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Synthesis and Characterization of Chemically Cross-Linked CMC/Acrylamide Hydrogels

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Carboxymethyl cellulose/poly(acrylamide) (CMC/Amm) hydrogels were synthesized by the chemical cross-linking method. Ammonium persulfate used as an initiator, while aluminium sulfate used as a cross-linking agent. The structure and morphology of the hydrogels were characterized by FTIR and scanning electron microscopy (SEM) analysis. The swelling behaviour of the hydrogels can be studied by using acids (CH_3COOH, HCl and $HClO_4$) and also in the pH of the buffer solutions at different temperature (room temperature, 30 and 37 ºC) was studied. Swelling of hydrogels increased with an increase in the concentration of aluminum sulfate up to 20 %, above 20 % it has found to be decreased. The effect of four series of cationic different concentrated salt solutions on the swelling had found to be the following order $K^+ > Na^+ > Ca^{2+} > Mg^{2+}$.

Keywords: Acrylamide, Carboxy methyl cellulose, Cross-linking agent, Hydrogels, Initiator.

INTRODUCTION

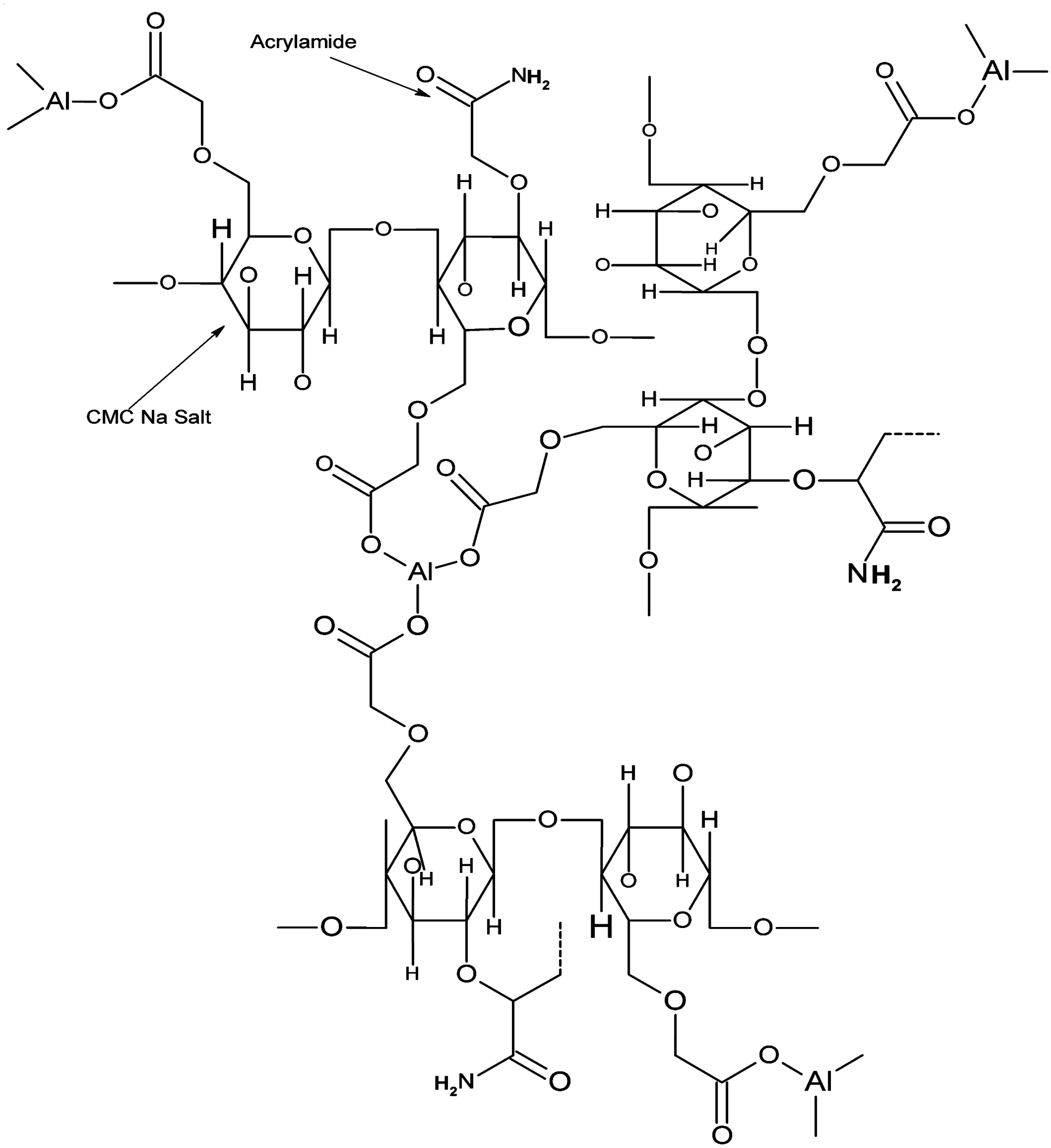
Hydrogels are characterized as a three-dimensional network system of hydrophilic polymers, which assimilate and hold a significant amount of water. Carbohydrate polymers, *e.g.*, cellulose, starch, alginate and chitosan are good skeleton candidates for hydrogels due to their excellent biocompatibility, bio-degradability and bio-activity [1,2]. They have been widely used in the synthesis of hydrogels for drug delivery, pharmaceutical, medical, biomedical and tissue engineering applications [3,4]. The anionic polyelectrolyte carboxymethylcellulose (CMC) is an important derivative of cellulose with good water-solubility resulting from the $–CH_2COOH$ groups in its skeleton [5]. It has attracted considerable attention because of its excellent properties, like transparency, hydrophilicity, high viscosity, biocompatibility, biodegradability, good film-formability and non-toxicity [6,7]. CMC is a natural derivative of cellulose polymer of an anionic, highly water-soluble, widely used in potential applications in the field of oil exploration, food, paper, textile and detergent industries due to its unique high viscous properties [8]. Due to its excellent and exclusive characteristics and behaviour properties, these materials are widely used in various fields like agricultural, horticultural, pharmaceuticals, food packaging applications, disposable nappies, sanitary pads, feminine napkins, pampers, medical and paramedical and biomedical applications [2,3]. Hydrogels prepared through the cross-linking of CMC are highly absorbable with excellent dynamic, physical, chemical and visco-elastic properties [9]. Most of the CMC hydrogels are prepared through chemical cross-linking reactions. The cross-linking network prepared by a chemical reaction is usually robust, exhibit good mechanical properties and excellent water absorption properties. The monomer, acrylamide is used for the synthesis of poly(acrylamide) of desired molecular weights of different physical and chemical properties up to a wide variety of applications. PAAm hydrogel is an inert, neutral and more hydrophilic material. These hydrogels have enough hydrophilicity but they are poor mechanical properties and hydrolytic stability. PAAm hydrogels are reversible and stimuli responsible for temperature, pH and other external agencies hence they are used for application as smart materials [10].

