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Cassava waste pulp – poly(acrylamide-acrylic acid) based hydrogels using gamma irradiation

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Abstract. Cassava waste pulp (onggok) based hydrogels were synthesis with added of acrylic acid (AA) and acrylamide (AAM). The used of onggok promises superior properties such as eco - friendly, lower prices because the raw materials are available naturally in abundant quantities compared to synthetic polymers, and biodegradable. Gamma irradiation method at doses of 0, 10, 20, and 30 kGy was carried out to produced cross-linked structures. Swelling, gel fraction, physical strength, and morphology were parameters of the hydrogels. The most optimum result was obtained from the addition of 25% acrylic acid and irradiation dose of 10 kGy, with swelling of 9423%, diffusion coefficient of 0.2393, gel fraction of 75.98%, the hardness of 8.53 mJ, and elasticity modulus of 7.01 kPa.

1. Introduction

Hydrogel is a three-dimensional polymer that has ability to absorb water superiorly without dissolved in water [1]. High water absorption ability and its unique characteristic cause hydrogel have been used in the many factory, such as planting media, fever plaster, urea immobilization, biomedical application, biosensor, hygienic products, etc [2-4]. from synthetic materials such as PVA, acrylic acid (AA), acrylamide (AAM), acrylonitrile, etc [1-5]. The use of synthetic materials leaves waste that was difficult to decompose in nature. Therefore, natural polymer-based hydrogel have been most issued of many researchs because of its abundance in nature. Natural polymer can be obtained from carbohydrates such as starch, cellulose, pectin, and chitosan. [6-8].

However, natural polymer-based hydrogels have weaknesses, including unstable mechanical strength and low absorption capacity [9]. Therefore, to obtain same properties to synthetics, natural polymer-based hydrogel must be grafted with synthetic material to produce hydrogel with good absorption and easy to decompose in nature [10]. This research synthesized hydrogel from onggok which were waste of tapioca production. Tapioca production leaves a solid residue in the form of onggok as much as 75% of the raw material. Onggok contained high amount of natural carbohydrate, around 82.7% [7-11]. Carbohydrates consist of amylose and amylopectin which along the chain were composed of hydroxyl groups, so they were hydrophilic [12]. Hydrophilicity of onggok become the main factor that causes onggok suitable as a raw material for hydrogels.



The characteristics of onggok in this study were improved by grafting acrylic acid (AA) and acrylamide (AAM). AA and AAM are commonly used in hydrogel production because of their hydrophilicity because they have a carboxylic anion that can form bonds with water using hydrogen bonds [1]. The monomer grafting process in the onggok polymer chain was carried out by the cross-linking method. In addition, cross-linking method produces three-dimensional structures that allow hydrogels to be insoluble in water [13].

The cross-linking process can be carried out using chemical methods and gamma ray. The use of gamma ray was more favored because the process takes place quickly, does not require catalysts, crosslinkers, and initiators which were generally toxic during the polymerization process, there was no residue in the process, and radiation doses can be determined as needed [14]. The aims of this study was to produce onggok-based hydrogels that were grafted with AA and AAM with high absorption ability and stable mechanical properties using gamma irradiation.

2. Experimental

2.1 Materials

The cassava waste pulp (onggok) of this study were received from local industry in Bogor, West Java, Indonesia. Acrylic acid from Sigma Aldrich, Germany. Acrylamide from Merck, Germany. The others materials were Aquadest, ethanol with 95% of concentration, potassium hydroxide with 10% of concentration, aseton, and methanol. Crosslinking process initiated with gamma irradiation from Irradiator ^{60}Co Gammacell 220 at Isotope and Radiation Application Center BATAN, Lebak Bulus, South Jakarta Indonesia. Instruments used for testing the characteristics of hydrogels were Scanning Electron Microscopy (SEM) JSM – 6510LA JEOL at State University of Jakarta, texture analyzer Brookfield CT 3 at Indonesian Center for Agriculture Postharvest Research and Development, Bogor, West Java, Indonesia.

