Synthesis and characterization of swelling properties superabsorbent Hydrogel Carboxymethylcellulose-g-Poly (Acrylic Acid)/Natrium Alginate cross-linked by gamma-ray irradiation technique

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Abstract. Superabsorbent hydrogel (SAH) is a hydrophilic polymer network that can absorb and retain large amounts of water in swelling conditions. SAH has been synthesized from Carboxymethylcellulose (CMC) with variations in weight (1.0 g, 1.5 g, 2.0 g, 2.5 g) grafted by poly (acrylic acid) (PAA) and composited with natrium alginate (NaAlg). Samples were cross-linked using gamma-ray irradiation technique in a dose 10 kGy (at a dose rate of 5 kGy/hour). CMC can increase porosity and influence swelling properties. Characteristics of swelling properties in aqueous solutions which are influenced by weight variations of the CMC were evaluated, such as aquadest, urea solution with various concentrations (0.25%, 0.50%, 0.75%, and 1.00%) and salt solution of NaCl and CaCl₂ with variation of concentrations (0.09%, 0.9%, and 1.9%). The Swelling ratio in salt solutions decreased significantly compared to in aquadest. Swelling capacity increased with decreasing cation content (Ca²⁺<Na⁺). A different phenomenon occurs in urea solution, because urea is a neutral molecule, so the presence of urea molecules are not influenced by the electrostatic repulsion of COO⁻ groups in the polymer chain. FTIR measurement showed PAA grafting reaction on CMC and NaAlg cross-linked with gamma-ray irradiation. From SEM measurement the SAH had heterogeneous porosity.

1. Introduction

In recent years, superabsorbent hydrogel (SAH) has great interest for researchers because it can absorb and retain large amounts of water, salt solution or physiology solution and defend physical dimension structure [1, 2]. SAH is a macromolecular network with the ability to absorb and release water reversibly based on external stimulants [3, 4]. SAH can change its volume (swelling/shrinkage) because it’s responsive to the environmental stimulants, such as temperature, pH and ionic strength [5].

SAH can be made from Carboxymethylcellulose (CMC) which has the basis of natural cellulose polysaccharide [6]. CMC is a linear cellulose polymer ether and as an anionic compound obtained by inserting CH₂COOH groups into cellulose molecular chain [7, 8]. CMC has an average degree of substitution (DS) that varies from 0.5 – 1.5 [8]. CMC is in great demand by researchers because of its
unique nature, such as high viscosity, transparent, hydrophilicity, non-toxic, biocompatible and biodegradable [9, 10].

Many studies have been carried out to modify polymers with other materials to increase their absorption ability and physical endurance [11]. Some of the CMC-based SAH studies include, CMC-g-poly (AA-co-AM) [12], CMC-g-poly (NaAA) [13], CMC-g-poli (NaAA)/Kaolin [14], CMC/poli (AA) [15], CMC-g-poly (AM-co-AMPS) [12], dan CMC-g-poly (AA-co-AMPS) [16], CMC-g-poly (AA) [17], CMC-g-poly (NaAA)/Celite [2], CMC/starch [18], CMC/HEC [19], CMC/PVA [20], CMC/Gelatin [21], CMC/NaAlg [22, 23], CMC/Alg [24], CMC/PANI [25], CMC/PE [26], CMC/PVP [27], CMC/CMS [28], CMC/Methacrylate [29], CMC/PVAm [30], CMC-g-PNaA/APT [31], CMC/OMMT [32], CMC/PEG [33], CMC/PEGDE [34], CMC/PEO [35], CMC/Sericin [36], CMC-g-PAAm-co-PNIPAm [37], CMC-g-poly(acrylic acid)/Bentonite [38], β-CD/CMC-co-Poly(AA) [39], Alginate/CMC-g-AA[40], CMC-g-poly (AA-co-AM)/carclyze [41]. The research has shown that CMC-based SAH can increase water absorption and maintain it in swelling conditions.

Grafting copolymerization of acrylic acid (AA) monomers to polysaccharides is an efficient route for SAH preparation where polysaccharides act as the main part because of their properties, such as biocompatible, biodegradable and non-toxic [42]. Neutralization of AA with potassium hydroxide (KOH) results in a more chewy gel. When acrylic acid is neutralized with potassium hydroxide, it forms potassium acrylate [11].

Alginates is a polysaccharide block copolymer which is obtained naturally and studied extensively for tissue engineering applications as an encapsulation matrix and cellular culture [43]. Natrium alginate (NaAlg) is a renewable and biodegradable natural polymer which is specifically used in a variety of commercial applications because of its capacity for gelatinization [12].

