

# Removal of Congo Red Dye using Synthesised Copper(II) Activated Carbon from Date Seeds

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Azo dyes are recognised as contaminants from the textile and printing industries that lead to human toxicity. Copper(II) activated carbon (CuAC) is an effective removal agent of dyes in these industries. The purpose of this study is to synthesise and characterise CuAC from date seeds. In addition, the efficiency of CuAC as a removal of Congo red (CR) in aqueous solution is also studied. Activated carbon (AC) was prepared from date seeds using phosphoric acid as activating agent, followed by activation process in a furnace at 500 °C for 2 hours. Copper(II) nitrate was used in the impregnation of AC to produce CuAC. The AC and CuAC were characterised using Attenuated Total Reflectance Fourier-Transform Infrared Spectroscopy (ATR-FTIR), BET surface area (SBET), Scanning Electron Microscope-Energy Dispersive X-Ray Spectroscopy (SEM-EDX), Atomic Absorption Spectroscopy (AAS) and X-Ray Diffraction (XRD). UV-VIS Spectroscopy was used to determine dye concentrations after treatment with removal agent of CuAC. The characterisation data proved that the CuAC has been successfully synthesised with 0.33% Cu(II) loaded onto AC and its surface area increased from 8.37 m<sup>2</sup>/g to 384.82 m<sup>2</sup>/g. The dye removal study was conducted at 10 ppm concentration of dye. Result revealed that 0.2 g of CuAC at pH 2 in 90 min removed 100% of CR dye.

**Keywords:** Copper(II) activated carbon; date seeds activated carbon; congo red; dye removal

## I. INTRODUCTION

Azo dyes are prepared by diazotisation of an aromatic amine and coupling with either phenol or naphthol. The aromatic amine (often N-alkylated) contains an acidic proton or active methylene group. This type of dye absorbed by the human body can undergo reduction decomposition to form carcinogenic amines due to enzymes in the body that have reduction properties. It causes health problems such as respiratory problems, skin sensitisation and carcinogenicity (Chavan, 2011). In addition, colours in dyes cause limit the underwater vision, restrain the aquatic plants from undergo a photosynthesis as well as toxic to aquatic life when large amount of dyes been release into the water stream (Chen *et al.*, 2019).

According to Berkane *et al.* (2019), dyes can be divided into three categories: anionic, cationic and non-ionic. Congo red is one type of azo dyes which is a benzidine-based anionic dye composed with two azo groups that been widely reported used in the industries. Congo red dye with the molecular formula of C<sub>32</sub>H<sub>22</sub>N<sub>6</sub>Na<sub>2</sub>O<sub>6</sub>S<sub>2</sub> can cause various health and environmental problems such as cancer and gene alteration if large amount of congo red was consumed (Shah *et al.*, 2020).

Industries such as textile, cosmetic, food and paper usually use the dyes and release the wastewater into water bodies (Kaur *et al.*, 2012). Before that, wastewater treatment for dye removal is strongly needed to prevent the negative effect of dye to the environment. The dyes such as congo red has a complex structure that is consequently difficult to remove

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and degrade in natural condition (Chen *et al.*, 2019). There are a few popular methods to remove dyes from wastewater such as ultra-filtration, biosorption, oxidation, biodegradation, adsorption, and ion-exchange (Wang *et al.*, 2011; Jamion *et al.*, 2017).

The adsorption process is one of the significant methods of advanced wastewater treatment to minimise the dangerous organic and inorganic compounds left in effluents (Khodaie *et al.*, 2013). This adsorption treatment gains a wide attention due to greater efficiency, non-toxic components, inexpensive, and simple removal (Shu *et al.*, 2017). The adsorption phenomenon depends on the interaction between the adsorbent's surface and the adsorbed species. The interaction may be due to chemical bonding, hydrogen bonding, hydrophobic and van der Waals forces. Activated carbon as adsorbent is an example of adsorption occurrence which is widely used in water treatment.