2.2 Methods

This process was modified from Erizal [14]. Raw material, 10 grams of onggok dissolved in 50 ml of distilled water at a temperature of 90 °C at a speed of 300 rpm using a magnetic stirrer to gelatinize. Gelatinized cassava was cooled at room temperature, then the addition of 2.5 g of acrylamide has been dissolved in 25 ml of distilled water, followed by the addition of neutralized acrylic acid which has been neutralized with varying percentages of 0, 5, 15, 25, and 35% of the total mixed solution while stirring using a magnetic stirrer with a speed of 400 rpm at room temperature for 60 minutes until homogeneous.

The suspension were put into plastics and then the plastics were sealed. Subsequently, gamma irradiation exposure from Cobalt 60 irradiator with the dose varied to 0, 10, 20, and 30 kGy were given at a rate of 5 kGy h⁻¹. Then the suspension were dried in oven at 60 °C to a constant weight.

2.3 Gel Fraction

Gel fraction shows the degree of cross-linking formed in the hydrogel. Measurement of gel fraction begins with weighed the hydrogel in dry conditions. Furthermore, soaking was done in water for 24 hours. After soaking, the hydrogels were dried in an oven at 60 °C for 6 hours. Furthermore, the dry mass after immersion is weighed using a digital scale. Gel fraction was determined by the equation 1.

$$\text{GelFraction (\%)} = \frac{m_g}{m_0} \times 100\% \quad (1)$$

m_g is mass of gel after immersion (gram) and m_0 is mass of dry gel (gram) [15].

2.4 Equilibrium Degree of Swelling (EDS)

Equilibrium Degree of Swelling (EDS) was measured by the percentage of the ratio of the mass of water absorbed to the initial mass of the hydrogel. Swelling can be calculated by the equation 2.

$$EDS (\%) = \frac{m_s - m_0}{m_0} \times 100\% \quad (2)$$

m_s is mass of hydrogel after soaked for 6 days (gram) and m_0 is mass of dry hydrogel (gram) [15].

2.5 Swelling Kinetics and Diffusion Coefficients

Swelling kinetics were done by soaking the hydrogel in water and 0.04 M urea solution for 144 hours. Data retrieval was done in the first 5 hours and subsequent data retrieval was done every 24 hours. Fick's law was used to determine the rate of diffusion of water and to categorize hydrogels based on the rate of outflow and entry of water. This category was determined from the swelling exponent value obtained from the equation 9.

$$F = \left(\frac{M_t}{M_\infty} \right) = Kt^n \quad (9)$$

The rate of diffusion in hydrogel was determined using equation 10.

$$F = \frac{M_t}{M_\infty} = 4 \left(\frac{Dt}{\pi L^2} \right)^{0.5} \quad (10)$$

F is swelling fraction ($g\ g^{-1}$), M_t is hydrogel mass at immersion time t (h), M_∞ is equilibrium mass of hydrogel (g), D is diffusion coefficient ($m^2\ h^{-1}$), n is swelling exponent, and K is swelling constant (h^{-1}). The constant K was determined from the intercept of the regression equation of the curve between $\ln F$ and $\ln t$, while n is determined from the slope of the curve. The diffusion coefficient (D) is determined from the slope of the curve of F and $t^{1/2}$ [3-16].

2.6 Mechanical Properties

The testing of hydrogel mechanical properties in this study was carried out using a texture analyzer instrument. Hydrogels that have been dried were soaked in water for 24 hours. Then the samples were pressed using a probe, and the maximum load data (m , grams), hardness (mJ), and strain ε (mm^{-1}) were obtained. Stress σ (kPa) was calculated using equation 11.

$$\sigma = \frac{F}{A} = \frac{m \times g}{A} \quad (11)$$

Modulus of elasticity (Y) was calculated by the ratio of stress to strain, as in the equation 12.

$$Y = \frac{\sigma}{\varepsilon} = 3Nk \quad (12)$$

F is maximum weight were given to sample (N), A is the cross-sectional area of probe = $1.27 \times 10^{-4} m^2$, m = maximum load were given to sample (kg), g is gravity = $9.8\ m/s$, N is density of crosslink (crosslink m^{-3}), k is Boltzmann constant = $1.38065 \times 10^{-23}\ J\ K^{-1}$, and T is room temperature = $300\ K$.

