Article

Assessing the Efficiency of Green Absorbent in Treating Nutrients and Heavy Metal in Wastewater

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Abstract: This study is aimed to determine the performances of zeolite-water hyacinth ash (WHA)-clay (ZWC) pellets on removing nutrients (nitrogen (N) and phosphorus (P)) and heavy metal (HM). In this study, the adsorption experiments were conducted for various pellet compositions and sizes, the application of pre-treatment on pellets, and tested with different concentrations of synthetic wastewater. The results identified that the maximum removal capacities of ZWC pellet were 0.112 mg/g, 0.08 mg/g, 0.171 mg/g, and 0.151 mg/g for phosphate (PO_4), total phosphorus (TP), nitrate-nitrogen (NO_3), and total nitrogen (TN), respectively. The optimization experiment indicated that smaller pellet sizes and those treated with calcium hydroxide solution exhibited better N&P and HM removal performances. The optimized ZWC pellet was able to remove up to 90%, 86%, 94%, 90%, 84%, 86%, and 91% for PO_4, TP, NO_3, TN, zinc (Zn), copper (Cu), and lead (Pb), respectively, after 3 h of contact time. The ZWC pellet proved that it can be used as an effective adsorbent for wastewater treatment.

Keywords: nitrogen; phosphorus; wastewater treatment; water hyacinth; zeolite

1. Introduction

The increasing trend of the conversion of land use into agricultural areas significantly increases the introduction of eroded soil into water bodies [1–3]. Soil erosion, landslides, agriculture activities, urbanization, and unplanned development associated with land-use change have significantly influenced surface runoff and river water quality [4–7]. Eutrophication or algal bloom is the phenomenon where the enrichment in the water body due to excessive nutrients causes structural changes to the ecosystem [8,9]. Water hyacinth is one of the problematic weeds in many developing countries that grow rapidly due to eutrophication conditions in the lake or reservoir water bodies. High loading of phosphorus with a soluble reactive concentration greater than 0.1 mg L^{-1} can trigger the high productivity of water hyacinth associated with favorable environmental conditions [10]. The uncontrolled growth of water hyacinths can have adverse effects on the lake ecosystem, leading to clogged irrigation systems and disrupting fishing and transportation activities within the lake. To address this issue and combat environmental pollution caused by water hyacinth, it is crucial to identify suitable control measures to reduce nutrient levels in the water.

Various nutrient removal technologies, including ion exchange, precipitation, ultrafiltration, reverse osmosis, electrodialysis, and adsorption, have been proposed [11–15]. However, these methods often demand high energy consumption and advanced operational requirements and face challenges in recovering metals and materials. Local communities and farmers find it difficult to afford the development or ownership of such technologies for treating wastewater from residential and agricultural areas. In light of this, it is preferable to explore cost-effective water treatment technologies, such as absorbents

derived from local natural materials. Numerous studies have emphasized the significant role of environmental technology in enhancing wastewater treatment processes [16–20]. By employing locally sourced absorbents, it becomes more feasible for communities to address the water hyacinth issue and manage wastewater effectively without imposing a substantial financial burden.

Different pollutant removal technologies have been applied to the removal of nutrients and heavy metal from wastewater [21]. Biosorption is one of the effective, eco-friendly, and low-cost removal technologies. In recent years, different available-in-nature, abundant, and eco-friendly agricultural by-products or wastes have been utilized to replace the conventional and expensive sorbents in the industry [22–24]. These adsorbents have shown promising nutrient removal efficiencies. Different sorbents have their sorption capacity, and the absorption rate is affected by factors such as the pH of the solution, agitation speed and contact time, flow rate, bed height of the adsorbent, initial concentration of the adsorbent, and mass and particle size of the adsorbent [25,26]. Recently, numerous studies revealed that natural and modified zeolites and bentonites, as well as zeolites synthesized from waste materials, have been widely used in wastewater treatment due to their large pore space, high resistivity to extreme temperatures, and chemically neutral structure [27,28]. The use of zeolites or modified zeolites in wastewater treatment has been recognized as an environmentally friendly and sustainable technology for the removal of wastewater pollutants such as ammonium ions, heavy metal ions, inorganic anions, and organics (humic substances, phenolic compounds, surfactants, pesticides, and pharmaceuticals).