Aluminum sulfate with its cationic charge forms of Al^{3+}, $Al(OH)^{2+}$ and oligomeric species in aqueous solution when pH < 5, these multivalent positive charge leads to cross-linking with enormous sites of opposite charge anionic groups (COO-). $Al_2(SO_4)_3$ has been widely used as a cross-linking agent in the preparation of bio-materials and exhibits excellent performance and little toxicity. It is excellent candidate for the electrostatic complexion with the polymer containing carboxyl -COOH- groups to form hydrogel structure [11]. In the present investigation, a novel electrostatic complexion method is planned for the preparation of CMC/AAm hydrogels with Al^{3+} ions as a cross-linking agent [12]. The structure of the prepared hydrogels was characterized and the effects of preparation condition of its properties were investigated. Because of its exclusive characteristics and behaviours, CMC/AAm-based hydrogel is a versatile and potential candidate for wound dressing/healing.

EXPERIMENTAL

Sodium salt of carboxymethylcellulose of high viscosity (15 Mpas) was purchased by the Titan Biotech. Ltd. Bhiwadi, India. Acrylamide and potassium chloride (Sisco Research Lab. Pvt. Ltd. Mumbai, India), aluminum sulfate (S.D. Fine-Chem Ltd. India), ammonium persulfate (Thomas-Baker (Chemicals), India. Sodium chloride was purchased from E. Merck (India). All the chemicals and reagents used without further purification.

Preparation of carboxymethylcellulose sodium salt/acrylamide hydrogels: Carboxymethylcellulose sodium salt (1 %, 25 mL), 6 g of acrylamide, 1 g of ammonium persulfate and 3 g (12 %) of aluminum sulfate were taken in 100 mL and dissolved completely in water by constant stirring. After the complete dissolution, a solution was kept on the hot plate at 40-45 °C for 20 min. After 20 min, the solution became a gel and then washed with ethanol to eliminate the unreacted monomer acrylamide and cross-linking agent aluminium sulfate and finally dried in an oven at 40-50 °C for 5-6 h. We have blended distinctive CMC/acrylamide hydrogels by fluctuating the cross-linking agent amount from 3 g (12 %) to 7 g (28 %) aluminum sulfate (**Scheme-I**).



