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Effects of Encapsulating Process Parameters of Magnetic Chitosan into Alginate Beads for Removal of Aspirin

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Aspirin is one of the drugs that is widely used to treat pain, reduce fever, and inflammation. Improper disposal of aspirin leads to pollution of the environment such as aquatic and human health by entering the food chain. Thus, this study uses incorporated magnetic nanoparticles (MNPs), chitosan (Cs), and alginate (AL) adsorbent (termed as MCsAL) beads through a cross-linking process by using glutaraldehyde as a linking agent for aspirin removal in a batch system. The influence of various parameters, including $CaCl_2$ molarity, drying time, impregnation time, and alginate concentration, on the adsorption performance was explored. The results demonstrate that the optimal conditions for MCsAL synthesis involve a $CaCl_2$ molarity of 0.05 M, a drying time of 120 minutes at 60 °C, an impregnation time of 120 minutes, and an alginate concentration of 2.8% v/v. MCsAL adsorbent bead exhibited significant adsorption capacity for aspirin. The obtained beads were characterized by Fourier-transform infrared spectroscopy to confirm the success of the synthesized beads. In addition, the study further discusses the parameters such as pH and adsorbent amount on the adsorption of the removing aspirin process. In conclusion, the study highlights the potential of MCsAL as a promising adsorbent for the removal of pharmaceutical contaminants from water bodies.

Keywords: Adsorption; chitosan; magnetic nanoparticles; alginate; aspirin

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Pharmaceutical waste, particularly the presence of aspirin, poses a significant threat to water quality. Aspirin, a common over-the-counter medication can enter water bodies through various pathways, including improper disposal of unused or expired medication, wastewater treatment plant effluent, and agricultural runoff. Once in aquatic environments, aspirin can disrupt the delicate balance of ecosystems, affecting aquatic organisms and potentially impacting human-well-being. Exposure of aspirin to humans directly can give a side effect such as skin irritation, respiratory and eyes tract. Aspirin can be absorbed and trigger allergic reactions when it contacts with the skin. Plus, prolonged use may lead to gastrointestinal bleeding [1]. Given the widespread use and improper disposal of aspirin, it is crucial to understand its potential impact on aquatic ecosystems. Aspirin was tested on fish (*Labio rohita*) at the lowest-observed-adverse-effect level (1 µg/L) for 7, 14 and 28 days. The outcome shows the vacuolization at hepatocytes as early as 7 days observation and it increased day by day until it became prominent at 28 days. Plus, bile stagnosis also occurs at 21 days which helps in the liver damage [2]. This shows that even at very low concentration, aspirin can harm aquatic life. In addition, a study conducted by [3] shows that aspirin significantly suppresses stem cell proliferation and hinders blastema growth in planarians. Planarians are tiny, flatworms with an extraordinary ability to regenerate. This makes them ideal organisms for monitoring water quality as their behavior and physical changes are highly sensitive to toxic substances. Their remarkable regeneration ability, however, can be disrupted by toxins, making them even more sensitive to water pollution. In recent times, the employment of pharmaceuticals has shown a significant increase. For instance, aspirin has been listed by the World Health Organization (WHO) as an important anti-inflammatory drug with high usage and production of around 35,000 metric tons every year [4]. According to Nordin et al. [1], the production of aspirin ends up being thrown away or improperly disposed and leading it to become highly present in water bodies. While pharmaceuticals are essential for public health, their improper disposal can lead to serious environmental consequences [5, 6]. Therefore, if the disposal of aspirin into water bodies becomes inevitable, it would be a major catastrophe for the ecosystems.

