



Survey of Efficiency of Dissolved Air Flotation in Removal Penicillin G Potassium from Aqueous Solutions

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Authors' contributions

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

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ABSTRACT

Antibiotics are a large group of pharma compounds which are stable in environment. Antibiotics are considered among the major pollutants in water environments. The aim of this study was to evaluate the efficiency of the dissolved air flotation in removal penicillin G potassium from aqueous solutions. This study was an empirical-lab study which the dissolved air flotation was applied in laboratory scale. After determination of the optimal condition of pH and the dosage of poly aluminum, the effect of the effective parameters including the concentration of the coagulant (20, 50, 75, 100 mg/L), penicillin G (25, 50, 100,200), flotation time (5, 10, 15 and 20 sec), saturation pressure (3, 3.5, 4 and 4.5 atm) on the removal efficiency of the penicillin G and COD by dissolve air flotation was studied. The results showed that the dissolved air flotation can reduce COD and

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penicillin G up to 70.41% and 67.45%, respectively. The optimum condition was as follows: pH=6, initial concentration of penicillin G = 25 mg/L, coagulation time = 10 min, flotation time= 20 sec, saturation pressure= 4 atm and PAC concentration = 20 mg/L. Also, the results showed that the removal efficiency is reduced by increasing the turbidity in the flotation process. The results of this study revealed that the dissolved air flotation process can be effective method to remove the penicillin G from aqueous solution.

Keywords: Dissolved air flotation; aqueous solution; penicillin G potassium; removal.

1. INTRODUCTION

Drugs are used to prevent or treat disease. Most drugs include wide variety of chemicals such as antibiotics, analgesics, hormones, vaccines, vitamins, dietary supplements, disinfectants, etc [1]. Antibiotic refers to a material that can be used for the elimination of microorganisms such as bacteria, fungi, and parasites [2]. Antibiotics are among the most beneficent drugs, however, they have potential harmful effects on environment, including entrance into soil and water resources and cause to develop antibiotic resistance microorganisms [3]. Penicillin G is a common antibiotic used for the treatment of different kinds of infectious diseases (Table 1). Penicillin G is formed by 6-amino-Penicillin acid and a benzyl chain. This antibiotic consists of a beta lactam (B-lactam) and is very sensitive to the pH, heat, and beta-lactam enzyme [4]. Penicillin G potassium is a colorless or white crystal, or a white crystalline powder which is odorless, and moderately hygroscopic. Penicillin G potassium is highly soluble in water. The pH of the reconstituted product is in the range of 6.0–8.5. The mechanism of action of penicillin G is the destruction of bacteria's cell wall by preventing the peptidoglycan production [5]. The chemical structure and general characteristics of aniline are presented in Table 1 and Fig. 1. This generally makes antibiotics as one of the important drugs for the treatment of bacterial infection, which are very useful in medicine.

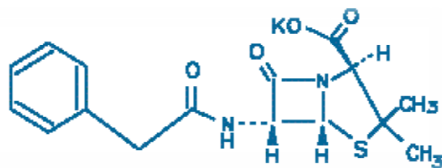


Fig. 1. The chemical structure Penicillin G potassium

Several treatments methods have been proposed for the removal of penicillin G from

