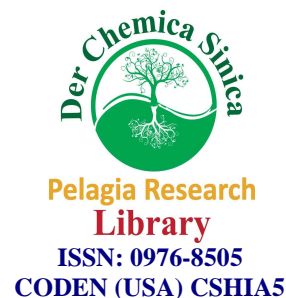




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Acrylic acid and methacrylic acid based hydrogels-A review

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ABSTRACT

Hydrogels are three-dimensional polymeric networks those swell quickly by imbibing a large amount of water or deswell in response to even miniscule changes in their external environment. The water uptake or swelling may be thousands of times of their initial volume in water. The volume phase transitions as a response to different stimuli make these materials interesting objects of scientific observations and useful materials for use in advanced technologies. These materials have earned the sobriquet of stimuli responsive or smart gels. The stimulus response of the gels makes these potential materials for uses in variety of applications. Due to their stimuli responsiveness these gels are used in diverse fields such as environment management technologies, controlled drug release, chemomechanical acuator, bioreactor, solute recovery reagent, environment sensitive membrane, mass separation devices, chemical valves, biosensors and in separation and enrichment technologies. In the present review, an attempt has been made to highlight the recent advances in the hydrogels of acrylic acid (AAc) and methacrylic acid (MAAc).

Key words: acrylic acid; methacrylic acid; hydrogel; swelling.

INTRODUCTION

Stimuli sensitive gels undergo reversible changes in their volume and shape under certain physicochemical factors [1]. Due to the range of properties these gels are used in diverse fields such as controlled drug release [2], chemomechanical acuator [3,4], bioreactor [5], solute recovery reagent [6], environment sensitive membrane [7], mass separation devices [8], chemical valves [9], biosensors [10] and in separation and enrichment technologies [11]. This wide spectrum of applications emanates mainly from their capacity to undergo large volume phase transition in response to slight changes in their external environment [12]. The large volume changes reflect conformational transitions in their network chains from an expanded state to a collapsed one. These changes can be induced by changing the surrounding medium composition [13], solvent nature [14], pH [15-17], temperature [18], electric field [19-21], mechanical force [22] and by adding charged and uncharged linear polymer to the gel system [22, 23]. Many a published works reports fundamental as well as experimental studies on hydrogels [24-26]. Dusek, Patterson and Prins proposed a theoretical analysis of equilibrium swelling of swollen network in a single solvent system [27-28]. These studies led to the conclusion that the swollen network phase in equilibrium with pure solvent undergoes a transition into two phases those differ in degree of swelling. Later Tanaka [12] and Ilavsky [29] demonstrated the practical relevance of these theoretical considerations. In the recent past a number of reviews have been published in which this phenomenon is generally attributed to the polyelectrolyte nature of the networks [30].

ACRYLIC ACID BASED HYDROGELS

AAc and MAAC are monomer acids. These on crosslinking afford high water absorbing hydrogels both as single component and as part of the multi-component systems. Due to the ionizable carboxylic acid, these hydrogels are sensitive to pH and ionic strength [31]. These monomers have been used as second component, grafted on polymer surfaces or used as blends to obtain a wide variety of the hydrogels. Zouahri *et al.* [32] have reported synthesis and characterization of graft copolymers prepared from ozonized high-density polyethylene (HDPE) followed by the copolymerization with monomers such as AAc and N,N-Dimethylamino-2-ethyl methacrylate. Most of the copolymers based on 4'-(6-acryloxy)hexyloxy-4-methoxyazobenzene and AAc exhibit *trans-cis-trans* isomerization in a solid film [33]. Poly[(AAc-*co*-N-vinyl pyrrolidone (NVP))] forms hydrogen bonding between the -C=O of the NVP and the -CO₂H of the poly(AAc). Swelling of poly(AAc-*co*-NVP) in a higher pH medium is significantly different from that in low pH solutions. This pH responsiveness of the hydrogels makes it suitable for gastric drug delivery systems [34].