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Presently, conventional wastewater treatment technologies are inadequate in eliminating pharmaceuticals from wastewater completely [7, 8]. This is because they are designed based on primary and secondary practices, which are only efficient for removing suspended particulates, inorganic nutrients, and biodegradable organic debris. Therefore, an advanced process (tertiary) is needed to treat pharmaceutical wastewater efficiently. However, the addition of a tertiary step, which works as a wastewater refinement stage prior to the discharge into the environment, is an uncommon action in most wastewater treatment plants. The main restriction lies with the operational process complexity associated with tertiary treatments and the formation of by-products at the end of the treatments [9]. In these regards, adsorption has been considered a promising approach due to its simplicity (compared to other technologies) and efficiency without undesired by-product generation in wastewater.

Despite its advantages, most of previous researchers used carbon-based adsorbent [10-12] to remove aspirin effectively. However, due to some limitation to produce carbon-based adsorbent such as time consuming, required high energy and consume chemical, thus, natural-based adsorbents such as chitosan [13], cellulose [14] and alginate [15, 16] were introduced to treat water from improper disposal of aspirin. This natural adsorbent has not been explored in detailed. As noted in the literature, the usage of natural polymers has drawn particular attention as a result of their appealing qualities, such as their abundance and environmental friendliness. In addition, the plentiful functional groups in their structures make them possible for physical and chemical modifications to enhance their adsorption capacity. For example, chitosan, the second most abundant biopolymer on the earth, contains amino and hydroxyl groups which possibly to take part in the complexation reaction with many pharmaceutical pollutants, such as acetylsalicylic acid [17] and acetaminophen [18]. Moreover, regarding its unique properties, such as biocompatibility, biodegradability and non-toxicity, chitosan has also been utilized widely in removing other pollutants, such as dyes [19, 20], heavy metals [21, 22]. However, it is challenging to directly apply chitosan in wastewater treatment applications due to its poor physicochemical properties, such as weak acid resistance and low volume density [23]. To address the drawbacks, an appropriate modification of chitosan surface chemistry can be done as a necessary approach to improve its adsorption properties and efficiencies.

Tao et al. [23] emphasized that sodium alginate gel beads are great bases for the development of novel composite adsorbents. It is known that sodium alginate is a secure polymer that may boost the ability of composite materials to adsorb substances in aqueous solutions, resulting in hydrogel [24]. However, alginate exhibits increased swelling in alkaline environments. This property, while advantageous in certain applications, can lead to mechanical instability and bead breakage when employed as filter column packing. Such structural failure can result in secondary contamination or even complete clogging of the filter [25]. The combination of Cs and alginate as an adsorbent has the potential to mitigate the individual limitations of both components [26]. By synergistically leveraging their properties, this approach aims to enhance adsorption capacity for aspirin without producing harmful chemicals into the environment while ensuring the stability and durability of the adsorbent material [27]. Moreover, magnetic nanoparticles (MNPs) compared to more traditional methods like centrifugation and filtration, magnetic separation is claimed to be more effective in separating solids from liquids. Therefore, the incorporation of magnetic nanoparticles (MNPs) onto chitosan-alginate bead makes the separation easier with an external magnetic field.

Magnetic separation is advantageous because the separation steps require a simple procedure without tedious and high energy consumption [28]. Therefore, this study presents the encapsulation of magnetic chitosan by alginate in bead form as an adsorbent for the efficient removal of aspirin in a batch system. To the authors' knowledge, the study on the effects of parameters in the synthesis of magnetic chitosan within alginate beads for the removal of aspirin has not been previously investigated. In this regard, this work investigates the effects of parameters, such as calcium chloride molarity, drying time and impregnation time on aspirin removal.

EXPERIMENTAL

Chemicals and Materials

The materials used, such as chitosan powder and sodium alginate were acquired from QReC. Other chemicals like glutaraldehyde and aspirin were purchased from Sigma Aldrich, and the MNPs (powder) were purchased from Merck Millipore. Without any further purification, all chemicals were used as supplied.

