



Diffusion and leachability index studies on stabilization of chromium contaminated soil using fly ash



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HIGHLIGHTS

- Reduction followed by S/S using FA is an effective treatment option.
- Effective diffusion coefficient was between 2.04×10^{-11} and 9.03×10^{-11} cm² s⁻¹.
- Leachability index was between 8 and 9.
- Characteristic compressive strength was between 24.47 and 40.49 kg/cm².
- To transform the laboratory studies to field implementation this data is useful.

ARTICLE INFO

Article history:

Received 24 January 2015

Received in revised form 14 April 2015

Accepted 16 April 2015

Available online 18 April 2015

Keywords:

Calcium polysulfide

Chromium

Leachability index

Solidification and stabilization

Toxicity Characteristic Leaching Procedure

ABSTRACT

Experiments were performed to establish a feasible treatment process for the solidification and stabilization (S/S) of soil contaminated by leaching of Cr(VI) from Chromite ore processing residue (COPR). Reduction of the highly mobile Cr(VI) was performed using calcium polysulfide (CaS₅) with a dosage of 3 times the molar stoichiometric ratio for the initial concentration of Cr(VI) present in the chromium contaminated soil (CCS). The CCS was solidified and stabilized (S/S) using fly ash (FA) in various proportions i.e., 1:1, 1:2, 1:3 (FA: CCS) with and without using reducing agent i.e., CaS₅. Leachability tests such as Toxicity Characteristic Leaching Procedure (TCLP) and semi-dynamic long term leachability tests indicated that the CaS₅ was effective in reduction of Cr(VI) followed by the S/S process. Leachability Index was derived from the results of the semi-dynamic long term leachability tests and was between 8 and 9, indicating that FA is an effective treatment for disposal into secured landfills for CCS. The characteristic compressive strength of the CaS₅ treated CCS with FA mortar blocks were between 24.47 and 40.49 kg/cm². Considering the cost of CaS₅ and FA, a total expenditure of Rs. 7826 i.e., US \$ 130.4 would be required for remediation of one tonne of CCS.

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1. Introduction

Chromium contamination of soil and water is a serious environmental hazard and its health impact on human beings is severe and manifold. Chromium exposure through ingestion of food, drinking water, inhalation or dermal contact can cause acute or chronic health hazards such as reproductive and developmental issues [1]. Among its two major states of natural occurrence, Cr(VI) is highly mobile and carcinogenic whereas Cr(III) is less toxic [1,2]. Chromium finds its way into the environment as a result of both

natural phenomena and anthropogenic activities. The major industrial sources of chromium are steel and alloy producing industries, chrome plating and leather processing [3]. Chromite ore processing residue (COPR) is a hazardous waste rich in chromium, a by-product during dichromate production from the chromite ore [4]. Open dumping of COPR, rich with the highly mobile Cr(VI), leads to leaching of the mobile Cr(VI) into the soil, groundwater and adjoining water bodies. In-situ and ex-situ clean-up technologies to clean-up such polluted sites are discussed by Guertin et al. [5]. Chromium mobilization from polluted soil by washing and remediation of total chromium using chelating agents was investigated by Palma et al. [6].

Several attempts have been made in the past for the remediation of superfund sites in USA contaminated with COPR. Cao and

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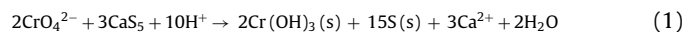
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Zhang [7] conducted laboratory bench scale studies for remediation of COPR using nanoscale iron particles and reported that one gram of nanoparticles was able to reduce 69.3–72.7 mg of Cr(VI) present in the COPR, which is 50–70 times greater than that of iron powders under the same experimental conditions. Bioremediation of Cr(VI) contaminated aquifer using bio-barrier and a permeable-reactive zone technology was investigated by Jeyasingh et al. [8]. Solidification/stabilisation (S/S) is a containment technique which is widely used across the world for immobilisation/remediation of hazardous wastes. This process inhibits the transport of pollutant elements into the surrounding environment and improves the physical characteristics, thus reducing the transport and leaching of contaminants [9,10]. S/S process remains the best available technology demonstrated for remediation of sites contaminated by hazardous wastes with nuclear and radioactive remains and heavy metals; it has been applied in remediation at more than 200 superfund sites [11,5]. In S/S technique, chemical transformation of the contaminant into a state that remains stable and non-toxic to the environment, called stabilization followed by solidification that controls the mobility of the contaminant by encapsulating within the materials of low permeability [12].

