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RESEARCH ARTICLE

Bio-Adsorption of Aniline from Aqueous Solutions using Activated Raw Sludge

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Abstract

Aniline is a simple aromatic ring compound with wide application in several industries. Since it is soluble and highly toxic, aniline is threatening to human and environment and can disturb aqueous species life. Aniline also has mutagenic and carcinogenic properties. The aim of the present study was to eliminate aniline from aqueous solutions by the use of activated raw sludge. In this experimental study, the efficiency of activated raw sludge in eliminating aniline along with the impact of other influential variables such as pH (3, 5, 7, 9, 11), contact time (0-120 minutes), adsorbent dose (0.2, 0.4, 0.6, 0.8, 1.5 mg/L) and aniline initial concentration (10-250 mg) was investigated. Adsorption isotherm was assessed using Freundlich and Langmuir isotherms. The results indicated that the maximum efficiency of activated raw sludge in eliminating aniline was 80% which was achieved at the optimum condition of pH 11, contact time of 30 minutes, 0.2 mg/L adsorbent dose and aniline initial concentration of 200 mg/L. In addition, isotherm data showed the most fitness into Langmuir adsorption model (R2 0.990). Therefore, activated raw sludge possesses a high capacity to eliminate aniline from aqueous solutions and chemical industry wastewater.

Keywords: *Aniline, adsorption, sludge, adsorption, isotherms, Langmuir*

Introduction

Growing number of chemical pollutants enter the environment and natural resources through increased volume of wastewater flow with industrialization. Due to the huge petroleum resources and petrochemical and related industries, the standard environmental condition regarding disposal of wastewater is a major concern in Iran (1). With chemical formula of $C_6H_5NH_2$, aniline (also called amino benzene) is a simple aromatic ring compound composed of a benzene ring bound to an amine group (2, 3).

Aniline is fat and colorless in nature and turns to reddish-brown color once oxidized. It is toxic and easily flammable and produces smoky flame during burning (3). Aniline has over 150 derivative compounds (4) and is used as raw material in petrochemical, plastic, paint, pharmacy and agricultural pesticide production industries and is a byproduct of paper and textile industries (2, 5).

Aniline is water soluble at concentrations up to 35g/L (6, 7) and this high solubility has led to increased pollution probability of industrial wastewater and water resources (1, 2). Aniline is a toxic chemical compound which can cause various problems to human and environment (6). Presence of aniline in aqueous environments can disturb aqueous species life due to its mutagenic and carcinogenic properties and because it facilitates the conversion of hemoglobin to methemoglobin causing methemoglobinemia (cyanosis) (6, 7).

In cases of acute contact, aniline leads to convulsion, coma and death and causes eye and skin complications when directly contacted (8) and can even cause nervous, kidney, liver and bone marrow disorders if the contact is frequent and prolonged (6, 9).

In addition, aniline increases the incidence of cancer tumors in animals and increases the risk of bladder cancer in human (10). Aniline is easily adsorbed to sediments and is easily spread and stabilized in the environment (11).

The maximum allowable concentration of aniline has been determined to be 2ppm for 8 hours of direct skin contact and 5ppm for 8 hours of exposure to polluted air (12). The US environmental protection agency (USEPA) has included aniline in the class of persistent organic pollutants (13). The USEPA has determined the maximum allowable concentration of 5 μg/L aniline in water (14).

Regarding its health and environmental hazards and chemical properties, the prompt and advanced elimination of aniline from the environment is necessary (2, 6). Common methods of aniline filtration are catalytic oxidation, biodegradation, electrolysis, advanced oxidation processes, membranous processes and adsorption (15, 16).

Each method has its own advantages and disadvantages. For instance, the application of biodegradation and electrolysis is limited by their extremely high cost (2). Furthermore, biologic processes are not applicable in wastewater with higher concentrations of aniline due to incomplete degradation and toxicity to other biologic species and also because these processes degrade other chemical substances (17, 18).

Among all methods, bio-adsorption that is useful for the isolation of wide range of organic material has become more common in recent years. In this adsorption process, activated carbon, coal, coconut shell, wood, bamboo shoots are used as the adsorbent (6). Industrial activated carbon is, among others, one of the most effective adsorbents, but it has limited application due to the high cost of initial production and reactivation (19, 20).

Activated sludge has been recently gained much attention as a highly effective bio-adsorbent for the isolation and recovery of pollutants and metal ions and to reduce toxicity in several studies due to its advantages such as lack of toxicity, the potential for recovery of metals, low cost, availability, reusability, application in wide environmental conditions, lack of production of chemical sludge and high efficiency (21, 22). Therefore, the present study was carried out with the aim of evaluating activated sludge bio-adsorbent for the elimination of aniline from aqueous solutions.

