

# Unique Stimuli Responsive Characteristics of Electron Beam Synthesized Bacterial Cellulose/Acrylic Acid Composite

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**ABSTRACT:** The formation and swelling behavior of bacterial cellulose/acrylic acid hydrogel prepared from aqueous mixture consists of 20 : 80 (v/v) acrylic acid (AAc) and 1% bacterial cellulose dispersion under accelerated electron beam was investigated. Gel fraction of hydrogel increased with the increasing dose suggesting a denser composite at 50 kGy compared to 35 kGy. SEM photomicrographs revealed a homogenous pores distribution at higher dose with pore sizes ranging from 1 to 5  $\mu\text{m}$ . Hydrogel synthesized at lower dose of electron beam exhibited higher swelling ability and the degree of swelling increased as the pH of surrounding medium

increased and it reached the optimum swelling at pH 7. While swelling of the hydrogel decreased with the increasing ionic strength of solution, swelling at different temperatures ranging from 25 to 50°C revealed a unique character where the hydrogel shrunk at 37°C. Moreover, hydrogel synthesized at higher dose exhibited a higher degree of swelling in methanol with respect to water. © 2010 Wiley Periodicals, Inc. *J Appl Polym Sci* 116: 2920–2929, 2010

**Key words:** hydrogels; swelling behavior; electron beam irradiation

## INTRODUCTION

Hydrogels are hydrophilic polymer networks that can retain at least 20% of water than its dried weight. It can be formed by reacting one or more monomers through chemical or irradiative techniques and may be chemically stable or disintegrated over time.<sup>1</sup>

Permanent hydrogels structures formed by covalent bonding in between polymer chains through irradiative technique have several advantages. Firstly, reactions may occur at room temperature without the necessity to introduce initiators or additives to initiate reaction that might leave undesirable residues. Secondly, the penetration ability of the radiation would also mean, a readily finished prod-

uct with proper sealed packed polymer could be cross linked at the same time providing a terminally sterilized product for medical uses.<sup>2</sup>

Hydrogel can be grafted onto surfaces and pores of materials like silicone wafers to achieve desirable properties.<sup>3,4</sup> On the other hand, hydrogels can be reinforced by grafting it onto certain type of fabrics and enhanced the mechanical strength of the hydrogels,<sup>5,6</sup> while crosslinking natural polymer such as polysaccharides will produce biodegradable devices.<sup>7</sup>

Swelling behavior and biocompatibility properties of hydrogels makes it an important device in biomedical and pharmaceuticals applications. Combination of polymers (Polyvinyl alcohol/fibers, Polyvinyl pyrrolidone/AAc, Polyvinyl pyrrolidone/CMC, Polyvinyl pyrrolidone/Poly Acrylic Acid, Polyvinyl pyrrolidone/Citric Acid)<sup>8–11</sup> can be fabricated into composite which could match the mechanical properties of soft tissues especially in the tissues replacement therapy.<sup>12</sup>

An enormous study has immersed in the past few decades where intelligent hydrogels were being synthesized and characterized. These hydrogels have the ability to respond towards changes in its environmental factors such as pH, temperature and ionic strength. Ionic hydrogels will have their swelling

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degree affected by changes of pH in surrounding medium. Anionic hydrogels swells even more when the external pH is higher than the pKa of the ionisable group, whereas in cationic hydrogels, the degree of swelling increases when the external pH is lower than the pKb of the ionisable group.<sup>13,14</sup> Acrylic acid (AA) and metacrylic acid (MAA)<sup>13,15</sup> are the most commonly used monomer to fabricate anionic hydrogels, while 2-dimethylamino ethyl methacrylate (DMAEMA)<sup>16</sup> has been used in cationic hydrogel formation. Their major applications are in delivering relatively large molecules such as peptide and proteins through oral administration.<sup>15</sup> As reported previously, repeated contraction and expansion of hydrogels has been used as a mode of delivery for insulin and the release of this hormone is due to the changes in the blood glucose level. An *in vivo* study into diabetic induced rats using such hydrogel has demonstrated promising results as the device gave a significant reduction in their hyperglycaemic levels for about 2 months.<sup>17</sup>

Another type of intelligent hydrogel is the one that response towards temperature changes. It is well known that poly(*N*-isopropylacrylamide) (PNIPAAm) hydrogels possess lower critical solution temperature (LCST) at 34°C. This is where the network expanded with large mesh size enabling diffusion process to occur, whereas above its LCST the pores shrunk, deswelled and restructured into a smaller mesh size network. Although the LCST could be modified by adding other monomers into the system, this hydrogels have little applications in biomedical area where a few pathological condition manifest as a drop in body temperature.<sup>14,18</sup>

In the effort of manipulating the properties of hydrogels, a lot of attempts have been carried out to produce a rather more complex composite. Several studies have been reported to have used polysaccharides with polyvinyl alcohol (PVA)<sup>7</sup> to improve the mechanical strength and thermal properties of the composite, while others have used cellulose fibers such as bacterial cellulose to increase the stiffness of the composite.<sup>12</sup>

In this study, bacterial cellulose, a natural and renewable biopolymer that is known for its biocompatibility was used as a major constituent of the hydrogels composite with acrylic acid as a crosslinking monomer. Under the bombardment of accelerated electron beam, a more cost effective hydrogels were formed and characterized.

## EXPERIMENTAL

### Materials

Acrylic acid (AAc) from Sigma Aldrich, sodium chloride, methanol, disodium hydrogen phosphate,

potassium dihydrogen phosphate, orthophosphoric acid, potassium hydroxide and trisodium phosphate (all from R&M Chemical) were used without further purification.

Nata de coco as the source of bacterial cellulose was obtained from local food industry, purified and characterized as described in British Pharmacopoeia prior using.