Experimental Work

The synthesis of the adsorbent followed the same method previously reported by Khalil et al. [29] with some modifications (**Figure 1**). Briefly, an alginate solution (2.8 % v/v) was prepared by mixing 2.8 g of sodium alginate powder in 100 ml of distilled water and stirred at 80 °C until it was completely dissolved. Then, the alginate solution was rapidly mixed for 60 minutes with a combination of

MNPs (2.8 g) and chitosan powders (5.6 g). Next, 1 ml of glutaraldehyde (GTA) was added into the solution for crosslinking for 1 hour. Under magnetic stirring, the crosslinked mixture was dropped into 0.1 M of calcium chloride ($CaCl_2$) solution using a peristaltic pump and left for 30 minutes to solidify. Finally, the adsorbent beads (termed as MCsAL) were filtered and rinsed with distilled water for several times before drying in the oven at 60 °C for 60 minutes. All the steps were repeated to investigate the percentage removal of aspirin by varying the parameters involved during the adsorbent synthesis process as presented in **Table 1**. In this setting, the adsorption experiment was conducted under the following conditions; adsorption time was 1 hour, shaking speed of the shaker was 180 rpm, adsorbent dosage was 2 g, and aspirin solution volume and concentration were 30 ml and 0.1g/L, respectively. The MCsAL was removed from the solution once adsorption was complete, and the absorbance of the filtrate was assessed using a UV-Vis spectrophotometer at an absorption wavelength of 226 nm. Following that, aspirin concentrations before and after adsorption were measured. Equation (1) was used to compute the percentage removal of aspirin.

$$\text{Percentage removal (\%)} = \frac{C_o - C_f}{C_o} \tag{1}$$

where C_o and C_f (mg/L) are the initial and final concentrations of aspirin, respectively.

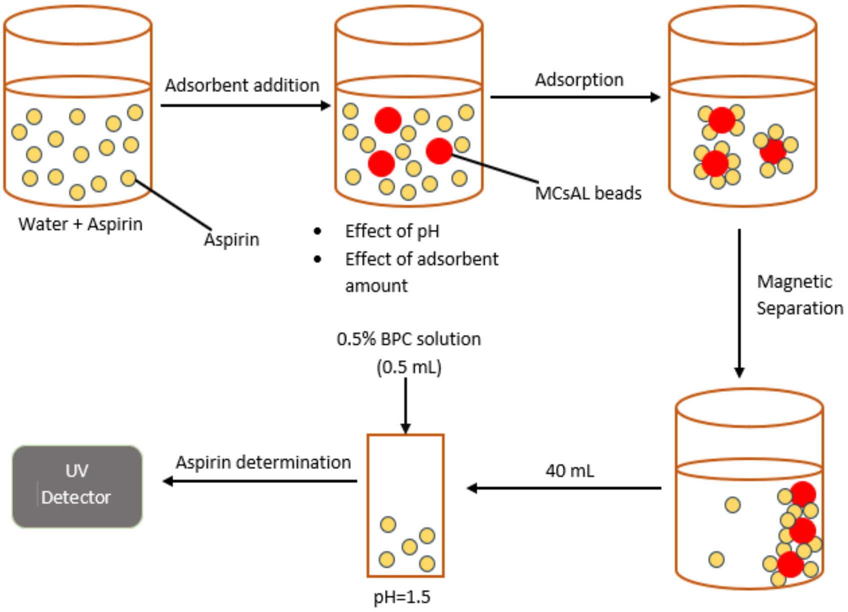

Figure 1. Schematic diagram of adsorption process.

Table 1. Parameter for experimental work.

Effects	Conditions
$CaCl_2$ molarity	0.05-0.5 M
Beads drying time	5-120 minutes
Impregnation time	5-300 minutes
Alginate concentration	08-8.4 %v/v

