https://doi.org/10.33261/jaaru.2019.26.2.002 *Association of Arab Universities Journal of Engineering Sciences (2019) 26(2): 6–11*

Association of Arab Universities Journal of Engineering Sciences
مجلة اتحاد الجامعات العربية للدراسات والبحوث الهندسية

Removal of Reactive Green 12 Dye and COD from Simulated Wastewater Using Different Coagulants

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Received: 30 June 2019

Abstract— The ability of using each of the following: aluminum sulphate (Al2(SO4)3.16H2O), ferric chloride (FeCl3), and ferrous sulphate (FeSO4) as chemical coagulants was investigated for removing of reactive green 12 (RG 12) dye and chemical oxygen demand (COD) from simulated wastewater. Best pH , coagulants dosages, and initial concentrations were obtained by jar test. The maximum efficiency for removing RG-12 and COD recorded by ferric chloride were 98% and 88 %, by alum were 95% and 88%, and by ferrous sulphate were 70% and 50%. All these results obtained at the best pH 6, dosage 100 mg/l and initial concentrations for RG-12 and COD 50 mg/l and 600 mg/l respectively. The maximum volume of sludge was for alum coagulant 14 ml/l, 12 ml/l for ferric chloride and 0.5 ml/l for ferrous sulphate. The study improved that it is possible to use each of aluminum sulphate, ferric chloride and ferrous sulphate as an economical coagulant to treat the wastewater which it is polluted with RG 12 dye and COD.

Keywords— Reactive Green12, COD, Alum, Ferric chloride, Ferrous Sulphate.

1. Introduction

During the last five decades, there was a special attention about the colored water that is wasted from the wet processing of textile industries because it generates a highly polluted wastewater. Textile wastewater is a source of pollution, because it has color, suspended solids, pH, high temperature, and chemical oxygen demand as a high rates [18]. There are about forty thousand kinds of dyes and pigments which consist of over seven thousand chemical structures. Most of them are difficult to remove their colors due to their synthetic origin and high water solubility [7]. Literatures reviews declare many methods of color removing consist of physiochemical [2], chemical [2,15] and biological process [11] as well as advanced methods like oxidation process [13,5], still there are no dependent economically and technically kind of methods, otherwise the combination between two or three methods generate good treatment for color removal. High removal efficiencies have been got when chemical treatment by coagulation was utilized [1]. There are many merits when coagulation process is used, such as not toxic, economic, easy to use without any special device, very effective to eliminate color and COD, and it is good alternative method for other methods [10].Turbidity, chemical oxygen demand (COD), biological oxygen demand (BOD), total dissolves solids (TDS) and chloride were removed with good results using alum, ferrous sulphate and polyacrylamide as an inorganic coagulants from dairy wastewater [9,8]. Reactive dyes have a complex chemical structures which share electrons between the reactive groups of cellulose. Reactive dyes give a stable structures and shining colors. Reactive colors react easily with cellulose fiber such as cotton, wool and polyamide fibers .Reactive dyes have many diverse collections that react to form shared electrons[17]. Reactive dyes could be classified as azo dyes, due to presence of azo bonds (N=N), some examples for reactive dyes are drimaren brilliant green Z-3G (Reactive Green 12), reactive black 5, remazol red RB, cibacron brilliant red B, reactive yellow 2, reactive green 19 etc.[6]. Experiments were carried out to find the best operating factors pH, dosage and dye concentration to remove reactive green 12 dye and COD from simulated wastewater using chemical coagulation process with three different coagulants alum, ferric chloride and ferrous sulphate.

2. Materials and methods

Drimaren brilliant Green Z-3G (Reactive Green 12) was used, provided from Al-Khadmya State Company for Textile, Baghdad, having the chemical structure C60-H29-

Cl3-N16-Ni-O21-S7.6Na and molecular weight 1839.4499 g/mol, the structure of reactive green dye 12 shown in fig.1.The solution of dye was ready after dissolving 1000 gm of dye in one liter of distilled water. COD synthetic wastewater was prepared using commercially sucrose ($C_{12}H_{22}O_{11}$) (Molecular weight: 342 g/mol). For a concentration of sucrose = 500 mg/L COD concentration approximate 560 mg/L. Chemical coagulants were used listed in Table 1.