contaminated waters, including photo-decomposition [6,7], electrolysis [6,8], adsorption [6], oxidation [6,8], biodegradation [6], and other processes, involving advanced technologies to destroy the Penicillin G structure. However, these treatments cannot be widely applied because they are expensive and time-consuming. Thus; there is a need for developing technologies to rapidly remove Penicillin G from wastewater [9,10]. Flotation is an operation used to separate particles from a liquid phase. Flotation has a great ability to efficiently separate low density particles, compared to the sedimentation process. Dissolved air flotation (DAF) is a physicochemical process by which mineral particles used to be separated in the early 1900s [10,11]. In other words, dissolved air flotation is a solid/liquid separation process in which microscopic air bubbles (10–100 μm) become attached to solid particles suspended in liquid, causing the solid particles to float [12]. Gravity settling is not usually a feasible treatment method for stable oil emulsions due to the presence of extremely fine droplets, which have a very low settling rate. DAF is used to enhance the buoyancy difference of emulsified-oil droplets by the attachment of small air bubbles [13]. The DAF process is more efficient and faster than sedimentation techniques and produces lower sludge volumes [14]. Air flotation is widely used to treat oil-bearing effluents from various sources: refineries, ship's bilge and ballast waste, deinking operations, metal plating, meat processing, laundries, iron and steel plants, soap manufacturing, chemical processing and manufacturing plants, barrel and drum cleaning, wash rack and equipment maintenance, glass plants, soybean processing, mill waste, and aluminum forming [13]. The main purpose of this study was to assess the proper operating parameters, PAC concentration, and initial concentration of penicillin G potassium, saturation pressure, coagulation time and time flotation for the full-scale DAF system for efficient removal of penicillin G potassium.

Table 1. Some physical and chemical properties characteristics of penicillin G [5]

Biological half-life	Solubility	Excretion method	Efficiency mechanism	Molecular weight	Chemical formula
30-60 minutes	Soluble in water completely	Kidney	Prevention from wall -cell synthesis	372.48	C ₁₆ H ₁₇ KN ₂ O ₄ S

2. MATERIALS AND METHODS

The experiments were carried out in duplicates in the batch mode. The study parameters included pH, PAC concentration, initial antibiotic concentration, saturation pressure, coagulation time, time flotation, and turbidity.

2.1 Chemicals

Penicillin G (C₁₆H₁₇KN₂O₄S) with 100% purity was purchased from Sigma- Aldrich Company (Germany). Other chemical products were purchased from Merck (Germany). In this study, Polyaluminum chloride (PAC) [(Al₁₃O₄ (OH)₂₄]⁺⁷ 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 (Volume =2.5 L, diameter =6 cm, height =85 cm), a

compressor, an Unpack saturator column (Diameter=10 cm, Height=170 cm) and a pressure gauge as shown in Fig. 2.

The purpose of experiments was to study the removal of penicillin G from aqueous solution. Stock solutions were prepared in accordance with the “Standard Methods” [15]. The stock solution of penicillin G was prepared with a concentration of 1000 ppm in double distilled water. Then standard curve of penicillin G was plotted (Fig. 3).

Turbid water for DAF tests was prepared by adding 1 g kaolin to 1 L tap water. The suspension was stirred for 1 h to achieve uniform dispersion of kaolin particles, and then was kept for 24 h for completing the hydration of the particles. This suspension was used as the stock suspension. Turbid water with 50 mg/L kaolin

