

Efficiency of Washing Techniques for Removal of Heavy Metals from Industrial Sludge

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ABSTRACT: The effluent treatment plant sludge is one of the major sources of contamination with toxic metals. Since the sludge contains heavy metals, it must be pretreated to reduce the contamination. The heavy metals from the sludge can be reduced/separated by washing it with a suitable leaching solution. In the present study, the efficiencies of three leaching solutions to remove the contaminants from an industrial sludge were studied and the leaching process was modeled. The leaching solutions used are 0.1 N HCl, 0.1 N EDTA and 0.1 N FeCl₃. The efficiencies of leaching solutions were assessed by conducting column leaching experiments on the sludge and the migration rates of heavy metal ions of Cu, Zn, Ni, Cd, Pb, Fe and Cr present in the sludge were estimated. These migration rates are useful to estimate the quantity of leaching solution required at the site to achieve the required levels of concentrations in the sludge.

Keywords: Contaminants, Leaching solution, Dispersion coefficient, Retardation factor

INTRODUCTION

The enormous rate of increase in waste generation across the world is a serious threat to the future generation, if not handled properly. The disposal of liquid waste and sludges by industries has led to contamination problems for both the soil and groundwater. The treatment of industrial effluents as part of pollution control technologies results in the generation of large volumes of solid phase pollutants (sludge). The sludge is retained at the effluent treatment plant (ETP). ETP sludge creates a lot of environmental problems due to lack of disposal methods (Rajkumar & Hema, 2011). The sludges often contain toxic metals such as lead, chromium, cadmium,

nickel, and zinc. Unlike organic contaminants, most of the heavy metals do not undergo biodegradation and will remain in the ecosystem. These heavy metals cause serious threats to human health by direct contact with sludge, through the food chain (soil-plant-human) and by contaminating the ground and surface water resources (Gusiatin & Klimiuk, 2012; McLaughlin et al., 2000a; McLaughlin et al., 2000b; Ling et al., 2007; Wuana & Okieimen, 2011). The indiscriminate disposal of this ETP sludge leads to soil, groundwater and surface water pollutions. Due to the seriousness of this, the ETP sludge was listed as hazardous waste in India by the Ministry of Environment and Forests (MoEF, 2008). As the costs associated with transport of this large volume

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of sludge and construction of landfills are very high, recycling of this waste as a substitute for building materials (like bricks and low-cost concrete) is a better option (Balasubramanian et al., 2006; Baskar et al., 2006; Mary Lissy & Sreeja, 2014; Shivanath et al., 2015). Kakati et al. (2013) have studied the use of textile ETP sludge as a fertilizer at low concentration to enhance the plant growth. They found that 10% sludge plus 90% farmyard enhances the plant growth and stated that the accumulation of toxic materials in the plant system after the treatment should be studied in detail. Washing is the most widely used technique for flushing the contaminants from the sludge (Wood, 1997). Most metal ions in natural stage may not be easily soluble in water. Thus, dilute acids such as hydrochloric acid are used to enhance the solubility of metal ions. The commonly used chemical extractants are surfactants, solvents, acids, bases and chelating agents. Chemical extractants are the most cost-effective and less damaging (Gebreyesus, 2015). Biosurfactants and biologically produced surfactants enhance the removal of metals from contaminated soils and sediments (Mulligan et al., 2001). The commonly used chelating agents for washing include Ethylenediaminetetraacetic acid (EDTA), Nitritotriacetic acid (NTA), Diethylenetriaminepentaacetic acid (DTPA), Ethylenediaminedisuccinic acid (EDDS) and citric acid (CA) which form stable complexes with most of the heavy metals over a broad pH range (Bilgin & Tulun, 2015). EDTA has the ability to chelate with almost all heavy metals (Zou et al., 2009). Khalkhaliani et al. (2006) used various concentrations of EDTA for evaluating the washing technique on an artificially contaminated sandy loam soil.