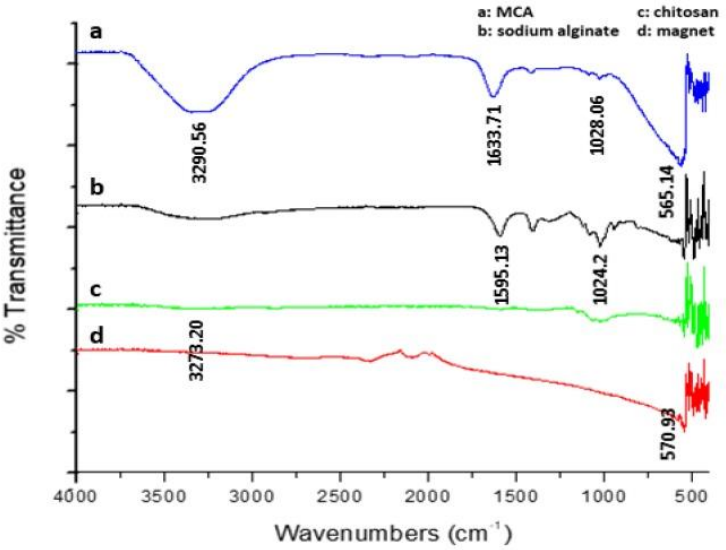

Figure 2. FTIR spectrum of (a) MCsAL, (b) sodium alginate, (c) Cs, (d) MNPs.

RESULTS AND DISCUSSION

Fourier Transform Infrared Spectroscopy (FTIR)

The changes in surface-modified composite beads, MCsAL can be observed due to alterations in the intermolecular bonds and verified by FTIR spectroscopy. As shown in **Figure 2**, the FTIR spectrum of the MCsAL beads exhibited a characteristic absorption peak for protonated Cs at 3290.56 cm^{-1} with a broad peak owing to the O-H stretching vibration of hydroxyl groups and 1028.06 cm^{-1} associated with extension of CO- bond [30]. The peak at 1633.71 cm^{-1} corresponds to the symmetric stretching peak of the carboxylate group in sodium alginate [31]. Additionally, the peak at 565.14 cm^{-1} confirms the presence of iron oxide from MNPs within MCsAL bead structure [25]. Overall, the FTIR spectrum of MCsAL indicates the successful incorporation of the functional groups of sodium alginate, Cs, and MNPs into the synthesized adsorbent.

Effect of Calcium Chloride Molarity

Figure 3 shows the effect of calcium chloride ($CaCl_2$) molarity on the percentage removal of aspirin. It can be observed that the molarity of $CaCl_2$ in adsorbent preparation showed an insignificant effect on aspirin removal. Through an ion exchange process, calcium ions from $CaCl_2$ replace sodium ions in the guluronate (G) blocks of alginate. This crosslinking reaction results in the formation of a three-dimensional alginate structure [32].

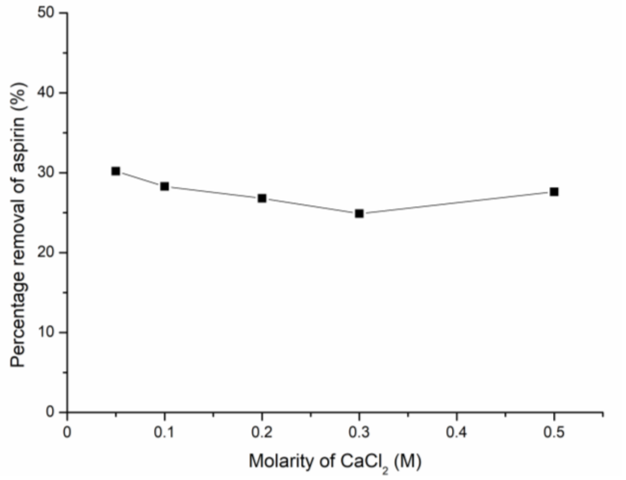

Figure 3. The effect of molarity of $CaCl_2$ on aspirin removal. (Conditions: drying time, 60 minutes; impregnation time, 60 minutes; alginate concentration, 2.8% v/v).

