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<https://doi.org/10.5109/4738583>

出版情報 : Proceedings of International Exchange and Innovation Conference on Engineering &
Sciences (IEICES). 7, pp.161-169, 2021-10-21. 九州大学大学院総合理工学府

バージョン :

権利関係 :

Mini Review on Recent Applications of Nanotechnology in Nutrient and Heavy Metals Removal from Contaminated Water

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Abstract: Several toxic substances, such as nutrients, heavy metals, radionuclides, and pharmaceuticals, are continuously discharging to the environment, as a result of unaccountable industrial and agricultural activities, and contaminating huge quantities of soil and water. Nanotechnology was employed over the past years to remediate the contaminated waters and clean the environment from these pollutants to protect the life on the planet. Nanoscale zerovalent iron (nZVI) is one of the most applied nanomaterials in the field of water and wastewater treatment and it is extensively utilized to remove a wide range of contaminants from water. Thus, this mini review intends to summarize the latest applications of nZVI and its composites in eliminating several nutrients and heavy metals, namely nitrate (NO_3^-), phosphorous (P), chromium (Cr) and arsenic (As).

Keywords: Arsenic (As); Chromium (Cr); Nanoscale zerovalent iron (nZVI); Nitrate (NO_3^-), Phosphorous (P).

1. INTRODUCTION

The irresponsible anthropogenic activities and the unwise application of agricultural chemicals (e.g. pesticides, fertilizers, herbicides, and insecticides) resulted in contaminating water and soil with toxic contaminants such as nutrients (i.e. Nitrate and phosphorous), heavy metals (e.g. Chromium, arsenic, nickel, cadmium, copper, mercury, zinc, palladium, etc.), and organic compounds [i.e. polyaromatic hydrocarbons (PAHs), perfluorinated organics (PFOs), etc.] [1,2]. Furthermore, the inadequate treatment of different types of wastewater such as domestic wastewater, industrial wastewater, nuclear wastewater, and hospital wastewater is another source for these contaminants and it can introduce another lethal contaminates, for instance, radioactive elements (i.e. Cesium, uranium, iodine, strontium, etc.), personal care products (PCPs), and pharmaceuticals (e.g. antibiotics) [3,4]. Discharge of these contaminants surely will threat all forms of life on the earth (i.e. Humans, animals, microorganisms, etc.) and negatively affect the surrounding environment, especially the aquatic environments (e.g. rivers, lakes, groundwater, etc.) [5,6]. Therefore, it is essential to develop an effective treatment technology to remove these contaminants from the limited water resources and provide a clean drinking water.

Nanotechnology is a promising approach that is getting the attention of many researchers in field of water and wastewater technologies because it has the potentials to overcome the limitations of traditional treatment technologies and it has the ability to meet the strict water regulations [7,8]. Nanoscale Zero-Valent Iron particles (nZVI) are one of the most promising nanomaterials for environmental remediation. Due to its small size, high surface area and high reducing potentials, nZVI, as a versatile material, is widely applied to remove a wide range of contaminants and pollutants [9,10]. Thus, this mini review aims to provide a recap for the state-of-the-art applications of nZVI and its composites in removing nitrate, phosphorous, chromium and arsenic from water as representative contaminants for nutrients and heavy metals.

2. NUTRIENT REMOVAL

2.1 Nitrate removal

Water contamination with nitrate (NO_3^-) causes many health and environmental complications. For instance, infant methemoglobinemia (blue baby), cancer, and liver diseases can result from drinking water contaminated with nitrate [11]. Moreover, high levels of nitrate contamination can deteriorate the quality of surface water bodies by promoting eutrophication [12]. Hence, the world health organization (WHO) defined the maximum contamination level of nitrate in drinking water as 50 mg L^{-1} (NO_3^-), whereas the Japanese authorities reduced this level to 10 mg L^{-1} (N) [13].