The activated carbon (AC) is a type of carbonaceous material that is rich in carbon and has large surface areas, non-toxic property, high porosity, and the ability to contain different functional groups (Hussein *et al.*, 2015). These features make activated carbon a very versatile and resourceful material (Aldawsari *et al.*, 2017). The development of economical adsorbents using waste materials has gained a lot of attraction. Several research studies reported a great deal on removal of heavy metals and dyes from water using waste material as an adsorbent. It can be prepared from agricultural waste such as wood, coal, peat, date seeds, coconut shells, nutshells, bones and fruit stone by chemical and physical activation (Salman *et al.*, 2015). Date seeds possess natural structure which contain lignocellulosic composition as well as low in ash content that nominating it to become one of the best starting materials in production of activated carbon (Krishnamoorthy *et al.*, 2019).

The chemical activation is a better method with single-stage process using phosphoric acid, nitric acid, zinc chloride, and bases such as sodium hydroxide and potassium hydroxide. An advantage of chemical activation is it will decelerate the formation of tar during thermal degradation of lignocellulosic material in the presence of a dehydrating agent. Moreover, it has been well recorded that the activation temperatures are low, while the porous carbon yield is high in chemical activation process compared to physical activation. It will also

break down the cellulosic material and suitable pore structure will be created from the carbonisation process as a result of dehydration (Obob *et al.*, 2018). Phosphoric acid is more preferred as a chemical activation agent compared to zinc chloride and others since it is eco-friendly and widely used in pharmaceutical and food industries (Al-Balushi *et al.*, 2017; Reddy *et al.*, 2015).

Deposit of metal element such as copper(II) into activated carbon has improved the adsorption capacity of dye in aqueous solution (Shu *et al.*, 2017). This type of dye removal is responsive as adsorbent and catalyst to remove dye due to its photocatalytic activity. However, commercially activated carbon is relatively expensive, so it is an interesting challenge to synthesise and make a relatively cheap activated carbon from agricultural waste (Hussein *et al.*, 2015).

This study focused on the synthesis of copper(II) activated carbon for dye removal. The AC was synthesised from date seed waste and believed to be a good, low-toxic and economical dye removal agent.

## II. MATERIALS AND METHOD

### A. Materials

Fresh Ajwa Dates Grade AA (1 kg) from Bani Hasyim Farm, Medina, Saudi Arabia were purchased and used as raw material. The seeds were collected by removing the date pericarps. The chemicals used were 30% and 85% aqueous solution of phosphoric acid ( $H_3PO_4$ ), copper nitrate ( $Cu(NO_3)_2$ ), congo red (CR), 2.0 M sodium hydroxide (NaOH) solution, 12.0 M hydrochloric acid solution (HCl), silica gel, deionised water and distilled water.

### B. Dye Removal Agent Preparation

#### 1. Preparation of activated carbon (AC)

Date seeds (100 g) were washed with water and dried in an oven at 90°C for 2 h. Dry samples were crushed with hammer and ground using a food grinder to get powdered date seeds. The pre-treated process continued when the powdered date seed was immersed in 250 mL of 30% aqueous solution of phosphoric acid for 24 h to give acidic conditions and washed using distilled water, filtered using vacuum filtration technique and dried under room temperature. The dried

powder date seed was pre-treated by impregnating it with 85% of concentrated phosphoric acid with the ratio of 2:1 (200 mL of phosphoric acid: 100 g of date seed). The mixture was heated using a hot plate by double boiling at a temperature range between 70°C to 90°C for 4 h. The drying procedure was carried out for about 24 h in the oven at 110°C. The activation process continued in the muffle furnace at 500°C for 2 h. The product was activated carbon (AC) and washed with distilled water using the vacuum filtration technique. The prepared AC was dried in the oven at 90°C for 3 h. Lastly, the sample was kept in a desiccator and ready for the preparation of copper(II) activated carbon (CuAC) (Hilal *et al.*, 2012; Jamion *et al.*, 2017; Ogungbenro *et al.*, 2018).

