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Adsorptive removal of Erythrosine dye from aqueous solutions using used cigarette filter ash loaded with Al/ Fe oxides

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ABSTRACT

Dye contamination in water is a serious environmental problem around the world. Erythrosine is a water-soluble xanthenes class of dye that widely used as colorant in many industries. Used cigarette filter ash loaded with Al/ Fe oxides was synthesized by a low-cost, simple, and environmentally benign procedure. The adsorbent was characterized by several methods including energy dispersive X-ray spectroscopy (EDX), scanning electron microscopy (SEM) and Brunauer Emmet and Teller (BET) analysis. Then, the potential of used cigarette filter ash loaded with Al/ Fe oxides was investigated for adsorption of erythrosine. The effect of different parameters including contact time, pH, adsorbent dosage and initial dye concentration on the removal yield was studied. The experimental data were fitted well with the Langmuir isotherm model. Maximum monolayer adsorption capacity based on Langmuir isotherm is 32.68 mg g^{-1}. The process for purifying water treatment presented here is clean and safe. Therefore, this adsorbent was considered to be applicable for managing water pollution caused by erythrosine.

Keywords: Adsorption; Erythrosine; Used cigarette filter ash; Al/ Fe oxides

INTRODUCTION

Dye contamination in water is a serious environmental problem around the world. Erythrosine is a water-soluble xanthenes class of dye. It is widely used as colorant in food, textile, drug and cosmetics. In large doses it causes various types of allergies, thyroid activities, carcinogenicity, DNA damage behavior, neurotoxicity and xenoestrogen nature in the humans and animals. The photochemical and biochemical degradation of the erythrosine is not recommended due to formation of toxic by-products [1]. Adsorption of dyes using different absorbents is widely studied. Adsorption was established as an important and economically practical treatment technology for removing the dyes from water and wastewater. Activated carbon is usually used adsorbent for the removal of erythrosine from aqueous solution. Despite the abundance applications of activated carbon, its uses are sometimes limited due to its high cost and also for loss during its re-formation [2-4]. Therefore, the researchers are on the

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search for new low-cost substitute adsorbents for the water pollution control, especially, where cost acts an important role. Many efforts have been done towards the development of another adsorbents that are effective and low-cost. They can be produced from a wide diversity of raw materials, which are abundant and have high carbon and low inorganic content. Owing to the low cost and high accessibility of these materials, it is not essential to have complex regeneration processes. Such low cost adsorption methods have attracted many researchers. Often, the adsorption capabilities of such adsorbents are not large, therefore the study and investigation of more and more new adsorbents are still under development. Several adsorbents have been used for removal of erythrosine: bottom ash and de-oiled soya [1], hen feathers [5], activated carbon prepared from bael tree bark [6], de-oiled mustard [7], modified palladium nanoparticles [8], lemon citrus peel active carbon [9] and ZnS nanotubes [10]. In recent years, due to economic problems, creating a cheap and efficient alternative methods of wastewater treatment instead of expensive and inefficient methods is of great importance. Ash due to the low cost of production is a good alternative to activated carbon [11-14]. Ashes can be produced from a wide range of carbon materials such as wood, coal, shell, walnut shell, fruit stones, agricultural waste, etc. [15]. Disposed cigarette butts are one of the biggest solid wastes produced worldwide each year. They are known to be the most-collected waste item on the beach [16]. They pose significant environmental contamination. Used cigarette filters are toxic and hazardous waste [17]. This contamination by cigarette butts thrown out of the windows of moving cars, dropped on sidewalks and left on the beach is serious when they eventually find their ways to the street drains and then to the streams, rivers and oceans [18]. Nearly all cigarette filters produced are made up of cellulose acetate which is a plastic product. Each filter consists of around 12,000 cellulose acetate fibers [19]. These fibers contain delustrant titanium dioxide. A plasticizer, triacetin (glycerol triacetate) is also added to the fiber as a fiber binder [20]. Cellulose acetate is one of the most important cellulose derivatives [21]. It has a molecular formula of $C_{76}H_{114}O_{49}$ with an average molecular weight of 1,811.68896 g/mol. It contains a high degree of carbon atoms and can potentially be nominated as an initial carbonaceous raw material for the production of porous carbon [22]. In this context, we became interested to investigate the capability of the used cigarette filters ash loaded with Al/Fe oxides as a low-cost adsorbent for removal of erythrosine from aqueous solution and also to study the adsorption mechanism of erythrosine onto this adsorbent. For this purpose, a set of batch adsorption experiments (contact time, pH, adsorbent dosage and initial erythrosine concentration, on the erythrosine removal using this adsorbent) were carried out at optimum conditions. The characterization of the adsorbent was described by EDX, BET and SEM analyses.

