Improvement of Iron Nanoparticles Reactive Performance via Coating Modification towards Contaminants Removal from Water

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論 文 内 容 の 要 旨
Thesis Summary

Water pollution is currently the main concern all over the world in whether developed or developing countries, especially with the continued population inflation which accompanied by the continuously increasing water demand. Meanwhile, nanotechnology has been emerged recently in several environmental remediation aspects owing to the excellent functional characteristics of the nanomaterials. The behavior of zero-valent iron nanoparticles (Fe^0) in water treatment from pollutants was comprehensively examined along the recent years. Fe^0 has the ability to react with most of the soluble contaminants due to its particular core-shell structure in addition to the dual redox potential. Furthermore, Fe^0 performance as an efficient adsorbent is highly comparable to other materials because of the relatively large surface/volume ratio of the nanoparticles. However, still more investigation is needed to overcome the drawbacks of Fe^0 especially in the real water treatment applications.

In this work, nanotechnology with innovative coating techniques was utilized for the removal of different contaminants (i.e., phosphorus, nitrate, and hexavalent chromium (Cr(VI)) from aqueous solutions. Three different metal hydroxides (i.e., magnesium, aluminum, and calcium hydroxides ($Mg/Al/Ca(OH)_N$)) were considered for the coating of iron nanoparticles (Fe^0) to enhance the aqueous characteristics of the coated particles (C-Fe^0) within different aspects, including transportability within porous media, aqueous suspension stability, reactivity release, crystallographic and morphological characteristics. The findings showed great superiority of magnesium hydroxide ($Mg(OH)_2$) as a non-toxic coating material for Fe^0, comparing to other investigated metal hydroxides. Magnesium hydroxide coated iron nanoparticles (MgC-Fe^0), comparing to that of bare Fe^0, showed 95% suspension enhancement, 5 times increased mobility in porous media, 47% finer size, 48% higher crystallinity, and improved anti-aggregation effect due to the non-magnetic coating. The Multi-functional effect of $Mg(OH)_2$ shell was represented by the progressive shell-dissolution in water and preventing the rapid corrosion of Fe^0-core, which resulted in a controlled release of Fe^0 reactivity towards Cr(VI). Synergetic effect of MgC-Fe^0 was achieved via the electrostatic attraction of Cr(VI) species onto the positively charged surface of $Mg(OH)_2$ shell, and selective co-precipitation towards Cr(III) species. MgC-Fe^0 showed great performance in preserving the long-term reactivity of Fe^0 within a wide range of pH and temperature, indicating its advantage in the real water treatment applications. The long-term investigation of the MgC-Fe^0 performance towards Cr(VI) removal confirmed the continuous release of Fe^0 reactivity to reach achieve 100% removal efficiency over 50 days of reaction, to be reported for the first time in the literature. The injection of MgC-Fe^0 into the 3-D lab-scale groundwater treatment system for 30 consecutive cycles resulted in a clear enhancement in preventing the rapid corrosion of the iron core and around 20% improvement in the final Cr(VI) removal efficiency comparing with that of Fe^0. The promising potential of the proposed MgC-Fe^0, provides a great contribution to the in-situ water treatment field as a reactive nanomaterial with enhanced features, to be a perfect candidate for the nanotechnology applications in water treatment.

This dissertation has a framework composed of six chapters that investigating improving the characteristics and the reactive performance of iron nanoparticles (Fe^0) via environmentally friendly coating techniques, backed up with a scientific background and theories related to Fe^0, synthesis, characterizations, analyses, batch experiments, mechanisms, modeling, 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 Fe^0 in decontaminating a wide range of pollutants. The literature survey on Fe^0 covering most of its aspects especially synthesis techniques, modifications, treatment implementation and its environmental impact. Detailed survey on the applications of Fe^0 based materials in groundwater treatment applications is also provided. Generally, the chapter identifies the goals of this research.

Chapter 2 presents the common chemicals, materials and procedures performed prior and post conducting batch and bench-scale experiments of this research involving chemicals preparation, synthesizing method of pristine nanomaterials, characterization of properties of the produced nanomaterials and analytical inspections.

Chapter 3 focuses on developing three types of coated iron nanoparticles (C-Fe^0), using three different layered hydroxide coatings ($Mg/Al/Ca(OH)_N$) with different coating ratios. Thus, various features of Fe^0 and C-Fe^0, including aqueous suspension stability, transportability, and reactivity, are investigated through different sets of experiments. Moreover, surface, and crystalline properties of the synthesized materials are examined considering different characterization techniques. Detailed interpretation of the obtained results is presented in the light of the correlative relations between the structural and morphological peculiarities as well as the reactive performance towards nutrients removal. Hence, this study represents a crucial contribution in terms of integrating such modified nanoparticles in the real water treatment applications.

Chapter 4 introduces the results of investigating the reactive performance of the promising magnesium coated iron nanoparticles (MgC-Fe^0) towards the removal of hexavalent chromium (Cr(VI)) from aqueous solutions. The short- and long-term controlled release of the coated nanoparticles' reactivity is evaluated through several batch tests and using several analytical and characterization techniques. The results are thoroughly explained with deep insights into the effect of different reaction factors on Cr(VI) removal by MgC-Fe^0. The findings of this chapter represent a great contribution to the field of water treatment applications, in terms of proposing a high-potential reactive material for long-term groundwater remediation.

Chapter 5 reviews all the obtained results of the injection of iron nanoparticles (Fe^0), and magnesium hydroxide-coated iron nanoparticles (MgC-Fe^0) into porous media, within 3-D bench-scale aquifer treatment system for the removal Cr(VI) from contaminated water.

Finally, **Chapter 6** lists the major findings and conclusions included in this thesis and the anticipated future research work