

## **Remediation Modeling of Metal Contaminated Water and Soil Using Water Soluble Polymers – An Overview**

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**Abstract:** *The toxic heavy metal ions are non-degradable and are persistent in the environment. Therefore, the elimination of heavy metal ions from wastewater is important to protect public health. Vegetables grown in such lands, therefore, are likely to be contaminated with heavy metals and unsafe for consumption. Polymer enhanced Ultra filtration technique offers a promising option for the heavy metal contaminated soils and water. Polymer - metal ions interactions are a subject of great interest as they have potential analytical and technological applications in fields like environmental science, industrial effluent treatment, metallurgy industry, etc., as well as their analogous implications in biological systems. With increasing industrialization, heavy metal ions undergo several technical processes and finally pass by way of effluent, sewage, dumps and dust, into the water, the earth and the air and thus into the food chain as finely dispersed and/or in solutions. Functionalized polymers with higher retention capacity and stability, offered great application prospects in heavy metal waste water and soil treatment. Moreover it is an economically viable option*

**Keywords:** *Water Soluble Polymers (WSP); Polymer Assisted Ultra Filtration (PAUF); liquid-phase polymer-based retention (LPR) technique; Remediation, green approach*

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### **1. INTRODUCTION**

Heavy metals e.g., Pb, Cd and Zn constitute an important and significant class of soil and water contaminants originating from industrial effluents, agricultural wastes, agrochemicals and fossil fuels. Metals are non-biodegradable, recirculating in the environment and have long residence time. The hazard imposed by these trace element contaminants is dependent on their ability to migrate from soil systems into water systems and their biological uptake. This becomes possible due to existence of metal in various forms e.g., soluble, adsorbed and chelated with different components of soil. It is the ability of these metal ions from solid phase to soil solution that governs their mobility and bioavailability. A novel approach of ‘chemoremediation’ using immobilized linear polymer chains permits an extremely strong metal binding. Here cleanup is done in aqueous homogeneous phases of water and soil system using polymer reagents as a green technology with ‘Liquid Phase Retention’. The hydrophilic polymers ‘polychelatogens’ should be non-cross linked, with complex-forming moieties, capable to bind, enrich, and separate metal ions from aqueous solutions. These polymers provide strong binding to the metal ion in water and soil matrix, with a minimum free energy. These metal – polymer bonds can be cleaved by altering the intra molecular interactions (e.g., H-binding, electrostatic attractions) using simple pH adjustments, thus providing easy and quantitative metal recovery.

Metal selectivity can be improved by selecting the side chain functionality, metal selectivity can be enhanced. Designing of poly (methacrylic acid) with side chain functionalities and novel co-polymers (with methacrylic acid, acrylamide, and ethylene imine as monomers) and further investigation of their improved remediation abilities opens several possibilities. The effectiveness of remediation can be tested by studying the leachate chemistry using analytical methods like Atomic Absorption Spectroscopy, Elemental analysis and Infrared Spectroscopy. The ‘polychelatogens’ can be regenerated for reuse. It is not only important for high cost materials, but also for inexpensive systems as it reduces waste disposal costs and limits the environmental

impact. The recovered metal can also be reused after purification. One can carry out chemical regeneration as it is simplest and economic. Polyelectrolytes containing acidic and basic groups can be regenerated by protolysis i.e., changing the pH of the solution in order to cleave the polymer-metal bond. Batch and column studies can be undertaken to check the effectiveness. The bioavailability of the metal ions can be checked using tests e.g., TCLP and PBET

Effective remediation and clean-up of polluted soils and water needs a thorough understanding of the mechanism(s) by which toxic heavy metals interact with these remediating agents. Further very limited information is available about the interactions of soil components with metals and remediating polymers, we propose to carry out kinetic and thermodynamic studies of these interactions. Research on providing basic insight information of the remediation designing based on binding of some selected heavy metals in water and on to soil/clay mineral surfaces should be carried out.

In order to reduce the environmental footprints of industrial and other agrochemical activities, the designing of remediation technologies using green amendments are really significant. The kinetic and thermodynamic studies of these interactions throw light on the selection, effectiveness and longtime feasibility of these remediation technologies.