EXPERIMENTAL

Materials

Erythrosine (Ery, CI 45,430) (Fig. 1), was purchased from Sigma–Aldrich. Erythrosine is an anionic dye with IUPAC name as Disodium 2-(2, 4, 5, 7-tetraiodo-6- oxido-3-oxo-3H-xanthen-9-yl) benzoate (Figure 1). For the erythrosine, λ_{max} is 526 nm. A 1000 mg/L stock solution of the dye was prepared in deionized water. All working solutions were made by consecutive diluting method with deionized water. Deionized water was prepared using a Millipore Milli-Q

(Bedford, MA) water purification system. All reagents ($FeCl_3$, $Al(NO_3)_3$, NaOH and HNO_3) used in the study were of analytical grade and purchased from Aldrich. Surface morphology and particle size were investigated by a Hitachi S-4800 SEM instrument. The specific surface (S_{BET}) of adsorbent is determined by a Micrometrics apparatus (Gemini 2375) by adsorption of nitrogen at 77 K according to the traditional method of Brunauer Emmet and Teller or BET. An energy dispersive X-ray XL30 philips microscope was used in the EDX analyses. All the erythrosine concentrations were measured spectrophotometrically (Hach DR 6000 UV–visible Spectrophotometer) at the maximum absorption wavelength of 526 nm, which corresponds to the maximum absorption peak of the erythrosine.

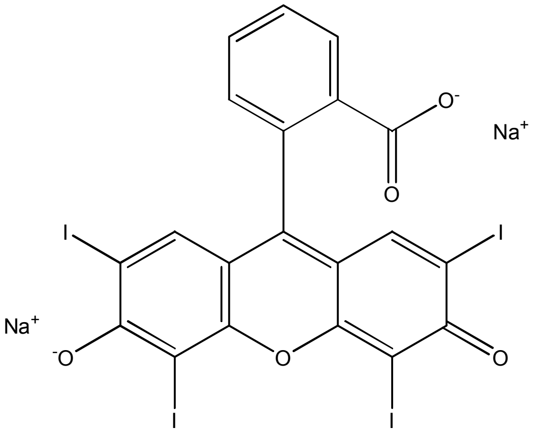

Fig. 1. Chemical structure of erythrosine.

Preparation of used cigarette filters ash (UCFA)

The collected Used cigarette filters were unwrapped, washed and dried in an air oven at 80 °C for 24 h and then were carbonized in a furnace at 900 ºC at heating rate of 10 °C/min for 2 h. The method for producing carbon materials are similar to other studies also have been used [19, 22, 23].

Synthesis of used cigarette filters ash loaded with Al/Fe oxides (UCFA/Al-Fe)

Al and Fe oxides were prepared by the chemical co-precipitation method [24]. In a typical procedure, 20 mL ferric chloride (0.4 M) and 40 mL aluminum nitrate (0.4 M) were mixed in hydrochloric acid (0.01 M). Used cigarette filters ash (5.0 g) and the prepared solution (100 mL) were added to a beaker on a heated magnetic stirrer at 60 °C and 300 rpm. Sodium hydroxide solution (2.0 M) was added dropwise to regulate the solution pH to 5.0, with vigorous stirring for 30 min. The resultant suspension was then filtered and dried at 110 °C for 3 h. Then the loaded used cigarette filters ash was washed by distilled water several times and dried at 100 °C for 5 h.

