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Saif S. ALQUZWEENI, Afrah A. HASSAN, Rasha S. ALKIZWINI
University of Babylon, College of Engineering

A novel application of building demolition waste for removal benzene from aqueous solutions

Key words: building demolition waste, benzene, equilibrium, FTIR analysis

Introduction

The environment of the planet of Earth is deteriorating severely nowadays because of the increase in human activities, including industries and agriculture, which results in the production of enormous gaseous and liquid pollutants (Grmasha, Al-Sareji, Salman, Hashim & Jasim, 2020; Zubaidi et al., 2020). For example, the cement industry pollutes the air with a wide range of gases and particulates (Kadhim, Sadique, Al--Mufti & Hashim, 2020) that led to global warming (Salah, Abdulkareem et al., 2020; Zubaidi, Al-Bugharbee, Muhsin, Hashim & Alkhaddar, 2020) and climate change (Salah, Ortega-Martorell et al., 2020). However, water sources are severely polluted due to the recent growth in industrial activities, where several contaminants, including petroleum hydrocarbons, have polluted many water bodies from various industries, such as textile, petroleum, and fertilizers industries (Hashim, Al-Saati, Hussein & Al--Saati, 2018; Omran et al., 2019). Benzene is the commonest compound used for fractions of petroleum in a number of applications, such as raw materials in a number of industries and solvents in a number of industrial processes (Bakather, 2020); nevertheless, benzene is a toxic groundwater pollutant that has adverse impacts on the environment and hence endangers on the health of human. Based on the reports of the U.S. Environmental Protection Agency (EPA), benzene does not exceed 5 □g·L^{-1}. Therefore, safe, effective methods are needed to remove benzene from water. Benzene and organic matter were removed from water by a number of methods as wet air oxidation, photocatalytic degradation, and adsorption (Elsayed et al., 2017; Alquzweeni & Alkizwini, 2020). Adsorption is one of the areas of surface chemistry that relies on the aggregation of contaminants called adsorbates on the

surface of adsorbents (Bakather, 2020). The literature demonstrates that adsorption is the most powerful and widespread way to remove water contaminants, such as electro-chemical methods (Mohammed et al., 2020; Abdulhadi et al., 2021; Hashim, Shaw, AlKhaddar, Kot & Al-Shamma'a, 2021), coagulation and electrocoagulation (Aqeel et al., 2020; Emamjomeh, Kakavand et al., 2020; Emamjomeh, Mousazadeh et al., 2020; Hashim, Kot et al., 2020), and hybrid methods (Alnaimi et al., 2020; Al-Marri et al., 2020; Zanki et al., 2020). Several adsorbents were used adsorb benzene and components from water, including beer bran (Legrouri et al., 2018), raw and thermally modified lignite (Aivalioti, Pothoulaki, Papoulias & Gidarakas, 2012; Abdulla et al., 2020), and activated carbon (Perrich, 2018; Alyafei et al., 2020), and zeolite (Alenezi et al., 2020; Alhendal et al., 2020), natural adsorbents (Abdulraheem et al., 2020; Alenazi et al., 2020), and industrial by-products (Hashim, Ewadh et al., 2020). However, various waste materials (e.g. biomass and sewage of sludge) were effectively used to minimize the cost of processing nanoporous carbon and controlling the accumulation of atmospheric waste materials (Abdulraheem et al., 2020). Adsorption may, therefore, be used to remove aromatic compounds dissolved in water. With the current trend of using methods that minimize losses and are sufficiently efficient, research has been conducted to evaluate the use of waste or by-products, in particular those generated by industrial processes among the various solid wastes. The prospect of using construction by-products (from the demolition process) as adsorbents would be a significant point for the present study to assess how suitable it is for removing benzene from aqueous solutions.

Materials and methods

Adsorbent and contaminant

The building materials waste from bricks used in this study has been collected and grinded, and then in the oven, drying is done for 24 h at 80°C. The powder was placed in a container for further use. Distilled water is polluted with refined benzene (C_6H_6) at room temperature (purity > 99.5%, produced at a specific concentration by Sinopharm Chemical Reagent Co., Ltd.). This benzene's physical properties are 1,750 $mg \cdot L^{-1}$ water solubility, 0.88 specific gravity, and 35 dynes per 1 cm interfacial tension (Elsayed et al., 2017).

