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Review Article

Reactive oxygen species-related activities of nano-iron metal and nano-iron oxides[☆]



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ABSTRACT

Nano-iron metal and nano-iron oxides are among the most widely used engineered and naturally occurring nanostructures, and the increasing incidence of biological exposure to these nanostructures has raised concerns about their biotoxicity. Reactive oxygen species (ROS)-induced oxidative stress is one of the most accepted toxic mechanisms and, in the past decades, considerable efforts have been made to investigate the ROS-related activities of iron nanostructures. In this review, we summarize activities of nano-iron metal and nano-iron oxides in ROS-related redox processes, addressing in detail the known homogeneous and heterogeneous redox mechanisms involved in these processes, intrinsic ROS-related properties of iron nanostructures (chemical composition, particle size, and crystalline phase), and ROS-related bio-microenvironmental factors, including physiological pH and buffers, biogenic reducing agents, and other organic substances.

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1. Introduction

Reactive oxygen species (ROS), resulting from the transfer of energy or electrons to oxygen, are highly reactive and potentially harmful to living organisms [1]. These ROS, such as singlet oxygen, superoxide, hydrogen peroxide, and hydroxyl radical, are essential intermediates in certain physiological processes (e.g., photosynthesis, respiration, and cell

signaling), and their levels within cells are tightly controlled via enzymes (e.g., superoxide dismutase, glutathione peroxidase, and catalase) or antioxidants (e.g., ascorbic acid, cysteine, glutathione, bilirubin, carotenoids, and bilirubin). However, this redox homeostasis can be perturbed in many circumstances, and a burst of ROS, a condition termed oxidative stress, can induce deleterious effects to cells through oxidative damage of biomolecules (e.g., proteins,

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lipids, and nucleic acids) or disruption in cell signaling mechanisms [2].

Advances in nanotechnology, which deals with materials with dimensions of the order of 100 nm or less, have resulted in increasing commercial applications of engineered nanoparticles in consumer products, such as electronics, medicines, dietary supplements, food, clothing, cosmetics, and goods for children. The use of nanomaterials in diverse categories of consumer products suggests that broad populations of consumers will be increasingly exposed to nanomaterials [3,4]. A large number of studies have been conducted to evaluate the biological effects of nanoparticle exposure, and ROS-induced oxidative stress has been well recognized as one of the most important mechanisms when toxicity is observed [5].

Iron, which plays an active redox-catalytic role in many energy-transfer or electron-transfer processes due to its partially filled d orbitals and variable oxidation states, is inextricably linked to ROS chemistry. Engineered nanostructures of iron, including nano-iron metal and nano-iron oxides, have attracted commercial interest in areas of medicine (e.g., intravenous iron preparations, iron supplements, magnetic resonance imaging contrasting agents, drug and gene delivery, tissue engineering, and hyperthermia), food (e.g., iron fortificants), environment (e.g., remediation of soils and water by removal of organic pollutant and heavy metals), and agriculture (e.g., plant fertilizer and animal feed). Therefore, understanding ROS-related activities of nano-iron metal and nano-iron oxides could help address concerns over increasing incidences of biological exposure to these engineered nanostructures [6–11]. There are also widespread naturally-occurring iron nanostructures in diverse parts of terrestrial and aquatic ecosystems (e.g., soils, sediments, rivers, lakes, springs, and marine) mainly in the form of iron oxides, including ferrihydrite ($\text{Fe}(\text{OH})_3 \cdot 4\text{H}_2\text{O}$), lepidocrocite ($\gamma\text{-FeOOH}$), goethite ($\alpha\text{-FeOOH}$), hematite ($\alpha\text{-Fe}_2\text{O}_3$), maghemite ($\gamma\text{-Fe}_2\text{O}_3$), and magnetite (Fe_3O_4), thus making ROS-related activities of nano-iron oxides critical in comprehending ecological roles of naturally-occurring iron nanostructures [12].