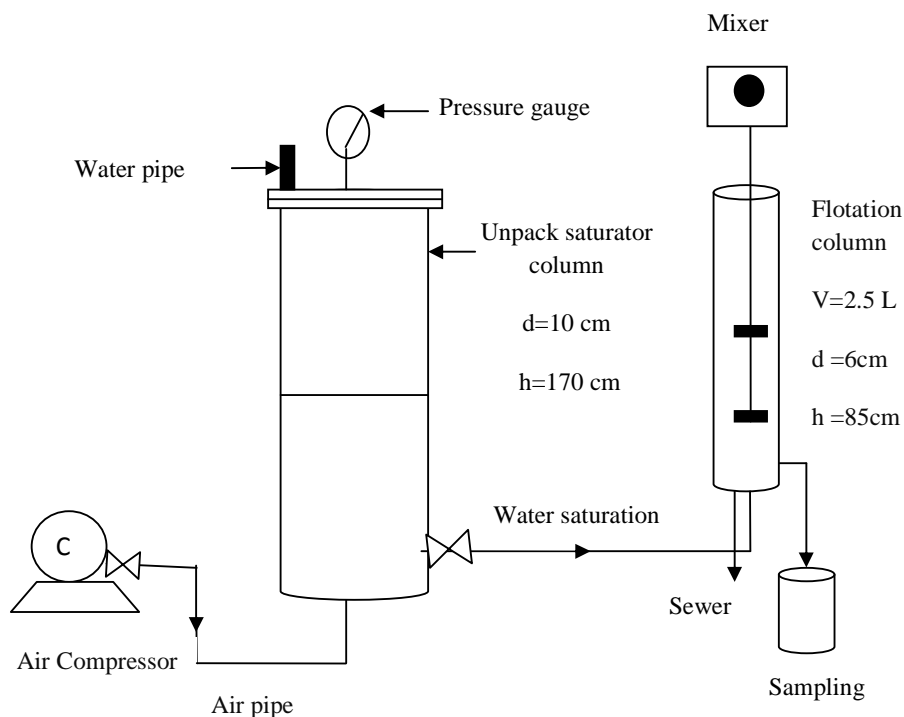


Fig. 2. Schematic of the experimental DAF system

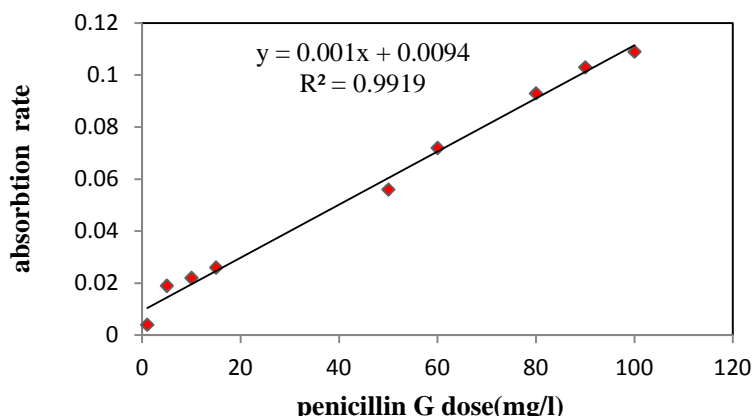


Fig. 3. Penicillin G standard curve

(about 35 nephelometric turbidity units NTU) was prepared by diluting 50 mL of stock suspension to 1000 mL tap water just before the coagulation test [16].

2.2 Application of DAF

A bench-scale DAF unit was employed as shown in Fig. 2. It usually consists of an air compressor; a stainless steel unpacked saturator vessel, and a flotation cell. The unit was designed so that coagulation and flotation can be performed in the same vessel. The optimum coagulant dose concentration and pH were determined by DAF. The coagulant was added to each 1 L of sample water containing various concentrations of Penicillin G. Prior to addition of coagulant, the pH of the water sample was adjusted by adding 0.1 N HCl (hydrochloric acid) or 0.1 N NaOH (sodium hydroxide) solutions in each bottle. The coagulation and DAF processes were performed as follow: (i) the pH of the penicillin G (Volume = 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 penicillin G and PAC were rapidly mixed (380 rpm for 2 min) as well as slowly mixed (30±2 rpm for 10 min); (iv) water saturated with air was injected through the saturator into the flotation cell for 5s; and (v) flotation was allowed to occur and samples were collected From the sampling point. After flocculation, appropriate quantities of saturated water were added flotation continued, and samples were collected for analyses.

2.3 Analyses

Penicillin G was analyzed by spectrophotometric method. The initial and final penicillin G

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 $\lambda_{max} = 248$ nm. 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. Turbidity was measured using a turbid meter (TURB 355 IR) and it was expressed in nephelometric turbidity units (NTU). COD was measured according to Method 5220D (closed reflux, colorimetric method) [15]. The removal of the studied parameters from penicillin G was calculated based on the following formula:

$$\text{Removal Percentage} = \left[\frac{C_i - C_f}{C_i} \right] \times 100 \ .$$

Where C_i and C_f are the initial and final.