2.7 Data Analysis

This research was designed used a complete randomized design (CRD) of two factors. The first factor was the percentage of acrylic acid, which were 0, 5, 15, 25, and 35%. The second factor was the dose of gamma ray with variations of 0, 10, 20, 30 kGy. Each treatment consisted of 3 replications, so that $5 \times 4 \times 3 = 60$ experimental units were obtained. The test data were analyzed using SAS software

and continued with the Significant Difference test at the 5% level. If there is a significant difference, Duncan's Multiple Range Test was performed.

3 Results and Discussion

3.1 Proximate Analysis

Proximate analysis was carried out to analyze the characteristics of the onggok raw material used in this study. The proximate test is a chemical analysis used to identify the content of a substance. The proximate test results shown in Table 1 show the onggok used in this study has a carbohydrate content of 82.41%. According to Srinorakurata et al. [17], the carbohydrate content of 82.41% consists of starch around 61.84 - 69.9% and cellulose around 10.61 - 14.35%. The content of starch and cellulose indicates the number of hydroxyl groups along the main chain of onggok polymer, so it is suitable as a raw material of hydrogels.

Table 1 Proximate analysis of onggok

Moisture	Ash	Fat	Protein	Carbohydrate
% (ww ⁻¹)				
12.45 ± 0.08	0.56 ± 0.00	3.98 ± 0.04	1.69 ± 0.11	82.41 ± 0.06

3.2 Gel Fraction

Gel fraction showed the efficiency of the crosslinking process. As can be seen in Figure 1, the addition 5% of acrylic acid increased gel fraction at each gamma-ray dose. However, in the sample with 15 and 25% of acrylic acid, the gel fraction decreased and then increased when the percentage of acrylic acid became 35%. Acrylic acid that has been neutralized will produce carboxylic anions that easily bind to water using hydrogen bonds [1]. The increasing of acrylic acid percentage caused the carboxylic group contained in the hydrogel polymer increased so that the absorption ability of the hydrogel will be higher. However, acrylic acid was a weak acid having a double bond carbon and a carboxylic acid or aliphatic ester which has high reactivity. This high reactivity caused acrylic acid had a high sensitivity to binding reactions [18]. This causes the amount of acrylic acid added to have a saturated value. When it reaches the saturation point, the addition of acrylic acid in a larger amount causes the bonds between the polymer chains to become tighter. The tightness of the cross-linking that is formed will result in a higher gel fraction value, but it causes an insignificant reduction or addition

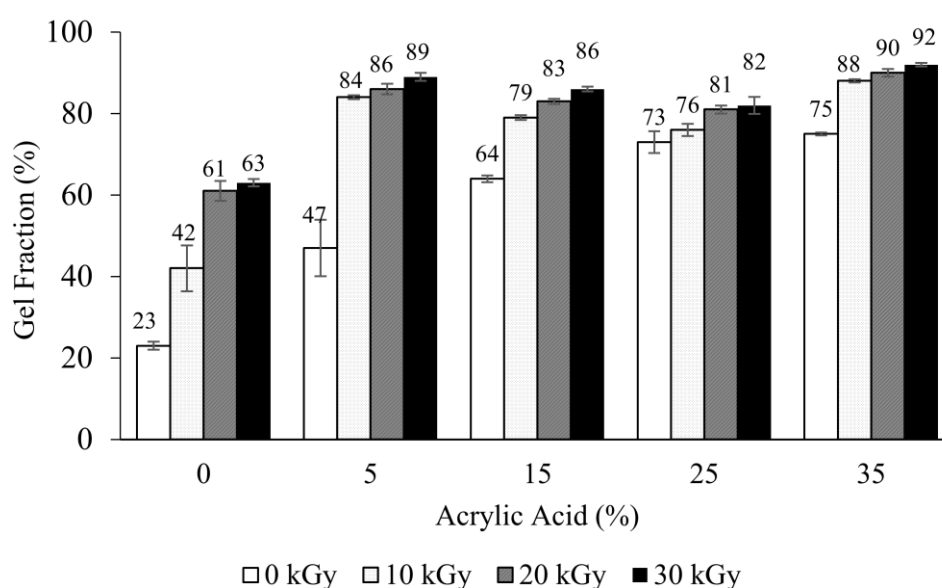


Figure 1 The effect of the acrylic acid and gamma-ray dose to the gel fraction

of the swelling value.