The application of synthesized zeolitic material as an absorbent for water treatment is gaining increasing attention in recent decades. However, there is limited study on the synthesis of zeolitic material from water hyacinth for wastewater treatment. The growth rate of water hyacinth is very fast and easily available, and it has high pollutant-adsorption characteristics. Therefore, the potential of a water hyacinth-based absorbent is worth exploring, as this will contribute to sustainable water management as the source-specific removal strategy. More studies are needed to be carried out to determine the optimum effectiveness of zeolitic material synthesized from water hyacinth in water treatment. This may include nutrient removal performance under various operating conditions, wastewater characteristics, and various design configurations. Therefore, this study aimed to investigate the nutrients and heavy metal removal efficiencies of zeolite-water hyacinth ash (WHA)-clay-based pellets and optimize the absorption capacity by modifying its pellet size and pre-treatment. The remainder of the paper will proceed as follows. Section 2 describes the materials and methods utilized in this study, including materials and pellet formation, batch sorption experiments, pellet optimization process, and removal efficiency analysis. The preliminary batch sorption results are described in Section 3.1. The performance of the optimized pellet in terms of pellet size, pre-treatment with calcium hydroxide (hydrated lime), and removal performance of heavy metals are presented in Section 3.2. The conclusion of this paper is stated in Section 4.

2. Materials and Methods

2.1. Materials and Pellet Formation

Bentonite clay, Kaoline clay, zeolite powder, water hyacinth ash, and deionized distilled water (DDW) were selected as the materials for producing the absorbent pellet. The composition of bentonite clay consists of 90–100% bentonite, 1–10% Crystalline Silica, and Quartz. On the other hand, Kaoline clays consist of 96–99.9% of Kaolinite and 0.1–4% Crystalline Silica. Both Bentonite and Kaoline clays were purchased from the local manufacturer—Kaolin (Malaysia) Sdn Bhd. Meanwhile, zeolite powder consists of 37.97% SiO_2, 4.53% Al_2O_3, 4.12% Na_2O, and 2.12% K_2O. The zeolite powder was purchased from Trio Garden Supplies Sdn Bhd. The preparation of water hyacinth ash (WHA) was started by obtaining water hyacinth plants from nearby lakes, followed by extensive washing with distilled water to remove the soil and dust. The water hyacinth plants were then sliced into pieces and dried for one week. Generation of WHA was performed by heating the dry

water hyacinth plants in the oven at a temperature of 950 °C in the presence of excess air for 5 h. The resultant ash was mixed to obtain a composite sample.

For zeolite–clay pellet formation, a total of 200 g of zeolite and 160 g of clays (80 g Bentonite clay; 80 g Kaoline clay) were added in a medium steel container. These materials were mixed to become evenly distributed. Beginning with a clay: water ratio of 1:4, DDW was added into the container at 2 to 4 increments until measurable workability was achieved into a paste consistency. The paste was kneaded and formed into a 1–2 inch diameter cylinder. Then, the paste was extracted by using a melon baller of the interior volume of a ¾ inch sphere. The paste was removed from the melon baller and was reshaped to form a sphere of ¾ in diameter. The pellets were then air-dried for 24 h before they were placed into the furnace for preheating process at approximately 100 °C for 24 h. After that, the furnace temperature was increased to approximately 400 °C, and 1 hour later, to approximately 600 °C for a calcination process of 24 h. Finally, the furnace was turned off and allowed to cool down to room temperature before the pellet was removed and stored in open sealable plastic bags. These pellets were exposed to high-temperature heat treatment to produce appropriate mechanical stability and high sorption properties. This process also enhances the development of meso/macropores within the zeolite pellets. The same procedure was applied for the zeolite-WHA-clay pellet (Figure 1), where 100 g of zeolite was mixed with 160 g of clay and 100 g of WHA to form the pellet. Different types of components, including zeolite–clay, WHA-clay, zeolite-WHA-clay, reservoir sediment–zeolite, and clay-only pellets, were formed for the preliminary absorption testing.

Figure 1. Zeolite-WHA-clay pellet.

2.2. Batch Sorption Experiments

The batch sorption experiments were conducted to study the nitrogen (N) and phosphorus (P) removal characteristics of absorbent pellets. The pellets were categorized as pellet I (clay only), pellet II (zeolite and clay), pellet III (water hyacinth ash and clay), pellet IV (zeolite and water hyacinth ash and clay), and pellet V (reservoir sediment and clay). The aqueous solution was prepared with deionized water spiked with standard stock solutions (100 mg/L) of N and P. According to the previous study, two different concentrations of 5.0 mg/L and 35 mg/L were prepared for N and P, respectively. In the experiments, the pellets were placed into the 300 mL flasks with 200 mL of an aqueous solution for N and P. After the pre-determined contact times of 15, 30, 45, 60, 90, 120, and 180 min, the solutions were sampled and analyzed for N and P concentrations. Various variables, including contact time, pellet composition, and initial concentration, were investigated for their effects on N and P sorption performances. The maximum sorption capacities for N and P by absorbent pellets were estimated by using equilibrium analysis. The difference between the initial concentration and equilibrium concentration was considered as the amount of

