Zeolite as a natural adsorbent for nitrogenous compounds removal from water

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Abstract. Water is a vital element to the survival of humans and other life forms. Yet, this source is being contaminated due to pollution leading to significantly limited freshwater, which threatens humans’ existence. Nitrates and ammonium are water contaminants and their concentration has vividly increased owing to their applications as farm nourishments. High concentrations of such contaminants in water can lead to health issues. Thus, controlling the concentration levels of these pollutants in water grows into the main task for environmentalist. Thus, a natural zeolite filter was employed in this study to minimize the traces of contaminants from water. Samples of synthetic water have been prepared and used in the laboratory tests that contain 50 mg/l of each contaminant. Using natural zeolite dose ranged from 1-5 g/l at various pH level (between 3-10) showed that the zeolite filter significantly improved the water quality for initial concentrations of each pollutant ranged from 10 to 50 mg/l. The outcomes showed that more than 93 % of both contaminates (nitrate and ammonium) were separated after using 5 g/l dose of zeolite in neutral pH level range for 120 minutes.

1. Introduction

Massive amount of water is available on planet earth covering about 70% of its surface, however, the freshwater is about 2.5% while the remaining water is salty [1-3]. It is undeniable that less than 1% of the total quantity of freshwater is available for human consumption as the majority of freshwater is captured in the ice cover or groundwater [4-6]. Furthermore, industrials sector expansion has led to a significant increase in global pollution, which dispose huge quantities of polluted wastewater and significantly minimises the quantity of freshwater available for human use [7-9].

Recent studies highlighted that freshwater is being polluted in an increasing trend with different pollutants, such as biological pollutants [10-12], heavy metals [13, 14], phosphates [15, 16], nitrate [17, 18], fluoride [19], turbidity [20, 21], phenols [22] and dyes [23-25]. Nitrogenous ions pollutants such as
nitrates and ammonium are a group of the main water pollutants which could be the source of serious health and environmental issues [15, 17]. Water pollution nowadays is more severe than ever before due to the global warming that unbalanced water consumption [26-28], precipitations [29-31], and urbanisation [32-34]. It is worthy to mention that the human activities are also the main reason of the global warming [35-38]. Therefore, researchers currently employing different treatment techniques including biological, chemical-coagulation, physical, and combined techniques to remove contaminants from the water or the wastewater [39-45].

Biologically, the contaminants are converted into gaseous using microorganisms, which removed from the water. Biological approaches were used by several researchers for water/ wastewater treatment. For instance, Bidhendi [46] utilised anaerobic microorganisms to remove nitrate from water and found that 120 minutes treating the polluted water was able to eliminate more than 75 % of nitrate and about 80 % of chemical oxygen demand (COD). Additionally, anaerobic microorganisms were applied to reduce nitrogenous contamination in water and highlighted that more than 93 % of nitrogenous contamination were removed. However, the biological methods have several drawbacks such as treatment duration, the required area for construction, sensitivity to ambient temperature, and pH of water which significantly limit their applications [15].

Chemical and physical de-nitrification are commonly used for nitrates removal from water. In these processes, chemicals including iron or aluminium salts are used to covert the nitrogenous ions into gases [47]. Although chemical methods remove large quantities of nitrates, experiments specified that these methods consume a high amount of metallic salts and produce large volumes of sludge [18], which in turn requires a complex solid wastes management plans [48-51]. On the other hand, the contaminants are removed in physical methods through the trapping them on the filter media [11, 14]. The literature pointed out that many industrial constituents can be used to remove nitrogenous contamination from water including clay adsorbents and activated carbon.

Other scholars combined both chemical and physical methods to remove pollutants from water. Dosta [52] employed the coagulation-flocculation to improve ammonium and other pollutants removals from wastewater. Additionally, the authors highlighted that the application of the coagulation-flocculation removed 1.17 kg /m³.day of the COD. Yet, using technique combination is expensive and needs several controlled environments.

