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Removal of Aniline and from Aqueous Solutions by Coagulation/Flocculation–Flotation

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Authors' contributions

This work was carried out in collaboration between all authors. All authors read and approved the final manuscript.

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ABSTRACT

The aim of this study was to evaluate the efficiency of the coagulation/ flocculation and flotation process for aniline removal from aqueous solutions, which the dissolved air flotation (DAF) was applied in laboratory scale. After determination of the optimal condition of pH and the dosage of Polyaluminum chloride by coagulation/ flocculation process, the effect of the effective parameters including the concentration of the coagulant, coagulation time, flotation time and saturation pressure on the removal efficiency of the aniline and COD By dissolved air flotation was studied. The DAF process can reduce COD and aniline up to 89.6% and 95%, respectively. The optimum condition was as follows: pH=6, initial concentration of aniline= 200 mg/L, flocculation time = 10 min, flotation time= 20 sec, the pressure= 4 atm and PAC concentration of 20 mg/L. The coagulation/ flocculation -flotation (DAF) processes can an effective method to remove the aniline from aqueous solution.

Keywords: Aniline; coagulation; flocculation; flotation; removal.

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1. INTRODUCTION

Aromatic compounds are common pollutants in the effluents of several industries. Among these aromatic pollutants, aniline (slightly yellow-to-brown, clear oily liquid, Specific gravity: 1.02) is very hazardous compound to human as well as the environment [1,2]. Aniline is an organic compound with diverse applications in a broad range of industries, including in manufacturing of pharmaceuticals, dyestuffs, rubbers, pesticides, plastics and paints [2,3]. It, therefore, is frequently observed in the production line and wastes of the industries [3]. It can enter human body from water through their skin, by food, breathing of contaminated air and skin touch. Aniline vapor is heavier than air and may cause asphyxiation in enclosed, poorly ventilated or low-lying areas. When aniline was released, it disturbed the aqueous life cycle and cause cancerous tumors in animal and increases the risk of bladder cancer in human. Aniline decreases the blood's oxygen-carrying capacity by converting hemoglobin to Met hemoglobin [4]. Thus, due to its negative environmental impacts and threats to organism health, aniline removal is a major concern [2].

There are several methods for the elimination of aniline, including the application of photodecomposition [5], electrolysis [6], adsorption [7], oxidation [4,8], biodegradation [9] and other processes. However, these methods are mostly cost-effective and produce toxic by-products [2-8]. Aniline must be properly treated before their discharge to the water bodies, there is a need for developing technologies to remove aniline from wastewater rapidly [9].

Flotation is a physical approach in which the solid particles are transported to the surface layer from the liquid phase. The main preference of this approach over the sedimentation is that in the former method, fine particles with densities close to that of water and slow settlement can be withdrawn more efficiently and in shorter period of time [10]. Four main stages of flotation are as follow [11,12]:

1. Bubble generation in the wastewater
2. Contact between the gas bubble and the particle or oil droplet suspended in the water
3. Attachment of the particle or oil droplet to the gas bubble
4. Rise of the air/solids combination to the surface where the floated materials are skimmed off.

Dissolved air flotation (DAF) is one of the novel techniques for industrial wastes and wastewater treatment [2]. It is successfully used in purification of drinking water, and the removal of algae, humic substances and dense colors, and also in separation of volatile suspended materials, lipids and organic compounds; in conjugation with coagulation process [2,11]. Flotation is performed by releasing air bubbles into the flotation tank using pressure and then, by decrease in the pressure as it encounters the atmospheric pressure [13]. Coagulation and flocculation chemistry is an important factor in DAF process. Particle mobilization for bubble-particle attachment requires coagulation. This will result in the efficient removal of the particles. To make flotation method more effective for waste treatment, addition of coagulants is necessary for settlement or surface absorption of the organic substances [11,13]. Several chemicals are generally used to facilitate the flotation process. Polyaluminum chloride (PAC) and organic polymers have been widely used in more recent years [14,15]. Some of the advantages the PAC offers over simple salts are broader pH operating range, lower temperature sensitivity, lower residual metal ion concentrations, smaller sludge production and better sludge dewater ability [15]. Dissolved air flotation (DAF) process is more efficient and faster than sedimentation techniques, and it produces lower sludge volumes [16].

