



## Synthesis And Characterization of Cellulose-Poly (Acrylamide-Co-Acrylic Acid) Graft Copolymer Hydrogel from Rice Straw

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### ABSTRACT

In this work, cellulose-poly(acrylamide-co-acrylic acid) graft copolymer hydrogels were prepared by free radical copolymerization of acrylamide and acrylic acid onto cellulose that had been extracted from rice straw, using Potassium persulphate as initiator and N,N'-methylenebisacrylamide as crosslinker. The extracted cellulose and synthesized graft copolymer hydrogel were characterized using Fourier transform infrared (FTIR) spectroscopy, X-ray diffraction (XRD), Scanning electron microscopy (SEM), Energy dispersive x-ray spectroscopy (EDX) and Differential scanning calorimetry (DSC). The FT-IR spectrum of the graft copolymer shows a band at  $1048\text{cm}^{-1}$  which correspond to C-O-C linkage confirming the successful grafting of the polymer on to the cellulose fibre. The diffraction pattern of raw rice straw obtained from XRD has an amorphous pattern and extracted cellulose obtained from XRD shows that, it has an amorphous pattern in raw rice straw and the extracted cellulose has a crystalline pattern. Morphologies of raw rice straw, extracted cellulose and that of synthesized hydrogel obtained shows that, the raw rice straw has a rough surface, the extracted cellulose has a smooth surface, and the hydrogel has a triangular shape with smooth surface. The glass transition temperature ( $T_g$ ) and the melting temperature ( $T_m$ ) of the hydrogel were found to be  $31.43^\circ\text{C}$  and  $92.44^\circ\text{C}$  respectively. (EDX) analysis shows a decrease in weight concentration of Silicon from 77.59 to 23.90, while that of Carbon increased from 22.41 to 57.03 after bleaching treatments. The hydrogel shows high water absorbency of 1141.28% at  $\text{pH} = 7$ . The synthesized hydrogel has the potential to be applied in waste water treatment, metal-ion removal, drug carrier and other fields where effective water utilization is essential.

**Keywords:** Rice straw; Cellulose; Acrylamide; Acrylic acid; Hydrogel

### 1.0 INTRODUCTION

Hydrogels, known as superabsorbent polymers (SAPs), are three-dimensional (3-D) crosslinked network structures composed of hydrophilic copolymers, that exhibit the ability to absorb and retain large amounts of water and biological fluids in relation to their body mass and without dissolution (Dai *et al.*, 2019). SAPs are commonly based on acrylamide (AAM), acrylic acid (AAc) and various vinyl monomers. The monomers can be polymerized by free radical and solution polymerization techniques (Raju and Raju, 2001). The polymerization is normally initiated by free radicals using thermal initiators. The crosslinked poly(Acrylamide-co-Acrylic Acid) PAAc has emerged as an important absorbent, because AAc is cheap and easy to polymerize to products of high molecular weight (Li and Wang, 2005). SAPs can share similar characteristics such as high water

content, porosity and soft consistency with natural living tissues more than other classes of synthetic biomaterials (Shivani *et al.*, 2015). These characteristics enable wide application of SAPs in several areas such as agriculture, architecture, drug-delivery system, hygienic products, coal dewatering, wastewater treatment and metal-ion removal (Li *et al.*, 2014). Recently, studies on the preparation of superabsorbents have focused on cellulose-graft copolymer hydrogel of acrylic acid (AAc) and acrylamide (AAM) monomers (Erizal, 2012). Cellulose from rice straw is a suitable candidate for the preparation of hydrogels by grafting or crosslinking reactions, as it has been shown to be a good precursor material for the preparation of hydrogels due to its great crosslinking and grafting ability attributed to abundant hydroxyl ( $-\text{OH}$ ) groups (Hosseini, 2011). The main compounds in rice straw are

cellulose, hemicelluloses, and lignin. Cellulose is a linear macromolecule consisting of (1–4) glycosidic linkages of  $\beta$ -D-glucopyranosyl monomers, and this  $\beta$ -(1–4) configuration contributes to its rigid structure and to the formation of aggregates through intra- or inter-molecular hydrogen bonds via hydroxyl groups. By chemical modification of cellulose through graft copolymerization with synthetic monomers many different properties, including water absorbency, elasticity, ion-exchange capabilities, thermal resistance and resistance to microbiological attack can be improved (Swantomo *et al.*, 2013).

### 1.1 Paper Objectives

The primary aim of this study was to extract cellulose from waste rice straw and graft it onto acrylamide and acrylic acid monomers to develop a graft copolymer hydrogel, by varying the composition of the extracted cellulose, acrylamide, acrylic acid, potassium persulphate (initiator), and N,N'-methylenebisacrylamide (MBA). The results obtained would provide information on the potential application of the hydrogel in waste water treatment, metal-ion removal, drug carrier and other fields where effective water utilization is essential.

## 2.0 MATERIALS AND METHODS

### 2.1 Materials

Materials used in this study are: Rice straw, Sulfuric acid (98% Sigma Aldrich, USA) Deionized water, Sodium hydroxide (97%, LobaChemie). Acrylic acid monomer (98.5%, Burgoyne, India), Acrylamide monomer (98%, Shanfau Guanghua, China), N, N'-methylenebisacrylamide (97%, Sigma Aldrich, USA) and Potassium persulphate (98%, Qualichem), Ethanol (96%, LobaChemie), all other reagents are of analytical grades and were used as received. All glasswares were cleaned thoroughly, rinsed with deionized water and dried in an oven at 100°C overnight.

