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# Elimination of Congored from Aqueous Solution by the using of gamma alumina nanoparticles

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## ABSTRACT

These days, the removal of various dyes from industrial wastewater has become an important concern. Synthetic dyes used in industries such as textiles are complex, toxic and mutant organic materials. Congored is also an acidic dye that is considered a high- risk source of contamination and threatens the lives of humans and other living organisms. Different methods, such as electrochemical, physical and biological processes, are used to remove organic pigment from aqueous samples. Adsorption is one of the most effective modern methods of industrial wastewater treatment, which is a relatively simple and inexpensive process that produces non- toxic and low- risk products. This research focuses gamma alumina nanoparticles was used for the adsorption of the cationic dye congored from aqueous solutions. Batch adsorption studies carried out to study various parameters included pH, gamma alumina nanoparticles dosage, temperature and contact time. The concentration of dye congored was measured using a UV-vis Spectrophotometer at the wavelength of 498 nm. The optimum adsorption conditions were found to be pH=6, adsorbent dose=0.04 g, temperature=298 K and contact time=40 min. The experimental results of this work were compared with Langmuir, Friendlich and Tamkin's isotherm models. The results obtained from isotherm models showed that the surface adsorption of these dye on the adsorbent used better follows the Langmuir isotherm model. Analysis of thermodynamic data showed that the adsorption process of the studied dye on the adsorbent surface is spontaneous and exothermic.

**Keywords:** Congored; Adsorption; Thermodynamic; Gamma Alumina.

## INTRODUCTION

Transitional aluminas, particularly γ-alumina and θ-alumina, which are usually obtained by calcination of boehmite at varioustemperatures, are among the most significant oxides used in the industrial applications. The gamma alumina are widely used as adsorbents, catalysts and catalysts supports, due to their large specific surface areas, well – defined pore size distributions, stability within a wide temperature span, ability to stabilize and disperse the active phases as

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well as moderate acidities. There are several reports representing key roles of gamma alumina as catalysts support. Gamma-alumina is a very important material industrially due to their use in adsorption and catalysis. The nanoparticles of aged pseudoboehmites materials with high specific surface area are being recently studied for the controlled release of drugs [1-8].

The release of huge volume of dyes containing waste water from different industries like textile, leather, paper, rubber, printing, food and cosmetics is a dangerous global concern. These harmful dyes containing waste water are disposed directly into the river without proper treatment causing carcinogenic and mutagenic effects. Therefore, the massive development towards remediation of such problems is highly desirable. Nowadays, the effect of harmful dyes is a serious health problem due to their negative impact on both flora and fauna. Dye elimination from wastes has been the object of many researches in the past few years because of the potential toxicity of dyes and visibility problems. These compounds are used in large quantity in many industries including textile, leather, cosmetics, paper, printing, plastic, pharmaceutical and food to color their products. As a result, considerable amounts of colored wastewater are generated. Dyes commonly have complex aromatic structures which make them stable and difficult to decompose. The dyes present in wastewater absorb sunlight, leading to a decrease in the efficiency of photosynthesis in aquatic plants due to reduced light penetration. Many of the industrial dyes are toxic, carcinogenic, mutagenic and teratogenic. Their elimination from wastewater is of great regard. Considering both the volume and chemical composition of the discharged effluent, the textile, dyeing, pulp and paper, and printing industries are the major polluters among the industrial sectors [3-14].

Over the past few decades, conventional waste water treatment methods for removing dyes include biological (aerobic and anaerobic), chemical and physiochemical methods such as coagulation and flocculation, solvent extraction, membrane filtration ozonation, ion exchange, electrochemical techniques, fungal decolonization and adsorption. The removal of dyes from wastewater or industrial effluent using adsorption method has recently gained more favour than the other methods of removal of dyes. Adsorption process has simplicity of design, more efficient, easy to operate, insensitive to toxic substances and cost effective. It therefore provides an alternative method to other expensive existing physical/ chemical/ biological methods for the removal of dyes from industrial effluents or waste water [5-20].