Adsorption experiments

Batch adsorption of erythrosine onto the adsorbent (UCFA/Al-Fe) was investigated in aqueous solutions under various operating conditions viz. pH 2 – 12, at a temperature of 298 K, for an initial erythrosine concentration of 5 mg L^{-1}. About 0.10 g adsorbent was added to 50 mL of erythrosine solution (2 g L^{-1}). Then the mixture was agitated on a shaker at 250 rpm. The initial pH values of the dye solutions were adjusted from 2 to 12 with 0.1 mol L^{-1} HNO_3 or 0.1 mol L^{-1} NaOH solutions using a pH meter. After equilibrium, the samples were centrifuged and the adsorbent (UCFA/Al-Fe) was removed from the solution removed from the solution. The erythrosine concentration in the supernatant was measured by UV-visible spectrometer. The effects of several parameters, such as contact time, initial concentration, pH and adsorbent dose on extent of adsorption of erythrosine were investigated.

The erythrosine removal percentage was calculated as Eq. (1):

$$\%\text{Removal} = \frac{C_0 - C_t}{C_0} \times 100 \quad (1)$$

where C_0 and C_t (mg L^{-1}) are the concentration of erythrosine in the solution at initial and equilibrium time, respectively.

The amount of erythrosine adsorbed (Q_e) was calculated using the Eq. (2):

$$Q_e = \frac{(C_0 - C_e)V}{m} \quad (2)$$

where C_0 and C_e are the initial and equilibrium concentrations of erythrosine (mg L^{-1}), m is the mass of adsorbent (g), and V is the volume of solution (L).

Adsorption Isotherms

Adsorption isotherms were obtained by using 0.20 g of adsorbent and 50 mL of erythrosine solution with different concentrations (5-100 mg L^{-1}) at 298 K. These solutions were buffered at an optimum pH (pH = 7) for adsorption and agitated on a shaker at 250 rpm until they reached adsorption equilibrium (30 min). The quantity of erythrosine adsorbed was derived from the concentration change.

RESULTS AND DISCUSSION

Characterization of used cigarette filters ash loaded with Al/Fe oxides (UCFA/Al-Fe)

The particle size and morphology of adsorbent was investigated by SEM technique. The SEM photograph of sample (Figure 2) shows that average size of UCFA/Al-Fe is approximately less than 10 µm. To further confirm the effective loading of the Fe and Al oxides onto the used cigarette filter ash, EDX spectra were investigated (Figure 3). Corresponding signal peaks were detected on the surface of the adsorbent samples. As expected, the main element found in the original used cigarette filter ash was carbon (Figure. 3a). In contrast, new Fe and Al peaks were found in Al–Fe loaded samples (Figure 3b). Such observation is direct evidence of Fe and Al loading on the surface of used cigarette filter ash.