3. RESULTS AND DISCUSSION

The effects of saturation pressure, Poly aluminum chloride, initial concentration of Penicillin G, flocculation time and time flotation on DAF performance were investigated. Coagulants play an important role in reducing electrostatic barriers, which leads to aggregation of particles and bubbles with a density lower than that of water. 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 Penicillin G and COD removal. Separation of particles by flotation follows the same laws as the 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. 4) [17,18]. 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 [17]. 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) the bubbles trapped in a floc structure as the bubbles rise through the liquid media, and (iii) the bubbles adsorbed in a floc structure as the floc is formed [19]. The theory presented above regarding particle-bubble collisions suggests that flocs of 10 to 30 microns (pinpoint floc) should be prepared for flotation [17].

3.1 Effects of Coagulant Dose PACL

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 PACL coagulant for the penicillin G in DAF reaction. The study also showed that a pH of 6 was optimal for the penicillin G removal. The data in Fig. 5 show the effect of coagulant dose (polyaluminum chloride PACL) on the flotation of penicillin G at pH 6. Polyaluminum chloride $[(Al_{13}O_4(OH)_{24})^{+7}]$ at 20, 50, 70, and 100 mg/L was used to determine the effect of coagulant concentration. Some of the PACL advantages include: coagulation efficacy is from 2 to 3 times higher than ordinary Aluminum salts. Flock formation occurs rapidly so that a short time is needed for the reaction and sedimentation. Using a similar dose of coagulants, the reduction of pH after the PACL consumption is less than other mineral coagulants. It means less sensitivity to temperature and less residual than other metallic coagulants, which lead to the reduction of produced sludge and facility of dewatering. These are factors that increased the PACL consumption for water treatment [20]. Coagulation chemistry is the most important

operating control variable affecting the flotation performance. Without coagulation, the particles carry a negative charge and are often hydrophilic so that the bubble attachment is poor. Good coagulation chemistry depends on the coagulant dose [17]. The data in Fig. 4 show the effect PAC dose on DAF process with a flocculation time of 10 min, flotation time of 5s and pressure of 3.5 atm, which has the best COD and penicillin G removal efficiencies of about 60% and 54.5%, respectively. The optimum coagulant was 20 mg/l PAC so that the removal decreased as the concentration of coagulant increased. Sedimentation processes requires large floc particles (100 μ m) with densities higher than water. Flotation does not require large and heavy floc particles, which leads to a reduction in chemicals dosage. Floc particle densities less than water are required and achieved by the attachment of air bubbles to floc particles [17,13]. Generally, dissolved air flotation systems require a less dose of coagulant material of poly-aluminum chloride (PACL). Biesinger et al. tested the efficiency of a DAF recycle system on beef-packing-plant wastewater with and without the use of chemicals. Without chemicals, 73% of the influent oil and grease (initial concentration of 300 mg/L) was removed whereas with the addition of 10 mg/L coagulant, the removal level was increased to 86% [21].

3.2 Effect of Initial Penicillin G Concentration

The effect of initial antibiotic concentration of penicillin G and COD on various doses (50, 100, 150, 200 mg/L) as well as the effect of removal rate of the penicillin G at the optimal condition (pH = 6, PAC = 20 mg/L) are shown in Fig. 6. As found from this figure, the antibiotic removal rate decreased from 59.5% to 45% as the initial penicillin G concentration was increased from 25 to 200 mg/L. The results show that the removal percentage of the penicillin G and COD were 50% and 54.5% for the initial penicillin G concentration of 25 mg/l and the COD concentration of 165.9 mg/l, respectively. The dissolved air flotation method is considered for the removal of materials with a lower density or similar to water density. Most of biodegradable organic and chemical materials have densities less than water and then are favorable in terms of removal variation (1 gram per cubic centimeter) [17]. So for desired flotation, two conditions are necessary to neutralize the charge of the particle and hydrophobic particles. The

decreasing penicillin G concentration can provide favorable conditions to form a complex of floc-bubble which cause the penicillin G to float [17, [13].