Oustan et al. (2011) used two natural, low molecular weight organic acids, oxalic acid (OA) and acetic acid (AA) to decontaminate a highly contaminated calcareous soil. They concluded that this

method is efficient in the removal of contaminants such as Zn, Pb and Cd and also cost effective compared to other chelating agents. Ma et al. (2014) have used 0.01 N FeCl_3 in thermal treatment of mercury contaminated soil. Bilgin & Tulun (2016) have also used 0.01 N FeCl_3 to treat coarse grained and fine-grained soils which showed higher removal of zinc in the presence of iron. The adsorption capacity of cationic heavy metals increases with pH and this plays a primary role in adsorption. The concentration of sorbate and ferric hydroxide play a secondary role in adsorption of these heavy metal ions (Farley et al., 1985). Akahane et al. (2013) and Hirokazu et al. (2010) have used FeCl_3 to treat cadmium contaminated soils. The ferric salts have the capability to adsorb metal ions (both cations and anions) by co-precipitation. The precipitated ferric hydroxide facilitates incorporation of heavy metal ions directly into the precipitate matrix in place of the similarly sized and charged ferric cations (Patoczka et al., 1998). Although experimental conditions were favorable for the homogeneous distribution of metals throughout it is suggested that Cu and Zn are known to segregate within Fe oxides (Martinez & McBride, 1998). Gitipour et al., 2016 have studied soil washing technique on contaminated sludge with HCl and EDTA and found that the removal efficiencies of these solutions were in the range of 66.81% - 82.69%.

The objective of this study is to demonstrate the efficiency of washing technique to remove the heavy metals from the sludge using selected leaching solutions. The leaching solutions used for this study are 0.1 N HCl, 0.1 N EDTA and 0.1 N FeCl_3 . The column leaching tests were conducted with these leaching solutions. The advantage of column tests compared to the usual batch tests is that bulk density and the porosity of the sample are closer to field conditions (Susset &

Grathwohl, 2002). The column leaching tests are modeled, and the contaminant transport parameters are estimated which can be used to design a washing program at the site to treat the similar type of sludges.

MATERIALS AND METHODS

The industrial sludge was collected from KIADB Industrial Area, Doddaballapur, Bangalore, India (Fig.1). M/s Eco Green Solutions Systems (P) Ltd, which handled this sludge, is located in the villages of Doddaballapur Taluk, and Bangalore rural District. The area lies in the northern latitude of 12°59'03" N and eastern longitude of 77°35'16.2" E. Doddaballapur is about 40

km from Bangalore towards North on Bangalore-Hindupur state highway (SH-9).

In order to observe the chemical properties of industrial sludge, samples were collected from Eco Green Solutions Systems (P) Ltd. plant in the month of November 2013. The initial concentrations of various metal ions in the sludge were analysed with the standard methods given by USEPA 3050B (USEPA, 1996). The heavy metals present in the sludge and their chemical properties are shown in Table.1.

The wet sludge was air-dried and then sieved through 2 mm sieve to determine the index properties which are given in Table 2.