### 2. Preparation of Copper(II) Activated Carbon (CuAC)

Mass of 30 g of the prepared AC was immersed in 150 mL of 0.05 M copper(II) nitrate ( $\text{Cu}(\text{NO}_3)_2$ ) solution. The mixture was shaken with 100 rpm of rotation rate for 24 h. The CuAC was filtered and dried at 105 °C for 24 h (Huang *et al.*, 2006).

### 3. Characterisation

The functional groups of AC and CuAC were observed using Perkin Elmer Spectrum 100 FT-IR spectrometer with a reaction range of experiment between 4000  $\text{cm}^{-1}$  to 650  $\text{cm}^{-1}$  as attenuated by the total reactance (ATR).

The pore structural analysis of prepared AC and CuAC was done using nitrogen adsorption at 77 K surface area analyser, Micromeritics ASAP 2010. The surface of the sample was physically adsorbed by nitrogen gas and out-gassed under vacuum at 100 °C for 12 hours (Shu *et al.*, 2017).

The morphology of AC and CuAC was analysed using SEM Tescan VEGA3. The samples were coated with gold in a thickness range of 30-40 nm and equipped with EDX Oxford instruments x-act Model Series 118-0101. The EDX is an elemental analysis technique that gives the information of the element present in the sample.

Percentage of  $\text{Cu}^{2+}$  deposited onto the CuAC was analysed by AAS. Mass of 0.1 g of CuAC was digested with 5 mL concentrated HCl and heated for 30 min. Then, the solution was transferred into a 25 mL volumetric flask and deionised water was added to the mark. The sample was heated for boiling and filtered through filter paper. The filtrate was determined by the Perkin Elmer Atomic Absorption

Spectrometer Analyst 700 at 324.8 nm (Huang *et al.*, 2006).

The crystalline structure of Cu and CuAC were observed using Powder XRD at room temperature. The samples were scanned from 5° to 70°  $2\theta$  angles using a step interval 0.02° and scan speed of 5  $\text{min}^{-1}$ .

### C. Dyes Removal Study

The CR solution with concentration of 10.0 ppm were prepared. The 1.0 M sodium hydroxide (NaOH) and 12.0 M hydrochloric acid (HCl) were added to the dye solution to control the pH condition (Nasron *et al.*, 2018). The prepared CuAC was added into 10.0 ppm of 100 mL of dye solution in beaker at pH 2. The mixture was stirred with speed of 300 rpm for 90 min. Then, the dye solution was taken at each 15 min interval and keep for further analysis (Obboh *et al.*, 2018). The process was repeated at different pH which are pH 7 and 10, respectively. The effect of light and the effect of CuAC mass (0.2 g and 0.3 g) also conducted. The residual concentration of dyes in the solution were determined from UV-vis and the % removal of dye was calculated by using the Equation (1) (Obboh *et al.*, 2018).

$$\% \text{Removal} = \frac{C_1 - C_2}{C_1} \times 100 \quad (1)$$

Where,  $C_1$  was initial concentration while  $C_2$  was final concentration of dyes solution.

## III. RESULTS AND DISCUSSION

### A. Synthesis of AC and CuAC

Powdered date seed turned from brown colour to black paste when heated with 85% concentrated  $\text{H}_3\text{PO}_4$  at temperature range between 70-90°C for 4 h. The black paste turned to dry carbon when the mixture was heated in the oven for 24 h at 110 °C and continued being heated in a muffle furnace for 2 h at 500°C, resulting in hard brittle black carbon called as AC. After impregnation of AC with copper to form CuAC, the colour changed from black to grey and gained 91.30% of yield.