Characterization of building material waste

Figure 1 shows the results of X-ray diffraction, which indicates that O–H ranges from 3,700 to 3,000 cm^{-1}. The very strong and sharp band appearing at 3,644 cm^{-1} corresponds to calcium hydroxide stretching vibrations. The C–H stretching bands measure 3,300––2,800 cm^{-1}. In water solution, however, the C–H vibration frequency is much lower than in non-polar solution. Extending vibration frequencies between hydrogen and other heteroatoms are 2,600–2,000 cm^{-1}, including 2,250––2,100 cm^{-1} Si–H. Regions from 1,300 to 910 cm^{-1} also have skeleton C–O and C–C a contribution to vibration, providing additional molecular structural de-

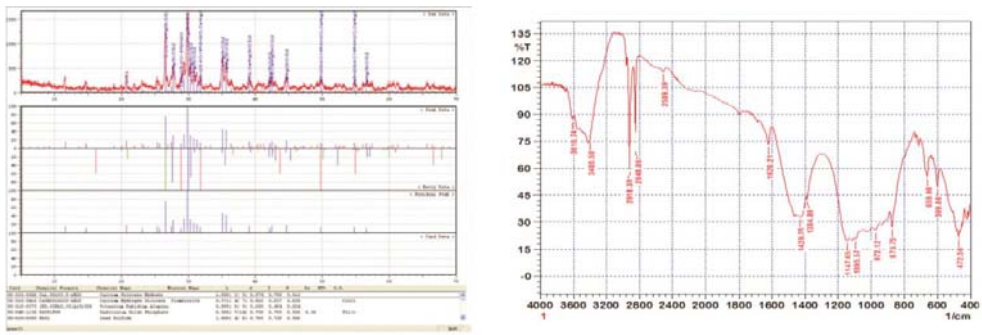

FIGURE 1. XRD and FTIR analysis for natural building material waste

tails relevant to higher frequency regions. Bands between 450 and 600 cm^{-1} are typical features of oxides not detected as major constituents. Due to the complex structure of building material waste, additional structural knowledge from this spectrum area is very difficult to obtain, as the bands characteristic of the various components overlap.

Batch experimental study

It is put in place to get the aforementioned single benzene equilibrium and kinetic data with prepared sorbents. This data will define the best-operating conditions such as initial pH, sorbent dose, and shaking speed for some initial concentration (C_o) that achieves the high performance of the process of treatment. Batch experiments are required for preparing a series of 250 mL flasks, and 100 mL of the contaminated water must be put in every flask with an initial 300 ppm concentration. Different doses of sorbent were applied and the flasks were kept agitated for 180 min at 250 rpm. Then each flask solution was filtered to isolate the water from the solid particles. High-performance liquid chromatography can be used to calculate the residual concentration (C_e) of benzene in the supernatant (HPLC, Shimadzu 2010 Japan). The principle of mass balance can be applied to locate the contaminant sorbed quantity per unit mass of the sorbent. Sorption analysis with pH levels of 3.0–7.0, contact time of 0–180 min and finally C_o at 300–700 $mg \cdot L^{-1}$ was performed. The amounts of pollutants sorbed onto the sorbent (q_e) have been calculated for the best conditions as follows:

$$q_e = (C_o - C_e)\frac{V}{m} \tag{1}$$

where:
V – volume of water in a flask [L],
m – sorbent mass in a flask [g].

The adsorption isotherm is plotted between calculated q_e and C_e.

Equilibrium sorption process models

The model of sorption is the relationship between the quantities of the chemicals sorbed on solids (q_e $mg \cdot g^{-1}$) and final concentrations of the compound (C_e $mg \cdot L^{-1}$). At a certain temperature and

pH values were determined; however, several isothermal models, including Freundlich and Langmuir, were fitted.

Freundlich model

The present model may cover multilayer sorption and heterogeneous surfaces but may, generally, be the following (Alquzweeni & Alkizwini, 2020):

$$q_e = K_F C_e^{1/n} \tag{2}$$

where:
K_F – maximum quantities of contaminant sorbed on reactive material,
$1/n$ (< 1) – indicated by the intensity of the sorption.

