



Comparison between Acrylic Acid and Methacrylamide on Release and Swelling Properties for Hydrogels based on PVP

Mokhtar Heidari Naghdeali and Maryam Adimi
Department of Chemical Engineering, Farahan Branch,
Islamic Azad University, Farahan, IRAN

(Corresponding author: Maryam Adimi)

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ABSTRACT: In this experimental study, eight hydrogels based on Polyvinyl pyrrolidone were prepared. The four hydrogels based Polyvinylpyrrolidone/acrylic acid by changing the concentration of acrylic acid monomer was prepared. The swelling and release test of these hydrogels were examined. And other hydrogels based Polyvinyl pyrrolidone/Methacrylamide by changing the concentration of Methacrylamide monomer was prepared. The swelling and release test for all of samples were done. Anti-asthma drug aminophylline was used to test for drug delivery in the phosphate buffer solution as the same body pH. The results showed that the acrylic acid monomer had higher swelling and controlled released.

Keywords: Hydrogels, PVP, Controlled Release, Swelling Rate, Acrylic Acid, Methacrylamide.

INTRODUCTION

Hydrogels are polymer networks extensively swollen with water. Researchers, over the years, have defined hydrogels in many different ways. The most common of these is that hydrogel is a water-swollen, and cross-linked polymeric network produced by the simple reaction of one or more monomers (Buchholz and Graham, 1998, Brannon and Harland, 1991, Li *et al.*, 2013). The ability to swell depends on pH, temperature and ionic strength environment, type of solvent and the polymer structure (Gaharwar *et al.*, 2011). Hydrogels have numerous applications, including cosmetic products, preservatives, water, moisture retention, coagulant, sludge thickener, drug delivery systems and lenses in ophthalmology. This polymer gets big part of the global markets due to the economic value and technical interest of it (Ganji and Farahani, 2009). So if we can obtain useful information about the synthetic hydrogel dehydration, that information can be used in making a controlled drug delivery system (Sirousazar *et al.*, 2008, Jones *et al.*, 2002). Polyvinylpyrrolidone has excellent water solubility, absorbency and biocompatibility. PVP has low toxicity and it is used in medical, food, cosmetics and as a film-forming agent (Yeh *et al.*, 2006). PVP is a component of hydrogels that is widely used for biomedical applications (Rostak and Olejniczak, 1993). Hydrogels of PVP have limited applications because of its inferior mechanical properties. To increase its mechanical properties, Polyvinylpyrrolidone and its monomer, N-vinylpyrrolidone have been copolymerised with acrylic acid, methacrylates and other vinyl monomers (Ghilzai,

2003). Acrylic acid (AA) is a pH and electrically sensitive material. It forms complexes with poly bases (Ray *et al.*, 2008). Poly acrylic acid (AAc) is known to be a good muco adhesive and may increase the transit time of formulation. The polymers, composed of acrylic acid, have the ability to absorb a large amount of water and are used in many applications including ion exchange resins, personal hygiene products and membranes for hemodialysis, ultrafiltration, and controlled drug release (Ekici and Isikver, 2003). Methacrylamide-based hydrogels are attended more than the other hydrogels because of their high water absorbency (due to existing hydrophilic amide groups), high mechanical stability of the swollen hydrogels and their green characteristics (Barati *et al.*, 2010). For these reasons Recently a great deal of interest has been shown by few workers on synthesis, characterization and swelling studies of PVP based hydrogels (Serano *et al.*, 2002, Zhai *et al.*, 2000, Park and Nho, 2003, Bajpai, 2000). This work focused on synthesizing pH-sensitive Polyvinylpyrrolidone/Acrylic acid and Polyvinylpyrrolidone / Methacrylamide hydrogels with in situ polymerisation for colon targeting. Different formulations with variation in the content of monomers were evaluated in terms of swelling and drug release and attempts were made to correlate these with the network parameters. Finally Aminophylline drug delivery of hydrogel samples (Aminophylline known as an anti-asthma drug) was measured using UV devices at pH = 7.4 and was used the data obtained from the release for developed of mass transport model to calculate the penetration rate of the hydrogel.