Distilled water was used to prepare aqueous solution and dispersion while plastic trays with dimension 12 cm × 12 cm × 1 cm was used as hydrogel mould.

### Preparation of bacterial cellulose/AAc hydrogels

Purified bacterial cellulose was dried and ground to powder form with particle size ranging from 20 to 200 μm. Bacterial cellulose powder was then dispersed in distilled water to produce 1% (w/v) dispersion. Acrylic acid was added to this dispersion to make a 1 : 4 acrylic acid:bacterial cellulose mixture. The mixture was then poured into a plastic tray and subjected to electron beam irradiation of 35 and 50 kGy in air, using an accelerator facility at Malaysian Nuclear Agency.

### Determination of gel fraction

Freshly prepared hydrogels was cut into pieces of size 2 cm × 2 cm and dried to a constant weight in a convection oven at 60°C. Dried hydrogels were then extracted at room temperature in distilled water for 7 days. The extraction process was done to remove any sol that may be present in the hydrogel. The extracted hydrogels were then dried again in the oven at 60°C until a constant weight is achieved. The percent (%) of gel fraction was calculated using the equation as below<sup>19</sup>:

$$\frac{G_d}{G_o} \times 100$$

where  $G_o$  is the dried initial weight before extraction and  $G_d$  is constant dried weight of hydrogels after extraction.

### Infrared spectroscopic analysis

Infrared spectra of the hydrogel were recorded using FTIR Spectra 2000 (Perkin-Elmer) at room temperature. Samples were prepared as thin films and analyzed over the range of 550–4000 cm<sup>-1</sup> using Diamond ATR.

### Morphological analysis

For morphological analysis, dried extracted hydrogels were swollen in distilled water at room

temperature (25°C) for 48 h to reach equilibrium swelling state. Hydrogels were then cut into pieces of 0.5 cm × 0.5 cm × 0.5 cm to be placed in samples chamber. Samples were then observed in its hydrated state under ESEM mode for wet samples. The bound water in hydrogel sample was prevented from vaporizing by cooling the sample's temperature down to 5°C to preserve the open microporous structures before the image was taken. The procedures were completed in less than 30 s to avoid the hydrogel from drying out due to over exposure to the electron bombardment. Scanning electron micrographs were taken using Quanta200F instrument (FEI, USA) with magnification ranging from 400 to 8000×.

### Swelling behavior of hydrogels

The swelling behavior of hydrogels was studied under various conditions such as different solvents, pHs, ionic strengths and temperatures. A known weight of dried extracted hydrogels ( $G_d$ ) was immersed in distilled water and methanol for 48 h at room temperature (25°C). Swollen samples were weighed ( $G_s$ ) at every hour where excess water was pad dried. The percent of swelling were calculated by using the following equation:

$$\text{Swelling (\%)} = \frac{G_s - G_d}{G_d} \times 100$$

The effect of external stimuli towards swelling behavior was evaluated by using phosphate buffer medium of pH 2, 5, 7, and 10 with different concentration of sodium chloride and temperatures ranging from 25 to 50°C. The swelling percentages of the hydrogel under these conditions were also calculated as mentioned earlier.

### Differential scanning calorimetry

Differential scanning calorimetry was used to study the heat capacity of the hydrogels. The analysis was done using Metler Toledo DSC STAR<sup>e</sup> and hydrogels was swelled in distilled water at 5°C.<sup>20</sup> The hydrated samples were weighed and hermetically sealed and scan was done at temperature range of 0°C and 50°C with constant heating rate of 2°C/min.

## RESULTS AND DISCUSSION

### Gel fraction of crosslinked bacterial cellulose/AAC composite

Findings from the experiment showed that 35 kGy of electron beam was found to be the lowest dose required to solidified a 0.5 cm thickness of gel with-

out any temperature elevation that might cause bubbling within the composite samples. The percentage of gel fraction for hydrogel synthesized at 35 kGy and 50 kGy was found to be 60% ± 3.44% and 77% ± 1.17%, respectively. Even though the initial formulation for both selected dose of electron beam were the same, a higher dose of electron beam yields a higher gel fraction. Electron irradiation will lead to radiolysis of water which later caused the formation of hydrated electrons, hydroxyl and hydrogen radicals as reactive species that probably create some active sites on AAC to be crosslinked.<sup>21</sup> In this case, a higher dose will produce more reactive species which would increase the degree of crosslinks.