Table 1 summarizes the latest utilization of iron nanoparticles and its composites in removing nitrate from water. Eljamal et al. attempted to improve the performance of nZVI in removing nitrate from water by coating nZVI with polyacrylamide (PPA) and the coated nZVI (PPA-nZVI) completely removed 300 mg L^{-1} of nitrate within 2 h [14]. In addition, **table 1** shows that the nZVI-based composites prepared by Song et al. [15], Zhang et al. [16], Zhang et al. [17], Tang et al. [18], Anbia and Kamel [19], and Babaei et al. [20] succeeded to accomplish full removal of different concentrations of nitrate (20-100 mg L^{-1}), whilst the nZVI-based materials proposed by Khoshro et al. [21], Maamoun et al. [22], Diao et al. [23], He et al. [24], Mokete et al. [25], Li et al. [26], and Valiyeva et al. [27] exhibited a comparable performance with efficiencies ranges from 91.5% to 99.8%.

2.2 Phosphorus removal

Phosphorous (P) is an essential nutrient for the growth of all organisms such as plants, animals, microbes, algae, etc. [28,29]. However, similar to nitrate, excessive concentrations of phosphorus in water will escalate the phenomena of eutrophication and will negatively affect the quality of water and stress the aquatic life [30,31]. Effluents of wastewater treatment plants, landfill

leachate, fertilizers, and industrial activities are some of the sources for phosphorus contamination [32,33].

Table 2 sums up the recent articles that used nZVI and its composite to remediate phosphorus contaminated water. Eljamal et al. found that the addition of copper chloride salt (CuCl_2) stimulated the full elimination of 50 mg L^{-1} of phosphorus by nZVI [34]. Moreover, Chen et al. proved that stabilizing nZVI with starch strengthened the performance of nZVI to achieve a full uptake of 100 mg L^{-1} of phosphate (PO_4^{3-}) within an hour [35]. In addition, Marková et al. illustrated that doping a noble metal such as silver (Ag) on the surface of nZVI improved the reactivity of nZVI and the bimetallic (Fe–Ag) nanoparticles attained a 100% removal efficiency of 10 mg L^{-1} of phosphate (PO_4^{3-}) [36]. **Table 2** also demonstrates that several nZVI-based materials for instance, polyacrylamide coated nanoscale zero-valent iron particles (PPA-nZVI) [14], activated carbon supported nanoscale zero-valent iron particles (AC-nZVI) [37], magnesium hydroxide coated-nanoscale zerovalent iron nanoparticles ($\text{Fe}^0 [\text{Mg/Fe}]^{1.0}$) [38], encapsulated nanoscale zero-valent iron [39], nanoscale zero-valent iron/nickel supported on zeolite (Z-Fe/Ni) [40], sepiolite-nano zero valent iron composite [41], clay-supported nanoscale zero-valent iron [42], nanoscale zerovalent iron particles supported on spondias purpurea seed waste (S-NaOH-NZVI) [43], and nanoscale zerovalent iron particles (nZVI) derived from reducing natural goethite [44], were reported in the literature to efficiently remove high and low concentrations of phosphorus from water.

3. HEAVY METALS REMOVAL

3.1 Chromium removal

Chromium (Cr) is a heavy metal which used in a wide range of industries for example, pigments and paints, metal finishing, lumber protection, leather tanning, etc. [45]. The excessive industrial application of chromium and the inadequate treatment of industrial wastewater resulted in contaminating surface and groundwater with chromium [46]. Drinking contaminated water with chromium has lethal health issues such as cancer, disorder of nervous system, destruction of liver and kidney and impairment of male reproductive organs [47]. The WHO set the maximum allowable limit of chromium in drinking water and surface water to $50 \mu\text{g L}^{-1}$ and $100 \mu\text{g L}^{-1}$, respectively [48].