Scheme-I: Proposed mechanism between aluminum sulphate ions and CMC Na salt/acrylamide

FTIR: The FTIR spectra of the CMC sodium salt, CMC Na salt/acrylamide cross-linked hydrogel blend samples have been recorded in the range of 4000 to 500 cm^{-1}.

SEM: The surface morphology of CMC sodium salt and aluminum sulfate cross-linking hydrogels were investigated by using scanning electron microscopy (SEM Zeiss, LS15).

Swelling behaviour of CMC/AAm hydrogels: The swelling ratio of hydrogel sample was measured at different temperatures in different solvents by using the gravimetric method. The previously weighed dry hydrogel samples were soaked in excess of different solutions and kept undisturbed for 8 h at a different temperatures until the constant values were obtained. Degree of swelling rate can be calculated as follows:

$$DS\,(\%) = \frac{W_2 - W_1}{W_1} \times 100 \quad (1)$$

where DS is the degree of swelling expressed in percentage, W_1 and W_2 are the masses of the sample before and after swelling, respectively.

Swelling at various pH: The various solutions having pH 1.2, 2.2, 4.0, 6.8, 7.0 and 9.5 were prepared using phosphate buffer solution at room temperature, 30 and 37 °C. The dried hydrogel samples were used for the swelling measurement according to eqn. 1.

Swelling in salt solutions: The swelling capacity of hydrogels was determined in the different salt solutions (KCl, NaCl, $CaCl_2$ and $MgCl_2$) and also with different concentrations *viz.* 0.4, 0.6, 0.8, 1 and 1.2 M.

RESULTS AND DISCUSSION

FTIR analysis: Fig. 1 shows the FTIR spectrum of pure sodium salt of CMC in which the broad absorption band at 3315.02 cm^{-1}, due to the stretching frequency of -OH group [13]. The band around 1317.14 cm^{-1} is assigned to OH bending vibration. The high intense band appears at 1588.09 and 1416.45 cm^{-1} may attribute the stretching and bending vibration of anion groups (-COO-) in CMC. The band around 1320 cm^{-1} is assigned to OH bending vibration. CMC samples showed a band at 1052.94 and 941.09 cm^{-1} which dominates the spectrum of cellulose linkages and α-glycosidic linkage [14-17]. Pure poly(acrylamide) PAAm and CMC-PAAm hydrogel FTIR spectra are also showed in Fig. 1. A broad peak at around 3289 cm^{-1} is attributed to OH stretching and the symmetric stretching vibration of N-H group. A stretching frequency at 2887.22 cm^{-1} confirmed the presence of CH_2 group in PAAm. An absorption peak at 1667.15, 1590.98 and 1444.42 cm^{-1} were attributed to amide-I (C=O) on PAAm chains, a overlap of (-COO-) on the CMC backbone and the amide-II (N-H) on the PAAm chains