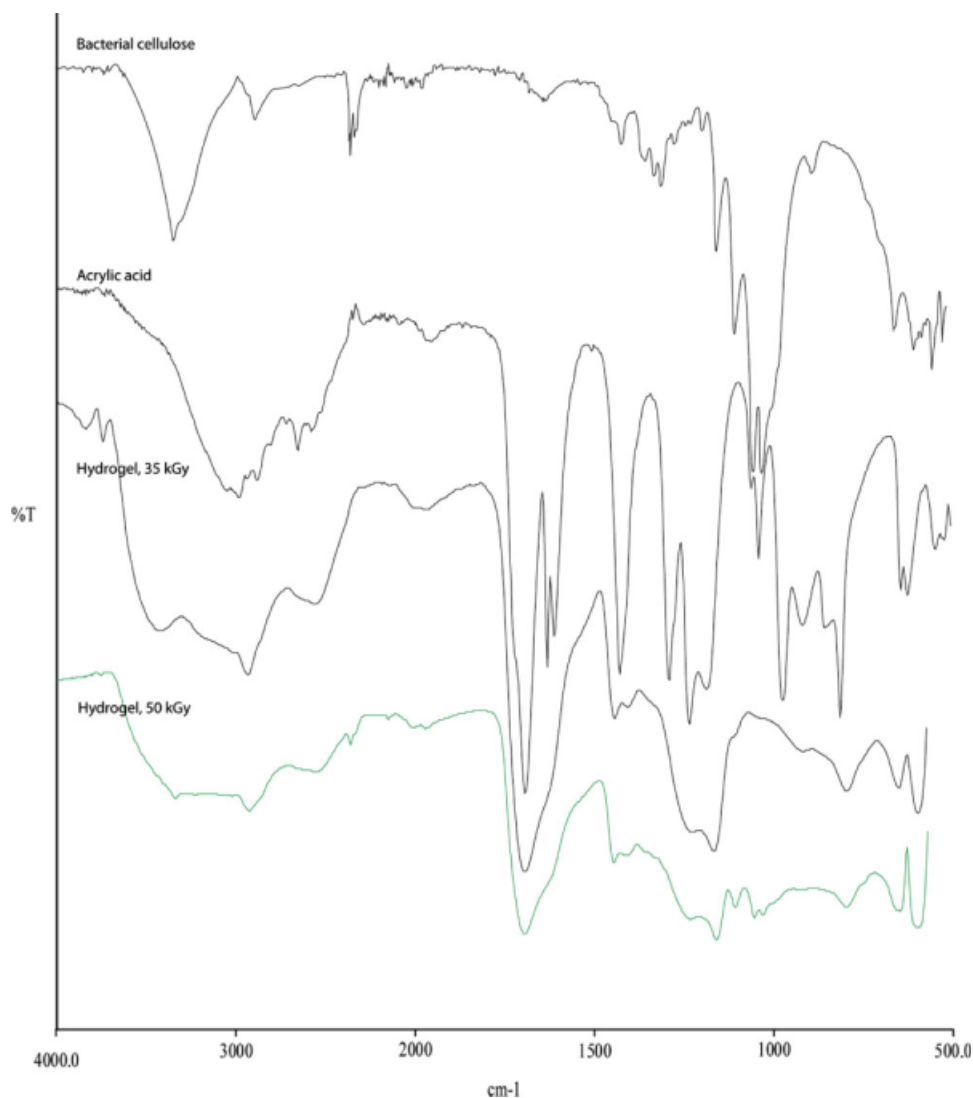
Although the hydrogel consists of a large amount of water, it was found that the selected dose of electron beam would still be able to crosslink the acrylic acid monomer and the bacterial cellulose in the mixture into a solidified form. Previous studies have reported that the gelation dose for poly (acrylic acid) alone was at 50 kGy<sup>22</sup> but from this study it was found that for this mixture, the complete crosslink was dependent on the size and surface of hydrogel exposed to the radiation.

When the dried hydrogel was immersed in distilled water the network chains became solvated and the hydrogen forces between the polymer and the solvent causes the absorption of the solvent molecules.<sup>22</sup> However, the crosslinking prevented the structure from dissolving and only allows the expansion of the hydrogel network.

### Infrared spectroscopic analysis

The IR spectra of bacterial cellulose, acrylic acid and bacterial cellulose/AAC hydrogels synthesized at 35 and 50 kGy are shown in Figure 1. It shows that all peaks for both hydrogels spectra from two different doses are the same but with different intensity suggesting that the hydrogel formation for both doses involves the same interaction between the fibers and the AAC polymer chains. The bacterial cellulose IR spectrum has peaks at 1040 cm<sup>-1</sup> due to C—O stretching, 1163 cm<sup>-1</sup> due to C—O—C stretching of the ether linkage [1,4-β-D-glucoside] of cellulose, 2926 cm<sup>-1</sup> due to C—H stretching and 3440 cm<sup>-1</sup> due to O—H stretching of intermolecular hydrogen bonding.<sup>23,24</sup>

AAC spectra showed distinctive peaks at 1600 cm<sup>-1</sup> due to C=C stretching, 1700 cm<sup>-1</sup> due to C=O stretching and broad peak at 3040–2340 cm<sup>-1</sup> due to O—H stretching.<sup>25</sup> The present of C=C stretching in the hydrogel spectra as a shoulder suggesting the polymerization of acrylic acid to polyacrylic acid. As reported by Scherzer et al. and Knolle and Mehnert<sup>26,27</sup> the irradiation of acrylates and methacrylates with electrons leads to curing of the



**Figure 1** FTIR spectra of bacterial cellulose/AAc hydrogels synthesis at 35 and 50 kGy of electron beam. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]

polymer by formation of radicals that start the polymerization and crosslinking. While the absence of C—O stretching in the hydrogel indicated the grafting of the acrylic acid on to the bacterial cellulose fibers. During the irradiation process, the water molecules were turned into reactive species such as hydrated electrons, hydroxyl radicals and hydrogen atoms.<sup>28</sup> These species then created active sites on the AAc and bacterial cellulose to form grafting.<sup>21</sup> The C=O stretching at around  $1700\text{ cm}^{-1}$  in the hydrogel spectra was obviously caused by the presence of carboxylic group from AAc and a very broad peak found at the region of  $2500\text{--}3500\text{ cm}^{-1}$  indicating the presence of extensive hydrogen bonding. This may result from the increasing hydrogen bonding of bacterial cellulose towards the AAc polymerization where the bonding was formed between the carboxylic group of AAc and the nonsubstituted

hydroxyl group of the cellulose which could also explain the formation of the hydrogel.<sup>21</sup>

The predictive reaction mechanism of the above process is illustrated in Figure 2. When the radiolysis of water occurs, all reactive transients from water are expected to react with the monomers. The formation of the radicals in the eqs. (1) and (2) are very likely to occur and may initiate the polymerization of the acrylic acids.<sup>28</sup> The formation of mid-chain radicals (crosslink sites) by intramolecular 1,5 H-shift was reported and found in electron pulse radiolysis (EPR) investigation of acrylates, polyacrylic acid and 2,4-dimethylglutaric acid.<sup>29</sup> This would be the possible way of crosslinking polyacrylic acid during the irradiation process. It is well known that the OH and H radicals are capable of extracting hydrogen atoms from polymers when the addition to double bond is not possible.<sup>28</sup> In this study, the H radicals

