



## Reduction of noxious Cr(VI) ion to Cr(III) ion in aqueous solutions using H<sub>2</sub>O<sub>2</sub> and UV/H<sub>2</sub>O<sub>2</sub> systems



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### ABSTRACT

Conversion of the toxic Cr(VI) to the less toxic Cr(III) in aqueous solutions by chemical and photochemical reduction was carried out using H<sub>2</sub>O<sub>2</sub> and UV radiation. The whole mechanism was well elucidated and investigated using potassium dichromate (K<sub>2</sub>Cr<sub>2</sub>O<sub>7</sub>) as a standard model compound. The effects of potential factors affecting the reduction efficiency were well studied and optimized, the optimized parameters such as concentration of H<sub>2</sub>O<sub>2</sub> (from 0.5 to 1.5 M), pH (from 5 to 9), irradiation time (0 to 140 min) and reaction time (0 to 120 min). Initial concentration of Cr(VI) in the solution was varied from 0.1 to 15 mg/L plays a significant role in the mechanism involves for the reduction of noxious Cr(VI) to less toxic Cr(III).

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Chromium occurs in the nature mainly in three common oxidation states, i.e. Cr(III) and Cr(VI) and Cr(0), the former being the most stable form and available naturally in the environment while the latter are generated due to several hazardous industrial processes [1]. Cr(VI) ion is noxious to most of the biotic organisms as well abiotic organisms, higher than 0.05 ppm concentration of Cr(VI) lead to severe detrimental and lethal effect on the prevailing flora and fauna [2], it act as carcinogenic agent in animals and causes severe irritation problems in humans. Cr(VI) ion is very soluble in water and forms divalent oxy-anions such chromate (CrO<sub>4</sub><sup>2-</sup>) and dichromate (Cr<sub>2</sub>O<sub>7</sub><sup>2-</sup>). As it is a sorbent, Cr(VI) ion can easily flow through sediments and cause contamination of aquifers, groundwater and various other forms of aquatic source. Cr(III) is readily precipitated or adsorbed on a variety of inorganic and organic substrates at neutral or alkaline pH [2,3]. Reduction of noxious Cr(VI) to stable Cr(III) plays a vital role in the remediation

of environmental sites contaminated by chromium. Indeed, Cr(III) is an essential nutrient (especially in glucose metabolism) for all the biotic creatures including flora as well as fauna [2–4], on the other hand Cr(VI) is perilous to humans, animals, plants and microorganisms and lead to severe detrimental effect on the liver and kidney, causes internal hemorrhage, dermatitis, respiratory damages and lung cancer [2–5]. According to the World Health Organization (WHO), the metals which are required to remove from the environment and need immediate concern are Cr, Zn, Fe, Hg and Pb [6]. The maximum allowed limits for contaminants in “treated” wastewater are enforced in developed and many developing countries [3,4]. Several techniques and adsorbents such as electrocoagulation [7], activated carbon from logan seeds [8], zeolite prepared from raw fly ash [9], exfoliated polypyrrole-organically modified montmorillonite clay nanocomposite [10], illuminated ZnO/TiO<sub>2</sub> composite [11], alumina [12] were used for the sorption and reduction of the Cr(VI) species.

In the presented work, the rapid reduction of Cr(VI) ion by UV, H<sub>2</sub>O<sub>2</sub> and UV/H<sub>2</sub>O<sub>2</sub> was well studied and elucidated using K<sub>2</sub>Cr<sub>2</sub>O<sub>7</sub> solutions as a model contaminant. The impact of the crucial parameters such as irradiation time with UV, concentration of H<sub>2</sub>O<sub>2</sub>, pH, initial concentration of Cr(VI) ion and reaction time were well examined and optimized. Hydrogen peroxide has been used as

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the reduction reagent.  $\text{H}_2\text{O}_2$  is both not toxic and effective and it converts to  $\text{O}_2$  and  $\text{H}_2\text{O}$  in the reaction with  $\text{Cr(VI)}$  ion. The purpose of this study was to investigate the reduction of  $\text{Cr(VI)}$  ion in aqueous solutions using  $\text{H}_2\text{O}_2$  and UV/ $\text{H}_2\text{O}_2$  systems.

## Materials

Analytical grade (AR) reagents were purchased from different suppliers and were used without any further purification. Potassium dichromate ( $\text{K}_2\text{Cr}_2\text{O}_7$ ) was used as the model solution for the production of  $\text{Cr(VI)}$  ion. Hydrogen peroxide ( $\text{H}_2\text{O}_2$ ) solutions 2% (30%, v/v) were prepared by dilution of 30%  $\text{H}_2\text{O}_2$  (Merck) in double distilled water.

## Experimental

The experiments were carried out in a cylindrical glass reactor with 12 cm in inner diameter and 30 cm in height (Fig. 1). A 1000-mL  $\text{Cr(VI)}$  ion solution (adjusted at different physiological pHs according to the experiment) was introduced into the reactor with a predetermined amount of  $\text{H}_2\text{O}_2$ . The reactor was tightly closed and the reaction solution was stirred by means of a magnetic stirrer to ensure complete mixing. For the photo reduction experiments, an ultraviolet lamp fixed at 254 nm and 8 W power) was attached into a quartz glass tube at the center of the reactor. Samples were withdrawn at different time intervals and analyzed to monitor the progress of the reaction. The reactor temperature was maintained constant at  $20 \pm 1^\circ\text{C}$  by circulating chilled water.