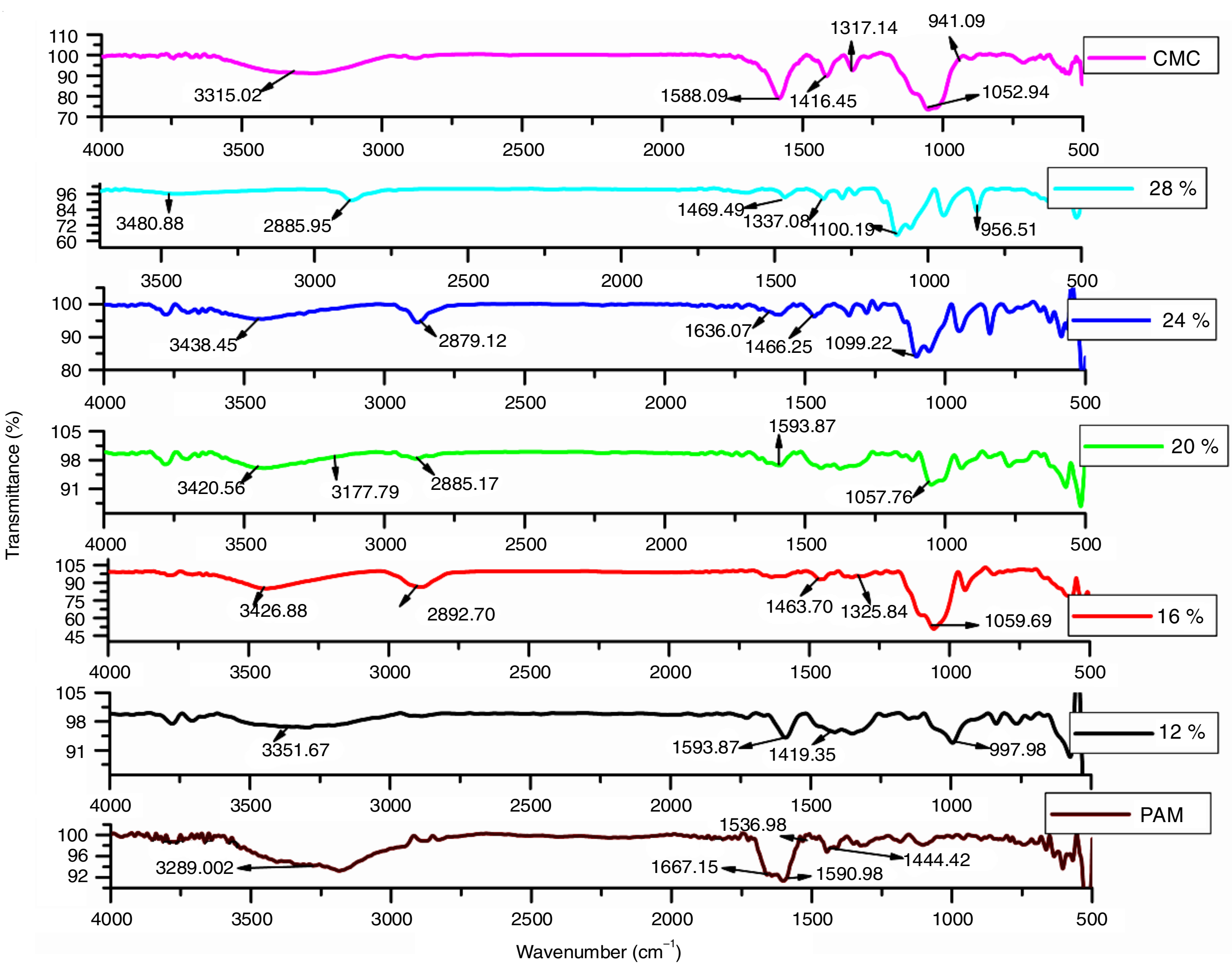


Fig. 1. FTIR spectra of sodium salt of CMC, PAM and CMC Na salt/acrylamide cross linked hydrogels

and the C-N stretching in hydrogel [18], respectively. A band at 1100.19-1059. 69 cm^{-1} represents an important (-CH-O-CH_2) unit of CMC-Am hydrogels.

SEM analysis: Fig. 2 represents the surface morphology of CMC Na salt/acrylamide. Fig. 2a shows that the CMC sodium salt has a smooth, uniform and non-porous surface. The pore size also depends on the concentration of aluminium sulfate. The pore size increases up to the certain extent of concentration of the crosslinking agent. Here, a pore size increases up to 20% of crosslinking agent after that pore size reduces because of the increases in the concentration of cross-linking agent [19,20].

Effect of crosslinking agent on swelling rate: The effect of crosslinking agent [$Al_2(SO_4)_3 \cdot 16H_2O$] on the swelling rate of hydrogel has been studied in the concentration range of 12-28 %. Table-1 clearly indicate that a swelling rate decreases with increasing crosslinking agent. The maximum absorbency (407 g/g, 1 M HCl) was observed at 20 % of crosslinking agent. According to Flory classical theory of elastic gel, crosslinking density increases with increase in the amount of crosslinking agents, this is more favourable for the absorbing and retaining fluid [20]. At low concentration, crosslinking agent leads to a low degree of crosslinking density and difficult for 3D network structure formation, so water absorbency/swelling rate was low. However, when it is higher than the optimum value (20 % $Al_2(SO_4)_3$), more molecules of crosslinking molecules were available and the size of pores becomes smaller in the network, which leads to the macroscopic decreases of the absorbency [21]. It was found that increasing the concentration of acidic crosslinking agent [$Al_2(SO_4)_3 \cdot 16H_2O$] or $Al_2H_{32}O_{28}S_3$ up to a certain limit enhance the swelling behaviour of CMC hydrogel (Table-1). Above 20 % of crosslinking agent, a swelling rate of hydrogel decreases significantly, which may be due to decrease in flexibility of the polysaccharide chains, which resulted in the segmental motion of chain. Diffusing of hydrolyzing agent (water) into the polymer network also decreases, thereby giving rise to a more rigid structure of the polymer network [22].