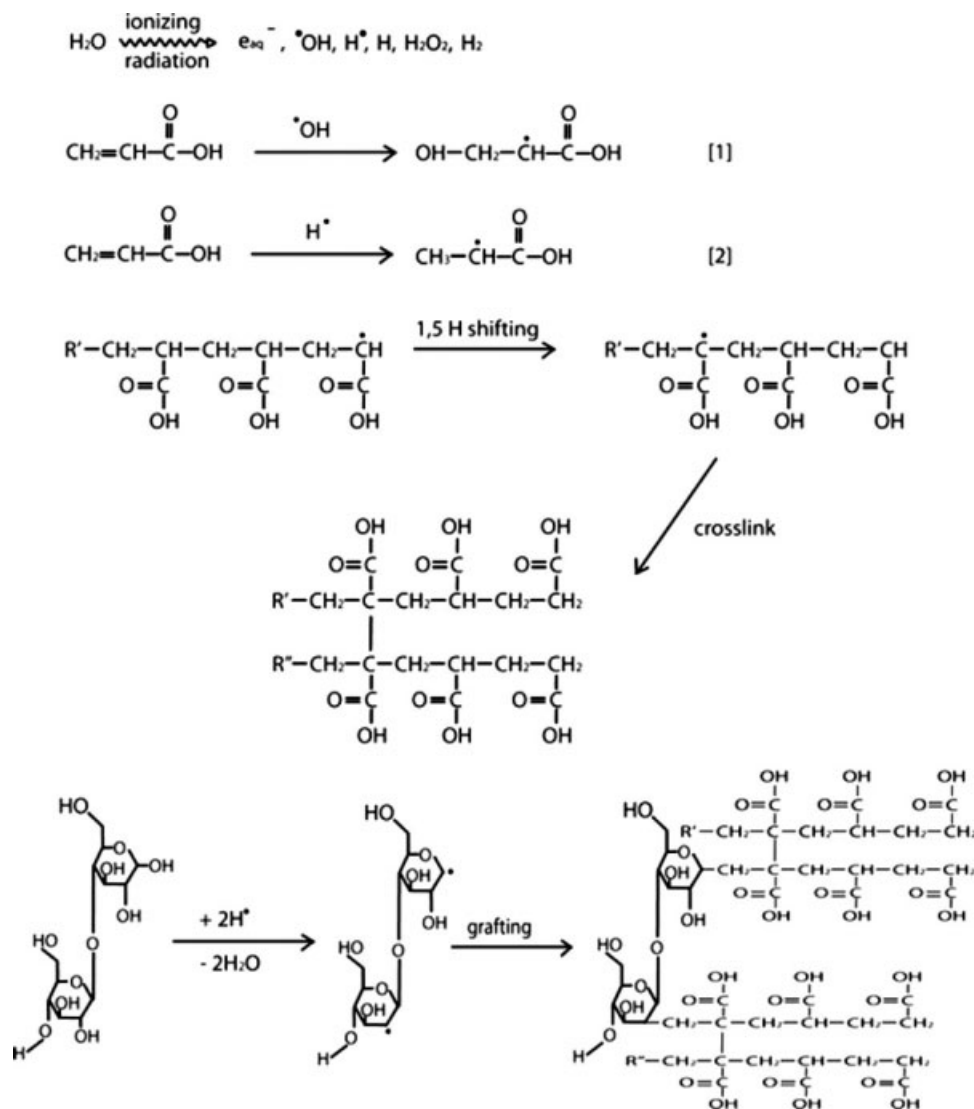


Figure 2 Predicted reaction mechanism of hydrogels synthesis.

would probably extract the hydroxyl group from the cellulose fibers leading to the radical sites for grafting which explain the absent of peak at  $1040 \text{ cm}^{-1}$  of C—O stretching in the hydrogel spectra.

### Morphological analysis

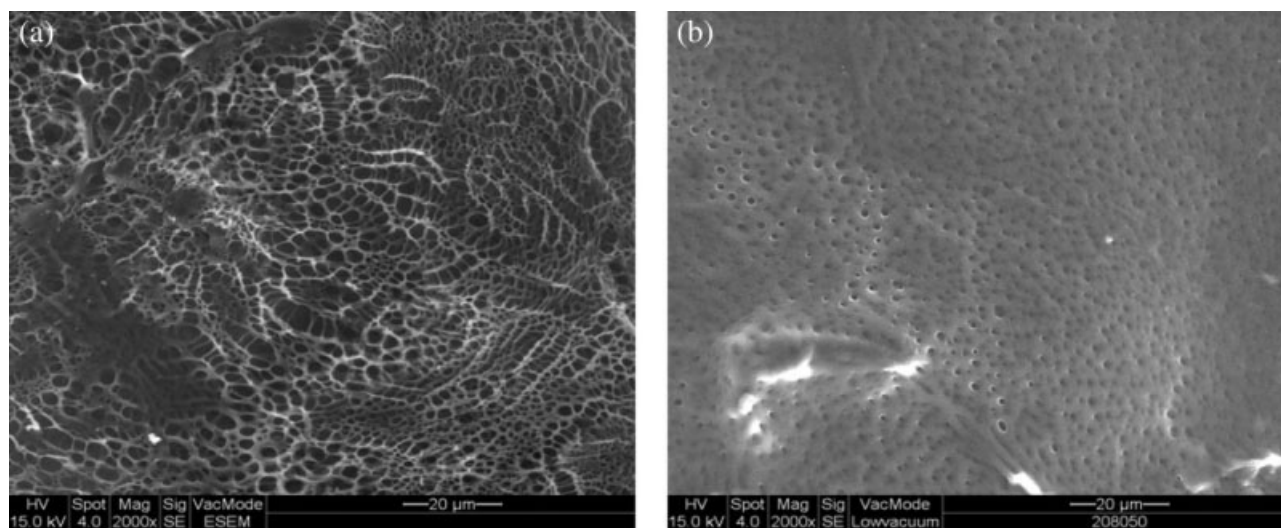
Scanning electron micrographs of two hydrogel networks synthesized at different doses are shown in Figure 3(a,b). At  $2000\times$  magnification, 35 kGy hydrogels revealed pores ranging from as small as 2–20  $\mu\text{m}$  while hydrogel of 50 kGy dose showed smaller and fine pores with a homogenous distribution with the size ranging between 1 and 5  $\mu\text{m}$ . The morphological condition is highly influenced by the degree of crosslinking of the hydrogel. At higher dose of electron beam, more inter polymeric network are formed between polyacrylic acid and grafted bacterial cellulose thus making the dense network to