There are several techniques for synthesis SAH namely, frontal polymerization [44], grafting polymerization [17], Cross-linked Agent [23] and cross-linked by gamma-ray irradiation [45]. The cross-linked method with gamma-ray irradiation can be carried out in solid, liquid and gas monomer phases and more superior because they produce pure SAH which consists of only one material thus reducing the risks associated with biocompatibility [46]. The technique does not need the addition of a catalyst, initiators, cross-linkers, and others which may be dangerous and difficult to remove [42, 43].

In this study, variations in CMC mass addition were carried out to determine the effect on AA monomers (neutralized with KOH) which were composites with NaAlg. Polymer composite solution is irradiated by gamma ray for crosslinking. sweling properties of SAH were studied to determine the mechanical properties of SAH in aquadest, salt solution, and urea solution.

2. Experimental Methods

2.1. Time and Place of Research

This research was conducted in March-April 2018 in the laboratory of biomaterials, isotope, and radiation center, National Nuclear Technology Agency, South Jakarta, Indonesia.

2.2. Tools and Materials

Tools used in this research includes beaker glass, measuring glass, tea filter, weigh paper, magnetic stirrer, oven Heraeus Instruments Vacuterm, analytical scales Mettler Toledo type AB 204 and Preciso 3000D, Stopwatch, Sealer machine, spatula, aluminum foil, FTIR Shimadzu Prestige-21, scanning electron microscopy (SEM) Carl Zeiss type EVO MA 10, and Irradiator 60Co (Rubber Irradiator, IRKA). The used Materials cover Acrylic acid (AA) with M = 72.06 g/mol from Germany, Potassium Hydroxide (KOH) with M = 56.11 g/mol from Germany, natrium chloride (NaCl) with M = 58.44 from Denmark, calcium dichloride (CaCl2), Urea, CMC-Soluble water with degree of substitution (DS = 0.7 – 0.8), Natrium alginate (NaAlg) and aquadest.
2.3. Synthesis of SAH CMC-g-PAANaAlg
15 mL AA monomer solution was dissolved in 50 mL aquadest and neutralized with ±5.6 g KOH to obtain the degree of neutralization (Dn) Dn = 0.5, afterward stirred until homogeneous solution using a magnetic stirrer [47]. CMC with weight variations of 1.0 g, 1.5 g, 2.0 g, and 2.5 g was dissolved into neutralized AA, stirred again until the homogeneous solution using a magnetic stirrer. Add 1.0 g NaAlg as control and aquadest reach the total volume of 100 ml mixture. Finally, each mixture of solution was packaged in a polypropylene (PP) plastic bag 20 × 10 × 0.5 cm³ [48] using a sealer machine and the sample irradiated at a dose of 10 kGy (dose rate 5 kGy/hour) [49] with gamma isotope source 60Co-γ at room temperature [50]. The final product of irradiated samples was appeared like an elastic transparent gel then cut into pieces [51] and dried in a vacuum oven at 60°C to up to constant weight dried [52]. The dried samples hydrogel was milled using a blender until the powder was formed and filtered to separate the fine and coarse samples.

2.4. Measurement of the Swelling ratio in water
0.1 g of sample poured into 100 ml aquadest at room temperature for 60 minutes with 5 minutes swelling ratio measurement interval. Swelling measurement can be calculated using the equation (3.1).

\[ Swelling\, ratio = \frac{W_1}{W_0} \]  

(3.1)

W₁ is the aquadest mass absorbed by the gel sample and W₀ is the weight of the dry gel [53]. Measurements are made with three repetitions to be averaged [54]. The SAH, which is swelling, is filtered using a tea filter (±200 mesh). Then, filtered water that comes out of the filter is collected in a beaker at an interval of ±1 hour until there is no more dripping water. The volume of water collected in a measuring glass is measured [55].

2.5. Measurement of the Swelling ratio in the salt solution of NaCl and CaCl₂
0.1 g SAH immersed in 100 mL of NaCl and CaCl₂ solutions with variations of concentrations 1.9%, 0.9% and 0.09% for 60 minutes at room temperature. Measurement of the swelling ratio can be calculated using the equation (3.1).

2.6. Measurement of the Swelling ratio in the Urea solution
0.1 g SAH was immersed in 100 mL urea solutions with a varying mass of Urea 0.25 g, 0.50 g, 0.75 g, and 1.00 g for 60 minutes at room temperature. Measurement of the swelling ratio can be calculated using the equation (3.1).

