

## Effects of Plasticiser on the Morphology and Swelling Properties of Cellulose-based Hydrogels Derived from Wastepaper

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**ABSTRACT:** Cellulose was successfully extracted from wastepaper with a pre-treatment using sodium hydroxide (NaOH) and hydrochloric acid (HCl). The cellulose was then used to fabricate two types of hydrogels: cellulose/carboxymethyl cellulose (CMC)/polyvinyl alcohol (PVA) and cellulose/CMC/glycerol. Epichlorohydrin, an ECH solution was used as a cross-linker for the fabrication of both types of hydrogels. NaOH/urea/ultrapure water (NU) solutions were used to dissolve all the materials needed for the formation of hydrogels. The attenuated total reflectance-Fourier transform infrared (ATR-FTIR) showed the presence of hydroxyl group (O-H), hydrocarbon group (C-H) and carbonyl group (C=O) stretching. Scanning electron microscope (SEM) showed different porosities for both types of hydrogels, while the equilibrium swelling,  $swelling_{eq}$  of cellulose/CMC/PVA hydrogels were better compared to the cellulose/CMC/glycerol hydrogels. The highest  $swelling_{eq}$  was found for the cellulose/CMC/PVA hydrogel (3/3/4 ratio), CCP334 with 6.33 g/g, while the lowest  $swelling_{eq}$  (g/g) was obtained from the cellulose/CMC/glycerol hydrogel (3/2/5 ratio), CCG325 with 1.49 g/g.

**Keywords:** cellulose, hydrogels, carboxymethyl cellulose (CMC), polyvinyl alcohol (PVA), glycerol

### 1. INTRODUCTION

Paper recycling has been one of the main activities in the paper industry where the recycling rates of paper increased yearly. The recovery rates of paper and

paperboard doubled from 1990 to 2018 in the United States where it was estimated almost 53 million tons were recovered in 2018.<sup>1</sup> However, the consciousness of recycling wastes including papers in Malaysia is generally quite low i.e., 4.5% recycling rate in Kuala Lumpur in 2015.<sup>2</sup> As one of the alternatives to recycle the wastepaper, they are used as raw materials to prepare cellulose-based hydrogels in this study.

Hydrogels are composed of copolymers and homopolymers with hydrophilic properties through a cross-linking process that forms water-swollen networks, where even in the swollen state, the stability of their networks is not affected and the hydrogels have a high swelling ratio which is more than 100 indicating that they can absorb a large amount of biological fluids or water.<sup>3,4</sup> The hydrogels are also known to be stimuli-sensitive, which means that environmental stimuli i.e., pH, composition of solvents, light and temperature enable the reversible changes in the properties and shapes of the hydrogels.<sup>5</sup> It was found that the presence of various functional groups including the hydrophilic groups attached to the polymeric chains can also affect the properties of natural polymer-based hydrogels.<sup>6</sup>

The degree of cross-linking affected the level of equilibrium swelling of hydrogels while cross-linking with higher molecular weight elements increased the tendency of the hydrogels to resist mechanical deformation.<sup>7</sup> Mechanical properties of hydrogels were generally found to be increased through chemical cross-linking of cellulose from sodium hydroxide (NaOH)/urea solutions using epichlorohydrin (ECH), which is a common cross-linking agent used for polysaccharides.<sup>8</sup> Cellulose-based hydrogels can be fabricated using ECH cross-linker by different methods including heating the mixtures for 20 h at 50°C followed by freezing for 20 h at -20°C.<sup>9</sup>

However, the fabrication of hydrogels usually involves repeated cycles of the freeze-thawing process.<sup>10</sup> In this study, the time to fabricate the cellulose/carboxymethyl cellulose (CMC)/polyvinyl alcohol (PVA) hydrogels was significantly shortened as the freeze-thawing process was only performed in one cycle. Besides that, the use of reused papers which are abundantly produced, as raw materials to prepare the cellulose can also reduce wastage and increase the efficiency of wastepaper management especially in Malaysia. Two plasticisers namely PVA and glycerol were also used to compare the resultant hydrogels' properties in terms of swelling and morphology.