3.3 Effect of Flocculation Time

The flocculation time is one of the operating parameters. Fig. 6 represents the effect of flocculation time using 20-mg/L dose of coagulants for removal of penicillin G and COD. The consistence increment of removals was revealed with increasing the flocculation time up to 10 min, and then the removal was decreased. Fig. 7 also shows the impact of the flocculation time on the contaminant removal performance

during from 5 to 20 min (PAC concentration = 20 mg/l, flotation time =5s and pressure= 3.5 atm, penicillin G concentration =25 ppm).

The optimum flocculation time was found to be 10 min. The highest removal of penicillin G and COD at 10 min were btained as 54.5% and 59.55% using DAF, respectively. In DAF process, the aim is to produce small "pin points" flocs of about 30 um. So flocculation time of about 10 min is recommended for DAF process. Studies show that the best flocculation time is between 5 to 20 min for DAF process [10,17]. Han and colleagues also suggested that the best removal could be provided at a flocculation time of 10 min [22].

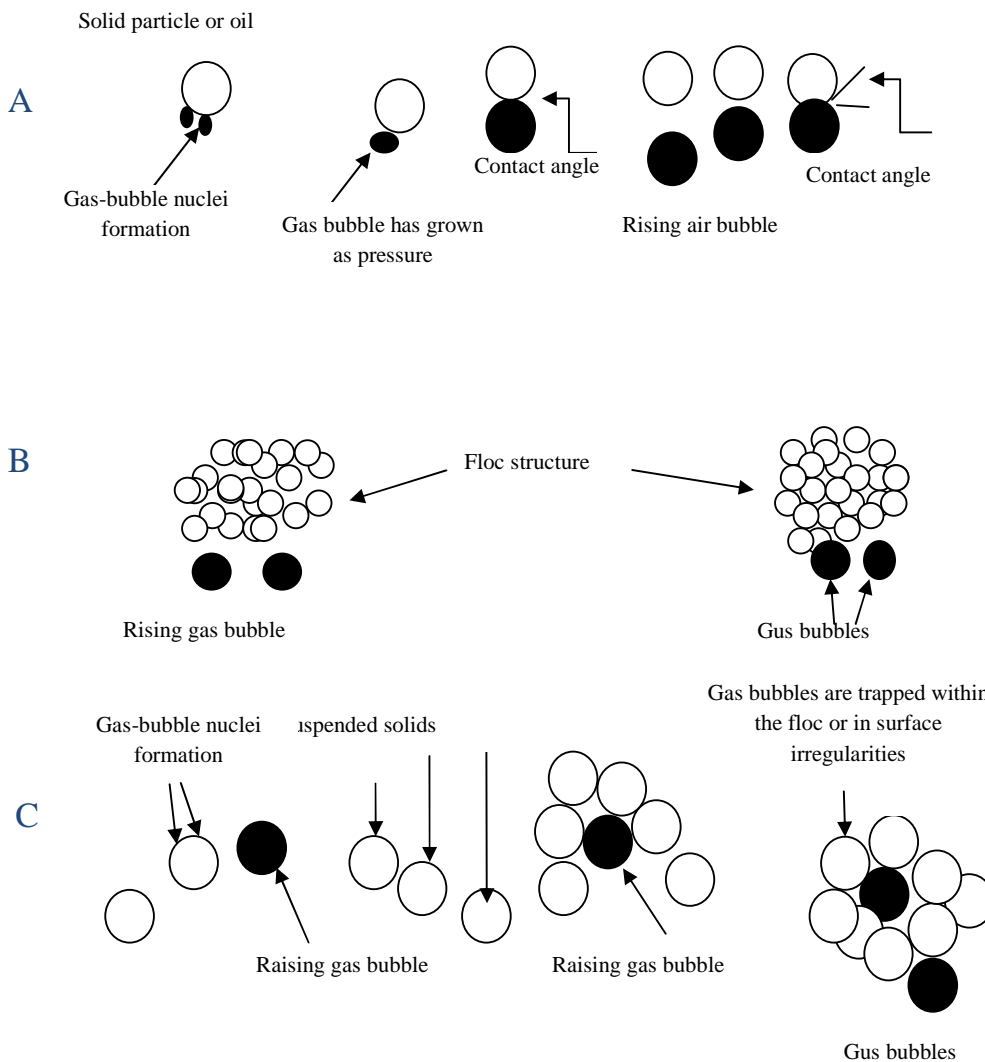


Fig. 4. Mechanisms of bubble/droplet formation and adhesion in dissolved air flotation

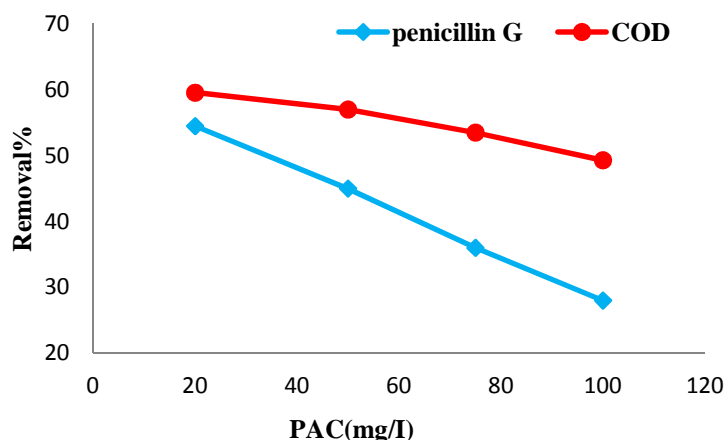