N and P retained by the pellets. The effect of pellet composition on N and P absorption performance was also determined from the residual levels in the solutions of both N and P. Different contact time durations were used for the batch experiments to determine the equilibrium time of the sorption reaction. Five flasks filled with aqueous solutions of N and P and different pellet categories were prepared and left for contact times of 15, 30, 45, 60, 90, and 180 min.

The nitrate-nitrogen (NO_3) as an N component in the aqueous solution was analyzed using the Cadmium reduction method (Hach Method 8039). The total nitrogen (TN) concentration was analyzed according to the Persulfate digestion method as described by the standard method for water and wastewater examination [29], followed by the nitrate determination method. HACH spectrophotometer DR6000 was used to determine the NO_3 concentration with a measurement wavelength of 500 nm. In the batch experiments, the soluble reactive phosphorus or phosphate (PO_4) was measured by using the high-range test 'N tube vials (Hach Method 8114). The PO_4 concentration in the aqueous solution was tested by using HACH spectrophotometer DR6000 with a measurement wavelength of 420 nm. In this study, total phosphorus (TP) was measured by using the Acid Persulfate Digestion method followed by the phosphate method (Hach Method 8190). Total phosphorus (TP) encompasses all the different forms of phosphorus present in a sample, whether dissolved or particulate. Soluble reactive phosphorus, on the other hand, specifically quantifies orthophosphate, which represents the filterable and soluble inorganic fraction of phosphorus directly utilized by plant cells.

2.3. Pellet Optimization Process

The pellet type that performed the best in the preliminary batch sorption experiment was selected to undergo the optimization process to enhance the performance of N and P removal efficiencies. Different pellet sizes of 1.5 cm, 2.0 cm, and 2.5 cm (diameter) were tested in this optimization process. A total of 120 pellets were formed and heated in the furnace for 6 h at 100 °C and 600 °C according to the method proposed by [30]. The pellets were also conditioned by using calcium hydroxide (hydrated lime) to explore the ion exchange capacity. The pellets were rinsed once with deionized water to wash off any debris before being soaked in the calcium hydroxide solution for approximately 24 h. Before testing, the pellets were rinsed again with deionized water after being taken out from the calcium hydroxide solution. The pellets were then put into the aqueous solutions of N and P with low and high concentrations of 5 mg/L and 35 mg/L, respectively. The impact of different initial concentrations on the performance of N and P removal from the prepared solutions was assessed in this study. The pellets were left to be soaked for 15, 30, 45, 90, and 180 min before the aqueous solution was sampled for N and P analysis.

The final optimized pellet was tested for its performance in removing heavy metals such as zinc (Zn), copper (Cu), and lead (Pb) from an aqueous solution. The aqueous solutions for Zn and Cu were prepared in two different concentrations of 0.5 mg/L and 3.5 m/L. Meanwhile, the concentration of aqueous solution for Pb was determined as 100 μg/L. The final optimized pellets were soaked in the aqueous solutions of heavy metals for contact times of 15, 30, 45, 60, 90, 120, and 180 min. After that, the aqueous solutions were sampled and digested using the TNT890 Metals Prep Set with acid, oxidizing agent, and buffer solution for 1 h at 100 °C in a DRB200 reactor. Then, the residual concentration of Zn was determined using HACH Method 8009; Cu was determined using HACH method 8506, and Pb was determined using Leadtrak® Fast Column Extraction (HACH Method 8317).

2.4. Removal Efficiencies Analysis

The efficacy of nutrient and heavy metal removal by the absorbent pellet was determined for different pellet compositions, contact times, pellet sizes, pre-treated with calcium hydroxide, and initial concentrations. The percentage of N and P removal was calculated by using Equation (1).

$$R = \left(\frac{C_0 - C_e}{C_0} \right) * 100\% \tag{1}$$

where

C_0 = Initial concentration of sorbate in solution (mg/L);

C_e = Equilibrium concentration of sorbate (mg/L).

The amount of N and P absorbed onto the pellet was calculated by using Equation (2) as below:

$$q = (C_0 - C_e) \frac{V}{m} \tag{2}$$

where q = amount of pollutant adsorbed by the pellet (mg/g);

C_0 = Initial concentrations of aqueous solution for N and P (mg/L);

C_e = Equilibrium concentration of aqueous solution for N and P (mg/L);

m = Sorbent mass (g);

V = Aqueous solution volume (L).

