8 days, respectively. No change in the weight of dried Ku-g-PAM was observed even after three months. These observations confirmed that the grafting of acrylamide chains increases the shelf life of the polysaccharide without destroying its biodegradable nature.

The biodegradability of Okra-*cl*-poly(AAm) could not be tested by measuring intrinsic viscosity as it was insoluble in many polar, non polar and mixed solvents due to its crosslinked structure. Therefore, the biodegradability of Okra-*cl*-poly(AAm) copolymer samples was tested by soil burial test. One of the few systematic studies of soil biodegradability was performed in the early 1990 (Barak, Coquet, Halbach, & Molina, 1991). As polymers show different rates of degradation according to the location of the experiment and the specific characteristics of each polymer, therefore the soil condition was maintained throughout the soil burial experiment. The pH of the soil was in the range of 7.4–7.9. The sample was kept buried under the soil for three months. After three months, it was taken out, washed with water, dried in hot air oven and weighed. A negligible reduction in polymer weight was observed.

4. Conclusions

Grafting of polyacrylamide with Okra mucilage, a polysaccharide of vegetable origin, offer new polymeric materials with properties that can be exploited industrially. A redox initiator system of CAN/HNO_3 was efficiently used to graft and crosslink mucilage using AAm and NN-MBAAm, respectively. The remarkable variation in extent of grafting and crosslinking was seen with the variation in the concentrations of monomer and initiator and reaction time and temperature. Various analytical techniques such as FTIR, SEM, and XRD patterns confirmed grafting and crosslinking of acrylamide onto mucilage chains. Water absorption capacity of crosslinked hydrogels was dependent on pH variation. As the crosslinked polyacrylamides are non biodegradable, the inclusion of polysaccharide in polyacrylamides increases the possibility of formation of biodegradable polymers. Graft copolymers of the Okra mucilage and polyacrylamide seem to be more advantageous as compared to pure polyacrylamides. Grafting only improves the properties of mucilage by introducing more reactive sites and without making any change in the molecular mobility of chelating groups of polysaccharide.

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