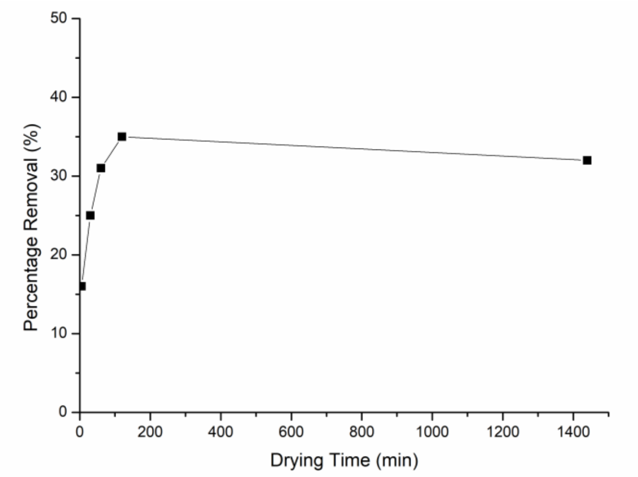

Figure 4. The percentage removal of aspirin at different drying time of MCsAL beads. (Conditions: $CaCl_2$ Molarity, 0.05 M; impregnation time, 60 minutes; alginate concentration, 2.8% v/v).

However, it is worth mentioning that the MCsAL adsorbents did not display in bead form at low $CaCl_2$ molarity (<0.05 M). This is due to an insufficient amount of Ca^{2+} ions at low $CaCl_2$ molarity. As a result, a low degree of crosslinking between calcium ions and alginate chains made them unable to develop good bead's structure [30]. Thus, the best molarity of $CaCl_2$ obtained for MCsAL preparation was 0.05 M, as the adsorbent could form in bead structure at low molarity.

Effect of Drying Time

In this study, the effect of drying time of beads on the percentage removal of aspirin was carried out, and the result is presented in **Figure 4**. Based on the outcome, it can be observed that the percentage removal of aspirin increases from 16 % to 35 % as the drying time increases from 5 to 120 minutes, respectively. According to Kaur et al. [32], the drying rate might influence the size and shape of the bead-based adsorbent as it causes shrinkage of the structure (water was removed from the beads). As a result, the size of the beads decreased, and the surface area increased [33]. In this regard, more surface sites on the beads were viable for the pollutant attachment, thus, increasing the adsorption performance. However, as the drying time was further increased to 24 hours (1440 minutes), the percentage removal of aspirin decreased to 32%. This might be due to the poor encapsulation efficiency of the beads upon prolonged heating. Consequently, chitosan, which is a pH-sensitive polymer, might not be encapsulated well. Hence, it leached and dissolved in acidic aspirin solution (pH 3) [34]. Therefore, chitosan (in its liquid state) is unable to provide the active sites for aspirin attachment and leads to a low percentage of removal. The finding reveals that the optimal drying time for MCsAL beads is 120 minutes.

Effect of Impregnation Time

Figure 5 shows the effect of impregnation time between chitosan, alginate and MNPs on the percentage removal of aspirin. The result shows that the percentage removal of aspirin increased as the impregnation time increased from 5 to 120 minutes, with the percentage removal of 19% and 58%, respectively. The low percentage of aspirin removal in a short period of impregnation time may be due to the mixture not mixed homogeneously (as shown in **Figure 6**) with the inadequacy of time until it reached homogeneity at 120 minutes. As a result, chitosan, which carries the amine groups, was not spreading well on the surface of the adsorbent. Thus, less number of active sites are available for the attachment of aspirin molecules during adsorption. Though the effects of the carboxyl group in alginate structure may associate with attracting aspirin molecules [35], the main active groups lie in the amine functions. Also, as the duration of impregnation time increased over 120 minutes, the percentage of removal was slightly decreased. It was observed that, at a longer impregnation time, the solution was exceedingly viscose and challenging to pour dropwise by syringe in $CaCl_2$ solution. Therefore, the best impregnation time between the alginate, chitosan and MNPs was 120 minutes, with adequate workability of the solution.