smaller pores size.<sup>2</sup> These two morphological observations are expected to influence the swelling profiles of hydrogels as in terms of water and solutes uptake.<sup>2,14,30</sup>

As reported previously a sponge like structure with interconnectivity pores will allow diffusion to occur at every direction,<sup>14</sup> resulting in higher swelling capability as exhibited by 35 kGy hydrogel. This highly porous structure would also facilitate water molecules to diffuse in the hydrogel through the pores leading to increase of water uptake.<sup>31</sup>

### Swelling behavior of hydrogels

Despite crosslink density and pores size of network, the swelling behavior of the hydrogel was greatly influenced by the nature and the presence of other components in the swelling medium.<sup>2</sup> The effect of different solvents towards swelling kinetics of the



**Figure 3** Scanning electron micrographs of hydrogels synthesis at 35 kGy at 2000 $\times$  magnification. Scanning electron micrographs of hydrogels synthesis at 50 kGy at 2000 $\times$  magnification.

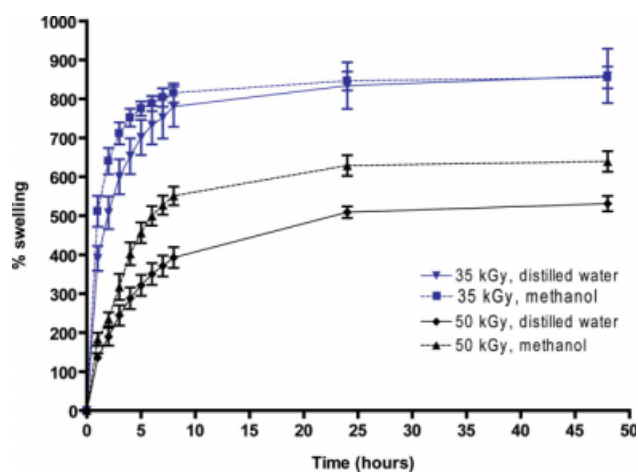
hydrogel is shown in Figure 4. It is clearly revealed that both hydrogel synthesized at 35 and 50 kGy have higher swelling rate in methanol than in distilled water. It is also observed that hydrogel prepared at 35 kGy has higher swelling percentage rate as compared to the 50 kGy hydrogel with respect to the swelling mediums. This may be concluded from the lower crosslink density of the hydrogel which formed a porous structure and thus allowed the water molecules to diffuse easier<sup>31</sup> hence faster response time.<sup>32</sup> Hydrogel synthesized at higher dose always have lower swelling ratio probably due to the higher crosslink density which resist the swelling of the matrix.<sup>32</sup> Previous study has also reported changes in morphology are associated with significant changes in the swelling behavior of the hydrogel.<sup>14</sup> This phenomenon agrees well with the morphological finding as discussed earlier.

Besides this character, both hydrogel were found to have a greater swelling capability in methanol in comparison to water. This is probably due to the nature of the hydrogel. It is suggested that the hydrogel are formed through a grafting process of the AAC on to the cellulose fibers and by hydrogen bonding between polyacrylic acid long chains polymer. The strong hydrogen bonding makes hydrophilic group less available for the hydrogen bonding with water molecules<sup>2</sup> making the network structure to be less polar. A higher dose of electron beam produced more radical species that will create more active sites<sup>21</sup> to allow grafting process to occur and lead to a higher degree of crosslink<sup>32</sup> and even less polar hydrogel. This relatively hydrophobic hydrogel exhibited higher swelling rate in methanol as compared to distilled water at room temperature

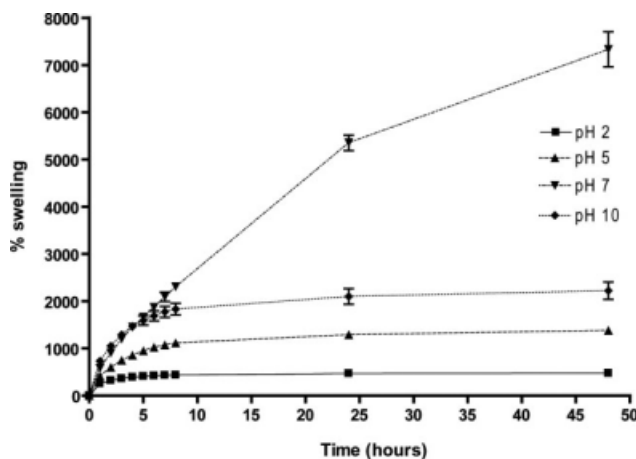
and the effect is more significant for hydrogel synthesized at 50 kGy.

