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Removal of hexavalent chromium from contaminated waters by ultrasound-assisted aqueous solution ball milling

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ABSTRACT

Batch mode experiments were conducted to study the removal of hexavalent chromium (Cr(VI)) from aqueous solutions using ultrasound-assisted aqueous solution ball milling. The results show that the reduction rate of Cr(VI) by ultrasound-assisted aqueous solution ball milling was significantly faster than that by ball milling or ultrasound treatment alone, and an initial Cr(VI) concentration of 166 mg/L could be decreased to 0.35 mg/L at 120 min. The decisive factors, including initial concentration of Cr(VI), pH value, ultrasonic frequency and filling gas, were studied. It was found that the optimal ultrasonic frequency for ultrasound-assisted aqueous solution ball milling device was 20 kHz, and the rate of Cr(VI) reduction as a function of filling gas followed the order: Ar > air > N₂ > O₂. Samples were characterized by X-ray diffraction, fluorescence measurements, atomic absorption and the diphenylcarbazide colorimetric method. The Cr(VI) transformed into a precipitate that could be removed from the contaminated water, after which the water could be reused.

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Introduction

Chromium has been widely used in multiple industries and in agriculture. Among the extensive historical applications, metallurgy, electroplating, leather tanning, fungicides and corrosion inhibitors have been most common. Cr(VI) is generally soluble over a wide pH range and has been shown to exert toxic and carcinogenic effects on humans and experimental animals (Costa, 1997; Flury et al., 2009; Zazo et al., 2008). Leakage, poor storage practices and improper disposal of waste related to chromium uses have released Cr(VI) into the environment, causing contamination of groundwater and surface water (Flury et al., 2009). Therefore, Cr(VI) must be removed from wastewaters before their disposal to natural aquatic environments. Among chromium oxidation states ranging from (–IV) to (+VI), only the (+III) and (+VI) states are stable in the natural environment (Pechova and Pavlata, 2007). Cr(III) is considered to be an essential

nutrient for the human body, and the toxicity of Cr(III) is 500–1000 times smaller than Cr(VI) for living cells.

During recent years, various methods have been employed for removing Cr(VI) from contaminated water (Dima et al., 2015; Janoš et al., 2009; Liang et al., 2014; Shirzad-Siboni et al., 2014; Ritter et al., 2002; Lai and Lo, 2008), such as adsorption (Hu et al., 2005, 2007a, 2007b; Shen et al., 2009a, 2009b; Park et al., 2006), separation with reverse-osmosis (Ozaki et al., 2002), and reduction by zero-valent iron (Liu et al., 2009; Lee et al., 2010), in which reducing Cr(VI) to Cr(III) is considered to be a satisfactory solution to eliminate the toxicity of Cr(VI).

Using zero-valent iron for *in situ* reduction of Cr(VI) has become increasingly popular (Gheju and Iovi, 2006; Chen et al., 2008). The conventional process using zero-valent iron powder in aqueous solution has two disadvantages: low removal efficiency and large iron loss. The reduction rate of Cr(VI) is improved when the pH value is lowered, but the adjustment of pH adds to the complexity of the process (Gheju and Balcu, 2011;

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Chen et al., 2007). Although nano zero-valent iron powder has high efficiency, it presents some shortcomings, such as tendency to agglomerate, difficulty of preservation, and complex preparation process (Ponder et al., 2000; Geng et al., 2009).

This study investigated the use of ultrasound-assisted aqueous solution ball milling, which is a combination of ultrasound and ball milling techniques. This process cannot only provide fresh surfaces on the iron milling balls by impaction–abrasion, but also generate radicals and H^+ ions to participate in the Cr(VI) reduction reactions. Therefore, ultrasound-assisted aqueous solution ball milling can remove Cr(VI) from high concentration contaminated waters in a short time, and form a stable precipitate. Reaction with zero-valent iron is the main principle of the removal of Cr(VI) by ultrasound-assisted aqueous solution ball milling. If other toxic substances can be removed by zero-valent iron, they can be removed by ultrasound-assisted aqueous solution ball milling, too. So the continuous production of new zero-valent iron will make ultrasound-assisted aqueous solution ball milling applicable to not only removal of Cr(VI), but also removal of other toxic substances, such as Pb(II) (Ponder et al., 2000), arsenic(III) (Liu et al., 2016), Methyl Orange (Yuan et al., 2016), and dinitrotoluenes (Patapas et al., 2007). Besides, the iron balls can be reused in the experiment, which can lower iron loss. The resulting precipitate can be removed from the contaminated water, and the treated water can be used again. High efficiency, no adjustment of pH and simple operation process make this method green and practicable.