### 2.2 Methods

#### 2.2.1 Sample Collection and Purification

Rice straw was obtained from a rice farm located at Kwalkwalawa area of Sokoto North Local Government, Sokoto State of Nigeria. The undesired impurities were first removed by hand-

picking and the rice straw was crushed with mortar and pestle into fine powder. It was then sieved in order to ensure homogeneity. The prepared sample was stored in airtight polythene bag prior to use.

#### 2.2.2 Extraction of Cellulose from Rice Straw

#### 2.2.3 Acid Treatment of Rice Straw

The acid treatment was carried out using a procedure reported by Ibrahim and El-Zawawy, (2013). About 50 g of the rice straw was weighed and transferred into a pressure cooker pot, 500 ml of 5% H<sub>2</sub>SO<sub>4</sub> (w/w) was added in order to obtain a 10:1 liquor to fibre ratio. The fibre was cooked at 170°C for 2 h. At the end, the pressure was released to atmosphere and the fibre was washed with distilled water until neutrality, then air dried for 48 h.

#### 2.2.4 Alkaline Treatment of Rice Straw

The alkaline treatment was carried out according to a procedure reported by Ibrahim and El-Zawawy, (2013). About 20 g of rice straw powder was weighed and transferred into pressure cooker pot. 200 ml of 10% of NaOH (w/w) related to the fibre was added in order to obtain a 10:1 liquor to fibre ratio. The fiber was cooked at 170°C for 2 h. The pressure was then released to atmosphere and the fibre was washed with distilled water until neutrality, then air dried for 48 h.

#### 2.2.5 Bleaching of Rice Straw with H<sub>2</sub>O<sub>2</sub>

Bleaching was carried out for the treated lignocellulose fibre using a similar method reported by AbdulRazak *et al.* (2014). In this procedure, the alkaline treated lignocellulose fibre was further treated with hydrogen peroxide solution. The treated fibre (17 g) was measured into a 1000 ml beaker into which 170 ml of H<sub>2</sub>O<sub>2</sub> was added. The fibre was placed in a water bath and maintained at 80°C for 60 min. The procedure was repeated twice in order to obtain total bleaching. The fibre was then washed with distilled water until neutrality, and air dried for 72 h.

#### 2.3.0 Synthesis of Cellulose-Poly(Acrylamide-co-Acrylic Acid)-Graft Copolymer Hydrogel from Rice Straw

Cellulose (Rice Straw)-poly(Acrylamide-co-Acrylic Acid) graft copolymer hydrogel was synthesized according to a procedure reported by

Shivani *et al.*, (2015). In a typical sample preparation under nitrogen atmosphere, 0.5 g of cellulose was dissolved in 15 ml of distilled water in a three-necked round bottom flask fitted with magnetic stirrer and, a reflux condenser. The mixture was stirred at 40°C for 10 min. Thereafter, 0.6 g acrylamide and 0.6 g acrylic acid were separately dissolved in 7 ml of distilled water, they were added and allowed for 15 min at 200 rpm and at 60-70°C to mixed completely, followed by the addition of 0.1 g potassium persulphate and 0.1 g

N-methylenebisacrylamide as initiator and crosslinker respectively. Then the reaction was allowed to proceed for 2 h. At the end of the reaction, the prepared hydrogel was removed carefully, cooled down to room temperature and washed with ethanol and distilled water to remove the unreacted monomers and impurities. Finally, the hydrogel was dried in an oven at 50°C until a constant weight was obtained. The chemical composition of all the prepared hydrogels is presented in Table 1.0

**Table 1.0 Chemical Composition of Synthesized Hydrogels**

Polymer	AAM	AAc	KPS	MBA	Cellulose
H <sub>1</sub>	0.6	0.6	0.1	0.1	0.5
H <sub>2</sub>	0.6	0.6	0.1	0.2	0.5
H <sub>3</sub>	0.6	0.6	0.1	0.3	0.5
H <sub>4</sub>	0.6	0.5	0.1	0.1	0.5
H <sub>5</sub>	0.6	0.5	0.1	0.2	0.5
H <sub>6</sub>	0.6	0.5	0.1	0.3	0.5
H <sub>7</sub>	0.6	0.5	0.1	0.4	0.5
H <sub>8</sub>	0.4	0.6	0.2	0.1	0.5
H <sub>9</sub>	0.4	0.6	0.2	0.2	0.5
H <sub>10</sub>	0.4	0.6	0.2	0.3	0.5

These compositions were chosen/arrived from the preliminary study carried out prior to the synthesis.

### 2.3.1 Percentage Yield (%)

The percentage yield (%) of cellulose from pretreated rice straw was calculated using the equation

$$\text{Percentage Yield (\%)} = \frac{W_f}{W_i} \times 100 \quad (3.0)$$

Where  $W_i$  = initial weight of rice straw before treatment and  $W_f$  = final weight of rice straw after treatment.

### 2.3.2 Water Absorbency Measurement and pH Response of the Hydrogel

The procedure for water absorbency measurement was adopted from Priya *et al.* (2019). Pre-weighed hydrogel sample was immersed in 250 ml of distilled water (at various pH ranging from 2-12) for 48 h and at room temperature. The water absorbency was determined by weighing the swollen gel after it had been allowed to drain on a sieve for 10 min. The water absorbency  $Q$  (gH<sub>2</sub>O/g sample) was calculated using the following equation;

$$Q (\text{gH}_2\text{O/g sample}) = \frac{W_s - W_d}{W_d} \times 100 \quad (3.1)$$

Where  $W_d$  and  $W_s$  are weight of dry and swollen hydrogel respectively.