MATERIALS AND METHODS

A. Materials used in the preparation of hydrogels

Polyvinylpyrrolidone K25 (PVP, Base polymer: Sigma-Aldrich, $[C_6H_9NO]_n$, $M = 2.5 \text{ g/mol}$). Acrylic Acid 99% (AA, Monomer: Merck Chemical Company, $C_3H_4O_2$, $M = 72.06 \text{ g/mol}$). Methacrylamide (MAAm, Monomer: Penta Chemical Company, C_4H_7NO , $M = 85.11 \text{ g/mol}$). N,N' -Methylenebisacrylamide 99% (MBA, Cross link: Merck Chemical Company, $C_7H_{10}N_2O_2$, $M = 154.17 \text{ g/mol}$). Potassium Persulfate 99.99% (KPS, Initiator: Sigma-Aldrich, $K_2O_8S_2$, $M = 270.32 \text{ g/mol}$). Aminophylline (Drug: Caspian Pharmaceutical Co, $C_{16}H_{24}N_{10}O_4$, $M = 420.427 \text{ g/mol}$). Deionized water was used in all experiments.

B. Method of preparation of hydrogels based on PVP/AA

Beginning Powder Polyvinylpyrrolidone and monomers of acrylic acid were poured in beakers and mixed very well with the N,N' -methylenebisacrylamide and distilled water by stirring, and then the initiator potassium persulfate added and continue until

ingredients are dissolving well in distilled water. Then the beakers put in a water bath to 373 (K) after 20 minutes a hydrogel produce. Then take them in distilled water for 24 hours to put up the unreacted monomers are removed from the hydrogel and then put it in a vacuum oven with a temperature of 30 degrees will cause it to dry. Compositions of various formulations are given in Table 1 (Naghdeali *et al.*, 2014). Possible structure of polyvinylpyrrolidone/acrylic acid hydrogel was showed in (Fig.1), (Lei *et al.*, 2009, Dafader *et al.*, 2012).

C. Method of preparation of hydrogels based on PVP/MAAm

The Powder Polyvinylpyrrolidone and monomer of Methacrylamide were mixed distilled water and synthesis of hydrogel samples have been conducted in a manner that was. Compositions of various formulations are given in Table 2. Possible structure of Polyvinylpyrrolidone/Methacrylamidehydrogel was showed in (Fig. 2).

Table 1: Materials and wt% of hydrogels construction PVP/AA

Sample designation	AA (wt%)	PVP (wt%)	MBA (wt%)	KPS (wt%)
AA (1)	2.84	0.948	0.948	0.379
AA (2)	3.75	0.939	0.939	0.375
AA (3)	5.53	0.922	0.922	0.367
AA (4)	6.39	0.914	0.914	0.365

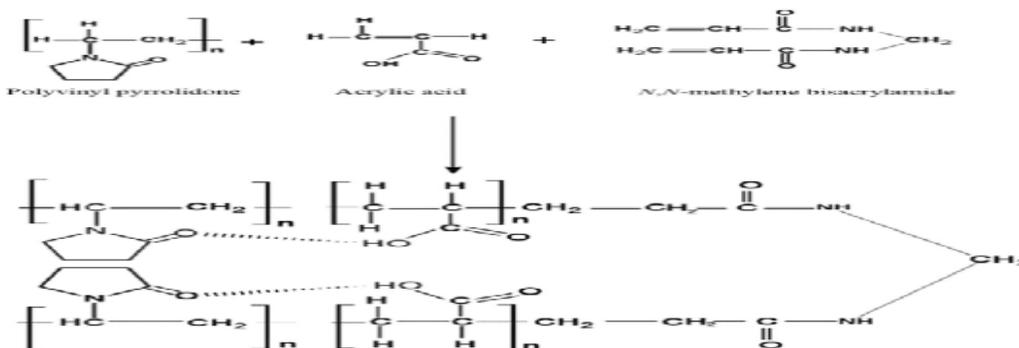


Fig. 1. Possible structure of polyvinylpyrrolidone/acrylic acid hydrogel.

Table 2: Materials and wt% of hydrogels construction PVP/MAAm.

Sample designation	MAAm (wt%)	PVP (wt%)	MBA (wt%)	KPS (wt%)
MAAm (1)	2.835	1.323	0.945	0.378
MAAm (2)	3.745	1.310	0.936	0.374
MAAm (3)	5.514	1.286	0.919	0.367
MAAm (4)	7.220	1.263	0.902	0.361