1. Methods

1.1. Devices and materials

A special stainless steel milling pot 165 mm in diameter and 158 mm in height was designed to remove the Cr(VI) by ultrasound-assisted aqueous solution ball milling. The details of this device are shown in Fig. 1. The Cr(VI) solution was added to the milling pot, and air was introduced into the solution for 3–5 min. Then iron milling balls ($\Phi = 1\text{--}1.5$ mm) with a total weight of 2 kg were added into the milling pot. Moreover, the stirring rod was adjusted to a height of 5 mm from the bottom of the pot before sealing the milling lid and tightening the screws. Either the ball milling stirring motor or ultrasonic generator were activated to remove Cr(VI) by ball milling or by ultrasound treatment. Both functions were activated to remove Cr(VI) by ultrasound-assisted aqueous solution ball milling. The stirring speed was kept constant at 250 r/min. The average temperature in the milling pot was 50°C , and the power of the ultrasonic generator was 66 W. Samples were extracted at set time intervals, then centrifuged and filtered with a $0.45\ \mu\text{m}$ membrane. The precipitate was dried at room temperature. The clear liquor and precipitate were tested respectively. Table 1 shows the parameters for the different experiments. Analysis of Cr(VI) and Cr(total) concentration, fluorescence intensity, and X-ray diffraction (XRD) were carried out in experiment I. Analysis of Cr(VI) was implemented in experiments II–V.

Cr(VI) solutions were prepared by dissolving $K_2Cr_2O_7$ in deionized water. All experiments were performed in a

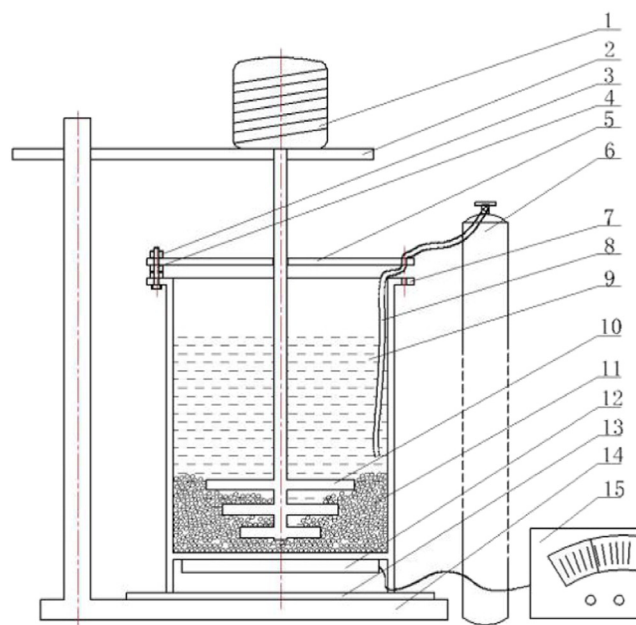


Fig. 1 – Schematic diagram of ultrasound-assisted aqueous solution ball milling. 1—electromotor; 2—lift platform; 3—nut $\times 9$; 4—bolt $\times 3$; 5—cover of milling pot; 6—gas pump; 7—stainless steel milling pot; 8—snorkel $\times 3$; 9—reaction medium; 10—stirring rod; 11—iron milling balls; 12—ultrasound transducer; 13—base of milling pot; 14—supporter; 15—ultrasonic generator.

background electrolyte mixture (50 mg/L Ca^{2+} ; 20 mg/L Mg^{2+} ; 128 mg/L Cl^- ; 104 mg/L Na^+ ; 293 mg/L HCO_3^-) to maintain a constant ionic strength. H_2SO_4 or NaOH was used for changing the pH in experiment III. The pH values were measured using a pH meter Model PHB-1. All chemicals used were of AR grade. Before the experiment, the iron balls were washed with HCl.

1.2. Measurement of fluorescence intensity

In order to achieve pH = 7.3, the raw materials, phosphate buffer solution (0.2 mol KH_2PO_4 and 0.2 mol NaOH) and terephthalic acid (0.16 mol) and free-radical indicator solution were put into the milling pot in the fluorescence determination experiments. After every 20 mL samples were extracted using disposable pipettes at specific time intervals, and an equivalent amount of buffer and indicator solutions were added. Then samples were filtered several times to obtain a clear solution. A Fluorescence Spectrophotometer (Model F2500, Hitachi, Japan) was used to record and monitor the fluorescence intensities of samples at 310 nm excitation and 426 nm emission.

1.3. Measurement of Cr(VI) concentration and Cr(total) concentration

The analysis of Cr(VI) concentration in solution was carried out by the diphenylcarbazide colorimetric method with a Shimadzu UV-2550PC spectrophotometer (APHA, 2005). The purple color was fully developed after 15 min and the sample solutions were transferred to a 1 cm path length glass cell, and the absorbance of the solution was measured at 540 nm.

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