REMEDIATION TECHNOLOGIES FOR HEAVY METALS CONTAMINATED SOIL

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ABSTRACT

Mining, manufacturing, and the use of synthetic products (e.g. pesticides, paints, batteries, industrial waste, and land application of industrial or domestic sludge) can result in heavy metal contamination of urban and agricultural soils. The environmental safety of soil has become severe with the boost of industrialization and urbanization. In this paper, on the basis of investigating the status of soil contamination, the remediation technologies of soil contaminated by heavy metals, including physical remediation, chemical remediation and biological remediation were focused. The mechanisms of remediation, strengths and drawbacks, developing trend were reviewed in order to supply reference to the study in this field.

KEYWORDS: Heavy Metals, Contaminated Soil, Remediation, Ecosystem, Bio Toxicity

INTRODUCTION

In ecosystem, soil is considered as basic environmental elements and also the important material basis for human being survival and development. But this soil also become a sink for contaminants entering the environment, independently of if the contamination derives from air pollution, water pollution or soil pollution itself. This soil Contamination is caused due to various agricultural and industrial activities e.g. agriculture use of fertilizers and pesticides, fossil fuel combustion, mining waste, and landfill leaching. Potential harmful contaminants have been accumulated in the upper soil during thousands of years, starting from the mining for hematite and later for copper. In terms of their mobility and bio toxicity on living ecosystem, removing metals from soil is an essential task. Soil contamination has become a serious problem with the economy and industry development. Heavy metal soil contamination is more serious than any other soil contamination. Soils may become contaminated by the accumulation of heavy metals and metalloids through emissions from the rapidly expanding industrial areas, mine tailings, disposal of high metal wastes, leaded gasoline and paints, land application of fertilizers, animal manures, sewage sludge, pesticides, wastewater irrigation, coal combustion residues, spillage of petrochemicals, and atmospheric deposition [1, 2]. Heavy metals are toxic to all organisms if present in high concentrations. The concentrations of heavy metals in soils can vary widely, even in uncontaminated soils. Marked differences in the geochemical composition of the rocks which form the parent materials of soils and variations in the intensity of soil-forming processes can result in wide ranges of total and available concentrations of most elements in soils, even in those unaffected by contamination. Nevertheless, contamination from many sources can often give rise to some very high concentrations of heavy metals which can cause toxicity in soil organisms and susceptible plants, but this depends on the factors affecting the bioavailability of the elements.

The European countries have invested a lot to remediate the contaminated soils [3]. In the 1980s, the U.S. Congress has passed the Comprehensive Environmental Response, Compensation & Liability Act (CERCLA), namely superfund program, in order to protect the human health and remediate the environmental pollution. There are many other laws and regulations, such as The Resource Conservation and Recovery Act (RCRA), Superfund Amendments and Reauthorization Act (SARA), emphasize the standard and behavior of soil remediation. From 1982 to 2002 year, the area of remediated land has reached 18.35 million m_3. The Britain also passed Environmental Protection Act in the 1990s and in which the second part clearly stated that the principle of polluter responsibility. As compared to the developed countries, the investment and research in the remediation of contaminated soil was not far enough. The prevention of heavy metals contaminated soil is not only needed to control the sources, but also enhance the remediation of contaminated soil [4].

1. Physical remediation

The physical remediation mainly includes soil replacement method and thermal desorption. The soil replacement is to clean soil to restore or partly restore the contaminated soil in order to dilute the pollutant concentration, increase the soil environmental capacity, and thus remediate the soil [5, 6]. The soil replacement is also divided into three types, including soil replacement, soil spading and new soil importing. (1) Soil replacement is removing the contaminated soil and putting into new soil. This method is suitable for contaminated soil with small area. Besides, the replaced soil should be treated possibly, or else it will incur the second pollution. (2) Soil spading is deeply digging the contaminated soil, to spread the pollutant into the deep sites and achieving the aim of diluting and naturally degrading. (3) New soil importing is adding lots of clean soil into the contaminated soil, covering it at the surface or mixing to make the pollutant concentration decreasing. The soil replacement can effectively isolate the soil and ecosystem and thus decrease its effect on environment. However, this technology is suitable for soil with small area and polluted severely and is large in working volume, costs a lot [7]. The thermal desorption is on the basis of pollutant's volatility and heat the contaminated soil using steam, microwave, infrared radiation to make the pollutant (e.g. Hg, As) volatile. The volatile heavy metals are then collected using the vacuum negative pressure or carrier gas and attain the aim of removing the heavy metals [8-13]. According to the temperatures, the traditional thermal desorption can be classified into high temperature desorption (320~560°C) and low temperature desorption (90~320°C). This technology has advantages of simple process, devices with mobility and the remediated soil being reused. A company of mercury collection and service in USA has used this technology for in-situ remediation and developed commercial service. However, the limited factors, such as the expensive devices, long desorption time, limit its application in the soil remediation [14].

