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Phenol removal from aqueous solution by adsorption process: Study of the nanoparticles performance prepared from aloe vera and mesquite (prosopis) leaves

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Adsorption kinetics.

Abstract. This study was performed to measure the potential utilization of agro-waste to generate nanoparticles and evaluate its capability as a low-cost adsorbent for phenol removal. Adsorption studies for phenol removal using aloe vera and mesquite leaves nanoparticles were carried out under various experimental conditions including pH, nano-bioadsorbent dosage, phenol concentration, contact time, temperature, and ionic strength in a batch reactor. The adsorption kinetics were applied by pseudo-first order and pseudo-second order models and isotherm technique by Freundlich and Langmuir isotherms models. The results showed that the rate of phenol adsorption increases in both nano-bioadsorbents with an increase in pH up to 7, adsorbent dosage up to 0.08 gL^{-1}, phenol initial concentration up to 32 mgL^{-1}, contact time up to 60 min, and a raise in temperature. The adsorption data followed the Freundlich isotherm model. The kinetic studies indicated that the adsorption of phenol with nano-bioadsorbents was best described by the pseudo-second order kinetics. We found that the nanoparticles prepared from aloe vera and mesquite leaves had a high capability in adsorption of phenol, besides the point that they could be accessed at low cost. These agro-wastes can be used to remove phenol from aqueous environments.

1. Introduction

The presence of persistent toxic substances within environment is a major environmental problem. One of the classified pollutant compounds in environment is phenol due to its specific properties, such as toxicity, carcinogenicity, taste, and odor problems, in drinking water, causing adverse effects on human health and living organisms, according to reports of the Environmental Protection Agency of America (US EPA) [1,2]. Phenol or hydroxyl benzene (C_6H_6O) is one of the aromatic hydrocarbons and derivatives of benzene that has high solubility in water. Phenol solubility in water is 93-98 gL^{-1} (depending on the temperature between 20-25°C) [3,4]. Phenol and its derivatives are

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aromatic compounds widely used in many industries such as petrochemical, paints, textiles, resins, plastics, and so forth. Hence, they are widely released into the environment as air pollutants and cause major problems even in low concentrations [5-7]. The threshold concentration of phenol in drinking water is recommended (to be) less than 1.0 μgL^{-1} by both World Health Organization (WHO) and U.S.EPA regulations; they constantly request for lowering phenol content in wastewaters to less than 1 mgL^{-1} [8,9]. Eventually, due to their harmful effects, wastewaters containing phenol and other toxic compounds must be treated before being discharged into receiving water bodies. Several methods have been investigated for the removal and degradation of phenolic compounds from aqueous solutions. These include physicochemical treatment processes, chemical oxidation, biological degradation, and combined techniques [10-13]. Many of these processes have disadvantages such as the generation of dangerous by-products, low efficiency and applicability to limit concentrations of pollutants [9]. Among them, adsorption technique is one of the effective methods, quite popular in removing various pollutants due to its simplicity, high efficiency, easy handling, high selectivity, minimization of the production of chemical or biological sludge as well as the availability of a wide range of adsorbents [11,12]. A number of adsorbents have been used for phenol removal; for example, activated carbon, red mud, rubber seed coat, fly ash, organobentonite, surfactant-modified natural zeolite, and organomodified tirebolu bentonite. It is considered that adsorption of phenol by activated carbons is a well-known process, because it has a large surface area and results in high adsorption capacity [8,12]. However, high cost of production, reclamation, purification, and irreversible nature of adsorption reduce its usage as an adsorbent, particularly in developing and low-income countries. Therefore, these issues have led many scientists to work on cheap and locally available absorbents for substituting activated carbon to remove different biodegradability pollutants and organic chemicals, such as phenol. Hence, in recent years, special attention has been drawn to the use of industrial and agricultural wastes to remove toxic and dangerous pollutants from water and wastewater as potential adsorbents [14,15]. This is likely to be the first attempt to synthesize nanoparticles from aloe vera and mesquite leaves as new, available and low-cost materials for the removal of phenol. Unique properties of bioadsorbents provide new opportunities for the removal of pollutants with high efficiency and cost-effective method. On the other hand, it is shown that nanoparticles efficiently have a promising adsorption related to the removal of various pollutants because of the high surface area, more active sites, and special properties [11,15]. Aloe vera or aloe barbadensis is an herbaceous plant with thick and fleshy leaves, mainly growing in tropical and subtropical dry zones. aloe vera belongs to the Liliaceae family and aloiede group. This plant contains anthraquinones derivatives such as aloin, barbaloin, isobarbaloin, and anthranol. Other important components of aloe vera consist of sugars (such as glucose, mannose, and cellulose) and enzymes (including oxidase, amylase and catalase, and folic acid), and minerals (such as calcium, sodium, magnesium, and zinc). This plant is mostly used in pharmaceuticals and cosmetic industries [16]. Prosopis plant is from Mimosaceae families which are common in many arid and semi-arid climates as trees and shrubs. It can grow in different countries including India, Pakistan, Nigeria, Ghana, Brazil, Peru, and South coast of Iran (Sistan and Baluchestan, Hormozgan, Bushehr, Khuzestan, and south of Fars). This plant is relatively considered a good source of crude protein, crude fat, crude fiber, ash, calcium, and phosphorus. Fuel and fencing, sand dune stabilization, stabilization of nitrogen in the soil, and forage source for livestock feeding can be cited as its applications. Prosopis genus covers 44 different species including four species of prosopis julifilora, prosopis farcta, prosopis cineraria, and prosopis koelziana found in Iran [17,18]. Prosopis cineraria leaves (because of its abundance in Sistan & Baluchestan province) were used in this study as a nano-bioadsorbent. The objectives of this study are: (i) To determine the feasibility of developing a new and inexpensive alternative of nano-bioadsorbents based on agricultural wastes for the removal of phenols from aqueous solutions; (ii) To investigate the main effects of experimental parameters on the removal of phenols, such as pH, nano-bioadsorbent dosage, initial concentration, contact time, temperature, and ionic strength; and (iii) To study the adsorption capacity of the nano-bioadsorbents using the adsorption isotherm technique and kinetics of adsorption of phenol on the nano-bioadsorbents using different models.