## 2. EXPERIMENTAL

### 2.1 Materials

Cellulosic wastes were procured from used papers from various sources which had been shredded into smaller pieces. NaOH pellets, CMC and PVA (99+% hydrolysed) were obtained from R&M Chemicals (Essex, UK) while the ECH solution was purchased from Merck (Darmstadt, Germany). The HCl solution, 33% (w/w) was purchased from Progressive Scientific (Selangor, Malaysia), urea was acquired from HmbG Chemicals (Hamburg, Germany) while the glycerol solution was obtained from Chemiz (Selangor, Malaysia). Ultrapure water with a resistivity of 18.2 MΩ.cm at 25°C was obtained from the Water Purifying System (ELGA, Ultra Genetic).

### 2.2 Extraction of Cellulose from Wastepaper

The procedures for extraction of cellulose from wastepaper were based on the methods used by Voon et al. with few modifications.<sup>11</sup> The shredded wastepaper were ground using a grinder machine before soaking with ultrapure water overnight to remove dirt and impurities. The slurry was then filtered and dried followed by stirring in NaOH (12% w/v) for at least 2 h to eliminate inks, hemicellulose and fillers. Then, the slurry was filtered and rinsed using ultrapure water to remove the NaOH. The slurry was then placed in an oven for 24 h at 60°C to remove the moisture due to the ultrapure water. Subsequently, the slurry was refluxed in 6% HCl for 4 h at 80°C followed by centrifuging with ultrapure water at 4000 rpm for 5 min in 4 cycles to remove the HCl from the slurry. It was then dried in an oven at 60°C until the ultrapure water was completely removed from the cellulose. The cellulose was cooled in a desiccator and kept there until further use.

### 2.3 Preparation of Cellulose-based Hydrogels

Cellulose-based hydrogels were prepared following the procedures as reported by Pang et al. and Chin et al. with some modifications.<sup>12,13</sup> Aqueous solutions with weight ratios of 7/12/81 of NaOH/urea/ultrapure water (NU) were prepared and stored in the freezer overnight until the temperature of the solutions reached -13°C. The frozen solutions were then thawed and different weights of cellulose i.e., 3 g and 4 g were added to the solutions before stirring to form heterogeneous mixtures. Next, CMC of two different weights which were 2 g and 3 g were added to the mixtures of each different concentration of cellulose. After that, PVA of different weights i.e., 3 g and 4 g were added to the mixtures which were heated gradually until it reached 90°C for 1 h to dissolve the PVA. Subsequently,

7.50 mL of ECH cross-linker was added drop by drop until the hydrogels were formed. In the case that no formation of hydrogels occurred, the cross-linking process was allowed to take place by placing the mixture in a freezer at 5°C overnight followed by thawing for 4 h. A total of 8 cellulose/CMC/PVA hydrogels of various ratios were successfully fabricated.

Similar procedures were used for the fabrication of hydrogels using glycerol where the PVA was replaced by glycerol which was added to the mixtures in the volume of 5 mL. However, different concentrations of cellulose and CMC were used where 2 g, 3 g and 4 g of each cellulose and CMC were chosen. The freeze-thaw method was also used where it was repeated (3 cycles) until the hydrogels were fabricated. A total of 17 cellulose/CMC/PVA and cellulose/CMC/glycerol hydrogels of different ratios were successfully fabricated.

## 2.4 Characterisation of Cellulose-based Hydrogels

The characterisation of cellulose-based hydrogels was conducted using the attenuated total reflectance-Fourier transform infrared (ATR-FTIR) and scanning electron microscope (SEM) instruments. The ATR-FTIR analysis was carried out using Perkin Elmer FTIR Frontier model spectrophotometer to obtain the spectrum in the wavenumber range of 400 cm<sup>-1</sup> to 4000 cm<sup>-1</sup> in order to identify the functional groups present in the cellulose-based hydrogels. SEM (JEOL, JSM-6390 LA) was used to observe the morphologies of the cellulose-based hydrogels.

