A comparative study to estimate the ability of three waste organic materials to remove methylene blue and Congo red Dyes from aqueous solutions

M E Al-Defiery¹  A K Al Muttairi²  A Omran³  A M Yasin⁴  and M S Kazam⁵

¹ 1 Biology Department, Collage of Science for Women, University of Babylon.
²,³ 2,3 Environmental Research and Studies Center, University of Babylon.
⁴,⁵ 4,5 Collage of Basic Education, University of Babylon.
E-Mail: al_defiery2004@yahoo.com

Abstract. The current study was done in order to find the efficiency of (spent tea leaves, orange peels, pomegranate peels) as adsorbents to remove industrial dyes; Congo red and methylene blue dyes. the results show that the three proposed adsorbents have a low to moderate ability to remove the dyes. The results show that depleted the tea leaves have a higher ability than orange peels and the pomegranate peels being the lowest in removing the methylene blue dye, while for the Congo red dye removal the orange peels were the highest followed by pomegranate peels and the lowest was the depleted tea leaves.

keyword: plant organic wastes, depleted the tea leaves, orange peels, pomegranate peels.

Introduction

During the different stages of the textile industries a tremendous amount dyes are being released. these dyes are composed of aromatic compounds that need treatment before released, otherwise, they can cause many environmental problems [1]. among these dyes is methylene blue (MB) which is usually applied for dying textile such as cotton and silk. MB is an ionic dye can cause a variety of diseases to human beans such as cyanosis, convulsions, and irritation to epidermis as well as subsequently cause nausea, vomiting and diarrhea when ingested [2].
Another dye that is important from health respect that is found in the effluents discharged to the environment is Congo red (CR) which is an anionic diazo dye that has a brownish-red crystalline solid as well as high solubility in water. It contains a benzidine base with \(-\text{NH}_2\) and \(-\text{SO}_3\) functional groups. CR can be carcinogenic since it can be metabolized to benzidine, it also can be toxic all life form and must be treated before released to the water bodies \([4,5]\).

Many techniques and methods can be used to remove pollutants from the effluents before their released to the environment, these methods can be classified generally into three major classes: biological, chemical and physical. Between these techniques adsorption is quite familiar method to be used to remove these dyes from water due to its low cost and applicability \([7]\).

In this study three types of adsorbents were used to remove MB and CR dyes which are spent tea leaves (STL); orange peels (OP) and pomegranate peels (PP). Tea is available around the globe and been consumed extensively. tea is presented in many flavor and colors and different aroma; the leaves been picked and dried to produce the dried tea. Tea contain many important chemical components such as tannins, proteins and amino acid as well as contain minerals such as Ca, Na, K, Mg, by far the most known constituent of tea is caffeine \([8]\).
Citrus plants are known around the world and there is about 1300 species. Mediterranean contain one of the largest farms for citrus plants including orange (*Citrus sinensis*). They are major source of income since they play part in the food, medicine as well as cosmetics industries. The fruit have two part generally the pulp and the peel [9].

Pomegranate (*Punica granatum*) belonging to family Lythraceae is found originally in areas of Northern Africa and the Caucasian. Pomegranate is used in great amounts in industries such as juice and food industry and that resulted in an enormous amount of Pomegranate peels as waste. The Pomegranate peels contain many important components such as enzymes, antioxidants and antimicrobial agents [10, 11]. In this study STL, OP and PP were used as adsorbent.

Many studied have been carried out in this area with different degree of success [12,13,14]. The aim of present work is to test which of the proposed low-cost adsorbents (STL, OP and PP) is a better adsorbent for removing these dyes.

**Materials and methods**

**Adsorbents and adsorbates**

The organic wastes (STL, OP and PP) were used in this study as adsorbent. All three were dried and then grinded to fine powder by using an electrical grinder without any activation and kept for later use.

Standard solution of both dyes was prepared by diluting the dry dye in D.W. the concentration used in this study were 5, 10, 15 ppm respectively with three replicates for each concentration.

**Experimental work**

The experiments were carried out as follow: three concentration (5,10,15ppm) of methylene blue and congo-red dyes were prepared and 100 ml of each concentration was kept in flask with three replicates was used in the experiments. Then 5 gm of STL, OP and PP was added to each flask with pH-6.0 and temperature was (28±2 °C). The pH value of each sample was adjusted to 7. After that the samples were kept in the shaker for 1 hour for better mixing and at 80rpm. The adsorbent was separated by using the centrifuge at 1000 rpm, the resulted supernatant was taken and
the absorbance of the dyes were measured in the UV-visible spectrophotometer (Shanghai, Ykm1005068), for methylene blue with $\lambda_{max}$ 663nm and for the congo-red dye with $\lambda_{max}$ 497 nm. Removal efficiency ($R\%$) was calculated by using the following equation [15]:

$$R\% = \frac{\text{Initial dye concentration} - \text{Final dye concentration}}{\text{Initial dye concentration}} \times 100$$

**Results and discussion**

This study was carried out in order which of the three adsorbents (STL, OP, PP) was better in removing the methylene blue and Congo-red dyes. These three adsorbents are considered as waste and generally produce in large amount so putting them in use will provide both economical and practical advance.

For the removal of methylene blue dye removal, all three adsorbents show a reverse correlation between their removal efficiency ($R\%$) and the increase in the dye concentration, and that can be explain by the fact that as the concentration of the dye increase the site of adsorption available on the adsorbent will be saturated which led to decrease the removal efficiency of the dye [16]. The three adsorbents used in this study show moderate ability to remove MB dye. STL show the highest ability to remove MB dye followed by the orange peels and the lowest was pomegranate peels with $R\%$ 51, 47 and 26 at dye concentration of 5 ppm respectively as illustrated in Figures 3-5 respectively. The results of this study agree with the findings of [14] which find that the equilibrium removal of MB by using the spent tea leaves was decreased from 95.2% to 62.1% as the initial MB concentration increased from (50-500) mg/l. the study findings agrees with the findings of [17] which found that Spent tea leaves were more effective in removing MB dye followed by orange peels but with different percent of removal, on the other hand the results of this study disagree with the results of [18] which state that the removal capacity of MB dye increase with the increase of initial dye concentration.
Figure 3: Removal percentage of different Methylene blue concentrations by spent the tea leaves.

Figure 4: Removal percentage of different methylene blue concentrations by orange peels.

Figure 5: Removal percentage of different methylene blue concentrations by pomegranate peels.
While for the removal of CR dye, as shown in figures 6-8 there was also a reverse correlation between the R% and the initial dye concentrations. The result show that the OP was the most efficient in removal the dye followed by STL and the lowest was PP with R% 61, 53, 35 respectively at concentration 5 ppm of the dye. The ability of these organic waste for the adsorption of pollutants may be attributed to their cellulose content which has hydroxyl group which act as active site for adsorption [19]. The results of this study agree with the findings of [20] which state that OP efficiency for removing Congo red dye was decrease as the initial dye concentration increase but with much lower removal efficiency percent for the OP used in this study. also, the results for STL consent with findings of findings of [21].

Figure 6: Removal percentage of different Congo red concentrations by Orange peels.

Figure 7: Removal percentage of different Congo red concentrations by deplete the tea Leaves
Figure 8: Removal percentage of different Congo red concentrations by pomegranate peels

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