Gel fraction rise with the increase of gamma-ray dose as shown in Figure 1. Gamma-ray crosslinking used high energy to change the structure of the material into radicals. The higher the gamma irradiation dose that was exposed to the material made the more radicals formed. The greater radicals caused the tighter bonds formed, so the hydrogel water solubility decreased and the value of the gel fraction increases. The gel fraction of the samples in this study had a gel fraction value of 22% in the control treatment, and the highest was 92.44% in the hydrogel with 35% acrylic acid and 30 kGy. Hydrogels with 25% acrylic acid and 10 kGy, which was the treatment with the highest swelling, had a gel fraction of 75.98%. The hydrogel gel fraction value of the gamma irradiation method in the study was almost the same as the results of several similar studies which ranged from 70 to 98% [14-20].

3.3 Equilibrium Degree of Swelling (EDS)

The EDS of hydrogels measured based on the percentage of water absorbed. According to Figure 2, EDS increased with an increasing of acrylic acid percentage. Acrylic acid that has been neutralized can ionize to form carboxylic anions which have the ability to bind water using hydrogen bonds [13]. The increase of acrylic acid caused an increase in the number of carboxylic anions which are hydrophilic in the hydrogel polymer chain so the amount of water that can be absorbed also increases. According to Erizal et al. [1], when the hydrogel was immersed in water, an interaction occurs between water molecules and anions in the hydrogel. This causes repulsion between the polymer

