



# Separate and simultaneous removal of phenol, chromium, and cyanide from aqueous solution by coagulation/precipitation: Mechanisms and theory



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## HIGHLIGHTS

- A conformational mechanism for organics and inorganics interactions is proposed.
- Performance of coagulation declines in simultaneous presence of cyanide and chromium.
- Enhanced coagulation relies on rising surface area and destroying cyano-complexes.

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## ABSTRACT

Presence of heavy metals, phenols, and cyanides is highly dangerous, and in the simultaneous presence, the effect is compounded. Chemical precipitation of such pollutants from industrial waste has gained momentum in recent years, as it promise to surpass the major drawbacks laid by the other physicochemical and biological methods and can be practically carried out in all conditions. The present work illustrates the possible removal mechanisms and interactions between organic and inorganic compounds during this process. Jar test experiments, using ferric chloride, were conducted to compare the removal efficiencies of phenol, chromium, and cyanide, either in separate or simultaneous presence. Moderate removal of Cr<sup>+6</sup> was observed during conventional coagulation; though removal efficiencies were insignificant in the case of cyanide and phenol. The presence of cyanide ions in the solution showed an antagonistic influence on the removal of chromium. Enhanced coagulation outperformed conventional coagulation in terms of Cr<sup>+6</sup> removal and 97% elimination was attained; however, phenol removal was slightly augmented. Although the results provided no evidence for possible removal mechanisms, electrostatic attraction was considered as the dominant removal mechanism in separate coagulation. Indeed, removal efficiencies of cyanide and chromium decreased in simultaneous presence, which can be attributed to the cyanide and Cr complexes. Higher removal efficiencies which achieved during enhanced coagulation might be due to the increase of specific surface area, aggregation rate, and settling efficiencies, as well as cyano-complexes destruction. The results of Mann–Whitney test revealed a significant difference ( $p_{\text{value}} \leq 0.05$ ) between separate and simultaneous removal performances of phenol and chromium.

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## 1. Introduction

Wastewater discharged by industrial activities is often contaminated by a variety of toxic or otherwise harmful substances. The tendency to combine industrial effluents for treatment in centralized waste treatment facilities, like in industrial parks,

indeed increases the possibility of introducing toxic, hazardous or non-hazardous metal and organic bearing wastes into the wastewater treatment plant's influent. This can be economically profitable but requires a specific treatment cycle that is quite often a combination of physical, chemical, and biological stages. Such an approach has become an increasing cause of concern for environmental protection authorities and scientific community around, especially, the developing world as they attempt to implement affordable and robust wastewater treatment practices. Even in

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highly industrialized countries, the costs and time needed to develop state-of-the-art conventional wastewater treatment facilities make it arduous to address all the problems. Fortunately, a recent flurry of activity in water/wastewater treatment research offers hope in the removal of various individual organic and inorganic compounds, either at laboratory or pilot scale, from aqueous environments [1–5]. While each contaminant has its best available technology (BAT) for removal by itself, these pollutants are present simultaneously in most industrial effluents and also in the influent of centralized waste treatment facilities. There are indeed interests to understand the impacts of interacting factors on simultaneous removal of these pollutants in aqueous solutions. Thus, studies to seek the BAT(s) for simultaneous removing these contaminants from water/wastewater and to increase our understanding of the fate of these organic and inorganic pollutants during a special treatment process are warranted. Nonetheless, studies of this kind for widespread and important environmental pollutants either are lacking or are of uncertain quality.

Chemical precipitation by coagulation/flocculation using hydrolysable trivalent metal salts is a century old and proven technology. This process has traditionally been used in water treatment to decrease turbidity and color and to remove pathogens [1]. It has also been applied for metals capture as well as phosphorous removal in industrial wastewater treatment. Various mechanisms involve in pollutant removal including a combination of charge neutralization, entrapment, adsorption, and complexation with coagulant ions into insoluble aggregates [6–7]. However, coagulation is also capable of some degree of organic matter removal [7]. In recent years, process optimization with respect to organics removal has gained a lot of attention [8]. Conventional coagulation mainly concerns turbidity particles removal though with enhanced coagulation, another type of coagulation, greater removal of organics is expected [8]. Enhanced coagulation is the practice of using coagulant dose in excess of what is normally required for turbidity removal, to achieve a specific reduction of total organic matter by conventional treatment [8–9]. Thus, the concept of enhanced coagulation involves broadening coagulation objectives from turbidity removal to both turbidity and organic matter removals. These dual objectives require modifying coagulation conditions for type of coagulant, coagulant dosage, and coagulation pH [8–9]. Because chemical coagulation/precipitation is a cost competitive, common method especially in developing world, it is logical to investigate ways to improve its performance of simultaneous toxic organic and inorganic contamination removal from aqueous solutions.