Buffer solution at pH of 2, 4, 6, 8, 10, 12, and 14 was also used to determine water absorbency of the hydrogel.

## 2.4 Characterization

### 2.4.1 Fourier Transform Infra-Red – Analysis

The FT-IR spectra of the pure and bleached rice straw, acrylamide monomer, acrylic acid monomer and synthesized graft-co-polymer hydrogels were recorded on a CARY 630 FTIR spectrophotometer (Agilent) at wavelength range of 4000 – 650 cm<sup>-1</sup>.

### 2.4.2 Differential Scanning Calorimetry (DSC) Analysis

The DSC thermogram of the hydrogel was recorded using DSC 180 Star system-Mettler Toledo instrument at a heating rate of 0 – 124.54°C

### 2.4.3 X-ray Diffraction (XRD) Analysis

The X-ray diffraction (XRD) analysis of raw rice straw and bleached rice straw cellulose were carried out using Panalytical Emperean XRD analyzer.

### 2.4.4 Scanning Electron Microscopy (SEM)/ Energy Dispersive X-ray (EDX) Spectroscopic Analyses

The morphologies and elemental weight composition of raw rice straw, bleached rice straw cellulose and synthesized hydrogel were carried out using Phenom proX model.

## 3.0 RESULTS AND DISCUSSION

### 3.1 RESULTS

The percentage yield of the pretreated rice straw cellulose was presented in table 2.0 below. The acid-alkaline pulping method was selected as a pretreatment method for the rice straw. The action of the dilute acid was to solubilized hemicellulose and remaining lignin and cellulose. While alkaline

treatment is basically a delignification process, in which a significant amount of the lignin was dissolved and separated in a resulting black liquor. These colour changes during the pulping could have resulted from the degradation of the cell wall components and extractives as well as incomplete lignin removal (Ibrahim *et al.*, 2010). The pulp cellulose was bleached using hydrogen peroxide in order to remove the remaining lignin content that were not removed from the previous treatment, as it might inhibit crosslinking during grafting. The percentage yield of cellulose obtained after the acid, alkaline and bleaching treatments were 46.4%, 56.5% and 78.2% respectively. Similar percentage yield had been reported by Ibrahim and El-Zawawy (2013).

Table 2.0 Percentage Yield (%) of Cellulose from Pretreated Rice Straw

Rice Straw	Initial Weight (g)	Final Weight (g)	Percentage Yield (%)
Acid treatment using 5% H <sub>2</sub> SO <sub>4</sub>	50.00	23.18	46.36
Alkaline treatment using 10% NaOH	20.00	11.30	56.50
Bleaching using 30-31% H <sub>2</sub> O <sub>2</sub>	17.00	13.30	78.24

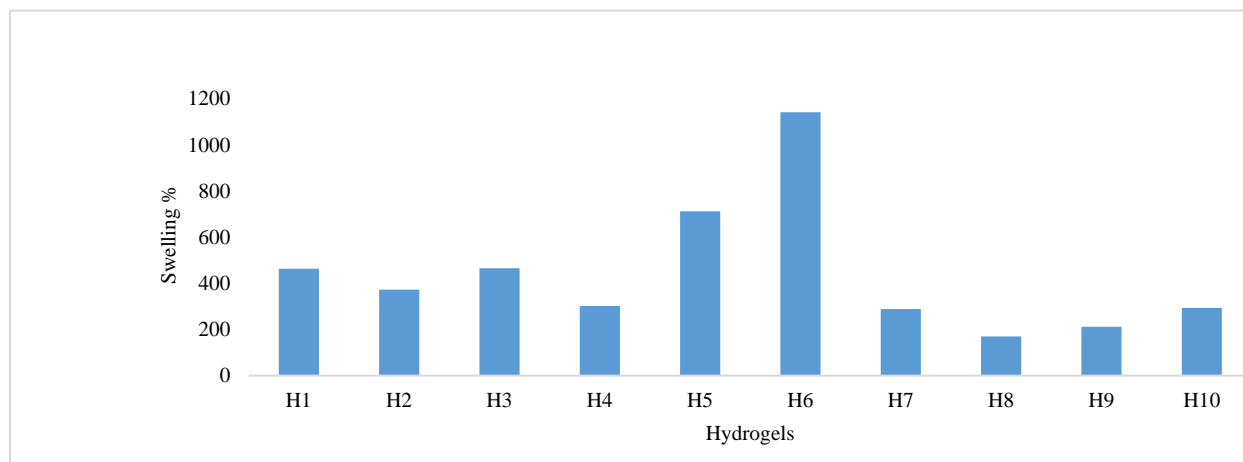


Figure 1a. Percentage Swelling (%) of Cellulose-g-Poly(AAm-co-AAc) Hydrogels at pH 7.