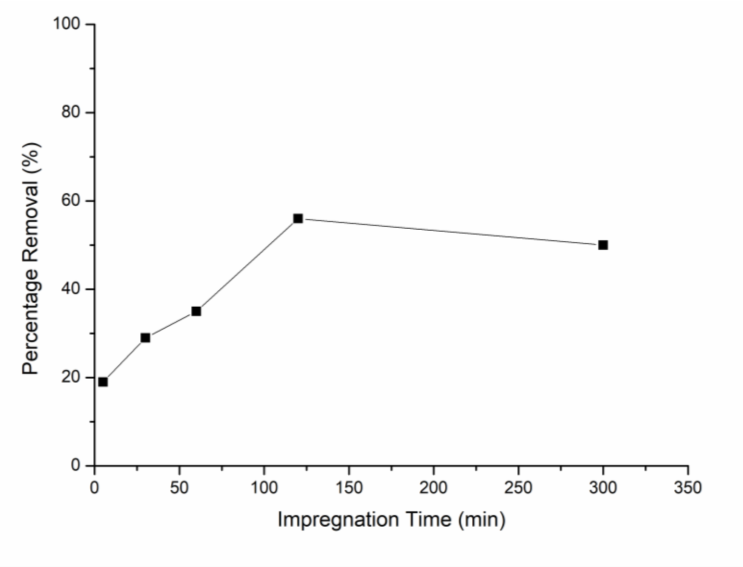

Figure 5. The percentage removal of aspirin at different impregnation time. (Conditions: $CaCl_2$ Molarity, 0.05 M; drying time, 120 minutes; alginate concentration, 2.8% v/v).

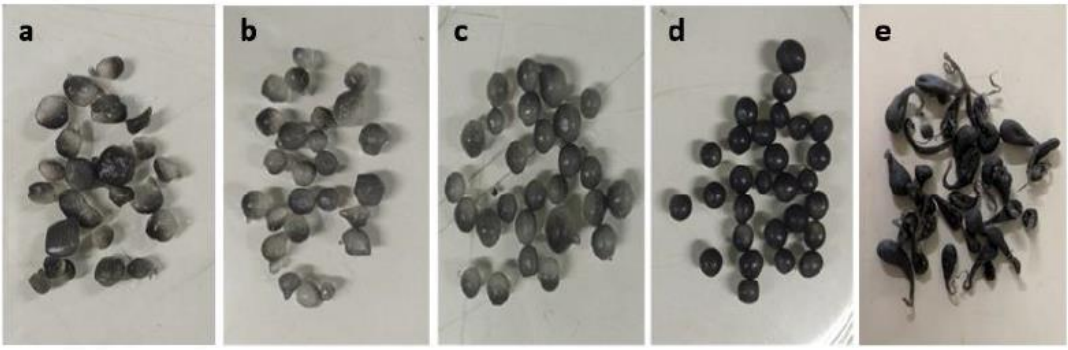

Figure 6. The bead structure at different impregnation time (a) 5 minutes (b) 30 minutes (c) 60 minutes (d) 120 minutes and (e) 300 minutes.