Some of the advantages of this method are its quick start, more solid wastes elimination, less dependence on coagulants and mostly on auxiliary polymers, suitable control of separation process, and high amounts of surface loads in small units [2,13]. Previous studies have reported that the organic matter removal of this process can be higher than 98% if the main working parameters were under control, such as the diameter of the bubbles, saturation time and saturation pressure [17]. The main purpose of this study was to assess the proper operating parameters, PAC concentration, and initial concentration of aniline, saturation pressure, coagulation time and time flotation for the full-scale DAF system for efficient removal of aniline.

2. MATERIALS AND METHODS

2.1 Chemicals

Aniline ($C_6H_5NH_2$) (molecular weight, 93.13 g/mol), was supplied by Merck, bought from Sigma Aldrich Co, Germany (99% purity). In this study, Polyaluminum chloride (PAC) $[(Al_{13}O_4(OH)_{24})]^{+7}$, Merck (Germany) was used as coagulant; hydrochloric acid (HCl) and sodium hydroxide (NaOH) was used to adjust pH in this study. The experimental DAF system consisted of a flotation Column, a compressor, an Unpack saturator column and a pressure gauge as shown in Fig. 1.

2.2 Stock Solutions

The experiments were performed to study the removal of aniline from aqueous solution. Stock solutions were prepared in accordance with the "Standard Methods" [18] the stock solution aniline (99% purity) was prepared with a concentration of 1000 ppm in double distilled water. Then aniline standard curve was plotted. By obtaining the uptake and use of linear regression obtained (y=0.097x+0.1535) in the calibration curve. The stock solutions of aniline [$C_6H_5NH_2$] were prepared daily and stored in amber colored glass bottles. The stock solution of the polyaluminum chloride [PAC, 1 g PAC/L] used for the experiments was 1000 mg/L. The stock solutions of aniline species were stored at 4^oC and that of PAC was stored at room temperature in a dark glass container.

2.3 Coagulation/Flocculation

Jar test experiments are conducted under controlled laboratory conditions. Coagulation/ flocculation was performed by the standard jar test method (Phipps and Bird, JLT6) at 380±2 rpm for 3 min rapid mixing and 20 min of slow mixing at 30±2 rpm After that the samples were allowed to sediment for an hour [19]. The optimum coagulant dose concentration and pH were determined for coagulation. The coagulant was added to each 1 L of sample water containing various concentrations of aniline. Prior to addition of coagulant, the sample water pH was adjusted by adding 0.1 N HCl (hydrochloric acid) or 0.1 N NaOH (sodium hydroxide) solutions in each bottle previously, jar test procedures were also accomplished for the optimal coagulation dosages and pH determination for aniline removal from aqueous solutions. All jar tests were carried out in a temperature controlled room at 20–23°C and each experiment was carried out in (at least) duplicate.