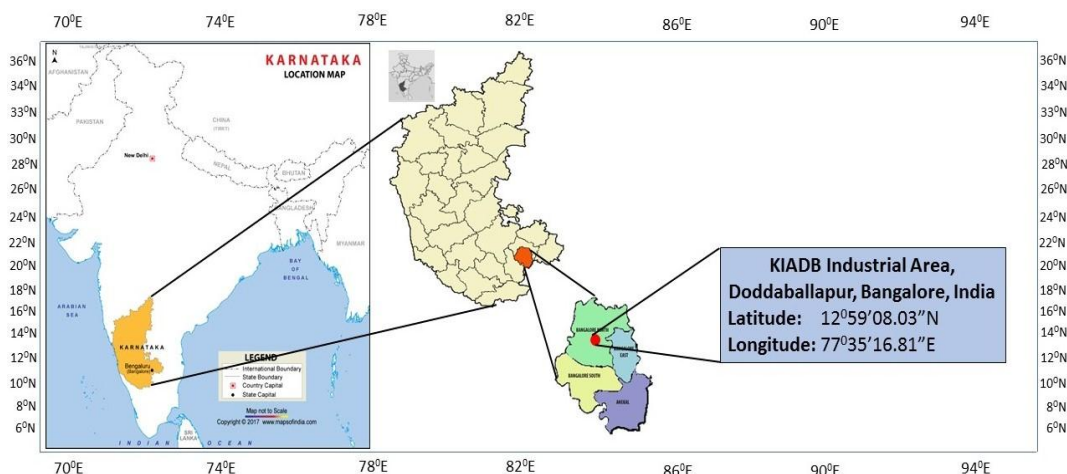


Fig. 1. Site location

Table 1. Chemical properties of industrial sludge

Pollutant	Quantity of metals in industrial sludge (mg/kg)
Copper (Cu)	92.3
Zinc (Zn)	152.7
Iron (Fe)	414.5
Chromium (Cr)	74.2
Cadmium (Cd)	4.7
Nickel (Ni)	52.3
Lead (Pb)	11.2

Table 2. Characteristics of industrial sludge

Specific Gravity (G)	Liquid Limit (%)	Plastic Limit (%)	Shrinkage Limit (%)	Plasticity Index (Ip)	Maximum dry Density (MDD), (g/cc)	Optimum moisture Content OMC, (%)
1.7	38	Non-plastic	9.80	-	0.98	33.50

The column leaching tests were conducted on the sludge samples (according to Rowe et al., 1988) to know the removal efficiencies of three leaching solutions, i.e. 0.1 N HCl, 0.1 N EDTA and 0.1 N FeCl₃. The samples were compacted to a density of 0.98 g/cc (MDD) and a water content of 33.5% (OMC). The void ratio and porosity of the test samples were found to be 0.82 and 0.45, respectively. The volume of voids (V_v) was estimated by multiplying the porosity (n) with total volume (V) of the sample which is called as pore volume (pore volume = $n.V$). The pore volume of the samples was estimated to be 56.55 cm³ and the average seepage velocities with a hydraulic gradient of 10 were estimated to be 5.2×10^{-5} , 5.5×10^{-4} and 1.2×10^{-4} with 0.1 N HCl, 0.1 N EDTA and 0.1 N FeCl₃, respectively. During the column leaching test, the seepage velocity increased with time due to the removal of contaminants and hence an average value of seepage velocity was taken for the analysis.

RESULTS AND DISCUSSION

From the effluent concentrations obtained from the column leaching tests, the mass of contaminant leached with number of pore volumes of flow was determined. The number of pore volumes is estimated from the cumulative volume of effluent leached by taking the ratio of cumulative volume of effluent to one pore volume of soil sample. The cumulative leaching mass ratio (LMRm) was estimated corresponding to

each time period by taking the ratio of cumulative mass of contaminant leached to the initial mass of contaminant present in the soil sample. The elution curves were prepared by taking number of pore volumes on x-axis and LMRm on y-axis. The elution curves are nothing but breakthrough curves in leaching which are useful to assess the efficiency of leaching solution and to select the suitable leaching solution for the soil washing treatment.

The experimental elution curves (number of pore volumes vs. LMRm) of the metal ions with different leaching solutions are given in Figs. 2 – 4.

As most of the metals are soluble in HCl, it has been used as a leaching solution to remove the pollutants from the sludge. From the Fig. 2, it can be observed that all the seven metals were leached out with this solution in different quantities. The metals Cd, Cu, Zn and Ni have almost reached a removal efficiency of 50% after passing around 25 pore volumes of the solution through the sludge. It can also be observed that after 25 pore volumes of flow, the leachability of metals was reduced, and the elution curves were almost flat for all the metal ions tested. The order of removal efficiencies achieved with 0.1N HCl is as given below.