2. Materials and methods

2.1. Supply and preparation of adsorbent

mesquite and aloe vera leaves were used as natural adsorbents. In fact, known as agricultural waste, it was prepared in Iranshahr city located in Sistan-Baluchistan Province. The preparation process of the desired nano-bioadsorbents was as follows. After collecting wastes, they were washed with distilled water several times to remove impurities. Then, leaves dried for 3 hours at 105°C in the oven. They were burned in the oven to be converted into ash at a temperature of 700°C for 1 h. Then, it was milled by the Los Angeles abrasion to prepare the adsorbents in nanometer size and shed them to the mill (NARYA MPM-2* 250 model). Stearic acid equaling 3% of adsorbent weight was added to convert adsorbent particles to nanoscale. Steel balls were used in this study in a ratio of 1 to 7 (adsorbent weight to balls).

2.2. Batch adsorption experiments

The chemicals and reagents were applied to analytical grade and used without extra purification with an exception of deionized water, which was used as the solvent. All experiments were done by the use of double-distilled water. A stock phenol solution of 1000 mgL^{-1} was prepared by dissolving 1.0 g of phenol obtained from Merck (Darmstadt, Germany) in 1 L of deionized water. The required concentration of phenol solutions was prepared by diluting the appropriate volumes of the stock solution. The pH of the solutions was adjusted by the addition of 0.1 M HCl or 0.1 M NaOH solutions. A digital pH meter (DHP-500, SICO, UK) was used to measure pH values. Phenol-spiked synthetic water samples were used to determine and measure the effective parameters as well as the adsorption capacity and percentage of phenol removed from aqueous solutions using nanoparticles from aloe vera leaf extract and mesquite bioadsorbents. Effective parameters include pH, nano-bioadsorbent dosage, initial concentrations of phenol, contact time, reaction temperature, and ionic strength; their amounts are cited in Table 1. To examine the influence of ionic strength on phenol removal, $NaNO_3$, NaCl, Na_2SO_4, and $NaHCO_3$ salts and various concentrations were individually added to water solutions. Batch adsorption experiments were carried out in 100 ml sample flasks. To start each adsorption test of phenol, synthetic produced water samples with predetermined conditions were loaded into a 100 ml polythene vial (Table 1). The sample vials were shaken on a rotary shaker (GFL 137) with 120-rpm speed in order to appropriately mix the absorbent and adsorbate. The samples were then withdrawn at predetermined time intervals, and the mixture was filtered with the filter paper (Whatman No. 45 μm) with an aim of measuring the residual phenol concentration using a UV-vis spectrophotometer (Shimadzo-1700, Japan) at wavelength of 520 nm with Standard Methods [19]. The equilibrium adsorption capacity, q_e (mgg^{-1}), and the percentage removal were calculated using Eqs. (1) and (2), respectively:

$$q_e = \frac{V}{M} \times (C_0 - C_e), \tag{1}$$

$$E = \frac{C_0 - C_e}{C_0} \times 100, \tag{2}$$

where C_0 is the initial concentration of phenol in solution (mgL^{-1}), C_e is the equilibrium concentration of phenol in aqueous solution (mgL^{-1}), V is the volume of solution (L), M is the mass of adsorbent (g), q_e is the amount of absorbed phenol at equilibrium time (mgg^{-1}), and E is the removal efficiency.

The adsorption isotherms were described by Langmuir and Freundlich isotherm models for understanding the adsorption mechanism. Adsorption kinetics were also applied by the pseudo-first-order and pseudo-second-order models for understanding the adsorption rates of phenol on the surfaces of nano-bioadsorbents and determining the equilibrium times of adsorptions.