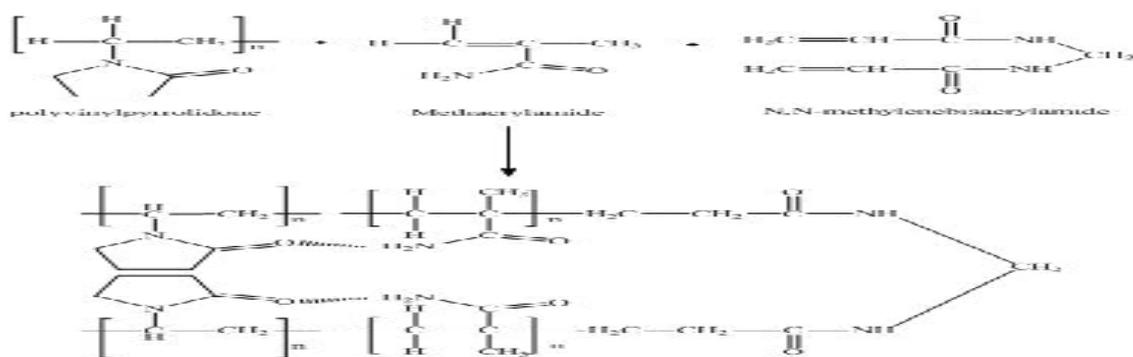


Fig. 2. Possible structure of Polyvinylpyrrolidone/Methacrylamide hydrogel.

D. Uploading drug

Since there is likely to reactions between initiator and drug, the present study used indirect method. For loading drug, the amount of hydrogel (1gr) in 100 mL of solution with a specification concentration of the drug was soaked for 5 hours. To prevent degradation of the drug during this period, the solution was placed in the dark and at room temperature. The drug concentration in the environment was measured by using UV spectrometer Model UVD-3200 made in Labomed companies in America. The amount of drug loaded was determined between initial and final concentrations of the drug in solution.

E. Drug delivery

The drug-loaded hydrogels were placed in the phosphate buffer solution pH = 7.4 for 5 hours and the drug release rate was measured by the UV device at successive time and Delivery rates were fixed for 3 hours in duration.

RESULT AND DISCUSSION

A. Swelling measurements

First, blank hydrogels (1cm length, 2.5mm diameter) were freshly made and then dried in freeze drier for 24 hr at -20°C . The swelling studies were carried out in triplicate by placing of blank gels in 50 mL water at room temperature. In time intervals, the gels were removed, gently dried with a kim-wipe and weighed, and then returned to the vials with 1 mL of fresh water or solution. The swelling percent (%ESR) was estimated by comparing the ratio of the wet hydrogel weight (M_{wet}), which was measured at the various time intervals, to the initial dry hydrogel weight (M_{dry}), which was measured before the swelling study. Results of swelling for hydrogels were showed in Fig. 3 and Fig. 4 (Brahim *et al.*, 2003):

$$\% \text{ ESR} = \left(\frac{M_{\text{wet}} - M_{\text{dry}}}{M_{\text{dry}}} \right) \times 100\% \quad (1)$$

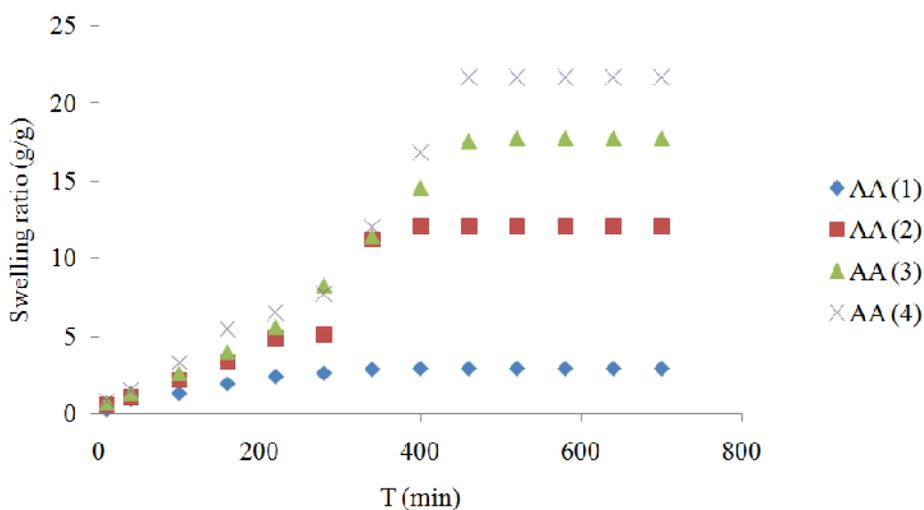


Fig. 3. Swelling ratio Polyvinylpyrrolidone/Acrylic acid (PVP/AA).