Materials and Methods

In this experimental-practical study, the effectiveness of activated raw sludge bio-adsorbent in eliminating aniline from samples of artificial wastewater was evaluated in laboratory scale. All chemicals including aniline were obtained from Merck (Germany) and the spectrophotometer (model PD-303 UV) at 300nm wavelength was used to determine aniline residual concentration (23). The efficiency of the adsorbent was assessed by follow up experiments with given concentrations of aniline (50, 10, 100, 150, 200 and 250 mg/l), pH conditions (3, 5, 7, 9 and 11), adsorbent doses (0.2, 0.4, 0.6, 0.8, 1.0 and 1.5 mg/l) and contact time (15, 30, 60, 90 and 120 minutes). 250 cc Erlenemayer flask shaker set at 150 rpm containing 100 ml of sample was used. pH of the solutions was adjusted using chlorhydric acid or hydroxide 1%. Samples were removed at different contact times, filtered through 0.45 wattman filter papers and finally were analyzed aniline concentration was determined spectrophotometrically at λmax= 300 nm according to the Lambert–Beer law using an UV–VIS spectrophotometer (T80 PG Instruments Ltd.) using standard calibration curve (equation 1).

Subsequently, the adsorbed aniline qe (mg/g) was determined by the following equation1 and 2:

$$q_e = \frac{(C_0 - C_e)V}{M}$$

$$RE(\%) = \frac{(C_0 - C_e)}{CO} * 100$$

Where Ce and Co are the initial and equilibrium concentration of aniline, respectively, in aqueous phase (mg/l), V is the volume of solution (L) and M is the amount of adsorbent used (g). The aniline elimination percentage was calculated as following (equation 2):where C_F and C_O are the initial and final (pre and post-absorption) concentrations of aniline, respectively (23, 24). All experiments were performed in duplicate to insure the reproducibility of the results; the mean of the two measurements is reported. The linear regression statistical test was used to determine the equations of adsorption isotherm.

Adsorption Isotherm

In the current study, Langmuir, Freundlich and Temkin isotherm models were used for the analysis of experimental data and description of the absorption equilibrium between solid and aqueous phases, and fitness into each model was determined by drawing equilibrium curve for each model and comparing the correlation coefficient (R^2) of each curve with experimental data (25).

Temkin isotherm: In the Temkin model, taking into account the probable adsorbate-adsorbate and adsorbate species-adsorbent interactions, adsorption theory is corrected. The adsorption heat of all molecules in adsorption layer decreases linearly with the degree of coverage (26). Equation (3):

$$q_e = \mathrm{B} \ln \mathrm{K_T} + B \ln \mathrm{C_e}$$

K_T is constant and B1 has been calculated by plotting qe against ln. K_T is the constant stunning equilibrium (mg/L) associated with the maximum energy of binding and B is related to adsorption heat.

Langmuir isotherm: Langmuir isotherm model is based on scientific assumptions. The most important assumption is that adsorbates (atom, molecules or ion) bind to specific sites on the adsorbent surface and single layer adsorption process occurs and also that all adsorption sites have identical affinity to adsorbate molecules and no transfer of adsorbate occurs at adsorbent surface Equation (4), (25, 27).

$$q_e = \frac{q_m K_l C_e}{1 + K_l C_e}$$

In this equation, qe represents the amount of adsorbed aniline per unit of adsorbent used Ce (mg/g), Ce is the equilibrium concentration of solution (mg/L) and qm is the amount of aniline required for the formation of double layer (mg/g). At the same time, Langmuir equation was drawn linearly and Langmuir adsorption constant (K_L) and the adsorbent maximum adsorption capacity (q_m) were linearized as following. The qm and K_L values are obtained by plotting 1/q against 1/Ce. Equation (5):

$$\frac{1}{q_e} = \frac{1}{q_m} + \frac{1}{q_m K_l} \frac{1}{C_e}$$

Freundlich isotherm is only an empirical model described based on heterogeneous adsorption on adsorbent surface by the following equation (6):

$$q_e = K_f C_e^{\frac{1}{n}}$$

where Kf and 1/n are adsorption constants for capacity and intensity of adsorption, respectively. By plotting ln q_e against ln C_e, based on experimental data, K_f and 1/n were obtained from intercept and slope of the regression line, respectively. The Freundlich equation for determination of Freundlich adsorption constants can be linearized as following (27, 28):

$$\ln q_e = \ln K_f + \frac{1}{n} \ln C_e (Eq7)$$

Results and Discussion

As the first step of the present study, the effect of pH (3, 5, 7, 9 and 11) on aniline adsorption capacity of sludge, with the aniline initial concentration of 100 mg/l and adsorbent dose of 0.6 mg/L for 30 minutes at room temperature (22±2 C) was investigated. The results are presented in figure 1. There was an increasing trend in aniline adsorption capacity of sludge with increasing pH from 3 to 11, with the maximum efficiency of 80% (133.3 mg/g aniline adsorbed) at pH=11. Therefore, pH of 11 was selected for remainder of the experiment.