Table 1: List of chemical coagulants were used

Coagulant chemical name	Coagulant chemical structure	Molecular weight (g/mol)
aluminum sulphate	$Al_2(SO_4)_3.16H_2O$	630.39
ferric chloride	$FeCl_3$	162.21
ferrous sulphate	$FeSO_4$	151.908

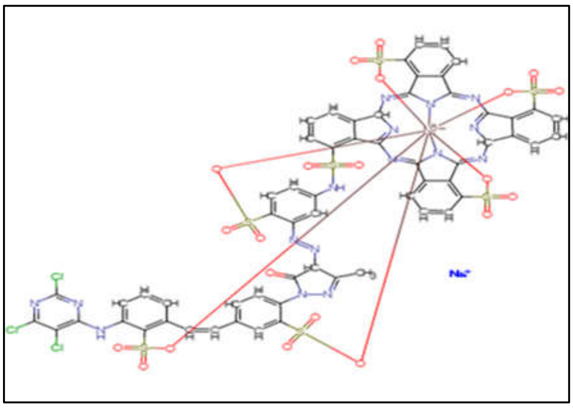

Figure 1: Reactive green 12 dye chemical structure[15]

Stock solutions were prepared by dissolving 10 g of these salts in 1L of deionized water. Jar test was conducted for coagulation process. It was carried out as a batch test, accommodating a series of six beakers together, each beaker used for testing was filled with 1000 ml of dye solution with 10, 25, 50, 75, 100 and 150 mg/L concentration and COD solution with concentration 200, 400, 600, 800, 1000, 1200 mg/L, in order to find the optimum pH, 0.1 M NaOH or 0.1M H_2SO_4 was added to each beaker for pH adjustment, pH meter was used thermo- orion 3 star, USA, final values of pH were 2, 4 , 6, 8 and 10 in beakers as a sequence, coagulant solution was added to each beaker with dosage of 100 mg/L. The initial rapid agitation speed was adjusted to be 200 rpm for 1 min, thereafter, the slow agitation speed was 30 rpm for 30 min. Settling time was made to be 30 min. After settling , about 2 mL of supernatant was withdrawn from the point located about 2 cm below liquid level for the determination of removal percentage of dye and COD. All tests were conducted at room temperature. The study was achieved by main conventional, but important control factors which were coagulants dosage, pH and dye concentration in order to study their effect in flocculation and obtain the optimum condition for each factor. The volume of sludge was estimated depending on the amount of precipitation. At the end of each run, the solution was filtered and the filtrate was centrifuged at 4000 rpm for 10 minutes, and then, was analyzed, The concentration of dye remnants were measured (λmax = 660 nm) using a spectrophotometer (thermo-genesys 10 UV, USA). The efficiency of COD removal were measured using a Lovibond Checkit direct COD Photometer (Germany), % Removal, after each run was estimated as follows:

$$R(\%) = \frac{(C_i - C_e)}{C_i} \times 100 \quad ....(1)$$

Where Ci is the initial concentration (mg/L) and Ce is the final dye concentration (mg/L).

3. Results and discussion

3.1 *pH results*

Effect of pH on RG-12 reduction from synthetic wastewater was found to be remarkable. The dosing rate of all coagulants was kept uniform as 100 mg/l , dye concentration 75 mg/l and COD concentration 800 mg/l. All trials were taken at different initial pH values of 2, 4, 6, 8,10. Coagulants were added and rapidly mixed for one minutes by a stirrer at 200 rpm and thereafter slowly mixed for 30 minutes at 30 rpm and sample was left over for 30 minutes. The supernatant was decanted off and its value for RG12 and COD was measured. From fig. 2 and fig.3, for Alum, RG12 and COD reduction were 95% and 86%, respectively at pH 8. For ferrous sulphate the removal was 70% and 50% for RG12 and COD, but, these results were at pH 2. Finally, for ferric chloride RG12 and COD reduction were 98% and 88% at pH 6. Ferric chloride showed more color and COD reduction. As it is clear with ferrous sulphate that the best value of pH for RG12 and COD was two which might be due to strong precipitation at acidic pH. As the precipitation increased, solution will more clearly give very low value of percentage removal. However, the percentage removal of RG12 and COD with alum was maximum at basic condition. This means that when applying ferric chloride and ferrous sulphate as coagulant favorable pH is acidic (12). However, for alum the favorable pH is basic [4].