Researchers painted that filtration techniques are a more attractive option to remove the nitrogenous ions due to their cost-effectiveness and easy operating. Additionally, some literatures employed a mixture of zeolite and clay to remove ammonium from water and showed that 61.10 % of ammonium were removed after 120 minutes of filtration at a pH level of 5.5.

According to the literature, filtration methods can provide an attractive option to remove pollutants from water and wastewater such as nitrogenous ions. The filtration methods can efficiently remove the nitrogenous ions form water and wastewater in addition to other attractive advantages including low cost, ease of operating, and eco-friendly techniques. Therefore, the current research investigates the usability of natural zeolite filter (clinoptilolite) in the removal of both nitrogenous ions’ pollutants from water. The clinoptilolite is used in this research as it is available and inexpensive natural material.

2. Methodology

2.1. Materials
The chemicals and zeolite used in this study were provided by the Department of Civil Engineering, Liverpool John Moores University. All materials were implemented in the experiments without adjustment or decontamination. The filter used in this research is Clinoptilolite. The Clinoptilolite has been selected in this research due to its wide application in water and wastewater treatment [15].

2.2. Solution
Initially, the polluted solution that contains 200 mg/l of nitrates and 50 mg/l ammonium has to be produced for lab experiments. A suitable amount of KNO₃ has to be dissolved in 4 litters of deionised water to have 200 mg/l of nitrates. Then, the 50 mg/l of ammonium in the solution was achieved by adding anhydrous ammonium-chloride salt NH₄Cl. Subsequently, the prepared polluted water was
cooled to produce samples with fewer pollution concentrations such as 10, 30, and 50 mg/l for both pollutants. Additionally, HCl and NaOH were used to control the pH value in the solution.

2.3. Batch filtration process

Batch experiments were employed to remove both pollutants (nitrates and ammonium) from the polluted water by zeolite filter. 500 ml of contaminated water was filled in a 1000 ml plastic vessel that comprises the filter media (zeolite) and kept for one hour and 40 minutes. Then, 10 ml samples to test the removal efficiency of the filter were collected every 20 minutes. The concentration of both pollutants was measured using Hach-Lange spectrophotometer (DR-2700), standard nitrate cuvettes (LCK 339 and LCK 340), and standard ammonium cuvettes (LCK 304 and LCK 303).

The impact of pH level on removal efficacy of the filter media was studied by altering the pH level of polluted water from acidic (3) to neutral (6.5) and basic (10). In addition, the initial concentrations’ impact of both pollutants on the elimination competence of the filter was examined by changing the concentrations from 10 to 50 mg/l while the influence of filter dose was analysed by adding several doses of zeolite including 1, 3 and 5 g/l.

Finally, the removal efficiency of the zeolite filter for nitrogenous pollutants (nitrates and ammonium) was determined using the following equation [47]:

\[ \text{Filter efficiency (\%)} = \frac{A_1-A_2}{A_1} \times 100 \]

A₁ and A₂ are the primary and ending contaminants’ concentrations, respectively.

3. Results and discussion

3.1. Influence of pH level

The influence of pH level on the removal efficiency of nitrogenous pollutants has been investigated by treating 500 ml water using 3 g/l of zeolite filter for 30 minutes at pH level values ranged from 3 to 10. Figure 1 provides a graphic presentation on the impact of pH on the removal efficacing of nitrogenous pollutants. It can be seen that the removal efficiency of both pollutants increased with the decrease in the wastewater acidity. The best removal efficiency was reached in neutral pH level (from 5 to 7). When the wastewater becomes alkaline (the pH more than 7), the removal efficiency starts to decrease with the increase in water pH level. The variation in the elimination efficiency happens due to the impact of pH on the charge of the filter superficial. Researchers highlighted that the surface of the zeolite became negative at high pH values which in turn prevent the adsorption of the negatively charged nitrates and ammonium [15]. On the other hand, the surface of the filter media (zeolite) encompasses more protons at very low pH levels which in turn minimizes the removal efficiency of the filter media [53].