Networks of AAc and isophorone diisocyanate containing a poly(ethylene glycol) chain have been reported [35]. Membrane of poly(vinyl alcohol) (VA) and poly(AAc-*co*-MAAc) blend has been also reported [36]. Membranes prepared from the complexes of polyelectrolyte complexes containing poly(AAc) and their copolymers with acrylamide (AAm) or methacrylamide (MAAm) show remarkable transport properties in dialysis and in pressure driven processes. Low density polyethylene (LDPE)-*g*-poly(AAc) membranes prepared by the direct radiation grafting of aqueous AAc solutions onto LDPE films have good permeability to glucose and urea, which increases with the hydrophilicity of the membrane [37]. Temperature and pH sensitive microfiltration membranes prepared from the blends of the poly(AAc)-*g*-poly(vinylidene fluoride) and poly[(AAc)-*g*-N-isopropyl acrylamide (N-i-PAAm)] exhibit both pH dependent and temperature sensitive permeability to the aqueous solutions, with the most drastic change in the permeability being observed at permeate pH (between 2 and 4) and temperature around 32°C [38]. The stimuli sensitive properties of crosslinked polybetaines based on AAc and ethyl 3-aminocrotonate gels were studied as a function of pH, ionic strength, water-organic solvent mixture, electric, and combined electric and magnetic fields. Appearance of pH gradient within the polyampholyte gel matrix under the externally imposed DC electric field was observed [39].

Hydrogels based on lightly crosslinked poly(methacrylamidophenylboronic acid-*co*-AAc) swells monotonically at pH 7.4 with increasing glucose concentrations. At pH 10, these hydrogels first shrink and then reswell with increasing glucose concentration [40]. A series of superabsorbent hydrogels prepared from carrageenan and partially neutralized AAc are pH sensitive and even as low as 1% carrageenan (sodium salt) increases the equilibrium degree of swelling of the hydrogels many folds [41].

The effects of composition of the interpenetrating network (IPN), pH, and temperature of the swelling medium and salts were investigated on the kinetics of the swelling of the binary IPN of poly(VA) and poly[acrylamide (AAm)-*co*-AAc] by Bajpai *et al.* [42]. A series of temperature and pH sensitive elastomers based on poly(AAc-AAm) show very quick swelling rate [43]. A semi interpenetrating polymer network (SIPN) of carboxymethyl cellulose (CMC) and crosslinked poly(AAc) has been prepared and its water-sorption capacity was evaluated as a function of chemical architecture of the IPN, pH, and temperature of the swelling medium. The water uptake potential of the IPNs has also been investigated in solutions containing inorganic salts and simulated biological fluids [44]. The hydrogels based on poly[(AAm-*co*-AAc)-*cl*-N, N- methylene bisacrylamide (N, N-MBAAm)] exhibit good dimension stability and high yet reversible swelling at 60°C [45]. Synthesis of thermosensitive poly(N alkylacrylamide) hydrogels by amidation of poly(AAc) have been reported [46]. Crosslinked poly(1-vinyl imidazole-*co*-acrylic acid) hydrogel was synthesized by radical polymerization by Rivas *et al.* for uranyl ion adsorption [47]. Synthesis and swelling behaviour of poly(N-i-PAAm-*co*-MAAc) hydrogel were carried out and swelling of this hydrogel was found to be affected by hydrogen bond formation [48]. Synthesis and solution properties of comb like polymers from octadecyl methacrylate and AAc have been reported [49]. *In situ* synthesis of a suspension agent based on a poly(Sty-*co*-AAc) for the suspension polymerization of Sty has been reported [50]. Well-ordered micro-domain structures in polydisperse poly(Sty-*co*-AAc) di-block copolymers from controlled radical polymerization are reported [51]. Mixtures of a diethylamine-functionalized dodecyloxy methoxy biphenyl mesogen and poly(AAc) were investigated as a function of composition by infrared spectroscopy (IR), differential scanning calorimetry (DSC), polarizing optical microscopy, and X-ray diffraction. These components form a monophasic supramolecular complex for amine/acid molar ratios below about 0.5 and a biphasic system composed of the supramolecular complex and a mesogen-pure phase for higher molar ratios [52].

METHACRYLIC ACID BASED HYDROGELS

The nature of the pH dependent collapse of poly(MAAc) hydrogels is investigated using ^1H solid-state NMR. In aqueous solution, poly(MAAc) changes from an expanded conformation at high pH to a compact contracted form at low pH, where hydrogen bonds play a central role [53]. Poly(MAAc) and poly(2-ethyl-2-oxazoline) (PEOZO) are a polyacid/polybase pair capable of forming reversible, pH-responsive, hydrogen-bonding complexes stabilized by hydrophobic effects in aqueous media. At high pH, the expected conformational expansion of the poly(MAAc) units is enhanced at moderate levels of PEOZO incorporation [54]. Aqueous saline solutions of poly(AAc) and poly(MAAc) were studied by phonon correlation spectroscopy and viscometry over a wide concentration range and pH values. In solutions of poly(MAAc), the transition to the region of semi dilute solutions is preceded by the formation of stable clusters of macromolecules which is not observed in poly(AAc) solutions [55].