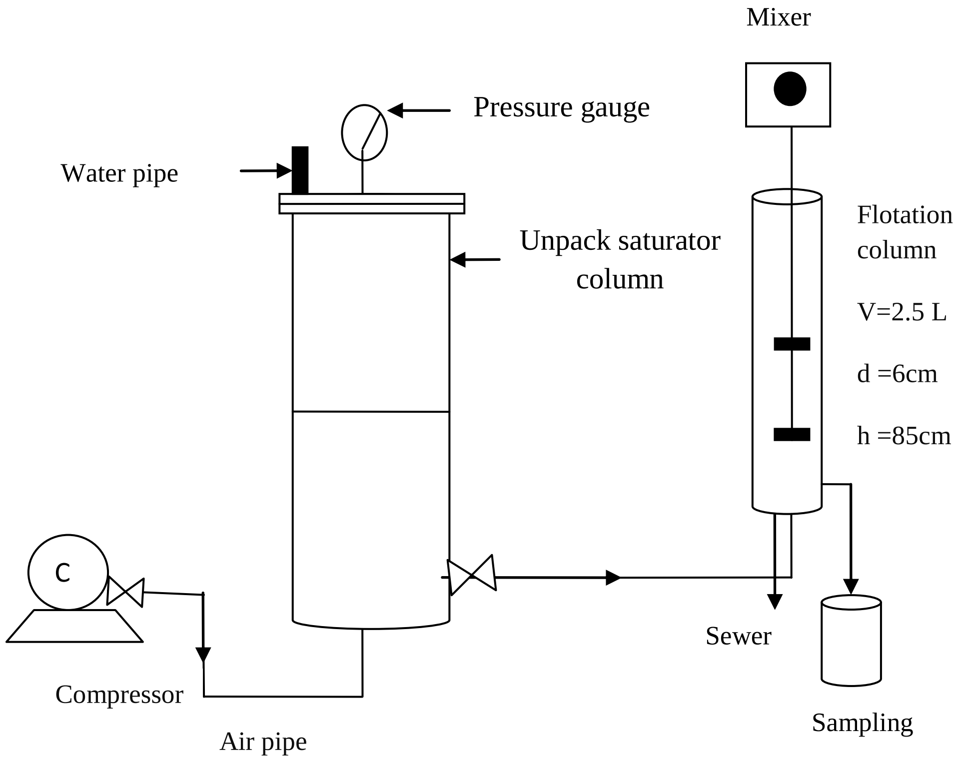

Fig. 1. Schematic of the experimental DAF system

2.4 Pilot DAF

A DAF pilot is made of three sections of combination-coagulation, flotation and saturation tank. Combination-coagulation unit is responsible for the production of flocs, and in saturation tank air-saturated water is produced. The DAF unit, as shown in Fig. 1, the bench scale DAF unit used for this study was designed to perform coagulation/flocculation and flotation operated in the same vessel. It can be consisted of an air compressor; a stainless steel unpacked saturator vessel and a flotation cell. The unit was designed so that coagulation, flocculation and flotation can be carried out in the same vessel. First, the distilled water was added to the pressurized tank. Then, coagulation and flotation processes were simultaneously accomplished in the following five steps to optimize the parameters. (i) the pH of the aniline (4 L)was adjusted according to the experiment and the sample was added to the flotation cell, (ii) PAC was added to the cell according to the design of experiment (iii) the aniline and PAC were rapidly mixed (380 rpm for 2 min) and slow mixed (30±2 rpm for 10min), (iv) water saturated with air was injected from the saturator into the flotation cell for 5s, and (v) flotation was allowed to occur and samples were collected from the sampling point.

Influx of the saturated water into the flotation tank produces very small bubbles. These bubbles stick to the clotted particles while moving to the water surface, by which will float the particles. The samples were then taken from the bottom of the cylinder at the depth of 3.5 cm through the sampling valve.

2.5 Analyses

The initial and final aniline concentrations remaining in solutions were analyzed by a UV–visible Recording Spectrophotometer, (Shimadzu Model: LUV-100A) Construction Japan was determined at a wavelength of maximum absorbance λ_{max} = 199 nm [2]. The pH was measured using a MIT65 pH meter respectively. Treated COD samples were analyzed using the standard method. The spectrophotometer used for measuring sample absorbance at a wavelength of 600 nm was a COD Reactor DR 5000 HACH spectrophotometer Germany. COD was measured according to Method 5220D (closed reflux, colorimetric method) [18]. The removal of the studied parameters from aniline was calculated based on the following formula:

$R = [\frac{(C_i - C_f)}{C_i}] \times 100$ Where C_i and C_f are the initial and final concentrations of the studied parameter.

3. RESULTS AND DISCUSSION

In order to investigate the effects of saturation pressure, Polyaluminum chloride, initial concentration of aniline, flocculation time and time flotation on DAF performance. Coagulants are useful in overcoming or reducing electrostatic barriers in order to allow closer approach between individual particles and particles and bubbles. There is an optimum chemical additive concentration at an optimum wastewater pH range. Therefore, the subsequent experiments were designated to find the most efficient combination of parameters, which would give a high level of aniline and COD removal. Separation of particles by flotation adheres to the same laws as sedimentation but in a "reverse field of force." The governing equation in air flotation separation, as in all gravity controlled processes, is Stoke’s Law (at least in laminar flow), which is used to compute the rise rate of bubble flocs, agglomerates, and bubble-oil aggregation (Fig. 2) [11,13].Studies have shown that conditions are necessary for favorable flotation such as charge neutralization of the particles, production of hydrophobic particles, floc diameter, bubbles diameter and rising velocity [13]. This results in the formation of flocs, which lead to more bubble-particle attachment during the DAF process. The bubble-particle attachment involves three mechanisms; (i) precipitation or collisions, (ii) bubbles trapped in a floc structure as the bubbles rise through the liquid media and (iii) bubbles adsorbed in a floc structure as the floc is formed [20]. theory presented above regarding particle-bubble collisions suggests that flocs of 10 to 30 microns (pinpoint floc) should be prepared for flotation [13].