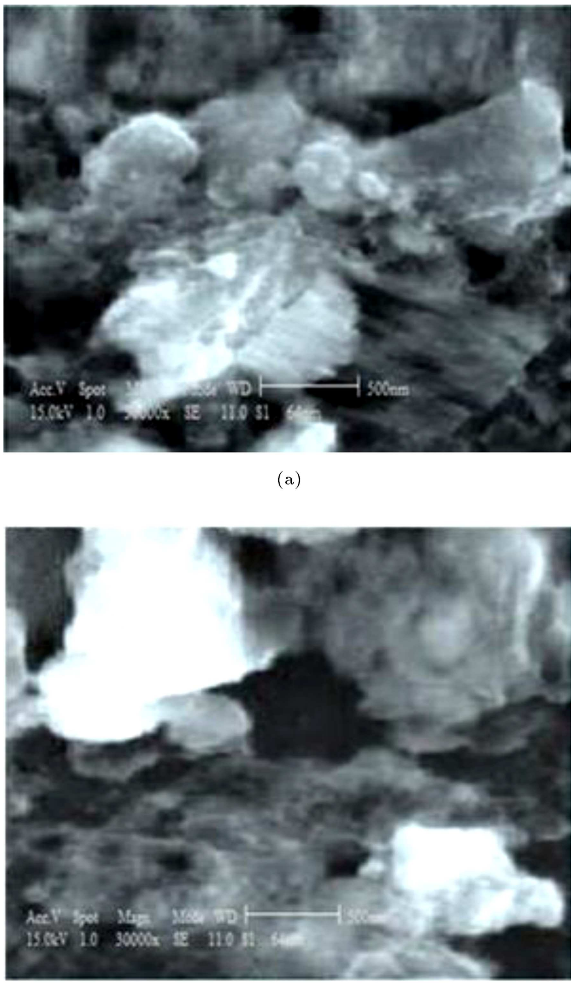
3. Result and discussion

3.1. Morphology and determination of specific surface area, particle distribution, methylene blue, and iodine numbers of nano-bioadsorbent

The morphologies of adsorbent as nanoparticles were obtained using a Field Emission Scanning Electron Microscope (FESEM, FEINova-Nano SEM-600, Netherlands), shown in Figure 1(a) and (b). Nano-bioadsorbents surface area was determined according to Brunauer-Emmett-Teller Method (BET) using Tristar 3000 Micromeritics device (Micromeritics Instrument Corp., USA) [20,21]. The results of BET method showed that average surface areas of aloe vera and mesquite nanoparticles were 41.25 and m^{2}g^{-1} and 38.76 m^{2}g^{-1}, respectively. Average particle size of the nano-bioadsorbent was 95 nm according to the Barret, Joyner, and Halenda method (BJH) through adsorption and desorption curves of nitrogen at 77 degrees Kelvin using Micromeritics Tristar 3000 device [20,22]. Two other important parameters used in indicating

Table 1. Evaluation of parameters and their range.

Number	Parameter	Value
1	pH	3, 5, 7, 9, 11
2	Adsorbent dose	0.01, 0.02, 0.04, 0.08, 0.1, 0.3 gL^{-1}
3	Initial concentrations of phenol	0.1, 0.5, 1, 2, 4, 8, 16, 32, 64 mgL^{-1}
4	Contact time	10, 20, 30, 40, 50, 60, 70, 90 min
5	Temperature	25, 35, 45, 55°C
6	$NaNO_3$, NaCl, Na_2SO_4, and $NaHCO_3$	0.01, 0.05, 0.1, 0.15, 0.2, 0.25 M



(a)

(b)

Figure 1. SEM micrograph of nanometric adsorption nanoparticles of (a) mesquite and (b) aloe vera leaves.

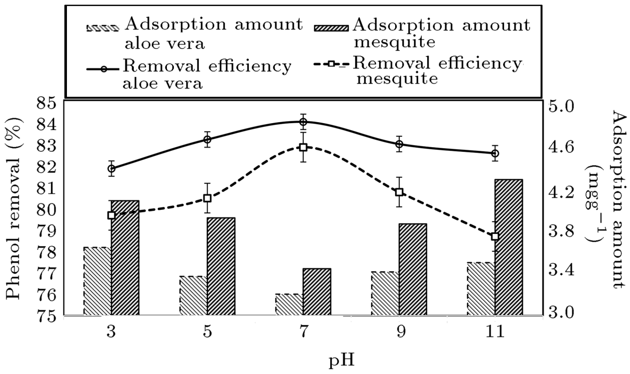


Figure 2. Effect of pH on phenol adsorption by mesquite and aloe vera nanoparticles. Condition: The initial concentration of phenol: 8 mgL^{-1}, nano-bioadsorbent dosage: 0.08 gL^{-1}, and contact time: 40 min.

adsorptive property are methylene blue and iodine numbers determined by standard procedures [23]. The aloe vera and mesquite nanoparticles have methylene blue numbers of 130 and 119, respectively. The aloe vera nanoparticle has iodine number of 375, whereas mesquite nanoparticle has 346.

3.2. Effect of pH

The pH of the solution is considered to be one of the most important parameters in adsorption processes, affecting the surface charge of the adsorbent as well as the degree of ionization and speciation of the adsorbate [24,25]. In order to evaluate the effect of pH on the adsorption of phenol on nano bioadsorbents, the adsorption experiments were carried out with 8 mgL^{-1} phenol concentration, 0.08 gL^{-1} nano-bioadsorbent dose, and 40 min contact timeat 35°C at different initial pH values in the range of 3-11 (Figure 2). Figure 2 shows the effects of pH on phenol adsorption in aloe vera and mesquite nanoparticles, where an increase in pH up to 7 would lead to adsorption efficiency of up to 84% and 82% for both nanoparticles, respectively. As the pH is increasing to 11, adsorption amount increases gradually, whereas adsorption efficiency tends to decrease. The minimum adsorption capacities of nanoparticles of aloe vera and mesquite at pH 7 were equal to 3.2 mgg^{-1} and 3.4 mgg^{-1}, respectively. The optimum amount of adsorption was determined at pH 7 for both nano-bioadsorbents due to the difference in the concentrations of H^+ and OH^- in the solutions.