Fig. 5. Removal efficiencies of penicillin G and COD by DAF at different doses of coagulant PACL

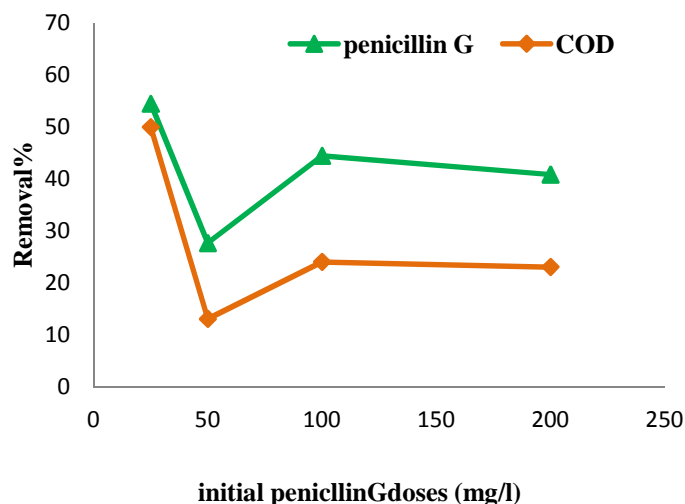


Fig. 6. Effect of initial penicillin G different doses on dissolved air flotation in removal penicillin G and COD

3.4 Effect of Flotation Time

Air flotation systems are normally treating large volumes of water. Hence, the detention time in the air flotation chamber becomes a very important processing variable. The removal efficiency of Penicillin G and COD were increased with increasing the flotation time (Fig. 8). The effect of flotation time under the optimal conditions (PAC=20 mg/L, pH=6, initial Penicillin G dose = 25 mg/l) indicates that the highest removal ratio of COD and Penicillin G was about 59.55% and 54.5%, respectively.

On the other hand, the optimum flotation time for the DAF process was at the flotation time of 20s.

This result shows that the long flotation increases the removal rate of aniline and Arsenic [10,17]. At longer flotation times, due to sufficient time for connecting bubble to particle and production of larger bubbles, more rising the bubble-particle happens. Karhu in 2014 reported that the maximum removal was at the flotation time of 27s [23].