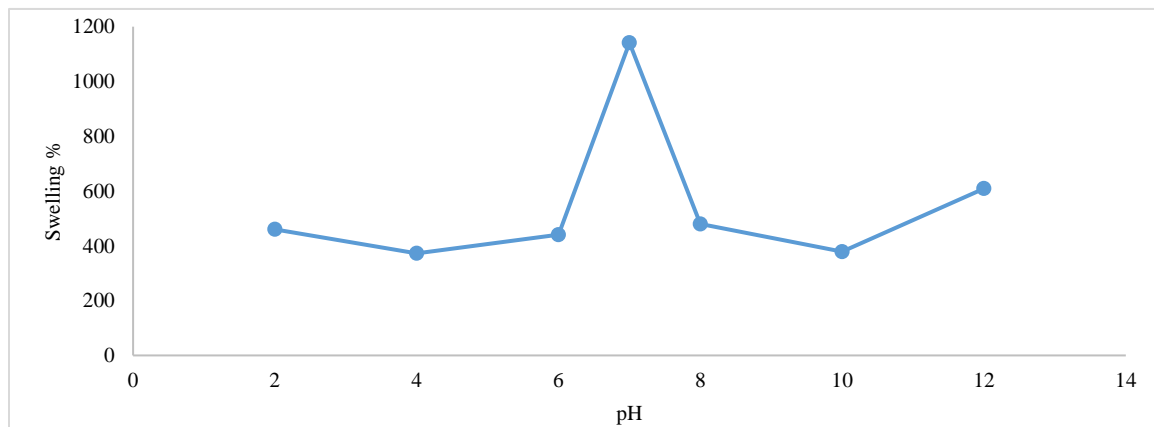


Figure 1b. Percentage Swelling (%) of Cellulose-g-Poly(AAm-co-AAc) Hydrogel (H6) at various pH level