Various methacrylamides have been synthesized in good yields directly from MAAc using microwave irradiation in a solvent free environment [56]. The swelling of poly(ethylene oxide-*co*-MAAc) hydrogels is characterized by the existence of a collapse in the 4-6 pH range, which is related to the formation of a stable interpolymer complex [57]. The dynamic swelling behaviour of poly(MAAc-*co*-methacryloxyethyl glucoside) and poly(MAAc-*g*-ethylene glycol) hydrogels has been investigated. The mechanism of water transport through these gels is significantly affected by the pH of the medium [58].

Hydrogen bonding effects on the dynamic swelling of poly(N-i-PAAm-*co*-MAAc) has been reported [59]. Chemically induced dynamic electron polarization studies of a pH dependent free radical cage formed in a photo initiator labeled poly(MAAc) has also been reported [60]. Synthesis, characterization and swelling behaviour of poly(AAm-*co*-MAAc)-*g*-Gum ghatti superabsorbent hydrogels was reported [61]. Thermal stability of the poly(HEMA-*co*-MAAc) blend by the intermolecular bond between poly(HEMA) and poly(MAAc) has been reported [62]. Thermal degradation of hydrogels of different carbazolyethyl methacrylate with MAAc has been reported [63]. Poly(AAm-*co*-MAAc) and poly(AAm-*co*-AAc) have thoroughly been investigated. Poly(AAm-*co*-AAc) has been found to be homogeneous while poly(AAm-*co*-MAAc) is not [64]. Poly(N-vinylcaprolactam-*co*-MAAc) hydrogel microparticles for oral insulin delivery has been reported [65]. Swelling behaviour of poly(N-vinylcaprolactam-*co*-MAAc) have been investigated as a function of pH and surfactant concentration [66]. Interpolymer complex microparticles based on polymethacrylic acid-chitosan for oral insulin delivery have been investigated [67]. Kumar *et al.* reported the synthesis and characterization of pH sensitive poly(Ethylene glycol dimethacrylate -*co*-MAAc) microparticles for oral insulin delivery [68,69]. Synthesis and characterization of poly(MAAc) hydrogels for metoclopramide delivery has been reported [70]. An investigation of fluorescence correlation spectroscopy into the diffusion of fluorescein-tagged dextran in a poly(MAAc) hydrogel was reported [71].

CHARACTERIZATION OF HYDROGELS

Elemental analysis and spectroscopic methods such as FTIR has been frequently used to deduce composition of the networks [72]. Similarly, other chemical and physical attributes of the polymers those are affected after the network formation and post polymer reactions, include surface morphology, thermal stability, pore structure and pore distribution and water uptake among other things [73]. Various attempts have been made to specify a gel with simple parameters such as the branching fraction [74] or the correlation length [75]. The thermal characterization of the networks can be investigated by differential scanning calorimetry, thermogravimetric analysis and dielectric analysis [76]. The spin-spin relaxation time (T_2) of hydrated samples to probe the states of the imbibed water in the hydrogels is an important method of characterization [77].

Swelling study of various polymeric gels is one of the most extensively used methods of characterization [78-80]. Small angle neutron scattering studies have also been used as a method of characterization [81]. Kofinas and Cohen [82] have reported a variety of methods which satisfactorily characterize the structure of polymeric hydrogels beyond the level of information provided by a single swelling test. These methods include wide-angle X-ray diffraction, which verifies the structure and reproducibility across synthesized hydrogel batches and measurements of the temperature and frequency dependent dielectric relaxation provide information on the network topology. Thermogravimetric analysis is another useful method used for the characterization of polymers [83-84].