Cd (64.5%) > Cu (57.9%) > Zn (52.3%) > Ni (45.3%) > Pb (28.5%) > Fe (22.6%) > Cr (18.3%)

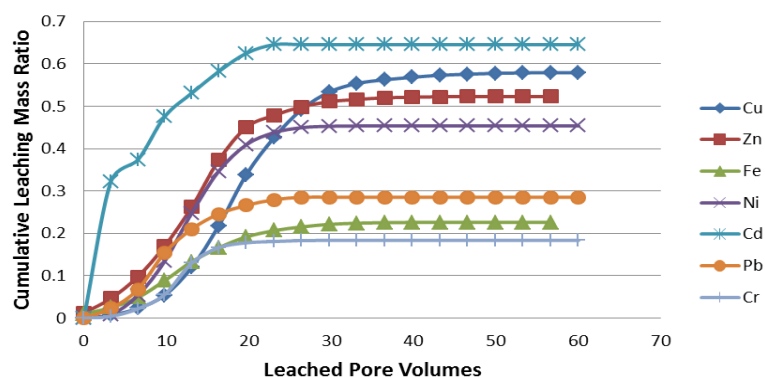


Fig. 2. Experimental elution curves with 0.1N HCL

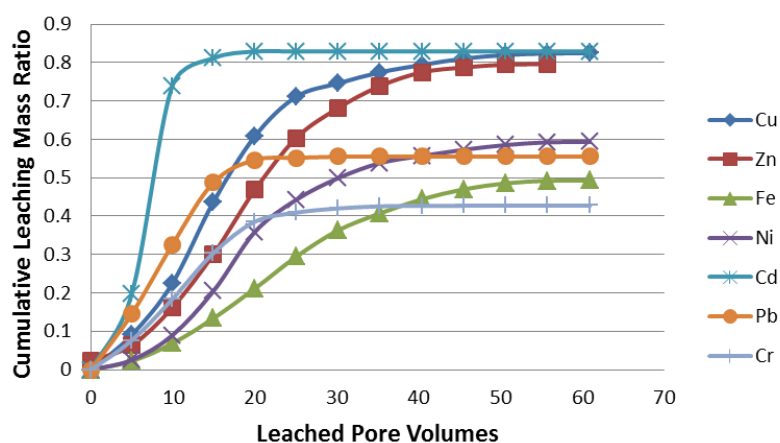


Fig. 3. Experimental elution curves with 0.1N EDTA

As most metal ions form stable complexes with EDTA, its efficiency in removing pollutants from ETP sludge was studied by passing 0.1N EDTA through this sludge. It can be observed from the elution curves of different metals (Fig.3) the curves were steep up to 20 pore volumes and were with a moderate slope up to 30 pore volumes followed by a flat portion. The metal ion Cd has reached its equilibrium much earlier compared to other metals at a pore volume of 10. The metal

ions Cd, Cu and Zn have attained a removal efficiency of 80% and other metals have attained almost 50%. Hence this solution can be used to reduce the levels of contaminants in the sludge. The order of removal efficiencies achieved with 0.1N EDTA is as given below.

Cd (82.9%) > Cu (82.5%) > Zn (79.6%) > Ni (59.3%) > Pb (55.5%) > Fe (49.3%) > Cr (42.7%)