This research focuses on the use of gamma alumina nanoparticles as a biosorbent for the removal of dye congored from aqueous solution, was studied in batch equilibrium conditions. The effects of different parameters including pH, initial metal ion concentration, contact time, gamma alumina nanoparticles dosage and temperature were investigated. Langmuir and Freundlich isotherm models were used to analyze the equilibrium data.

## EXPERIMENTAL

### *Apparatus and Materials*

An Lambda 25 UV/VIS Spectrophotometer From PerkinElmer was used to measure the concentration of dye congored in the studied solutions, a 820 A model pH meter (Metrohm Co.) was used to measure pH of solutions and a thermostatic orbit incubator shaker neolab model (India) was used to measure contact time in the solutions. All chemical

materials used in this study were of analytical grade.

***Batch Adsorption Experiments***

Batch adsorption experiments were carried out to characterize the dye congored adsorption isotherm onto gamma alumina nanoparticles and its thermodynamic properties. Dye congored stock solution was prepared by dissolving the appropriate quantity of dye congored in deionized water. Adsorption isotherms were obtained by using initial dye congored concentration, $X_o$, and its equilibrium concentration, Xe. The effect of pH on the dye congored adsorption onto gamma alumina nanoparticles was evaluated in a pH range of 2-12. The pH of solutions was adjusted by 0.1 M HCl and 0.1 M NaOH solutions. For every experiment, 50 ml of the solution dye congored was mixed with gamma alumina nanoparticles in a 250 ml glass conical flask which was shaken in a thermostatic orbit shaker at 220 rpm. The mixture was filtered through a 0.45 μm membrane filter. The filtrate was measured by UV/VIS Spectrophotometer then, the adsorption percentage (%A) was determined as:

$$\%A_e = \frac{A_0 - A_e}{A_0} \times 100 \qquad (1)$$

$Q_e$, amount adsorbed per unit weight of adsorbent at equilibrium (mg/g) was calculated using the following equation. Where $X_o$ and $X_e$ are the initial and final concentrations of dye congored in solution (mg/L), respectively [3-23].

$$Q_e = \frac{(X_0 - X_e)V}{M} \qquad (2)$$

Where M is the mass of gamma alumina nanoparticles (g) and V is the volume of the solution (L). To evaluate the thermodynamic properties of the adsorption process, 0.04 g of gamma alumina nanoparticles was added into the 50 ml solution with pH of 3.0 and initial dye congored concentration ranging from 5 mg/L in every experiment. Each solution was shaken continuously for 50 min [3-23].

## RESULTS AND DISCUSSTON

***The Effect of pH***

The solution pH was very important parameter for the biosorption studies. Batch studies at different pH (2-12) were conducted by soaking the 0.01 g adsorbent in 6 mg/L of dye congored in each microcosm. Each container was agitated (156 rpm) for 40min at 25$^o$C. Table 1 illustrate the effect of the pH of the solution on the adsorption percentage of dye congored, adsorbed onto gamma alumina nanoparticles. The best results were obtained at pH=6 for dye congored [10-27].

Table 1. The effect of initial pH of the solution on the adsorption percentage (%A) of dye congored (Xo=6 mg/L, M gamma alumina nanoparticles =0.01 g, T=298 K, tc=40 min)

| pH | %Ae |
|---|---|
| 2 | 47.16 |
| 4 | 53.51 |
| 6 | 59.32 |
| 8 | 47.97 |
| 10 | 38.37 |
| 12 | 38.21 |

**The Effect of Adsorbent Dosage**

Microcosms with different adsorbent doses (0.01-0.07 g) were amended with 6 mg/L of dye congored in aqueous solutions. The rate of adsorption was monitored at the following optimum conditions: pH of 6, for 40 min at 25$^o$C.The effect of gamma alumina nanoparticles dosage on the adsorption percentage of dye congored is shown in table 2. The best results were obtained at M gamma alumina nanoparticles =0.04 g for dye congored.