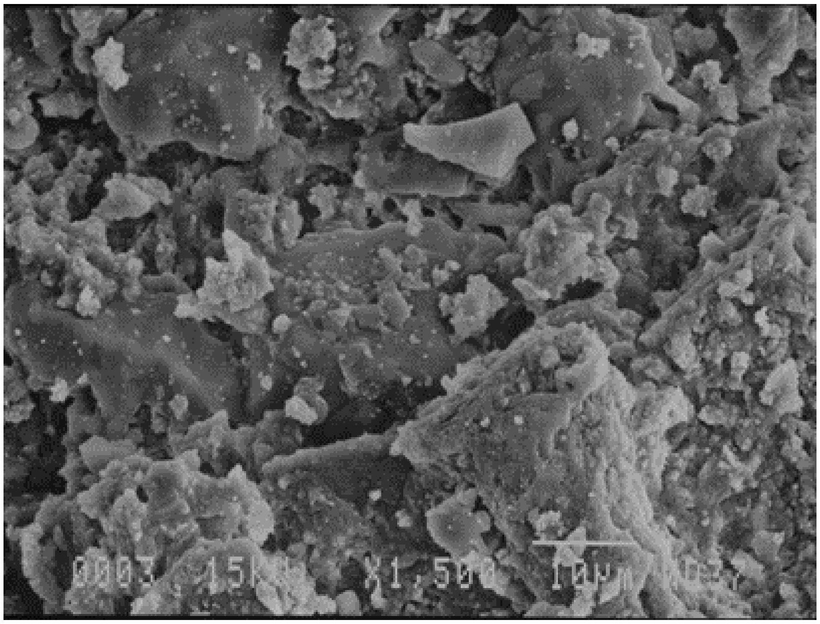

Fig. 2. The SEM image of UCFA/Al-Fe.

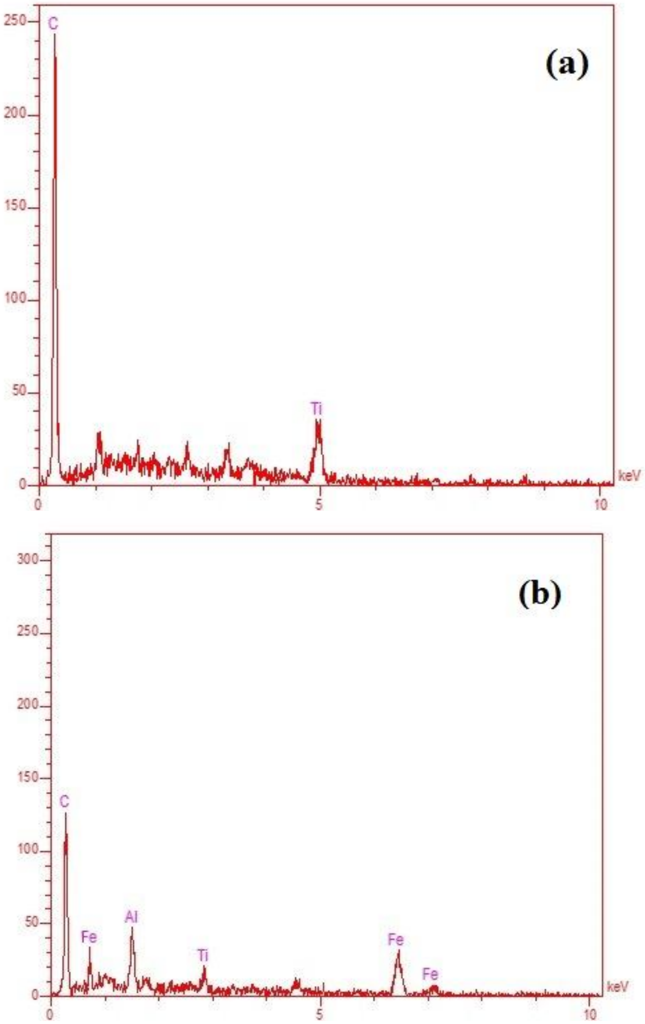

Fig. 3. EDX spectra of the original used cigarette filter ash (a) and Al – Fe loaded used cigarette filter ash (b).

Some properties of the UCFA/Al-Fe are presented in Table 1. The UCFA/Al-Fe has a surface area about 854.01 m^2/g and a porous volume equal to 0.726 cm^3/g.

Table 1. Some properties of UCFA/Al-Fe

Parameter	Value
Specific surface area (m^2/g)	854.01
Porous volume (cm^3/g)	0.726
Water content (%)	Trace

Adsorption and removal of erythrosine from aqueous solution

Effect of contact time

The effect of contact time on the amount of erythrosine adsorbed was studied at 5 mg L^{-1} initial concentration of dye. It could be observed from Figure 4 that with the increase in contact time, the removal percentage also increased. Minimum adsorption was 17.32 % for time 5 minutes to maximum adsorption value 98.5 % for the time 30 minutes. The adsorption characteristic indicated a rapid uptake of the dye. The adsorption rate, however, reached to a constant value with an enhancement in contact time because of all available sites were covered, and no active site was present for adsorbing.

