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Textile dyes removal by low cost SA-g-(PAAc-co-AM)/AC hydrogel nanocomposite

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ABSTRACT

Hydrogels were prepared by the free radical polymerization method successfully to remove textile dyes from the aqueous solution, due to the good properties of the hydrogels as being environmentally friendly, having a high ability to swell, high adsorption efficiency and good chemical stability. These hydrogels include surface SA-g-(PAAc-co-AM) hydrogel and its nanocomposite. The properties of the prepared hydrogels were studied using several techniques, including FESEM, TEM and XRD. Adsorption tests for these surfaces were performed on textile dyes including Brilliant green (BG), Methyl violate (MV), Malachite green (MG), Rhodamine B (RhB) and Brose Bengal (RB). We compared the surfaces of SA-g-(PAAc-co-AM) hydrogel, its nanocomposite and activated carbon (AC) as adsorbents. The best results of the percentage of removal (E%) of textile dyes were in the order of BG, RB, MV, MG and RhB. Arranging in the increasing order are as follows: SA-g-(PAAc-co-AM)/AC, SA-g-(PAAc-co-AM) and activated carbon (AC). The good results of the percentage of removal (E%) of SA-g-(PAAc-co-AM)/AC were also as follows: BG = 98.91, MG = 93.43, MV = 90.98, RhB = 77.55, RB = 70.66.

Keyword: Hydrogel, Textile dyes, Acrylic acid, Acryl amide, Brilliant green, Brose Bengal, Methyl violate, Malachite green, Rhodamine B.
Article type: Research Article.

INTRODUCTION

Brilliant Green (BG) dye is odourless yellow-green to green powder and is one of the commonly known cationic dye (its structure is shown in Fig. 1 a). Its colour causes severe allergic reactions in the digestive and respiratory systems. It also causes severe diarrhoea, nausea and vomiting, shortness of breath and severe coughing. In addition, when the dye comes into contact with the skin, it causes irritation with redness. Therefore, BG is considered as dangerous products such as nitrogen oxides, carbon oxides and sulfur oxides when heated to decompose (Alhattab 2023). Methyl Violet (MV) dye is 4-[4-(dimethylamino) phenyl] -(4-methyliminocyclohexa-2,5-diene-1-ilidine) Methyl-N, N-dimethylaniline (Fig. 1b). It is a family of organic compounds that have many wide uses, the most important as dyes. This is due to its composition that contains methyl groups, through which the colour of the dye is distinguished, and by changing the associated methyl groups, the color of the dye changes. This dye has many uses, including as an indicator for hydration of silica gel (Walli *et al.* 2018; Bader *et al.* 2019; Kadam *et al.* 2021; Alshamri *et al.* 2021). Rhodamine B (RhB) is a chemical compound and a dye (as shown in Fig. 1c). It is often utilized as a tracer dye within water to locate the rate and direction of flow and transport. Rhodamine dye fluoresce. Thus can be revealed easily and is low-cost with fluorimeters. RhB is a cationic dye with a high solubility in water, commonly used as drugs (in animals), in the staining of trials biological, printing, and textile manufacture. It is famous to have cancerous, and harmful, respiratory diseases as well as kidney failure. RhB reason eye irritation to humans as this compound contains a

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cationic dye. Malachite green (MG) dye is considered as organic compound, utilized as a dye stuff and controversially as an anti-microbial in aquaculture (Fig. 1d). MG is traditionally utilized as a dye for materials like leather, silk, and paper. However, its oral consumption is carcinogenic. Therefore, its detection in animal milk, fishes, and other food stuff designed for human consumption are of great alarm for the human health. Studies too confirmed that the products formed after the degradation of MG are also not safe and have carcinogenic potential (Kadam *et al.* 2021; Radia *et al.* 2022). Thus it becomes essential to remove it from waste water before releasing into aquatic environment. Rose Bengal (RB) dye (4,5,6,7-tetrachloro-2',4',5',7'-tetraiodofluorescein) is a stain, belonging to the class of organic compounds called xanthene's (as shown in Fig. 1e). Its sodium salt is usually utilized in eye drops to stain the damaged conjunctival and corneal cells and thereby, identifying damage to the eye (Radia *et al.* 2022). The stain is as well utilized in the preparation of Foraminifera for microscopic analysis, allowing the distinction among forms that were alive or dead at the time of collection (Aljeboree *et al.* 2019; Aljeboree & Alkaim 2019).