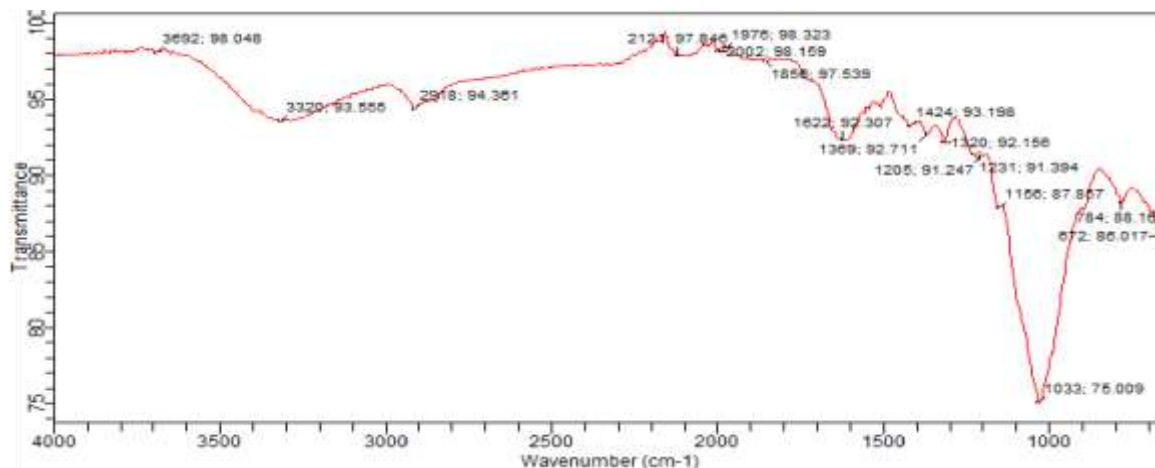


Figure 2a. FTIR Spectrum of Raw Rice Straw

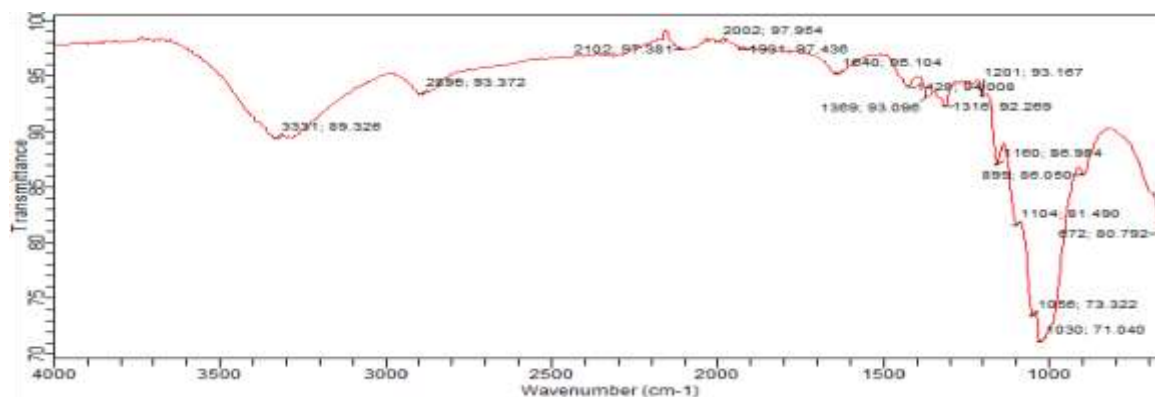


Figure 2b. FTIR Spectrum of Bleached Rice Straw Cellulose

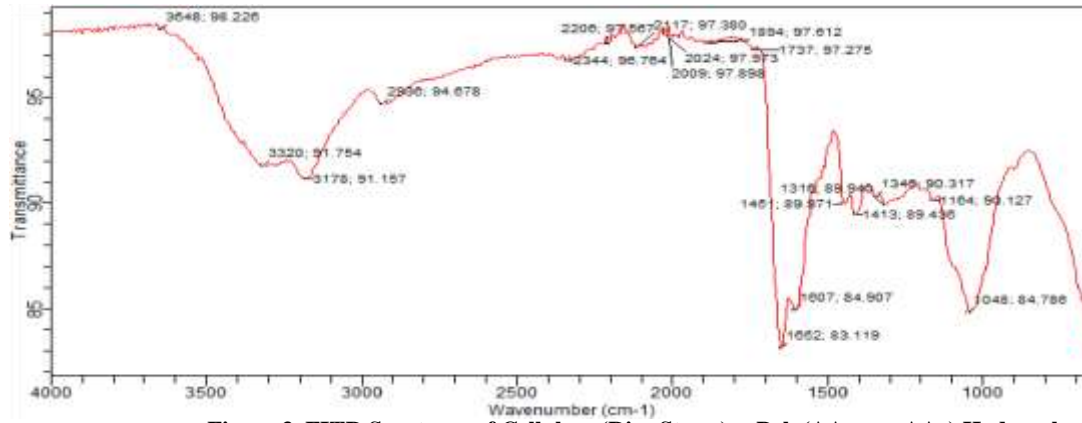


Figure 3. FTIR Spectrum of Cellulose (Rice Straw)-g-Poly(AAm-co-AAc) Hydrogel

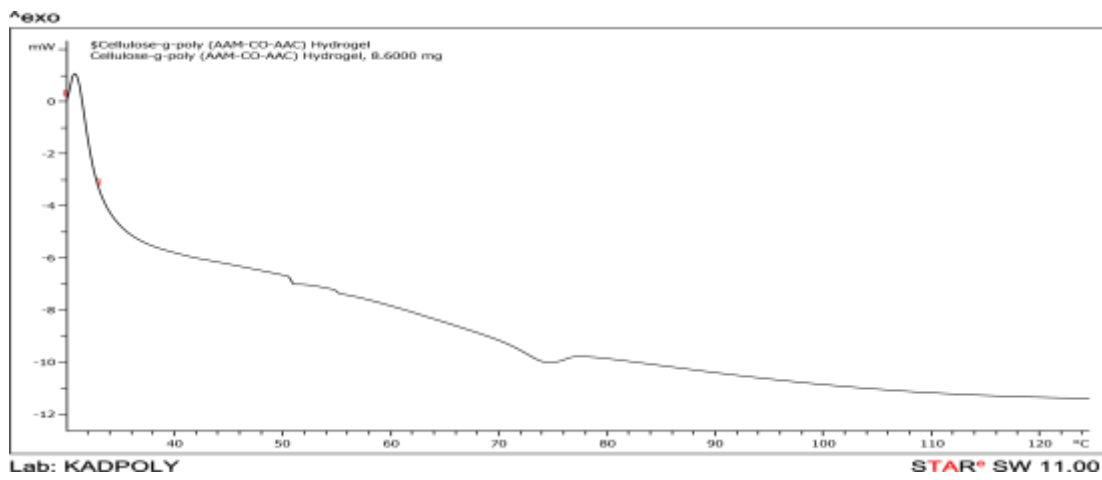
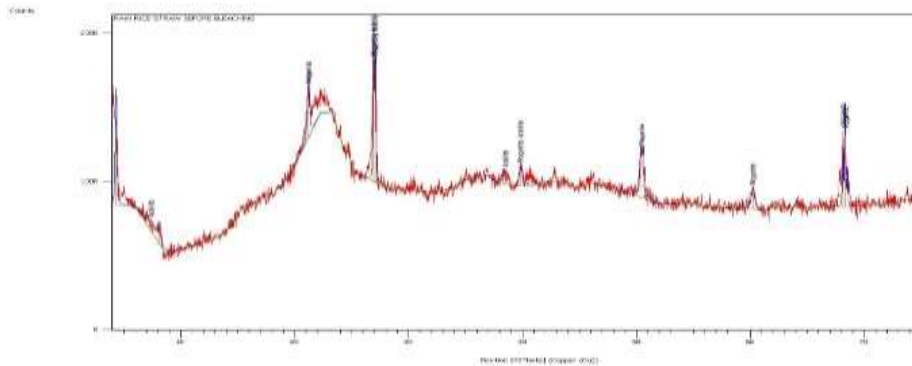
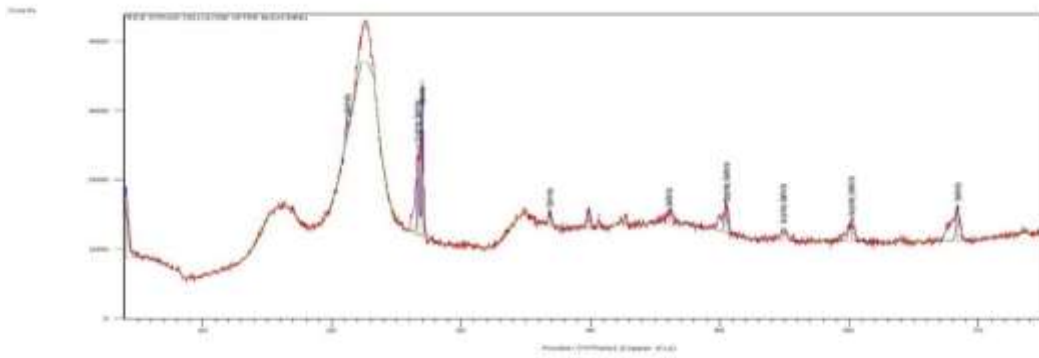