Some recent works on the hydrogels of the polymers based on AAc, MAAc and their acrylamides and methacrylamides are reviewed in Table 1

Table 1: Recent Progresses in AAC, MAAC Based Hydrogels

Polymer Composition	Remark	Ref.
poly(AAc-co-AAm)	Preparation and characterization of macromolecular nanostructures and ultra-thin hydrogels were done.	[85-86]
Poly(N-vinyl caprolactam-co-AAc)	The phase separation behaviour of copolymers shows a temperature, pH and molecular weight dependence.	[87]
IPN hydrogel of chitosan- poly(AAc)	Synthesis and characteristics was carried out.	[88]
Poly(AAc-g-ethylene)	Thermo and pH sensitive similar to the IPN hydrogel.	[89]
Poly(AAc-co-isoctylacrylate)-cl-N,N-MBAAm	Reversible, mass-changing pH responsive hydrogel used as a pH sensor.	[90]
Poly(N-i-PAAm-co-AAc)	Micropatterned, temperature responsive, self-assembled polymers were prepared.	[91]
Poly(N-i-PAAm-co-AAc)	Temperature and pH sensitive hydrogels.	[92]
Poly(AAc-co-AAm)	Synthesis and swelling was carried out.	[93]
Poly[(AAc)-g-poly(2-ethyl-2-oxazoline)]	Synthesis, complex formation, and dilute-solution associative behaviour were studied.	[94]
Poly(N-i-PAAm-co-AAc)	Preparation and electrochemical characterization of gels, swollen by non-aqueous solvents was carried out.	[95]
Poly(AAc-co-styrene)	<i>In situ</i> syntheses of copolymer for the suspension polymerization of styrene was carried out.	[96]
EGDMA crosslinked poly(AAc-co-2-(N-ethylperfluorooctanesulfoamido) ethyl methacrylate, poly(AAc-co-stearyl acrylate), poly(AAc-co-lauryl acrylate)	Synthesis and characterization was carried out.	[97]
poly(Styrene-co-AAc)	Well-ordered, micro-domain structures in polydisperse controlled radical polymerization was observed.	[98]
Poly(AAm-co-AAc)	Synthesis and rheological properties of hydrogel.	[99]
Poly(N-i-PAAm-co-AAc)	Forms conjugates with alanine mono, di and tripeptide.	[100]
poly(AAc-g-polypropylene)	Pervaporation and separation of water/ethanol mixtures were carried out.	[101]
Poly(AAc-co-2-[(methacryloyloxy)-ethyl]trimethylammonium chloride)	The formation of polyelectrolyte complexes of different molecular weights hydrogels has been studied.	[102]
Chitin/poly(AAc)	Synthesis and orientation study of a magnetically aligned liquid crystal was carried out.	[103]
Poly(AAc-g-ethylene glycol) hydrogel	Drug transport mechanisms, release kinetics studied.	[104]
poly(vinyl alcohol)/poly(AAc) IPN	Synthesis, characteristics and swelling studies.	[105]
Poly(Propylene glycol-co-AAc)	Synthesis, characteristics and water sorption of IPN hydrogels were carried out.	[106]
Poly(AAc-co-N-butyl acrylate)	Amphiphilic cylindrical brushes with shell to obtain a narrow length distribution of the brushes were observed.	[107]
poly(AAc-co-butyl acrylate)	Prepared by reversible addition fragmentation chain transfer polymerization.	[108]
poly(AAc-co-hexamethyldisilazane)	Prepared by plasma copolymerization.	[109]
Poly(AAc)	Preparation of hyper-branched polymer <i>via</i> self-condensing atom transfer radical copolymerization.	[110]
Poly(MAAC-g- lignocellulose) (jute)	Preparation by UV radiation induced polymerization, shows 42% increase in hydrophilicity.	[111]
Poly(MAAC) and poly(AAc) multilayers	Preparation and modification has been carried out.	[112]
Poly(AAc-g-CMC)	Graft copolymers show high water absorbency.	[113]
Poly(AAc) crosslinked by γ -radiations	Synthesis, characteristics and swelling performed.	[114]
Poly(AAm-co-AAc/polyethyleneimine)	Swelling and mechanical properties of the superporous hydrogel studied. Gelation kinetics changes according to the acidic nature of the reactant.	[115]
Poly(AAm-co-AAc)-cl-N, N-MBAAm	Swelling behavior of hydrogel have been investigated. Average molecular weights between crosslinks and diffusion/swelling characteristics have been evaluated.	[116]
Poly(AAc-methyl-co-methacrylate-co-butyl acrylate)	The pervaporation ability increases with swelling.	[117]
Carrageenan/ AAC superabsorbent hydrogels	Preparation, swelling kinetics and the equilibrium degree of swelling of the hydrogels were carried out	[118]
AAm and AAc grafted on to different celluloses such as wood pulp, films and non-woolen fabrics	Super absorbents, used as carriers in drug release and as sensors in to stimuli as heat, electrical field and pH changes.	[119]
Poly(AAc)	Uptake of Ag ⁺ ions was studied.	[120]
Poly(AAm-co-AAc)-cl-N, N-MBAAm	Synthesis, characterization, and swelling behaviour of superabsorbent hydrogels have been carried out.	[121]
Poly(maleic anhydride-alt-AAc)	Synthesis of Poly(maleic anhydride-alt-AAc) and its transition-metal complexes with Ni(II), Cu(II), and Cd(II). The temperature dependence of the conductivity of the copolymer and polychelates of transition metals was investigated.	[122]