#### Swelling behavior towards changes in pH

The swelling kinetics of bacterial cellulose/AAC hydrogel prepared at 35 and 50 kGy are presented as in Figures 5 and 6. It has been found that both hydrogel exhibited very low swelling rate in the media of pH 2 and 5. The swelling rate slightly increased within 2 to 8 h before it reached swelling equilibrium after 24 h. As the pH increased, the swelling rate of the hydrogel greatly increased with the highest swelling rate was at pH 7. The results



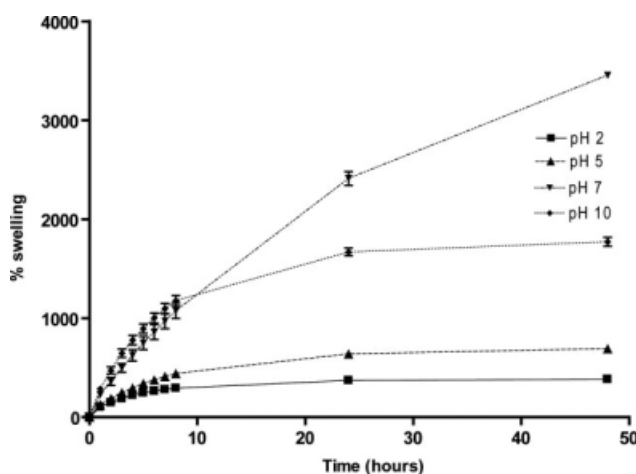
**Figure 4** Swelling of hydrogels synthesis at 35 and 50 kGy in methanol and distilled water at room temperature. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]



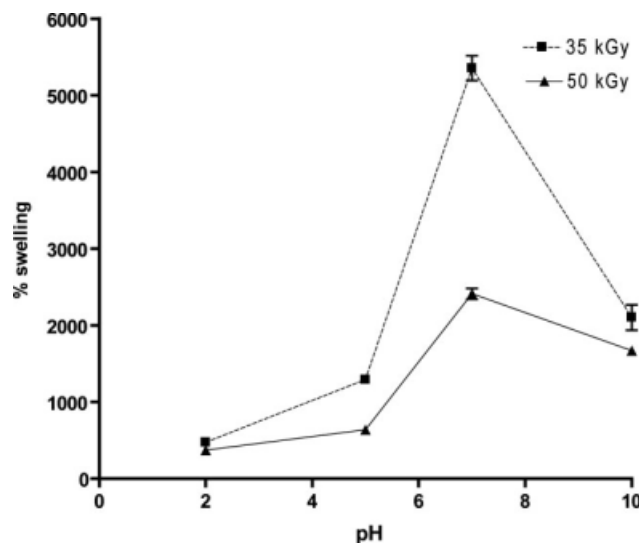
**Figure 5** Swelling kinetics of 35 kGy hydrogels at different pH.

also showed that after 24 h the hydrogel soaked in the medium at pH 7 had swollen up to eight times of its original size whereas at lower pH, the swelling process reached equilibrium after 8 h. The state of equilibrium for the hydrogel at pH 7 could not be determined as swelling kept continuing even after 48 h where eventually the hydrogel abruptly. However when the pH is further increased to pH 10, the swelling rate for the first 8 h were slightly higher than the swelling rate in pH 7 for the same respective time interval. Nevertheless after 8 h, the swelling process slowed down and eventually reached equilibrium after 24 h.

This sharp volume transition on the changes of pH from pH 2–7 was probably due to the hydrophilicity of carboxylic group from AAc. It is well known that below the pKa values (pKa: 4.25), the carboxylic groups of acrylic acid are in the form of COOH. When the pH is increased, the COOH was ionized



**Figure 6** Swelling kinetics of 50 kGy hydrogel at different pH.

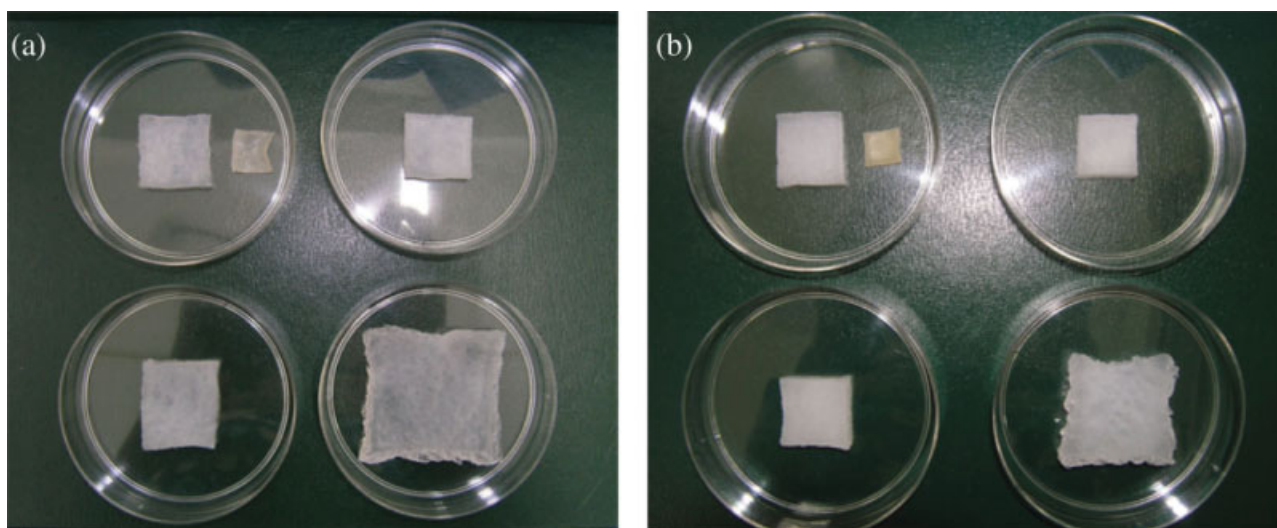


**Figure 7** Swelling dependency of 35 kGy and 50 kGy bacterial cellulose/AAC hydrogels on different pH values at 24 h.

and deprotonated to a negatively charged  $\text{COO}^-$ .<sup>14,22</sup> As a result, the electrostatic repulsion causes the swelling of the hydrogel<sup>18</sup> and greater expansion of the network thus gives a higher swelling ratio.<sup>33</sup> However, in an alkaline environment, the hydrogel interacted with the basic species, restricting interaction between the water molecules with the acid molecules. The nonionized condition of the carboxylic groups and the restricted interaction with water would result in the reduction of the swelling percentage particularly at  $\text{pH} < 5$  and  $\text{pH} > 10$ .<sup>2</sup> Figure 7 showed the swelling dependency towards different pH values at 24 h while a photo of swollen hydrogel at a series of PBS medium is shown in Figure 8(a,b).

