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Enhancement of the Reduction and Adsorption Mechanism of nZVI using an effective polyacrylamide as a nonionic polymer

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Abstract: Nano Scale Zero Valent Iron (nZVI) has been considered as an effective material for contaminated water cleanup due to the large surface area and its high reducing power. However, due to its high magnetic force, the particles tend to form aggregates resulted in decreasing its high surface area. To overcome the aggregates problem and increase the particle's surface area, polyacrylamide has been used as an effective polymer for synthesizing a stabilized nZVI particles. Comparing with non-stabilized nZVI, the polyacrylamide stabilized nZVI exhibited an excellent performance for both of the nitrate reduction and phosphorus adsorption. Different polyacrylamide dosages varied between 0.2 to 2 (g/L) were investigated, a dose of 0.6 (g/L) was the optimum ratio of polyacrylamide to nZVI to achieve the highest removal efficiency for both of phosphorus and nitrate removal. The polyacrylamide stabilized nZVI improved the nitrate and phosphorus removal efficiency by 43.6 and 21.9% respectively with respect to non-stabilized nZVI. The polyacrylamide was proposed as an effective polymer for enhancing the nZVI particles reactivity to treat the nitrate and phosphorus in the contaminated water.

Keywords: Polyacrylamide; nZVI particles; Stabilization; Reactivity.

1. INTRODUCTION

Nano-scale Zero-valent iron have been widely used for water cleanup in different sites including river water, groundwater and waste water. The nZVI particle has a strong reducing power due to its core electron-donor and large surface area for rapid uptake and transformation of a huge number of water pollutants [1]. Therefore, numerous studies have found that nZVI can effectively degrade most of the water pollutants including inorganic, organic dyes and chlorinated compounds [2]. Due to the magnetic attraction and the high intensity of iron, bare nZVI tend to be agglomerated. Therefore, the bare nZVI has very limited surface area and hence low reactivity toward the water pollutants. Moreover, the nano-iron particles have stronger tendency to form aggregates with respect to micro sized particles. This has been ascribed to smaller particle size and reduced surface potential. Its likely that surface area and particle size highly depends on the synthesis method of nZVI and the possibility of applying a desorption method whether by using physical dispersion techniques or chemical stabilizers during the synthesis process. In order to create a dispersive particles and maintain the reactivity of nZVI particles a wide range of polymers has been investigated including anionic, neutral and nonionic polymers. Cho and Choi et al [3], they found that carboxyl methyl cellulose (CMC) stabilized nZVI in the pre-agglomeration approach showed improved reactivity towards TCE and decreased the mean size to 2.8 nm. He and Zhao et al [4], investigated the dechlorination of TCE and PCBs using starch stabilized Pd/nZVI and they found an improved reactivity for both of TCE and PCBs. Kanel et al [5], they followed the post agglomeration approach to prepare nZVI particles using nonionic polymers (Tween 20) and observed a high arsenic removal. The investigation of stabilized nZVI in terms of reactivity for contaminant degradation has not been extensively studied. Therefore, this study had three goals: 1. preparation of bare and

stabilized nZVI flowing the pre-agglomeration approach using nonionic polyacrylamide polymer. 2. Investigate the nitrate reduction and phosphorus adsorption behavior using both of bare and polyacrylamide nZVI. 3. To enhance the performance of nZVI particles by using an environmental friendly polymer to achieve the highest degradation for the water contaminants.

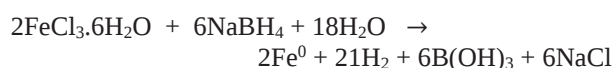
2. MATERIALS AND METHODS

2.1 Materials

Ferric chloride hexahydrate ($\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ > 99.0%, Junsei Chemical Co., Japan) and sodium borohydride (NaBH_4 > 99.0%, Junsei Chemical Co., Japan) were used as a source of iron and as a reductant respectively in order to prepare non-stabilized nZVI. PAA (Polyacrylamide, Sigma-Aldrich Co., USA) was purchased in order to stabilize the nZVI particles. Sodium nitrate (NaNO_3 > 99% Junsei Chemical Co., Japan) and potassium dihydrogen phosphate (KH_2PO_4 , 99.0% Junsei Chemical industries, Ltd, Japan) were used to prepare the nitrate and phosphorus stock solution respectively.

2.2 Synthesis of bare and PAA stabilized nZVI

The preparation of nZVI particles was carried out by following the chemical reduction approach according to the following chemical equation.



Briefly, 1.1 g of sodium borohydride was dissolved in 50 mL deoxygenated deionized water which was pumped with flow rate 20 mL/min into 500 mL four-neck open flask containing 2.5g ferric chloride dissolved in 100mL deoxygenated deionized water and the synthesis temperature was set at 30 °C. Stirring was continued 5 min after the full injection of borohydride as an aging time. Afterwards, the produced particles were collected and dried using vacuum-filter. In case of synthesis PAA stabilized nZVI, the ferric chloride was mixed for 30 min

with different concentration of PAA varied between 0.2 to 2 g/L prior to adding the borohydride solution.