### B. Characterisation of AC and CuAC

The FTIR spectra of AC and CuAC are displayed in Figure 1. Both spectra show similar vibration band characteristics (Shu *et al.*, 2017). Absorption band of C-H can be observed at 2763

$\text{cm}^{-1}$  in AC and  $2792 \text{ cm}^{-1}$  in CuAC spectra. The existence of carbonyl group (C=O) can be seen at absorption band of  $1615 \text{ cm}^{-1}$  in AC spectra and  $1593 \text{ cm}^{-1}$  in CuAC spectra. Meanwhile, the presence of P-O-C appeared at  $1286$  and  $993 \text{ cm}^{-1}$  in AC and  $1277$  and  $1066 \text{ cm}^{-1}$  in CuAC spectra. The presence of P-O group in the structure of AC and CuAC was due to the phosphorus acid used in the preparation process of activated carbon (Jamion *et al.*, 2017). The intensity of P-O absorption band of CuAC was lower compared to AC which could be due to the reduction in the number of P-O attached to the surface of AC after Cu(II) loading in the formation of CuAC.

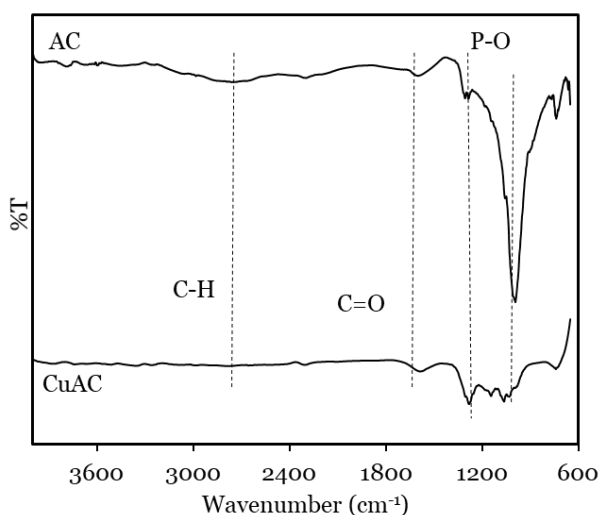


Figure 1. FTIR spectra of AC and CuAC

The data are listed in Table 1. From the table, the BET surface area (SBET) of CuAC was higher which was  $384.8175 \text{ m}^2/\text{g}$  compared to AC which was  $8.3742 \text{ m}^2/\text{g}$ . This indicated that the Cu(II) loaded on the surface of activated carbon helped in the evolution of higher surface area. The total pore volume of AC was  $0.03632 \text{ cm}^3/\text{g}$  with the average pore diameter of  $20.4707 \text{ nm}$ . Meanwhile, CuAC had  $0.38847 \text{ cm}^3/\text{g}$  of total pore volume with the average pore diameter of  $5.8393 \text{ nm}$ .

Table 1. The specific surface area, total pores volume and average pores diameter of AC and CuAC

Sample	Specific surface area, $S_{\text{BET}}$ ( $\text{m}^2/\text{g}$ )	Total pores volume, V ( $\text{cm}^3/\text{g}$ )	Average pores diameter, d (nm)
AC	8.3741	0.03632	20.4707
CuAC	384.8175	0.38847	5.8393

Figure 2 shows the pore size distribution graph. The AC has the mesopore type represented by larger peaks between pore diameters of  $4 \text{ nm}$  to  $50 \text{ nm}$  (Dias *et al.*, 2007). This occurrence was due to the effectiveness of phosphoric acid producing larger pores. Meanwhile, for CuAC, the majority of pore volume was at  $5\text{-}10 \text{ nm}$  due to the  $\text{Cu}^{2+}$  loaded on the surface. Thus, AC and CuAC exhibited well-developed porous structures which contained mesopores.