## 2.5 Water Absorbency Measurements

Determination of the water absorbency was performed based on the procedure by Chin et al.<sup>14</sup> Firstly, the hydrogels were dried and weighed prior to soaking in distilled water at room temperature. The swollen hydrogels were taken out from the distilled water at predetermined time intervals (12 h to 37 days) where their weights were measured after wiping their surface with filter paper to remove the excess water. Equation 1 was used to calculate the equilibrium swelling (g/g).<sup>14</sup>

$$\text{Swelling}_{\text{eq}} \text{ (g/g)} = (m_s - m_i) / m_i \quad (1)$$

where  $m_s$  refers to the mass of swollen hydrogels, while  $m_i$  is the initial mass of dried hydrogels.

### 3. RESULTS AND DISCUSSION

#### 3.1 Fabrication of Cellulose-based Hydrogels

The hydrogels were formed using the cross-linking agent, ECH through the formation of covalent bond between the O-H group of cellulose with either PVA or glycerol. This process formed three-dimensional network that can trap water which reinforced the formation of gel with swelling property. A total of 8 cellulose/CMC/PVA hydrogels and 9 cellulose/CMC/glycerol hydrogels were fabricated as shown in Tables 1 and 2. The textures of hydrogels formed from PVA were harder and firmer compared to the hydrogels formed from glycerol which were stickier. The colour of both types of hydrogels were yellowish as shown in Figure 1, due to the heat treatment in order to ensure a complete dissolution of CMC and PVA. Observation on the cellulose/CMC/PVA shown that higher percentage of cellulose and PVA increased the hardness of the hydrogels while adding CMC reduced the hardness of the hydrogels. Higher concentration of PVA can increase the stiffness of the hydrogels due to their mechanical strength while increased concentrations of CMC can lead to softer and more flexible hydrogels due to their swelling properties.

Table 1: Composition of cellulose/CMC/PVA hydrogels in 100 mL NU solutions.

Hydrogels	Cellulose (g)	CMC (g)	PVA (g)	ECH (mL)
CCP323	3.00	2.00	3.00	7.50
CCP333	3.00	3.00	3.00	7.50
CCP324	3.00	2.00	4.00	7.50
CCP334	3.00	3.00	4.00	7.50
CCP423	4.00	2.00	3.00	7.50
CCP433	4.00	3.00	3.00	7.50
CCP424	4.00	2.00	4.00	7.50
CCP434	4.00	3.00	4.00	7.50

Table 2: Composition of cellulose/CMC/glycerol hydrogels in 100 mL NU solutions.

Hydrogels	Cellulose (g)	CMC (g)	Glycerol (mL)	ECH (mL)
CCG225	2.00	2.00	5.00	7.50
CCG235	2.00	3.00	5.00	7.50
CCG245	2.00	4.00	5.00	7.50
CCG325	3.00	2.00	5.00	7.50
CCG335	3.00	3.00	5.00	7.50
CCG345	3.00	4.00	5.00	7.50
CCG425	4.00	2.00	5.00	7.50
CCG435	4.00	3.00	5.00	7.50
CCG445	4.00	4.00	5.00	7.50