At lower pH values, the adsorption of phenol is less likely to happen due to both the presence of H^+ ions and increased H^+ adsorption on the carbonyl sites, suppressing phenol adsorption [26]. At higher pH range, the ionization degree of phenol increases and phenol forms salts, readily ionizing and causing negative charge on the phenolic group. Simultaneously, the quantity of OH^- ions also increases. Thereby, the presence of OH^- ions on the adsorbent hampers and the diffusion/uptake of phenolic ions increase the electrostatic repulsion between the negatively charged surface sites of the adsorbent and phenolate ions. These results are in line with the findings of the previous studies. Banat et al. (2000), for example, reported that in the presence of bentonite, the proportion of phenol adsorption is decreased by increasing pH [27]. A similar study by Senel et al. showed that when using hollow fibers, the maximum removal rate of both phenol and chlorophenols pollutants from water was observed at pH 2, and no significant decrease was found in the adsorption capacity when pH increased to 6 [1]. Likewise, the study showed that when using low-cost adsorbents (coconut fiber activated carbon), the removal efficiency of phenolic compounds [2-chlorophenol (2-CP)] from solution decreased from 50% to 15%. The removal efficiency of phenolic compounds [2, 4, 6-trichlorophenol (TCP)] also decreased from 55% to 19%. Using acid treated coconut fiber activated carbon for the removal of 2-CP showed a decline from 58% to 23%; for the removal of TCP, a decrease from 78% to

30% was revealed due to changes in pH from 2 to 10 [28]. PH 7.0 was selected as an optimum pH value from the experimental results.

3.3. The effect of nano-bioadsorbent dosage

In any adsorption process, another important parameter is the amount of adsorbent that plays an important role. This parameter determines the capacity of adsorbent for a given phenol concentration and also determines sorbent-sorbate equilibrium of the system [29]. Thus, the study of a wide range of aloe vera and mesquite nano-bioadsorbents dosage at constant conditions showed that phenol removal from the solution depends on the dosage of adsorbent in the solution. The result of the study demonstrates that as the dosage of nanoparticles of aloe vera and mesquite increases, the amount of solute adsorbed increases and reaches a maximum value corresponding to a certain dosage (Figure 3). This can be observed in Figure 3. For nanoparticles of aloe vera and nanoparticles of mesquite, phenol removal reached 97.13% and 86.2%, respectively, when their dosage was 0.08 gL^{-1} and the initial phenol concentration was 8 mgL^{-1}. As nano-bioadsorbent dosage increases at a fixed phenol, the initial concentration provides greater availability for the exchangeable sites or surface area for phenol; hence, the removal has enhanced, whereas the adsorption amount has decreased due to the partial aggregation or overlapping of nanoparticles of aloe vera and mesquite, resulting in a less effective surface area for the adsorption. Beyond a certain dosage, the adsorption efficiency did not increase significantly. In other words, the system has reached equilibrium, indicating that the adsorption and desorption rate of phenol is equal [30]. In addition, nanoparticles have high reactivity due to its high specific surface area, and by increasing the amount of adsorbent solution (up to 0.08 gL^{-1}), the particles interact with each other instead of adsorbing pollutants and convert to hunk. In this case, the specific surface area reduced, and then the efficiency of adsorption of pollutants reduced by these particles [31]. Therefore, the optimum amounts of nanoparticles of aloe vera and mesquite for further adsorption experiments were selected 0.08 gL^{-1} for both nano-bioadsorbents. Rengaraj et al. (2002) conducted one study on the adsorption of phenol from water and wastewater using activated carbon produced from palm kernel, and observed that phenol removal efficiency increased up to the optimum adsorbent dosage of 1 mgL^{-1} [32]. Sarkar and Acharya, in another research, studied on the phenol removal and its derivatives from the contaminated water using fly ash optimum adsorbent dosage of 0.02 mgL^{-1} [33].

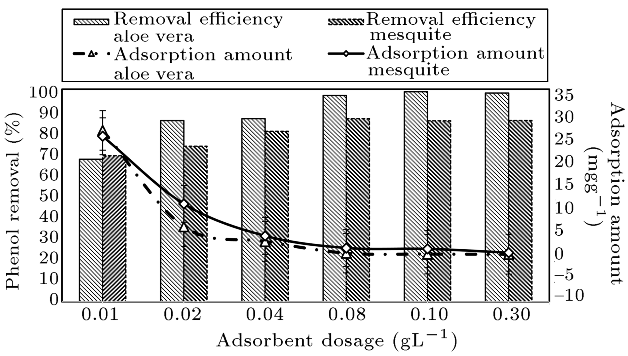


Figure 3. Effect of mesquite and aloe vera nanoparticles dosage on phenol adsorption. Condition: pH: 7.0, the initial concentration of phenol: 8 mgL^{-1}, and contact time: 40 min.

3.4. The effect of particle size of the nano-bioadsorbent

The size of particle provides valuable information on achieving optimum usage of adsorbent and adsorption capacity [34]. Particle size of 95 nm was applied to the adsorption kinetic experiment of this study to remove phenol. The smaller the adsorbent particles, the faster the diffusion, because more tiny pores are exposed to adsorbate molecules. In addition, the existence of large number of smaller particles would provide the adsorption system with a greater surface area for adsorption of phenol from the solution. The adsorption capacity is directly proportional to the total surface area exposed, whereas it is inversely proportional to the particle size diameter for non-porous adsorbents [35,36]. In a study conducted by Roostaei and Tezel (2004) on phenol removal through the adsorption using silica gel, HiSiv^{TM} 3000, activated alumina, activated carbon, Filtrasorb-400, and HiSiv^{TM} 1000, it was revealed that powdered HiSiv^{TM} 1000 (particle size < 100 μm) has the best kinetics result with the highest rate of adsorption [37]. Sarkar and Acharya (2006) conducted another study on the removal of phenol and its derivatives using fly ash. The results showed that by resizing the adsorbent particle from 0.125 to 0.053 mm, the adsorption of phenol, 1,2-dihydroxy benzene, 1,3-dihydroxy benzene, and 4-nitrophenol increased from 31% to 70%, 19% to 40%, 22% to 48%, and 26% to 56%, respectively [33].