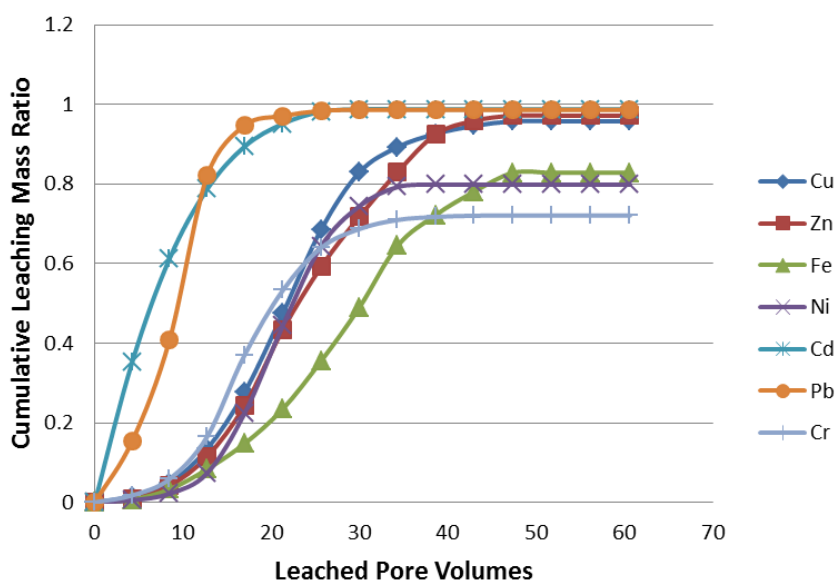


Fig.4. Experimental elution curves of different metal ions with 0.1N FeCl₃

In the present study, the ETP sludge was also treated with FeCl_3 to know the removal efficiencies of different metals from this sludge. The experimental elution curves of different metal ions are as shown in Fig 4. It can be observed from these curves that the removal efficiencies were much higher with this solution compared to all the leaching solutions used. All the metals have reached their equilibrium much earlier with this solution compared to that of 0.1N EDTA. The order of removal capacities for removing different ions using this leaching solution is as given below.

Cd (98.8%)>Pb (98.7%) >Zn (97.2%)>Cu (95.8%)>Fe (82.8%)>Ni (79.9%)>Cr (72.1%)

For a continuous point source of the contaminants leaching across the soil surface boundary for infinite duration, Ogata & Banks (1961) developed the analytical solution of Eq. 1. The governing linear partial differential equation for one-dimensional linear advection-dispersion-reactive process in vertical direction z for the unsteady state contaminant transport in a saturated homogeneous porous medium with uniform velocity field is given as:

$$R \frac{\partial C}{\partial t} = D_z \frac{\partial^2 C}{\partial z^2} - \bar{v}_z \frac{\partial C}{\partial z} \quad (1)$$

where R is the dimensionless retardation factor defined as the ratio of the mean velocity of non-adsorbed solute to that of the retarded solute, c is the concentration of contaminant in the medium (M/L^3), $\partial c / \partial t$ is the change in concentration of contaminant with time ($(\text{M/L}^3/\text{T})$), D_z is the longitudinal dispersion coefficient in z direction which takes into account both the molecular diffusion and the mechanical dispersion and is along the direction of flow and considered constant in this analysis, $\partial c / \partial z$ is the change in concentration of contaminant with depth i.e. concentration gradient ($\text{M/L}^3/\text{L}$), and \bar{v}_z is the average linear velocity in

direction z which was considered steady and uniform in time and space. They used initial condition of $c(z, t = 0) = 0$ for $z \geq 0$; and the Diriclet boundary conditions of $c(z = 0, t) = C_0$ for $t \geq 0$ and $c(z = \infty, t) = 0$ for $t \geq 0$ and solution was obtained as:

$$C(z,t) = \frac{C_0}{2} \left\{ \operatorname{erfc} \left(\frac{Rz - \bar{v}_z t}{2\sqrt{D_z R t}} \right) + \exp \left(\frac{\bar{v}_z z}{D_z} \right) \operatorname{erfc} \left(\frac{Rz + \bar{v}_z t}{2\sqrt{D_z R t}} \right) \right\} \quad (2)$$

where C_0 is the concentration of the contaminant at the soil surface i.e., the upstream boundary

The last boundary condition can only be appreciated mathematically. However, this is required to arrive at the analytical solution (Eq. 2). The $\operatorname{erfc}(u)$ is the complementary error function which is equal to $1 - \operatorname{erf}(u)$. For estimation of contaminant transport parameters, the method as reported by Rowe et al. (1988) was used. A computer program was prepared for the above equation using MATLAB v7 software tool to perform the iterations. The experimental values of the seepage velocity (\bar{v}_z), thickness of the sample (z), time periods (t) and the effluent concentrations along with the trial values of dispersion coefficient D_z and retardation factor (R) were incorporated into the computer program. The trial values of D and R were taken from the literature values (Almani et al., 2013). By running this program, the theoretical effluent concentrations were determined, and the plot was generated with the theoretical breakthrough curves. The theoretical effluent concentrations were compared with experimental values and the iterations were continued till the theoretical curve match with the experimental values. The transport parameters were obtained by matching the theoretical elution curves (with a set of assumed diffusion coefficients and retardation factors) with the experimental elution curves (i.e. breakthrough curves for leaching).

As the 0.1N FeCl_3 solution gave comparatively better results, the transport

parameters were determined with respect to this solution. The theoretical elution curves

obtained are shown in Fig 5 and the estimated transport parameters are given in Table 3.