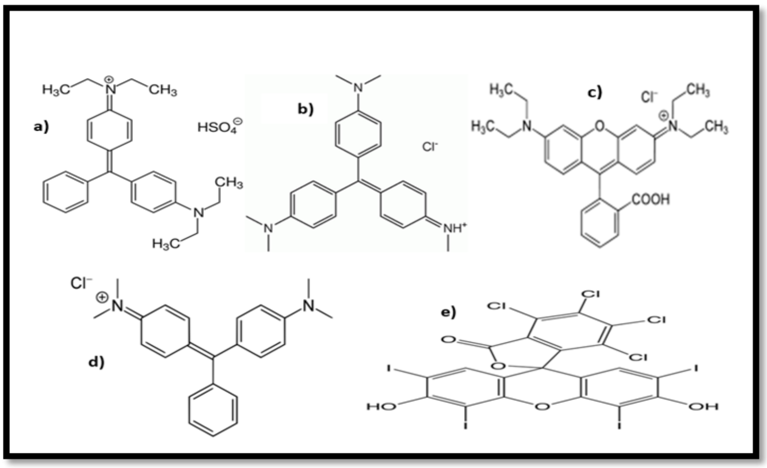

Fig. 1. chemical structure: a) Brilliant green (BG); b) Methyl violet (MV); c) Rhodamine B (RhB); d) Malachite green (MG); e) Rose Bengal (RB).

MATERIALS AND METHODS

Experimental part

Alginate sodium (Ag), $C_6H_9NaO_7$, Acrylic acid (AAC) C_3H4O_2, N, N'-methylene bisacrylamide $C_7H_{10}N_2O_2$, potassium per sulfate (KPS) $K_2S_2O_8$, all chemicals of the highest analytical purity were supplied by Sigma–Aldrich. Textile dyes (Brilliant Green, Methyl Violet, Roadmen B and Rose Bengal) were obtained from the Hila-Iraq factory. A stander solution (1000 mg L^{-1}) was prepared via dissolving 1 g of all textile dyes in 1000 mL of distilled water. The calibration curve of several concentrations for abovementioned textile dyes were prepared in serial dilutions (2-50 mg L^{-1}). Absorbance was measured at the λ_{max} for dyes and plotted against the concentration values of these textile dyes as appeared in Fig. 2.

Preparation of Activated Carbon (AC)

Orange peels (OP) were washed with distilled water, cut into small pieces and dried in the sun. Then it was ground by a special mill to obtain powder, and by using hydrochloric acid (0.1 N) the ground peel (powder) was activated and placed in a 250-mL beaker with stirring it for 1 h, followed by washing it several times with DW and drying in the oven at a temperature of 70 °C for 24 h. Afterward, it was burned at 300 °C for 1 h to obtain activated carbon. Acid-activated orange peels (AACOP) were ground to obtain powder (as shown in Fig. 3).