Table 3 epitomizes the most recent applications of nZVI and its composites to detoxify chromium-contaminated water. It can be seen from **table 3** that most of the applied modifications on nZVI enhanced the elimination of chromium by nZVI with removal efficiencies exceed 99%. For instance, Cheng et al. reported that CaCO_3 -coated nZVI removed 100% of 10 mg L^{-1} of hexavalent

chromium [Cr(VI)] [49]. However, Xu et al. found that chitosan-stabilized biochar supported S-nZVI (CS@BC/S-nZVI) was proficient in removing high concentrations of Cr(VI) (99.83% of 100 mg L^{-1} of Cr(VI)) [50]. Also, a similar performance was reported by Zhang et al. where more than 99% of 100 mg L^{-1} of Cr(VI) was eliminated by nanoscale zero valent iron supported by biomass-activated carbon (BC-nZVI) [51].

3.2 Arsenic removal

Arsenic (As) is a natural heavy metal that exists in earth crust. Beside its natural presence, mining activities and agricultural practices are the main sources for As contamination in the environment [52]. The high toxicity of arsenic made the WHO to consider it as one of chemicals of major public health concerns [53]. Thus, the WHO strictly set the maximum allowable limit of arsenic in drinking water to 10 ppb [54].

Some of the latest utilization of nZVI and its composites in arsenic removal from water are summarized in **table 3**. According to **table 3**, most of the fabricated nZVI-composites were efficient to remove more than 90% of different concentrations of arsenic from water. For example, stabilization with starch and carboxymethyl cellulose reinforced nZVI to achieve a complete removal of 2 mg L^{-1} of arsenate [As(V)] [55]. Likewise, supporting nZVI with activated carbon succeeded in removing 100% of 2 mg L^{-1} of As(V) [56]. On the other hand, using chitosan fiber as a supporting material for nZVI did not improve the removal of As(V) as chitosan fiber-supported zero-valent iron nanoparticles only removed 70% of 0.01 mg L^{-1} of As(V) [57].

4. CONCLUSIONS.

The objective of this mini review is to sum up the recent published articles on the application of nZVI and its composites in cleaning contaminated water with nitrate (NO_3^-), phosphorus (P), chromium (Cr) and arsenic (As). It can be noticed from this review that several improvement techniques were applied on nZVI to overcome the disadvantages of nZVI and to enhance its reactivity towards the removal of the contaminants of interest. For instance, some of the researchers used supporting materials, such as activated carbon, zeolite, multi-wall carbon nanotubes, etc. While, some scientists coated nZVI particles with polymers or dopped another metal (i.e. Copper, nickel, silver) on the surface of nZVI. The purpose of these methods is to distribute and disperse nZVI particles, reduce their aggregation and improve their performance in wastewater treatment applications. This short review concludes that nitrate, phosphorus, chromium and arsenic were efficiently removed by nZVI and its composite from water with removal efficiencies ranges from 90% to 100% under different removal conditions.

Table 1. Application of nanoscale zerovalent iron particles and its composites in nitrate removal from water.