Figure 4. DSC Thermogram of Cellulose (Rice Straw)-g-Poly(Acrylamide-co-Acrylic Acid) Hydrogel

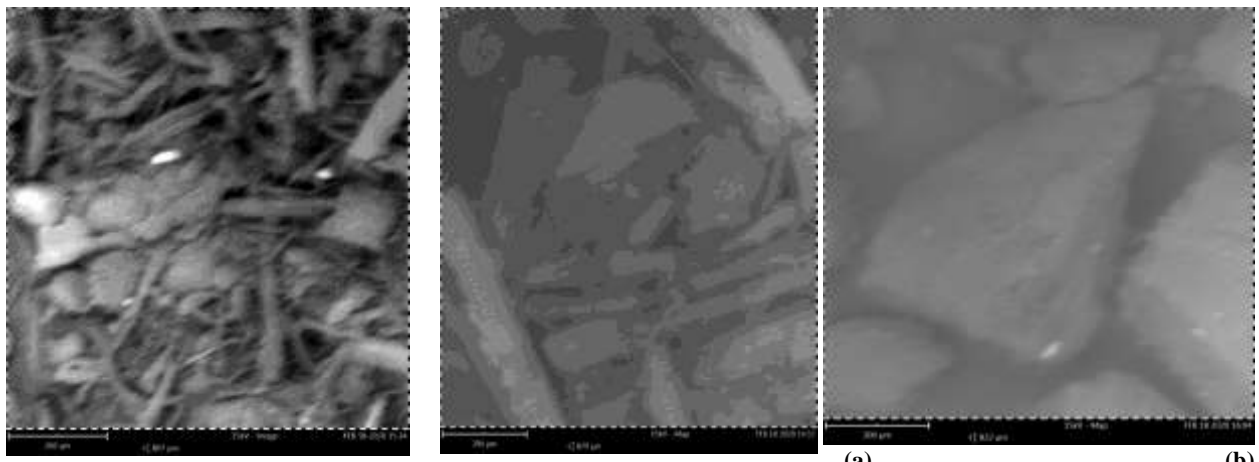


(a)



(b)

Figure 5. X-ray Diffractogram of (a) Raw Rice Straw (b) Bleached Rice Straw Cellulose

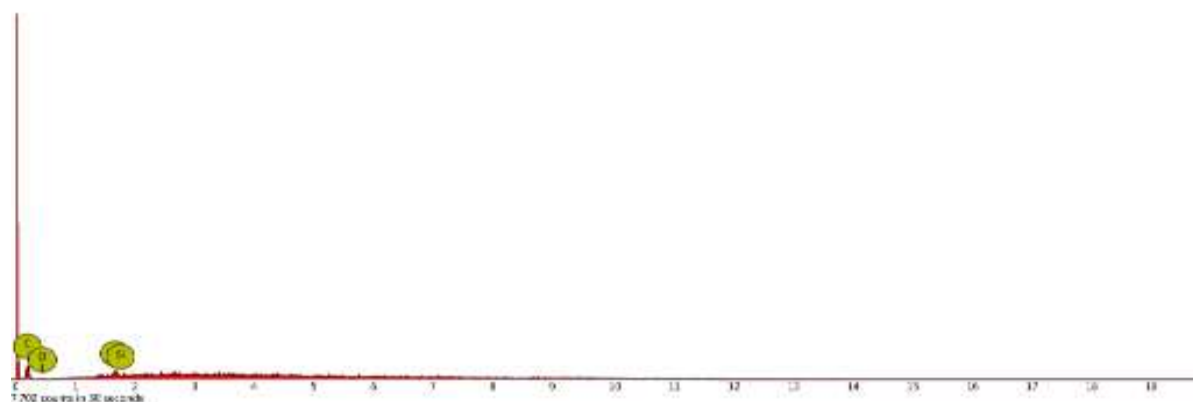


(c)

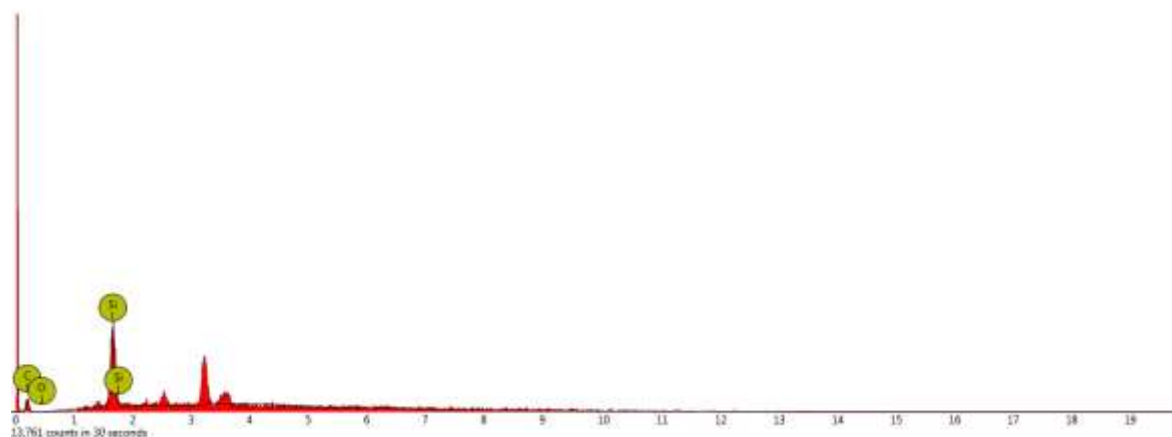
(a)