Effect of pH

The acidity of the aqueous solution applies a considerable effect on the adsorption process owing to it can alter the solution chemistry of contaminants and the state of functional groups on the surface of adsorbents [25, 26]. The effect of solution pH on erythrosine adsorption was studied at pH 2 – 12 at 298 K. As shown in Figure 5, the general observation is that as pH increases, dye adsorption decreases and the best pH for adsorption is at range 2-7. At pH range 2–7, the surface of adsorbent and adsorbate molecules are differently charged making adsorption favorable over this range, while at the basic range 7–12 dye molecules ionize to have a negative charge making adsorption process less favorable over this range as shown in Figure 5. The reduction in erythrosine removal at alkaline pH ranges could be attributed to the competition of OH^- ions for the adsorption sites. Since excessively strong acidity was not appropriate for the practical handling of water samples, neutral solution (about pH 7) was selected for further experiments.

Effect of adsorbent dosage

The effect of change in the adsorbent amount on the process adsorption of erythrosine was investigated, with different adsorbent dose in the range of 0.05 – 1.0 g. The results obtained are shown in Figure 6. From Figure 6, it is considered that as the adsorbent dose enhances, the percentage removal also increases, until it approaches a saturation point, where the enhancement in adsorbent dose does not alter the percentage of removal. An increase in adsorption rate with adsorbent quantity can be ascribed to increased surface area and the availability of more adsorption sites. The best removal of erythrosine is at about 98.4 %, using an adsorbent dosage of 0.20 g in 50 mL of 5 mg L^{-1} erythrosine solution.

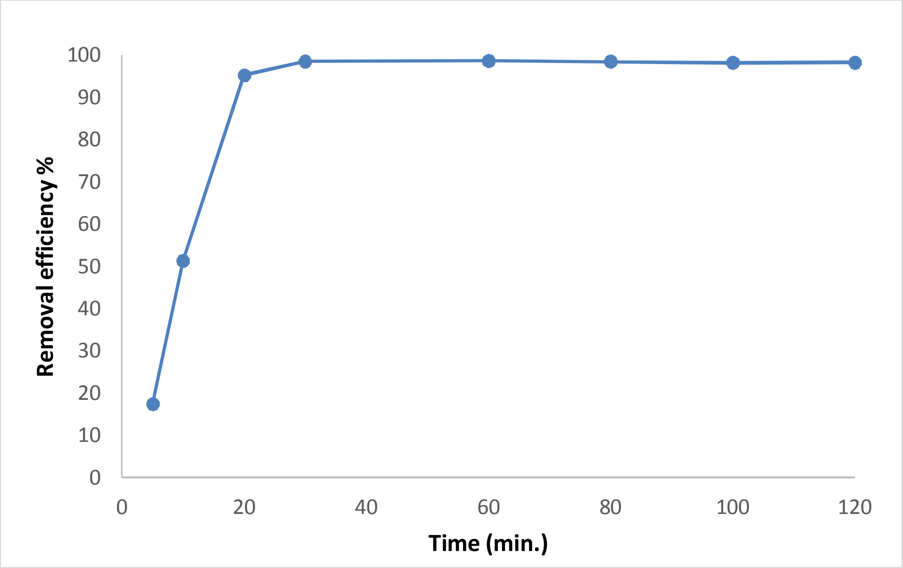

Fig. 4. Effect of contact time on erythrosine removal (initial concentration = 5 mg L^{-1}, pH native, adsorbent dose = 2 g L^{-1} and T = 298 K)