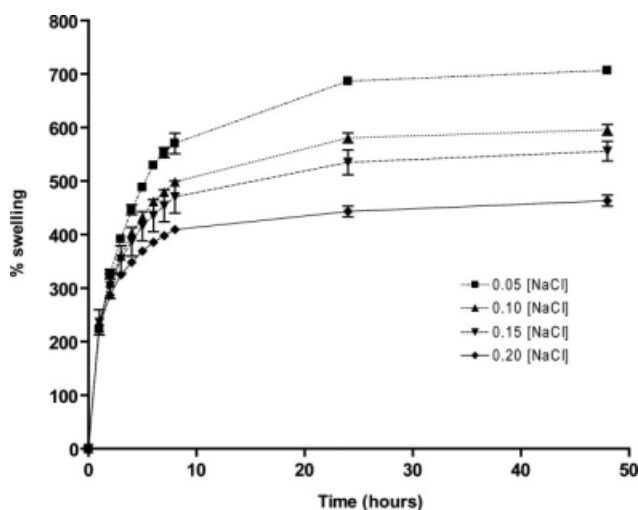
### Swelling behavior towards changes in ionic strength

Figures 9 and 10 represent the swelling behavior of the hydrogel at different concentrations of sodium chloride to evaluate the responsive character towards changes in ionic strength in the surrounding medium. The percentage of swelling for both hydrogel increased with time and reached the equilibrium swelling state after 24 h. Although the trends are the same, the hydrogel synthesized at 35 kGy has higher swelling percentage as compared to the hydrogel at 50 kGy due to the compact nature of the network and smaller pores. The results agreed well with the morphology study of the hydrogels as shown in Figure 3(a,b). However, the swelling dependency of the hydrogels towards the external stimuli (Fig. 11) showed that increasing sodium chloride concentration will reduce the swelling



**Figure 8** (a,b). Hydrogels synthesis at 35 kGy and 50 kGy in different medium from top left, dried and swollen hydrogels in distilled water, PBS pH 2, bottom left PBS pH 5 and PBS pH 7. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]

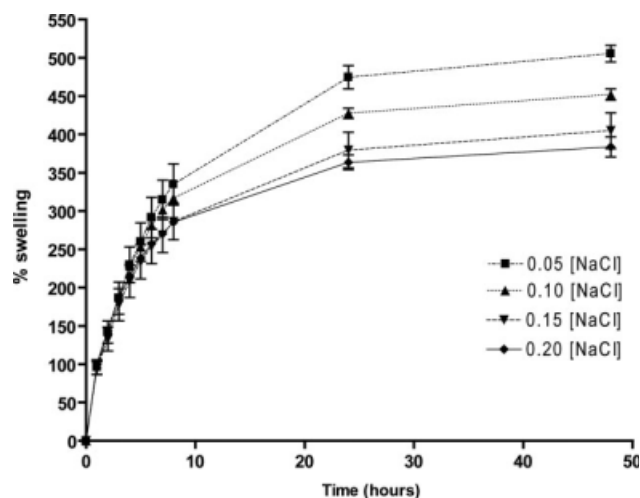
percentage in both hydrogels. This is because at higher concentration, the carboxylate groups ( $-\text{COO}^-$ ) are partially neutralized by the presence of sodium ions, reducing the network contact with water molecules and causing the hydrogels to shrink and expel water molecules from its network structure.<sup>14,18,22</sup> Decreasing the ionic strength means decreasing the concentration of counterions thus decreasing the shielding of the fixed charges on the polymer chains. In this condition, the concentration of ions outside the gels was also decreased resulting in increased differences in the concentration of the mobile ions between the hydrogels and the medium. This phenomenon creates an increased of osmotic pressure that favors swelling and known as Donnan effect.<sup>33,34</sup>



**Figure 9** Swelling kinetics of hydrogels synthesized at 35 kGy in different ionic strength.

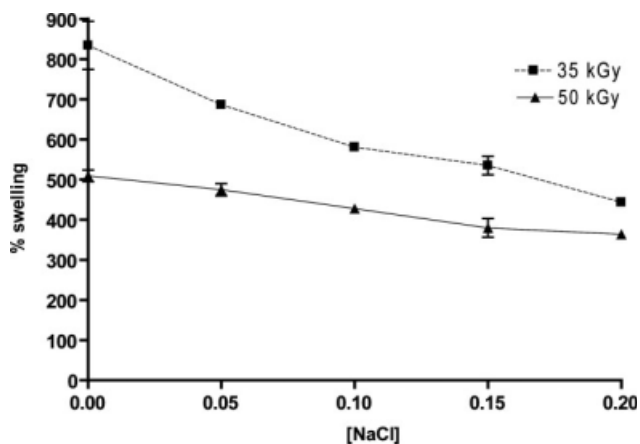
#### Swelling behavior towards changes in temperature