Poly(MAAc) doped with tin dioxide	The effect of UV irradiation on antimony-doped tin dioxide thin films derived from poly(MAAc) modified precursors was investigated.	[123]
Poly(ethylene-co-MAAc)	Measurement of diffusion of paramagnetic species, Mn ²⁺ was carried out.	[124]
Poly(MMA-co-MAAc)	Synthesis and characterization of a UV-sensitive aqueous base developable lithographic plate was carried out.	[125]
Poly(MAAc)-g-poly(vinylidene fluoride)	Preparation of membranes with pH responsive separation characteristics.	[126]
Poly(Ethylene-ran-MAAc)	Imaging and X-ray microanalysis was studied.	[127]
poly(AAc-co-AAm)	The preparation and characterization of macromolecular nanostructures possessing an amphiphilic core-shell morphology was carried out	[128]
poly(AAc-co-AAm)	physicochemical properties of the ultra-thin hydrogels prepared by the sequential formation of amide linkages between commercially available poly(AAc-co-AAm) hydrogels was carried out	[129]

FACTORS AFFECTING PROPERTIES OF HYDROGELS

Apart from the environmental factors, polymerization conditions such as nature and concentration of monomer and crosslinker affect hydrogel structure, hence swelling behaviour and end-uses of the hydrogels. According to Elliott, the effects of ionic strength and pH on the network structure are interrelated [130]. Different factors those affect water uptake of the hydrogels, being relevant in the present study are discussed.

Effect of Polymer Composition and Structure

The initial AAm/AAc mole ratio and crosslinker concentrations affect swelling behaviour of the random copolymer poly(AAm-co-AAc)-*cl*-N,N-MBAAm as the swelling capabilities of the hydrogels decrease with the increasing AAc and crosslinker concentrations [131]. Network parameters such as molecular mass between crosslinks, crosslink density, mesh size and porosity affect swelling properties of the hydrogels [132]. Non-ionized groups of poly(N-i-PAAm-co-sodium acrylate) gel form hydrogen bonding between non-ionized (-CO₂H) and/or between the -COOH and -CONH- groups. The replacement of the counter ion (Na⁺ by H⁺) and formation of intermolecular hydrogen bonding explains macroscopic polymer network shrinkage [133].

A super fast swelling response in hydrogels can be developed by taking THF as a foaming and a precipitating agent during the polymerization. Such response is attributed to the macroporous network and heterogeneous matrix, and it may contribute significantly to the construction of novel intelligent devices in the future [134]. Chen *et al.* [135] have reported AAc based superabsorbent polymers with partially neutralized AAc synthesized by inverse suspension polymerization. Further, to improve the properties of salt resistance and anti-electrolyte of the polymer, AAm containing non-ionic and hydrophilic groups were introduced into the system, which can suppress the salt and homion effect. The water absorption was observed to be proportional to the reciprocal of ionic strength. Gradual phase transition of poly(N-i-PAAm-co-AAc) gel induced by electric current has demonstrated that temperature-induced gradual phase transitions of poly(N-i-PAAm-co-AAc) gel are mainly driven by transport before polymerization and protonation of the AAc, caused by the electric current [136]. Hydrogen bonding and electrostatic interactions increase with MAAc content in poly(MAAc-co-methacryloxyethyl glucoside) prepared by free radical polymerization [137].

Linear poly(MAAc) modified with methoxy-terminated, long-chain (MW = 5000) poly(ethylene glycol) (PEG) represent a polyacid/polybase pair form reversible, pH responsive, hydrophobic complexes stabilized by hydrogen bonding. Acidic conditions produce coil collapse, arising from the formation of intramolecular hydrogen-bonded complexes along the polymer backbone. Under basic conditions, coil expansion takes place due to the conformational changes (polyelectrolyte effect), as well as the reduction of the intermolecular complexation as the number of carboxylate anion increase [138]. Isoelectric point of the copolymers changes continuously from 2.8 to 6.7 with increasing the 1-vinylimidazole (VIm) content in the polyampholytes of VIm and sodium salts of AAc and MAAc [139]. The volume transition of hydrogels is adjusted by varying the crosslink density of the shell [140].