Langmuir model

Langmuir developed the following relationship for monolayer sorption and homogeneous surfaces (Alquzweeni & Alkizwini, 2020):

$$q_e = \frac{q_{\max} b C_e}{1 + b C_e} \tag{3}$$

where:
$q_{\max}$ – maximum capacity of adsorption [mg·g^{-1}],
b – intensity of contaminants to the solid phase.

Kinetic models

For the design of the appropriate sorption process, the rate of transfer is used from the watery to the solid phase. The following kinetic models predict this rate:

Pseudo-first order model

It has a formula to explain sorption rates as a function of time (Alquzweeni & Alkizwini, 2020):

$$\frac{\mathrm{d}q}{\mathrm{d}t} = k_1 (q_e - q_t) \tag{4}$$

By applying $q_t = q_e$ at $t = t$ and $q_t = 0$ at $t = 0$, Eq. (2) is integrated to produce the following model:

$$\ln(q_e - q_t) = \ln q_e - k_1 t \text{ or } q_t = q_e \left(1 - e^{-k_1 t}\right) \tag{5}$$

where:
q_t, q_e – amounts of contaminant sorbed on the solid matrix in time t [mg·g^{-1}],
t – time of equilibrium [min],
k_1 – rate constant [min^{-1}].

Pseudo-second order model

The same energy of sorption for uninteracted sorbents and sorbed chemical species, as well as the contaminant's monolayer attached to the sorbent surface, are the general assumptions to guide this model as in Eq. (4):

$$\frac{\mathrm{d}q}{\mathrm{d}t} = k_1 (q_e - q_t)^2 \tag{6}$$

where:
k_2 – rate constant [g·mg^{-1}·min^{-1}].

Eq. (5) is integrated under the same conditions as the previous model, and the equation will take the forms in Eq. (5):

$$\frac{1}{(q_e - q_t)} = \frac{1}{q_e} + k_2 t \text{ or } q_t = \frac{\mathrm{t}}{\left(\frac{1}{k_2 q_e^2} + \frac{t}{q_e}\right)} \tag{7}$$

Results and discussion

Effect of adsorbent dosage

The adsorbent dose effect on the adsorption of a fixed benzene concentration is shown in Figure 2a. As the solvent dose increased over the range of 0.4–2.4 g per 100 mL, the percentage of benzene sorbent increased reached 98%. The number of available adsorption sites increases by raising the adsorbent dose, which increases the removal efficiency (Alquzweeni & Alkizwini, 2020). Higher adsorption with the sorbent dose may also be due to increased surface area. These frequent findings with Langmuir's theories that the adsorbent particle is increasingly compatible with organic material with a growing amount of adsorbent particles per unit volume.

Effect of the time of contact

Determining the time of contact needed to achieve an equilibrium status takes into account the possible point of the batch study. Accordingly, as a function of the time of contact, the sorption of benzene on sorbents was controlled, and results were plotted in Figure 2b. Operational conditions include C_o, agitation rate of 300 ppm, and 250 rpm, respectively, adsorbent dose of 1.2 g added at room temperature to 100 mL of polluted water. Figure 2b shows that the removal will increase in the early times of the rapid rate significantly with the contact time, and this rate slows after approximately 150 min. The explanation for slower sorption may be the decline in vacant sites for adopting sorbent. Kinetic data shows that 1 h is sufficient for benzene removal. After this equilibrium period, there is no noticeable shift in the final concentration up to 180 min (Faisal & Naji, 2019).

Effect of initial pH

Effects of the initial pH on benzene's adsorption by the adsorbent were measured at a pH range of 3.0–7.0. As shown in Figure 2b, the pH influences significantly on adsorption of benzene by the adsorbent of 3–6. This means the adsorbent has absorbed benzene. It can be shown that the adsorbent's adsorption capacity decreased rapidly to pH 7.0. It can decrease the driving force of benzene, including the force of van der Waal to the adsorbent's active sites.