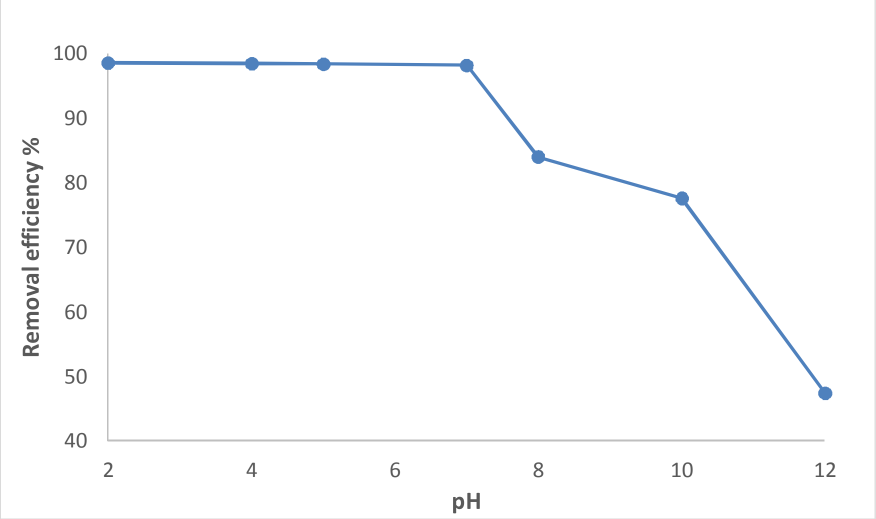

Fig. 5. Effect of pH on erythrosine removal at different pH values (initial concentration = 5 mg/L, adsorbent dose = 2 g L^{-1}, contact time = 30 min. and T = 298 K).

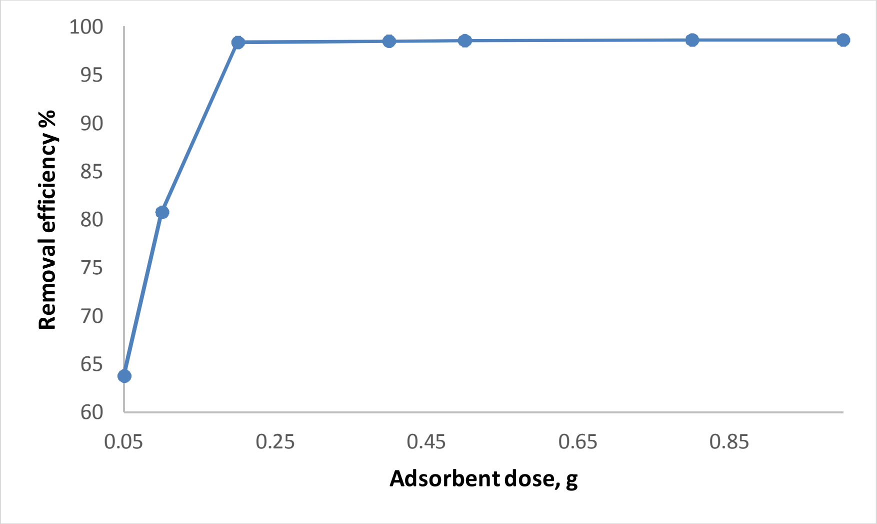

Fig. 6. The effect of adsorbent dosage on the removal percentage of erythrosine (initial concentration = 5 mg L^{-1}, pH = 7, contact time = 30 min. and T = 298 K).

Effect of initial erythrosine concentration

Batch adsorption experiments were performed at different initial dye concentrations (5, 10, 25, 50, 75 and 100 mg L^{-1}), while other experimental parameters were constant. Figure 7 shows that adsorption capacity of erythrosine increases, but the removal percent (%R) of erythrosine decreases with the increase in initial concentration, indicating that the adsorption of dye onto UCFA/Al-Fe is highly related to initial dye concentration. This observation can be described considering the fact that by increasing the initial erythrosine concentration, the more dye molecules are available, while the amount of active sites on adsorbent is constant which leads to decrease %R.