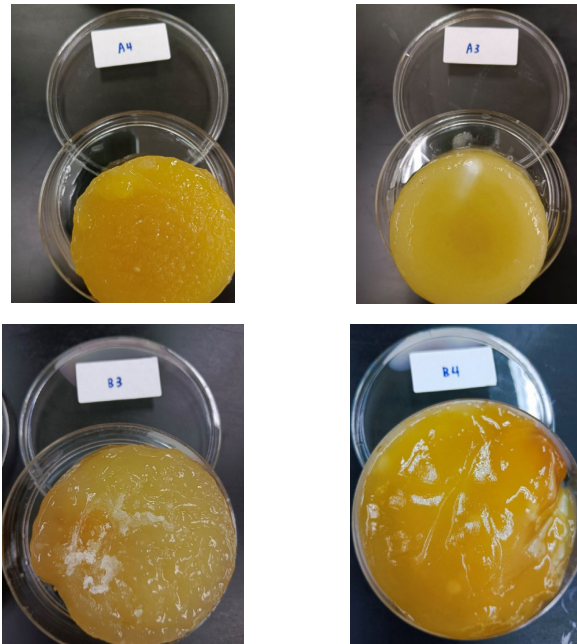


Figure 1: Photographic images of cellulose/CMC/PVA (A4: CCP334 and A3: CCP324) and cellulose/CMC/glycerol hydrogels (B3: CCG245 and B4: CCG325).

### 3.2 Fourier Transform Infrared (FTIR)

The FTIR spectra of cellulose fibre; cellulose/CMC/PVA and cellulose/CMC/glycerol hydrogels are shown in Figure 2. It was observed that there was a broad absorption peak at  $3,337\text{ cm}^{-1}$  for the cellulose, which corresponded to the O-H stretching. Similar absorption peaks were observed for the hydrogels with a slight shifting to the left for cellulose/CMC/PVA hydrogels while for cellulose/CMC/glycerol hydrogels, the O-H absorption peaks shifted slightly to the right. Strong hydrogen bonds between PVA and cellulose can be indicated by the shifting of the O-H peak to a higher wavenumber.<sup>15</sup> It was also found that the O-H band in cellulose had lower intensity compared to O-H bands in both types of hydrogels. This was due to the presence of O-H bands in both CMC and PVA that increased the intensity of the band, which was in accordance with a study by Das et al. while a study by Danish et al. showed there was a broad absorption peak of O-H for crude glycerol at  $3,322\text{ cm}^{-1}$ .<sup>16,17</sup>

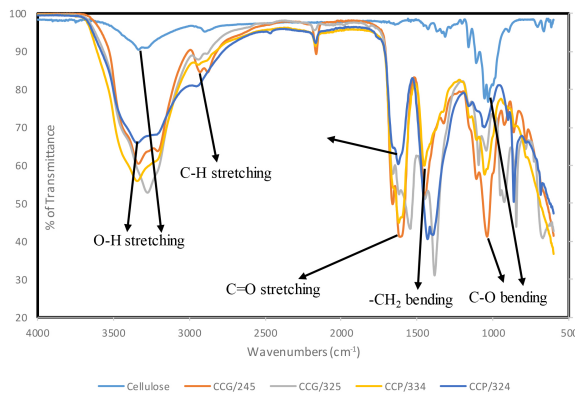
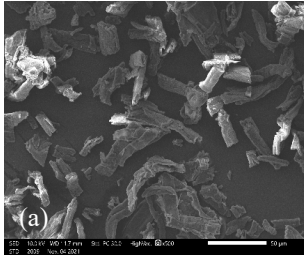


Figure 2: FTIR spectra of cellulose fibre; cellulose/CMC/glycerol and cellulose/CMC/PVA hydrogels.

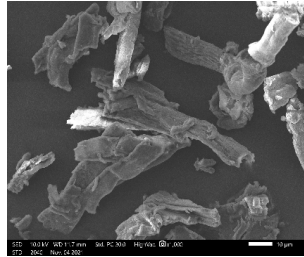
There was also an absorption peak at  $2,844\text{ cm}^{-1}$  at low intensity for cellulose assigned to C-H stretching which also appeared for all hydrogels, with slightly higher intensities and the peaks were also shifted to more than  $2,900\text{ cm}^{-1}$ .<sup>18</sup> The lower intensity of the C-H stretching at similar range of wavenumbers was also similar to the findings by other researches for cellulose extracted from different sources including bamboo in a study by Kassinov et al. and sago pith waste by Jampi et al. C=O stretching was not observed for cellulose but the peak appeared in both types of hydrogels at the range of  $1,546\text{ cm}^{-1}$  to  $1,619\text{ cm}^{-1}$ .<sup>19–21</sup> Another peak with low intensity was observed at  $1,428\text{ cm}^{-1}$  for cellulose which was assigned to the  $-\text{CH}_2$  bending.<sup>15,22</sup> A peak at  $1,000\text{ cm}^{-1}$  was also observed for cellulose which corresponded to the C-O stretching.<sup>19</sup>