3.1 Effects of Coagulant Dose PAC

Coagulants are useful in overcoming or reducing electrostatic barriers in order to allow closer approach between individual particles and bubbles. The initial stage of the experiment was designed to determine optimum amount of PAC coagulant for the aniline in coagulation/ flocculation and DAF reaction. The data in Fig. 3 show the effect of coagulant dose (polyaluminum chloride/ PAC) on the sedimentation and flotation of aniline at pH 6. The study also showed that the polyaluminum chlorides can the color, turbidity and algae remove effectively [21]. Polyaluminum chloride $[(Al_{13}O_4\ (OH)_{24})]^{+7}$ at 10, 20, 30, 40 and 60mg/L was used to determine the effect of coagulant concentration. The study also showed that a pH of 6 was optimal for the aniline removal [2,19]. The system was maintained at pH 6 optimal. Poly Aluminum Chloride is great power in the removal of organic contaminants and COD. Another the PAC characteristic is included hydroxide ions that, the hydroxide ions cause a small collection polymer of AL in the PAC. The main part of PAC sets up of AL_3^{+7}; these polymer structures have a better effect on unstable colloids [22]. Fig. 3 show aniline and COD highest removal efficiency in the sedimentation process is 93.34% and 88% at 60 mg/L polyaluminum chloride dose. Coagulation chemistry is the most important operating control variable affecting flotation performance. Without coagulation, the particles carry a negative charge and are often hydrophilic so that bubble attachment is poor. The data in Fig. 4 show the effect PAC dose on DAF process by flocculation Time 10 min, Flotation time 5 s and Pressure 3.5 atm and Concentration 20 mg/L of the PAC coagulant, the optimum coagulant for the wastewater was PAC, which has the best COD and aniline removal efficiencies about 80.63% and 91%, respectively, with increasing concentration of coagulant decreases removal. The sedimentation processes require large floc particles (100 µm) with densities greater than water. Flotation does not require large floc particles and heavy, which leads to a reduction in chemicals dosage. The floc particle densities less than water are required and are achieved by attachment of air bubbles to floc particles [13,19]. In total dissolved air flotation systems is required a dose less than material polyaluminum chloride/ PAC.