## 2. INTERNATIONAL SCENARIO

It is worth noting how the soil as a resource is poorly cared for under international conventions, in spite of its importance. Soil contamination by heavy metals is increasing. Soil pollution with toxic metals has become an important social and environmental challenge. When the polluted area is large and the site poses little immediate risk to health or environment, *in situ* remediation technologies can be envisaged.

Rivas B.L. *et al* (2012) the retention properties of commercial chelating water-soluble polymers, for different metal ions in aqueous solution using a liquid-phase polymer-based retention (LPR) technique. The polymers studied were poly(ethyleneimine) or P(EI) (water-free and a 50 % aqueous solution) and poly(ethyleneimine epichlorohydrin) or P(EIE) (a 17 % aqueous solution). These commercial polymers were fractionated by ultra filtration membranes and then characterized by Fourier-transformed infrared spectroscopy. The extraction process was performed using the following metal ions:  $\text{Cu}^{2+}$ ,  $\text{Cd}^{2+}$ ,  $\text{Co}^{2+}$ ,  $\text{Ni}^{2+}$ ,  $\text{Zn}^{2+}$ ,  $\text{Pb}^{2+}$  and  $\text{Cr}^{3+}$ . In the washing studies, we varied the pH (3, 5 and 7) and retention time. The results showed that P(EI) showed high retention for all the metal ions at pH 7 and for selective retention of  $\text{Cu}^{2+}$  at pH 5, while P(EIE) showed selective retention of  $\text{Cu}^{2+}$  ions at pH 7. Using the enrichment method, the maximum retention capacity of  $\text{Cu}^{2+}$  and  $\text{Cd}^{2+}$  was achieved using a 50 % aqueous solution of P(EI) at pH 5 and 7, respectively. Finally, charge-discharge experiments for  $\text{Cu}^{2+}$  were analysed by changing the pH from basic to acidic over three cycles. These results showed that it is possible to remove metal ions and regenerate the removal capacity of the polychelators using the LPR technique.

Adsorption-desorption of copper ( $\text{Cu}^{2+}$ ) at contaminated levels in two red soils was investigated by Yu *et al* (2002). The red soil derived from the Quaternary red earths (clayey, kaolinitic thermic plinthite Aquult) (REQ) adsorbed more  $\text{Cu}^{2+}$  than the red soil developed on the Arenaceous rock (clayey, mixed siliceous thermic typic Dystrochrept) (RAR). The maximum adsorption values ( $M_A$ ) that are obtained from the simple Langmuir model were 25.90 and 20.17 mmol  $\text{Cu}^{2+}$   $\text{kg}^{-1}$  soil, respectively, for REQ and RAR. Adsorption of  $\text{Cu}^{2+}$  decreased soil pH, by 0.8 unit for the REQ soil and 0.6 unit for the RAR soil at the highest loadings. The number of protons released per  $\text{Cu}^{2+}$  adsorbed increased sigmoidally with increasing initial  $\text{Cu}^{2+}$  concentration for the RAR soil, but the relationship was almost linear for the REQ soil. The RAR soil released about 2.57 moles of proton per mole of  $\text{Cu}^{2+}$  adsorbed at the highest  $\text{Cu}^{2+}$  loading and the corresponding value for the REQ soil was 1.12. The distribution coefficient ( $K_d$ ) decreased exponentially with increasing  $\text{Cu}^{2+}$  loading. Most of the adsorbed  $\text{Cu}^{2+}$  in the soils was readily desorbed in the  $\text{NH}_4\text{Ac}$ . After five successive extractions with 1 mol  $\text{L}^{-1}$   $\text{NH}_4\text{Ac}$  (pH 5.0), 61 to 95% of the total adsorbed  $\text{Cu}^{2+}$  in the RAR soil was desorbed and the corresponding value for the REQ soil was 85 to 92%, indicating that the RAR soil had a greater affinity for  $\text{Cu}^{2+}$  than the REQ soil at low levels of adsorbed  $\text{Cu}^{2+}$ .