### 3.3 Scanning Electron Microscope (SEM)

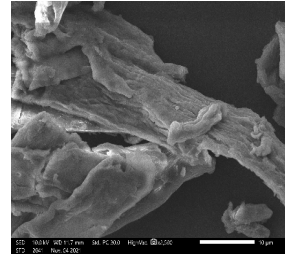
Figure 3 shows the SEM micrographs of cellulose fibre: cellulose/CMC/PVA and cellulose/CMC/glycerol hydrogels at different magnifications. Extraction of cellulose fibres from the wastepaper was successfully done as shown by the SEM micrograph of cellulose. Figure 3(a) showed rough surface morphology of the short fibrous cellulose-rich fibres with average length of 20  $\mu\text{m}$ , which was slightly different from a study by Sridhar and Park who found their cellulose fibres to be longer around 400  $\mu\text{m}$  to 450  $\mu\text{m}$  in length.<sup>23</sup> However, their method of pre-treatment for the extraction of microfibrillated cellulose from wastepaper only used the NaOH/urethane aqueous solution without HCl. Treatment of cellulose using HCl acid can break the hydrogen bonds between the cellulosic fibres, thereby producing shorter length of fibres due to the removal of the amorphous structure of the cellulose.



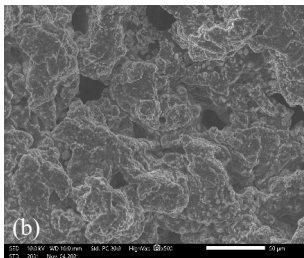
a(i): SEM (magnification 500 $\times$ , bar 50  $\mu\text{m}$ ) of cellulose



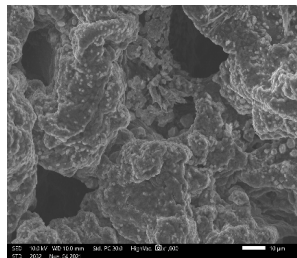
a(ii): SEM (magnification 1,000 $\times$ , bar 10  $\mu\text{m}$ ) of cellulose



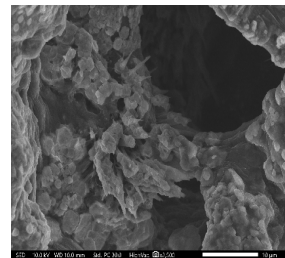
a(iii): SEM (magnification 2,500 $\times$ , bar 10  $\mu\text{m}$ ) of cellulose



b(i): SEM (magnification 500 $\times$ , bar 50  $\mu\text{m}$ ) of cellulose/CMC/PVA (3/3/4 ratio) hydrogel

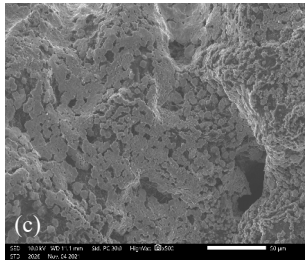


b(ii): SEM (magnification 1,000 $\times$ , bar 10  $\mu\text{m}$ ) of cellulose/CMC/PVA (3/3/4 ratio) hydrogel

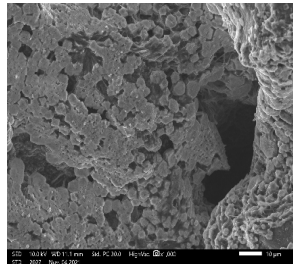


b(iii): SEM (magnification 2,500 $\times$ , bar 10  $\mu\text{m}$ ) of cellulose/CMC/PVA (3/3/4 ratio) hydrogel