2.7. FTIR (Fourier Transform Infrared) Analysis
Spectrophotometer FT-IR Shimadzu Prestige-21 was expended to determine the binding function of SAH molecules at spectral of interval 400 - 4000 cm⁻¹ with a resolution of 2 cm⁻¹. FTIR analysis was carried out with mixing powder samples with KBr grade IR (1:100) [56] in a mortar and pressed in tablets [57].

2.8. SEM (Scanning Electron Microscopy) Analysis
SEM was used to determine the morphology and porosity of SAH. SAH was soaked in water for 24 hours then frozen for 24 jam. SAH was freeze-dried at a temperature of -105°C [11] and analyzed using scanning electron microscopy (SEM) Carl Zeiss type EVO MA 10.

3. Result and Discussion
The weight ratio of the hydrogel in the swelling state to its dry weight or swelling ratio is one of the main parameters of the hydrogel especially for testing material as absorbance [58].
Figure 1. SAH swelling process scheme in a water medium. The red circle shows some characteristics of the reactive SAH functional group in the swelling process [59].

Hydrogels form polymers that have hydrophilic functional groups in polymer structures, such as amines (NH$_2$), hydroxyl (-OH), amide (-CONH-), -CONH$_2$ and sulfate (-SO$_3$H). The hydrophilic group allows the hydrogel to absorb water and aqueous liquids which produce a hydrogel expansion and a large volume is absorbed. This process is called swelling. During the swelling process, a crosslinked hydrogel structure prevents the separation and damage of the crosslinks in the hydrogel system as in Figure 1.

3.1. *Swelling ratio in water*

Determination of swelling ratio in water is one of the main parameters of the polymer especially for testing an absorptive material [58]. SAH synthesized in this study was tested the swelling ratio in the water against CMC mass differences. The relationship of immersion time variation with swelling ratio can be seen in Figure 2.

Figure 2. The swelling ratio of SAH in water with a variation of time from 5 to 60 minutes with 5 a minute immersion interval.

The swelling ratio of SAH shows the mass effect of CMC from 1.0 g, 1.5 g, 2.0 g, and 2.5 g on maximum capacity for 60 minutes respectively, which is 491 g/g, 485 g/g, 460 g/g and 327 g/g in water. It’s showed that the water uptake of SAH was influenced by CMC weight. The increase of CMC weight will reduce porosity and increase the density of SAH consequently the water uptake decreased.
However, the increase of density crosslink of SAH induced stronger power to maintain water capacity in the SAH network.

### 3.2. Swelling ratio in a salt solution of NaCl and CaCl$_2$

NaCl and CaCl$_2$ solutions with 1.9%, 0.9%, and 0.09% concentration, respectively, are prepared to observe the effect of salt environment on the swelling ratio. Swelling ratio in salt solution increased significantly compared to aquadest [53]. The swelling ratio is often associated with additional cationic screening effect that induces imperfect anion electrostatic repulsion, which leads to decrease in osmotic pressure (difference in ionic pressure) resulting from differences in ion concentration resulting in network shrinkage [16]. The relationship of variations in the concentration of NaCl and CaCl$_2$ salt solutions with swelling ratio can be seen in Figure 3.

![Figure 3. Swelling ratio of SAH in NaCl and CaCl$_2$ solutions with variations in the concentration 1.9%, 0.9%, 0.09%](image)

The swelling ratio of SAH in salt solution shows the effect of CMC mass from 1.0 g, 1.5 g, 2.0 g and 2.5 g to maximum durability in NaCl at concentration 0.09 % in a row namely 156.6 g/g, 170 g/g, 173 g/g, 213.3 g/g. While in CaCl$_2$ solution at 0.09% is 160 g/g, 173.3 g/g, 180 g/g and 186.6 g/g in a row. Swelling capacity is increased with decreased of cation content (Ca$^{2+}$$<$$Na^+$) [41]. Increased of ionic strength in NaCl solution, water uptake from SAH decrease, because the cation concentration (Na$^+$) in salt solution will neutralize carboxyl groups and sulphonate [60]. Moreover, multivalent cation Ca$^{2+}$ can neutralize several content in the gel by complex formation of carboxamide or carboxylic groups, which induce an increase of ion crosslinking content and consequently, eliminate the swelling [62]. Ion valence influenced swelling capacity, that is the smaller size of cation, the swelling capacity is great [1].

### 3.3. Swelling ratio in the Urea solution

The most important chemical properties to be tested from SAH on a commercial scale as a personal care ingredient is the value of swelling ratio in urine [63]. The urine content is dominated by urea compounds so that the swelling test is carried out with a urea solution at various concentrations of 0.25%, 0.50%, 0.75%, and 1.00%. The relationship of variations in the concentration of urea solution with the swelling ratio can be seen in Figure 4.