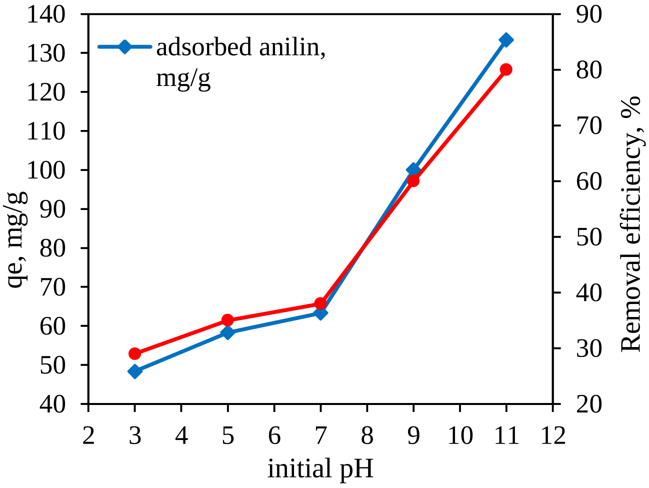


Fig 1:The effect of initial pH on aniline adsorption capacity of activated raw sludge adsorbent (initial aniline concentration: 100 mg/g; adsorbent dose: 0.6 mg/l; contact time: 30 minutes and temperature: 25 °C)

The solution affects ionization of aniline and thus its reaction with adsorbent surface during adsorption process. Aniline is a week base which can convert to positive charged anilinium ion.

Aniline is present as neutral aniline molecule at pH values below its pKa and at pH values above pKa it is converted to positive charged ion (29). In agreement with our results, abdossalam et al reported the maximum elimination of aniline by carbon/ferrite nanotube nanocomposite at pH of 10 (30).

In order to determine the effect of adsorbent dose on the adsorption process, different levels (0.2-1.5 mg/L) of adsorbent was used at a constant pH of 11, aniline initial concentration of 100 mg/L, 30 minutes of contact time at room temperature. The results are summarized at figure 2. Aniline elimination capacity decreased rapidly with increasing adsorbent dose from 0.2 to 0.8 mg/L and the decline was continued with gentler slope with higher doses until 1.5 mg/L. On the other hand, the amount of adsorbed aniline decreased with increasing adsorbent dose. The maximum efficiency of aniline elimination on the sludge was 80% at 0.2 mg/L adsorbent dose that corresponded to 400 mg/g of total aniline adsorbed above which there observed a dramatic decline in elimination efficiency.

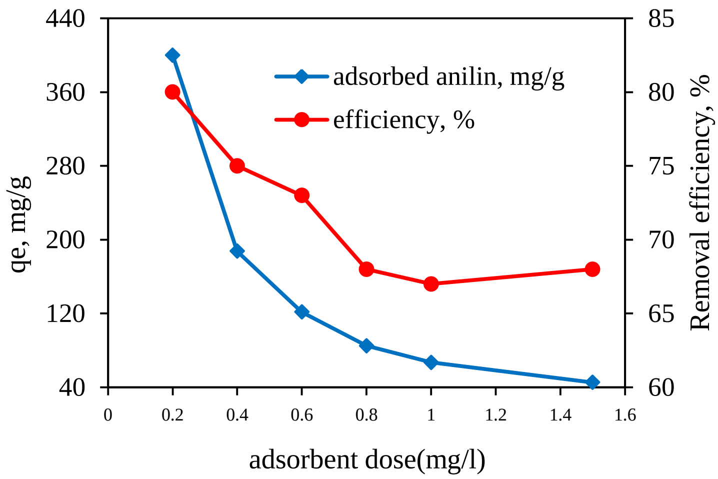


Fig 2: The effect of adsorbent dose on the elimination of aniline using activated raw sludge adsorbent (aniline initial concentration: 100 mg/L; contact time: 30 minutes; temperature: 25 C and optimum pH:11)

The decline in the efficiency and capacity of aniline elimination at higher adsorbent doses might be attributed to increased adsorption sites and availability of aniline which decreases concentration and the number of aniline molecules and thus creating equilibrium between adsorbent and pollutant. This may have increased the probability of pollutant readsorption and accumulation of adsorbent particles (30).