Preparation of SA-g-(PAAc-co-AM)/AC hydrogel nanocomposite

Preparation of SA-g-(PAA-co-AM)/AC hydrogel: We dissolved 0.1 g of Activated Carbone (AC) in 10-mL distilled water and dissolved sodium alginate SA (about 1 g) in 20 mL followed by stirring it for 1 h at room temperature (25 ºC). Thereafter, AC solution was added to SA, followed by adding 4-mL acrylic acid (AAc) and 0.5 g acryl amide (AM) in 2-mL distilled water, then stirring for 15 min. Afterward, potassium per sulfate 0.03 g in 1-mL distilled water and N, N'-methylene bisacrylamide (0.05 g in 1-mL distilled water), all added to SA solution. Processes were taken place in the presence of nitrogen gas to form free radicals, placed in a water bath for 4 h at 65 ºC. The SA-g-(PAA-co-AM)/AC hydrogel was cut, and washed several times to remove any uncreative materials. Thereafter, it was dried in an oven at 40 ºC for 24 h, and well ground.to obtain the SA-g-(PAA-co-AM)/AC hydrogel. Note that SA-g-(PAA-co-AM) hydrogel was prepared in the same way as abovementioned, without the addition of AC (as shown in Fig. 4).

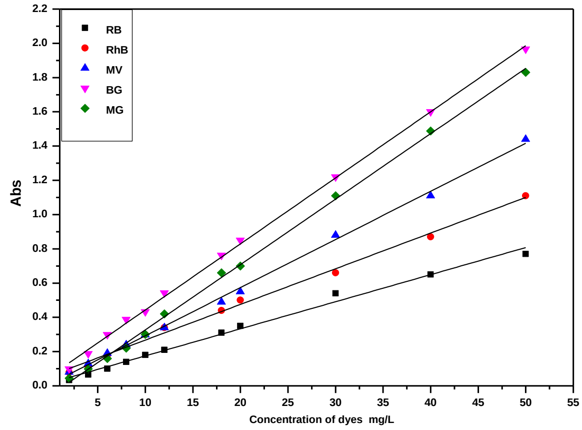

Fig. 2. Calibration curve of several concentrations for dyes.

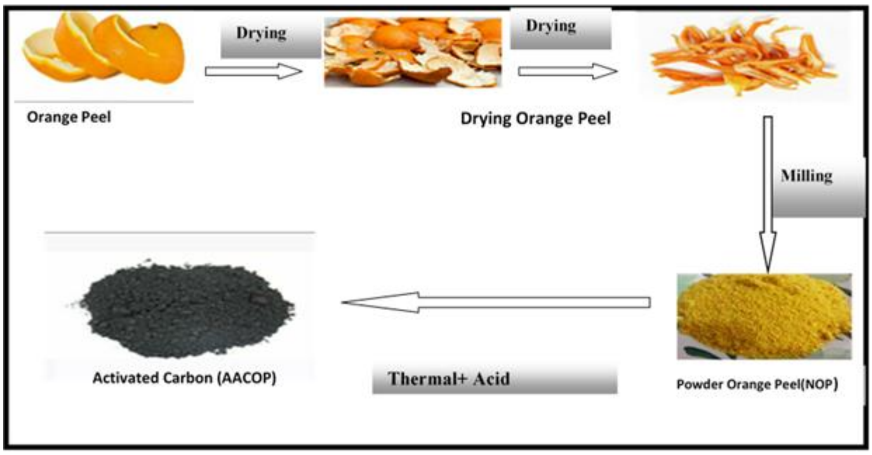

Fig. 3. Preparation of Activated Carbon (AC).

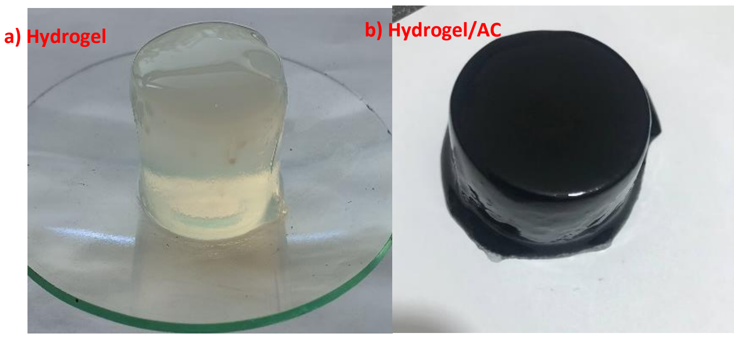

Fig. 4. Preparation of a) SA-g-(PAAc-co-AM); and b) SA-g-(PAAc-co-AM)/AC hydrogel nanocomposite.