3.5. The effect of initial concentration of phenol

The experiment's findings depicted in Figure 4 demonstrate that phenol adsorption is a function of initial concentration. The maximum phenol removal efficiency of both nano-bioadsorbents was found at initial concentration of 32 mgL^{-1}. In other words, the highest removal efficiency was equivalent to 99.4% and 98.65% for aloe vera and mesquite nanoparticles, respectively, where the rate of phenol removal decreased with higher concentrations. This efficiency reduction is due to a decrease in the number of available active sites on the surface of the adsorbent and the saturation of these

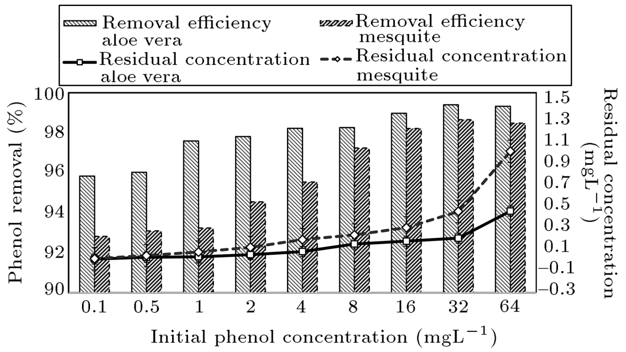


Figure 4. Effect of initial phenol concentration on its adsorption. Condition: pH: 7.0, nano-bioadsorbent dosage: 0.08 gL^{-1}, and contact time: 40 min.

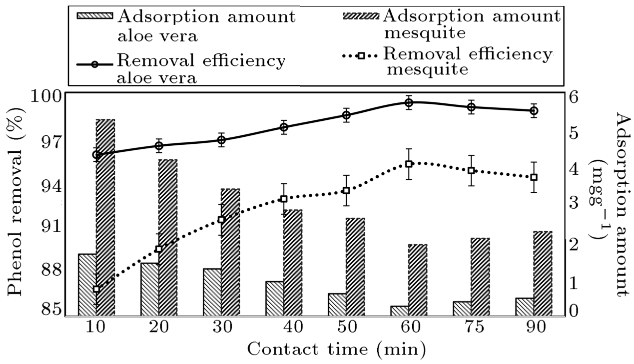


Figure 5. Effect of contact time on phenol adsorption. Condition: pH: 7.0, nano-adsorbent dosage: 0.08 gL^{-1}, and phenol concentration: 32 mgL^{-1}.

locations on the adsorbent surface. Inefficient removal of phenol in higher concentrations can be related to the reduced rate of mass transfer and subsequent reduction in adsorption capacity [32,38]. Differences between aloe vera and mesquite as nano-bioadsorbents revealed that the efficiency of phenol removal has greatly increased by mesquite with a steeper slope than aloe vera, whereas both nano-bioadsorbents had the maximum adsorbent capacity at the initial concentration of 32 mgL^{-1} of phenol. The increases in the level of phenol solution were shown to reduce the removal efficiency. This study also showed that adsorption capacity moved upward with increasing concentrations of phenol, and the maximum adsorbent capacity was obtained at initial concentration of 64 mgL^{-1} of phenol. In a study conducted by Mahvi et al. (2004) on the application of agricultural waste to remove phenol from aqueous environment, it was found that ash of rice bran had higher capacity (98%) for phenol adsorption than the rice bran (44%), and that the adsorption capacity of adsorbent was raised by increasing the initial concentration of phenol [39]. Similarly, Saitoh et al. (2011) investigated the phenol removal from water by applying bonded chitosan to heated polymers, and found that the removal efficiency remains constant by increasing phenol concentrations from 0.2 to 0.3 mM; however, it is still reduced by increasing the phenol concentrations from 0.3 to 1 mM [40].

3.6. The effect of contact time

Adsorption capacity decreased with increasing the contact time in both nano-bioadsorbents, where it reached the lowest value (0.4 and 1.86 mgg^{-1} for aloe vera and mesquite, respectively) with increasing the time up to 90 min. The rate of removal was speedier at the beginning of reaction. According to Figure 5, the maximum efficiency of phenol removal happened within 60 min, and these values were 99.32 and 95.2% for both aloe vera and mesquite nanoparticles, respectively. The system, however, reached an equilibrium state after this time period with no significant increase in the removal efficiency. Phenol reduction in the solution did not increase by rising the contact time between nano-bioadsorbents and phenol. Such an increase in removal efficiency is due to more empty sites available for phenol adsorption at shorter contact times where the phenol contact increases with empty sites. The phenol in solution occupied the empty sites until 60 min passed, and the removal efficiency reached its maximum capacity [34,41]. In their study, Mohanty et al. (2005) used activated carbon produced from sawdust tree in tropical regions, and found that the phenol adsorption and phenol removal efficiency increased from 36% to 63% by increasing contact time from 20 to 180 min [42]. In another study, Gutierrez et al. (2012) evaluated the usage of copper slag to catalyze the advanced oxidation processes for phenol removal from water, and it was revealed that the application of UV/H_2O_2/copper slag and H_2O_2/copper slag reduced phenol at 30 and 90 min time intervals, respectively. However, the removal of phenol in up to 120 min was not considerable [43].