Fly ash (FA) primarily consisting of silica, alumina and iron oxide has been widely used in the S/S process [13]. Parsa et al. [14] successfully used Class C FA as the only binding material for the solidification/stabilization of a simulated waste containing toxic elements such as chromium, cadmium and lead, to meet the US Environmental Protection Agency (US EPA) leachability requirements. They also reported that Class 'C' FA yielded better results than Class F. Use of FA improves strength, resistance to pH changes and essentially arrests leaching of the toxic elements from S/S waste material [15,16]. Leaching of trace elements viz., Fe, Mn, Ca, Na, K, Cu, Cr, Zn, As and Pb from coal fly ash was investigated by Dutta et al., [17] and it was reported that blending of fly ash with lime can avoid the water pollution if the fly ash is used as back-filling material under acid mine drainage conditions. The trace elements concentrations are more in acidic fly ash than alkaline fly ash [18]. The mobility of trace elements from selected Australian fly ashes was demonstrated by Jankowski et al. [19], where it was seen that the mobility of trace element depended on the acidic or alkaline nature of fly ash, concentration and pH of the leaching medium. The mobility of water-soluble major trace elements such as Al, Si, K, Na, Ba, Cr, Rb, Sr and V from Spanish fly ashes was assessed by following the open, closed and heated (90 °C) closed leaching systems and the leaching pattern was almost similar for both open and closed systems [20]. Approximately, 112 million tonnes of FA generated in India has great potential for bulk utilisation in developing building materials, road, embankment, mine filling and land development for techno economic augmentation [21]. So, FA was chosen as the inexpensive binding material in the current study.

As a pre-requisite, the Cr(VI) contaminated soil has to be reduced before S/S process. Chemical reduction of the Cr(VI) to Cr(III) for in-situ and ex-situ remediation is achieved using elemental iron, Fe(II), sulphite, sulphide or organic compounds [22]. Besides reduction, S²⁻ will prevent the remobilization of Cr from the S/S matrix [5]. Chrysochoou et al. [23] studied the application of calcium polysulfide (CaS₅) to treat Cr(VI) contaminated soil as an inexpensive and promising reducing agent. Unreacted CaS₅ in the soil dissociates rapidly to form calcium ions and elemental sulfur. After application, polysulfides enter the sulfur cycle immediately and being transformed into various stages of oxidation (sulfate and sulfite). Sulfates and sulfites are water soluble and thus made available for uptake by plants and animals. This is an active uptake process and sulphur is an essential element required for maintaining crucial life functions. Hence, the environmental fate of polysulfides in soil is beneficial [24].

In long term bench scale studies, Wazne et al. [22] were able to meet the New Jersey Department of Environmental Protection (NJDEP) and Toxicity Characteristic Leaching Procedure (TCLP) regulatory standards when CaS₅ was used as a reducing agent for COPR. The reaction between Cr(VI) and CaS₅ is shown below:



The Cr(III) precipitate is then encapsulated by the binder matrix as Cr(III), which is highly stable and favourably adsorbed in soil and cementitious materials [17].

The objective of the present study is to establish a treatment process for the solidification and stabilization (S/S) of CCS due to leaching of Cr(VI) from COPR. Based on the results of TCLP, long term semi-dynamic leaching test and unconfined compressive strength tests were conducted on the treated wastes. From the results, an optimized combination of the FA and contaminated soil, with the required dosage of the reducing agent, was ascertained for safe disposal.

2. Materials and methods

2.1. Sampling and materials used

About 1.5 million tonnes of COPR waste was dumped in the premises of Tamil Nadu Chromates and Chemicals Limited located at Ranipet, Tamilnadu, India. This was accumulated during the operation of the plant that manufactured sodium chromate, chromium salts and basic chromium sulphate till 1995; later on the factory was closed. The CCS samples were collected from different locations in the premises as reported by Kanchinadham et al. [25] Soil from the different sampling points was mixed thoroughly and homogenized. Class 'C' FA used for the remediation process was obtained from Neyveli Lignite Corporation, India that processes sub-bituminous and lignite coal.

2.2. Characterization of chromium contaminated soil and fly ash

The CCS and FA were characterized for pH as per ASTM method – D 4972 (ASTM) [26]. Chromium (VI) was determined following colorimetric Analysis – Method 7169A of USEPA [27] and total chromium by acid digestion followed colorimetric method (Method 3500 B, APHA) [28]. All tests were performed in triplicate and mean value of the results are reported. The SEM and chemical composition of the contaminated soil was analysed by Energy-dispersive X-ray spectroscopy (EDAX) [Model: Hitachi S- 3400N; Make: Germany]. Loss on ignition (LOI) tests was conducted by following ASTM method–D7348 [29].

2.3. Reduction studies for chromium contaminated soil

CaS₅ dosage of 3 times molar stoichiometric ratio of Cr(VI) was used based on the results of laboratory experiments for complete reduction of Cr(VI) to Cr(III) present in CCS [25]. The dosage was also ascertained based on the optimization studies conducted for remediation of COPR by Kameswari et al. [30]. Calcium polysulfide with 29% sulphide content was procured from M/s Classic Chemicals, Rajkot and a reaction time of 24 h was allowed for complete reduction. Chromium contaminated soil stabilized by CaS₅ is hence forth referred to as Chemically reduced chromium contaminated soil (CRCCS).

2.4. Details of mortar blocks preparation

The CCS was mixed well with FA in a pan and poured in 3 cm (diameter) × 5 cm (height) cylindrical moulds in 1:1, 1:2 and 1:3

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