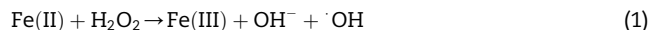
This review focuses on activities of nano-iron metal and nano-iron oxides in ROS-related redox processes. The known redox mechanisms involved in these processes are demonstrated first, followed by a description of intrinsic ROS-related properties of iron nanostructures. Finally, ROS-related bio-microenvironmental factors are also discussed.

2. ROS-related redox mechanisms of nano-iron metal and nano-iron oxides

Successive one-electron or two-electron reduction of molecular oxygen to water in the aqueous solution yields a series of ROS such as superoxide radicals ($\text{O}_2^-/\text{HO}_2^\bullet$), hydrogen peroxide (H_2O_2), and hydroxyl radicals ($\text{OH}^\bullet$). Nano-iron metal and nano-iron oxides can be involved in these redox reactions as the reactant or catalyst via a homogeneous or heterogeneous means, based on dissolved iron species or solid surfaces, respectively. The standard reduction potentials (E^0) of redox pairs covered in this review are cited from previous studies [13–15].

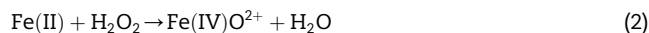
2.1. Homogeneous reactions

(1) The classic homogeneous Fenton reaction, which involves one-electron reduction of hydrogen peroxide by soluble ferrous iron species, generates hydroxyl radicals ($E^0 = +2.3 \text{ V}$) that are powerful enough to oxidize most organic molecules:



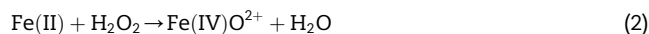
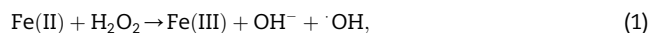
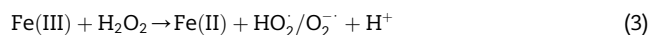
By convention, Fe(II) and Fe(III) represent all ferrous and ferric iron species, respectively.

(2) The non-radical mechanism for the homogeneous Fenton reaction involves the generation of ferryl-oxo complexes ($E^0 = +0.9 \text{ V}$), less powerful oxidants compared to hydroxyl radicals, via two-electron reduction of hydrogen peroxide with soluble ferrous iron species:

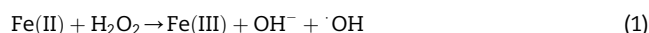
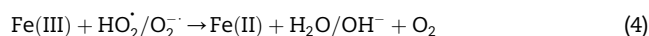


Based on the distinct oxidizing power of hydroxyl radicals and ferryl-oxo complexes, many studies have found that radical and non-radical Fenton reactions are competing reactions with a pH-dependent partitioning [16–18].

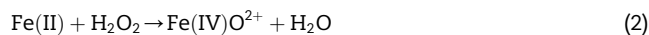
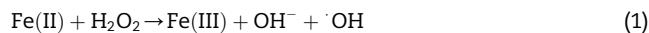
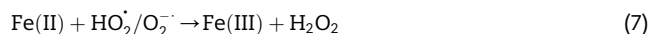
(3) The homogeneous Fenton-like reactions, which involve the generation of superoxide radicals, hydroxyl radicals, or ferryl-oxo complexes from hydrogen peroxide and soluble ferric iron species, consist of two steps; a slow one-electron reduction of ferric iron by hydrogen peroxide and a rapid generation of powerful oxidants via reaction (1) or (2):



(3) The Haber-Weiss reaction, which involves generation of hydroxyl radicals from hydrogen peroxide and superoxide, can be catalyzed by soluble ferric iron species through two steps:



(4) The homogeneous Fe(II) autoxidation in the presence of molecular oxygen in the aqueous solution via single-step two-electron transfer or stepwise one-electron transfer reactions can generate oxidants of ferryl-oxo complexes or a series of ROS [19,20]:



(5) The homogeneous auto-scavenging reactions involving evolution of molecular oxygen and oxidative loss of soluble ferrous iron species [21]:

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