3. Results and Discussion

3.1. Preliminary Batch Adsorption Results

The batch adsorption experiments began with testing different types of pellets at an initial concentration of 5 mg/L for contact times of 15, 45, 60, 90, and 180 min. Figure 2 shows the N and P removal results for all types of pellets after 180 min of contact time. The removal rate of NO_3 via the zeolite–clay pellet outperformed other parameters (TP, PO_4, and TN), which recorded a 72% removal percentage after 180 min. The removal percentages of TP, PO_4, and TN for the zeolite–clay pellet after 180 min of contact time were 60%, 62.4%, and 65%, respectively. Meanwhile, the WHA-clay pellet recorded removal percentages of 64%, 67.4%, 70%, and 77.2% for TP, PO_4, TN, and NO_3, respectively. The reservoir sediment–zeolite and clay-only pellets were two types that showed lower performances of N and P removal. The reservoir sediment–clay pellet recorded removal percentages of 50%, 51.4%, 55%, and 56.2% for TP, PO_4, TN, and NO_3, respectively. The clay-only pellet showed the lowest removal performances of N and P, 50% for TP, 52% for PO_4, 40% for TN, and 44% for NO_3, respectively.

The zeolite-WHA-clay-based pellet demonstrated the best N and P removal performance in the batch absorption experiment. A total of 67.4% of TP, 71.6% of PO_4, 74% of TN, and 80% of NO_3 were removed by the zeolite-WHA-clay pellet from the 5 mg/L concentration of aqueous solution after 180 min. The removal rate of PO_4 is higher than TP for all types of pellets in the batch absorption experiment. This phenomenon is probably because the bigger molecular weight of condensed phosphate and organic phosphate molecules induces a slower diffusion rate, hence causing a weaker affinity between sorbate and solid surface [31]. As proven by Razali et al. [31], the absorption capacity of different P species follows the sequence of orthophosphate > polyphosphate > organic phosphate. Therefore, it is recommended to transform other forms of P into orthophosphate as much as possible to achieve better removal efficiency in the treatment plant.

High adsorption of NO_3 and TN by zeolite-WHA-clay pellet could be due to the availability of organic carbon (OC) sources (electron donor). According to Carlson and Ingraham [32], an OC source is normally required by heterotrophic bacteria during the biological denitrification process. An OC source can be obtained from either the outside solution or the inside solid substrate. In this case, the feed synthetic solution did not possess organic matter, and therefore, it is not the source of OC for the denitrification process. The WHA contains high content of organic carbon that could be slowly released and supplied as an OC source for the denitrifiers to convert the NO_3 into nitrogen gases [33]. Mixing zeolite and WHA can achieve better removal efficiency for both N and P levels in wastewater, as WHA can enhance the denitrification process. This may also be due to the porous properties of the zeolite-WHA-clay pellet, which can further improve the absorption mechanism of P

onto the surface area of the pellet. The zeolite-WHA-clay pellet also showed a higher pH value compared to other pellet types during the absorption experiment. The pH values range from 5.5 (lowest) for the clay-only pellet to 6.6 (highest) for the zeolite-WHA-clay pellet during the batch absorption testing. It was found that lower pH levels may contribute to a lower absorption rate of pellets [34].
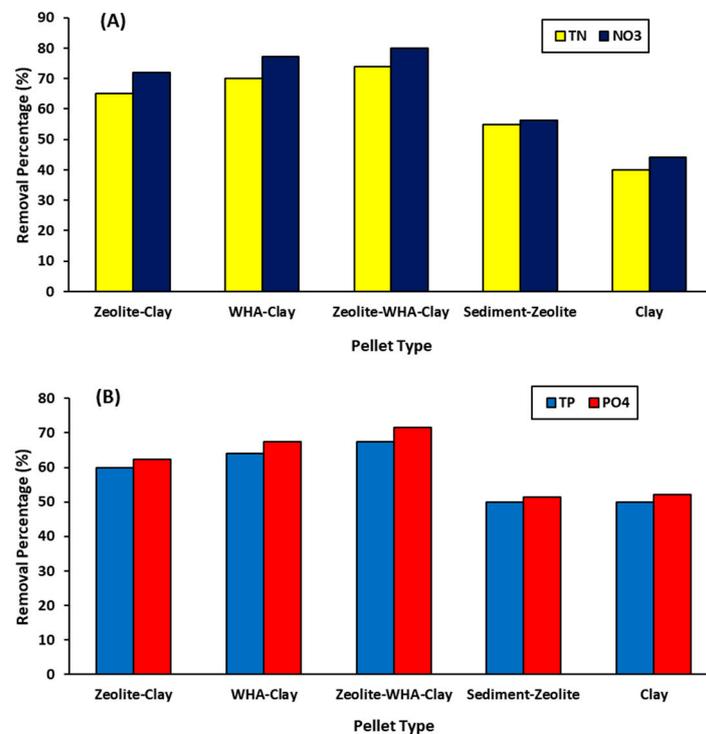
Figure 2. Comparison of (**A**) nitrogen and (**B**) phosphorus removal percentages for all pellet types after 180 min.