Effect of Polymerization Techniques

Living and controlled polymerization techniques have been used recently for the design and synthesis of novel complex polymers with MAAc segments. Solution behaviour and morphologies derived from their amphiphilic polymers having three-dimensional architectures have been reported [141]. Characterization of aqueous micellar solutions of amphiphilic block copolymers of poly(AAc) and poly(sty) toward the control of the number of particles

in emulsion polymerization has been reported [142]. A novel emulsion polymerization technique referred to as emulsifier-minor emulsion polymerization was achieved by the copolymerization of MAAC or AAc with a combination of methyl methacrylate, butyl acrylate, and Sty. The water resistance of the film can be improved using technique other than the conventional emulsion polymerization and its foaming capacity can be lowered [143].

The thermoresponsive phase behaviour of gels based on N-i-PAAm/AAc prepared with and without the use of ultrasound was investigated. The former showed thermoresponsive swelling behaviour with a large hysteresis over a wide range of temperatures around its phase-transition temperature and that increased with the AAc content. The former were also characterized from the viewpoint of chemical, hydration, and macroscopic physical structures [144]. The effect of the absorbed dose on the gel fraction and on the swelling in aqueous solutions at different pH in case of the hydrophilic films based on poly(AAc)/poly(vinyl methyl ether) blends crosslinked by gamma radiation has been reported. In acidic media, the films have low swelling degree due to the suppression of carboxylic groups ionization and formation of additional physical crosslinks via interpolymer hydrogen bonding [145]. Nanogels of poly(AAc) synthesized by intramolecular crosslinking of the linear macromolecules in aqueous solution and induced by a few microseconds pulses of fast electrons were reported to be much more resistant to free-radical induced degradation than the parent linear chains [146]. The free-radical copolymerization of AAc with AAm has been studied in aqueous media in the presence of ammonium chloride. The reactivity ratios r_1 and r_2 were reported to vary with the $[\text{NH}_4\text{Cl}]$. The reactivity of AAc depends on its dissociation degree controlled by the ionic strength of the solution [147]. The effect of aqueous medium containing poly(AAc) on the morphology of the composite polymer particle, produced by two stages of soapless emulsion polymerization, has been reported [148].

Effect of Swelling Conditions

A small ion concentration range within the hydrogel influences properties of hydrogels obtained by the free radical copolymerization of the methacrylated dextran and AAc. The swelling response of hydrogels to changes in external pH and ionic strength is governed mainly by the ionic osmotic pressure due to the accumulation of diffusible ions within hydrogels [149]. pH dependent growth and morphology of the poly(AAc) have also been reported [150]. Swelling of hydrogel prepared by grafting AAc on styrene-maleic anhydride copolymer both by chemical and radiation methods was performed as a function of temperature, time and pH. Maximum swelling (3290%) was observed at 37°C in 12h at 9 pH for the grafted sample prepared by radiation method [151]. Chauhan *et al.* synthesized superabsorbent, pH and temperature sensitive and moderately salt tolerant hydrogels namely poly(AAm-co-AAc) and poly(AAm-co-MAAc). These hydrogels respond quickly to the changes in their external environment [152].

A series of water-swellaible, temperature and pH sensitive elastomers based on poly(AAc-co-AAm) show very quick swelling rate. The polymer reaches its equilibrium state in only 30 min. The equilibrium swelling ratio increases with increase in the amounts of water absorbent resin. The higher the amount of poly(AAc-co-AAm), the higher is the swelling degree. The nature of the sorbed metal ions adversely affect swelling behaviour which decreases dramatically with the increase in the charge number of the cation, but was not influenced by radius and valence state of the anion [45]. Linear and nonlinear viscoelastic properties of poly(AAc) and poly[AAc-co-2-acrylamido-2-methylpropane sulfonic acid(AAmPSA)-*g*-poly(N,N-dimethylacrylamide)(N,N-DMAAm)] mixtures have been investigated as a function of pH. At $\text{pH} < 3.75$, strong hydrogen bonding interpolymer complexation between poly(AAc) and poly(N,N-DMAAm) side chains in semi dilute solution results in the formation of a transient network of the hydrogen bonded complex of poly(N,N-DMAAm/poly(AAc)) [153]. The effects of pH and ionic strength on the swelling behaviour of poly(dimethylaminoethyl methacrylate-co-AAc) synthesized by UV radiation have been reported [154].

