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ORIGINAL ARTICLE

Comparing the performance of granular coral limestone and Leca in adsorbing Acid Cyanine 5R from aqueous solution

Reza Shokoohi [a], Mohammad Taghi Samadi [a], Mohammad Reza Samarghandi [a], Mohammad Ahmadian [b], Kamal Karimaian [a], Ali Poormohammadi [a,*]

[a] *Department of Environmental Health Engineering, Faculty of Health, Hamadan University of Medical Sciences, Hamadan, Iran*
[b] *Department of Environmental Health Engineering, Faculty of Health, Bam University of Medical Sciences, Bam, Iran*

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Abstract The effect of granular coral limestone and Leca as adsorbents for removing Acid Cyanine 5R (AC5R) from aqueous solution was studied. The optimum pH and adsorbent particles size in both adsorbents were determined to be 3 and 297 μm, respectively. The optimum dosages of coral limestone and granular Leca were 0.150 and 0.145 g/mg of dye, respectively. Also, results have shown that the adsorption efficiency by both coral limestone and Leca increased with the decreasing adsorbent particles size. Moreover, under similar conditions, the maximum removal efficiency by granular coral limestone and Leca was 94% and 88%, respectively. The results revealed that the performance of granular coral limestone was better in AC5R removal than that of Leca granulated under such condition. In total, granular coral limestone and Leca act as suitable adsorbents for removing dye pollutants from an aqueous solution.

1. Introduction

Various industries such as cosmetics manufacturing industries, dyeing industries, paper and paperboard manufacturing industries, and textile industries produce colored effluents (Jayanthy et al., 2014). Dyes have complex molecular structures and are often toxic, carcinogenic (production of amine groups in anaerobic decomposition), non-biodegradable and sustainable (Asgher et al., 2013; Helmes et al., 1984). When dye effluents enter the environment, especially water resources, they disturb the esthetic aspects of the environment, cause eutrophication phenomenon in surface water and disturb the ecology of acceptor water (Ahmadian et al., 2012). The majority of dyes are resistant to heat and light. Moreover, the resistance of dyes to degradation causes them not to be removed by conventional wastewater treatment systems. Different methods have been examined to decolorize color

* Corresponding author. Tel.: +98 831 4216143.
E-mail address: apoormohammadi000@yahoo.com (A. Poormohammadi).
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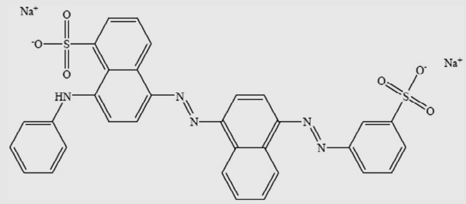
Table 1 Characteristics of AC5R dye.

Chemical structure	
Symbol	AC5R
Molecular formula	$C_{32}H_{21}N_5Na_2O_6S_2$
Molecular weight (g/mol)	681.6
Maximum absorption wavelength (nm)	494
Type of dye	Azo dye
CAS No.	3351-05-1
CB number	CB3250534

wastewater. Some of these methods include: coagulation, flocculation, chemical oxidation, electrochemical treatment, ion exchange, advanced oxidation, enzyme catalysis, adsorption, and making use of photo-catalysts. Of them, adsorption is one of the highly used processes. In the adsorption process, the main objective is to transfer contaminants from the liquid matrix to the solid matrix and to remove them completely (Magdalena et al., 2011). Activated carbon is one of the most effective adsorbents in removing dyes from aqueous solutions. Since it is expensive and costly to restore, researchers have always tried to find new adsorbents in this field (Ghosh and Bhattacharyya, 2002). In this regard, a lot of research has been carried out to apply and use cheaper adsorbents, which are easier to use. Some of these studies include application of kaolinite ore (Ghosh and Bhattacharyya, 2002), perlite (Arh-Hwang and Shin-Ming, 2009), bentonite (Gulgonu, 2012), rice bran (Guo et al., 2003), porous minerals (Ozdemir et al., 2004), and many other materials to remove various dye contaminants from water and wastewater. Coral has a limestone structure with abundant porosity and is incompressible, non-biodegradable, and resistant to heat. Owing to its abundance in Iran's water, its natural structure and lack of environmental impact in the case of limited use, it can be used as an adsorbent in industrial wastewater treatment (Ohki et al., 1996). Leca (Light Expanded Clay Aggregate) is produced from the expansion of a certain type of clay in rotating kilns in temperature of about 1200 °C. Leca has almost round particles with coarse and rough surfaces (Leca Co., 2006). Since Leca is light, incompressible against constant pressure, non-biodegradable, is a sound and temperature insulator, has a natural pH, is produced in Iran, and is cheaper than other industrial absorbents, it can be used as a convenient, inexpensive, and environmentally friendly adsorbent for removing dyes (Nkansah et al., 2012). According to what was said above, the main objective of this study is to evaluate and compare normal adsorbents of limestone coral and Leca in removing AC5R dye from aqueous solutions. Efficiency of experimental adsorbents was studied after changing pH, contact time, initial dye concentration and the amount of adsorbent; then, Freundlich and Langmuir isotherm models as well as pseudo first and pseudo second order models were examined to evaluate the equilibrium constant of the reaction and removal efficiency.

2. Materials and methods

2.1. The preparation of dye

AC5R dye used in this research was a product of Alvan Sabet Company in Hamadan. Properties of AC5R dye used in this research are presented in Table 1. To prepare a sample, AC5R with a purity degree of 98% was used. This azo dye is widely used in textile companies in Iran. All samples were taken in laboratory temperature using ion free water.