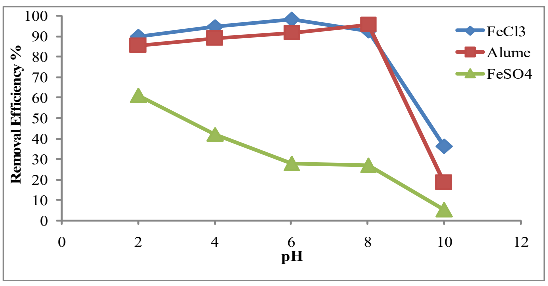

Figure 2: Reactive green 12 dye removal versus pH

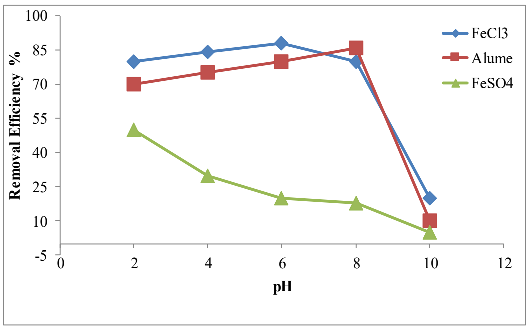

Figure 3: COD removal versus pH

3.2 *Coagulants dosage results*

According to fig.4 and fig.5, at constant pH 6, 8 and 2 for ferric chloride, alum and ferrous sulphate, and constant initial concentration of RG12 75 mg/l and COD concentration 800 mg/l. The coagulants dosage were 10, 25, 50, 100, 300, 400 mg/l. The highest degree of RG12 removal was obtained at 100 mg/l for ferric chloride, alum and 300 mg/l for ferrous sulphate, and the same dosage for all coagulants cause the highest degree of COD removal, from fig.4 and fig.5, it is clear that 100, 300 and 400 mg/l dosage of ferric chloride lead to the same removal percentage for RG12 98% and COD 85%, but for alum and ferrous sulphate their approach were slightly different as the RG12 and COD removal became less than 98% and 85%, i.e. there was a smallest difference in the efficiency of RG12 and COD removal. However, RG12 and COD removal were more percentage decreasing at 50, 25, and 10 mg/l dosage, especially at 25 and 10 mg/l, i.e. The results are increasing when the coagulants dosages increase. The result also shows that somewhat increase in coagulant dosage is desired for the removal of the wastewater containing reactive dyes and COD. As it is clear, the degree of dye removal is different from dye to another dye depending on their solubility in water [16], thus the degree of COD removal is also depend on the kind of dye. Therefore the demand of dye to the coagulant increase or decrease form one of dye to another. The flocculation process is also depending on the dye solubility in water [15]. These results can be agree with either researche [16,1,12].