No.	Name of the material or system	Removal conditions	Removal Efficiency (%)	Reference
1	Polyacrylamide coated nZVI particles (PPA-nZVI)	Nitrate (NO_3^-) con. = 300 mg L ⁻¹ , initial pH = 7, material dosage = 1 g L ⁻¹ , temperature = 25 °C, and contact time = 120 min.	100%	[14]
2	nZVI/activated carbon micro-electrolysis	Nitrate (NO_3^-) con. = 20 mg L ⁻¹ , initial pH = (3-7), nZVI dosage = 4 g/L, nZVI/C = 2:1, temperature = 25 °C, DO concentration <1 mg L ⁻¹ , and contact time = N/A.	100%	[15]
3	nZVI supported on pillared clay (NZVI/PILC)	Nitrate (NO_3^-) con. = 50 mg L ⁻¹ , initial pH = 7, material dosage = 0.5 g L ⁻¹ , temperature = 25 °C, and contact time = 120 min.	100%	[16]
4	nZVI supported on exfoliated graphite	Nitrate (NO_3^-) con. = 80 mg L ⁻¹ , initial pH = 6.7, material dosage = 1.67 g L ⁻¹ , Fe ⁰ loading percentage = 15%, temperature = 25 °C, and contact time = 90 min.	100%	[17]
5	Nanoscale zero-valent iron (nZVI) with copper deposited on chelating resin (D407) (Fe-Cu/D407)	Nitrate (NO_3^-) con. = 50 mg L ⁻¹ , initial pH = 3, Cu-Fe ratio = 1:2, material dosage = 2.5 g L ⁻¹ , temperature = 40 °C, and contact time = 120 min.	100%	[18]
6	Pyramids structured silicon (PSi) with nano zero valent iron (NZVI) (NZVI/PSi)	Nitrate (NO_3^-) con. = 100 mg L ⁻¹ , initial pH = 3, material dosage = 0.25 g L ⁻¹ , temperature = N/A, and contact time = 140 min.	100%	[19]
7	Multi-wall carbon nanotubes (MWCNTs) supported nano-zero valent iron (nZVI) particles (nZVI@MWCNTs)	Nitrate (NO_3^-) con. = 50 mg L ⁻¹ , initial pH = 3, material dosage = 1 g L ⁻¹ , temperature = 25 °C, and contact time = 240 min.	100%	[20]
8	nZVI-reduced graphene oxide composite (nZVI-rGO)	Nitrate (NO_3^-) con. = 64 mg L ⁻¹ , initial pH = 7, material dosage = 2.5 g L ⁻¹ , and contact time = 1 min.	96%	[21]
9	Coated-Fe ⁰ [Mg/Fe] ^{1.0}	Nitrate (NO_3^-) con. = 200 mg L ⁻¹ , initial pH = 7, material dosage = 1 g L ⁻¹ , temperature = 25 °C, and contact time = 180 min.	92.4%	[22]
10	Acid mine drainage-based nZVI (AMD-based nZVI)	Nitrate (NO_3^-) con. = 50 mg L ⁻¹ , initial pH = 4.02, material dosage = 3 g L ⁻¹ , power of ultrasonic irradiation = 40 W, temperature = N/A, and contact time = 120 min.	95%	[23]
11	Zeolite-supported nanoscale zero-valent iron/palladium (Z-Fe/Pd)	Nitrate (NO_3^-) con. = 20 mg L ⁻¹ , initial pH = 3, material dosage = 0.5 g L ⁻¹ , temperature = 25 °C, and contact time = 6 h.	91.5%	[24]
12	Bimetallic silver/nanoscale zerovalent iron (FeAg)	Nitrate (NO_3^-) con. = 200 mg L ⁻¹ , initial pH = 3, material dosage = 0.5 g L ⁻¹ , temperature = 25 °C, and contact time = 3 h.	99.5 %	[25]
13	Nanoscale zero-valent iron prepared from steel pickling waste liquor (S-NZVI)	Nitrate (NO_3^-) con. = 20 mg L ⁻¹ , initial pH = 5, material dosage = 2.8 g L ⁻¹ , temperature = 25 °C, and contact time = 4 h.	99%	[26]
14	Fe/Ni Bimetallic Nanoparticles	Nitrate (NO_3^-) con. = 300 mg L ⁻¹ , initial pH = free, material dosage = 0.2 g L ⁻¹ , temperature = 25 °C, and contact time = 15 min.	99.8%	[27]

Table 2. Application of nanoscale zerovalent iron particles and its composites in phosphorus removal from water.

