

# Synthesis and Characterization of Melamine-modified Hydrogels: The Study of Dye

# **Removal from Aqueous Solutions**

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**Abstract:** In the present study, a class of melamine-modified hydrogels were prepared for use as dye removal adsorbents from aqueous solutions. In the first step, poly (itaconic acid-*co*-maleic anhydride) and poly (vinyl acetate-*co*-maleic anhydride) were synthesized. Then the copolymers were modified by melamine to prepare the final adsorbents. The adsorbents were characterized by Fourier transform infrared spectroscopy (FT-IR). The dye removal studies were then carried out for the adsorbents and different parameters were investigated in order to reach an optimized condition for each of the adsorbents.

Keywords: Hydrogel, Melamine, Dye removal, Adsorbent

## Introduction

A three dimensional network of polymers made of natural or synthetic materials possessing high degree of flexibility due to large water content are called hydrogels. Under physiological conditions, they are able to retain a large amount of water or biological fluids and are characterized by a soft rubbery consistency similar to living tissues, making them an ideal substance for a variety of applications [1-3].

Colors are among the most dangerous chemical compounds in industrial waste water. These contaminants cause different environmental issues such as reducing the penetration potential of light and, consequently, disrupting the process of photosynthesis in water resources. Congo red color is azo color. Azo paints are one of the most important groups of artificial colors that are used in many industries because of low cost, high solubility and stability [4-6].

Treatment of water contaminated by dyes is one of the solemn environmental issues. Different procedure including biological treatment, congealing/flocculation, ultrafiltration, chemical oxidation, photocatalytic methods, membrane processes, ozone treatment, abstraction and adsorption methods are used to overcome this issue. Natural and commercial adsorbent such as activated carbons, zeolites, clays, stimulated slag, chitosan globules, cellulosic resins, polymer resins are used for this purpose [7].

Therefore, a polymer hydrogel was used to remove congeal paint from industrial waste water and effective factors such as pH, adsorbent amount, contact time and color rendering were investigated.

## **Experimental**

General

All the solvents, chemicals and reagents were purchased from Merck, Fluka and Aldrich. Concentration of the dye solutions were estimated using absorbance recorded on UV/VIS spectrophotometer model Agilent 8453 Diode Array, USA.

#### Synthesis of melamine-modified poly (vinyl acetate -co- maleic anhydride) (PVA-MAn) hydrogel

In a 100 ml two-necked round-bottom flask equipped with magnetic stirrer, reflux condenser and gas inlet and outlet, maleic anhydride (1 g, 10 mmol) and dry THF (40ml) were added fallowed by adding newly distilled vinyl acetate (0.88 g, 10 mmol). The contents were degassed with argon using capillary for 20 minutes. After that, AIBN (0.007g, 0.04mmol) was added to the flask and the contents were stirred for 8 hours at  $70^{\circ}$  c under inert atmosphere.to complete the polymerization reaction. After the mentioned time, a solution of melamine (1.286 g) in hot water (40 ml) was added to the flask and let to be stirred for additional 24 hours. The final product was filtered, washed several times with hot water and dried at  $60^{\circ}$  c.

#### Adsorption studies

Batch technique was performed to observe the effects of pH, amount of adsorbent, dye concentration, contact time. For adsorption study, 20mL of the dye solutions of desired concentration was taken into 100mL volumetric flasks and a fixed amount of *PVA-MAn* were added into it. The conical flask was then agitated on a mechanical shaker at an optimum pH achieve equilibrium. When equilibrium was thought to have been established, the adsorbent was filtered and the amount of dye adsorbed was monitored spectrophotometrically at  $\Lambda$ max 498 nm.

#### **Results and discussion**

First, PVA-Man was prepared as described in the materials and method section. To study the characterization of PVA-MAn, Fourier-transform infrared spectroscopy (FT-IR) was used. Fig. 2 show the FT-IR spectra of PVA-MAn. The spectrum of PVA-MAn has a peak at 1681 cm<sup>-1</sup> due to the overlap of carbonyl group (-C=O) amide and carboxylic acid. The broad peak at 3379 cm<sup>-1</sup> was attributed to the –OH band.

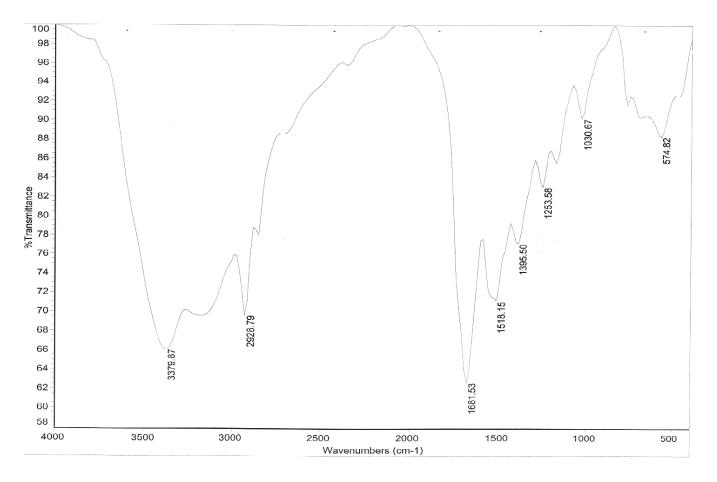


Fig. 1. FT-IR spectra of PVA-MAn

The potential of PVA-Man for removing Congo red dye was studied in this research. Also, factors affecting adsorption such as adsorbent amount, pH, contact time, and dye concentration were investigated.