RESULTS AND DISCUSSION

FESEM

The basic method for determining the characteristics of the surface morphology as well as the important physical properties of adsorption using FESEM technique. FESEM was taken for the adsorbents content on the three surfaces, i.e., AC, Hydrogel, and Hydrogel AC (Al Mashhadani *et al.* 2021) (as shown in Fig. 5). Through the exhibiting the FESEM images of AC, it was observed that small particles spread on the surface, resulting from the activation of the surface with acid, where the acid helps to activate the active sites of the surface. Also, the

FESEM Fig. 3(b) of hydrogel illustrates the occurrence of rough clusters on the surface, and this evidence confirms the roughness of the hydrogel surface (Wared & Radia 2021). In the case of Fig. 5c of the hydrogel after loading the activated carbon, it was observed that the activated carbon particles merged with the surface of the hydrogel and formed a surface containing many active sites, where this surface has high efficiency on adsorption (Shi *et al.* 2012; Radia *et al.* 2022). Figs. 5a-c illustrate images of TEM of three surfaces, i.e., AC, SA-g-(PAAc-co-AM) hydrogel and SA-g-(PAAc-co-AM)/AC hydrogel nanocomposite, where AC was observed embedded inside the SA-g-(PAAc-co-AM) hydrogel.

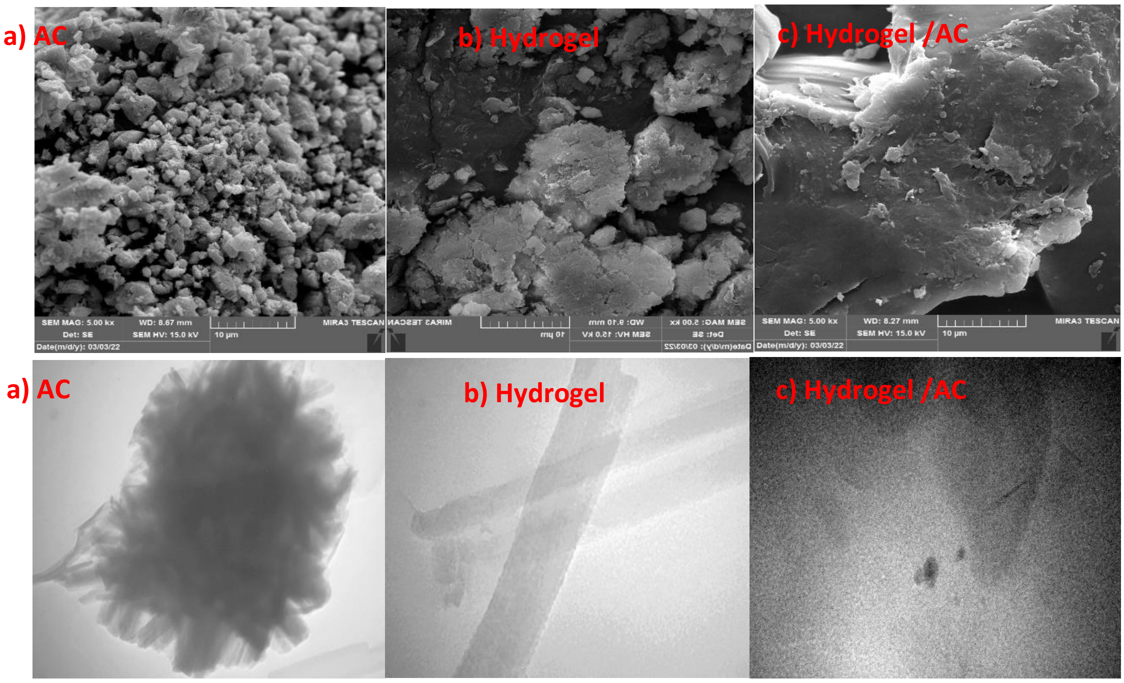

Fig. 5. FESEM and TEM images of three surface: a) Activated Carbone (AC); b) Hydrogel; and c) Hydrogel / AC.