Effect of initial benzene concentration

The effect of the initial concentration on benzene's sorption efficiency within the range of 300–700 $mg \cdot L^{-1}$ has been investigated. Figures 2c showed that the removal efficiencies of benzene on a sorbent at (pH 7.0) decreased from (> 90%) to lower values (> 50%) as a result of an increase in the C_o within the range described. The primary explanation for this activity may be the presence of sufficient sites to absorb even more benzene from an aqueous solution. The benzene present in the solution is capable of interacting with lower concentrations of vacant binding sites and, therefore, has been high intake performance relative to higher rates. Thus, the treatment yield can be improved by diluting the water polluted with high benzene concentrations.

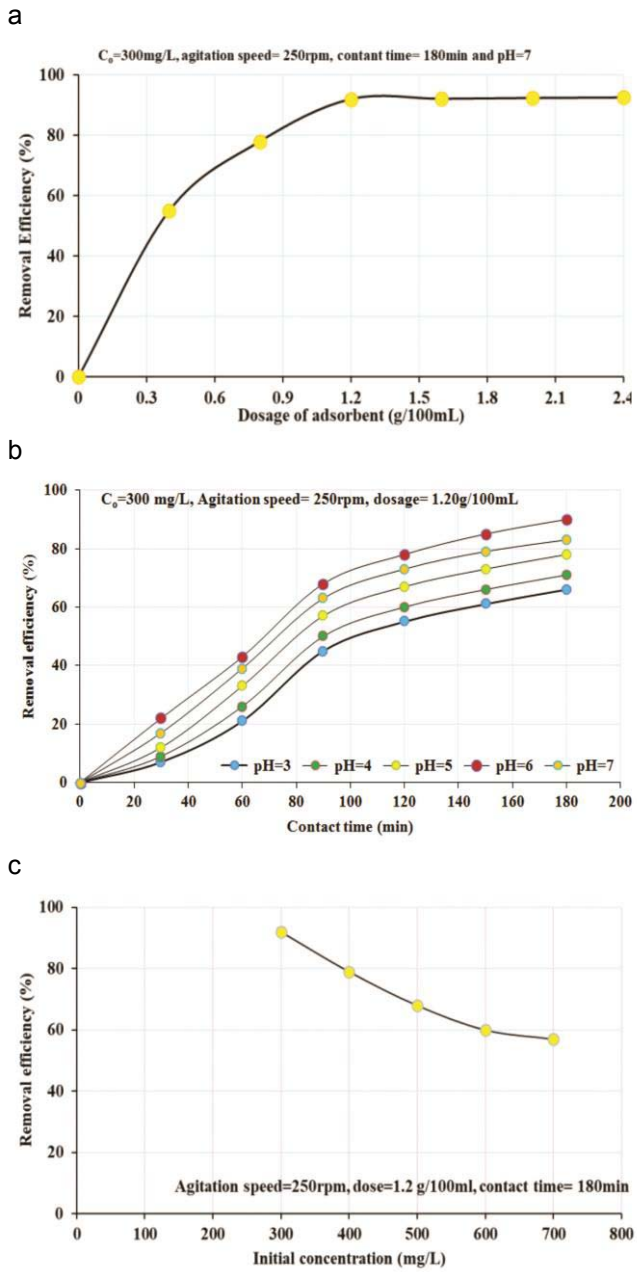

FIGURE 2. The relationship between benzene removal efficiency: a – dose of the adsorbent; b – time of contact, initial pH; c – initial concentration

Kinetic and isotherm analysis

Figure 3 shows the pseudo-first, second-order, and Weber–Morris models that are fitted with kinetic data. For this reason, Microsoft Excel 2016 used the nonlinear regression approach to find the models' constants. The benzene sorption is compatible with (pseudo-first order kinetic) model. This indicates that physical sorption is the primary Sorption mechanism under consideration (Alquzweeni & Alkizwini, 2020). Sorption mechanisms are difficult to characterize based on the previously described kinetic models. This function was thus performed based on (intra-particle diffusion) model concepts. As shown in Eq. (8), the model would be represented empirically where the sorbed quantity changing as a function of $t^{0.5}$ instead of t.

$$q_t = k_{\text{int}} t^{0.5} + C \tag{8}$$

where:
k_i – constant rate of stage i [mg·g^{-1}·h$^{-0.5}$], and is equal to the related slope of q_t to $t^{0.5}$.