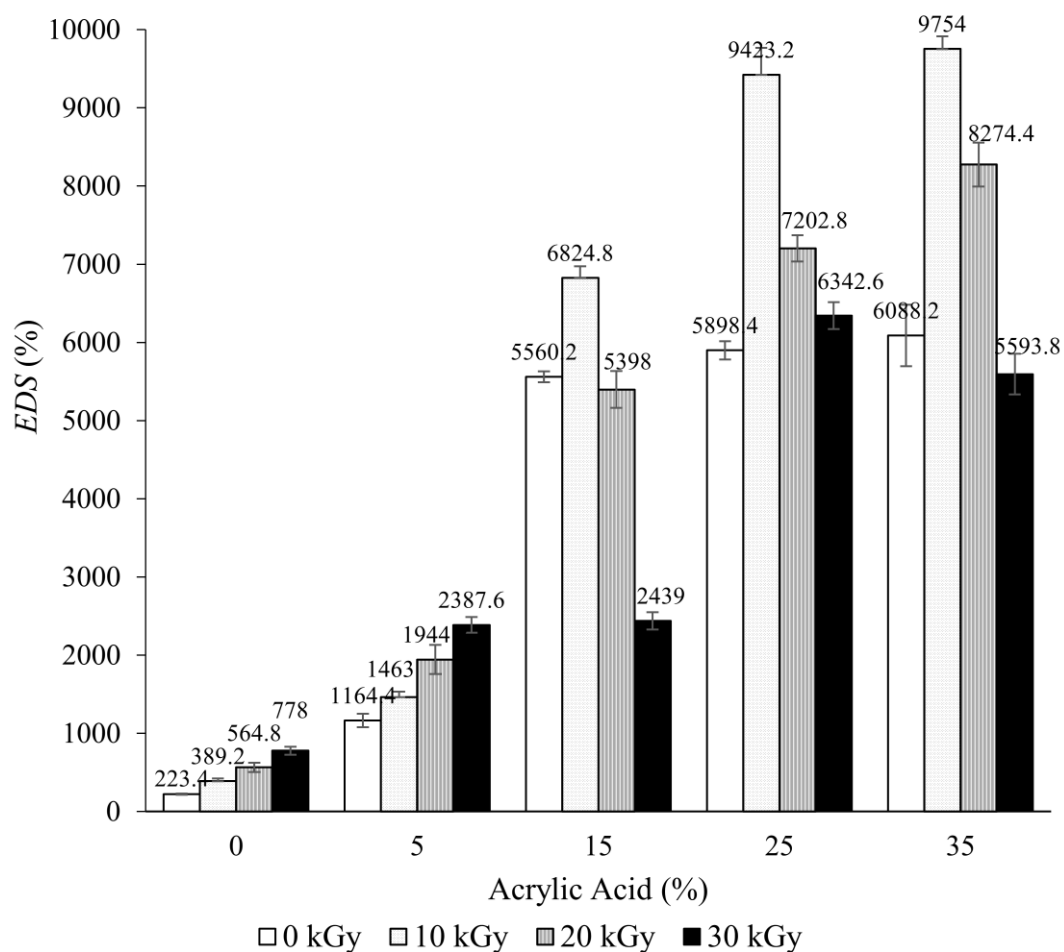


Figure 2 The effect of the acrylic acid and gamma-ray dose to the EDS

chains which forces the polymer chains to expand so the EDS increases. Hydrogels of Gamma-ray at 10 kGy showed a decrease in swelling when acrylic acid increased from 25% to 35%. This shows that the percentage of acrylic acid used in the hydrogel process has an optimum point. The addition of acrylic acid that exceeds the optimum point causes an insignificant increase in the swelling value or a decrease in the swelling value.

As represented in Figure 2 the rise of gamma-ray dose increase EDS in the control (without acrylic acid) and the 5% of acrylic acid. Besides that, in the sample with 15, 25, and 35% acrylic acid, EDS of 10 kGy hydrogels have a significant increase than the control treatment that did not get irradiated. However, when the gamma-ray dose increase to 20 and 30 kGy, the EDS decrease. The irradiation dose required to produce cross-linking on the material was highly dependent on the reactivity of the material to the crosslinking reaction using gamma-ray. Materials with high reactivity to the cross-linking reaction required the lower dose to produce a three-dimensional structure [21]. Acrylic acid was material that reactive to cross-linking reactions because it has double bonds with unsaturated acids and carboxylic acids or aliphatic esters [18]. This caused acrylic acid could react easily to the biopolymer matrix to form a crosslinked structure. The higher percentage of acrylic acid, the number of double bonds that will be bonded to the biopolymer matrix was greater, so the crosslinking process required lower gamma-ray dose. The higher of gamma-ray dose formed tighter cross-linked, so the polymer chain being difficult to stretch and the water penetration restricted [22]. The EDS in this study in the range of 200 to 9700%. Hydrogel with 25% acrylic acid and 10 kGy gamma-ray was the most optimal in this study with EDS of 9423%. Although the EDS of this treatment lower than the hydrogel with 35% acrylic acid which had EDS of 9754%, the results of Duncan's multiple interval test showed that the EDS difference was not significant between this two treatments. The EDS in this study close to the EDS of a similar study based on starch and acrylic acid - acrylamide which has an EDS between 10000 to 25000% [14].

3.4 Swelling Kinetics and Diffusion Coefficient

Figures 3a and b shows the swelling kinetics of hydrogel in water and 0.04 M urea solution. The process of water and urea solution penetrating into the polymer chain is due to osmotic pressure arising from differences ions concentration between the system in the inner membrane of hydrogel and the environment [1]. The EDS in water (Figure 3a) higher than EDS in urea solution (Figure 3b). However, EDS at equilibrium (day 6) did not show a significant difference in values. The urea solution is a non-polar which when dissolved in water did not produce ions which cause the osmotic pressure to decrease [4]. The difference in swelling when immersed for 5 hours is due to the difference in ion concentration which decreases when there is urea in the solution. This causes the osmotic pressure when the hydrogel is immersed in urea solution is lower than when immersed in water which results in a lower diffusion rate [4].

The diffusion ability of polymer chains can be categorized using Fick's Law of based on the