The swelling kinetics of a series of Poly (N-i-PAAm-co-MAAc) copolymers and their homopolymers, *i.e.*, poly(N-i-PAAm) and Poly(MAAc) under different acidic and neutral conditions have been studied to probe the hydrogen bonding ability of comonomers [155]. The swelling behaviour of poly(AAm-co-AAc-co-trimethyl-N-acryloyl-3-aminopropyl ammonium chloride) gel in electrolyte solutions of NaCl and MgCl_2 is induced by the ionic pressure due to Na^+ and/or Mg^{2+} ions in the acid region ($\text{pH} = 4$ to 7) as well as basic region, where Na^+ or Mg^{2+} ions added in solution are exchanged with H^+ ion supplied by the yet non-dissociated -COOH. In the pH region 1 to 4, a Cl^- ion generates the ionic pressure. In the case of bivalent cations, especially Mg^{2+} , it is observed that the interactions between the localized fixed charges and the bivalent cations may result in the formation of weak crosslinks [156]. For chitosan/poly(AAc) complex a high swelling ratio of about 600 times its dry weight is observed in water as well

as in low salt and solvent concentration [157]. The swelling capacities of poly(AAm-co-AAc) prepared by gamma irradiation were in the range of 1000-3000%, while poly(AAm) hydrogels swelled in the range of 450-700% [158].

Bending behaviour between two electrodes has been studied under various conditions for IPN hydrogels composed of poly(AAc) and polyacrylonitrile. The swelling ratio of the IPN hydrogels decreases with an increasing [NaCl] in an aqueous solution. As the applied voltage increases, the bending speed increases. The deformation of the IPN hydrogels is reversible when the applied voltage was turned on and off [159]. The effect of the concentration and molecular parameters of random copolymers of poly(AAc-co-sodium acrylate) on their performance as additives reducing the hydraulic resistance of turbulent flows in oil-in-water emulsions is quantitatively studied [160]. Water/ethanol pervaporation performance of asymmetric polyelectrolyte complex constructed by the diffusion of poly(AAc) in chitosan membrane, has been reported [161].

The ion specific swelling behaviour of poly(AAc) gels prepared by γ -rays irradiation in the presence of 0.01 M HCl have been reported. The anion specificity for the swelling ratio is similar to that for many kinds of hydrogels, i.e., $\text{Cl}^- < \text{Br}^- < \text{NO}_3^- < \text{I}^-$, while the cation specificity proved to be rather unusual (i.e., $\text{Mg}^{2+} < \text{Ca}^{2+} < \text{Li}^+ < \text{Na}^+ < \text{K}^+ < \text{Cs}^+$). According to the results, the marked deswelling of poly(AAc) gel in the presence of strongly hydrated cations was ascribed to the unfavourable hydration to the acidic proton of poly(AAc) due to the reduced availability of water oxygen as well as to the destabilization of hydrophobic hydration developing around the uncharged poly(AAc). Synthesis and solution behaviour of homo and copolymers based on N-substituted acrylamide reveals the formation of associates with inter and intramolecular hydrogen bonds. The selective interactions occur between high and low molecular mass components of the polymerization mixture and macromolecules pass through a set of conformational states, depending on the nature of the medium and the presence of acceptors of hydrogen bonds in the system [162].

Gradual volume phase transition of poly(N-i-PAAm-co-AAc) gel induced by electric current has been reported. Temperature induced gradual phase transition of the gel is mainly driven by transport before polymerization and protonation/ionization of the AAc, caused by the electric current [163]. Swelling behaviour of poly(N-i-PAAm)/poly(dimethyl siloxane) (PDMS) SIPNs show that incorporation of hydrophobic PDMS decrease swelling degrees of IPNs without affecting their LCST whereas addition of acrylated PDMS as crosslinker instead of N, N-MBAAm into the structures of these hydrogels change their LCST along with their swelling degrees [164]. Mun *et al.* reported that growth of the polyethylene glycol-MAAc copolymers gelation with increase in polyethylene glycol molecular weight is accompanied by decreasing of equilibrium swelling degree of the networks [165]. Water absorption by the superabsorbent poly(AAc-co-AAm) improves in the supercritical CO_2 [166].