**Table 2**. The effect of gamma alumina nanoparticles dosage on the adsorption percentage (%A) of dye congored ($X_o$=6 mg/L, pH=6, T=298 K, tc=40 min)

| M gamma alumina nanoparticles (g) | %Ae |
|---|---|
| 0.01 | 59.32 |
| 0.02 | 60.94 |
| 0.03 | 68.51 |
| 0.04 | 75.67 |
| 0.05 | 70.40 |
| 0.06 | 68.64 |
| 0.07 | 64.68 |

### *The Effect of Temperature*

The same preparation was made, except for the varying temperature conditions. The microcosm which was maintained at pH=8 was incubated at different temperatures (25-65 $^oC$) for a period of 40 min. Table 3 show that the adsorption percentage decreases with increasing temperature. Therefore, it may be concluded that the interaction between dye congored and gamma alumina nanoparticles is exothermic in nature.

**Table 3**. The effect of temperature on the adsorption percentage (%A) of dye congored ($X_o$=6 mg/L, M gamma alumina nanoparticles =0.04 g, pH=6, $t_c$=40 min)

| T (K) | %Ae |
|---|---|
| 298 | 75.68 |
| 308 | 75.00 |
| 318 | 74.16 |
| 328 | 73.33 |
| 338 | 72.66 |

### *The Effect of Contact Time*

The effect of contact time, tc, on the adsorption percentage of dye congored onto gamma alumina nanoparticles is shown in table 4. A rather fast uptake occured during the first 40 min of the adsorption. It became slower as the adsorbed amount of dye congored reached its equilibrium value. It can be seen that the adsorption process is rapid due to the availability of very active sites on the adsorbent surface at initial stage. This may be due to the special one atom layering the structure of dye congored. At first, adsorption capacity was a slow process then, increased rapidly, it attained equilibrium and saturation gave constant adsorption value. The optimum contact time was obtained at 40 min.

**Table. 4**. The effect of contact time, $t_c$, on the adsorption percentage (%A) of dye congored ($X_o$=6 mg/L, M gamma alumina nanoparticles =0.04 g, pH=6, T=298 K)

| Tc (min) | $\%A_t$ |
|---|---|
| 10 | 43.14 |
| 20 | 71.89 |
| 30 | 75.13 |
| 40 | 75.67 |
| 50 | 75.67 |
| 60 | 75.67 |

### *Adsorption Isotherm*

An adsorption isotherm is characterized by certain constant values, which express the surface properties of the adsorbent and so on the percentages adsorption of dye congored onto gamma alumina nanoparticles as a function of initial concentration of dye congored , shown in table 5. Adsorption isotherms represent the relationship of the amount of dyes adsorbed with the adsorbent dose. These provide information about the mechanism of adsorption and the adsorptivity of the composite towards the dyes of interest. In this study, Langmuir, Freundlich and temkin isotherms were investigated [5-29].

The simplest and still the most useful isotherm, for both physical and chemical adsorption, is the Langmuir isotherm. This model assumes that adsorption is limited to a monolayer: only a singlelayer of molecules on the adsorbent surface are absorbed, adsorbent surface is homogeneous and adsorption energy is uniform for all sites and there is no

transmigration of adsorbate in the plane of the surface. Once a pollutant occupies a site, no further adsorption can take place in that site; the intermolecular attractive for cesrapidly decrease as distance rises. There is no interaction between molecules adsorbed on neighboring sites, adsorption on surface is localized, which means that adsorbed atoms or molecules are adsorbed at definite and localized sites. The linear form of the Langmuir isotherm is described by the equation (3) [8-30]:

$$\frac{1}{Q_e} = \frac{1}{bQ_m}\left(\frac{1}{X_e}\right) + \frac{1}{Q_m} \qquad (3)$$