3.2. Performance of Optimized Pellet

Following the batch absorption experiment, the pellet type that achieved the highest nutrient removal percentages continued to go through the optimization process. The zeolite-WHA-clay pellet underwent an optimization process on different sizes (1.5 cm, 2.0 cm, and 2.5 cm) in diameter for different initial concentrations of N and P. After optimizing the pellet size, the determined pellet size was treated by using calcium hydroxide (hydrated lime) before testing for nutrient removal efficiency again.

3.2.1. Pellet Size

Figure 3 presents the results obtained from the absorption study using different sizes of zeolite-WHA-clay pellet with N and P concentrations of 5 mg/L, respectively. It was observed that the removal percentages increased inversely with the pellet size. The results show that the zeolite-WHA-clay pellet with a 1.5 cm diameter has the highest efficiency in removing the nutrients (TP, PO_4, TN, and NO_3) in the synthetic solution samples. About 71.4% of TP, 74% of PO_4 and TN, and 78% of NO_3 were removed by the 1.5 cm zeolite-WHA-clay pellet from the synthetic solution of 5 mg/L. For the 2.0 cm diameter pellet, the removal percentages were 67% for TP, 71% for PO_4, 70% for TN, and 71.4% for NO_3, respectively. The 2.5 cm diameter size of the pellet achieved removal percentages of 66%, 70%, 67%, and 70% for TP, PO_4, TN, and NO_3, respectively. The pH values observed did not have much difference compared to all tested pellet sizes. The pH values ranged from 6.1 to 6.6 for the 1.5 cm pellet size, 6.1 to 6.5 for the 2.0 cm pellet size, and 6.0 to 6.5 for the 2.5 cm pellet size, respectively. Since all pellets are made from the same material, changing pellet sizes does not affect the pH value significantly. A greater removal percentage is achieved

by a smaller pellet as a smaller pellet size will increase the surface area of absorbent that is available for nutrient uptake. Smaller particle sizes will also shorten the ions' diffusion distance to travel to the active site. Such findings in this study are comparable with the results obtained by [35,36].
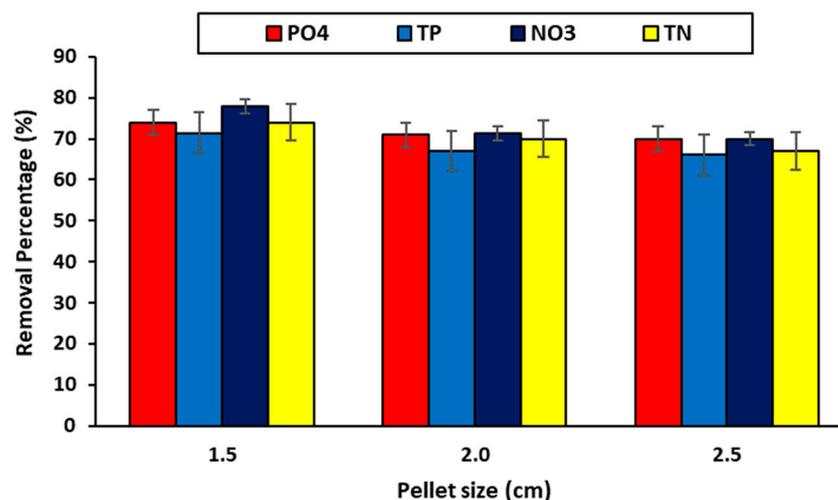
Figure 3. Removal rates of N and P for different pellet sizes in 5 mg/L concentration after 180 min.