Effect of pH: The swelling behaviour of CMC/AAm hydrogel has been investigated in the pH range of 1.2-9.5. The results (Table-2) revealed that rate of swelling (%) increased with higher pH. It is due to the fact that under acidic pH condition most of the carboxylate ions were protonated, so the main anion-anions COO-COO^- repulsive forces were eliminated and consequently, swelling rate of hydrogels decreased. However, some type of attraction or interactions like (H-O and H-N hydrogen bonding) lead to decrease the absorbance. Further increasing the pH of the medium, the swelling capacity surprisingly enhanced, which may be due to the fact that COOH groups of hydrogels were converted to carboxylate anions COO^- (deprotonation). This clearly results in an expansion of network chains leading to an increase in the absorbency rate. In other words, at higher pH, a ratio of COO^-/COOH on CMC also enhances because of increasing ionization of carboxylic COO- groups and this results in a greater repulsion among the COO- bearing CMC chains. The results may also be explained by the fact that with increasing pH of the swelling medium the extent of ionization of carboxylate groups of the CMC also increases, which produces the greatest number of carboxylate ions of the CMC molecules. These anionically charged centers repel each other and produce a rapid relaxation in the network. This clearly results in a rise in the swelling (%). Similar swelling pH dependencies have been reported in the case of other hydrogel systems [23-25].

Effect of temperature on swelling of hydrogels in different acids: The swelling rate of hydrogel was increased with the temperature increases up to 37 ºC. An increase in swelling rate values were dependent on the kinetic energy of polysaccharide chain in CMC, which leads to a lower solubility of the hydrogel as well as the diffusion rate of CMC backbone. The

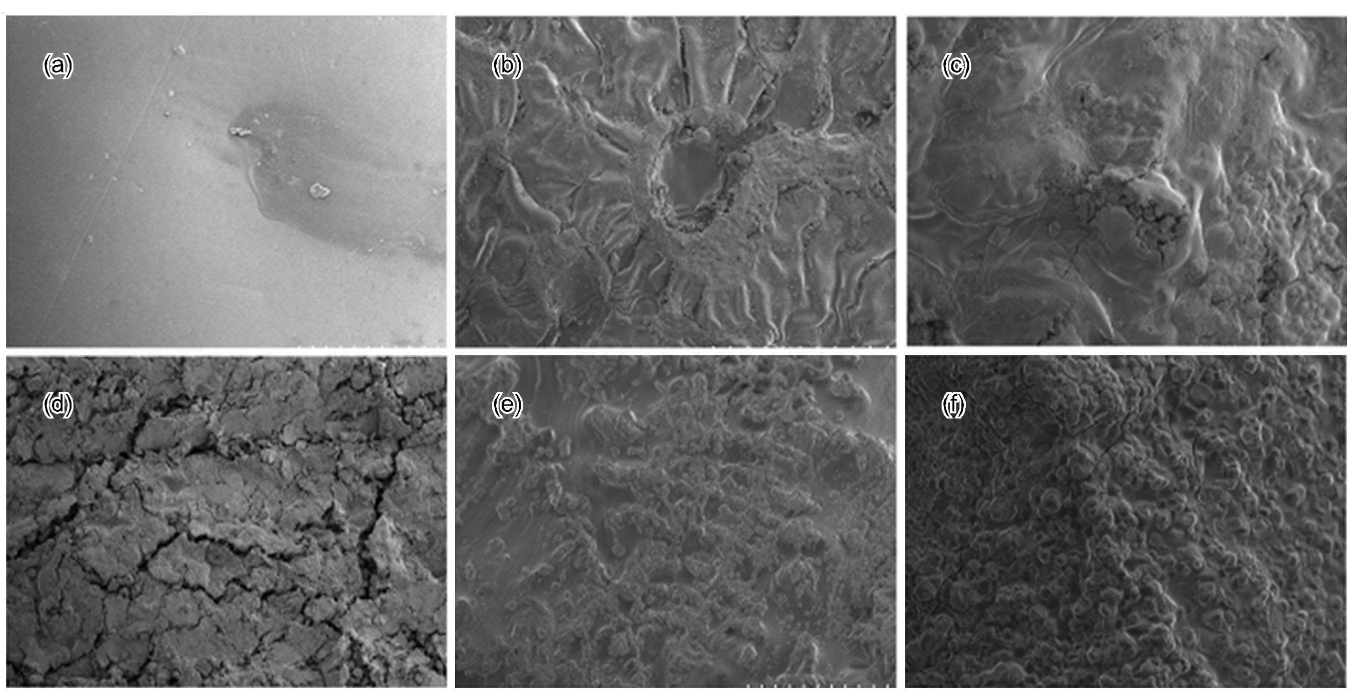


Fig. 2. SEM images of (a) pure CMC Na salt (b) 12 % (c) 16 % (d) 20 % (e) 24 % (f) 28 % aluminum sulphate cross linked CMC Na salt/acrylamide hydrogels