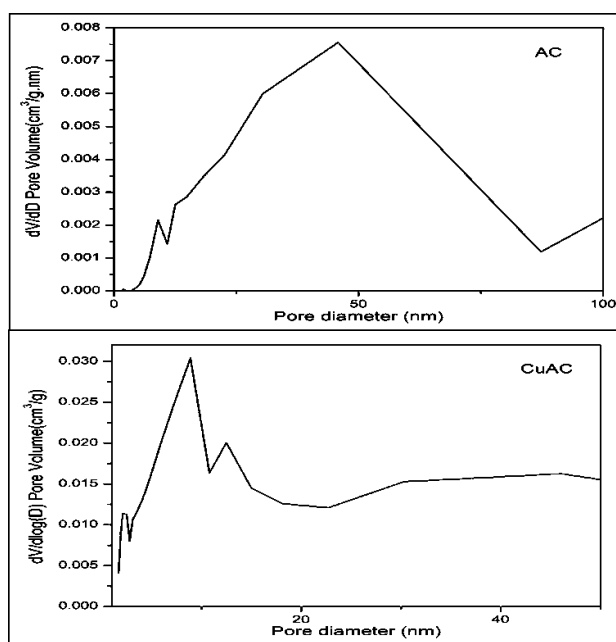


Figure 2. Pore size distribution of AC and CuAC

The  $\text{N}_2$  adsorption-desorption isotherms of AC and CuAC are shown in Figure 3 and can be assigned as Type IV according to IUPAC classification. This occurred due to the hysteresis loop which was associated with capillary condensation taking place in the mesopores. The graph approached the limiting value at  $P/P_0$ . The graph of AC was confirmed by hysteresis loops type H3. The shape was aggregated and had plate-like particles forming slit-like pores. Meanwhile, the hysteresis loops of CuAC were classified as H4 type. The shape was narrow slit-like pores, with irregular particles, broad size distribution and hollow spheres composed of mesopores (Foo & Hameed, 2010).

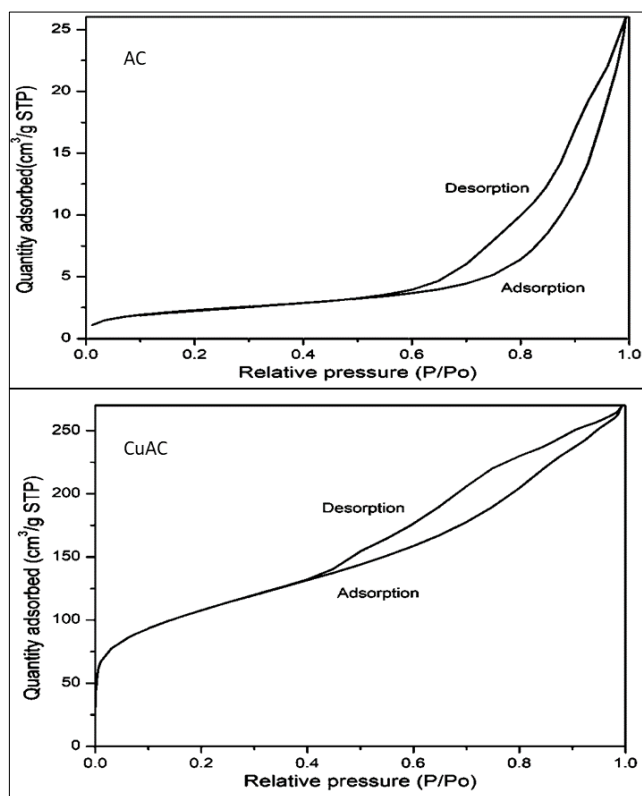


Figure 3. N<sub>2</sub> Adsorption-Desorption Isotherm of AC and CuAC

The SEM micrographs of AC and CuAC with magnification 1.97 Kx and 1.50 Kx are shown in Figure 4. Both micrographs show the pores have been well developed on the external surface with oval and circular patterns. This phenomenon occurred due to the activation process using phosphoric acid as activating agent to create pores on activated carbon and increase pore volume (Jamion *et al.*, 2017). The AC had a larger pore structure suitable for Cu(II) ions to be loaded on it, in the formation of CuAC (Shu *et al.*, 2017). After Cu(II) was loaded, the morphology was changed with the abundant pore formation with higher surface area suitable for the dyes trapped, leading to higher adsorption capacity.

