



Research paper

Radioactive iodine capture and storage from water using magnetite nanoparticles encapsulated in polypyrrole



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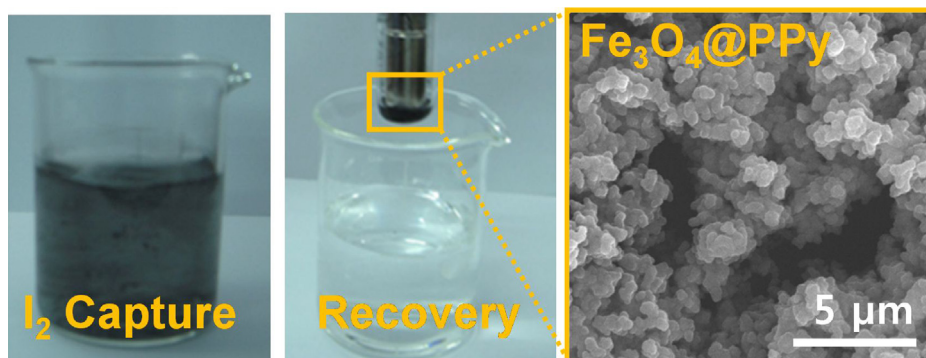
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HIGHLIGHTS

- The Fe₃O₄@PPy is environment friendly, ferromagnetic and can be regenerated using ethanol.
- Fe₃O₄@PPy shows aqueous radioactive iodine adsorption capacity 1627 mg/g.
- The adsorbed iodine shows 2% release of iodine during nitrogen gas sweeping test.
- The XPS spectrum confirms presence of polyiodides ions (I₃⁻, I₅⁻) on the surface of adsorbent.

GRAPHICAL ABSTRACT



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ABSTRACT

The effective capture and storage of radioactive iodine is of importance for nuclear waste storage during nuclear power station accidents. Here we report Fe₃O₄@PPy powder containing ~12 nm magnetite (Fe₃O₄) nanoparticles encapsulated in the polypyrrole (PPy) matrix. It shows 1627 mg/g uptake of iodine dissolved in water, within 2 h at room temperature. Fe₃O₄@PPy is ferromagnetic in nature and can be separated from water using external magnetic field. The nitrogen gas sweeping test at 30 °C shows release of 2% iodine from iodine adsorbed Fe₃O₄@PPy, revealing stable storage of iodine for a moderate period. The iodine-adsorbed magnetic powder can be regenerated by washing with ethanol. The XPS spectrum of iodine adsorbed Fe₃O₄@PPy confirmed the presence of polyiodides (I₃⁻ and I₅⁻) bound to the PPy surface. This excellent iodine capture and storage from iodine contaminated water is an environment friendly, inexpensive and large scale method.

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1. Introduction

There is an increasing concern on health and environmental risks, specifically on nuclear accidents and hazardous wastes,

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caused by wide applications of nuclear energy [1,2]. The Chernobyl disaster in 1986 and the Fukushima nuclear disaster caused by a tsunami in 2011 have attracted serious attentions on the efficient removal and reliable storage of radioactive nuclear wastes. Radioiodine wastes generated from fission of uranium in a nuclear reactor may disperse rapidly in air and water in the event of nuclear leakage, causing long-term health risks [3,4]. The main components of radioiodine waste are ¹³¹I and ¹²⁹I with half-lives of 8 days and 15.7 million years, respectively. In the waste streams of nuclear plant,

radioactive iodine I^{131} exists as molecular iodine (I_2), iodide (I^-), hypoiodite ion (IO^-), and iodate (IO_3^-) [5]. The iodine dissolved in water can be ingested through food or water consumption, affecting human metabolic processes [6].

So far, a variety of materials have been developed for effective capture and storage of volatile radioactive iodine [7,8]. The silver exchanged zeolites [9,10], metal organic framework [11,12], conjugated microporous polymer [13,14], polyacrylonitrile–chalcogen [15], melamine–citrazinic acid hydrogelator [16], deep eutectic solvents [17], nanoporous carbon [18] and graphene based sorbents [19] have been used in capturing gaseous iodine and iodine dissolved in non-aqueous solvents. However, their poor water stability, high cost and toxicity weaken their application in capturing and storing iodine.

The removal of iodine from water was carried out using silica coated magnetite nanoparticles [20], zerovalent iron nanoparticles [21], Mg–Al(NO_3) layered double hydroxide (LDH) [22], Ag_2O grafted titanate nanolamina [23] and layered bismuth–iodine–oxide [24]. However, these adsorbent showed poor adsorption capacity and recyclability. The powdered activated carbon has been used for removal of radioactive iodine from water [25,26]. Iodine adsorption isotherms on chemically activated *Salsola vermiculata* plant and Merck activated carbon showed iodine adsorbed concentration 1178 mg/g and 975 mg/g, respectively [27]. The bituminous coal-based granular activated carbon (GAC-B), the granular carbon made of coconut shells (GAC-C) and the activated carbon fabric (ACF) made from polyacrylonitrile showed aqueous iodine adsorption capacity 1016 mg/g, 940 mg/g and 889 mg/g, respectively [28]. Synthesis of chemically activated carbon generates carbon dioxide which is a major cause of global warming.