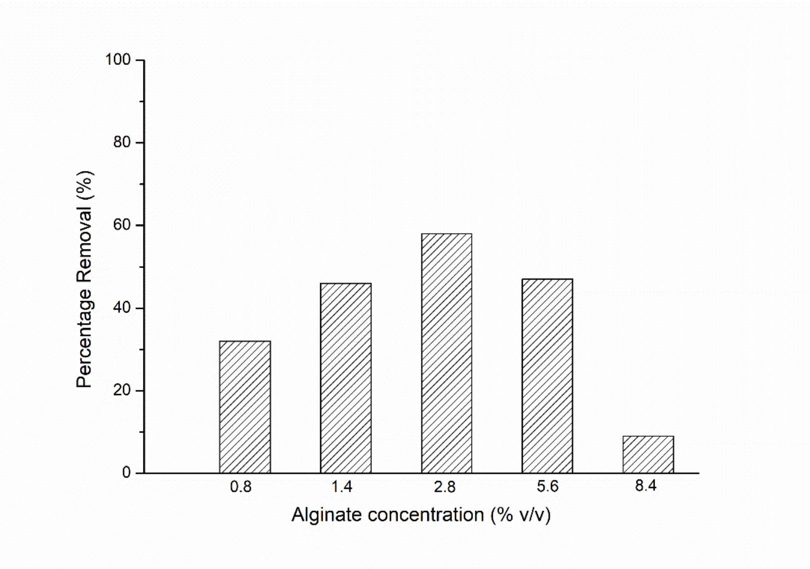

Figure 7. The percentage removal of aspirin at different concentration of alginate. (Conditions: $CaCl_2$ Molarity, 0.05 M; drying time, 120 minutes; impregnation time, 120 minutes).

Effect of Alginate Concentration

Figure 7 shows the effect of alginate concentration on the percentage removal of aspirin. It is observed that as the concentration of alginate increases from 0.8% v/v to 2.8% v/v, the percentage removal also increases from 32% to 58%, respectively. The observation may be due to a better structural bead developed as the alginate concentration increased. As a result, the chitosan's active sites are well protected in the acidic aspirin solution and become accessible for adsorption. However, the rate of aspirin removal decreases as the alginate concentration increases to 8.4% v/v with a percentage removal of 9%. This may occur due to a high alginate concentration leading to the amine sites becoming over protected, which restricts attracting the aspirin molecules. Although the carboxyl groups of alginates may attract the aspirin molecules through hydrogen bonding, the electrostatic attraction by amine functionals is more effective. This is because, in an aqueous solution, the measured pH of aspirin (without adjustment) was 3, which is acidic [36]. Therefore, the protonated amines have high affinity and stronger attraction towards negatively charged aspirin. Unfortunately, at a high alginate concentration, adsorption may lie on the weak forces of interaction (such as hydrogen bonding), which results in low percentage removal.

Effect of pH

The pH of the solution plays an important role in controlling the surface charge of adsorbent, the degree of ionization of adsorbate in the solution as well as dissociation of various functional groups on the active sites of the adsorbent. In most cases, pH is considered as a 'master variable' [37, 38]. **Figure 8** indicates the effect of the pH of the solution and the percentage of removal of aspirin. As shown, the percentage of removal of aspirin decreased with the increase in pH. The percentage of removal is highest at pH 3 corresponding to 22.27%. This situation is related to the point of zero charge (PZC) of the MCsAL [17]. The PZC of MCsAL is reported to be 6.8. The percentage of aspirin removal is higher when the pH is below pH_{PZC}, where MCsAL beads are positively charged [26]. In aqueous solutions, the aspirin is a weak acid (pKa=5) and can partially dissociate in water to form negatively charged ions [39]. This results in strong electrostatic interaction arising between the high protonation of MCsAL surfaces and the negatively charged aspirin anions. However, when the pH increases, the surface of MCsAL is deprotonated and becomes negatively charged in alkaline solution. The change in surface charges of MCsAL can be correlated with the decreasing in aspirin adsorption in higher pH of aspirin solution (more than pH 3) as the repulsion between negatively charged MCsAL and aspirin hindered the adsorption process [40]. Moreover, there was also competition between hydroxide (OH-) and the aspirin anions for positively charged adsorption sites at high pH [41]. This results in lower adsorption capacity, corresponding to a lower percentage of removal of aspirin. Hence, the adsorption of aspirin by MCsAL bead was more favorable at pH 3.