3.5 Effect of Saturation Pressure

The most important dependent variable in air flotation systems is bubble size. The bubble sizes reported by Vrablik ranged from approximately 45 to 115 mm with mean diameters of 75–85 mm at 20 and 50 psi

saturation pressures, respectively. They obtained an average bubble size of 66 mm and 42 mm at 30.5 and 40 psi, respectively [24].

In the results shown in Fig. 9, the effect of air pressure on the removal of COD and penicillin G at optimum condition (PAC = 20 mg/l, initial penicillin G dose= 25, flocculation time =10min, flotation time = 20s) was studied for the determination of the critical pressure. It is observed that the maximum removal of penicillin G and COD was 67.5% and 70.41%, respectively, at a pressure 4 atm. These

conditions were generally more favorable for collisions and adherence among the bubbles and suspended particles. However, by increasing the saturated pressure to 4.5 atm, the bubble diameter became smaller and the removal efficiency of the penicillin G was reduced. The generation of bubbles depended on the pressure in the air dissolving vessel. The amount of bubbles increased as the size of bubbles was decreased and the pressure was increased. At low saturation pressure, the dissolved air flotation process will produce bubbles with larger diameters, which have high rising velocity and

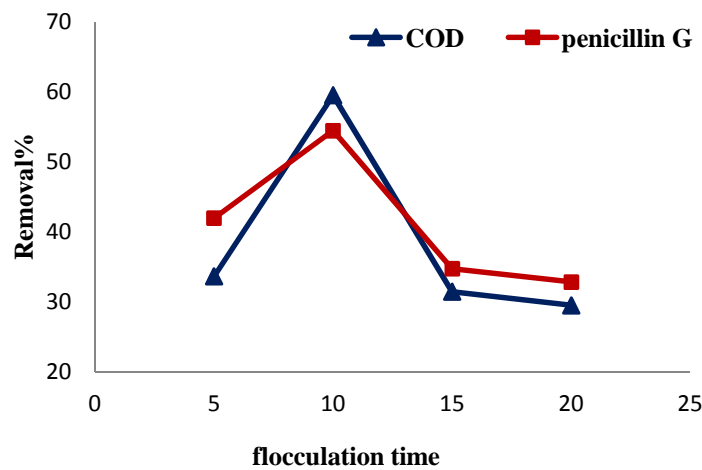


Fig. 7. Effect of flocculation time different on dissolved air flotation in removal penicillin G and COD

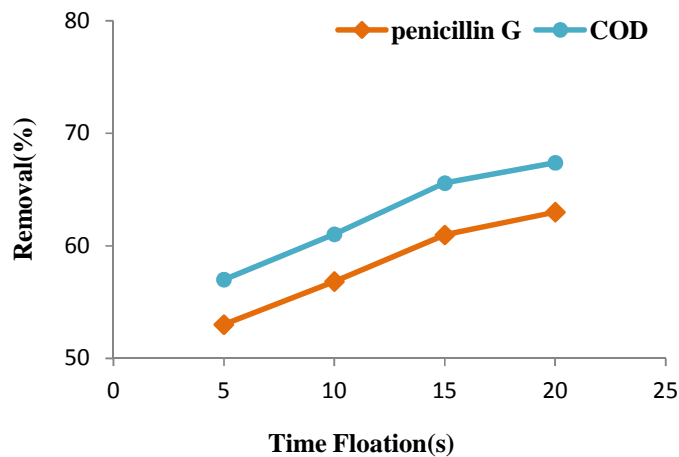


Fig. 8. Effect of time flotation different on dissolved air flotation in removal penicillin G and COD