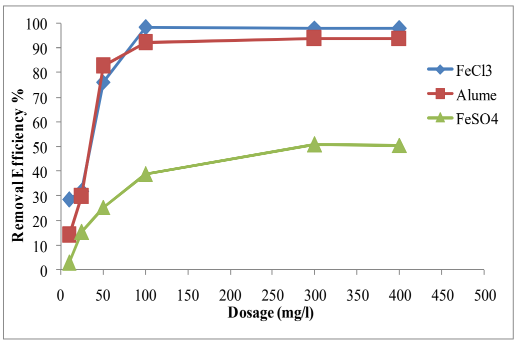

Figure 4: Reactive green 12 dye removal versus coagulants dosage

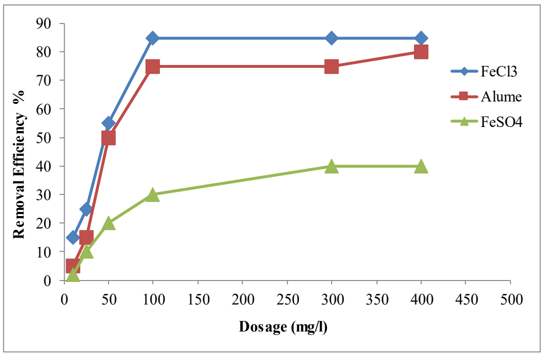

Figure 5: COD removal versus coagulants dosage

3.3 *Initial concentrations results*

The best concentrations for the selected dye solution was determined by varying the concentrations 10, 25, 50, 75, 100 and 150 mg/l and for COD concentrations 200, 400, 600, 800, 1000, 1200 mg/l and maintaining the best pH for Ferric chloride 6, alum 8, and ferrous sulphate 2, and dosage for ferric chloride and alum 100 mg/l, dosage for ferrous sulphate 300 mg/l. RG12 removal as a function of dye concentration is shown in Fig.6. It was observed that the percentage RG12 removal increases with the increasing in dye concentrations up to 100 mg/l to slow down at 150 mg/l. Nearly 99% RG12 removal was observed at 50 mg/l of ferric chloride, after that the percentage removal was constant, till the concentration reaches 150 mg/l, then, the removal was a little bit down. Alum was also found to be effective which gave 94% RG12 removal efficiency at a higher concentration of 150 mg/l. For the synthetic wastewater containing RG 12 dye had been observed to produce a normal percentage removal of 57% at 50 mg/l when it treated with ferrous sulphate. So, ferric chloride and alum were found to be quite effective more than ferrous sulphate. Some similar trends of COD removal were observed with increasing dye concentration as obtained in case of RG12 removal using ferric chloride, alum and ferrous sulphate (Fig.7). A maximum of 89% COD removal efficiency was observed at the optimum concentrations of 600 mg/L using ferric chloride. COD removal efficiency was 88% at 1200 mg/l by alum and 50% at 800 mg/l by ferrous sulphate. Ferric chloride gives better results for RG12 and COD removal. Limited researches were studied the dye concentration effect on coagulation process, therefore, smallest dye concentrations were studied as a result smallest removal efficiency obtained when dye was treated with polyaluminum chloride and alum[19].