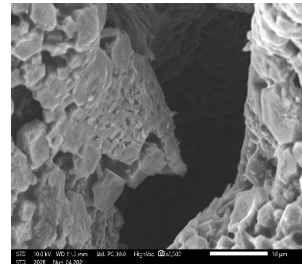




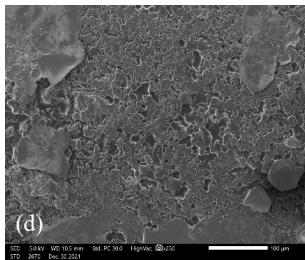
c(i): SEM (magnification 500×, bar 50 μm) of cellulose/CMC/PVA (3/2/4 ratio) hydrogel



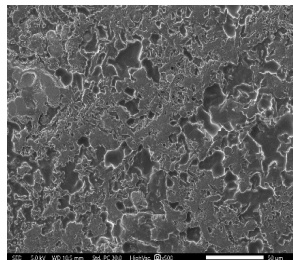
c(ii): SEM (magnification 1,000×, bar 10 μm) of cellulose/CMC/PVA (3/2/4 ratio) hydrogel



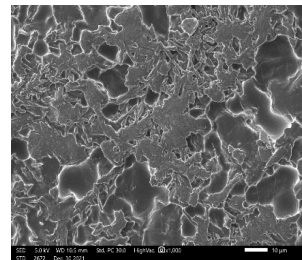
c(iii): SEM (magnification 2,500×, bar 10 μm) of cellulose/CMC/PVA (3/2/4 ratio) hydrogel



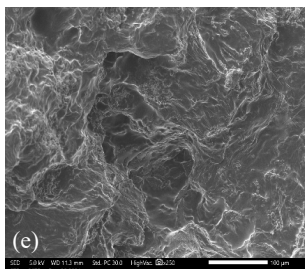
d(i): SEM (magnification 500×, bar 50 μm) of cellulose/CMC/glycerol (2/4/5 ratio) hydrogel



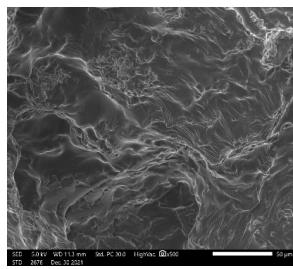
d(ii): SEM (magnification 1,000×, bar 10 μm) of cellulose/CMC/glycerol (2/4/5 ratio) hydrogel



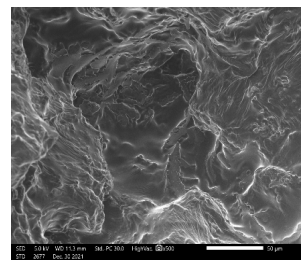
d(iii): SEM (magnification 2,500×, bar 10 μm) of cellulose/CMC/glycerol (2/4/5 ratio) hydrogel



e(i): SEM (magnification 250×, bar 100 μm) of cellulose/CMC/glycerol (3/2/5 ratio) hydrogel



e(ii): SEM (magnification 500×, bar 50 μm) of cellulose/CMC/glycerol (3/2/5 ratio) hydrogel



e(iii): SEM (magnification 500×, bar 50 μm) of cellulose/CMC/glycerol (3/2/5 ratio) hydrogel

Figure 3: SEM micrographs of (a) cellulose (b) CCP324 (c) CCP334 (d) CCG245 and (e) CCG325 at different magnifications.