3.7. The effect of temperature

Figure 6 shows that the removal efficiency of phenol on nano-bioadsorbents increases with increasing tem-

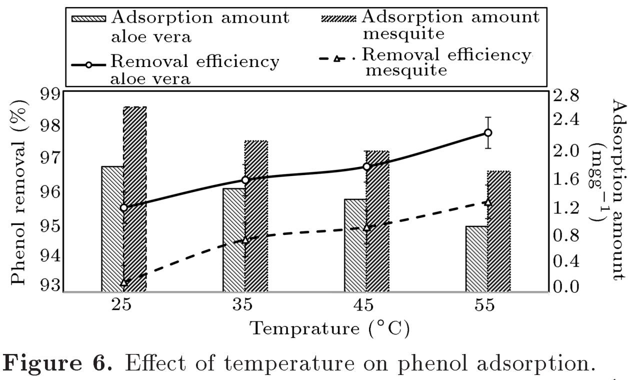


Figure 6. Effect of temperature on phenol adsorption. Condition: pH: 7.0, nano-bioadsorbent dosage: 0.08 gL^{-1}, and phenol concentration: 32 mgL^{-1}, and contact time: 40 min.

perature, particularly when the temperature increases from 25 to 55°C. The removal rate was the highest at 55°C where 97.62% and 95.62% of phenol were removed by aloe vera and mesquite nanoparticle, respectively. The increase in removal efficiency together with increased temperature can be caused by the expansion of adsorbent and increases in active sites available for phenol adsorption [44]. The 2% difference could be due to aloe vera available sites being available more than the mesquite sites. Similar findings have been obtained by the studies conducted on pollutant removals. Examples include the biological phenol adsorption using chicken feathers conducted by Qadeer, Indigo Carmine dye adsorption by poultry feathers carried out by Mittal, and Lead adsorption by applying chicken feathers done by Dela Rosa [45-47]. Srivastava et al. conducted a similar study (2006) on the phenol removal by bagasse fly ash and activated carbon. The results showed that phenol adsorption increases by an increase in temperature from 25 to 45°C [48], but another study by Kilic et al. (2011) on phenol removal, by the activated carbon prepared from residues of tobacco, indicated that adsorption efficiency decreased by increasing temperature from 20 to 50°C [49].

3.8. Effects of ionic strength

In the present study, NO_3^-, Cl^-, SO_4^{2-}, and HCO_3^- ions in the form of $NaNO_3$, NaCl, Na_2SO_4, and $NaHCO_3$ were separately used to evaluate the effect of ionic strength (0.01-0.25 M concentrations) of the solution on the phenol removal through aloe vera and mesquite nanoparticles. The results are shown in Figures 7 and 8. Experimental findings indicate that an increase in the salt concentration resulted in a decrease of phenol adsorption by nanoparticles obtained from aloe vera and mesquite, in that when the concentration of salts within the aloe vera nanoparticles increased from 0 to 0.25 M, the percentage of removal efficiency decreased from 99.4% to 18.62%, 27.15%, 32.19%, and 39.68% with $NaNO_3$, NaCl, Na_2SO_4, and $NaHCO_3$ salts, respectively. Likewise, with an increase in the concentration of salts within mesquite nanoparticles from 0 to 0.25 M , the phenol removal efficiency has decreased from 98.67% to 15.46%, 21.47%, 29.81%, and 34.32% with $NaNO_3$, NaCl, Na_2SO_4, and $NaHCO_3$ salts, respectively.

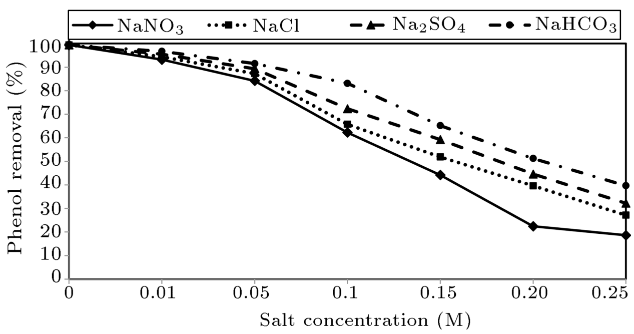


Figure 7. Effect of ionic strength on the phenol adsorption by using nanoparticles aloe vera. Condition: pH: 7.0, initial phenol conc.: 32 mgL^{-1}, nano-bioadsorbent dosage: 0.08 gL^{-1}, and contact time: 40 min.

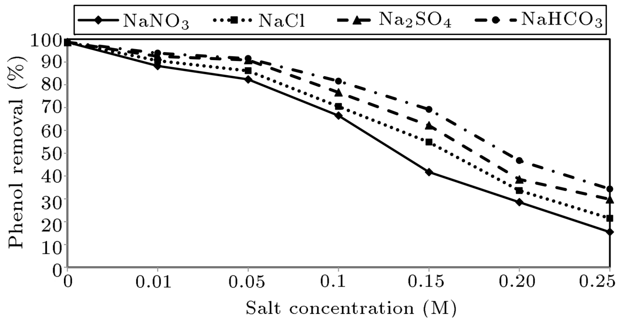


Figure 8. Effect of ionic strength on the phenol adsorption by using nanoparticles aloe vera. Condition: pH: 7.0, initial phenol conc.: 32 mgL^{-1}, nano-bioadsorbent dosage: 0.08 gL^{-1}, and contact time: 40 min.