The intercept for stage i is also expressed by the value of c_i and reflects the boundary layer's thickness. This implies the greater boundary layer effect can be recognized with the greater intercept value. The intra-particle diffusion occurs if the relationship is associate linearly between q_t and $t^{0.5}$. If the linear plot passes through the origin, the rate-limiting mechanism is defined as the only intra-particle diffusion. Otherwise, other processes of intra-particle diffusion must be considered. The adsorption mechanism is usually considered to include: (i) the mass transfer of adsorbents from bulk to the particulate surface, (ii) surface adsorption, finally (iii) intra-particle diffusion of the adsorbed molecules to an adsorption site through a pore diffusion and/or surface diffusion mechanism. Step (ii) is always presumed to be extremely fast; hence the adsorption of large molecules with long periods of contact to equilibrium is often considered to be regulated by diffusion through external film resistance and/or internal mass transfer of diffusion or intra-particle diffusion. A classical method for evaluating whether intra-particle diffusion regulates an adsorption mechanism is to plot the amount adsorbed versus the square root of time ($t^{0.5}$); when the plot is linear and passes through the origin, it implies that the adsorption rate is regulated by intra-particle diffusion. So, Figure 3 shows the amount of benzene adsorbed for the adsorbents as a function of $t^{0.5}$. It can be shown that the experimental data are satisfactorily fitted by the intra-particle diffusion model, obtaining a linear section that does not pass through the origin, suggesting that benzene adsorption on these adsorbents is not controlled by intra-particle diffusion. Sorption tests are carried out for various concentrations under conditions of pH 7.0, dose = = 1.2 g per 100 mL, speed of 250 rpm, and contact time of 3 h. Freundlich and Langmuir isotherms are shown graphically in Figure 3, and the table lists their constants, which were determined using non-linear fitting of these models with sorption measurements using the Microsoft Excel 2016 Solver option (Alquzweeni & Alkizwini, 2020). Based on Figure 3 and determination coefficient (R^2) in the table, in the definition of sorption measurements, Freundlich model is better than Langmuir one.