2.3 Assessment of nZVI reactivity

A batch experiments using bare and PAA stabilized nZVI were performed, to evaluate their reactivity on the nitrate and phosphorus removal. The batch experiments were carried out using a 300 mL conical flask which filled with 200 mL of nitrate solution (300 mg NO_3^-/L) and 200 mL of phosphorus solution (100 mg P/L) separately. A dose of 0.25 g of bare and 0.25 g of PAA stabilized nZVI prepared under different PAA concentration were added into the nitrate and phosphorus solution separately. Nitrate and phosphorus solution were purged with nitrogen gas before adding the bare and PAA stabilized nZVI dosage. Afterwards, the solutions were placed on the magnetic stirrer at the ambient temperature with shaking speed 1000 rpm. At different times, nitrate and phosphorus sample were taken and filtered through a 0.22 μm membrane filter and then measured using UV-Vis spectrophotometer.

3. RESULTS AND DISCUSSION

3.1 Nitrate reduction using bare and PAA/nZVI

The nitrate reduction was implement by using bare nZVI which was prepared based on the optimization results for the synthesis process in our previous study [6]. Additionally, the nitrate reduction was carried out using a PAA/nZVI which prepared using different PAA concentration varied between 0.2 to 2 g/L. As indicated in Fig.1, for the bare nZVI, the maximum nitrate reduction only reached 133 mg/L at 2 hours which means 55% of the nitrate reduction was achieved. On the other hand, nitrate reduction was improved by using PAA stabilized nZVI and the highest reduction of nitrate reached 2.3 mg/L at a dose of PAA 0.6 g/L which means 99 % of nitrate reduction was achieved. The improved reactivity is ascribed to the contribution of PAA to prevent the aggregation during the synthesis which resulted in decreasing the particle size and hence improving the reactivity. However, at a concentration of 1 and 2 g/L of PAA added, a lower reduction rate was observed and its attributed to the occupation of particle surface at that concentration.

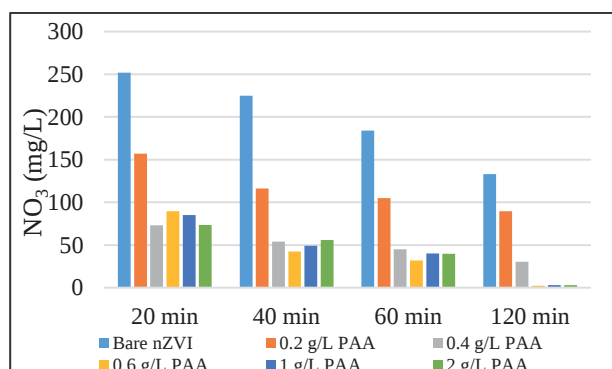


Fig.1. shows the nitrate reduction using bare and PAA stabilized nZVI at different PAA concentrations.

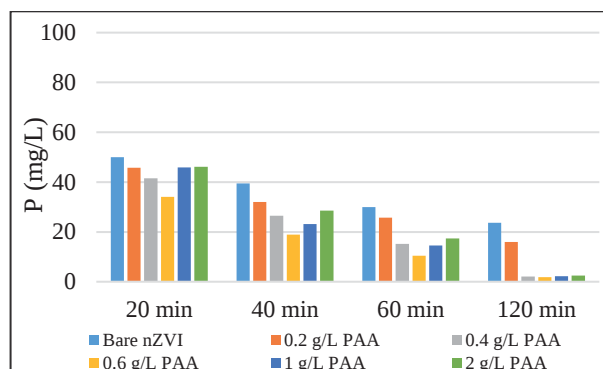


Fig.2. shows the phosphorus adsorption using bare and PAA stabilized nZVI at different PAA concentrations.

3.2 Phosphorus adsorption using bare and PAA/nZVI

A six experiments were conducted using bare and PAA stabilized nZVI to investigate the phosphorus adsorption behavior. Preliminary experiments indicated that phosphorus adsorption greatly enhanced by employing the PAA stabilized nZVI with respect to bare nZVI as it shown in Fig.2. However, a high concentration (1 and 2 g/L) of PAA resulted in slightly decreasing the adsorption rate of phosphorus. The removal percentage of phosphorus at different PAA concentration over a period of 120 min were 70, 89, 99.2, 99 and 99% for PAA concentration of 0.2, 0.4, 0.6, 1 and 2 g/L respectively. The improved reactivity of PAA stabilized nZVI is attributed to decreasing the agglomeration possibility of nZVI particles which resulted in decreasing the particle size and increasing the surface area which in turn leads to improve the adsorption capacity for phosphorus.

4. CONCLUSIONS

The behavior of nitrate reduction and phosphorus adsorption were investigated by preparing bare and PAA stabilized nZVI. PAA stabilized nZVI largely enhanced the nitrate reduction and phosphorus adsorption by 43.6% and 21.9 % respectively with respect to bare nZVI. Among varied dosages of PAA, a dose of 0.6 g/L was the best to achieve the highest removal efficiency for both of nitrate and phosphorus removal.

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