**Table 5**. Adsorption data for dye congored adsorption onto gamma alumina nanoparticles (pH=6, $t_c$=40 min, T=298 K, M gamma alumina nanoparticles =0.04 g)

| **Parameter** | **Value** | | | |
|---|---|---|---|---|
| $X_0$ (mg/L) | 2 | 3 | 4 | 5 |
| %A | 65.01 | 66.00 | 67.81 | 73.79 |
| Xe (mg/L) | 0.07 | 1.02 | 1.29 | 1.31 |
| $Q_e$ (mg/g) | 1.63 | 2.48 | 3.39 | 4.61 |
| $1/Q_e$ (g/mg) | 0.62 | 0.40 | 0.29 | 0.22 |
| 1/Xe (L/mg) | 1.43 | 0.98 | 0.78 | 0.76 |
| lnXe | -0.36 | 0.02 | 0.25 | 0.27 |
| lnQe | 0.40 | 0.91 | 1.22 | 1.53 |

Where $Q_m$ (mg/g) is the maximum dyes to adsorb onto 1 g adsorbent and b (L/mg) is the Langmuir constant related to adsorption capacity and energy of adsorption. The slope and intercept of plot of $1/Q_e$ versus $1/X_e$ are shown in fig. 1 that were used to calculate the values of b and $Q_m$ (table 6).

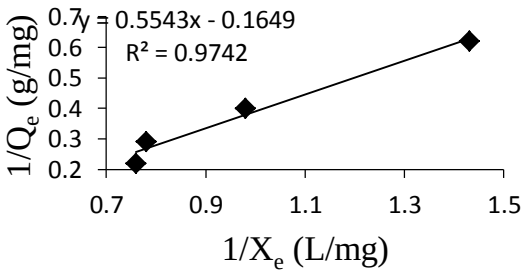


Fig. 1. Langmuir isotherm for dye congored adsorption onto gamma alumina nanoparticles.

The Freundlich isotherm model is an empirical equation and another form of Langmuir that can be applied to multilayer adsorption. This model assumes that the surface of the adsorbent is heterogeneous and active sites and their energies distribute exponentially. The stronger binding sites are occupied first, until adsorption energy is exponentially decreased upon the completion of adsorption process. The Freundlich isotherm is expressed as equation (4) [8-30]:

$$\ln Q_e = \ln P + \frac{1}{n}\ln X_e \qquad (4)$$

Where P (L/mg) and n are the Empirical Freundlich constant or capacity factor and adsorption intensity. The values of P and n are determined from the intercept and slope of a plot of ln Qe versus ln Xe (table 5 and fig. 2) that were used to calculate the values of P and n (table 6)).

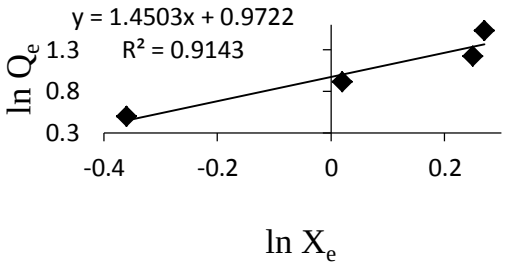


**Fig. 2**. Freundlich isotherm for dye congored adsorption onto gamma alumina nanoparticles.

Another empirical equation, the Temkin equation, describes theadsorption of hydrogen onto platinum electrodes within the acidicsolutions. The model is given by equation (5) [8-30]:

$$Q_e = B_T \ln A_T + B_T (\ln X_e) \qquad (5)$$

The Temkin isotherm equation assumes that the heat of adsorption of all the molecules in the layer decreases linearly rather than logarithmically as equilibrium adsorption capacity increases because the $B_T$ factor is related to adsorbent-adsorbate

interactions. The adsorption is characterized by a uniform distribution of the binding energies, up to some maximum binding energy. The Temkin equation is better for predicting the gas phase equilibrium rather than liquid-phase equilibrium [8-30].

Where $A_T$ (L/mg) is Temkin isotherm constant. $A_T$ is related to binding constant and $B_T$ (J/mol) is the Temkin constant that is related to the heat of sorption. $B_T$ and $A_T$ are determined from the slope and intercept of a plot of Qe versus ln Ce (table 5 and fig. 3). We used these results to calculate the values of $B_T$ and $A_T$ (table 6).