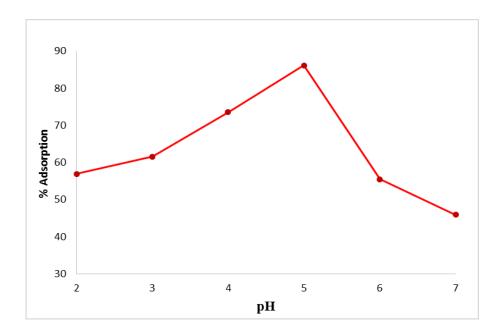


Fig. 3. Effect of pH on the dye adsorption

To determine the optimum pH for the removal of the dye Congo red, the adsorption studies were carried out over a wide pH range of the dye ranging from 2.0 to 7.0 as depicted in Fig.3.

According to the diagram, with increasing pH, the adsorption rate increases untill pH 5 and then decreases. At pH greater than 7, absorbent is also become a decomposable. The results showed that the pH 5 is optimum for removal the Congo red dye.

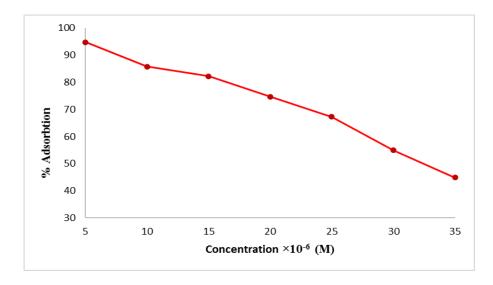


Fig. 4. Effect of dye concentration

The adsorption experiments were carried out in the concentration range of the dye ranging from 5 Mg/L to 35 Mg/L at a fixed pH 5.0 (Fig.4). The results shown that with the increase in the amount of adsorbent, the efficiency of dye removal increases (Fig. 5). But with the increase in the dye concentration, the number of available absorbent sites will be less and thus absorbance will have a decreasing trend.

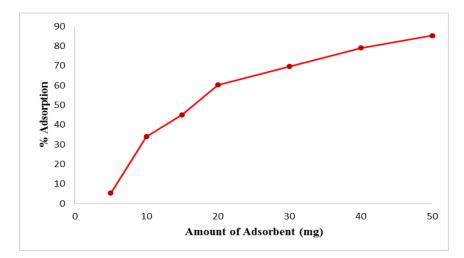


Fig. 5. Effect of adsorbent dosage.

Dye removal efficiency also increases with increasing contact time (Fig. 6). Dye removal rate by this polymeric adsorbent increases in the first minutes and decreases over time, which can be the result of decrease in concentration of Congo red dye and also color molecules occupy the adsorbent sites which reduce the number of this sites over time . In this case, the amount of adsorbed color and PVA-Man are in equilibrium.

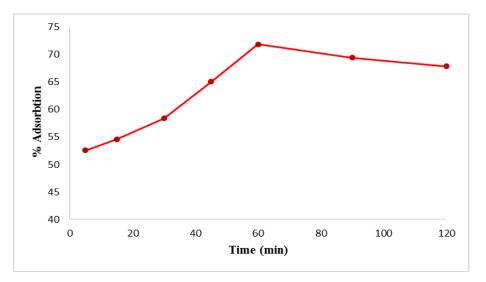


Fig. 6. Effect of time contact

#### Conclusions

In summary, in this study, a polymeric adsorbent poly (vinyl acetate-*co*-malic anhydride) modified by melamine was prepared by radical polymerization method. Also, the adsorption process of the PVA-MAn to remove hazardous azo dye Congo red from its aqueous solutions was studied. We found that the adsorption is very much dependent upon parameters like pH, contact time, amount of adsorbent and dye concentration. The highest adsorption of the Congo red can be achieved at 5.0 pH, 10 Mg/L dye concentration, 20 mg quantity of adsorbent and 60 min contact time.

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## References

- [1] Rosiak, J.M.; Yoshii, F. Nucl Instrum Meth B. 1999, 151, 56-64
- [2] El-Hefian, E.A.; Elgannoudi, E.S.; Mainal, A.; Yahaya, A.H. J. Turk. Chem. 2010, 34, 47-56.
- [3] Khan, A.; Othman, M.B.H.; Akil, K.A. J. Polym. Res. 2013, 20, 1-8.
- [4] Haspulat, B.; Gülce, A.; Gülce, H. J. hazard. Mater. 2013, 260, 518-526.
- [5] Ohtani, B. J. Photochem. Photobiol. 2010, 11, 157-178
- [6] Wu, J.; Liu, C.; Chu, K.H.; Suen, S. J. Membrane Sci. 2008, 309, 239-245.
- [7] Saad, M.; Tahir, H.; Khan, J.; Hameed, U.; Saud, A. Ultrason. Sonochem, 2017, 34, 600-608.