Promoting the Reactivity of Nano-scale Zero-valent Iron for Water Treatment: Mechanisms and Application

アハマド, モハマド, エルサイエド, アハマド, アリ, カリル

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氏　　名　:　Ahmed Mohamed Elsayed Ahmed Aly Khalil
Name　(アハマド モハマド エルサイエド アハマド アリ カリル)

論 文 名　:　Promoting the Reactivity of Nano-scale Zero-valent Iron for Water Treatment: Mechanisms and Application
Title　(水処理におけるナノスケール 0 価鉄の反応促進：メカニズムと応用)

Abstract of Thesis

Nano-scale zero-valent iron (nZVI) has been the research spotlight for numerous researches in water treatment and environmental applications over the last decade. It has proved its high pollutant removal efficiency and reactivity to treat several contaminants via diverse mechanisms. Owing to its unique properties such as excellent magnetic properties, surface modifiability, extremely small size and high surface area to volume ratio, nZVI is the emerging technology for water purification. However, the search for methods to improve the removal efficiencies of contaminants and their kinetic reduction rates by nZVI is unstoppable, especially in order to decrease the dosage amount required for decontamination. In addition, nZVI has some concerns about its reliability in environmental applications due to its limited mobility and fast agglomeration. Modifying nZVI surface and supporting nZVI particles on a carrier were the proposed solutions to boost the reactivity and solve the aforementioned issues. The nZVI-based reagents were synthesized and then characterized using a variety of analytical equipment, such as transmission electron microscopy, surface characterization analyzer, X-ray diffraction and laser diffraction particle size analyzer, then applied in different water treatment experiments to examine their performances. Nitrate and phosphate were chosen to be the under-investigated contaminants of this study to represent the enhanced mechanisms of removal on applying modifications, which are common mechanisms among numerous contaminants treated by nZVI. Their removal performance was carefully investigated via several batch experiments at various copper ions addition/loading ratios, different conditions of pH, presence/absence of oxygen, distinctive pollutant concentrations and so forth. Optimum addition ratio of $CuCl_2/Fe^0$ was collected from experimental results, which was used to conduct the rest of batch experiments.

The presence of copper ions during nitrate reduction imposes two electrochemical reactions; one stimulates iron corrosion and another reaction causes hydrogen-catalytic reduction of nitrate. Both reactions boosted removal efficiency and kinetics around 3.5 times more than that by ordinary pristine nano-Fe^0 alone, i.e. nitrate removal time was greatly reduced. In case of phosphate, the presence of copper chloride effectively enhanced the adsorption capacity of phosphorus as it produced copper ferrite spinel on nZVI particles' surface, which can adsorb phosphorus and increase its rate of adsorption, and also it stimulated nZVI corrosion. The adsorption capacity of phosphorus in case of nZVI in the presence of copper chloride was greater by ca. 80% than that of nZVI without copper chloride. This modification greatly boosted reaction kinetics and removal efficiency, and decreased the dependency of remediation on pH of medium and dissolved oxygen presence.

Among 25 composites of synthesized nZVI on activated carbon (AC) support under different preparation and treatment conditions, thermally-treated granular AC-supported nZVI was selected at optimum nZVI/AC mass ratio and treatment conditions. This study introduced thermal treatment of AC before supporting nZVI, which modified its textural and surface chemistry properties to attract contaminant anions with a higher affinity towards nZVI. The novel composite succeeded in increasing the removal efficiency of nitrate by 50% and of phosphate by 100% from their aqueous solutions and of nitrate by 170% along with a complete removal of phosphate from their solution. Regarding the contaminants, their removal mechanisms were carefully inspected, and their Interference studies with the treatment efficiency were conducted including the investigation of interference of domestic wastewater, humic acid, anions of sulfate and phosphate, cations of cuprous and cupric, and calcium carbonate (hardness). Results showed the significant negative impact of interference of most substances (except copper ions) on the nitrate reduction and the superiority of optimized nFe(0)/AC by ca. 27% to ca. 183% increase in treatment efficiency over nZVI. All removal profiles of most batch experiments were described by kinetic formulation models that fitted

experimental data with high accuracy and precision.

Concerning material conservation, a regeneration of nZVI materials and recovery of phosphate contaminant were successfully achieved. The regenerated nZVI, recovered from spent synthesized nanoparticles, regained full and complete removal performance.

Finally, the nZVI-based reagents were implemented in a developed application of a laboratory-scale continuous flow system (LSCFS) in order to test their performances and discover the challenges that can face actual operations. The results showed another improvement in removal efficiency and convenience in operation. To sum it up, the novel composite of combined modified copper ions addition and heat modified AC-supported nZVI can be a suitable reagent and a versatile material for applications in water treatment systems.