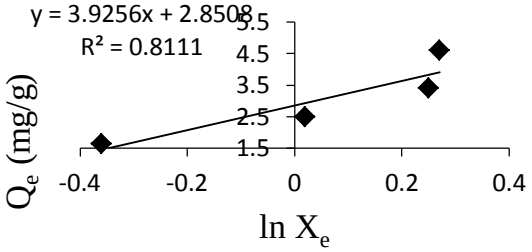


**Fig. 3**. Temkin isotherm for dye congored adsorption onto gamma alumina nanoparticles.

**Table 6.** The resultant values for the studied isotherms in connection to dye congored adsorption onto gamma alumina nanoparticles

| Isotherm | Parameter | Value |
|---|---|---|
| Freundlich | P (L/mg) | 2.64 |
| | n | 0.69 |
| | $R^2$ | 0.9143 |
| Langmuire | b (L/g) | 0.3 |
| | $Q_m$ (mg/g) | 6.06 |
| | $R^2$ | 0.9742 |
| Temkin | $B_T$ (J/mol) | 3.93 |
| | $A_T$ (L/mg) | 2.07 |
| | $R^2$ | 0.8988 |

### *Thermodynamic Parameters*

The amounts of adsorption of dye congored by gamma alumina nanoparticles in temperature 298-338 K. Analysis of thermodynamics of equilibrium adsorption data can give more important information on adsorption process. Thermodynamic parameters such as change in free energy (ΔG°), enthalpy (ΔH°) and entropy (ΔS°) were determined using the following equations. In the present study, thermodynamic parameters $\Delta H^0$, $\Delta S^0$ and $\Delta G^0$ were calculated by using the equation. The thermodynamic parameters of adsorption process can be determined from the variation of thermodynamic equilibrium constant, $K_o$, where $K_o$ is defined as follows [7-34]:

$$K_0 = \frac{a_S}{a_e} = \frac{Q_e}{X_e} = \frac{X_0 - X_e}{X_e} \quad (6)$$

Where $a_S$ and $a_e$ are the activity of adsorbed dye congored and the activity of dye congored in solution at equilibrium, respectively. The adsorption standard free energy change ($\Delta G^0$) is calculated according to:

$$\Delta G^0 = -RT \ln K_0 \quad (7)$$

The average standard enthalpy change ($\Delta H^o$) and the average standard entropy change ($\Delta S^o$) are obtained from the plot of equation (8) [7-34]:

$$\ln K_0 = \frac{\Delta S^0}{R} - \frac{\Delta H^0}{RT} \quad (9)$$

The ΔH° and ΔS° values obtained from the slope and intercept of plots are presented in table 7and fig. 4.

**Table 7**. The effect of temperature on $K_o$ values ($X_o$=6 mg/L, pH=6, M gamma alumina nanoparticles =0.04 g, $t_c$=40 min)

| T (K) | $\ln K_0$ |
|---|---|
| 298 | 1.14 |
| 308 | 1.10 |
| 318 | 1.05 |
| 328 | 1.01 |
| 338 | 0.98 |

The obtained values of thermodynamic parameters ($\Delta G^0$, $\Delta H^0$, $\Delta S^0$) are listed in table 8. The negative value of $\Delta H^0$ suggests that the interaction of adsorbed

dye congored with gamma alumina nanoparticles is an exothermic process, which is supported by the decreasing of the amount of dye congored adsorption with increasing temperature. The negative value of $\Delta S^o$ showed an decreased randomness during dye congored adsorption. The negative values of $\Delta G^0$ reveal the fact that the adsorption process is spontaneous.