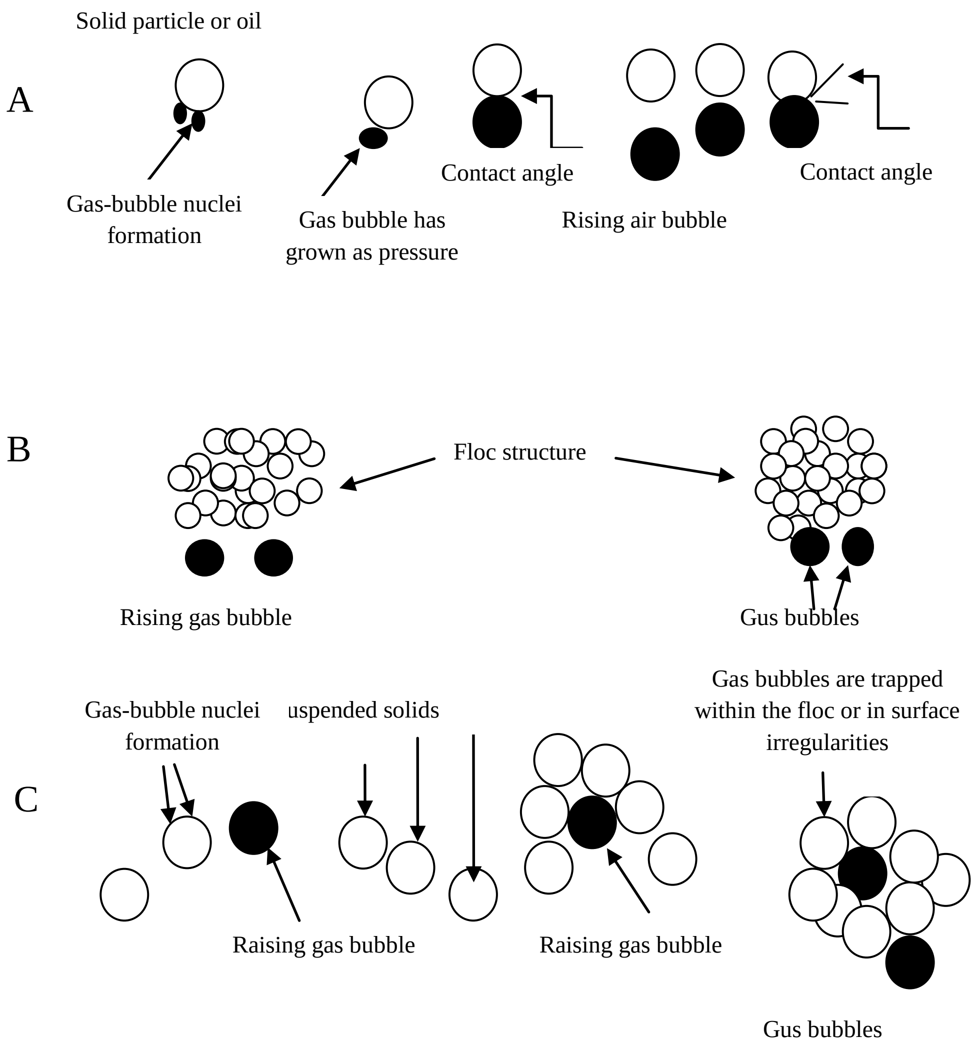

Fig. 2. Mechanisms of bubble/droplet formation and adhesion in DAF

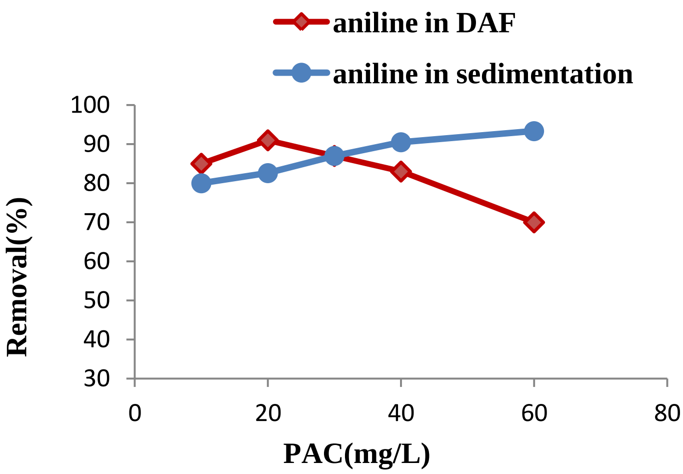

Fig. 3. Removal efficiencies of Aniline and COD by DAF and sedimentation at different doses of coagulant PAC

3.2 Effect of Initial Aniline Concentration

The effect of initial aniline concentration of aniline and COD the removal of DAF on various doses (50, 100, 150 and 200 mg/L) was studied as shown in Fig. 4. The system DAF with increasing concentration of aniline from 50 to 200 ppm, the percentage removal of the aniline was found to increase with the increase in initial aniline concentration. The results show that the percentage removal of the aniline and COD were 94.51% and 83.15% in initial aniline concentration 200 mg/L and the COD concentration 179 mg/l respectively. The dissolved air flotation method is considered of removal materials with lower density or equal water density. Most of biodegradable organic matter and chemical materials are favorable removal and particle accumulation with density less than water (1 gram per cubic centimeter) is expected to be floating [13]. So for desired flotation, two conditions are necessary to neutralize the charge of the particle and hydrophobic particles. The increasing aniline concentration can provide favorable conditions to form a complex of floc-bubble which cause the aniline to float. Variations in aniline removal rates from the water are due to the changes in the floc conditions in case of size and vitality, and charge interaction between bubbles and flocs. The reason could be that the aniline ion is also involved in Bubble-floc attachment, stabilizing their linkage. Bubble-floc binding strength can also be because of the bubble-floc charge interaction and the hydrophilic effect of the binding water on the particles surface [13,19]. Studies on arsenic removal by dissolved air flotation showed that increasing arsenic concentrations has been increase removal [19].