2.2. The preparation of adsorbents

Leca (3–10 mm in size and density of 380 kg/m^3) was bought from Leca Boton Manufacturing Company in Qom. Limestone coral was prepared from limestone substrata of Kish Island. To prepare granules of studied adsorbents, samples were first crushed manually. Then, they were washed with ion free water to remove dust and were placed in the oven for 12 h in 105 °C to remove their extra water and humidity (Ozdemir et al., 2004). After they were dried, they were kept in containers in the laboratory. To assess the chemical structure and to determine type of phases forming adsorbents qualitatively, X-ray fluorescence and X-ray diffraction tests were used.

2.3. The standard curve

To evaluate concentration of the studied dye, VIS–UV Spectrophotometer (model Shimadzu UV-1700), which was made in Japan, was used. Dominant wavelength was studied in the range 400–700 nm and its level was λ_{max} = 494 nm. Standard curve was plotted at this wavelength. That is, 10 certain concentrations of the studied dye were prepared; after absorbance level was read at maximum absorbance wavelength, its standard curve was plotted and dye concentration was determined by this curve.

2.4. Batch experiments

All experiments were carried in batch condition. At the end of each run and before measurement of the residual dye, all samples were centrifuged for 10 min using a centrifuge (Sigma-301, made in Germany) with a speed of 3000 rpm and then were passed through 0.45 μm filter paper. To increase the accuracy, all experiments were done in a triplicate basis. All reported results in this paper are the mean of three measurements. The effect of some parameters such as contact time, pH, adsorbent dosage, initial dye concentration and adsorbents particles size were investigated on the adsorption efficiency by both coral limestone and Leca.

2.5. Effect of contact time

To study the effect of contact time on adsorption process, different contact time in the range 0–180 min were investigated under selected conditions (dye concentration = 100 mg/dm^3, adsorbent dosage = 10 g/dm^3, particles size = 595 μm and pH = 7).

2.6. Effect of pH

To study the effect of pH on adsorption process, various pH values in the range 2–12 were tested under similar conditions (dye concentration = 100 mg/dm^3, adsorbent dosage = 10 g/dm^3, particles size = 595 μm and contact time = 180 min). The pH of the solutions was adjusted using sulfuric acid and caustic soda. Finally, in this part of the study, optimum pH was chosen.

2.7. Effect of adsorbent dosage

In the next step, the effect of adsorbent dosage on the adsorption efficiency was assessed in the range 1–10 g/dm^3 under similar conditions (pH = 3, dye concentration = 100 mg/dm^3, and particles size = 595 μm). In this step optimum adsorbent dosage was determined.

2.8. Effect of initial dye concentration

In order to determine the influence of initial dye concentration on the adsorption efficiency, various concentrations of dye (50, 100 and 150 mg/dm^3) were tested under same conditions (pH = 3, adsorbent dosage = 10 g/dm^3 and particles size = 595 μm).

2.9. The effect of adsorbents particles size

To evaluate the effect of the adsorbent particles size on dye adsorption, experiments were carried out in three different sizes of both absorbents (2000, 595 and 297 μm) under similar conditions (pH = 3, adsorbent dosage = 10 mg/dm^3 and dye concentration = 100 mg/dm^3).

2.10. Kinetic study

The four most common types of kinetics were examined to study the kinetic adsorption. Table 2 shows the simplified kinetic models of pseudo first order, pseudo second order, Elovich and intraparticle diffusion to analyze adsorption kinetic onto the granular coral limestone and Leca.

2.11. Isotherm study

In this part of study to describe the dye uptake capacity and its adsorption behavior onto granular coral limestone and granular Leca, isotherm data obtained were analyzed by four isotherm models including Freundlich, Langmuir-1, Langmuir-2, Langmuir-3 and Langmuir-4. In this step, the most commonly isotherms models were used for the explanation of the obtained data. The equations and linear forms of isotherms models are shown in Table 3 (Malakootian et al., 2011; Shokouhi et al., 2012).

Table 3 Equations and linear forms of isotherms.

Isotherms	Equation	Linear form
Freundlich	$q_e = K_f C_e^{1/n}$	$\log q_e = \log K_f + \left(\frac{1}{n}\right) \log C_e$
Langmuir-1	$q_e = \frac{Q_m K_L C_e}{1+K_L C_e}$	$\frac{C_e}{q_e} = \left(\frac{1}{K_L Q_m}\right) + \left(\frac{1}{Q_m}\right) C_e$
Langmuir-2		$\frac{1}{q_e} = \frac{1}{Q_m} + \left(\frac{1}{K_L Q_m}\right) \frac{1}{C_e}$
Langmuir-3		$q_e = Q_m - \left(\frac{1}{K_L}\right) \frac{q_e}{C_e}$
Langmuir-4		$\frac{q_e}{C_e} = K_L Q_m - K_L q_e$

3. Results

Table 4 shows results of X-ray diffraction and fluorescence analysis for Leca and limestone coral adsorbents. As shown in this table, the main components of Leca are silicon oxide and aluminum oxide, which are similar to constituents found in clinoptilolite zeolites which are found in most parts of Iran. Concerning limestone coral, the main component is calcium oxide, which is due to the presence of calcium ions and its sediment at the bottom of Persian Gulf. Concerning the components of both adsorbents, it seems that both adsorbents have the ability to make electrostatic bonds with negative ions such as the studied dye. Fig. 1 shows results of X-ray diffraction for limestone and Leca adsorbents. According to this figure, crystalline structure of limestone coral can be seen in all adsorption bonds. A dome-shaped curve observed in 26.0° bond can be attributed to the formation of the aragonite mineral which indicates the amorphous phase of limestone coral. Moreover, Leca crystalline structure can be found in 24.0°, 22.0°, 2θ = 9.0°, 49.0°, 46.0°, 38.0°, 32.0° and 28.0° bonds. The peak observed in 32.0° and 28.0° bonds can be attributed to the formation of quartz and feldspar minerals respectively; it forms the amorphous phase in Leca.