TABLE-1
SWELLING RATE VALUES OF HYDROGELS USING ALUMINUM SULPHATE (AS) AT 37 °C IN DIFFERENT ACIDS

AS	Time (h)	CH_3COOH				HCl				$HClO_4$			
		1.0 N	0.8 N	0.6 N	0.4 N	1.0 N	0.8 N	0.6 N	0.4 N	1.0 N	0.8 N	0.6 N	0.4 N
12 %	1	94.07	90.28	88.31	70.47	110.46	103.53	94.18	102.40	110.78	75.27	64.58	62.37
	2	171.62	160.15	156.28	153.28	134.79	125.66	116.82	114.28	132.49	113.58	100.76	88.26
	3	251.16	246.25	242.56	223.28	152.39	143.76	135.38	124.96	155.86	140.63	111.46	98.23
	4	261.09	234.86	251.74	250.99	176.62	154.48	143.91	141.27	167.27	151.34	138.56	127.76
	5	266.02	256.31	257.18	251.63	210.43	184.23	143.71	139.47	179.55	161.49	151.73	140.54
	6	298.27	286.95	260.61	256.14	241.39	222.76	182.29	158.64	183.67	173.58	161.49	152.65
	7	313.18	288.48	269.27	262.09	270.29	259.19	202.73	169.83	201.34	184.62	173.58	162.37
	8	329.31	298.64	264.78	261.27	298.34	269.18	214.79	192.64	213.82	194.29	184.62	173.28
16 %	1	105.56	103.45	97.56	82.59	127.86	68.20	80.17	93.48	124.79	90.19	79.86	78.34
	2	182.14	174.13	170.12	165.25	156.27	105.47	86.53	132.83	147.69	125.34	114.68	100.86
	3	264.96	258.17	248.15	234.12	175.93	142.54	95.76	143.58	166.38	152.49	123.64	102.58
	4	273.86	273.12	262.85	251.14	184.61	182.52	117.21	163.87	180.29	162.37	153.86	140.60
	5	278.96	267.45	258.45	263.45	183.65	223.58	167.33	193.43	192.37	172.68	163.49	152.77
	6	310.56	298.46	272.45	268.56	201.58	258.27	189.35	196.46	195.64	184.33	174.96	165.37
	7	325.76	302.36	280.12	275.15	221.37	292.38	195.47	228.59	209.44	199.06	187.72	180.54
	8	341.12	310.89	278.15	276.89	243.86	299.29	214.83	247.67	220.86	211.49	201.12	192.66
20 %	1	154.79	142.56	129.58	122.59	146.64	76.27	69.37	114.82	136.83	98.61	93.28	88.55
	2	190.45	175.86	152.88	133.35	187.46	126.63	71.52	115.68	157.29	112.06	103.16	101.71
	3	224.15	195.45	195.86	160.49	216.76	166.32	144.75	97.28	180.44	157.29	139.64	123.59
	4	285.45	242.15	202.74	176.36	247.18	215.34	144.62	95.17	187.23	168.34	148.71	135.63
	5	330.45	269.45	247.35	205.76	274.34	256.17	218.24	124.38	198.43	178.26	164.23	149.94
	6	371.69	292.45	258.56	231.45	293.79	258.34	221.38	133.89	218.29	193.92	176.34	160.88
	7	385.46	330.56	289.86	250.64	321.63	262.76	231.88	156.46	229.32	204.21	185.37	171.29
	8	407.45	339.78	328.45	262.36	354.39	305.28	236.46	168.66	244.16	215.08	201.34	186.20
24 %	1	101.45	95.96	85.63	93.56	132.86	64.29	56.48	102.92	130.55	92.03	86.34	82.19
	2	125.36	116.46	107.64	105.72	175.54	114.73	60.46	103.73	150.06	105.34	97.86	94.78
	3	146.45	134.26	127.87	116.47	203.62	153.49	132.64	85.49	174.34	151.83	132.47	116.37
	4	167.48	144.76	135.78	130.66	234.28	203.66	132.48	81.94	180.09	161.74	142.69	128.06
	5	202.47	175.92	154.59	139.86	262.28	243.37	204.86	116.48	191.59	172.30	157.07	141.99
	6	236.45	217.59	177.86	156.45	280.64	244.37	207.29	120.87	212.08	184.55	169.34	155.07
	7	267.45	254.79	199.48	164.86	207.19	286.11	220.79	155.86	223.19	197.82	178.61	169.07
	8	294.48	267.46	208.73	187.86	343.46	293.53	224.83	155.64	237.12	208.09	195.11	179.23
28 %	1	118.46	58.53	70.82	84.63	127.64	61.81	55.16	99.48	124.09	86.24	80.24	74.38
	2	147.89	96.59	77.97	123.58	171.53	110.26	57.12	99.57	146.34	99.21	91.86	88.49
	3	166.49	134.56	86.65	134.62	199.29	149.83	129.38	82.67	168.66	145.09	126.54	110.06
	4	173.64	171.79	107.86	153.76	231.92	198.29	130.27	79.26	175.27	154.29	136.39	121.76
	5	175.28	212.72	155.46	182.53	259.73	240.29	201.64	112.56	184.37	165.06	150.34	136.34
	6	189.81	250.84	180.77	190.69	276.29	241.62	203.53	115.82	206.03	176.31	178.29	148.34
	7	214.26	285.48	187.69	221.56	301.46	281.49	217.28	138.39	217.21	191.76	172.37	159.06
	8	237.45	290.63	209.63	244.54	339.83	289.92	220.47	153.56	230.86	201.37	189.36	172.82