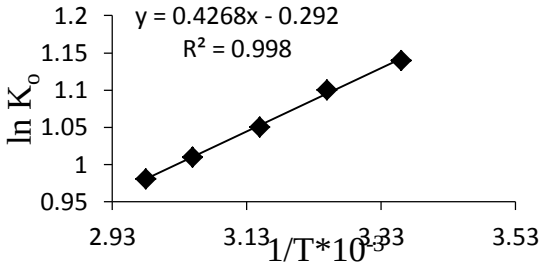


**Fig. 4**. The effect of temperature on equilibrium constant values.

**Table 8**. Thermodynamic parameters for adsorption dye congored onto gamma alumina nanoparticles

| T /K | $\Delta G^o$(J/mol) | $\Delta H^o$(J/mol) | $\Delta S^o$(J/mol K) |
|---|---|---|---|
| 298 | -2812.58 | | |
| 308 | -2813.43 | | |
| 318 | -2787.42 | -3.55 | -2.43 |
| 328 | -2758.17 | | |
| 338 | -2746.74 | | |

## CONCLUSION

Taking into account the results, we have considered it of great interest to assess the ability of gamma alumina nanoparticles for the adsorption of dye congored from aqueous solutions in the batch technique and optimization of conditions for its adsorption. The results of this work show that gamma alumina nanoparticles is an effective adsorbent for the removal of dye congored from aqueous solutions. The isotherm parameters were calculated and Results showed that the Langmuir isotherm model was fitted well with adsorption data, thus, indicating the applicability of monolayer coverage of dye congored on gamma alumina nanoparticles surface. The temperature variations have been used to evaluate the values of $\Delta H^0$, $\Delta S^0$ and $\Delta G^0$. Thermodynamic analysis revealed that the adsorption process is exothermic and spontaneous in nature.

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# حذف کنگورد از محلول‌های آبی با استفاده از نانوذرات گاما آلومینا

**غلامعلی حق دوست***

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## چکیده

این روزها حذف رنگ های مختلف از پساب‌های صنعتی به یک دغدغه مهم تبدیل شده است. رنگ‌های مصنوعی مورد استفاده در صنایعی مانند نساجی، مواد آلی پیچیده، سمی و جهش یافته هستند. کنگورد همچنین یک رنگ اسیدی است که منبع آلودگی پرخطر محسوب می‌شود و زندگی انسان و سایر موجودات زنده را تهدید می‌کند. روش‌های مختلفی مانند فرایندهای الکتروشیمیایی، فیزیکی و بیولوژیکی برای حذف رنگدانه آلی از نمونه‌های آبی استفاده می‌شود. جذب سطحی یکی از موثرترین روش‌های نوین تصفیه فاضلاب صنعتی است که فرایندی نسبتاً ساده و ارزان است و محصولات غیرسمی و کم‌خطر تولید می‌کند. در این تحقیق از نانوذرات گاما آلومینا برای جذب رنگ کاتیونی حاصل از محلول‌های آبی استفاده شده است. مطالعات جذب دسته‌ای برای مطالعه پارامترهای مختلف شامل pH، دوز نانوذرات گاما آلومینا، دما و زمان تماس انجام شد. غلظت رنگ کنگورد با استفاده از اسپکتروفتومتر UV-vis در طول موج ٤٩٨ نانومتر اندازه‌گیری شد. شرایط جذب بهینه pH=6، دوز جاذب، ۰/۰٤ گرم، دما، ۲۹۸کلوین و زمان تماس،٤۰ دقیقه بود. نتایج تجربی این کار با مدل‌های ایزوترم لانگمویر، فرندلیچ و تمکین مقایسه شد. نتایج بدست آمده از مدل‌های ایزوترم نشان داد که جذب سطحی این رنگ بر روی جاذب مورد استفاده از مدل ایزوترم لانگمویر بهتر پیروی می‌کند. تجزیه و تحلیل داده‌های ترمودینامیکی نشان داد که فرایند جذب رنگ مورد مطالعه بر روی سطح جاذب، خودبه‌خود و گرمازا است.

**کلید واژه‌ها**: کنگورد؛ جذب ترمودینامیکی؛ گاما آلومینا

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