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）

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論文内容の要旨
Thesis Summary

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. When adding an optimized amount of copper salt to nZVI during a remediation process of eutrophication-causing pollutants (nitrate and phosphate), electrochemical reactions occur one after another, which greatly boost reaction kinetics and removal efficiency, and decrease the dependency of remediation on pH of medium and dissolved oxygen presence. Supporting nZVI particles on thermally-treated granular activated carbon prevented agglomeration of nZVI particles and increased their mobility, stability and reactivity. This research carried out numerous experiments to acquire optimum conditions required to produce the best-modified and supported nZVI. The nZVI-based reagents were characterized using a variety of analytical equipment such as transmission electron microscopy, surface characterization analyzer, X-ray diffraction and particle size analyzer then applied in different experiments to examine their performances. Regarding the contaminants, their removal mechanisms were carefully inspected, and their Interference studies were conducted including the investigation of contaminants interference of domestic wastewater, humic acid, anions of sulfate and phosphate, cations of cuprous and cupric, and calcium carbonate (hardness) with the treatment efficiency. Concerning material conservation, a regeneration of nZVI materials and recovery of phosphate contaminants were successfully achieved. 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. All removal profiles were described by kinetic formulation models that fitted experimental data with high accuracy and precision.

The thesis has a framework composed of seven chapters that explains the way of promoting the reactivity of

nZVI particles towards a suggested composite, backed up with a scientific background and theory related to nZVI, synthesis, characterizations, analyses, batch experiments, mechanisms, modeling, interference studies, recovery and regeneration, and practical application. Hence, the framework was organized as follows:

Chapter 1 gives information about the current situation of water pollution problems, the nature of contaminants investigated in this research, informative overview that encompasses nanotechnology in general and specifically in water treatment, and the role of nano-scale zero-valent iron (nZVI) in decontaminating a wide range of pollutants. The literature survey on nZVI covers most of its aspects especially synthesis techniques, modifications, treatment implementation and its environmental impact. Then the chapter identifies the goals of this research.

Chapter 2 presents the common materials and procedures performed prior and post conducting batch experiments of this research involving chemicals preparation, synthesizing method of pristine nZVI, characterization of properties of produced nanomaterials and analytical inspections. Other non-common chemicals and steps are covered later in other chapters when needed.

Chapter 3 introduces the first suggested modification employed to nZVI surface during the remediation of eutrophication causing contaminants, nitrate and phosphorus, separately. The applied technique is a special one involving the addition of a certain contaminant of copper salt, which causes coating of nZVI surface rapidly throughout treatment. Batch experiments were carried out in different scenarios (conditions of pH, dissolved oxygen, higher contaminants concentrations and addition ratios of copper ions). The mechanisms of nitrate and phosphorus removal in addition to the promotion of nZVI reactivities were deeply discussed and explained. Kinetic models were proposed to describe reaction rates. This chapter also provides a technique for the recovery of phosphorus contaminant.

Chapter 4 suggests another modification that can increase both reactivity and mobility of nZVI particles. Different composites are produced of activated carbon supported nZVI under different synthesis and treatment conditions. The chapter shows the way of selecting the optimized composite from batch experimental studies against aqueous solutions of nitrate, phosphate and mixture of these contaminants, then discovers its properties and tests its performance in interference studies comprising organic matters and group of common ions at different compositions. The results of these tests are explained, compared to that of nZVI, plotted in well-fitted kinetic models that takes into account the passivation of nZVI due to interferences.

Chapter 5 presents the essential steps of handling a material used in a remediation process, the regeneration and recycle. The chapter manages to compare between different types of nZVIs. It shows the lack of stability of nZVI particles upon ordinary storage represented in an aged old-purchased nZVI, the effect of controlled acid treatment on this aged type of nZVI, and the potent influence of the regeneration process on a spent nZVI extracted from nitrate batch experiments with fresh prepared type. This presentation is demonstrated by methods of characterizations and batch experiments to compare performances of removal.

Chapter 6 displays the method of developing the application of nZVI reagent in a laboratory-scale continuous-flow system. It describes the importance of this procedure before application on a large scale as it depicts the contemporary issues related to the remediation of a certain contaminant (nitrate) by a specific nZVI reagent in a type of design at certain operational conditions. Process and equipment designs are illustrated and performances are reported.

Finally, Chapter 7 lists the major findings included in this thesis and the anticipated future work. One of recommendations is suggested based on these findings that the combination of both modifications of copper salt addition and supporting nZVI on heat-modified activated carbon (AC) at optimum loading ratios (0.1 wt $CuCl_2$/wt Fe^0 and nZVI/AC mass ratio of 2:1) and treatment conditions (of AC at 950 °C for 2 h) forms a promising material that suits groundwater and wastewater treatment systems.