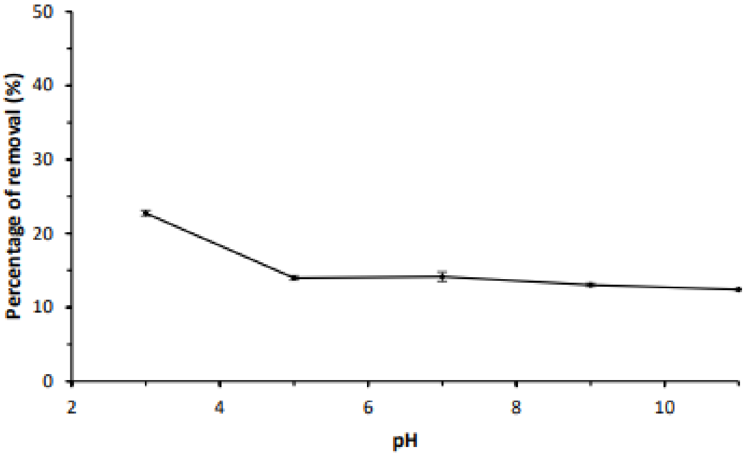

Figure 8. Effect of pH on aspirin removal (Conditions: contact time: 120 minutes, initial aspirin concentration: 0.1 g/L, solution temperature: 30 °C, adsorbent dosage: 1.0 g).

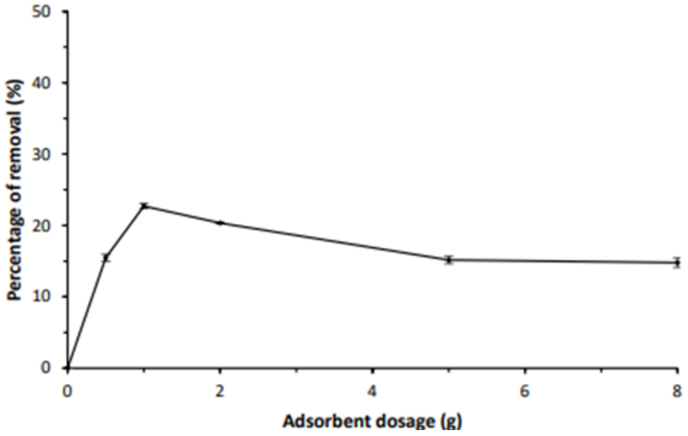

Figure 9. Effect of adsorbent amount on aspirin removal (Conditions: contact time: 120 minutes, initial aspirin concentration: 0.1 g/L, solution temperature: 30 ℃, pH: 3).

Effect of Adsorbent Amount

Adsorbent dosage is important in determining the adsorption capacity for a given amount of the adsorbent at the operating conditions. The effect of adsorbent dosage offers an economic point of view by recognizing the capability of the adsorbent to adsorb pollutants with the smallest amount of adsorbent. **Figure 9** shows the effect of adsorbent dosage on the adsorption performance of aspirin by MCsAL. As shown, 1 g of MCsAL dosage is the most effective where the percentage of aspirin removal is highest. This may be due to increased adsorbent surface area and the availability of adsorption active sites with higher doses [17]. However, the adsorption capacity decreases when the adsorbent dosage increases. This is consistent with [42] in the study on the adsorption of Pb(II) onto Fe_3O_4-GO/Ch beads. This is because at higher adsorbent amounts, the available active sites also increase, however, it cannot be occupied by the adsorbate due to its limited number.

CONCLUSION

In this paper, the effects of parameters involved during adsorbent synthesis processes on the percentage removal of aspirin were studied. The best $CaCl_2$ molarity obtained to synthesize the MCsAL adsorbent was 0.05 M, with an excellent bead structure observed. This study proved that the $CaCl_2$ had an insignificant impact on aspirin removal efficiency as it is only involved in the formation of the three-dimensional structure of the adsorbent beads. However, the adsorption of aspirin onto MCsAL is highly dependent on the drying time of the beads (120 minutes), impregnation time of materials (120 minutes) and alginate concentration (2.8 g). The adsorption study shows that the adsorption of aspirin is high at pH 3 and an adsorbent dosage of 1 g, with the percentage removals of aspirin was 22.27%. A further investigation into the optimization of adsorbent synthesis process and adsorption study is needed to enhance the adsorption of aspirin onto MCsAL.

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