The pellets were further tested with higher concentrations of 35 mg/L for N and P in the aqueous solution samples. Figure 4 shows the results of the adsorption study using different pellet sizes for higher concentrations (35 mg/L) of N and P. Obviously, the removal percentages of N and P via the 1.5 cm pellet are greater than that of 2.0 cm and 2.5 cm pellet sizes. The results indicate that a 1.5 cm diameter pellet is the most efficient pellet size for removing all N and P parameters in this study. About 40% of TP, 47.4% of PO_4 and TN, and 48.9% of NO_3 were removed by the 1.5 cm diameter zeolite-WHA-clay pellet in the aqueous solution with a 35 mg/L concentration at the end of the experiment. However, the magnitudes of removal by the 1.5 cm pellet were less effective for 35 mg/L of N and P concentrations compared to that of the 5 mg/L concentration. For the 2.0 cm diameter pellet, the final removal percentages were 34% for TP, 37.1% for PO_4, 40% for TN, and 41.7% for NO_3, respectively. Meanwhile, the 2.5 cm pellet had the lowest removal percentages, which were 30.6% for TP, 34.3% for PO_4, 32.9% for TN, and 36.3% for NO_3. The pH values ranged from 5.3 to 5.9 and 5.5 to 6.2, which were lower than that in the 5 mg/L concentration, respectively.

The statistical significance of differences among mean removal percentages of different pellet sizes was evaluated using one-way analysis of variance (ANOVA) at a significant level of 0.05. The removal percentages of pellets with different sizes were statistically different, with values of $p = 0.011$ and $p = 0.002$ for wastewater concentrations of 5 mg/L and 35 mg/L, respectively.

3.2.2. Pre-Treatment with Calcium Hydroxide (Hydrated Lime)

To enhance the adsorption performance, the zeolite-WHA-clay (ZWC) pellet with a 1.5 cm diameter was conditioned with calcium hydroxide for 24 h. Figures 5 and 6 show the results of N and P removal percentages in both low (5 mg/L) and high (35 mg/L) concentrations of an aqueous solution. The changes in pH values along contact times were also measured and plotted together with TP and PO_4 and TN and NO_3, as shown in Figures 5 and 6, respectively. The removal rates of TP and PO_4 in 5 mg/L of aqueous solution are significantly higher than that in 35 mg/L of concentration. The highest removal percentages of TP and PO_4 after 180 min are 86% and 90% for 5 mg/L of the solution and 60.9% and 65.7% for 35 mg/L of the solution. The pH values range from 6.5 to 6.8 for 5 mg/L of the aqueous solution and 6.2 to 6.5 for 35 mg/L of the aqueous