(b)

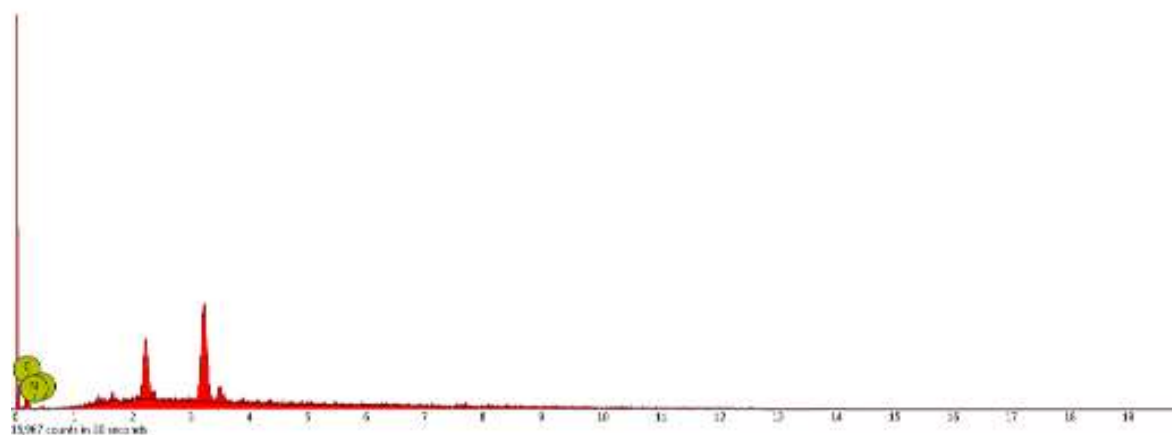
Figure 6. SEM Micrograph of (a) Raw Rice Straw (b) Bleached Rice Straw Cellulose (c) Synthesized Hydrogel.



(a)



(b)



(c)

Figure 7. EDX measurement of (a) Raw Rice Straw (b) Bleached Rice Straw Cellulose (c) Synthesized Hydrogel.

## 4.0 DISCUSSION

### 4.1 Swelling Behaviour and pH Response of the Synthesized Hydrogel

Swelling behaviour of copolymer hydrogels depends upon the composition of the gels, especially if one constituent monomer contains

ionizable groups (Nesrinne and Djamel, 2017). The percentage swelling of the synthesized hydrogels is shown in Figures 1a and 1b.

Figure 1a shows the percentage water uptake of all the synthesized hydrogels at neutral pH, it was observed that the percentage swelling increases with increase in crosslinker concentration (i.e. H1-H3, H4-H6 and H8-H10). This is because the increase in water absorbency before the optimum MBA content was attributed to the low crosslink density and hydrophilicity of MBA which enhanced the swelling ratio of the copolymer (Rop *et al.*, 2019). The maximum water uptake of all the hydrogels was found at H6 with about 1141.28 g/g. However, the percentage swelling decrease from H6 to H7, the decrease in water absorbency above the optimum MBA content may have resulted from a highly crosslinked network which decreased the porosity and flexibility of the polymer chains (Rop *et al.*, 2019).

Figure 1b shows the percentage swelling of H6 at various pH level. From the figure it shows that the hydrogel is pH sensitive, and the maximum water uptake was found at pH 7.0 as reported above. This result was found to be in agreement with the work of (Nesrinne and Djamel, 2017; Rop *et al.*, 2019 and Ariaenejad *et al.*, 2019; Mondal and Haque 2016). Because the equilibrium constant [pKa] of carboxylic acid contained in the copolymer is about 4.5, and the carboxyl group of hydrogels tend to dissociate at a pH > 4, the osmotic pressure inside the hydrogels increases resulting in a higher swelling (Nesrinne and Djamel, 2017). At pH 7-12, the system has a basic pH and the concentration of basic cations also increases. Finally, the decrease in swelling above pH 7.0 can be attributed to the fact that the -COOH groups may dissociate and further increase the amount of mobile ions leading to a decrease in osmotic pressure (Ariaenejad *et al.*, 2019).

#### 4.2 FTIR Analysis

Figures 2a and 2b shows the spectra of both raw and bleached rice straw. The band at 1622 cm<sup>-1</sup> which is assigned mainly to C=O stretching vibration of the carbonyl of the acetyl groups in the xylan component of hemicellulose and also typical for structural features of lignin (Ibrahim *et al.*,

2010), was present in the spectrum of raw rice straw, but its intensity has drastically reduced after bleaching with hydrogen peroxide. Other bands are well-known and specific to cellulose. O-H appears at 3320 cm<sup>-1</sup> in raw rice straw spectrum while after bleaching it appears at 3331 cm<sup>-1</sup>, C-H (stretching) appears at 2918 cm<sup>-1</sup> in raw while after bleaching it appears at 2895 cm<sup>-1</sup>, C-H (bending) appears at 1424 cm<sup>-1</sup> and 1428 cm<sup>-1</sup> in both raw and bleached rice straw respectively. Moreover, C-O-C stretching vibrations which are characteristic of β-(1-4)-glycosidic linkage (Ciolacu *et al.*, 2011), appears at 1033 and 1030 cm<sup>-1</sup> in raw and bleached rice straw respectively. Also, the increase in intensity observed in the spectrum after bleaching was as a result of the chemical treatment (Ibrahim and El-Zawawy, 2013). Thus, removal of lignin and hemicellulose by the chemical treatment was successful.