3.1. The effect of contact time

In first step of experiments the effect of contact time on the adsorption process was studied. The results are shown in Fig. 2. According to the results, the adsorption efficiency increased with the increasing contact time. The

Table 2 Equation and linear form of kinetics.

Kinetic	Equation	Linear form	References
Pseudo first order	$\frac{dq_t}{dt} = k_1(q_e - q_t)$	$\log(q_e - q_t) = \log(q_e) - \frac{k_1}{2.303} t$	(Ahmadian et al., 2012; Shokoohi et al., 2014)
Pseudo second order	$\frac{dq_t}{dt} = k_2(q_e - q_t)^2$	$\frac{t}{q_t} = \left(\frac{1}{k_2 q_e^2}\right) + \left(\frac{1}{q_e}\right) t$	
Elovich	$\frac{dq_t}{dt} = \alpha \exp(-\beta q_t)$	$q_e = \left(\frac{1}{\beta}\right) \ln(\alpha\beta) + \left(\frac{1}{\beta}\right) \ln t$	
Intraparticle diffusion	–	$q_t = K_{dif} t^{0.5} + C$	

Table 4 Component present in granular coral limestone and Leca based on XRF analyze % (w/w).

Component	SiO_2	Al_2O_3	MgO	P_2O_5	SO_3	K_2O	CaO	TiO_2	Cl	SrO	Na_2O
Coral limestone	0.109	0.636	0.634	–	0.997	–	92.85	–	0.36	2.75	1.46
Leca	66.06	16.57	2.99	0.21	0.23	2.69	2.46	0.78	1.7	0.13	0.69

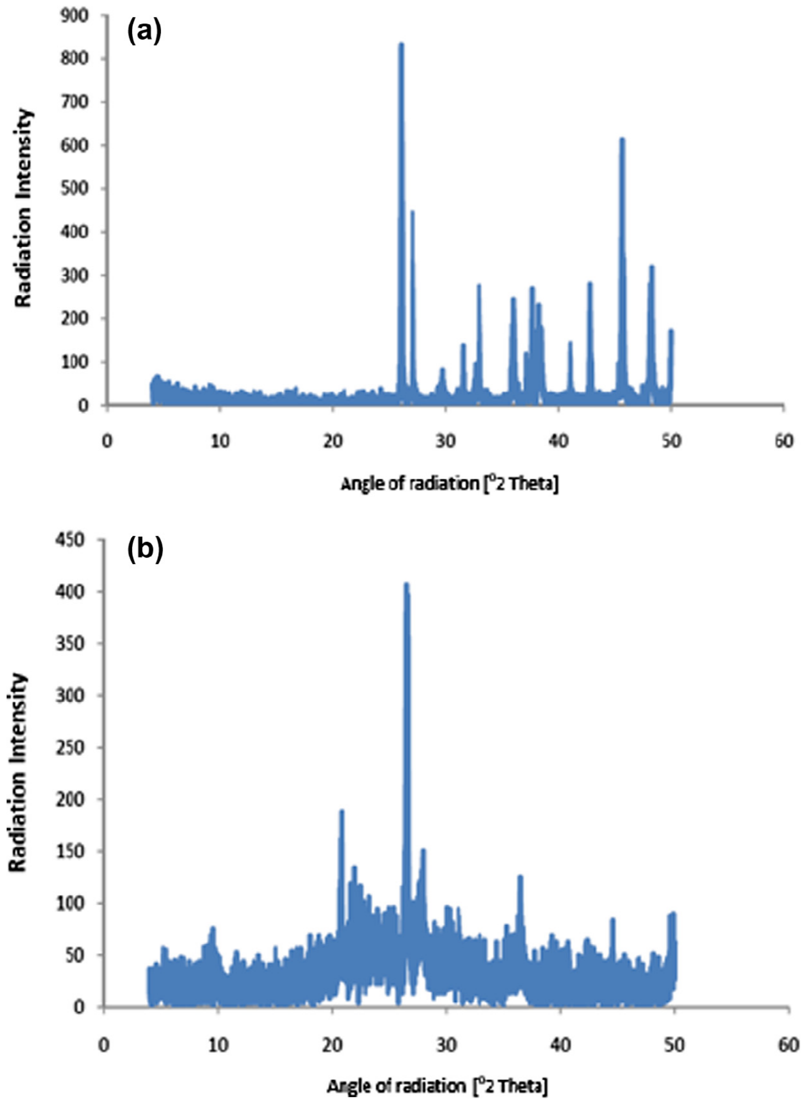


Figure 1 X-ray diffractograms of samples (a) limestone, (b) Leca.

maximum removal efficiency was achieved at 180 min of contact time, which was 72% and 74% for of limestone coral granules and Leca granules, respectively. The increase of dye sorption with the increasing contact time is associated with the increase in collisions between adsorbent and adsorbed. These results also indicate that the adsorption process of dyes in the first time is higher. This phenomena related with the large number of vacant surface sites (vacant surface sites) on the surface of the absorbent in the early stage. The results of this study are consistent with results from previous studies (Ozdemir et al., 2004; Shokoohi et al., 2015).