In the present study, phenol and chromium have been selected as the target toxic organic and inorganic contaminants, respectively. Phenol – an aromatic organic compound – is an important industrial commodity as a precursor to many materials and useful compounds. EPA (United States Environmental Protection Agency) has warned on the low control of phenol distribution in the environment and has classified phenol as a Group D, not classifiable as to human carcinogenicity, based on a lack of data concerning carcinogenic effects in humans and animals [10]. The primary uses of phenol are in the production of phenolic resins, caprolactam, and bisphenol A. Other uses of phenol include as a slimicide, as a disinfectant, and in medicinal products, etc. [11]. Therefore, due to these uses, phenol and phenolic compounds which are frequently discharged in industrial effluents are accumulated in ground and water bodies, making necessary the implementation of processes that allows its treatment. The industries which produce or use phenol generate wastewater contaminated by substantial amounts of this chemical (0.1–10% by weight) [12]. Chromium, on the other hand, is extensively used in industry and can be present in the environment as  $\text{Cr}^{+3}$  or  $\text{Cr}^{+6}$  [13]. The hexavalent chromium is 500 times more harmful than the trivalent type; thus, EPA has laid down the maximum contaminant level (MCL) for  $\text{Cr}^{+6}$  in domestic

water supplies to be 0.05 mg/L, while total Cr containing  $\text{Cr}^{+3}$ ,  $\text{Cr}^{+6}$  and other species of chromium is regulated to be discharged below 2 mg/L [14].

Furthermore, cyanide, a carbon–nitrogen highly toxic compound, was selected on the basis that cyanide treatment has been pointed out in scientific literatures for chromium removal [15–16]. Cyanide is usually present in various forms, such as ionic, molecular HCN, salt and metal-complexes in the effluent from various industries including metal electroplating, metal processing, steel tempering, photography, pharmaceuticals, coal coking, and plastics. Although EPA regulations for industrial effluents fixed the maximum contaminant level of cyanide at 0.2 mg/L [17–18], WHO (World Health Organization), EU countries, and National Bank recommend that the levels of cyanide in industrial wastewater should be reduced to 0.07, 0.05, and 0.05, respectively [18]. Presence of phenol, chromium, or cyanide is highly perilous, and in the simultaneous presence, the effects are piled. These pollutants are present simultaneously in the effluent of industries like iron and steel, coal mining, metal plating, etc. [19]. The current investigation explores the use of chemical precipitation with ferric chloride in separate and co-removal of organic pollutants, heavy metals, and cyanide as well as to look in depth at the mechanisms for their removal from aqueous environments.

## 2. Materials and methods

### 2.1. Materials

Phenol ( $\text{C}_6\text{H}_5\text{OH}$ ), chromium trioxide ( $\text{CrO}_3$ ), and potassium cyanide (KCN), as well as reagent grade ferric chloride ( $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ ) were purchased from Merck (Germany) and used without further purification. Stock solutions of chromium and cyanide were prepared by dissolving required quantities of these chemicals in deionized water (DW) ( $\text{EC} < 3 \mu\text{S}/\text{cm}$ ) and were stored in light resistant Pyrex bottles kept refrigerated at 4 °C. Cyanide solution was checked daily for any change in concentration. Similarly, daily stock solutions of phenol were applied. Methanol and ammonium acetate buffer (water solution) were used for high-performance liquid chromatography (HPLC) (CECIL, model 4010).

### 2.2. Procedure

Coagulation/precipitation experiments were performed in a series of jar tests using a conventional sedimentation jar-test apparatus (Velp Scientifica JLT6), equipped with six 1-liter beakers. Raw synthetic samples containing cyanide (10 mg/L), chromium (10 mg/L), and phenol (150 mg/L) were prepared. In order to facilitate the analytical procedures and to obtain a better accuracy of the measurements, relatively high concentrations of contaminants were used, compared to the levels in municipal and industrial wastewater. Adjustment of pH, in the range of 3–11, was performed by the addition of 1 N NaOH or  $\text{H}_2\text{SO}_4$  prior to rapid mixing and pH measurements were done by use of a HQ40d pH meter (Hach, Loveland, Colorado, USA). When needed, the synthetic samples were spiked with  $\text{NaHCO}_3$  as an alkalinity and pH buffer. The concentration of  $\text{NaHCO}_3$  was selected so that the alkalinity of water was in the range of 80–120 mg/L as  $\text{CaCO}_3$ , the typical alkalinity value of natural aqueous solutions [8]. The solution was mixed and kept at room temperature ( $\sim 25$  °C) for 24 h to reach equilibrium prior to jar testing. Before the jar test, volatilization/precipitation of studied contaminants was also examined over the initial pH range of 3–11, without coagulant addition, to investigate the possible effect of mixing and pH on their removal rates. During a typical jar test, a known volume of ferric chloride stock solution was added to each beaker and the solutions were stirred for

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