swelling rate of hydrogels in three different acids *viz.* acetic acid, perchloric acid and hydrochloric acid were also studied. Among these, acetic acid showed the highest swelling as compared to the other two acids and also the swelling increases with the increasing concentration of acids. Herein, 20 % aluminum sulfate cross-linked hydrogels showed a high swelling rate as compared to 12, 16, 24 and 28 % aluminum sulfate cross-linked hydrogels.

Effect of salt solution: Hydrogels are considered as polyelectrolytes, which suggest that the ionic strength of hydrogels increases with decreasing porosity. Further addition of cations leading to a non-perfect anion-anion repulsion because of charge screening effect. The amount of absorbent or swelling rate decreases with decreasing of the mobile ion concentration difference between the gel and aqueous phases [26]. The multivalent cations in salt-solutions, ionic cross-linking sites on the surface of particles lead to a significant decrease in swelling ability. It is evident that the decrease in swelling strongly depends on the medium.

The effect of cationic radius on swelling characteristics, an equilibrium swelling absorbency was measured in four series of 0.4, 0.6, 0.8 and 1.0 M salt solutions of K^+, Na^+, Ca^{2+} and Mg^{2+} and the results are summarized in Table-3. It is known that Mg^{2+} and Ca^{2+} can chelate with carboxylic (COO^-) group, leading to a compact network and causing further shrinking to the hydrogel. On the other hand, it was also found that smaller the radius of atoms of monovalent atomic cation, more the water absorption capacity. Thus, the swelling rate decreased quickly because of higher cationic charge of Ca^{2+} and Mg^{2+} than that of monovalent charge of K^+ and Na^+ [27].

Conclusion

Carboxymethylcellulose/poly(acrylamide) (CMC/Amm) hydrogels were prepared using ammonium persulfate as an initiator and aluminium sulfate as a cross-linking agent. Herein, aluminum sulfate was acted as an complexion for carboxylate anion of CMC Na salt. The swelling rate (%) of hydrogels has been studied by varying the concentration of aluminum sulfate