The effect of aniline initial concentration is presented in figure 3. According to the figure, at contact time of 30 minutes, pH of 11 and adsorbent dose of 0.2 mg/L, the efficiency of aniline elimination declines with increasing aniline initial concentrations and the amount of aniline adsorbed increases with increasing aniline initial concentration. When the initial concentration of aniline increases from 10 to 250 mg/ml, the amount of adsorbed aniline rises from 15 to 875 mg/g while the elimination efficiency decreases from 99.5 to 97.45%.

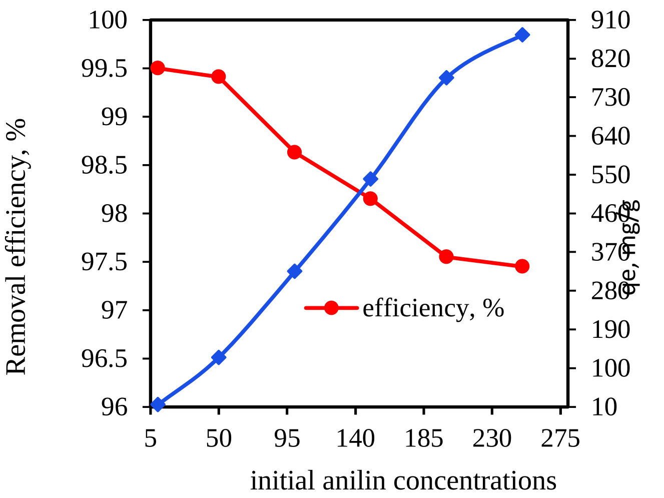


Fig 3: The effect of aniline initial concentration on aniline elimination by activated raw sludge adsorbent (adsorbent dose: 0.6 mg/L; contact time: 30 minutes; temperature: 25 C and optimum pH: 11)

The effect of aniline concentration indicates that the adsorbent possesses limited adsorption sites and in lower concentrations of aniline more adsorbent sites are available. This is responsible for the rapid increase in adsorption of aniline and the decrease in the efficiency of elimination with saturation of adsorption sites (32, 33).

Due to high probability of contact between

adsorbent and adsorbate, the adsorption capacity increases with higher initial concentrations (23). Other effective factor increasing the adsorption capacity in higher concentrations is the increased mass transfer force that overcomes the force opposing adsorption and provides considerable thrust force in favor of pollutant transfer from aqueous phase to the common border of adsorbent and liquid (34).

Results of the present study confirm those of Alipour (2014) studying the elimination of natural organic mattrials from aqueous solutions by oyster shell supported zero valent nano scale iron and those of Li who studied the application of cotton stalk for nitro-aniline elimination (30, 35). Contact time between adsorbent and aniline in another parameter evaluated in this study. According to figure 4, the greatest elimination efficiency was 80.52% (805 mg/g) which was achieved at 30 minutes of contact time.

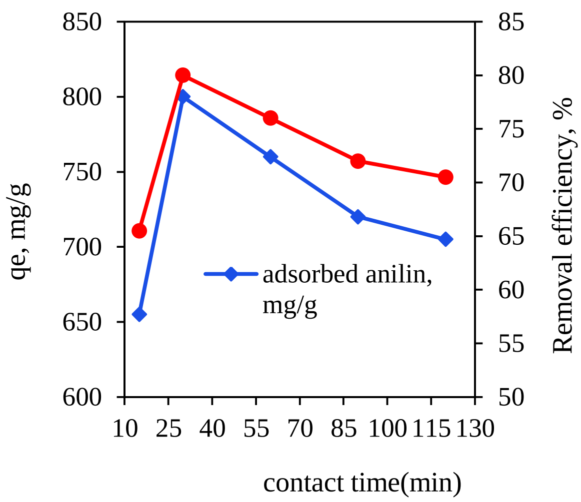
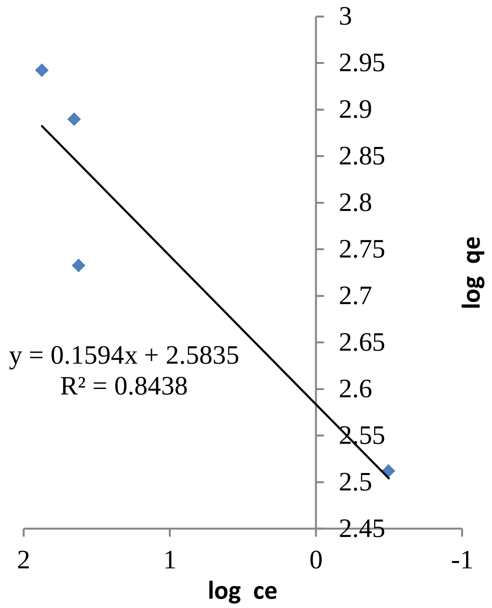