3.2. The effect of pH

pH is one of the significant factors in the adsorption process and this process is highly dependent on the pH of the solution. In this study, the effect of pH values on the adsorption efficiency was tested under selected condition (dye concentration = 100 mg/dm^3, adsorbent dosage = 10 g/dm^3, particles size = 595 μm and contact time = 180 min). The results of this part of the study were shown in Fig. 3. According to the obtained results, the removal efficiency of dye increased with the decreasing pH values, and the maximum removal efficiency of AC5R was seen at pH 3 under the presence of both

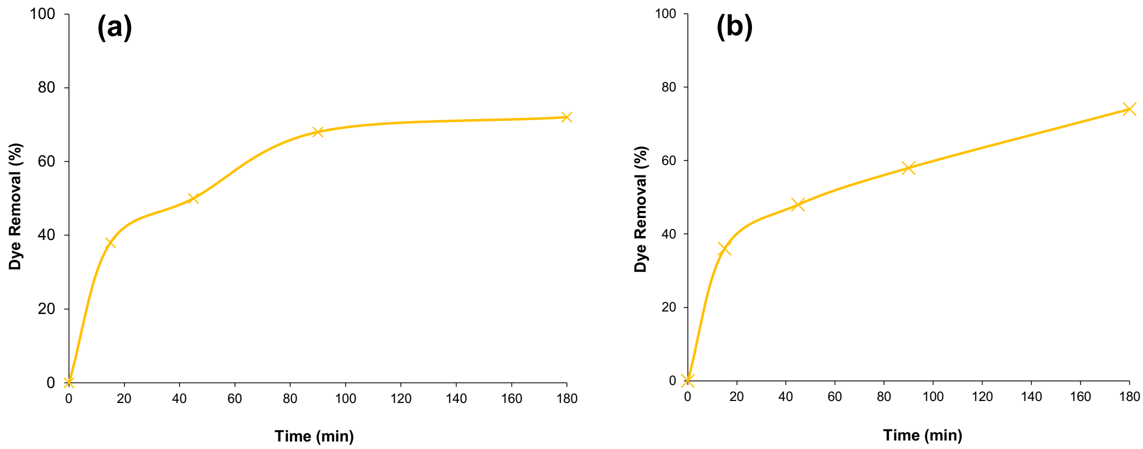


Figure 2 Effect of contact time on the adsorption efficiency (a) granular of coral limestone, (b) granular of Leca.

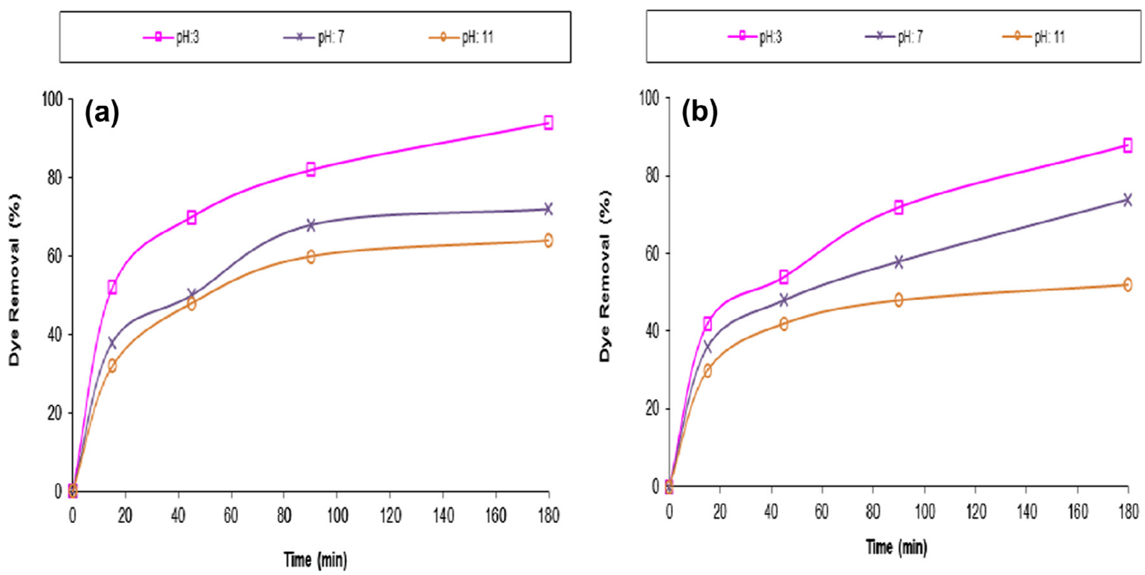


Figure 3 Effect of adsorbents dosage on the adsorption efficiency (a) granular of coral limestone, (b) granular of Leca.

adsorbents. As shown in Fig. 2 after 180 min and at pH 3, the maximum removal efficiency of dye by granular coral limestone and Leca was 94% and 88%, respectively. An increased efficiency of dye removal due to a decreased pH can be as a result of the presence of H_3O+ ion. In some cases in acidic pH, H+ ion loses a proton and establishes a bond with dye cations; this bond causes more cations to be removed compared with alkaline pH (Fernandes et al., 2007). In acidic pH, H+ ion gets positive load and establishes electrostatic bonds with dye anions and thus increases dye removal efficiency. In alkaline pH of aqueous solution, available negative adsorption sites of adsorbent increase and positive ones decrease; it decreases the removal efficiency at a higher pH (Xue et al., 2008). Results of this study are in line with the results of study carried out by Kim et al. (2008a, 2008b). One-way statistical test shows that there is a significant difference between changes in pH and the removal efficiency of dye by both adsorbents (p-value < 0.05). Results of comparing the efficiency of limestone coral granules and Leca granules show that limestone coral granules have higher efficiency in removing AC5R dye in all the pHs.