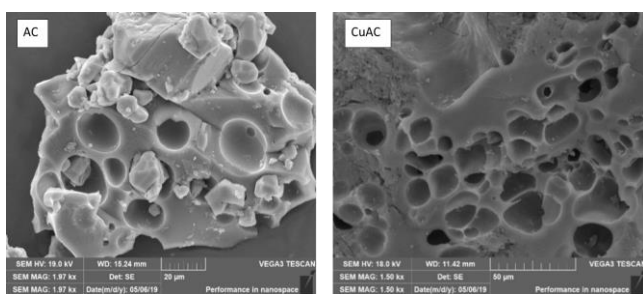


Figure 4. SEM micrograph of AC and CuAC

The Energy Dispersive X-ray (EDX) spectrum in Figure 5 shows the surface composition of AC and CuAC. Cu(II) was seen present in the EDX spectrum which indicated that Cu(II) was successfully loaded on the activated carbon surface. Meanwhile, the element percentages of AC and CuAC are tabulated in the inserted table. For AC, the presence of 37.34% of oxygen (O), 32.75% carbon (C) and 29.90% phosphorus (P) on AC surface can be seen. The higher percentage of oxygen and carbon determined that the date seed had higher content of fixed carbon. Phosphorus element was present due to the phosphorus acid used as activating agent. Phosphorus compound strongly bounded with oxygen and carbon (Jamion *et al.*, 2017). For CuAC, the presence of 79.43% carbon (C) and 20.57% copper (Cu) on AC surface was detected.

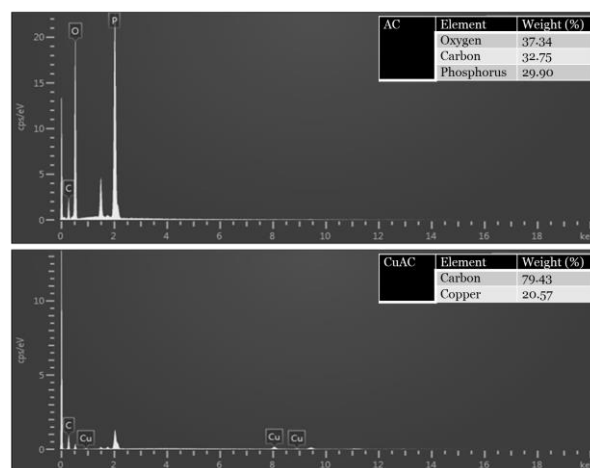


Figure 5. EDX spectrum of AC and CuAC

Mass of 30 g of AC was immersed in the 0.05 M (Cu(NO<sub>3</sub>)<sub>2</sub>) to become CuAC. AAS was used to analyse the amount of Cu(II) that was successfully loaded on the CuAC surface. In this study, 0.1 g of CuAC was used. The AAS result shows that in 0.1 g of CuAC, 0.327% Cu(II) was successfully loaded on AC. The good loading of Cu(II) on the surface of CuAC was due to the small diameter molecule of Cu(II) that can easily enter into the pore of AC and consequently increased the surface area after CuAC was formed. Cu(II) molecule penetrated deep into the AC pores to increase the pore volume (Shu *et al.*, 2017).

Figure 6 demonstrates the XRD spectra of AC and CuAC. The XRD analysis of CuAC shows one small diffraction peak corresponding to Cu(II) at 45.48° (Shu *et al.*, 2017; Manuel *et al.*, 2018), thus proving that the surface of activated carbon

was loaded with Cu(II). Furthermore, the diffraction peak shows sharp nature which contribute to the crystalline nature of synthesized CuAC (Iqbal *et al.*, 2019). Diffraction peak shown was very small due to only 0.327% Cu(II) loaded on 0.1 g CuAC surface as found in AAS analysis.