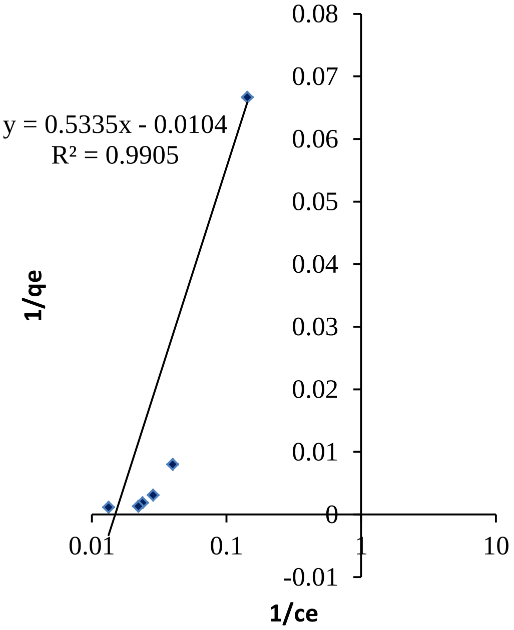
Fig 4. The effect of contact time on aniline elimination by activated raw sludge (Optimum aniline concentration: 200 mg/L; optimum adsorbent dose: 0.2 mg/L; temperature: 25 °C and pH: 11)

With increased contact time, there is an initial increase in elimination of aniline due to higher number of adsorption sites and higher adsorbate concentration gradient from aqueous phase to adsorbent surface. However, with further increases in contact time, the efficiency starts to decline with gentle slope due to occupation of adsorbent surface and decreased availability of adsorption sites (23).

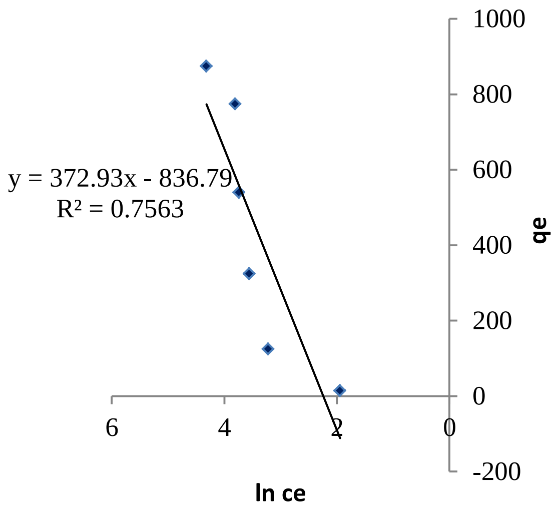
Adsorption isotherms are adsorption properties and equilibrium data which describe how pollutants interact with adsorbents and play a fundamental role in optimization of adsorbent indicator. Creating a proper relationship for equilibrium curve and optimal designation of an adsorption system for aniline is very important. The distribution of aniline molecule between liquid phase and adsorbent is an indicator of the position of the equilibrium in adsorption process and usually is expressed by one or more isotherm models (48, 50).



a) Freundlich adsorption isotherm



b) Langmuir adsorption isotherm



c) Temkin adsorption isotherm

Charts a, b and c are the result of regression coefficient of Langmuir, Freundlich and Temkin for both adsorbents and show that they fit Langmuir isotherm and indicate that aniline adsorption occurs at specific homogenous sites as a single layer process (36) that agrees with the study of Rahmani (2015) who investigated the aniline elimination by commercial activated carbon (24).

Conclusions

Results of the present study showed that bio-adsorption by activated raw sludge can effectively eliminate pollutant aniline from aqueous solutions.

The percentage of aniline elimination depends on its initial concentration, contact time between aniline and adsorbent, pH and the adsorbent. The greatest efficiency of activated raw sludge in eliminating aniline was 80% and the optimum condition for this efficiency was achieved at pH of 11, 30 min contact time, 0.2 mg/L adsorbent dose and aniline initial concentration of 200 mg/L.

Aniline adsorption to activated raw sludge fitted most the Langmuir adsorption isotherm. Therefore, activated raw sludge is highly potent in eliminating aniline from aqueous environments and can be considered as a practical strategy to eliminate these kinds of pollutants from industrial wastewater.

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