3.3. The effect of adsorbents dosage

To study the influence of adsorbent dosage on removal efficiency of AC5R, various dosages of granular coral limestone and Leca in the range 1–10 g/dm^3 were tested under selected condition (at pH 3 and 100 mg/dm^3 of dye initial concentration). The results of this part were shown in Fig. 4. According to the results, by increasing granular coral limestone dosages from 0.0285 g/mg of dye to 0.150 g/mg of dye the removal efficiency of AC5R was increased from 35% to 67%. Also, in such condition, by increasing Leca granular dosage from 0.0384 g/mg of dye to 0.145 g/mg of dye the removal efficiency of dye was increased from 26% to 69%. The obtained results indicated that the adsorption rate increased with the increasing adsorbent dosage. This can be attributed to the increasing adsorbent contact surface (Denise et al., 2008). On the other

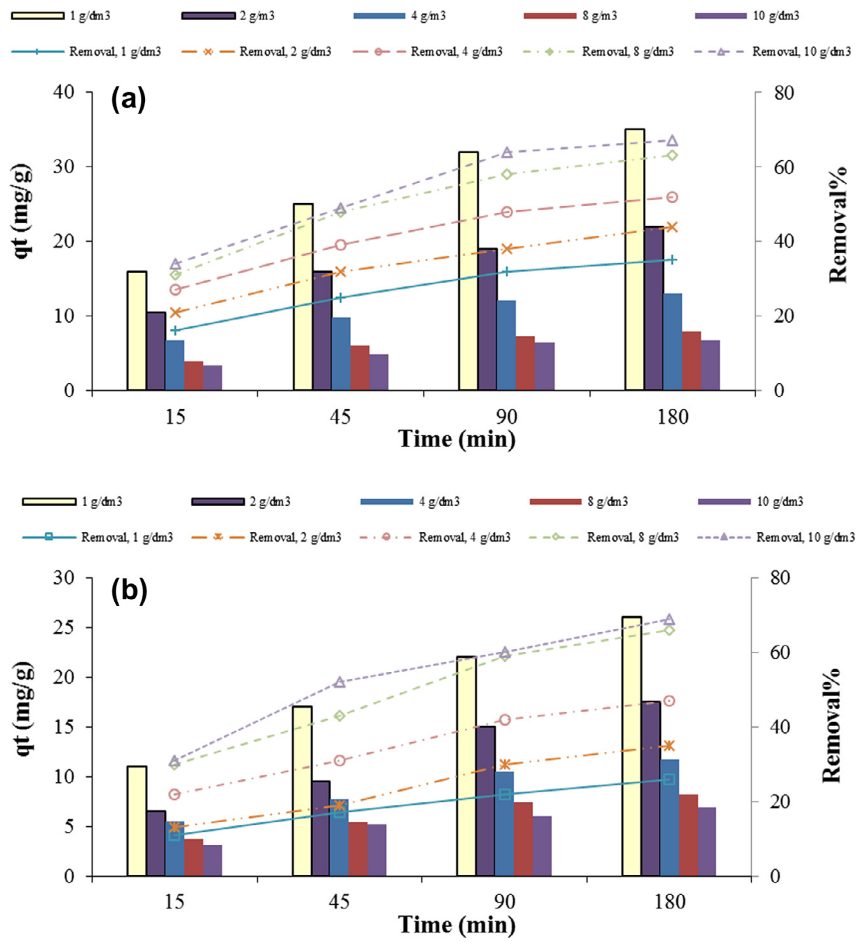


Figure 4 Effect of pH on the adsorption efficiency (a) granular of coral limestone, (b) granular of Leca.

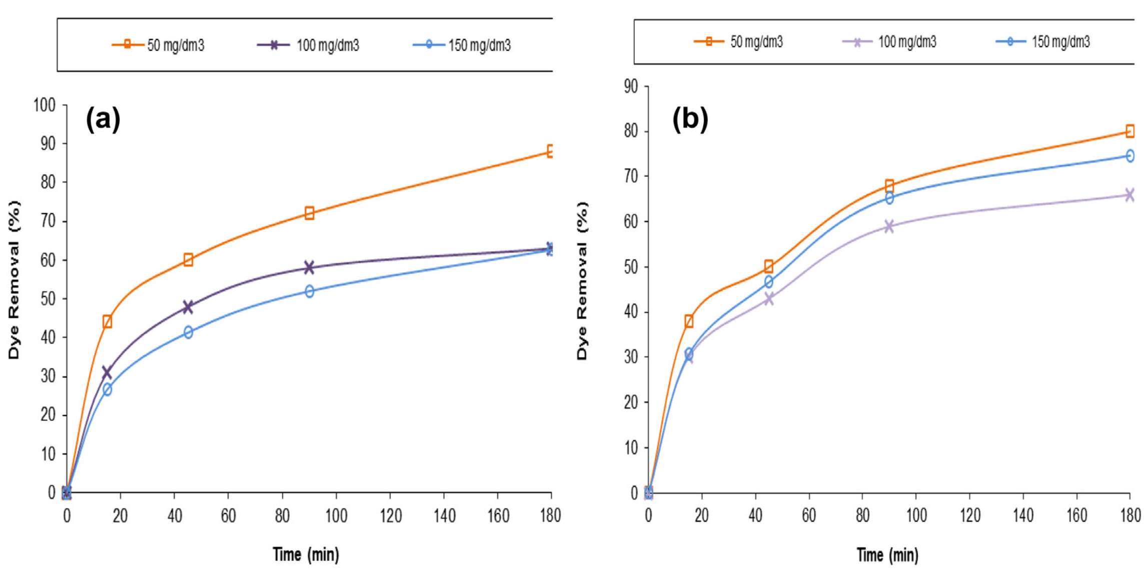


Figure 5 Effect of initial dye concentration on the adsorption efficiency (a) granular of coral limestone, (b) granular of Leca.

hand, adsorption capacity decreased with the increasing adsorbent dosage. This phenomenon can be related to the inside particle reactions such as aggregation that led to a decrease in the adsorbent area surface and an increase in distance between adsorbent and adsorbed (Tae et al., 2008). The results of the present study are in accordance with results of Xue et al.