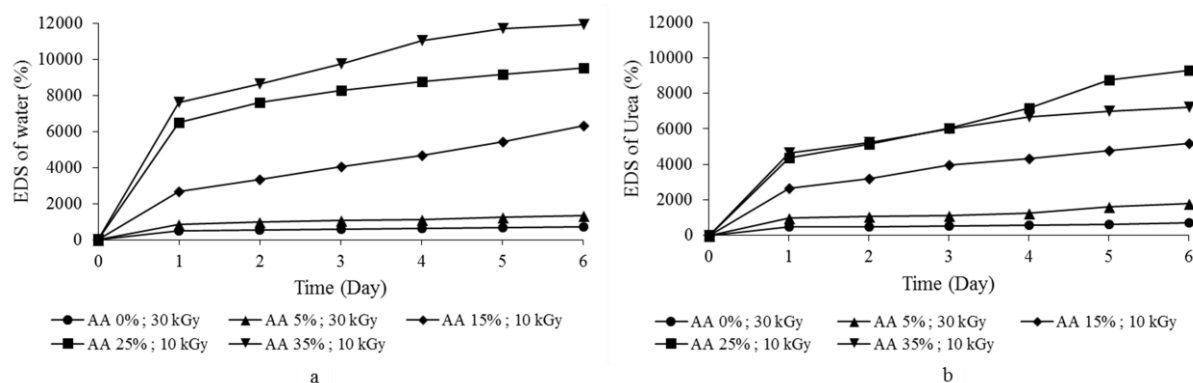


Figure 3 EDS interaction with immersion time in (a) water and (b) 0.04 M urea solution

diffusion exponential value (n). The first category (Fickian Diffusion) occurs when the diffusion rate of water lower than the relaxation rate of the polymer chain. This condition occurs when $n \leq 0.5$. The second category is the condition that occurs when water diffusion is very fast compared to relaxation so the diffusion rate is influenced by the ability of the hydrogel to stretch. The hydrogels categorized in this type when the diffusion exponent (n) ≥ 1 . The third category is the non-Fickian or anomalous diffusion. Anomalous diffusion occurs when the diffusion exponential value (n) is in the range 0.5 to 1 ($1 > n > 0.5$). This condition occurs when the diffusion rate is equal to the rate of relaxation of the polymer chain [4-16]. The swelling exponential (n) in Table 2 shows the hydrogel with 5, 15, 25, and 35% acrylic acid were categorized as non-Fickian or anomalous diffusion ($1 > n > 0.5$). However, when immersed in urea solution, 5% AA hydrogel was included in the Fickian diffusion category ($n \leq 0.5$). The controls treatment (AA 0%) included Fickian diffusion when immersed in water and urea solution. Therefore, the hydrogel of the gamma irradiation method without acrylic acid (control) and 5% acrylic acid had a lower diffusion rate than the relaxation rate of the polymer chain.

The other parameter used in the swelling kinetics analysis was the swelling constant. The swelling constant showed the characteristics of the polymer hydrogel chain during the diffusion process until the optimum swelling [16]. As shown in Table 2, the swelling constant increases with the increase of acrylic acid percentage. This resulted in accordance with the EDS which rise with the incline of acrylic acid percentage. The negative sign in the K value showed the diffusion process that flows from a higher water concentration to the lower concentration [23]. The swelling constant in this study is almost the same as the hydrogel based on acrylamide and methacrylic acid which has a k value from -2.1 to -0.6 [24].

The diffusion coefficient was analyzed using the second-order Fick's law equation obtained from the slope of the swelling fraction (F) to $t_1 / 2$ curve. From Table 2 it can be seen that the diffusion coefficient increase with the increasing of acrylic acid percentage. The diffusion coefficient showed the flow rate of the solution into and out of the polymer system. The greater diffusion coefficient value, the flow rate of water and urea solution into the polymer chain better. Based on the analysis using diffusion exponential values, almost all hydrogels in this study had anomalous or non-Fickian diffusion types. This type had an equal diffusion rate to the relaxation rate of the polymer chain. So that indicated the hydrogels with a high diffusion coefficient had more elastic.

Table 2 Parameters of swelling kinetics

Acrylic Acid (%)	Gamma-ray dose (kGy)	N_{water}	n_{urea}	K_{water} (h^{-1})	K_{urea} (h^{-1})	D_{water} (m^2h^{-1})	D_{urea} (m^2h^{-1})
0	30	0.2742	0.1928	-0.927	-0.836	0.0752	0.0701
5	30	0.5147	0.4534	-1.424	-1.960	0.1792	0.1276
15	10	0.7111	0.6744	-2.196	-2.272	0.1874	0.1700
25	10	0.8471	0.6568	-2.211	-2.212	0.2393	0.1766
35	10	0.7017	0.6535	-2.350	-2.149	0.1709	0.1842