XRD

The structural properties of hydrogel were investigated utilizing X-ray diffraction (XRD). The composition, crystal size, and spacing among the crystalline levels were all measured according to the broadband and the hydrogel non-crystalline (Chen *et al.* 2021; Alradaa & Kadam 2021), (as shown in Fig. 6).

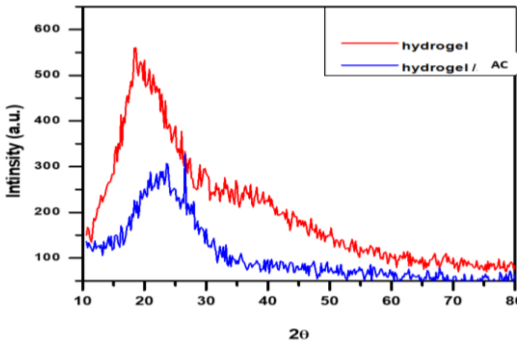

Fig. 6. XRD of hydrogel, hydrogel / AC nanocomposite.

Effect of weight

Figs. 7-9 exhibit the adsorption of the textile dyes (Brilliant Green, Methyl Violet, Malachite Green, Roadmen B, Rose Bengal) using three different weights (0.025-0.1 g) of hydrogel/AC, hydrogel and activated carbon (AC) for comparison as the surface efficiency (Wared & Radia 2021; Radia *et al.* 2022). Through the results shown, the elevation in weight, leads to the increased percentage of removal for the three surfaces. Therefore, the number of active adsorption sites upraises from which a mass adsorption enhancement can be obtained [8, 9]. Therefore, through the results, the hydrogel loaded with activated carbon about 0.1 g gave the best removal percentage for the abovementioned textile dyes (BG = 98.91, MG = 93.43, MV = 90.98, RhB = 77.55 and RB = 70.66) compared to hydrogel (BG = 88.25, MG = 81.54, MV = 77.78, RhB = 60.21 and RB = 53.23) and Activated Carbon (BG = 80.25, MG = 70.76, MV = 66.66, RhB = 56.33 and RB = 40.43). This is a clear evidence of the success in the process of loading activated carbon on the hydrogel, leading to an elevation in the surface area and an upraise in the surface efficiency (Aljeboree *et al.* 2019; Alkaim & Ajobree 2020; Gao *et al.* 2021).

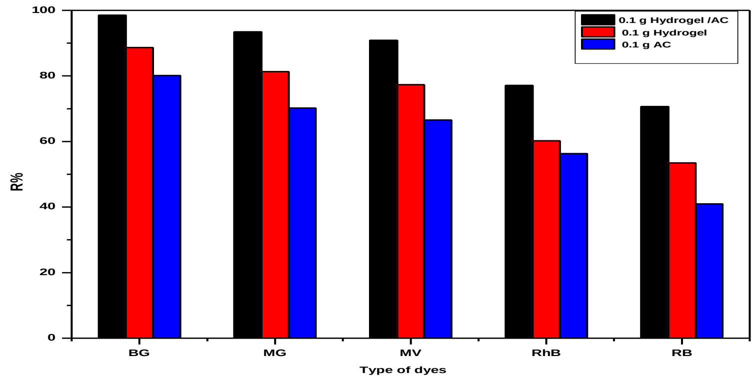

Fig. 7. Effect of 0.1 g of SA-g-(PAAc-co-AM)/AC, SA-g-(PAAc-co-AM) and Activated Carbon (AC) surfaces for removal the textile dyes (Brilliant Green, Methyl Violet, Malachite Green, Roadmen B, Rose Bengal).