The residual concentrations of  $\text{Cr(VI)}$  ions were measured spectroscopically in a Perkin-Elmer Lambda 25 UV-vis Spectrophotometer (Perkin-Elmer, Norwalk, CT) by the diphenylcarbazide method at 540 nm [13].  $\text{Cr(VI)}$  ion reduction was calculated and expressed in percentage.

The results of the rapid reduction of  $\text{Cr(VI)}$  ion by  $\text{H}_2\text{O}_2$  per time are presented in Fig. 2. The reduction rate of  $\text{Cr(VI)}$  ion was very fast

initially, leading to a rapid decrease in the concentration of  $\text{Cr(VI)}$  ion. About 86, 74 and 64% of the initial  $\text{Cr(VI)}$  ion was reduced within the first 5 min of reaction at different physiological pHs 5, 7 and 9, respectively. Then, the residual concentration of  $\text{Cr(VI)}$  ion became almost constant after 15 min of reaction time. Therefore, subsequent experiments were performed at this reaction time.

Fig. 3 shows a linear relationship between the logarithm of the  $\text{Cr(VI)}$  ions concentration and the reaction time, showing that the reduction of  $\text{Cr(VI)}$  ions by  $\text{H}_2\text{O}_2$  followed first-order kinetics.

The relationship between the reduction efficiency of  $\text{Cr(VI)}$  ion and the pH value is shown in Fig. 4. It reveals that the pH had a significant effect on the reduction of  $\text{Cr(VI)}$  ion using  $\text{H}_2\text{O}_2$ . Reduction of  $\text{Cr(VI)}$  ion in acidic and neutral solutions by hydrogen peroxide was more efficient than in alkaline solution. In general, the pH value of surface water and groundwater is between 5 and 9. Most of the reduction reagents reported in the literature was either less effective or non-effective in alkaline solutions [2–4,14]. However,  $\text{H}_2\text{O}_2$  reduced  $\text{Cr(VI)}$  ion in a wide range of pH.

In the physiological pH range from 1 to 6,  $\text{HCrO}_4^-$  is the predominant form of  $\text{Cr(VI)}$  ion, while above pH 6 the anion  $\text{CrO}_4^{2-}$  predominates [2]. As shown in Fig. 4 the pH value had a significant effect on the reduction of  $\text{Cr(VI)}$  ion. Thus, increasing the pH from 5 to 9 at an initial concentration of 0.1 mg/L of  $\text{Cr(VI)}$  ion, the percentage of reduction decreased from 83.0 to 54.8% (Fig. 4).

The reduction of  $\text{Cr(VI)}$  followed first-order kinetics:

$$\ln \left[ \frac{C}{C_0} \right] = k_{\text{red}} t \quad (1)$$

where  $k_{\text{red}}$  is the photo catalytic reduction rate constant,  $t$  is the reaction time,  $C_0$  is the concentration of  $\text{Cr(VI)}$  ion at time zero and  $C$  is the  $\text{Cr(VI)}$  ion concentration at a particular time. The observed first-order rate constants for the decrease in the concentration of  $\text{Cr(VI)}$  ion as a function of pH, are presented in Table 1, reveals that the pH of the reaction solution has a significant effect on the

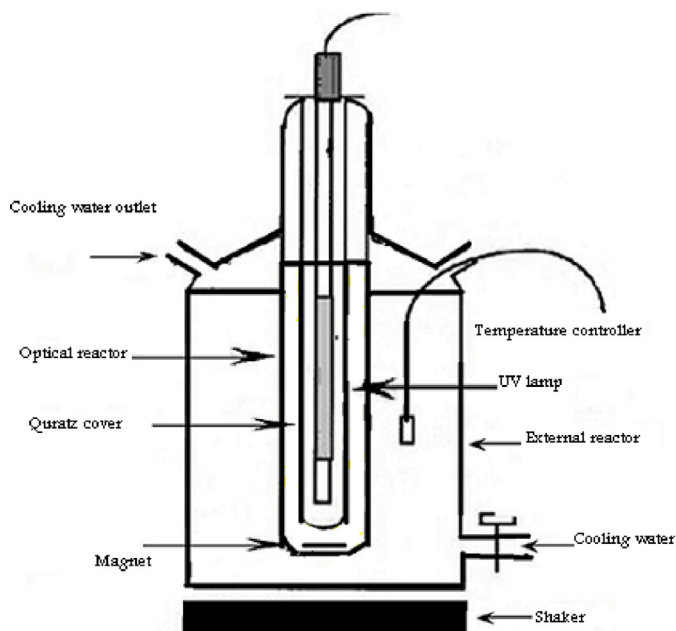


Fig. 1. Schematic of the reactor.

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