Based on the SEM micrographs, it was found that the hydrogels with higher swelling<sub>eq</sub> had more porosity compared to the hydrogels with lower swelling<sub>eq</sub>. Smaller pores can be found in hydrogels with tighter and denser structure.<sup>24</sup> Other study by Mulhbacher et al. also indicated that the water uptake capacity is decreased in hydrogels with tighter structure which restrict the mobility of polymer chains.<sup>25</sup> It was found that the cellulose/CMC/glycerol hydrogels showed the surface morphology which was more compact with tighter networks, smaller sizes of pores and more homogenous when compared to the cellulose/CMC/PVA hydrogels which had higher swelling<sub>eq</sub>. It was also found that the cellulose/CMC/glycerol hydrogels showed more flexibility compared to the cellulose/CMC/PVA hydrogels which is in accordance with a study by Pan et al. where it was indicated that glycerol hydrogels had greater flexibility and ductility due to the reduction of pore sizes.<sup>26</sup>

### 3.4 Equilibrium Swelling

The equilibrium swelling of the hydrogels were performed at various time intervals which are shown in Figure 4.

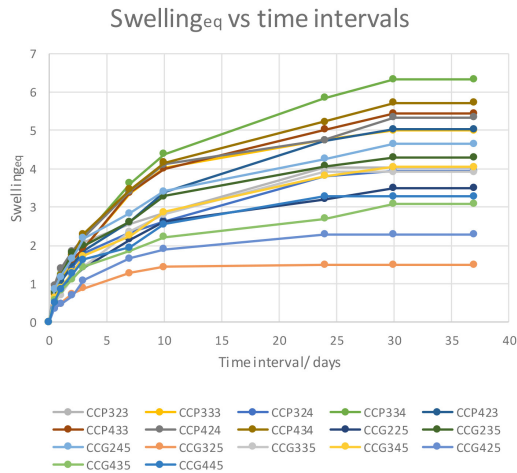


Figure 4: Swelling<sub>eq</sub> (g/g) of hydrogels at different time intervals (days).

It was found that the cellulose/CMC/PVA hydrogels had higher equilibrium swelling, swelling<sub>eq</sub> (g/g) compared to cellulose/CMC/glycerol hydrogels which has smaller sizes of pores as shown by the SEM analysis. These results are in accordance with a previous finding by Cai et al. which stated that the swelling ratio and hardness of hydrogels were decreased with addition of glycerol.<sup>10</sup> Highest swelling<sub>eq</sub> (g/g) of cellulose/CMC/PVA hydrogels was found for the hydrogel CCP334 (3/3/4 ratio) with 6.33 g/g, while the lowest swelling<sub>eq</sub> (g/g) was found for

the hydrogel CCP324 (3/2/4 ratio) with 3.94 g/g. As for cellulose/CMC/glycerol hydrogels, the highest swelling<sub>eq</sub> (g/g) of 4.65 g/g was found for the hydrogel CCG245 (2/4/5 ratio) while hydrogel CCG325 (3/2/5 ratio) was found with the lowest swelling<sub>eq</sub> (g/g) of 1.49 g/g. It was found that higher percentage of CMC increased the swelling as shown in Figure 4 which is similar to other findings by Roy et al.<sup>27</sup> This is due to the hydrophilicity of CMC with the presence of polar carboxyl (-COOH) functional groups.

#### 4. CONCLUSION

In conclusion, the cellulose was extracted from wastepaper and further used to fabricate cellulose/CMC/PVA and cellulose/CMC/glycerol hydrogels. The cellulose/CMC/PVA hydrogels were successfully formed in one cycle of the freeze-thawing method. It was found that the swelling<sub>eq</sub> was increased with higher content of CMC. It was also found that cellulose/CMC/PVA had greater swelling<sub>eq</sub> compared to cellulose/CMC/glycerol hydrogels as shown by the surface morphological of the hydrogels where more porous hydrogels had higher swelling<sub>eq</sub>. The optimum ratio of cellulose/CMC/PVA was found to be at 3/3/4 which formed hydrogel with highest swelling<sub>eq</sub> of 6.33 g/g. Based on this study, it can be suggested that the cellulose/CMC/PVA hydrogels have the potential to be used further as controlled-release device for agricultural applications.

#### 5. ACKNOWLEDGEMENTS

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