No.	Name of the material or system	Removal conditions	Removal Efficiency (%)	Reference
1	Nanoscale zero-valent iron (nZVI) plus copper chloride addition (CuCl_2)	Phosphorus (P) con. = 50 mg L^{-1} , initial pH = 2, material dosage = 1 g L^{-1} , CuCl_2 = 25 mg, temperature = 25 °C, contact time = 60 min.	100%	[34]
2	Starch-stabilized nanoscale zerovalent iron (SnZVI)	Phosphate (PO_4^{3-}) con. = 100 mg L^{-1} , initial pH = 4, material dosage = 1 g L^{-1} , temperature = 10 °C, contact time = 60 min.	100%	[35]
3	Magnetic bimetallic nanoparticles (Fe–Ag)	Phosphate (PO_4^{3-}) con. = 10 mg L^{-1} , initial pH = N/A, material dosage = 1 g L^{-1} , temperature = 25 °C, contact time = 5 days.	100%	[36]
4	Polyacrylamide coated nanoscale zero-valent iron particles (PPA-nZVI)	Phosphorus (P) con. = 100 mg L^{-1} , initial pH = 7, material dosage = 1 g L^{-1} , temperature = 25 °C, and contact time = 120 min.	100%	[14]
5	Activated carbon supported nanoscale zero-valent iron Particles (AC-nZVI).	Phosphorus (P) con. = 50 mg L^{-1} , initial pH = 2, material dosage = 8 g L^{-1} , room temperature, and contact time = 24 h.	99.5%	[37]
6	Magnesium hydroxide coated-nanoscale zerovalent iron nanoparticles (Fe^0 [Mg/Fe]$^{1.0}$)	Phosphorus (P) con. = 50 mg L^{-1} , initial pH = 7, material dosage = 1 g L^{-1} , temperature = 25 °C, and contact time = 180 min.	97.4%	[38]
7	Nanoscale zerovalent iron (nZVI)	Phosphorus (P) con. = 50 mg L^{-1} , initial pH = 7, material dosage = 1 g L^{-1} , temperature = 25 °C, and contact time = 180 min.	94.05%	[31]
8	Encapsulated nanoscale zero-valent iron	Phosphate (PO_4^{3-}) con. = 10 mg L^{-1} , initial pH = 6.5, material dosage = 20 g L^{-1} , temperature = 25 °C, contact time = 30 min.	42%	[39]
9	Nanoscale zero-valent iron/nickel supported on zeolite (Z-Fe/Ni)	Phosphate (PO_4^{3-}) con. = 5 mg L^{-1} , initial pH = 3, material dosage = 0.5 g L^{-1} , temperature = 25 °C, contact time = 6 h.	98%	[40]
10	Sepiolite-nano zero valent iron composite	Phosphorus (P) con. = 25 mg L^{-1} , initial pH = 4.5, material dosage = 0.8 g L^{-1} , temperature = N/A, contact time = 46.26 min.	99.43%	[41]
11	Clay-supported nanoscale zero-valent iron	Phosphorus (P) con. = 500 mg L^{-1} , initial pH = 2, material dosage = 5 g L^{-1} , temperature = room temperature, contact time = 24 h.	30%	[42]
12	Nanoscale zerovalent iron particles supported on spondias purpurea seed waste (S-NaOH-NZVI)	Phosphate (PO_4^{3-}) con. = 19 mg L^{-1} , initial pH = 4, material dosage = 1 g L^{-1} , temperature = 25 °C, contact time = 120 min.	99.9%	[43]
13	Nanoscale zerovalent iron particles (nZVI) derived from reducing natural goethite	Phosphorus (P) con. = 5.02 mg L^{-1} , initial pH = 6.8, material dosage = 0.6 g L^{-1} , particle size ≤ 75 μm . temperature = 23 °C, contact time = 120 min.	99.2%	[44]
14	Nanoscale zerovalent iron (nZVI)	Phosphate (PO_4^{3-}) con. = 10 mg L^{-1} , initial pH = N/A, material dosage = 1 g L^{-1} , temperature = 25 °C, contact time = 180 min.	99%	[58]

Table 3. Application of nanoscale zerovalent iron particles and its composites in heavy metals removal from water.