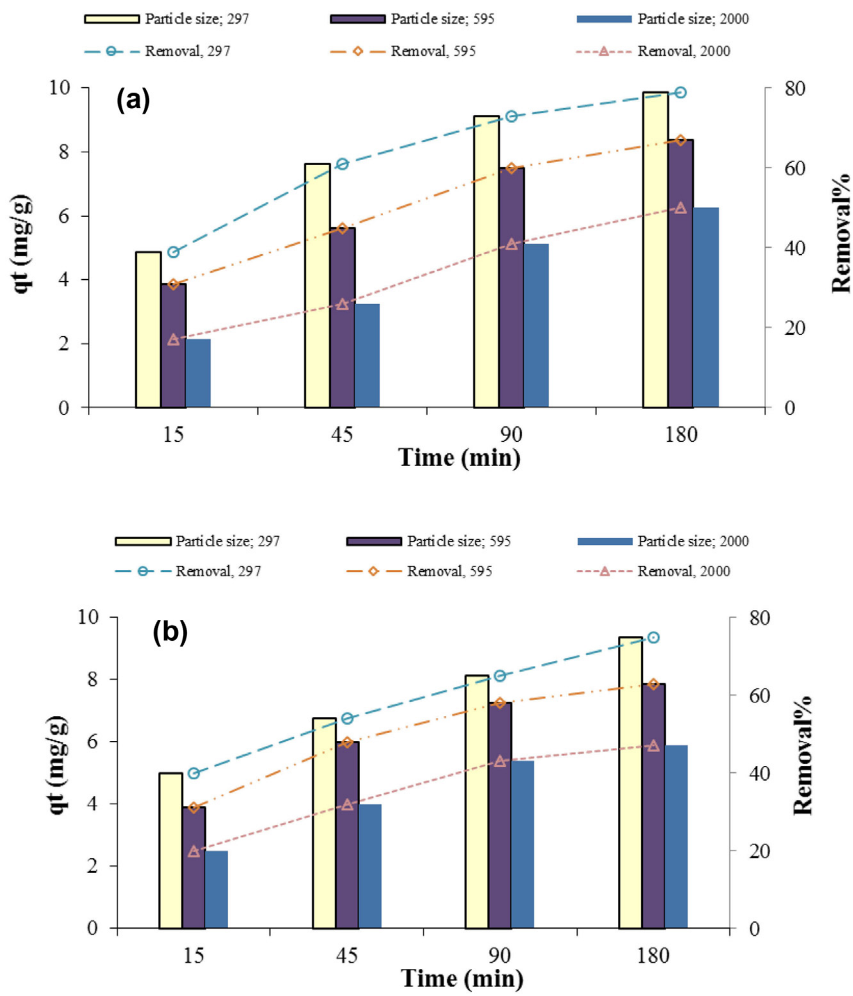


Figure 6 Effect of adsorbent particles size on the adsorption efficiency (a) granular of coral limestone, (b) granular of Leca.

Table 5 Parameters obtained from various kinetics models using different AC5R concentrations.

Kinetic	Kinetic parameter	AC5R concentration with coral limestone			AC5R concentration with granular Leca		
		50 mg/dm^3	100 mg/dm^3	150 mg/dm^3	50 mg/dm^3	100 mg/dm^3	150 mg/dm^3
Pseudo first order	k_1 (1/min)	0.025	0.033	0.004	0.025	0.034	0.01
	q_e cal (mg/g)	3.34	4.06	1.23	3.11	5.24	1.55
	R^2	0.7	0.6	0.26	0.64	0.75	0.26
Pseudo second order	k_2 (g/mg. min)	0.038	0.034	0.015	0.036	0.023	0.011
	q_e cal (mg/g)	5.7	8.23	12.39	5.25	8.73	14.97
	h (mg/g. min)	1.22	2.7	3.8	1.13	2.9	3.9
	R^2	0.98	0.99	0.97	0.97	0.97	0.97
Elovich	α (mg/g. min)	0.48	1.09	1.21	0.24	1.38	1.45
	β (g/mg)	0.77	0.5	0.35	0.83	0.49	0.29
	R^2	0.98	0.98	0.99	0.98	0.99	0.99
Intraparticle diffusion	K_{dif} (mg/g min$^{0.5}$)	0.68	1.019	1.5	0.63	1.072	1.83
	C (g/kg)	0.65	1.057	1.016	0.49	0.8	1.05
	R^2	0.93	0.9	0.96	0.95	0.94	0.96