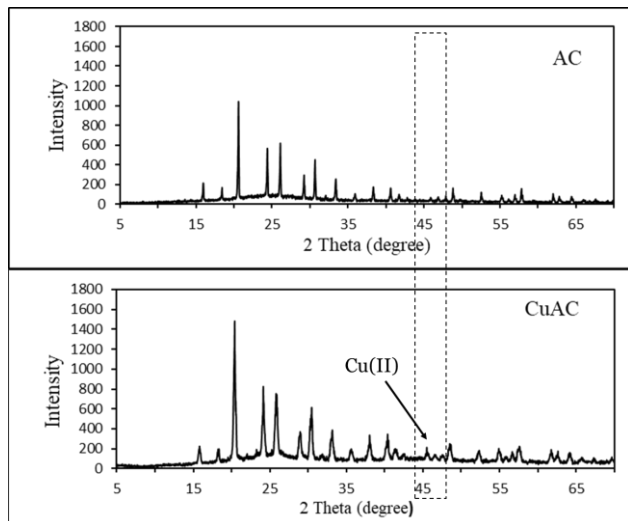


Figure 6. XRD spectra of AC and CuAC

### C. Removal of Dyes using CuAC

The 10.00 ppm of CR was used as dye sample. The control experiment has been done in 90 min without the presence of CuAC at pH 2. It was observed that the CR colour changed from orange to violet at pH 2 and no significant changes were seen from 15 min to 90 min at room temperature (Figure 7. (A)). The changes of dye concentration have been proven using UV-VIS spectroscopy. The result obtained showed that the concentration of dye slightly decreased from 10.00 ppm to 9.42 ppm at 90 min.

The experiment was repeated with the presence of 0.1 g of CuAC. The dye colour intensity clearly decreased in the Figure 7. (B) with the concentration decreasing from 10.00 ppm to 0.58 ppm. Consequently, it is shown that the CuAC had dye removal capability. Further study has been done to confirm the performance of CuAC as dye removal in the following discussion.

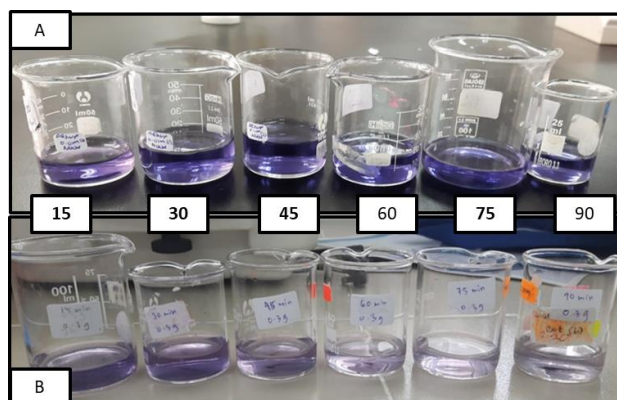


Figure 7. Changes of CR colour intensity in control experiment

### D. Optimization study

The experiments were conducted with various parameters such as effect of pH, the presence of light and different mass of CuAC. All experiments were conducted using 10.00 ppm of dyes and 90 min reaction time. The sample was taken at each 15 min interval.

#### 1. Effect of pH

The % removal of CR dyes at different pH conditions is illustrated in Figure 8. Within 90 min the % removal of dye using 0.1 g CuAC were 94.25%, 67.1% and 62.78% at pH 2, pH 7 and pH 10, respectively. The % removal of dye was higher in acidic condition (pH 2) because the abundant positive charges at the surface of CuAC were attracted to the negative charges of CR dye structure. The positive ions from  $\text{Cu}^{2+}$  were on the surface of CuAC itself and additional  $\text{H}^+$  ions were from HCl during the pH controlling process (Shu *et al.*, 2017; Sahoo *et al.*, 2019).

In neutral and basic conditions, the % removal of dye was lower.  $\text{OH}^-$  ion formed during the addition of NaOH led to deactivation of CuAC surface (Shu *et al.*, 2017). Increasing the basicity of reaction condition decreased the % removal of dye due to increasing electrostatic repulsion that occurred between the negative charge of CuAC surface with the negative charge in the CR dye structure (Sahoo *et al.*, 2019).