No.	Name of the material or system	Removal conditions	Removal Efficiency (%)	Reference
1	CaCO ₃ coated nanoscale zero-valent iron (nZVI) (C-nZVI)	Chromium(VI) [Cr(VI)] con. = 5 mg L ⁻¹ , initial pH = 3, Fe ⁰ dosage = 250 mg L ⁻¹ , temperature = 20 °C, CaCO ₃ /Fe ⁰ = 1:4, and contact time = 120 min.	100%	[49]
2	Chitosan-stabilized biochar supported S-nZVI (CS@BC/S-nZVI)	Cr(VI) con. = 50 mg L ⁻¹ , initial pH = 3, material dosage = 0.4 g L ⁻¹ , temperature = 25 °C, S/Fe mole ratio = 0.14, BC/Fe mass ratio = 0.25, and contact time = 120 min.	99.83%	[50]
3	Montmorillonite microparticles with nanoscale zero-valent iron (Mt-nZVI)	Cr(VI) con. = 20 mg L ⁻¹ , initial pH = 5.5, material dosage = 2.75 g L ⁻¹ , temperature = 25 °C, and contact time = 240 min.	99.95%	[59]
4	Nanoscale Zero Valent Iron Supported by Biomass-Activated Carbon (BC-nZVI)	Cr(VI) con. = 100 mg L ⁻¹ , initial pH = 3, material dosage = 400 mg L ⁻¹ , temperature = 25 °C, and contact time = 180 min.	99.12%	[51]
5	Starch coated nanoscale zerovalent iron particles supported on activated carbon (S-nZVI@AC)	Cr(VI) con. = 10 mg L ⁻¹ , initial pH = 5, mass ratio of S-nZVI@AC/Cr = 60:1, temperature = 25 °C, and contact time = 330 min.	99.96%	[60]
6	Magnetic iron nanoparticles assisted biochar (nZVI-BC)	Cr(VI) con. = 50 mg L ⁻¹ , initial pH = 4, material dosage = 2 g L ⁻¹ , Fe/BC ratio = 2:1, temperature = 20 °C, and contact time = 360 min.	99%	[61]
7	Humus-supported nanoscale zero-valent iron (H-nZVI)	Cr(VI) con. = 25 mg L ⁻¹ , initial pH = 6.5, material dosage = 6 g L ⁻¹ , NZVI loading = 10.4%, temperature = 25 °C, and contact time = 90 min.	99.9%	[62]
8	Biochar derived from nano zero-valent-iron (nZVI) and sewage sludge (nZVIB).	Arsenic(V) [As(V)] con. = 20 mg L ⁻¹ , initial pH = 2, material dosage = 10 g L ⁻¹ , temperature = 25 °C, contact time = 24 hr.	99%	[63]
9	Zeolite-supported sulfide nanoscale zero-valent iron (S-nZVI@ZSM-5)	As(V) con. = 5 mg L ⁻¹ , initial pH = 4, material dosage = 0.1 g L ⁻¹ , temperature = 25 °C, contact time = 300 min.	94.5%	[64]
10	Supported nano zero-valent iron on activated carbon (NZVI/AC)	As(V) con. = 2 mg L ⁻¹ , initial pH = 6.5, material dosage = 3 g L ⁻¹ , temperature = 25 °C, contact time = 72 h.	100%	[56]
11	Polyaniline/Fe ⁰ composite nanofibers (PANI/Fe⁰)	As(V) con. = 5 mg L ⁻¹ , initial pH = 4, material dosage = 0.1 g L ⁻¹ , temperature = 25 °C, contact time = 24 h.	99.9%	[65]
12	Magnetic Cellulose Nanocomposites (Cellulose@nZVI)	As(III) con. = 10 mg L ⁻¹ , initial pH = 8, material dosage = 1 g L ⁻¹ , temperature = 25 °C, contact time = 180 min.	100%	[66]
13	Chitosan fiber-supported zero-valent iron nanoparticles	As(V) con. = 0.01 mg L ⁻¹ , initial pH = 6, material dosage = 0.167 g L ⁻¹ , temperature = 25 °C, contact time = 60 min.	70%	[57]
14	nanoscale zero-valent iron stabilized with starch and carboxymethyl cellulose	Arsenic(V) [As(V)] con. = 2 mg L ⁻¹ , initial pH = 7, material dosage = 0.3 g L ⁻¹ , temperature = 25 °C, contact time = 120 min.	100%	[55]

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