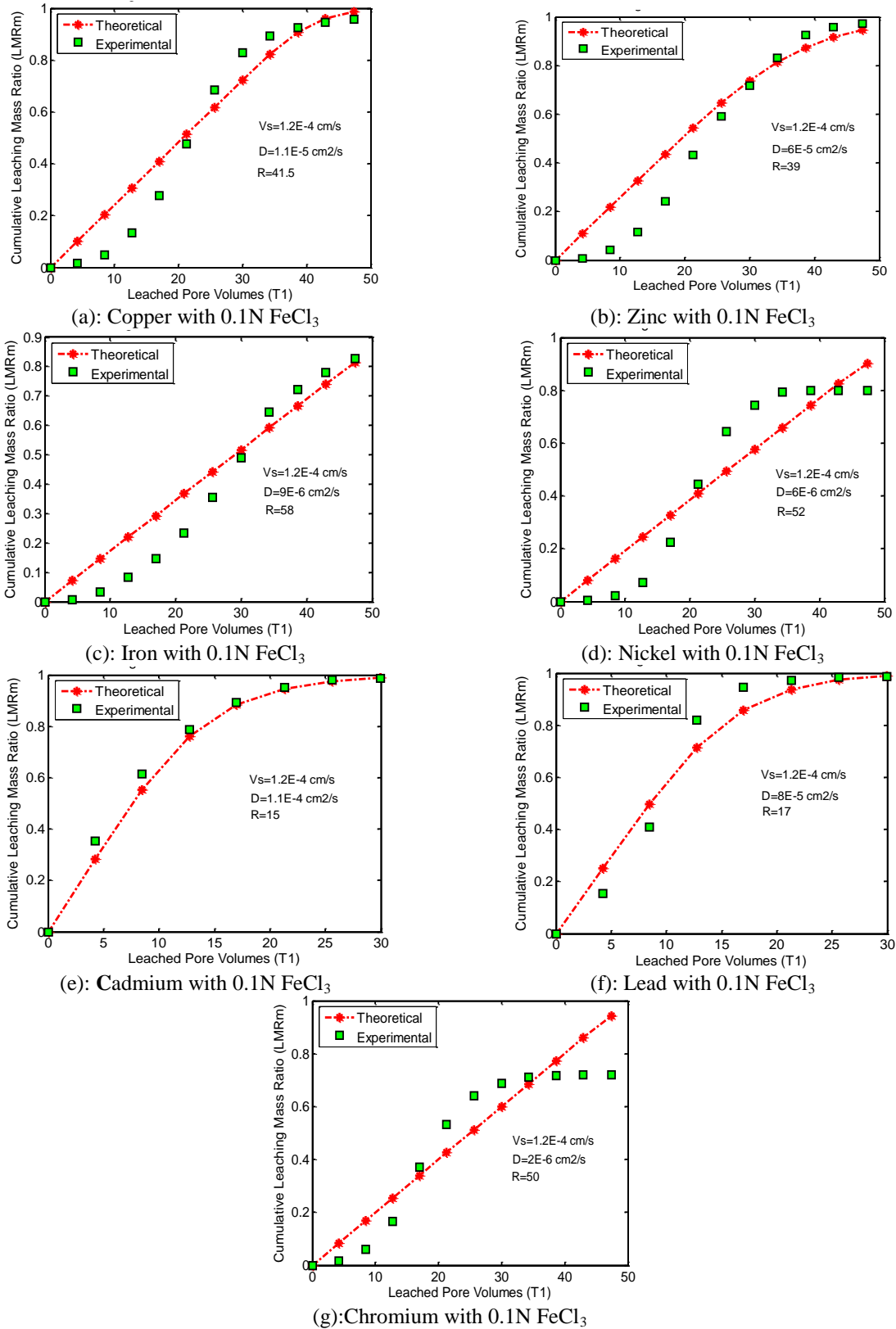
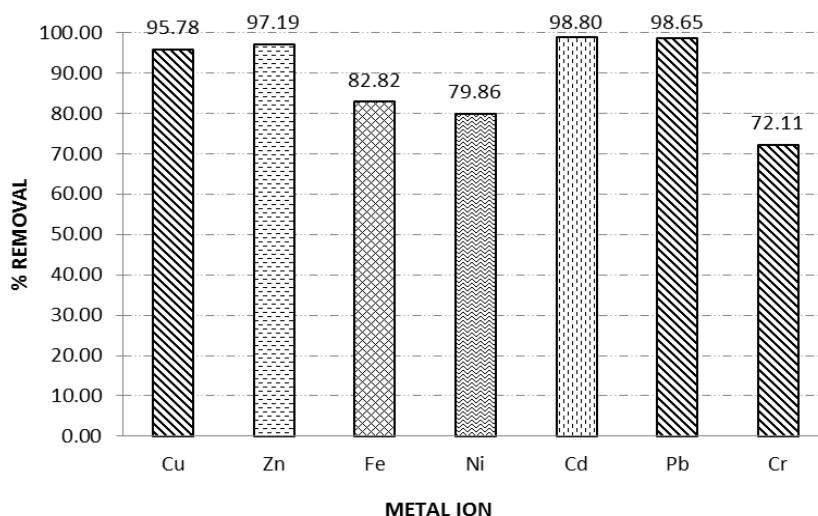