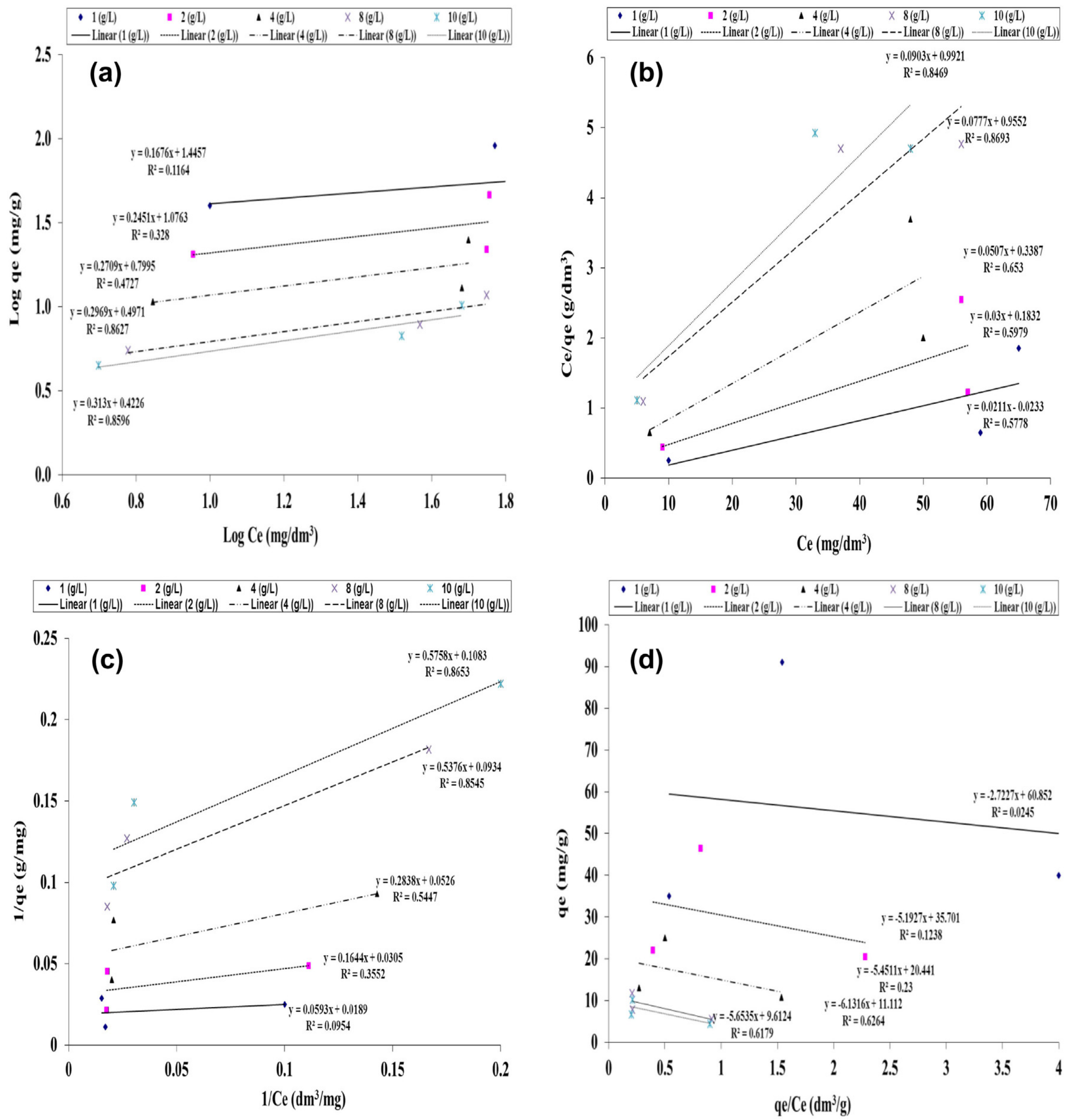


Figure 7 Isotherm models for adsorption of AC5R onto Coral limestone, (a) Freundlich, (b) Langmuir-1, (c) Langmuir-2, (d) Langmuir-3.

(2008) study and of Maximova and Koumnova (2008) study. The results of statistic analysis show that there was a significant difference between adsorbent dosage and dye removal efficiency (p-value < 0.05).

3.4. The effect of initial dye concentration

Since the industrial wastewater generally contains different concentrations of dyes, studying the effect of initial dye concentration on the adsorption process efficiency is practically of great importance. In this regard, three concentrations of (50, 100 and 150 mg/dm^3) AC5R dye were examined. Results of these tests are shown in Fig. 5. Results reveal that as initial dye concentration increases, efficiency of both adsorbents decreases. As shown in Fig. 5, as initial dye concentration increases from 50 to 150 mg/dm^3, efficiency of limestone coral granules and Leca decreases by 40% and 36%, respectively. It can be due to a decreased available adsorption surface due to saturation of the adsorption sites. Similar studies conducted by Fernandes et al. (2007) are consistent with the current study. Results of one-way statistical test indicate that there is a significant difference between change in dye initial concentration and removal efficiency of both adsorbents (p-value < 0.05). Generally, results reveal that in similar conditions in dye initial concentrations, limestone coral granules have a higher efficiency in removing AC5R dye compared to Leca granules.

3.5. The effect of adsorbents particles size

Based on previous studies (Mckay et al., 1988; Krishna and Swamy, 2012), particle size of the adsorbent is a factor affecting the efficiency of the adsorption process. Therefore, this