The negative effect of NO_3^- and Cl^-, SO_4^{2-} and HCO_3^- could be explained by the fact that the adsorbent active sites may have been blocked by salt, which in turn, has resulted in increasing the hindrance of phenol diffusion on the adsorbent surfaces. The decrease in adsorption with increased ionic strength may be also due to the decrease in hydrophobic nature of the dissociated phenol molecules at pH 2.0 [2,50]. Our findings showed that the $NaNO_3$ salt exhibited a higher inhibition of phenol adsorption compared with NaCl, Na_2SO_4, and $NaHCO_3$ salts. These findings are consistent with those of the study conducted by Senturk et al. on the phenol removal from aqueous solutions by adsorption on organomodified Tirebolu bentonite, and also the work of Mukherjee et al. on the removal of phenols from water environment by activated carbon, bagasse ash, and wood charcoal [2,50].

3.9. Adsorption isotherm

Equilibrium isotherms are the most important parameters in designing the adsorption process, referring, in this case, to the amount of adsorption on nano-bioadsorbent reaching equilibrium state. In fact, we can use the adsorption isotherm to determine the maximum adsorption and also the optimum conditions of the optimal adsorption. Analysis of adsorption isotherm is essential for achieving equation in order to show accurate results and design the adsorption systems [51,52]. In this study, two well-known models, i.e. Langmuir and Freundlich isotherm models, were chosen for evaluating the relationship between the amount of phenol adsorbed onto bio-adsorbents' nanoparticles and its equilibrium concentration in aqueous solution.

The Langmuir model assumes that adsorption occurs at specific homogeneous sites on the adsorbent surface. It also assumes that when a site is occupied by an adsorbate molecule, no more adsorption takes place at this site. The linear form of the Langmuir isotherm model can be presented as in Eq. (3) [35,36], where q_e is the equilibrium adsorption capacity of phenol on the adsorbent (mgg^{-1}), C_e is the equilibrium dye concentration in solution (mgL^{-1}), q_m is the maximum capacity of the adsorbent (mgg^{-1}), and k_L is the Langmuir adsorption constant (Lmg^{-1}):

$$\frac{C_e}{q_e} = \frac{C_e}{q_m} + \frac{1}{q_m K_L}. \tag{3}$$

A dimensionless constant (R_L) is also defined as a separation factor or equilibrium parameter derived from the Langmuir isotherm in accordance with the following equation. R_L is supposed to determine the optimal ability of phenol adsorption process on nanoparticles prepared from the agricultural waste [36,40]:

$$R_L = \frac{1}{(1 + K_l C_0)}, \tag{4}$$

where C_0 (mgL^{-1}) is the initial amount of adsorbate and K_l (Lmg^{-1}) is the Langmuir constant described above. R_L parameter is considered as a reliable indicator of the adsorption. The Freundlich isotherm model is valid for multi-layer adsorption on a heterogeneous adsorbent surface with sites that have different energies of adsorption. The Freundlich model in linear form is presented as in Eq. (5) [43]:

$$\ln q_e = \ln k_f + \frac{1}{n} \ln C_e, \tag{5}$$

where K_f (mgg^{-1}) is the constant related to the adsorption capacity, and n is the empirical parameter related to the intensity of adsorption.

Physically, Freundlich model provides more reliable descriptions for pollutants adsorbent than Langmuir model which can be explained by the presence of different adsorption bands on adsorbent [53,54]. Table 2 shows the results of the correlation and constants of the two Freundlich and Langmuir models. According to these findings, isothermal information indicates that Freundlich equation describes a better understanding of phenol adsorption by the nano-bioadsorbent than the Langmuir isotherm, mainly because of higher correlation coefficient (more than 0.95 for both nano-bioadsorbents). According to R_L, the adsorption process can be irreversible ($R_L = 0$), optimal ($0 < R_L < 1$), Linear ($R_L = 1$) or unfavorable ($R_L > 1$) [50]. The calculated RL value for phenol adsorption derived from the nanoparticles of the agricultural solid waste (leaves of the aloe vera plant and mesquite trees) was between zero and one, indicating desirability of phenol adsorption in adsorbents examined. These findings are consistent with the results of Mukherjee et al. (2007) who studied the phenol removal from water by activated carbon, bagasse ash and charcoal, and the results of Potgieter et al. (2009) who carried out another study on fly ash of coal. In line with these studies, we found that the adsorption of phenol by adsorbents better fits into Freundlich isotherms model [50,55]. Yet another study conducted by Srivastava et al. (2006) showed better results of phenol adsorption by the adsorbent [48]. These varied findings show that a single model cannot be provided for pollutants adsorption by adsorbents, and that choosing an acceptable adsorption model depends on the types of pollutants and adsorbents used in the process.