Because of the strong affinity between Lewis basic nitrogen and Lewis acidic iodine, different types of nitrogen containing adsorbents have been widely used in the capture and storage of radioactive iodine from non aqueous medium [13–17]. Polypyrrole (PPy) is an environment friendly conducting polymer and has been used for removal of toxic pollutants from water [29]. Polypyrrole exhibited high adsorption capacity for sulphate [30], bromate [31], chlorate [32], chromates [33], fluoride [34], nitrate [35], and many other anions but has not been used for capturing and storing of radioactive iodine dissolved in water. As compared to nonmagnetic PPy, the magnetic nature of adsorbent is beneficial as it can be guided using external magnetic fields; hence, there is no need to filter, and consequently capturing and storing can be simply and easily done at low cost [36–38]. Here we report synthesis of magnetite nanoparticles encapsulated polypyrrole ($Fe_3O_4@PPy$) powder for effective capture and stable storage of radioactive iodine dissolved in water.

2. Experimental section

2.1. Chemicals

Pyrrole (Sigma Aldrich), ammonium persulphate (Sigma Aldrich), hydrochloric acid (Merck), ferric chloride hexahydrate (Merck), ferrous chloride tetrahydrate (Merck), liquor ammonia (Merck), were obtained and used for synthesis of adsorbents and iodine (Loba) for making contaminated water sample.

2.2. Synthesis of polypyrrole (PPy)

The synthesis of polypyrrole was carried out via chemical polymerization using ammonium persulphate as oxidant. 1 g of freshly distilled pyrrole was added in 75 ml of distilled water and dispersed in ultrasonic bath for 10 min. 3 g of ammonium persulphate (APS)

was dissolved in 25 ml water and the resulting solution was added in the pyrrole solution with constant stirring at ice temperature ($0-5^\circ C$) and the reaction was carried out for 4 h. The reaction mixture was stand at room temperature for overnight to complete the polymerization process. The product was washed with copious amount of distilled water and methanol and dried at $90^\circ C$ for 24 h in a hot air oven.

2.3. Synthesis of Fe_3O_4 nanoparticles

Ferric chloride hexahydrate ($FeCl_3 \cdot 6H_2O$, wt = 1.3515 g) and ferrous chloride tetrahydrate ($FeCl_2 \cdot 4H_2O$, wt = 0.4971 g) were dissolved in 50 ml water. The resulting solution was kept on heating mantle and the temperature rose to $90^\circ C$. The required amount of aqueous ammonia solution was added to the reaction mixture slowly with continuous mechanical stirring and the pH of solution was maintained to be 10 using aqueous ammonia. The reaction mixture became black in color and after 2 h the suspension was cool down to room temperature. The black color product was separated by magnet and washed with distilled water till the filtrate becomes neutral.

2.4. Synthesis of $Fe_3O_4@PPy$

The required amount of Fe_3O_4 (10–40 wt% of pyrrole) was dispersed in 75 ml water. 1 g of pyrrole was added to this solution and dispersed in the mixture for 10 min in an ultrasonic bath. A solution of ammonium persulphate (3.0 g/25 ml) was added in the mixture with constant mechanical stirring at ice temperature ($0-5^\circ C$) for 4 h and kept at room temperature for overnight. The product was washed with distilled water and methanol and dried at $90^\circ C$ for 24 h.

2.5. Iodine adsorption

A stock solution of 200 ppm was prepared in deionized distilled water by dissolving 0.1 g of naturally occurring nonradioactive I_2 in 500 ml water and stored in the dark place. The pH of the solution was maintained at 7 using HCl and NaOH solution. The experimental solutions of desired iodine concentrations were obtained by successive dilutions of the stock solution. The required amount of adsorbent was added to iodine solution and mechanically stirred for different time intervals. At the end of adsorption experiment, the adsorbent was separated from solution using hand held magnet. The iodine concentration in the solution after adsorption was measured using UV–vis spectrophotometer by monitoring the absorbance change at maximum wavelength ($\lambda_{max} \sim 287$ nm). The equilibrium adsorption capacity was calculated q ($mg\ g^{-1}$) = $(C_0 - C_e) \times V/w$, where C_0 and C_e are initial and final concentration of iodine in water, respectively, w is the adsorbent dose (mg) and V (ml) volume of iodine solution.

2.6. Effect of Fe_3O_4 loading in PPy on iodine adsorption

The effect of Fe_3O_4 loading (wt%) in PPy on iodine adsorption was studied using 100 ppm solution, pH 7 and at temperature $30^\circ C$. 2 mg adsorbent was added in the 50 ml solution of iodine and stirred in the resulting solution for 3 h. After completion of adsorption the iodine concentration was measured spectrophotometrically.

2.7. Kinetics study

The kinetics of iodine adsorption on $Fe_3O_4@PPy$ was studied in 100 ppm solution at pH = 7 and $T = 30^\circ C$. 4 mg adsorbent was added in 100 ml of 100 ppm solution and kept under mechanical stirrer.

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