The swelling kinetics for the hydrogels at various temperatures is shown in Figure 12. It is observed that the swelling percentage for both hydrogels increases through time at all temperatures tested. However, the 50 kGy hydrogel was found to swell less than the 35 kGy hydrogel due to its denser structure that cause the mobility of the network more difficult. As crosslink density of the gel matrices increased with the dose, the gel became increasingly rigid to the thermal stimuli.<sup>32</sup> Although both hydrogels reached its equilibrium state after 24 h, the 35 kGy hydrogel exhibited the lowest swelling percentage at 37°C while the 50 kGy had a reduced swelling percentage at 25°C. Hydrogel synthesized at 35 kGy has increasing trends of swelling from 25

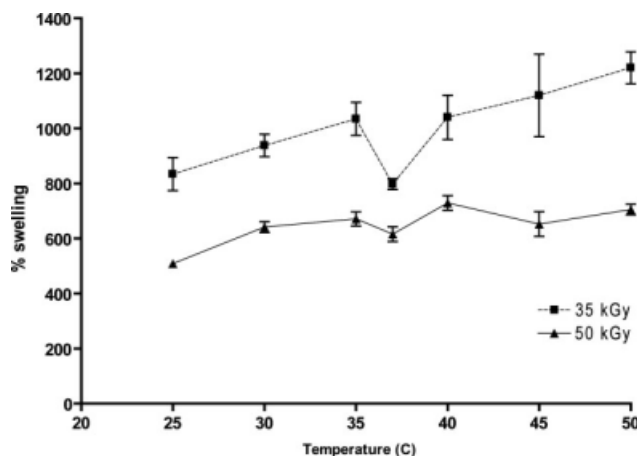


**Figure 10** Swelling kinetics of hydrogels synthesized at 50 kGy in different ionic strength.





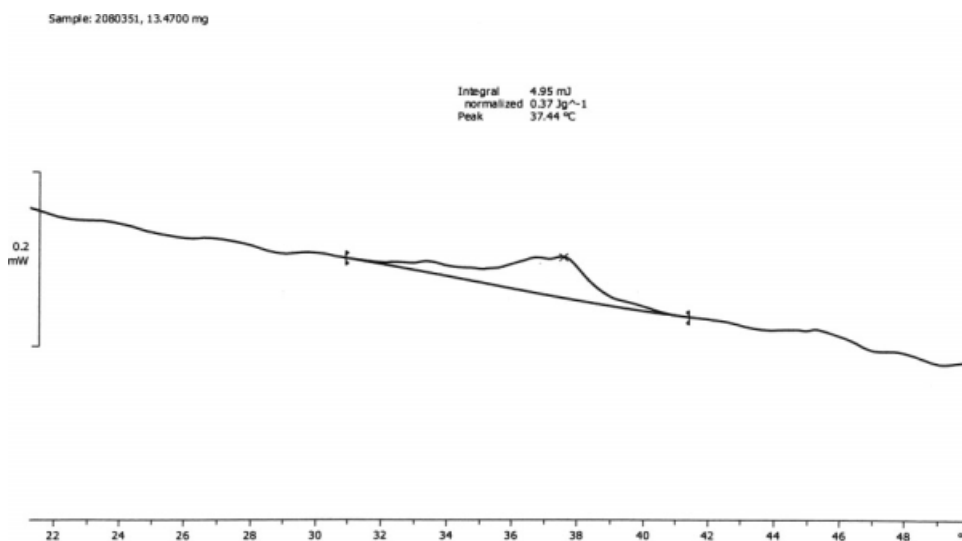
**Figure 11** Swelling dependency of 35 and 50 kGy cellulose/Aac hydrogels on different ionic strength at 24 h.



**Figure 12** Swelling dependency of 35 and 50 kGy bacterial cellulose/Aac hydrogels on different temperatures at 24 h.

to 35°C but the swelling percentage dropped at 37°C. The swelling started increasing again as temperature rises from 40 to 50°C. To confirm this finding, a differential scanning calorimetry was carried out for the 35 kGy hydrogel and a thermogram of hydrated hydrogel is shown in Figure 13. In this thermogram, the exothermic peak was found to appear at temperature 37.44°C which was approximately the same with the data obtained from the swelling equilibrium experiments. The exothermic peak was assumed to be caused by desolvation of the polymer chains.<sup>35</sup> This volume phase transitions is caused by the cleavage of hydrogen bonds between the hydrophilic groups of the polymer with the surrounding water molecules<sup>3</sup> thus eliminated the structured water and leading to the collapse of the hydrogel matrix.<sup>32</sup> It may also due to the strong hydrogen bonding of the polymer side chains that resulted in interpolymer complexation between the

polyacrylic acid and the cellulose fibers, forming a transient network.<sup>2</sup> However, a DSC analysis on the dried hydrogel did not show any exothermic peak, suggesting that the polymer chains have no contribution on the exothermic behavior. The exothermic peak was derived by the effect of water towards hydrophilic group in the matrix of the hydrogel.<sup>36</sup> The swelling pattern are slightly different for the 50 kGy where the swelling was the lowest at 25°C and the drop in swelling percentage can also be observed at two points which were at 37 and 45°C. While this phenomenon requires further evaluation, the swelling trend provides promising future in delivering active compounds. For instance at normal human body temperature (37°C) the active compounds would be retained in the network structure, but any changes of the pathological condition that may cause



**Figure 13** DSC thermogram of bacterial cellulose/Aac hydrogels synthesis at 35 kGy.

a raise in body temperature would trigger the release of the intended active compounds.

### CONCLUSION

This study has revealed that electron irradiation on the bacterial cellulose/AAC mixture is sufficient at 35 kGy for the formation of the hydrogel network. The crosslinking between bacterial cellulose fibers and poly AAC may probably due to the formation of the reactive sites by the hydroxyl/hydrogen radicals and strong hydrogen bonding. This stimuli sensitive hydrogel towards external changes may find many applications in drug delivery systems. As the hydrogels exhibits highest swelling percentage at pH 7, its unique characteristics at various temperatures makes it a very promising candidate for a pulse release carrier in the near future.

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