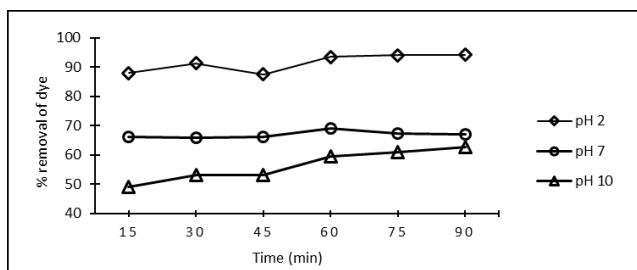


Figure 8. Effect of pH on % removal of dye study

### 2. Effect of light

The dye removal experiment was conducted with and without the presence of light using 0.1 g of CuAC at pH 2. The results are revealed in Figure 9. The % removal of dye showed no significant difference between with or without the presence of light. The result shows that light did not give a great impact in the removal of dyes. This result proved that the removal of dyes can be done independently with CuAC. In this study, the CuAC had two functions in dye removal which are as a catalyst with the presence of Cu<sup>2+</sup> ion on its surface that helped the dye degradation and as an adsorbent with a great number of active sites on CuAC that cause outstanding colourant adsorption (Shu *et al.*, 2017; Rabbani *et al.*, 2019).

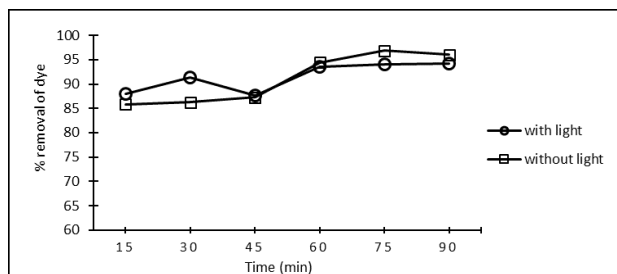


Figure 9. Effect of light on % removal of dye study

### 3. Effect of CuAC mass

The removal of dyes influenced by different mass of CuAC was studied at pH 2. The results obtained are shown in Figure 10. The % removal of dye at 90 min were 94.25% using 0.1 g of

CuAC, and 100% using 0.2 g and 0.3 g of CuAC. Within the time of reaction from 15 min to 90 min, there was no significant difference in % removal of dye using 0.2 g or 0.3 g of CuAC. Thus, the most suitable mass of CuAC for removal of dye at 10.00 ppm was 0.2 g which is cost-saving. The CuAC can act as dye removal at small amounts due to higher surface area and abundance of pores on its surface, leading to higher efficiency as dye removal. In addition, the capacity of CR adsorption increases with higher amount of CuAC used due to more adsorption sites was available for the process of adsorption (Chanzu *et al.*, 2019).

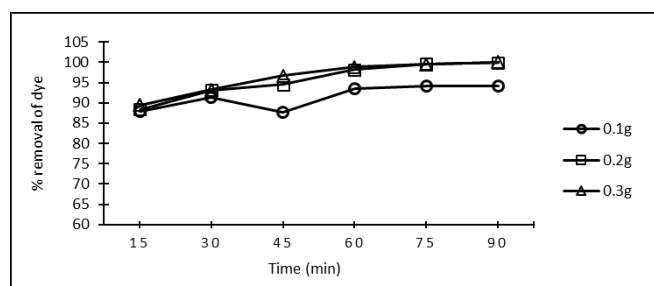


Figure 10. Effect of CuAC mass on removal of dye study

## IV. CONCLUSION

The CuAC from date seed has been successfully synthesised and used as CR dye removal. The optimum condition to remove 100 % of 10.00 ppm CR dye in aqueous solution was confirmed. The best condition was at pH 2, with mass of CuAC at 0.2 g and 90 min reaction time.

## V. ACKNOWLEDGEMENT

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