3.5 Mechanical Properties

The testing of mechanical properties was carried out using a texture analyzer instrument. The results obtained from this test was hardness, strain and stress. The modulus of elasticity was determined by the ratio of stress to strain. Furthermore, the crosslink density was determined using the elasticity theory of rubber [25]. Hardness is the amount of energy required to deform the sample [26]. The hardness was calculated based on the maximum energy loaded to the sample in the first compression cycle. The results in Table 3 shows hardness decreased from 10.95 mJ to 8.83 mJ when the acrylic acid increased from 5% to 25%. This increase leads by the increase in hydrogel EDS, as seen in the previous section. The increases of EDS caused the stretching of polymer chain length, so the pore size in the polymer chain structure larger. Therefore, the amount of energy required to deform the sample

lower as the swelling gets bigger. However, when the percentage of acrylic acid was 35%, the hardness increased dramatically to 24.89 mJ. Acrylic acid that has been neutralized had carboxylic anions which can form bonds with water, but AA is also sensitive to crosslinking reactions. The nature of acrylic acid which is sensitive to cross-linking reactions causes the addition of acrylic acid by 35% to produce a polymer structure with an increasingly dense number of cross-links. The tightness of this cross-linking will cause the hydrogel's strength to withstand loads to be better [26].

The second parameter observed in mechanical properties analysis was stress. Stress obtained from the force required to compress the hydrogels [27]. Stress was determined from the ratio between the maximum force loaded to the sample and the surface area of the sample being pressed. Stress had proportional value to the hardness. When the hardness was higher, the force required to compress the sample larger [28]. The stress of hydrogels was inversely proportional to strain. Strain shows the response of the sample to given stress [29]. Table 3 shows strain increased when the percentage of acrylic acid increases from 5 to 25%, but strain decreased when the percentage of acrylic acid becomes 35%. Based on the results of the diffusion coefficient analysis contained in the previous section, hydrogels with AA 25% have a greater diffusion coefficient than hydrogels with 35% AA. The greater diffusion coefficient causes a higher diffusion rate. Stress and strain values were used to determine the modulus of elasticity. The modulus of elasticity obtained in Table 3 is directly proportional to the hardness of the hydrogel. The modulus of elasticity of the hydrogel shown in Table 3 is higher than similar studies which have an elastic modulus of 4.53 kPa [30]. The modulus of elasticity was used to determine the number of active bonds per unit volume (crosslink density, N) based on the concept of rubber elasticity [25-31]. As can be seen in Table 3 crosslink density proportional to the hardness. These results were in accordance with the theory which states that a tighter cross-linking will produce a hydrogel with better mechanical properties [28].

Table 3 The parameters of mechanical properties

Acrylic Acid (%)	Gamma-ray dose (kGy)	Hardness (mJ)	Stress (kPa)	Strain (mm^{-1})	Modulus of elasticity (kPa)	N ($\times 10^{23} \text{ m}^{-3}$)
5	30	10.95	4.14	0.41	10.10	8.13
25	10	8.53	3.99	0.57	7.01	5.64
35	10	24.89	9.62	0.38	25.29	20.36

3.6 Morphology Analysis

Morphological analysis showed the surface structure of hydrogels. The analysis was carried out based on the image produced by a scanning electron microscope (SEM) at magnifications of 500 and 2000 times. The SEM results of the control treatment are shown in Figure 4, while Figure 5 shows the SEM image of the 25 % acrylic acid and 10 kGy hydrogel at magnifications of 500 and 2000 times. These results indicated that the addition of acrylic acid as a monomer and cross-linking of gamma irradiation resulted in pores in the hydrogel polymer chain. The pores contained in the hydrogel function as a pathway for the exit and entry of water into the polymer chain. The more pores the hydrogel has, the better the hydrogel's ability to absorb water [5]. In addition, according to Erizal [14], the pores contained in the hydrogel are the active side of the polymer which consists of hydrophilic groups, so that it can form bonds with water.