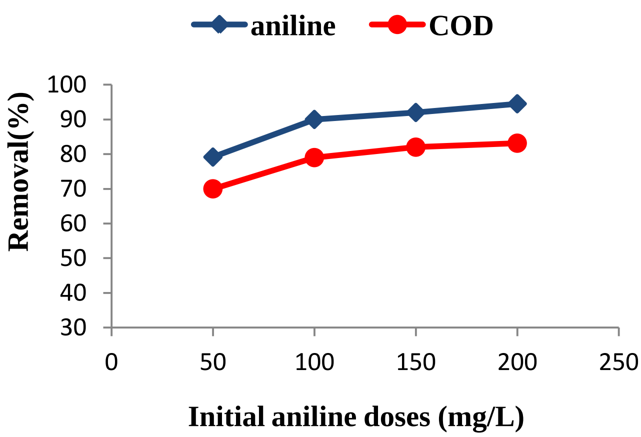

Fig. 4. The effect of initial aniline different doses on DAF in removal Aniline and COD

3.3 Effect of Flocculation Time

The flocculation time is one of the operating parameters. Fig 5 represents the effect of flocculation time using 20 mg/dose of coagulants, for removal of aniline and COD. Fig. 5 also shows the impact of flocculation time in this study of contaminant removal performance was evaluated in 5 to 20 min (PAC concentration of 20 mg/L, flotation time of 5s and presser 3.5 atm and aniline concentration of 200 ppm). The optimum flocculation time was found to be 10min. The highest removal of aniline and COD were found to be 95.41% and 85.36% using DAF respectively. An increase in the coagulation time from 10 to 20 has led to a decrease in aniline removal from 95.5% to 94%. Due to the increase in flocculation time, larger flocs are produced which cannot float, resulting in the reduced purification efficiency. Hence, DAF technique does not require long coagulation time, and shorter time would be more efficient and cost-effective [13]. A study has shown that DAF is successful when less-dense or pointed and spongy flocs are produced in the process [23].

3.4 Effect of Flotation Time

Aniline & COD removal efficiency increased with increasing flotation time (Fig. 6). Affect of flotation time under the optimal conditions (PAC 20 mg/L, pH 6, initial aniline dose 200 mg/L) the highest removal ratio of COD and aniline was about 89.2% and 95.57%, respectively. On the other hand, the optimum flotation time for the DAF process, at the time flotation 20s. This result shows that the long flotation increases removal rate of aniline [2]. At longer flotation times, due to sufficient time for connecting bubble to particle and production of larger bubbles, more raising the bubble-particle happens. Karhu in 2014 reported that the maximum removal was at the flotation time of 27 sec [24].

3.5 Effect of Saturation Pressure

The results shown in Fig. 7, the effect of air pressure on COD and aniline removal optimum condition PAC 20 mg/l initial aniline dose, flocculation time 10 min and flotation time 20s was studied for the determination of the critical pressure. It is observed that the aniline and COD maximum removal occurred at a pressure 4 atm of 95% and 86.6% respectively. These conditions were generally more favorable for collisions and adherence among the bubbles and suspended particles. In order to ensure the formation of bubbles at suitable diameters of 10 to 100 micrometers, saturation pressure of about 4 is recommended [25]. The bubble core diameter (d_{cb}) is calculated by the following formula [13]:

$$d = \frac{4\delta}{\Delta p}$$

Where δ is surface tension and ΔP is the change in the pressure in the nozzle.

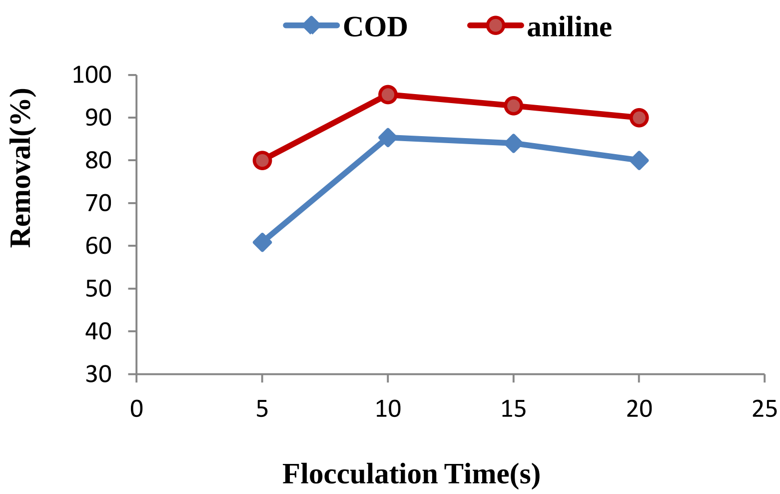

Fig. 5. The effect of flocculation time different on DAF in removal aniline and COD