3.10. The adsorption kinetics

Understanding the notion of the adsorption kinetics is also important for the mechanisms of adsorption and detection of adsorbents performance. In the current study, different Kinetic models were used for obtaining experimental results, including pseudo-first-order and pseudo-second-order adsorption kinetics [25]. The pseudo-first-order equation is shown in Eq. (6) [50]:

$$\ln(q_e - q_t) = \ln(q_e) - K_1 \times t, \tag{6}$$

where q_e (mgg^{-1}) and q_t (mgg^{-1}) are the amounts of phenol adsorbed at equilibrium at time t, and k_1 (min^{-1}) is the pseudo-first-order rate constant.

A straight line ln ($q_e - q_t$) versus t expresses the applicability of this kinetic model; q_e and K_1 can be determined from the intercept and slope of the plot. Pseudo-second-order model is expressed as follows [52]:

$$\frac{t}{q_t} = \frac{1}{k_2 \times q_e^2} + \frac{t}{q_e}, \tag{7}$$

where k_2 (gmg^{-1}min^{-1}) is the rate constant of the second-order equation.

Table 2. Isotherm parameters for the removal of phenol by aloe vera and mesquite as nanoparticles.

		Langmuir			Freundlich		
Bioadsorbent nanoparticles	**Q_0 (mgg^{-1})**	**K_L (L mg^{-1})**	**R^2**	**R_L**	**K_f**	**R^2**	**n**
Aloe vera	71.73	0.53	0.9	0.055	25.51	2.16	0.97
Mesquite	54.27	7.31	0.79	0.004	45.89	4.09	0.95

Table 3. Kinetic parameters for the removal of phenol by aloe vera and mesquite as nanoparticles.

Bioadsorbent nanoparticles	$q_{e.\exp}$ (mgg^{-1})	Pseudo first-order model			Pseudo second-order model		
		K_1 (min^{-1})	$q_{e.\exp}$(mgg^{-1})	R^2	k_2(g $mg^{-1}min^{-1}$)	$q_{e.\exp}$ (mgg^{-1})	R^2
Alo vera	2.66	0.033	0.05	0.458	0.04	1.87	0.793
Mesquite	5.27	1.002	1.76	0.671	0.19	5.24	0.968

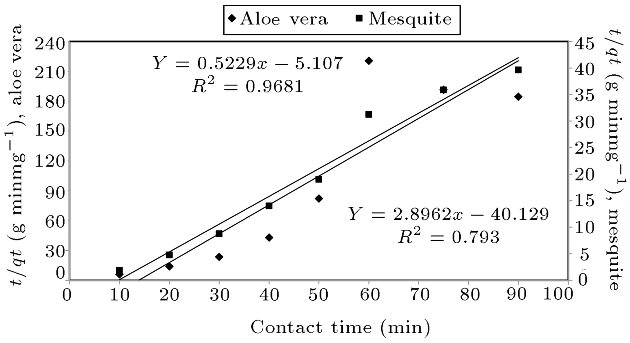


Figure 9. The pseudo-second-order kinetics for phenol adsorption by aloe vera and mesquite as nanoparticles.

The plot of t/q_t versus t would create a straight line if the pseudo-second-order kinetic model was applicable, where q_e and K_2 can be determined from the slope and interception of the plot. The rate constant of pseudo-first-order (K_1) and the calculated concentration of adsorbate of solid phase in equilibrium conditions ($q_{e,cal}$) were calculated through ln ($q_e - q_t$) plot versus t. The results are shown in Table 3. The correlation coefficient (R^2) was relatively low, which may indicate an inappropriate relationship. In addition, the amount of $q_{e,\text{cal}}$ from the model is very different from the concentration of adsorbate amount of solid phase in the equilibrium conditions ($q_{e,\exp}$). The phenol adsorption by bioadsorbents nanoparticles through pseudo-first-order model is thus inappropriate. The linear plot (t/q_e) versus t is shown in Figure 9 for the pseudo-second-order. The rate constant of pseudo-second-order (K_2) and amount of $q_{e,\text{cal}}$ were determined through the model, and the results are shown in Table 3. The correlation coefficient (R^2) was high, and the calculated amount of $q_{e,\text{cal}}$ was close to the amount of $q_{e,\exp}$. According to these results, Pseudo-second-order kinetics model is well correlated with phenol adsorption by bioadsorbents nanoparticles than using pseudo-first-order model. Senturk et al. (2009) studied the phenol removal from aqueous environments through adsorption by bentonite. Najam Khan et al. (2015) investigated the phenol removal by photocatalytic activity of zinc stannate particles and zinc stannate/zinc oxide composites. In addition, Gundogdu et al. (2012) investigated the ability of activated carbon produced from Tea industry waste in the adsorption of phenol. The results of this study are consistent with those reported by these authors [1,2,56].

4. Conclusions

The adsorption ability of nanoparticles obtained from the leaves of aloe vera plant and mesquite trees was investigated to explore the removal of phenol from aqueous solutions. The results indicated that aloe vera and mesquite nanoparticles are efficient as promising adsorbents in removing phenol from solution. A phenol removal efficiency of more than 96% was achieved under optimum conditions (pH = 7, adsorbent dosage = 0.08 gL^{-1}, initial phenol concentration = 32 mgL^{-1}, contact time = 60 min at 55°C) for both nano-bioadsorbents. Phenol removal from aqueous environment by bio-adsorbents nanoparticles follows the pseudo-second-order kinetics model. This method can be considered as a suitable method for phenol removal from industrial wastewater, reusing wastewater, reducing the irreversible health and environmental adverse effects of phenol, and the possibility of producing low-cost aloe vera and mesquite nanoparticles.

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