Lindim *et al* (2001) used a polyacrylate polymer to immobilize cadmium (Cd) in a sandy soil artificially contaminated with the metal. Growth of perennial ryegrass in pots with 9.5 kg of soil was stimulated in polymer-amended soil. Even in pots with the highest levels of Cd, growth was much less impaired than in pots without polymer. Shoot Cd concentrations were smaller in the plants cultivated in the amended soil. Because water soluble Cd was considerably reduced in the contaminated polymer-amended soil, the effect of the polymer on plant growth was attributed to the decrease of Cd concentration in soil solution.

The biosorption process for removal of the heavy metal Cd<sup>2+</sup> from contaminated soil was chosen by Lin *et al* (2005) due to its economy, commercial applications, and because it acts without destroying soil structure. The study is divided into four parts (1) soil leaching; (2) adsorption Cd<sup>2+</sup> tests of immobilized *Saccharomyces pombe* beads and polyvinyl alcohol (PVAL) were added to alginate (10 wt.%) and then blended or cross-linked by epichlorohydrin (ECH) to increase their mechanical strength; (3) regeneration tests of the optimum beads; and (4) adsorption model/kinetic model/thermodynamic property: the equilibrium adsorption, kinetics, change in Gibbs free energy of adsorption of Cd<sup>2+</sup> on optimum beads were also investigated.

Pure water is an essential requirement for the survival of human beings. To meet the requirements of potable, industrial and agricultural water, the immediate need is to treat waste water, particularly the sewage sludges and slimes from the municipal and industrial effluents respectively. These effluents are highly undesirable and unsafe. The removal of contaminants from waste water is a must before they can be reused. Tripathy *et al*(2006) discussed flocculation in detail.

The fundamentals and novel developments of water-based remediation processes for metals have been investigated and assessed (Green Chemical Reactions 2008). Recent developments in the area of metal remediation in connection with ultra filtration are the main focus. In addition, the LPR method is explained and highlighted in view of the application in different areas.

Work done by Luster *et al* (2009) needs a special reference as it seeks to develop controlled release biodegradable polymers that release chemical oxidants at controlled or sustained rates to extend the longevity of treatments for water and soil. This work explored the ability to encapsulate chemical oxidants in biodegradable polymers for environmental remediation. Several polymer/oxidant formulations were tested by combining hydrophobic and hydrophilic polymers to create pellets with various release rates for potassium permanganate. Potassium permanganate is a crystalline chemical oxidant that is currently dissolved in water for use in drinking water treatment and in the remediation of chlorinated solvents. This technology provides the potential for new methods for chemical remediation using biodegradable polymers including creation of subsurface permeable reactive barriers, wells packed with oxidation pellets, or reactors filled with pellets for pump and treat systems.

Birnbaum *et al* (2003) investigated water-soluble dendrimers and polymers as ligands for selective anion binding. PAMAM Starburst dendrimers and polyethylenimine were chemically modified to incorporate new ligands or hydrogen bonding groups at the primary amines. These molecules, along with the unmodified parent compounds, bind arsenate, chromate, and phosphate, even in the presence of competing anions such as chloride. Such systems could potentially be employed for remediation of contaminated water and soils using well-established ultrafiltration technology.

A water-soluble polymer, poly (2-acrylamido glycolic acid) was obtained by Rivas *et al* (2007) by radical polymerization and characterized by FT-IR, 1H NMR, and 13C NMR spectroscopy. The metal ion retention properties were investigated through the liquid-phase polymer based retention (LPR) technique at different pHs and filtration factor Z. The affinity of the ligand groups for the metal ions depends strongly on the pH. At lower pH, the retention is lower than 50%, which increased as the pH increased.

The resin poly(acrylamide-co-methacrylic acid) P(AAm-co-MA) by radical polymerization was also synthesized by Rivas *et al* (2010) and its metal ion binding was studied using the batch and column equilibrium procedures for: Cd(II), Zn(II), Pb(II), Hg(II), Al(III), and Cr(III). Experiments were carried out at different pH, metal ion concentration, temperature, and contact

time. The metal-ion retention properties were dependent on the polymer/metal ion ratio. Under competitive conditions of Pb(II), Hg(II), and Al(III), the resin showed a higher retention of Hg(II), allowing the selective separation of this metal.