![Figure 4. Swelling ratio of SAH in the Urea solution](image)
Figure 4. Swelling ratio of SAH to urea solution with various concentrations 0.25%, 0.50%, 0.75% and 1.00%

A large difference in osmotic pressure between the gel and solution phases results in osmosis of water into the gel phase is greater than in urea, NaCl and CaCl₂ solution. In urea solution occurs a difference phenomenon, because urea is a neutral molecule so that molecule is unaffected a reject electrostatic repulsion from COO⁻ in polymer chain [64]. moreover, urea molecules have a hydrophilic side such as NH2 that will interact with the solution. So that, the absorption polymer in urea solution increases compared to water which is 623.3 g/g, 610 g/g, 540 g/g and 473.3 g/g at concentration 0.25%.

3.4. FTIR Analysis

IR spectrum was obtained with KBr disc and recorded at a spectral range of 4000 - 500 cm⁻¹ that can be seen in Figure 5. IR spectrum of CMC is marked with a green line. The strong peak at 3442.93 cm⁻¹ indicated hydroxyl (O–H) stretching vibration, then strong peak at 2924.08 cm⁻¹ and 2854.64 cm⁻¹ shows methylene (C–H) groups vibration. the absorption peak at 1739.79 cm⁻¹ and 1637.56 cm⁻¹ representing carboxylic (C=O) groups and the signal at 1458.18 cm⁻¹ could be assigned stretching vibration of carbonyl (–COO) groups [40]. IR Spectrum of PAA is marked with a violet line. Vibration at 3439.07 cm⁻¹ indicated hydroxyl groups (O–H), peak at 2918.29 cm⁻¹ and 2848.86 cm⁻¹ shows methylene (C–H), CH₃ or CH₂, the absorption at 1631.77 cm⁻¹ representing carbonyl groups stretching from carboxyl (C=O), aldehyde or ketone, and then, peak at 1053.13 cm⁻¹ is (C–O) groups stretching vibration which is sign the form of polysaccharides [65]. IR spectrum of NaAlg is marked with a blue line. Wide vibration at 3001.23 cm⁻¹ until 3471.86 cm⁻¹ shows hydroxyl groups (O–H), sharp vibration at 2920.22 cm⁻¹ and 2908.65 cm⁻¹ indicated methylene (C–H), and carbonyl groups (–COO) detected at wave number 1598.98 cm⁻¹ [47]. While the graft process between CMC and PAA which is then composited with NaAlg raises several detected functional groups such as carboxylic (C=O), hydroxyl (O–H), and amide (N–H) in SAH which are shown at wave number 1729.37 cm⁻¹ [66] and 3653.18 cm⁻¹ [67, 68]. Carbonyl groups (–COO) contained previously in CMC at a wavelength of 1458.18 cm⁻¹ were not detected in SAH due to the grafting process, the more graft copolymers prepared, the more homopolymers content will be removed [40].
3.5. Morphological and Porosity of SAH

SAH is soaked in water to maximum swelling and freeze-dried before SEM analysis [69]. Then the sample is cut with a knife to expose the inner surface and coated with gold to increase conductivity [70]. SAH has a rough surface, a porous structure and forms a wide network. The pores in the composite structure help in water absorption [71].

SEM analysis is used to determine SAH porosity. Porosity affected the absorption rate of SAH [54]. Increased porosity size is affected by CMC [72]. It is assumed that porosity is the area of water permeation and the interaction of external stimulus with the grafted of copolymer hydrophilic groups [73]. The hydrophilic properties of hydrogels can be seen from the water absorption capacity caused by the level of porosity [74]. Morphology shows greater surface area indicating SAH has a higher
absorption capacity. The structure of closed tissue extends after being immersed in water which causes the volume to expand and increased space which can be seen in Figure 6. With 2.5 g CMC, the structure can accommodate many water molecules and form a barrier to hold the water molecules overflowing [69].

4. Conclusions
In this study, SAH CMC-g-PAA/NaAlg has been successfully cross-linked using gamma-ray irradiation technique. The weight gain of CMC in SAH has an effect on the swelling ratio in water, urea, NaCl and CaCl₂ solution. Swelling ratio in salt solution decreased significantly compared to distilled water. Swelling capacity increased with decreasing cation content (Ca²⁺<Na⁺). A different phenomenon occurs in urea solution, because urea is a neutral molecule, so the presence of urea molecules does not affect the electrostatic repulsion of the COO⁻ groups in the polymer chain.

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