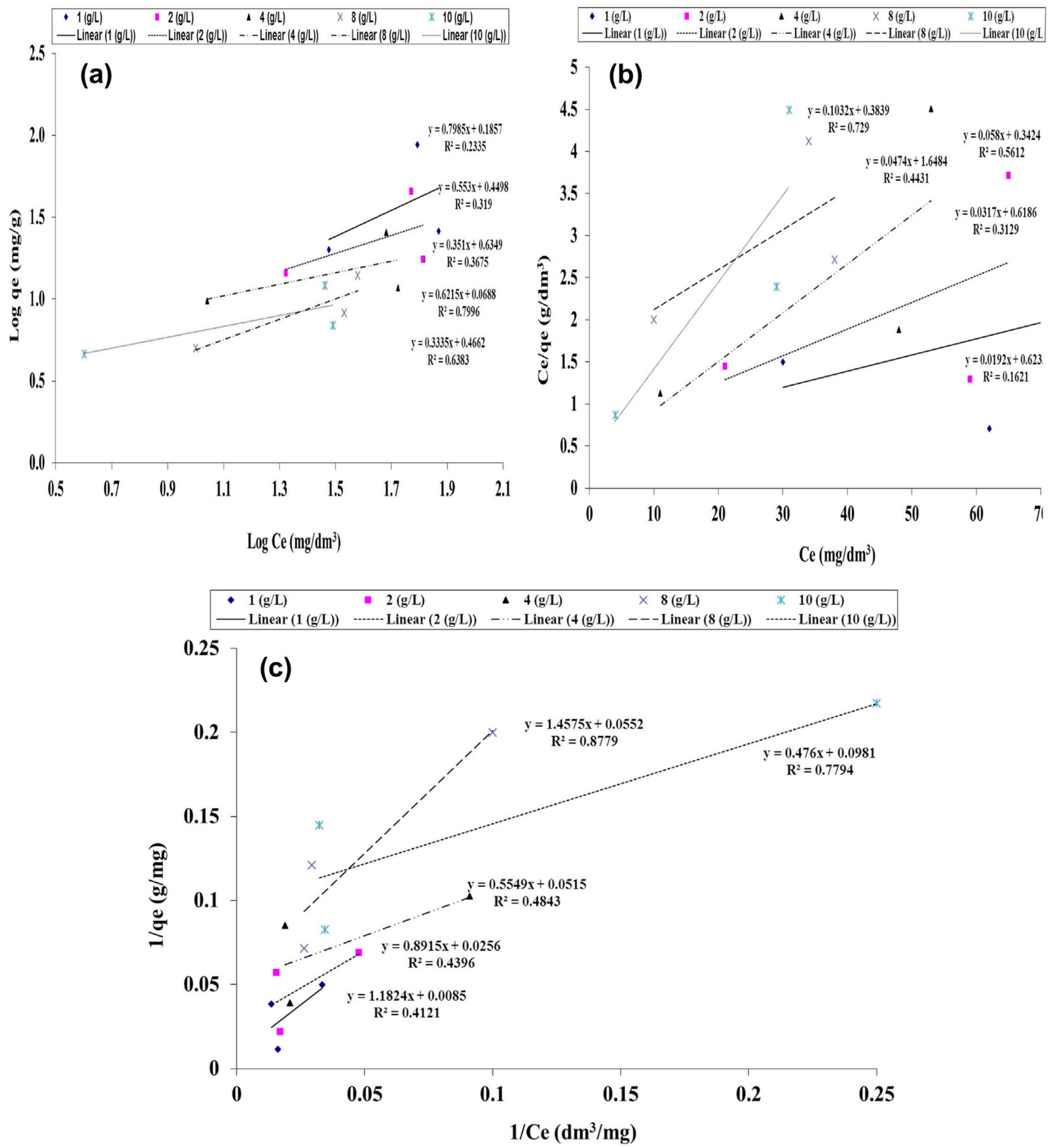


Figure 8 Isotherm models for adsorption of AC5R onto Leca, (a) Freundlich, (b) Langmuir-1, (c) Langmuir-2.

study aims to investigate the effect of particle size on the removal efficiency in three different sizes of limestone coral and Leca granules (2000, 595 and 297 μm). Results of these experiments are shown in Fig. 6. According to these results, as adsorbent size decreases, the dye removal efficiency increases; that is, as particle size decreases from 2000 to 297 μm, the removal efficiency of AC5R increases from 50% to 79% for limestone coral and increases from 47% to 75% for Leca granules, respectively. Increased efficiency can be due to the increased adsorbent physical surface. In addition, small-size adsorbent particles rotate more rapidly than bigger particles. In their research on dye adsorption by Mckay et al. (1988) they observed that as adsorbent size decreased, dye adsorption level increased drastically. Similarly, Krishna and Swamy (2012) obtained similar results. One-way statistical analysis shows that there is a significant difference between changes in adsorbent size and dye removal efficiency by both adsorbents (p-value < 0.05). As shown here, limestone coral granule is more effective in AC5R removal.

3.6. Kinetic study

Table 5 illustrates the parameters obtained from various kinetics models using different AC5R concentrations. It was found that the removal process of AC5R dye by both coral limestone and Leca followed the Elovich kinetic model, with a correlation coefficient higher than 0.99. In the study of Hashemian and Shahedi (2013) Ag/kaolin nanocomposite was used as adsorbent for removing AC5R from aqueous solution. The results of this study show that kinetic data were fitted with pseudo-second-order kinetic model and correlation coefficient equal to 0.99.

3.7. Isotherm study

Results of this study are shown in Figs. 7 and 8. Results obtained from the various linear forms of the isotherm models with coral limestone show that the isotherm data fitted to the Langmuir-2 with a correlation coefficient of 0.87. Moreover, the experimental results from the various linear forms of the isotherm models with Leca show that the isotherm data are best described by the Langmuir-2 isotherm equation, with a correlation coefficient of 0.88. The maximum absorption capacities of coral limestone and Leca towards AC5R were equal to 47.4 and 52.08 mg/g, respectively. Hashemian and Shahedi (2013) also observed that the adsorption isotherm was most consistent with Langmuir-2 model, and the maximum adsorption capacity of adsorbent in Acid Cyanine 5R removal was 12.65 mg/g. Shokouhi et al. (2012) also observed that the adsorption isotherm was most consistent with Freundlich model, and the maximum adsorption capacity was equal to 3.78 mg/g. Therefore, the adsorption capacities of coral limestone and Leca are higher than the maximum adsorption capacities reported for some other adsorbents.

4. Conclusion

Results of this study showed that both granular coral limestone and Leca could be used as natural adsorbents for removing AC5R from aqueous solution. The optimum pH and adsorbents particles size in both adsorbents were determined to be 3 and 297 μm, respectively. Moreover, the optimum dosages of coral limestone and granular Leca were 0.150 and 0.145 g/mg of dye, respectively. Also, obtained results showed that the adsorption efficiency by both granular coral limestone and Leca increased with the decrease in adsorbents particles size. In general, finding results indicated that performance of coral limestone granular was better in adsorption of AC5R than that of Leca granular under similar condition. Results obtained from the various linear forms of the isotherm model with coral limestone show that the isotherm data fitted to the Langmuir-2. Moreover, the experimental results from the various linear forms of the isotherm model with Leca show that the isotherm data are best described by the Langmuir-2 isotherm equation. Insomuch as, the granular of coral limestone and LECA were very low cost adsorbents, they can be used for water and wastewater treatments as a cost effective process compared to other expensive adsorbents. These findings suggested that the granular of coral limestone acts as a suitable adsorbent for removing dye pollutants from aqueous solution. However, it is obvious that farther studies are required to optimize process parameters for lager scales.

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