TABLE-2
SWELLING RATE VALUES OF HYDROGELS IN DIFFERENT pH SOLUTIONS AT 37 °C

Aluminum sulfate	Time (h)	pH 1.2	pH 2.2	pH 4.0	pH 6.8	pH 7.0	pH 9.5
12 %	1	83.76	100.43	148.52	207.91	220.47	232.83
	2	150.49	235.73	251.62	268.17	280.26	289.10
	3	170.27	242.71	262.19	269.09	276.30	280.18
	4	185.53	245.02	260.03	274.18	280.76	287.60
	5	196.18	252.09	265.19	277.40	285.38	294.37
	6	211.69	255.72	275.17	282.14	292.37	300.42
16 %	1	88.15	104.23	153.12	210.92	224.29	243.88
	2	153.66	240.21	263.09	276.19	280.31	271.56
	3	176.59	250.12	274.59	279.31	289.08	286.39
	4	191.18	257.44	279.07	283.06	294.19	297.72
	5	209.37	260.10	283.79	288.08	299.58	303.09
	6	220.93	267.19	286.93	291.76	304.73	307.12
20 %	1	95.27	109.32	157.92	219.57	230.88	235.71
	2	165.37	253.07	269.31	284.18	291.29	298.09
	3	177.86	263.19	280.22	289.32	298.67	306.44
	4	192.70	271.78	282.71	293.52	305.26	310.19
	5	208.18	277.15	288.17	295.27	310.78	316.79
	6	221.86	285.07	294.70	298.07	315.93	319.08
24 %	1	70.49	93.07	139.17	201.71	214.79	219.72
	2	110.28	241.23	261.19	273.23	275.37	281.09
	3	120.86	248.32	263.82	275.21	277.89	282.01
	4	128.27	253.17	274.61	284.92	286.79	287.23
	5	138.39	257.28	279.15	285.01	289.21	291.17
	6	145.86	264.15	284.92	292.32	293.67	294.17
28 %	1	61.34	88.23	134.21	194.29	203.76	199.19
	2	104.26	233.22	252.72	266.21	268.34	272.91
	3	113.51	237.21	255.21	268.31	271.09	275.72
	4	120.37	247.01	267.21	275.27	278.36	279.19
	5	131.28	249.28	271.34	277.19	280.19	284.62
	6	139.33	257.19	269.19	278.18	283.17	287.11

TABLE-3
SWELLING RATE VALUES OF HYDROGELS IN DIFFERENT SALT SOLUTIONS

Aluminum sulfate	Conc. (M)	12 h				24 h			
		KCl	NaCl	$CaCl_2$	$MgCl_2$	KCl	NaCl	$CaCl_2$	$MgCl_2$
12 %	0.4	370.86	340.09	310.23	288.29	377.46	345.62	314.30	293.83
	0.6	230.08	200.49	190.58	165.91	235.83	203.60	193.02	167.05
	0.8	200.20	185.29	140.18	117.82	205.76	188.09	142.16	119.48
	1.0	120.29	80.16	70.18	67.29	124.62	82.50	72.36	69.27
	1.2	75.09	60.18	40.08	35.29	78.27	62.79	42.09	37.37
16 %	0.4	382.86	351.79	319.27	297.27	395.14	359.03	326.01	403.07
	0.6	243.59	216.28	208.27	171.34	258.48	222.09	218.37	270.76
	0.8	216.38	198.49	153.64	128.21	229.09	206.93	159.38	239.05
	1.0	130.88	93.50	84.27	80.27	137.53	101.22	92.06	146.37
	1.2	88.27	73.64	54.21	50.28	65.90	82.07	62.18	75.55
20 %	0.4	403.07	365.86	334.33	220.31	414.21	376.73	346.22	231.34
	0.6	270.76	231.06	226.31	184.06	279.05	242.09	238.61	196.09
	0.8	239.05	215.28	167.24	150.89	250.92	227.49	170.88	162.89
	1.0	146.37	108.07	98.27	91.27	157.28	117.64	100.79	93.07
	1.2	75.55	88.34	70.09	67.29	84.87	99.08	81.20	78.33
24 %	0.4	411.53	372.01	346.88	223.62	413.21	376.30	349.91	227.32
	0.6	275.39	238.09	231.82	189.33	278.10	241.09	236.77	194.86
	0.8	246.08	223.16	163.04	154.20	249.16	226.60	167.08	158.86
	1.0	113.67	113.10	94.83	89.27	117.41	117.00	98.30	93.02
	1.2	95.22	94.06	73.24	70.11	99.57	97.20	77.28	77.28
28 %	0.4	409.37	373.21	345.07	221.30	412.36	375.53	347.06	224.30
	0.6	271.66	273.03	232.21	190.27	269.0	240.19	235.30	193.26
	0.8	246.04	222.10	163.00	154.37	248.90	225.30	166.96	157.28
	1.0	114.88	112.34	94.92	89.65	115.02	115.02	96.37	93.18
	1.2	95.09	94.91	72.19	62.08	96.00	96.30	75.96	65.06

from 12 to 28 %. The effect of prepared hydrogels were also investigated in different concentrations of some inorganic salts, where the swelling rates were found to be decreased with increasing concentration of salts.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

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