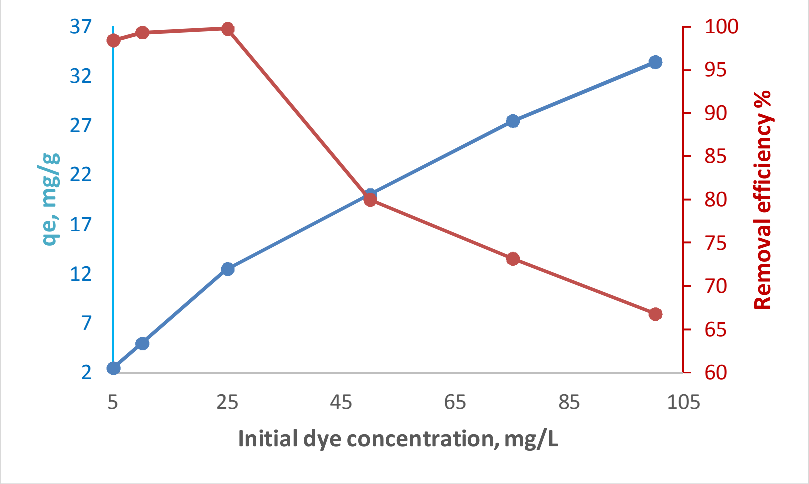

Fig. 7. Effect of initial dye concentration on the removal of erythrosine (adsorbent dose = 4 g L^{-1}, pH = 7, contact time = 30 min. and T = 298 K).

As it can see the LUMO orbital

Adsorption isotherms

Isotherms study can explain how an adsorbate interacts with adsorbent. The experimental data were corresponded by Langmuir, Freundlich and Dubinin–Radushkevich models as shown in Table 2. Langmuir isotherm model, which defines a monolayer adsorption, is given in Eq. (3):

$$\frac{C_e}{q_e} = \frac{1}{K_L q_m} + \frac{C_e}{q_m} \qquad (3)$$

where q_e = the amount of erythrosine adsorbed per unit mass at equilibrium (mg g^{-1});

q_m = the maximum amount of dye that can be adsorbed per unit mass adsorbent (mg g^{-1});

C_e = concentration of dye (in the solution at equilibrium (mg L^{-1});

K_L = adsorption equilibrium constant.

A plot of $\frac{C_e}{q_e}$ versus C_e gives a straight line, with a slope of $\frac{1}{q_m}$ and intercept $\frac{1}{K_L q_m}$.

The main characteristics of the Langmuir isotherm can be expressed in terms of a dimensionless constant separation factor R_L that is given by Eq. (4) [27]:

$$R_L = \frac{1}{1+K_L C_0} \qquad (4)$$

where C_0 is the highest initial concentration of adsorbate (mgL^{-1}), and K_L (L mg^{-1}) is Langmuir constant. The value of R_L shows the shape of the isotherm to be either unfavorable ($R_L > 1$), linear ($R_L = 1$), favorable ($0 < R_L < 1$), or irreversible ($R_L = 0$). The R_L values between 0 and 1 indicate favorable adsorption. In this study, value of R_L is 0.270 shows the favorable adsorption between UCFA/Al-Fe and erythrosine.

Freundlich isotherm is expressed by Eq. (5). This isotherm model defines a heterogeneous adsorption with different surface energy sites and supposes the change of uptake with exponential distribution of adsorption sites and energies [28-30].

$$\log q_e = \log K_F + \frac{1}{n} \log C_e \qquad (5)$$

where C_e (mg L^{-1}) and q_e (mg g^{-1}) are the equilibrium concentration of adsorbent in the solution and the amount of adsorbent adsorbed at equilibrium respectively; K_F

($mg^{1-(1/n)}$ $L^{1/n}$ g^{-1}) and *n* are the Freundlich constant which indicate the adsorption capacity for the adsorbent and adsorption intensity, respectively.

