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Environmental transformations and ecological effects of iron-based nanoparticles[☆]

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ABSTRACT

The increasing application of iron-based nanoparticles (NPs), especially high concentrations of zero-valent iron nanoparticles (nZVI), has raised concerns regarding their environmental behavior and potential ecological effects. In the environment, iron-based NPs undergo physical, chemical, and/or biological transformations as influenced by environmental factors such as pH, ions, dissolved oxygen, natural organic matter (NOM), and biotas. This review presents recent research advances on environmental transformations of iron-based NPs, and articulates their relationships with the observed toxicities. The type and extent of physical, chemical, and biological transformations, including aggregation, oxidation, and bio-reduction, depend on the properties of NPs and the receiving environment. Toxicities of iron-based NPs to bacteria, algae, fish, and plants are increasingly observed, which are evaluated with a particular focus on the underlying mechanisms. The toxicity of iron-based NPs is a function of their properties, tolerance of test organisms, and environmental conditions. Oxidative stress induced by reactive oxygen species is considered as the primary toxic mechanism of iron-based NPs. Factors influencing the toxicity of iron-based NPs are addressed and environmental transformations play a significant role, for example, surface oxidation or coating by NOM generally lowers the toxicity of nZVI. Research gaps and future directions are suggested with an aim to boost concerted research efforts on environmental transformations and toxicity of iron-based NPs, e.g., toxicity studies of transformed NPs in field, expansion of toxicity endpoints, and roles of laden contaminants and surface coating. This review will enhance our understanding of potential risks of iron-based NPs and proper uses of environmentally benign NPs.

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1. Introduction

With the development of nanotechnology, iron-based nanoparticles (NPs) are increasingly produced and used due to their superior physicochemical properties compared with bulk particles. Zero-valent iron nanoparticles (nZVI) have proved effective in the treatment of a variety of contaminants, especially for chlorinated solvents and metals/metalloids, leading to their prevalent application for *in situ* soil and groundwater remediation (Crane and Scott, 2012; Grieger et al., 2010; Yan et al., 2013). The

distinguished superparamagnetism property of magnetic iron oxide NPs such as γ -Fe₂O₃ and Fe₃O₄ permits their biomedical applications, such as magnetic resonance imaging, drug delivery, and cell labeling (Liu et al., 2013a). Iron oxide NPs are also used in water treatment because of their adsorption capacity (Hua et al., 2012). Various strategies have been adopted to improve the application potential of iron-based NPs, such as surface modification, protective shells, solid supports, and doping of a second metal (Crane and Scott, 2012; Stefaniuk et al., 2016). The wide applications of iron-based NPs result in their increasing release to the environment. It is worth noting that at least 3 g/L of nZVI is needed for successful field scale application, e.g., nZVI slurries with a concentration of 10 g/L was injected into a trichloroethylene (TCE) and trichloroethane contaminated site (Grieger et al., 2010; Karn et al., 2009; Phenrat et al., 2009c). As a result, the fate and potential impacts of iron-based NPs in the environment have aroused increasing

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attention (Lefevre et al., 2016; Mahmoudi et al., 2011a).

Lots of studies have emphasized the importance to understand transformations of nanomaterials following their release to the environment, such as aggregation, dissolution, redox reactions, and interactions with macromolecules, which ultimately alter their fate, transport and potential toxicity (Dwivedi and Ma, 2014; Lin et al., 2010; Lowry et al., 2012; Mitrano et al., 2015). The potential transformations of iron-based NPs are shown in Fig. 1. Of all transformations, aggregation is the most important and could decide whether the nanomaterials behave as particulates or aggregates (Hotze et al., 2010). Similar to other nanomaterials, bare iron-based NPs aggregate into micrometer-size clusters due to their high surface energy, especially in the presence of strong magnetic interactions for magnetic NPs (Kharisov et al., 2012; Stefaniuk et al., 2016; Yan et al., 2013). Engineered surface coating and naturally occurring coating (e.g., natural organic matter (NOM) and proteins) in the environment could effectively prevent the aggregation of iron-based NPs (Phenrat et al., 2008; Tombácz et al., 2013; Sheng et al., 2016). In addition to the potential reactions on surface coating (Fig. 1c), the complexity of NOM and the interactions between iron-based NPs and natural colloids (e.g., clays and microorganisms) further make it difficult to predict their aggregation behavior in the environment (Jung et al., 2014; Li et al., 2016; Philippe and Schaumann, 2014; Wang et al., 2015).