Figure 3 shows the FTIR spectrum of graft copolymer hydrogel of poly(acrylamide-co-acrylic acid). From the spectrum the band at 3320 cm<sup>-1</sup> is assigned to O-H, the N-H stretching appears at 3170 cm<sup>-1</sup> while N-H bending appears at 1607 cm<sup>-1</sup>. C-H (stretching) appears at 2938 cm<sup>-1</sup>, C-H (bending) appears at 1451 cm<sup>-1</sup>. The bands at 1346 cm<sup>-1</sup> and 1316 cm<sup>-1</sup> are assigned to C-N and C-O respectively. C-O-C appears at 1048 cm<sup>-1</sup>, C=O (acid) appears at 1737 cm<sup>-1</sup> and C=O (amide) appears at 1652 cm<sup>-1</sup>, which confirmed that the grafting reaction has occurred (Ariaenejad *et al.*, 2019).

#### 4.3 Differential Scanning Calorimetry (DSC)

DSC thermogram of cellulose (rice straw)-g-poly(acrylamide-co-acrylic acid) copolymer hydrogel sample is presented in Figure 4. The glass transition temperature (T<sub>g</sub>) and the melting temperature (T<sub>m</sub>) were observed around 31.43°C and 92.44°C respectively. The occurrence of a single T<sub>g</sub> in the blends is a strong indication of miscibility between acrylamide and acrylic acid (Daniliuc *et al.*, 1992; Vasquez-Torres *et al.*, 1993). The miscibility between acrylamide and acrylic acid has been attributed to the capacity of these monomers to form intermolecular H-bonding (Nesrinne and Djamel, 2017). This result indicates that the grafted copolymer hydrogel is thermally stable and it is endothermic, which agrees with the

work of Nesrinne and Djamel, 2017; Ramakrishna *et al.*, (2013).

#### 4.4 X-ray Diffraction (XRD)

The degree of crystallinity is a property of the fraction of crystalline content in the sample under consideration (Ibrahim and El-Zawawy, 2013). Crystalline regions are formed due to hydrogen bonds between the cellulose chains and Van der Waals forces between the cellulose molecules Bansal *et al.*, (2010). Figures 5a and 5b shows the X-ray diffractogram of raw rice straw and bleached rice straw cellulose respectively. Compared to the diffraction of raw rice straw and extracted cellulose bleached using hydrogen peroxide ( $H_2O_2$ ), there is an increase of intensity and sharpening of the major peaks which indicate that the bleaching of cellulose with  $H_2O_2$  does not reduced the crystallinity, this is in contrast to what was reported by Swantomo *et al.*, (2013) that the consumption of  $H_2O_2$  causes the breakage of hydrogen bonds in the crystalline part of cellulose chains, resulting in the reduction of crystallinity. The broad peaks diffraction pattern in Figure 5a represent the amorphous region which correspond to the impurities associated with the raw rice straw, while the sharp peaks diffraction in Figure 5b is the crystalline region of bleached cellulose (Feng and Wen, 2017). The X-ray diffraction pattern of bleached cellulose shows several sharp peaks between  $20-70^\circ$ , indicating the crystalline structure of the bleached rice straw.

#### 4.5 SEM Analysis

Figures 6a, 6b and 6c shows SEM micrographs of raw rice straw, bleached rice straw cellulose and the synthesized hydrogel respectively. The SEM indicate that treatment and bleaching process affects the morphological structure of the resulting samples, where from this morphological analysis, it is possible to observe the rough surface of the fibre before treatment. While the morphology of the bleached rice straw cellulose was found to have smooth surface, which corresponds to cellulose hornification. The bleaching treatments lead to fibre fibrillation indicating the presence of microfibrillated cellulose. While the morphology of the hydrogel shows a triangular shape with smooth surface. This result was found to be in conformity with the works of Ibrahim and El-Zawawy, (2013).

#### 4.6 Energy Dispersive X-Ray (EDX) Spectroscopy

The EDX spectra (Figure 7a) of raw rice straw indicate the presence of silicon with high weight concentration of 77.59, this correlates with the high ash content of raw rice straw (Ibrahim and El-Zawawy, 2013). However, the weight concentration of silicon reduced to 23.90 after bleaching (Figure 7b) indicating that the ash content has reduced. Moreover, the presence of oxygen indicates oxidation and the weight concentration of carbon increase from 22.41 (Figure 7a) to 57.03 (Figure 7b). This indicates that both the chemical treatment and bleaching processes played a vital role in the cellulose fiber formation. EDX measurements of the hydrogel (Figure 7c) shows the presence of elements expected from the graft copolymer of cellulose, monomers, initiator and crosslinker (C, K, O and N). This result is in conformity with the findings of Ibrahim and El-Zawawy, (2013); Feng and Wen, (2017).

#### 5.0 CONCLUSION

Cellulose has been successfully extracted from rice straw using acid-alkali pulping methods, and bleaching of the extracted cellulose was carried out by a hydrogen peroxide method. The extracted rice straw cellulose has a smooth surface morphology and good crystallinity as confirmed by SEM and XRD analyses. The synthesized graft copolymer hydrogel has excellent water absorption properties, high thermal stability and smooth surface morphology as confirmed by DSC and SEM analyses. The results further suggests that the hydrogel has the potential to be applied in waste water treatment, metal-ion removal, drug carrier and other fields where effective water utilization is essential.

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