Geckeler *et al* (2001) used heterogeneous methods for the separation of inorganic ions contained in natural waters, industrial fluids, or dissolved solid materials. In contrast to homogeneous reactions, these two phase distributions or heterogeneous reactions with diffusion-controlled processes have disadvantages, such as long contact times. Two-phase systems can be avoided by using separation methods based on pressure-driven membrane processes in the aqueous homogeneous phase.

Mocioi *et al* (2007) obtained New polymeric structures by chemical transformations of maleic anhydride/dicyclopentadiene copolymer with triethylenetetraamine, p-aminobenzoic acid, and p-aminophenylacetic acid were used for the removal Cu (II) ions from aqueous solutions. The experimental values prove the importance of the chelator nature and of the macromolecular chain geometry for the retention efficiency. The retention efficiency ( $\eta(r)$ ), the retention capacity ( $Q(e)$ ), and the distribution coefficient of the metal ion into the polymer matrix ( $K-d$ ) are realized by evaluation of residual Cu(II) ions in the effluent waters, by atomic adsorption. Also are discussed the influence of pH, the thermal stability of the polymer, and their polymer-metal complex, as well as the particular aspects regarding the contact procedure and the batch time. Based on the polymers and polymer-metal complexes characterization a potential retention mechanism is proposed. All polymer supports as well their metal-complexes are characterized by ATD and FTIR measurements.

Li *et al* (2008) focused on the effects of pH and organic ligands, namely ethylenediaminetetraacetic acid (EDTA), nitrilotriacetic acid (NTA), and citric acids, on the removal and recovery of Cd(II) in polyelectrolyte enhanced ultra filtration (PEUF). Polyethylenimine (PEI), which can bind with both positively charged metal ions by coordination bonding and negatively charged ligand-metal complexes by charge attraction, was employed as a chelating polymer. The removal and recovery of Cd species was greatly dependent on the chemistry of organic ligands according to solution pH, particularly being related to the distribution of Cd-ligand complexes at different pH levels.

### 3. NATIONAL SCENARIO

Rapid urbanization and industrialization created a great pressure on delicate ecosystem in and around urban areas. Aerial deposition and/or indiscriminate disposal of wastes from a variety of industries result in the contamination of urban soils with metals (As, Cr, Cu, Hg, Pb, Zn etc.). Persistence of these metals in soil and water poses a serious risk to environmental and human health. Information on metal distribution in urban environment may help to develop appropriate strategies for managing the uncontrolled dumping of industrial wastes and creating safer environment for urban populations. In this context, the metal contamination in Coimbatore urban environment, which is the second largest industrial city in Tamil Nadu, India was examined. Apart from textiles, hundreds of small scale industries like dyeing, foundry, metallurgy, smelting and chemicals are in operation in and around the city. Geographical Information System (GIS) was used to identify the 'hot spot' of the metal contamination. Through a laboratory closed incubation experiment the biotransformation of lead (Pb), a dominant metal in the urban soil, was examined to predict its possible groundwater contamination. (Singaram 2006)

The relative proportion of metal distribution in Coimbatore urban soils followed: Pb>Cr>Zn>Cu>Cd. The contamination of Pb and Cr was widespread in the urban soils and in many locations the concentrations exceeded the maximum permissible limits. Metal contamination in urban water showed greater variability as marked difference was observed between samples. Though the soils have shown varied degrees of metal contamination, ground waters remained relatively uncontaminated, possibly be due to the geological formation existing in the urban environment that would have effectively prevented the leaching of metals. However, water samples from ponds, ditches and drainage channels have had high concentration of metals which followed: Cr>Pb>Zn>Cu>Cd and the concentrations mostly exceeded the maximum permissible limit of the WHO. Joshi *et al* (1996) used sand to remove Arsenic from ground water.