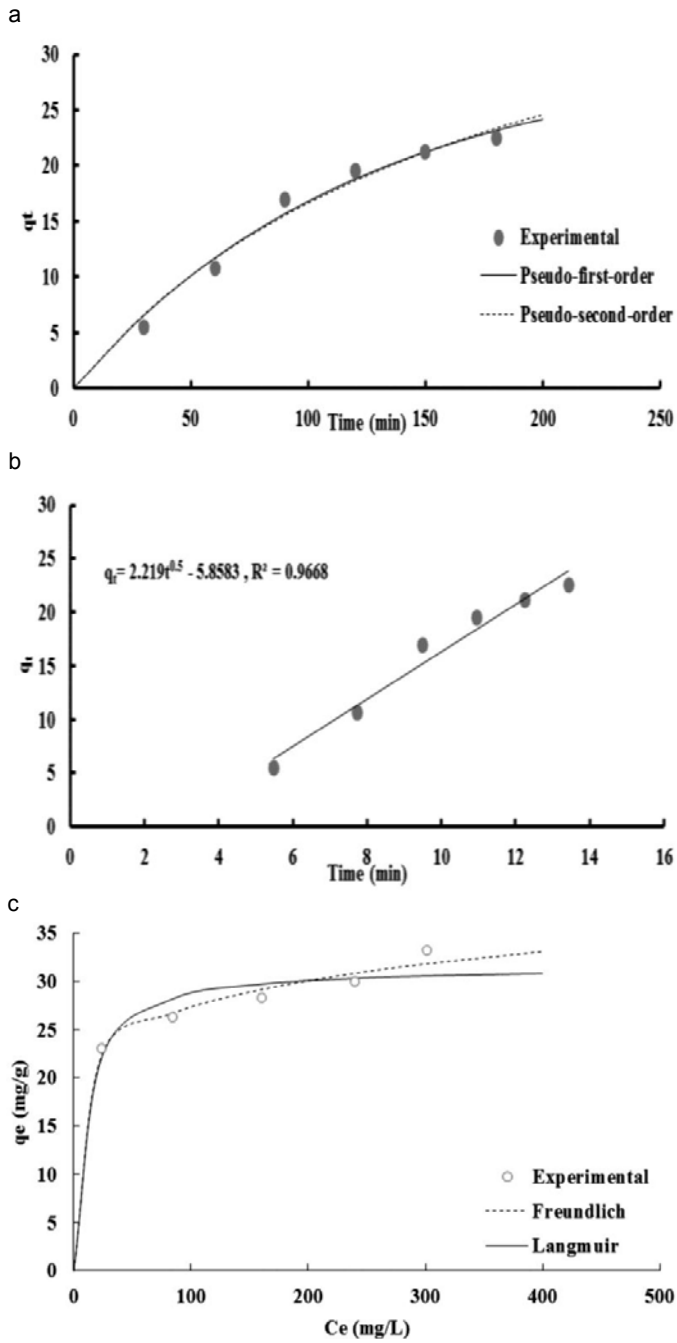

FIGURE 3. Experimental data on sorption fitted with (a) pseudo-first, pseudo-second order models, (b) intra-particle diffusion model, and (c) the isotherm models for the benzene's sorption

TABLE. Isotherm and kinetic modals' constant for benzene's sorption

Model		Parameter	Value
Isotherm	Freundlich	K_F [mg·mg^{-1}][L·mg$^{-1/n}$]	14.458
		N	7.237
		R^2	0.935
		SSE	13.277
	Langmuir	q_{max} [mg·g^{-1}]	31.602
		b [L·mg^{-1}]	0.097
		R^2	0.777
		SSE	3.868
Kinetic	pseudo-first order	q_e [mg·g^{-1}]	29.938
		k_1 [1·min^{-1}]	0.0083
		R^2	0.982
		SSE	4.632
	pseudo-second order	q_e [mg·g^{-1}]	47.04
		k_2 [g·mg^{-1}·min^{-1}]	0.0001
		R^2	0.977
		SSE	5.676
Intra-particle diffusion		k_{int} [mg·g^{-1}·min$^{-0.5}$]	2.219
		R^2	0.967

Conclusion

This has been researched the possible use of building material waste as an adsorbent to benzene. Building material was found to be more effective for removing benzene from the solution; the maximum benzene's adsorption efficiency was 31.601 mg·g^{-1}. The percentage of removal depended strongly on the contact time, the solution's initial pH, and adsorbent dose. Initial pH, contact time, and sorbent dose must be 6, 1 h and 1.2 mg per 100 mL for C_o of 300 mg·L^{-1} and a speed of agitation of 200 rpm, respectively, are the best conditions for achieving these effects. Sorption results are well illustrated by the Freundlich model for the interaction of benzene–water contaminated with sorbent. Finally, the kinetic process is consistent with the pseudo-second order equation.

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Summary

A novel application of building demolition waste for removal benzene from aqueous solutions. A novel application of building demolition waste for removal benzene from aqueous solutions. In this research, demolition waste from buildings has been studied for possible use as benzene removal adsorbent from aquatic solution. The effect of adsorbent dosage, contact time, initial benzene concentration, and initial pH on benzene adsorption capacity have been investigated in the batch adsorption experiments. The adsorption effects initially happened very rapidly and achieved equilibrium within 180 min. Benzene removal was observed to decrease by an increase in the initial concentration of benzene of 300–700 $mg \cdot L^{-1}$, an increase in the adsorbent dose of 0.4–2.4 g per 100 mL, where an optimum adsorbent dose equal to 1.2 g per 100 mL was found. The potential of adsorption increases with pH 3.0–7.0 to reach the maximum removal efficiency at pH 6.0. The findings showed that equilibrium data were adequately adapted and correlated with the Freundlich isotherm models. The average percentage of the removal at room temperature was about 98%. Results suggest that building demolition waste can be used effectively in industrial wastewater treatment for the removal of aromatic hydrocarbon, benzene, as a low--cost option.

Authors' address:
Saif S. Alquzweeni – corresponding author
University of Babylon
College of Engineering
Department of Civil Engineering
40-street, 009647812260838, 51001, Babil
Iraq
e-mail: saifalquzweeni@gmail.com

Afrah A. Hassan
Rasha S. Alkizwini
University of Babylon
College of Engineering
Department of Environmental Engineering
80-street, 009647812835911, 51001, Babil
Iraq
e-mail: eng.afrah.abood@uobabylon.edu.iq
eng.rasha.salah2018@gmail.com