As a redox-active nanomaterial, nZVI undergoes continuous structure and composition evolutions in the environment, i.e., aging, with the depletion of Fe^0 and formation of iron oxide shell (Fig. 1b), which is also considered as the most important transformation of nZVI (Dong et al., 2016b; Pullin et al., 2017b; Wang et al., 2010). In recent years, extensive efforts have been paid to elucidate the aging process and influencing factors (e.g., iron types, solution composition and coexisting contaminants), in view of their profound effects on the reactivity and toxicity of nZVI (Kim et al., 2012b; Reinsch et al., 2010; Liu et al., 2015; Xie and Cwiertny, 2012).

After release, iron-based NPs could also interact with various living organisms in the environment. On one hand, iron-based NPs might experience biologically mediated transformations, such as redox reactions (Fig. 1d), thereby altering their composition and behavior in the environment (Kirschling et al., 2011; Weber et al., 2006; Yan et al., 2008). On the other hand, the presence of iron-based NPs (both pristine and transformed) in the environment may impose threats to living organisms. Though iron is classified as non-toxic, increasing studies indicate negative impacts of iron-based NPs on living organisms, from DNA level to the whole

organism, where reactive oxygen species (ROS) generated by the NPs have been considered as the primary toxic mechanism (Liu et al., 2013a; Mahmoudi et al., 2011a; Stefaniuk et al., 2016). Other potential toxic mechanisms include metal ion release, cellular uptake, and physical interaction (Chen et al., 2012a; Keenan et al., 2009; Lewinski et al., 2008; Xie et al., 2017). However, contradictory toxicity results were reported by different studies due to inconsistent experimental conditions and the influence of transformations of iron-based NPs that would alter their toxicity.

Therefore, this review analyzed physical, chemical, and biological transformations of iron-based NPs in the environment, addressed toxic impacts of iron-based NPs on microorganisms, animals, and plants, discussed the potential effects of environmental transformation of iron-based NPs on their toxicity, and put forward future research directions. We believe that it will promote a proper use of iron-based NPs and better understanding of the associated risks.

2. Transformation of iron-based nanoparticles in the environment

2.1. Physical transformation

2.1.1. Challenges to the derjaguin-landau-verwey-overbeek theory

Following the release to the environment, aggregation of NPs with themselves (homoaggregation) or with natural minerals and organic colloids (heteroaggregation) occurs spontaneously, which would reduce available surface area of the NPs and influence their mobility, reactivity, and potential toxicity in the environment (Batley et al., 2013; Dwivedi et al., 2015). The Derjaguin-Landau-Verwey-Overbeek (DLVO) theory is commonly applied to predict the aggregation of NPs in the aqueous environment, which involves van der Waals forces and electrostatic repulsive forces as influenced by the size, Hamaker constant, and surface charge of the NPs and ionic strength (IS) of the solution (Dwivedi et al., 2015; Hotze et al., 2010). According to the DLVO theory, the initial NPs concentration and ambient environment conditions (pH, IS) play important roles in the aggregation of NPs. In general, increasing NPs concentration results in a greater number of particle-particle collisions and an increased level of aggregation (Dwivedi et al., 2015). Solution pH regulates surface charge of the NPs; the further the pH is away from the point-of-zero charge (PZC) of the NPs, the farther the surface charge is away from zero, favoring aqueous stability of the NPs due to the promoted electrostatic repulsion (Garner and Keller, 2014).

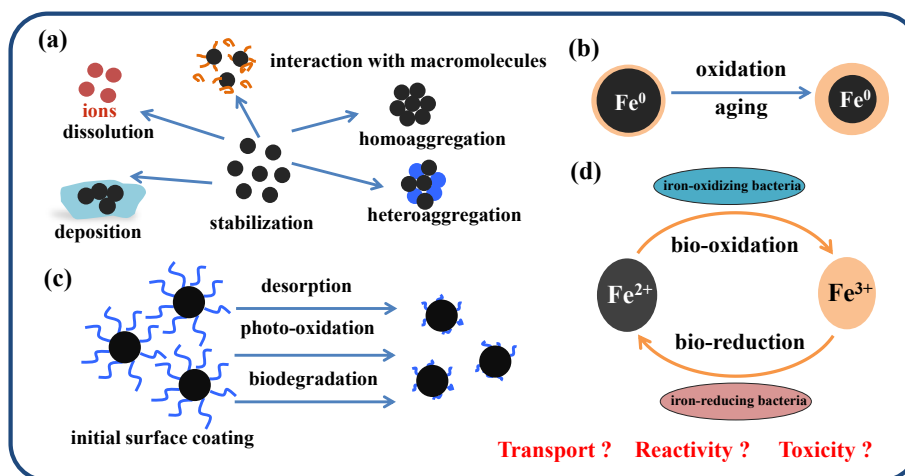


Fig. 1. Potential transformations of iron-based NPs in the environment.

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