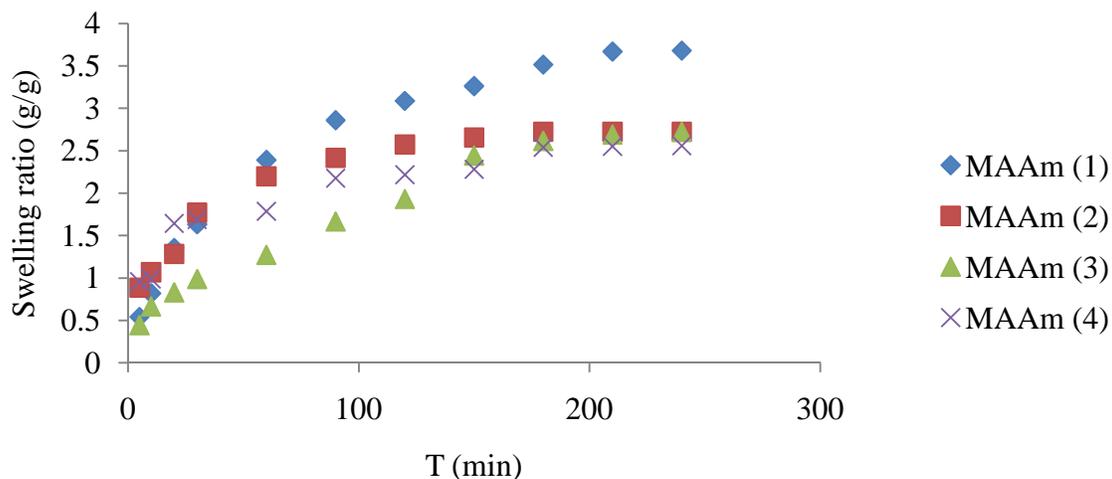


Fig. 4. Swelling ratio Polyvinylpyrrolidone/Methacrylamide (PVP/MAAm).

B. Results of drug delivery

Results of drug release from hydrogels based on Polyvinylpyrrolidone/Acrylic acid and Polyvinylpyrrolidone/Methacrylamide by plotting values of M_t / M versus t was showed in (Fig. 5 and Fig. 6) indicate that M_t concentrations of the drug

delivery solution that hydrogel has been put into it, M difference between the initial concentration of dissolved before putting hydrogel and secondary levels of concentration of Aminophylline solution after placing the hydrogel for 5 hours.

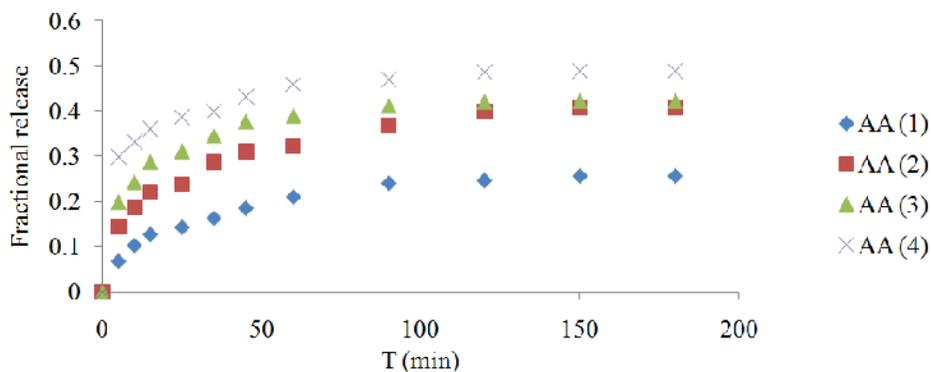


Fig. 5. Fractional release Polyvinylpyrrolidone/Acrylic acid (PVP/AA).

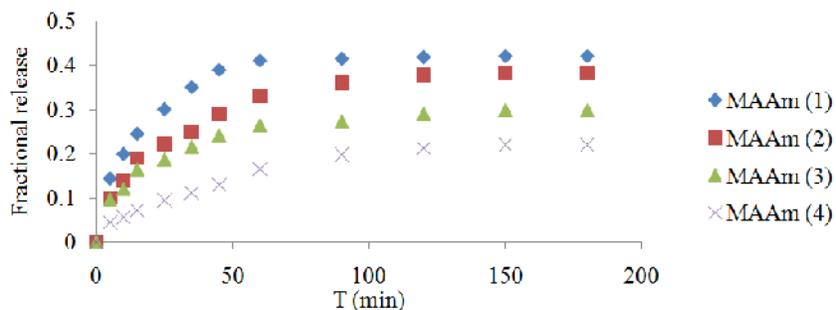


Fig. 6. Fractional release Polyvinylpyrrolidone / Methacrylamide (PVP/MAAm).

CONCLUSION

The results showed acrylic acid monomer have higher swelling and more controlled released from Methacrylamide this is because the acrylic acid monomer is more hydrophilic than Methacrylamidemonomer and acrylic acid water absorption is equal to 2100. On the other hand, Aminophylline is a anti-asthmatic drug, which has a short half-life, rapid onset and its peak effect is so fast and optimum percent of acrylic acid is (AA(4) = 6.398 wt%) that this sample had the most of controlled released.

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