solution. Meanwhile, a 90% removal percentage of TN and 94% removal percentage of NO_3 are observed for 5 mg/L of an aqueous solution. For pellets that were treated with 35 mg/L concentration, the results show that 71.4% of TN and 78.6% of NO_3 were removed after 180 min of contact time. The adsorption rates of the pellet in 5 mg/L concentration showed rapid increment during the first 90 min. Meanwhile, the adsorption rate in 35 mg/L concentration showed a gradual increment as the contact time was increased. The same trends of removal percentages are reported by all N and P parameters for both 5 mg/L and 35 mg/L concentrations. The removal efficiencies follow the sequences $NO_3 > TN > PO_4 > TP$. The results demonstrated that higher adsorption was found to take place at lower initial concentrations. This finding was similar to observations reported by other researchers [37–40]. The results of the removal efficiency of conditioned ZWC pellets are summarized in Table 1.
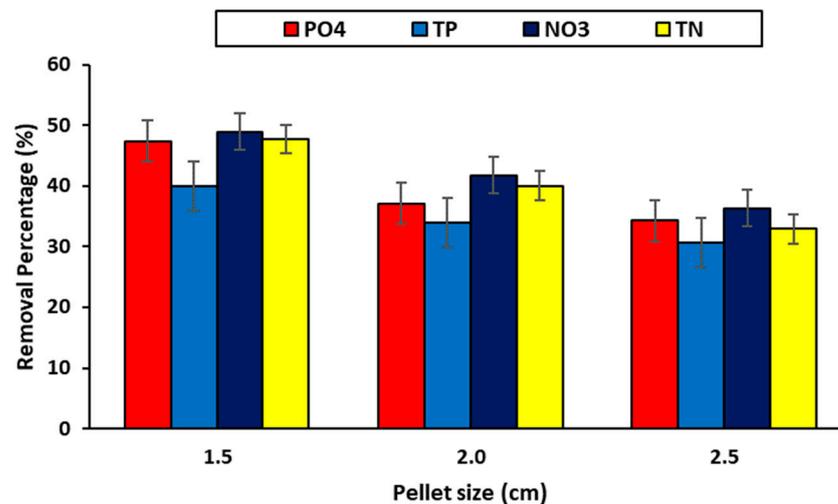
Figure 4. N and P removal rates for different pellet sizes in 35 mg/L concentration after 180 min.
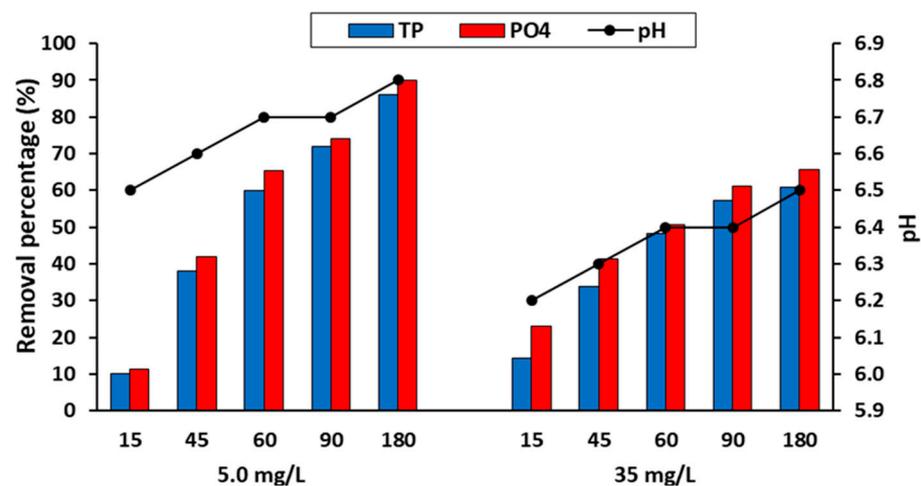
Figure 5. Removal percentages for pellets treated with P concentrations of 5 mg/L and 35 mg/L.
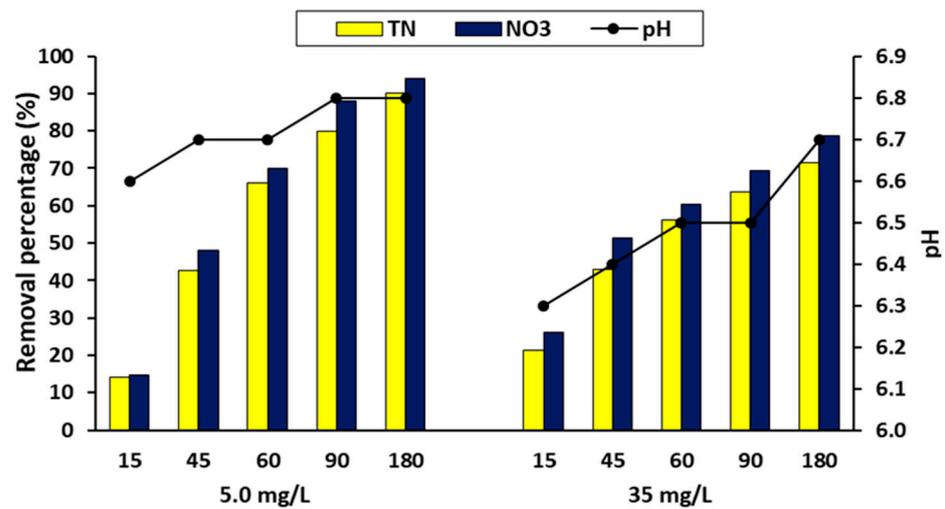
Figure 6. Removal percentages for pellets treated with N concentrations of 5 mg/L and 35 mg/L.

Table 1. Removal efficiency of optimized ZWC pellets.

Parameter	Q_{max} (mg/g)	Removal Percentage (%) for Wastewater Concentration	
		5 mg/L	**35 mg/L**
PO_4	0.112	90	65.7
TP	0.083	86	60.9
NO_3	0.171	94	78.6
TN	0.151	90	71.4

Interestingly, pH values also showed an increasing trend together with the N and P removal rates as the contact times increased. The pH values were monitored during the experiment. A low pH level in the aqueous solution means that high concentration and high mobility of H+ ions will compete with nutrient ions for the absorption sites. Protonated adsorption sites were incapable of binding nutrient ions due to electrostatic repulsion between positively charged nutrient ions and positively charged sites.