Figure 1. Impact of pH on the removal of nitrogenous pollutants (nitrates and ammonium).
While at moderate pH levels (between 5 and 7), the filter media is positively charged which attracts the negatively charged nitrogenous pollutants [53]. Accordingly, a pH level of 6.5 has been selected to identify the impact of initial pollutant concentration, the zeolite dose, and contact time.

3.2. The impact of initial pollutants’ concentrations
The influence of initial pollutants concentration (nitrates and ammonium) on the removal performance of the zeolite were analysed by treating 500 ml water with a pH level of 6.5 and 3 g/l of zeolite for 30 minutes at changing concentrations that are 10, 30, and 50 mg/l. as illustrated in figure 2, the removal of both pollutants declined with the growth of their initial concentrations. This happened due to the ions of the pollutants will contest with each other for the available adsorption sites [53]. With higher initial concentration and constant zeolite dose, the available site on the filter media is significantly low than the negative ions of the pollutants that have to be absorbed leading to untreated pollutants.

![Figure 2](image2.png)

**Figure 2.** The impact of the initial concentration of the removal of pollutants.

3.3. The influence of the zeolite dose
The dosage of media adsorbent strangely influences the removal of both nitrates and ammonium as it makes the most of the superficial area available for adsorption. Accordingly, the impacts of zeolite filter on the removal of both pollutants from the water were considered by treating the same sample size (500 ml) of water for the same duration (30 minutes) and the decided pH level of 6.5. The pollutants’ initial concentration was selected to be 50 mg/l and the doses of zeolite were 1, 3 and 5 g/l to understand the impact of the zeolite on pollutant treatment. Figure 3 highlighted that the increase in the zeolite dose significantly rises the removal of both nitrogenous pollutants. This confirms that the more the zeolite dose the more the available space to absorb pollutants which significantly enhances the removal both nitrates and ammonium.

![Figure 3](image3.png)

**Figure 3.** The impact of the zeolite dose on the removal of nitrogenous pollutants.
3.4. The influence of the contact time

Time plays a vital role in any water treatment activity such as filtration. The time allows longer contact between both the nitrates and the ammonium ions and the adsorbent. Thus, the influence of treatment duration on the removal of both pollutants has been analysed by treating samples that have a sample size of 500 ml, initial pollutants concentration of 50 mg/l, a pH level of 6.5 for 120 minutes using a zeolite dose of 5 g/l. The outcome showed in Figure 4 highlighted that the removal efficiency of the nitrogenous contamination has improved to reach more than 93% for both contaminants (nitrates and ammonium) for a duration of 120 minutes.

![Figure 4](image)

Figure 4. The impact of the treatment duration on the removal of nitrogenous pollutants.

The literature highlights the importance of using sensors in both civil and environmental engineering [54-57], thus; the use of sensors in the zeolite filters to obtain the best operation conditions.

4. Conclusions

The current study investigated the use of natural zeolite namely Clinoptilolite for the treatment of nitrogenous pollutants (the nitrates and the ammonium) from water and wastewater. According to the outcome of this experimental study, it can be said that the natural zeolite (Clinoptilolite) can be considered as a suitable option for the treatment of both the nitrates and the ammonium ions from water or wastewater. Higher zeolite dose and longer contact time provide better removal of the nitrogen’s contaminants from water and wastewater. Additionally, the removal effectiveness of the zeolite filter is affected by the pH value of the treated solution as the effectiveness of the zeolite filters is negatively affected by the variations in the pH level. It can be concluded that the neutral pH is the most favourable for using Clinoptilolite for the treatment of nitrogenous pollutants. Generally, the removal efficiency of the targeted pollutants using the zeolite was excellent (about 93%).

For future studies, zeolite could be used to remove other pollutants, such as lead and organic matter, from water or wastewater.

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