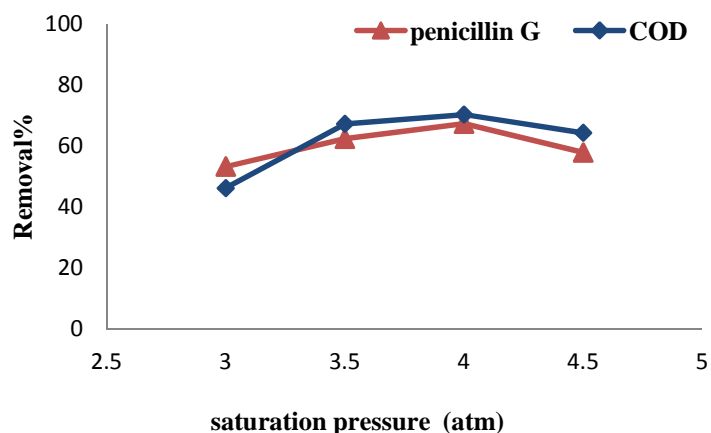


Fig. 9. Effect of saturation pressure different on dissolved air flotation in removal penicillin G and COD

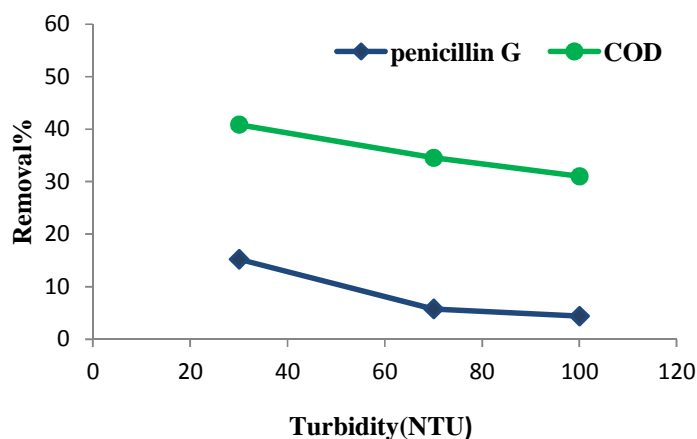


Fig. 10. Effect of Turbidity different on dissolved air flotation in removal penicillin G and COD

make the floc break down [17]. As the optimal bubble size for typical wastewater systems is in the order of 100 μ m or below (as will be described later), pressure differences of 4–5 atm are usually selected [13,25]. Sufficient concentrations of bubbles result in collisions between particles and bubbles, which then causes lowering the density of floc [10,13,17]. Overall, the pressure is the most factor in enhancement of removal efficiency of COD and penicillin G.

3.6 Effect of Turbidity

Fig. 10 above shows the residual turbidity observed as a function of contact time for the range of currents used in this study. The lowest residual turbidity of 100 NTU for the DAF occurred at a saturation pressure of 4 atm and a

flocculation time of 20s. When the turbidity was increased to 100 NTU, the decreased removal turbidity was observed. At high turbidity, the dissolved air flotation process will produce floc with larger diameters, which have high turbidity make the removal efficiency is reduced [10,26].

4. CONCLUSIONS

The experimental study shows that the dissolved air flotation process can give good efficiency in the removal of penicillin G potassium. The optimum conditions for parameters involved in the removal of COD and penicillin G as 70.41% and 67.5% was obtained at pH = 6, the initial concentration of penicillin G of 25 mg/l, the flocculation time of 10 min, flotation time of 20 s, saturation pressure of 4 atm, and the PAC concentration of 20 mg/l. Dissolved air flotation

process is influenced by different parameters. The results of this study indicate that the dissolved air flotation process requires a lower dosage of Poly Aluminum. Optimum conditions for the operation of the dissolved air flotation system with flotation time of 20 s and pressure of 4 atm can significantly affect the concentration of penicillin G in water. Also, the results showed that the removal efficiency is reduced by increasing the turbidity in the flotation process.

CONSENT

It is not applicable.

ETHICAL APPROVAL

It is not applicable.

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COMPETING INTERESTS

Authors have declared that no competing interests exist.

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