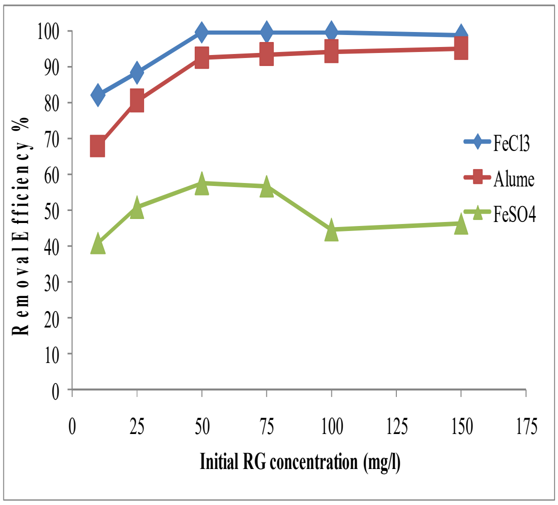

Figure 6: Reactive green 12 dye removal versus initial concentration

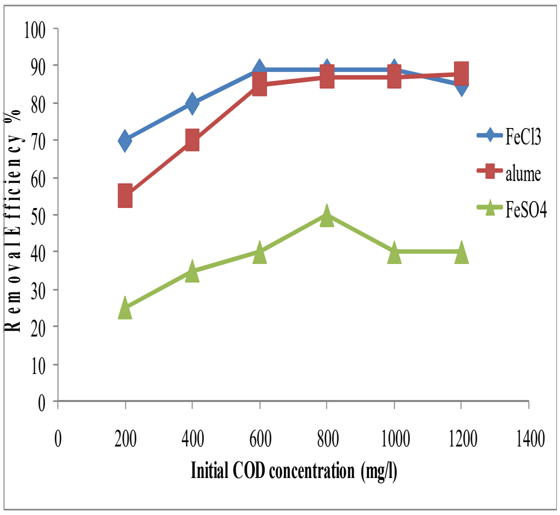

Figure 7: COD removal versus initial concentration

4. Sludge production results

In fact, coagulation treatment process produce a certain amount of sludge depending on the type of dye, type of coagulants and operating conditions [14], the relation between the removal efficiency and sludge production takes a direct relation. Sludge production was measured at the optimum conditions, it measured by the volume in ml in one litter of Imhoff cone after 24 hour of settling time. It can be seen in fig.8 the maximum volume of sludge was for alum coagulant 14 ml/l, 12 ml/l for ferric chloride and 0.5 ml/l for ferrous sulphate. In case of alum and ferric chloride the sludge production was approximate similar volume, which is more than ferrous sulphate, this mean the volume of sludge is proportion directly with the coagulation treatment efficiency. However, less sludge production related to less ability to reduction RG12 and COD. These results are similar to previous studies [3].

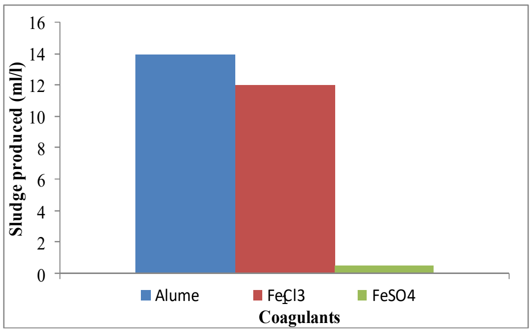

Figure 8: Volume of sludge produced versus coagulants

5. Conclusions

From this study the conclusions were:

• Each coagulant has got a different result of best pH and as follows : alum, ferrous sulphate and ferric chloride at 8, 2 and 6 pH respectively, this mean the removal efficiency was pH dependent.

•Ferric chloride performed more efficient than alum and ferrous sulphate, with low different in removal efficiency between ferric chlorite and alum. However, for the dosages more than 100 mg/L there is no increased in removal efficiency.

•With the increase of initial dye concentration more than 50 mg/L and initial COD concentration more than 600 mg/L, all coagulants showed similar

changing trends, however, the values of the removal efficiency for ferric chloride were remaining higher than other coagulants.

•The volume of sludge produced as a result of coagulant used follow the sequence as ferrous sulphate less than ferric chloride and ferric chloride less than alum.

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ازالة الصبغة الخضراء-12 والمتطلب الكيمياوي للاوكسجين من المياه العادمة المصنعة باستخدام عدة مخثرات

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نشر في: 30 حزيران 2019

الخلاصة – تم التحري عن قابلية استخدام كل من المواد الاتية :كبريتات الالمنيوم وكلوريد الحديديك وكبيرتات الحديدوزكمخثرات كيمياوية لازالة الصبغة الخضراء 12 والمتطلب الكيمياوي للاوكسجين من المياه العادمة المصنعة. اهم العوامل هي pH وجرعة المخثر وتركيز الصبغة تم ايجادها بواسطة فحص الجرة..اعلى كفاءة لازالة الصبغة الخضراء-12 والمتطلب الكيمياوي للاوكسجين مسجلة لكلوريد الحديديك كانت 98% و88% ولكبريتات الالمنيوم كانت 95% و88% ولكبرتات الحديدوز كانت 70% و50%.النتائج السابقة تم الحصول عيها لافضل القيم الاتية للدالة الحامضية 6 والجرعة 100 ملغم /لتر وتركيز لكل من الصبغة الخضراء -21 والمتطلب الكيمياوي للاوكسجين 50 ملغم /لتر و600 ملغم /لتر على الترتيب. حجم الحماة الناتجة كانت للمخثر كبريتات الالمنيوم بلغت 14 ملل/لتر و 12 ملل/لتر لكلوريد الحديديك و0,5ملل/لتر لكبريتات الحديدوز.الدراسة اثبتت انه من الممكن استخدام كل من كبريتات الالمنيوم وكلوريد الحديديك وكبريتات الحديدوز كمواد مخثرة اقتصادية لمعالجة المياه الملوثة بالصبغة الخضراء 12 والمتطلب الكيمياوي للاوكسجين.