Effect of Surfactants on Properties of Hydrogels

Many researchers have been trying to elucidate the effect of surfactants on the swelling equilibrium of polymer gels from both fundamental as well as technological aspects [167-170]. Many studies have been reported on the effect of surfactants on the swelling of polyelectrolyte gels [171-172]. Surfactants like sodium dodecyl sulfates (SDS) are reported to affect hydrogel water uptake by micelle formation [173]. The local mobility of the surfactant molecules in the complexes is much lower than their mobility in "free" micelles. As a result of increasing surfactant concentration in solution, the hydrophilic associates of the surfactant micelles with the polyacid having an unfolded coil conformation are formed. The local mobility of the surfactant molecules those enter association is considerably higher than in the complex with a compact conformation, and this mobility grows with the increasing number of micelles involved in the associate [174]. The viscosity and gelation of mixtures of modified poly(AAc) and a cationic photosensitive surfactant can be controlled reversibly by changing UV and visible light irradiation of the sample. At the critical aggregation concentration of the surfactant, micellar aggregates form on the polymer and solubilize the alkyl side chains grafted on the hydrogel, leading to physical crosslinking and gelation [175]. The swelling equilibrium of poly(AAc) and poly[AAm-co-(itaconic acid)(IAAc)] hydrogels was studied as a function of temperature in aqueous solutions of the surfactants such as SDS, and cetyltrimethylammonium bromide (CTAB) in water and re-entrant conformational transitions depend on temperature. The equilibrium volume swelling ratio of P(AAc/IAAc) increases sharply in SDS and CTAB solutions with an increase of the mole percent of AAc [176]. Constant shear and shear dependent viscosity measurements are reported in the terpolymers of AAm, N-alkylacrylamide and AAc with added anionic surfactant SDS. Surfactant mediated network formation with polymer coil expansion and hydrogen bonding between partially ionized AAc has been observed [177].

Viscosity data are reported for the aqueous solutions of a series of AAm and AAc based polymers with added surfactants, SDS, diethylhexyl sulfosuccinate, dodecyl trimethyl ammonium bromide. In the presence of anionic surfactants, the solutions exhibit a dramatic increase in the solution viscosity around the CMC, which is attributed to the interpolymer crosslinking through the formation of mixed micelles involving the hydrophobes from different polymer chains and the surfactant molecules. The viscosity enhancement increases with the increase of hydrophobicity of the hydrophobe and decreases with the increasing AAc incorporation in the polymer [178]. The molecular dynamics and the structure of the molecular complexes formed by the micelles of dodecyl substituted poly(ethylene glycol) with poly(MAAc) and poly(AAc) in aqueous solutions were studied by viscosimetry, pH measurement, and electron spin resonance spin-probe techniques. At low surfactant concentrations, the conformation of the complex is a compact globule. The local mobility of surfactant molecules in such a complex is much slower than that in the "free" micelle. At high surfactant concentration, the nonionic micelles and polyacids form hydrophilic associates [179]. A strong effect of the charge inhomogeneity on the degree of swelling has been observed in hydrogels obtained via inhomogeneous binding of anionic surfactant, SDS to lightly crosslinked poly(N-i-PAAm), while the other based on the physical entrapment of poly(AAc) within the poly(N-i-PAAm) chain network [180]. Macroscopic phase separation in poly(AAc) networks following the absorption of CTAB/chloride from the aqueous solutions has been studied. The gels show solvent swollen polyelectrolyte network (core), surrounded by a dense surface phase of polyion/surfactant complexes [181].

CONCLUSION

It is evident from the foregone discussions, that AAc and MAAc based hydrogels have attracted the attention of a large number of research groups in recent past. These gels have been synthesized and used in diverse fields worldwide by many researchers. One of the most important properties of these hydrogels is their ability to respond very quickly to the changes in their external environment by exhibiting volume phase transition. Due to their smartness or stimuli responsiveness these gels are referred to as *smart gels* or *stimuli responsive gels*. The stimuli responsiveness makes these gels an object of scientific observations and useful material for variety of applications. The most widely studied applications include drug delivery, selective ion uptake/sorption, ion exchange, enrichment and separation technologies, environment management technologies, sensors and many more. Properties of these gels are affected by a number of factors including polymerization technique employed, nature of crosslinker, crosslinking density, nature of comonomer/s swelling time, temperature, pH and presence of electrolyte and surfactants.

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