In summary, the optimal removal performances of the ZWC pellet were obtained at a pellet size of 1.5 cm, contact time of 180 min, pH of 6.8, and wastewater concentration of 5 mg L^{-1}. The optimized ZWC pellet was able to remove up to 90%, 86%, 94%, 90%, 84%, 86%, and 91% for PO_4, TP, NO_3, and TN, respectively. The maximum adsorption capacities of the ZWC pellet are 0.112 mg/g, 0.08 mg/g, 0.171 mg/g, and 0.151 mg/g for phosphate (PO_4), total phosphorus (TP), nitrate–nitrogen (NO_3), and total nitrogen (TN), respectively.

3.2.3. Removal Performance of Heavy Metals

The final optimized zeolite-WHA-clay pellet was further tested for its removal performance of heavy metals in the aqueous solution. Figure 7 shows the removal percentages of Zn, Cu and Pb for pellets treated with different concentrations of aqueous solutions. Based on the results, it is observed that all heavy metals showed much better removal performances in a low concentration (0.5 mg/L) compared to a high concentration (3.5 mg/L) of aqueous solution. The removal percentages of Zn, Cu, and Pb for the final optimized pellet after 180 min of contact time are 84%, 86%, and 91%, respectively. Lead exhibited a better removal rate compared to Zn and Cu in the aqueous solution of 0.5 mg/L. The efficiency of the pellet for treating Zn and Cu dropped significantly in the high concentration (3.5 mg/L) of aqueous solution. The removal percentage of Zn and Cu for pellets treated in 3.5 mg/L of aqueous solution only reached 29% and 6.9%, respectively, after 180 min. These results suggested that the zeolite-WHA-clay pellet is effective in treating heavy metals in low concentrations but might not be capable of treating the aqueous samples with a high concentration of heavy metal. A similar finding was also reported by [22] whereby

lower initial concentrations of the metal in the solution would lead to the highest removal efficiency of chromium.
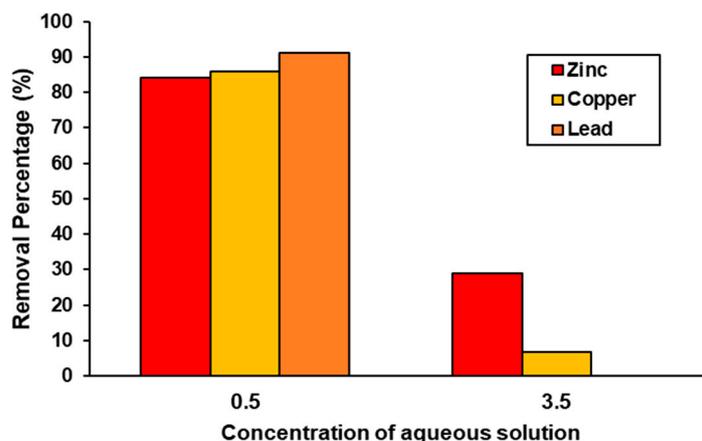
Figure 7. Removal percentages of heavy metals for pellets treated in different concentrations of aqueous solutions.

4. Conclusions

This study aimed to determine the efficiencies of nutrient and heavy metals removal performances by clay-based pellets consisting of different compositions of zeolite and water hyacinth ash (WHA). The batch adsorption experiment results showed that the zeolite-WHA-clay pellet had the best nitrogen and phosphorus removal efficiencies compared to other pellets. It was found that the combination of zeolite and WHA for clay pellet development performed better than the mono-mixture of clay pellets. The zeolite-WHA-clay pellets were enhanced in pellet size and pre-treatment with calcium hydroxide solution to optimize their removal performances. The findings revealed that the ZWC pellet demonstrated maximum removal capacities of 0.112 mg/g, 0.08 mg/g, 0.171 mg/g, and 0.151 mg/g for phosphate (PO_4), total phosphorus (TP), nitrate–nitrogen (NO_3), and total nitrogen (TN), respectively. The optimization experiment indicated that smaller pellet sizes and treatment with calcium hydroxide solution resulted in improved removal performances for nitrogen and phosphorus as well as heavy metals (HM). The optimized ZWC pellet exhibited remarkable removal efficiency, reaching up to 90%, 86%, 94%, 90%, 84%, 86%, and 91% for PO_4, TP, NO_3, TN, zinc (Zn), copper (Cu), and lead (Pb), respectively, within a contact time of 3 h. These results indicate the effectiveness of the ZWC pellet as a low-cost and efficient adsorbent for wastewater treatment. In the future, further experiments are suggested for consecutive input of various influent concentrations to determine the maximum adsorption capacity of the zeolite-WHA-clay pellet. Life-cycle analysis is also recommended for the zeolite-WHA-clay-based pellet to ensure the sustainable development of this product.

Conflicts of Interest: The authors declare no conflict of interest.

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