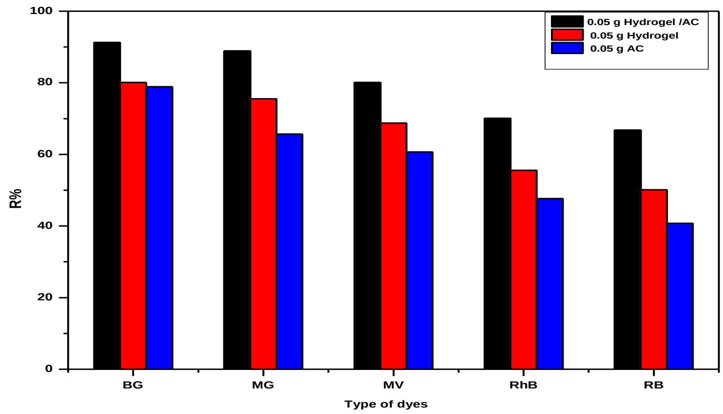

Fig. 8. Effect of 0.05 g of SA-g-(PAAc-co-AM)/AC, SA-g-(PAAc-co-AM) and Activated Carbon (AC) surfaces for removal of textile dyes (Brilliant Green, Methyl Violet, Malachite Green, Roadmen B, Rose Bengal).

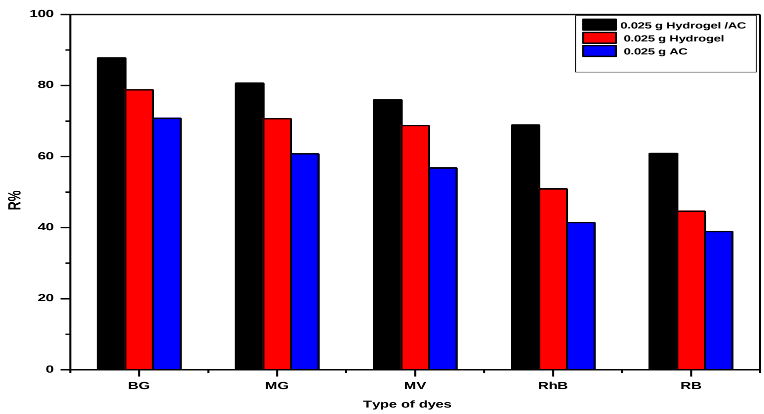

Fig. 9. Effect of weight 0.025 g of SA-g-(PAAc-co-AM)/AC, SA-g-(PAAc-co-AM), and Activated carbon (AC) surfaces for removal textile dyes (brilliant green, methyl violet, Malachite green, Roadmen B, Rose Bengal).

Comparative between different surfaces

Comparative study between AC, SA-g-(PAAc-co-AM) hydrogel, and SA-g-(PAAc-co-AM)/AC hydrogel nanocomposite, a sample of 100 mL of dyes at the concentration of 100 mg L^{-1} were used in this study, then added to a conical flask of 0.05 g from prepared AC, SA-g-(PAAc-co-AM) and SA-g-(PAAc-co-AM)/AC hydrogel nanocomposite. The better data of the E (%) for dyes were based on the increasing order: SA-g-(PAAc-co-AM)/AC > SA-g-(PAAc-co-AM) > AC (as shown in Table 1).

Dye	SA-g-(PAAc-co-AM)/AC	SA-g-(PAAc-co-AM)	AC
Brilliant Green (BG)	90.987	79.11	77.33
Malachite Green (MG)	89.12	77.23	72.22
Methyl Violet (MV)	85.44	72.33	70.11
Rose Bengal (RB)	82.23	68.22	56.55
Rhodamine B (RhB)	80.11	65.65	55.55

CONCLUSION

In this work, relied on preparing three surfaces from environmentally friendly materials, the first surface was activated carbon from orange peels, the second surface was hydrogel, and the third was hydrogel loaded on activated carbon. In addition, by comparing the efficiency of these surfaces in removal textile dyes, it was found that the hydrogel loaded with activated carbon exhibited the best results in removal textile dyes.

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