Heavy metals are found in their different forms in the environment. Because of the severity of heavy metal contamination and potential adverse health impact on the public, a tremendous effort has taken place to purify waters containing toxic metal ions. Traditional methods which have been employed prove to be costly and prohibitive for low level waste remediation. The distribution, mobility, and toxicity of metals are strongly related to these different forms. This necessitates the exploration of different methods for the remediation and speciation of heavy metals. Some direct and indirect physico-chemical methods such as filtration, chemical precipitation, ion-exchange, electro deposition, and membrane systems have been used for the last four decades. However, it is only in last few years that reliable biological methods have also been used. The biological methods include the use of microorganisms (fungi, algae, bacteria), plants (live or dead) and biopolymers. The use of these methods for the speciation of heavy metals was reviewed by Singh *et al* (2009)

Biosorption is presented as an alternative to traditional physicochemical means for removing toxic metals from ground and wastewaters. Most recently, plant based biomaterials have been of interest. The bark of *Hemidesmus indicus*, an extensively available plant biomass commonly called as Indian sarsaparilla was used as biomaterial by Chandra Sekhar *et al* (2004) for removal of lead from aqueous streams. The effectiveness of the IPBFIX for biosorption of lead ions was demonstrated using the wastewater samples emanating from a non-ferrous metal industry. The results from these studies will be useful for a novel phytofiltration technology to remove and recover lead from wastewaters and this can also be well adapted for secondary treatment or polishing of wastewaters. An attempt was made to remove lead from the lead polluted waters (both ground and surface) from an industrially contaminated sites.

Juwarkar *et al* (2007) focused on column experiments conducted to evaluate the potential of environmentally compatible rhamnolipid biosurfactant produced by *Pseudomonas aeruginosa* strain BS2 to remove heavy metals (Cd and Pb) from artificially contaminated soil. Results showed that di-rhamnolipid removes not only the leachable or available fraction of Cd and Pb but also the bound metals as compared to tap water which removed the mobile fraction only. This study showed that biosurfactant technology can be an effective and nondestructive method for bioremediation of cadmium and lead contaminated soil.

Sharma *et al* (2012a, 2012b) used this technique to remediate polluted water samples. Remediation upto 98% could be achieved. Sharma *et al* (2009a, 2009b, 2009c, 2010) used green amendments e.g., Rock Phosphate (RP) and Coal Fly Ash (CFA) to remediate heavy metal contaminated soils of Delhi. This research work focused on the batch, bench and column scale demonstration of metal immobilization in contaminated soils using green amendments like Rock Phosphate (RP) and Coal Fly Ash (CFA). The selected site is located at urban industrial area of NCR of Delhi and was contaminated with heavy metals like Lead (Pb), Cadmium (Cd) and Zinc (Zn) primarily due to industrial activities, battery recycling activities etc. The soil is characterized by alkaline pH as is typical of Delhi soil and high concentration of organic matter.

Lab research activities included screening of phosphorous sources and optimization of their application rates. Leachability of metals in treated soil was measured by using a combination of batch, bench and column tests to evaluate the immobilization capability of applied treatments. Various phosphate amendments evaluated, a mixture of  $\text{Ca}(\text{H}_2\text{PO}_4)_2$ ,  $\text{CaCl}_2$  and  $\text{H}_3\text{PO}_4$  For efficient immobilization, pH reduction with  $\text{H}_3\text{PO}_4$  became necessary to dissolve carbonate-bound metals and make them readily available for the geochemically stable metal phosphate formation.

The effectiveness of rock phosphate for immobilizing heavy metals (Pb, Cd, and Zn) from aqueous solutions was also investigated. The amounts of metals sorbed onto phosphatic clay decreased in the order  $\text{Pb} > \text{Cd} > \text{Zn}$ .

#### 4. CONCLUSIONS

Although current chelating polymers are extremely effective in remediation efforts, there are significant drawbacks in these existing polymer systems. The conventional insoluble polymers often have low surface area to volume ratios; thus are often not as effective remediation agents as their water soluble counterparts. In addition, many of the polymers are ineffective at acidic pHs; therefore, would be not be able to remediate the majority of industrial wastewater streams. In

these studies, we will try to remove these drawbacks by designing novel polymers with (i) reactive polymer blocks and (ii) inducting reactive moieties on the polymer chains. Another important feature is the regeneration of the polymer by simple pH adjustment. This offers a cost effective way of reusing the polymers.

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