A graph of log q_e versus log C_e provides a straight line with slope *1/n* and intercept log K_F. The value of 1/n mentions the adsorption intensity and the type of isotherm to be favorable (0.1 < 1/n < 0.5) or unfavorable (1/n > 2). The Freundlich parameter, 1/n, is related to the adsorption intensity of the adsorbent. When 0.1 < 1/n ≤ 0.5, the adsorption of the adsorbate is easy; when 0.5 < 1/n ≤ 1, the adsorption process is difficult; when 1/n > 1, adsorption takes place quite difficult [31, 32]. In our study, the value of 1/n (0.285) shows the favorable adsorption of dye on UCFA/Al-Fe.

In order to discern between physical and chemical adsorption, the sorption data were analyzed using Dubinin–Radushkevich (D-R) equation, which is given by the Eq. (6):

$$lnq_e = lnq_m - \beta\varepsilon^2 \quad (6)$$

where β is a constant related to the mean energy of adsorption ($mol^2\ kJ^{-2}$), q_m is the maximum adsorption capacity of erythrosine ($mg\ g^{-1}$), ε is the Polanyi potential given by Eq. (7):

$$\varepsilon = RTln(1+\frac{1}{C_e}) \quad (7)$$

where R is the gas constant (8.314 J mol^{-1} K^{-1}) and T is the temperature (K). By plotting ln q_e versus ε^2 with experimental data, a straight line is obtained. From the intercept and slope, the values of q_m and β are determined. With the value of β, the mean energy E, which is the free energy transfer of one mole of solute from infinity to the surface of adsorbent, can be obtained by the Eq. (8):

$$E = \frac{1}{\sqrt{2\beta}} \quad (8)$$

for E < 8 kJ mol^{-1}, the adsorption process might be performed physically, while chemical adsorption when E > 8 kJ mol^{-1} [33].

All the parameters are listed in Table 2. From Table 2, in which the Langmuir, Freundlich, D–R isotherm constants for the adsorption of erythrosine are summarized, it can be derived from R^2 that the Langmuir model matched the experimental data better than Freundlich and D–R models. Moreover, it is clear that the adsorption of erythrosine by UCFA/Al-Fe may be explained as physical adsorption process for the value of E is 4.1 kJ.

The adsorption capacity is a significant parameter which determines the performance of an adsorbent. Table 3 compares the maximum adsorption capacity of UCFA/Al-Fe for erythrosine adsorption with that of other adsorbents in the literature.

Table 2 Langmuir, Freundlich, D–R isotherm constants for the adsorption of erythrosine onto UCFA/Al-Fe

Langmuir

q_m ($mg\ g^{-1}$)	K_L	R_L	R^2
32.68	0.552	0.270	0.962

Freundlich

1/n	K_F	R^2
0.285	11.380	0.728

Dubinin–Radushkevich (D–R)

q_m ($mg\ g^{-1}$)	β ($mol^2\ kJ^{-2}$)	R^2	E ($kJ\ mol^{-1}$)
25.270	3×10^{-8}	0.652	4.100

Table 3 Maximum adsorption capacity of different adsorbents for erythrosine removal

Adsorbents	q_m (mg/g)	References
De-oiled soya	9.52	[1]
Hen feathers	15.43	[5]
Activated carbon from Bael Tree Bark	576.19	[6]
Modified palladium nanoparticles	38.76	[8]
Lemon citrus peel active carbon	296.00	[9]
Montmorillonite	578.03	[34]
UCFA/Al-Fe	32.68	This study

CONCLUSION

Used cigarette filters ash loaded with Al/Fe oxides was used in adsorption of erythrosine from aqueous systems and the maximum erythrosine adsorption occurred in the pH 7 with maximum adsorption capacity of 32.68 mg g^{-1} at 298 K. The adsorption isotherm fitted the Langmuir model well. The process of water treatment described here is clean and safe using the adsorbent. Thus, this adsorbent was found to be useful and valuable for controlling water pollution due to erythrosine dye.

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