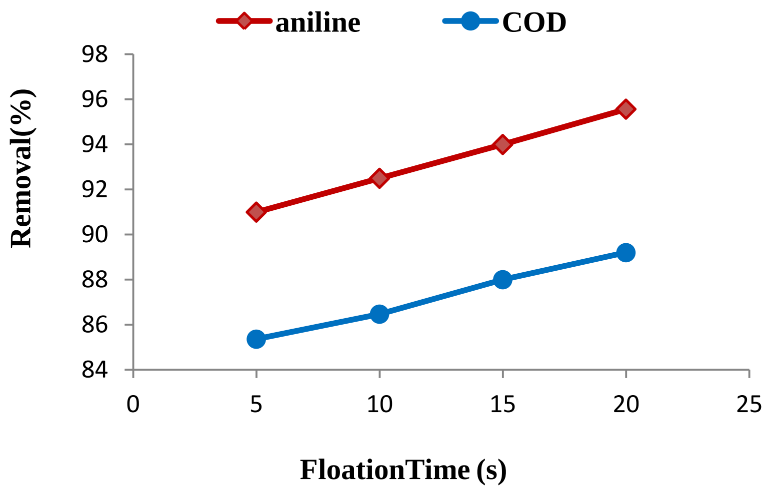

Fig. 6. The effect of time flotation different on DAF in removal aniline and COD

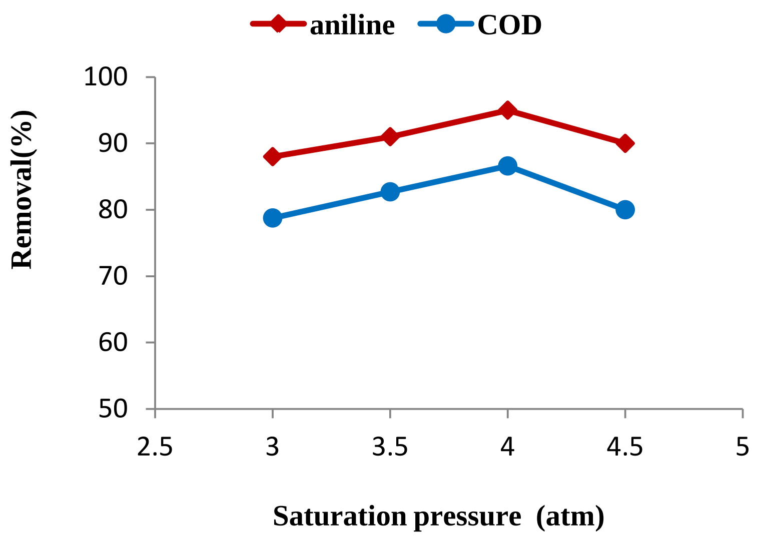

Fig. 7. The effect of saturation pressure different on DAF process in removal aniline and COD

Bubble diameter estimation in flotation system with dissolved air reveals that the bubbles with an average diameter of 40 micrometers are suitable for binding to the flocs [13,19]. In fact, with increase in the saturation pressure to up to 4.5 the bubble size decreases elevating spherically under the conditions of laminar flow and in accordance with the Stokes theorem. In contrast, in low saturation pressures, DAF produces larger elliptical bubbles with higher rising velocities that break the flocs [2,13]. As a result, the removal efficiency has decreased. Furthermore, DAF is effective when the bubbles have much higher volumes than the particles. This will help with more efficient removal of the particles by bubbles and lowers the density of flocs. Also, adequate concentration of the bubbles causes proper encounter of particles and bubbles and will finally decrease the density of the flocs [2,19]. Pressure to be as important as other factors indicate that slight increasing in these parameters to 4atm resulted in slightly enhanced removal of COD and aniline.

4. CONCLUSIONS

Dissolved air flotation (DAF) process is influenced by different parameters. The results of this study indicate that the dissolved air flotation process to conclusion requires a lower dosage

Poly Aluminum Chloride. Optimum conditions for the operation of the system dissolved air flotation with flotation time of 20 seconds and pressure of 4 atmospheres can remove a large impact on the concentration of aniline in water.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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