Fig. 5. Theoretical elution curves

Table 3. Contaminant transport parameters of different metal ions

Pollutant	With 0.1N FeCl ₃	
	Dispersion coefficient, D (cm ² /s)	Retardation Factor (R)
Copper (Cu)	1.1 x 10 ⁻⁵	41.5
Zinc (Zn)	6.0 x 10 ⁻⁵	39.0
Iron (Fe)	9.0 x 10 ⁻⁶	58.0
Chromium (Cr)	2.0 x 10 ⁻⁶	50.0
Cadmium (Cd)	1.1 x 10 ⁻⁴	15.0
Nickel (Ni)	6.0 x 10 ⁻⁶	52.0
Lead (Pb)	8.0 x 10 ⁻⁵	17.0

**Fig. 6. Removal efficiencies of 0.1N FeCl₃ for different metal ions**

The dispersion coefficients(D) of different metals were observed to be in the range of 2.0×10^{-6} - 1.1×10^{-4} cm²/s and retardation factors(R) were observed to be in the range of 15 – 58. The contaminant transport parameters obtained can be substituted in the contaminant transport equation (Eq. 2) to estimate the time required for reducing the contaminant concentrations to the required level. From the time estimated (t), the quantity of leaching solution required (Q) at the site can be calculated by multiplying the time with cross sectional area and velocity of soil. The percent removal of metal ions with 0.1N FeCl₃ have been compared and presented in Fig. 6. From this Fig., it is observed that the metal ions Cu, Zn, Cd and Pb have attained more than 95% removal efficiency. The highest removal

efficiencies of Fe, Ni and Cr were observed to be 82.8, 79.8 and 72.1%, respectively.

CONCLUSIONS

The contaminants in the sludge can be removed or reduced by washing it with suitable leaching solutions. From the column leaching tests conducted on the sludge it was found that 0.1N FeCl₃ solution is the most efficient leaching fluid to remove the metal ions Cu, Zn, Fe, Ni, Cd, Pb and Cr present in the sludge.

The percent removals obtained with the leaching solutions used are as follows.

With 0.1 N HCl:

Cd (64.5%)>Cu (57.9%)>Zn (52.3%)>Ni (45.3%)>Pb (28.5%) > e (22.6%) > Cr (18.3%)

With 0.1 N EDTA:

Cd (82.9%)>Cu (82.5%)>Zn (79.6%)> Ni (59.3%)>Pb (55.5%)>Fe (49.3%)> Cr (42.7%)

With 0.1 N FeCl₃:

Cd (98.8%)>Pb (98.7%) >Zn (97.2%)>Cu (95.8%)>Fe (82.8%)>Ni (79.9%)>Cr (72.1%)

The dispersion coefficients of different metals were observed in the range of 2.0×10^{-6} - 1.1×10^{-4} cm²/s and retardation factors were observed in the range of 15 – 58.

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