The SEM image results shown in Figure 5 also show a cross-linked structure. This cross-linked structure is produced by the bonds between the high-energy radical chains released by gamma irradiation [1]. This crosslinked polymer structure functions as a water absorber and a hydrogel structure strengthening so that it is not easily dissolved in water [5]. This is consistent with the results of the gel fraction analysis which showed an increase in the value of the gel fraction when the irradiation dose was increased. The resulting cross link also functions as an absorbent that holds water

in the polymer chain. This is supported by the results of the swelling hydrogel of the gamma irradiation method without 5% acrylic acid and acrylic acid which increases when the irradiation dose increases.

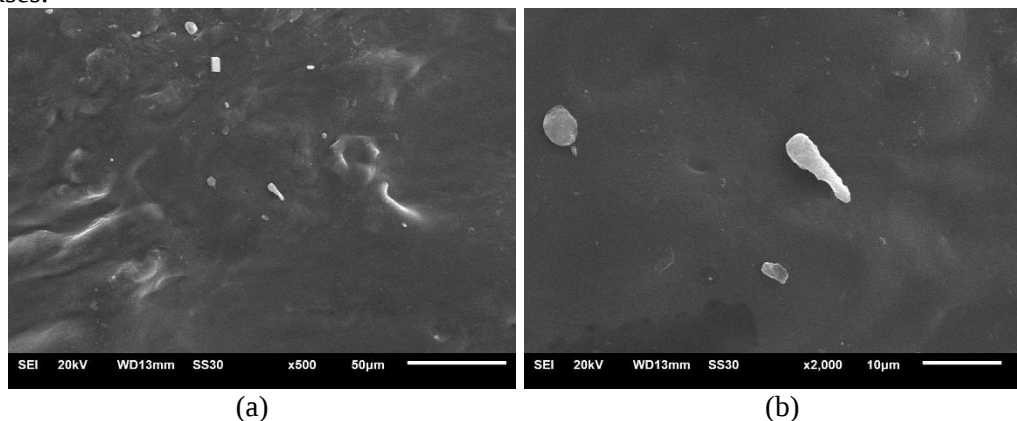


Figure 4 Morphology of control sample in the magnification of (a) 500 and (b) 2000 times

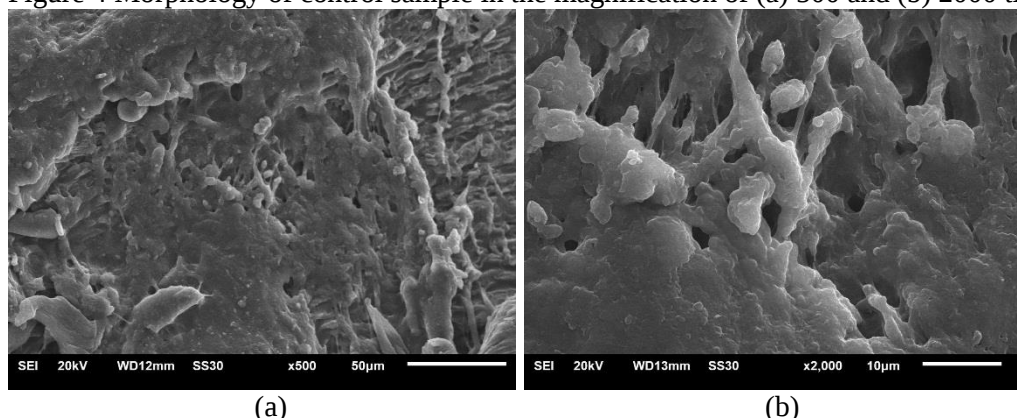


Figure 5 Morphology of 25% acrylic acid and 10 kGy in the magnification of (a) 500 and (b) 2000 times

4 Conclusions

Onggok based superabsorbent polymer (hydrogel) grafted with acrylic acid and acrylamide has been synthesized using the gamma-ray crosslinking method. The addition of gamma-ray and acrylic acid give a significant effect on swelling, gel fraction, and mechanical properties at the 5% significance level. The percentage of 25% acrylic acid and the irradiation dose of 10 kGy is the optimum conditions in this study. The optimum swelling obtained was 9423% with a gel fraction of 75.98%. The mechanical properties obtained were also optimum with a hardness value of 8.53 mJ, a modulus of elasticity of 7.01 kPa, and a crosslink density of 5.64×10^{23} active bond volume unity.

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