2. Chemical remediation

2.1 Chemical leaching

Chemical leaching is washing the contaminated soil by using fresh water, reagents, and others fluids or gas [15, 16] that can leach the pollutant from the soil. Through the ions exchange, precipitation, adsorption and chelation, the heavy metals in soil was transferred from soil to liquid phase, and then recovered from the leachate. The leachate using mainly include inorganic eluent, chelation agents, and surfactant, etc. Tokunage and Hakuta [15] investigated the effects of different concentrations of hydrogen fluoride, phosphoric acid, sulfuric acid, hydrogen chloride, nitric acid on As extraction from artificial polluted soil (As 2830mg/kg). It was found that phosphoric acid proved to be most promising as an extractant, attaining 99.9% arsenic extraction at 9.4% acid concentration in 6 h. Sulfuric acid also attained high percentage extraction. For the removal of arsenic from contaminated soil an environment-friendly and cost-effective extraction method has been studied [16-18]. A yellow-brown forest soil was contaminated with arsenic (V) and used as a model soil. Potassium phosphate was most effective in extracting arsenic among various potassium and sodium salts. Arsenic was proficiently extracted by phosphate solution of pH 6.0 at 300 mM phosphate concentration and at 40°C. Among the extractant, the EDTA can form stable composite with most heavy metals in the wide pH valve range. A soil washing process was applied to remediate arsenic (As)-contaminated stream sediments around an abandoned mine in Goro, Korea [19]. Removal efficiencies for fine sediments were >95% after 1 h of washing with 0.2 M citric acid. When 0.2 M citric acid was mixed with 0.1 M potassium phosphate, the As removal efficiency increased to 100%. It is well-known that, the effect is almost substandard when single extractant is used as the many different pollutants in soil. This let us join or successively use many different extractants. The results showed that, Na_2EDTA solutions were generally more efficient than $Na_2S_2O_5$ for removing heavy metals from the soil samples. Na_2EDTA preferentially extracted lead over zinc and cadmium but exhibited little impact on chromium removal. Cadmium and, especially zinc, removal by a 0.01 M Na_2EDTA solution were enhanced considerably by inclusion of 0.1 M $Na_2S_2O_5$, suggesting that a mixture of the two reagents may provide an economically optimum solution for certain contaminated soils [20]. Ehsan et al [21] evaluated the efficiency of a washing process with cyclodextrin together with EDTA for the simultaneous mobilization of heavy metals and PCBs from a field contaminated soil. These studies confirmed that PCB compounds and selected heavy metals can be coextracted efficiently from soil with three successive washes with the same washing suspension having EDTA and cyclodextrin. However, the chelation agents like EDTA is expensive and the biological degradability is bad. Thus, in order to promote the biological degradability of extractants and reduce the risk of second pollution, biological reagent was used to leach the heavy metals in soil. Biodegradable, synthetic organic chelate ethylenediaminedisuccinic acid (EDDS) was used for washing of soil contaminated with 1350 mg/kg of Pb [22]. Hong et al [23] evaluated the efficiency of saponin on remediating heavy metal contaminated soils. Three different types of soils (Andosol, Cambisol, Regosol) were washed with saponin in batch experiments. Utilization of saponin was effective for removal of heavy metals from soils, attaining 90-100% of Cd and 85-98% of Zn extractions. Li et al [22] also studied the efficiency of tea saponin on metal removal. The results showed that, the removal of Pb, Cd, Zn and Cu were 6.74%, 42.38%, 13.07% and 8.75%, respectively when using 7wt% tea saponin as the extractant. The tea saponin can effectively remove acid soluble and reductive metals, which will greatly reduce the environmental risk.

2.2 Chemical fixation

Chemical fixation is to add reagents or materials into the contaminated soil and to use them with heavy metals to form insoluble or hardly movable, low toxic matters, thus decreasing the migration of heavy metals to water, plant and other environmental media and to achieve the remediation of soil [5]. The soil conditioning materials used include clays, metallic oxides, biomaterials, etc. Hodson et al [23] evaluated the ability of bone meal additions (finely ground, poorly crystalline apatite, $Ca_{10}(PO_4)_6OH_2$) to immobilize pollutant metals in soils and reduce metal bioavailability through the formation of metal phosphates has been evaluated. Batch experiments and subsequent extraction of metals from controls and bone meal amended soils using 0.01 M $CaCl_2$ and DTPA indicated that bone meal additions decreased the availability of the metals in the soils. Lv [24] studied the efficiency of sodium bentonite, bentonite and diatomaceous earth on remediation of Cd contaminated soil. The results showed that, the concentration of Cd reduced 21.40, 27.63, 27.24 and 32.30% as compared with the control when the additive amount was 20, 30, 50 and 40g/kg, respectively. There was also report on the remediation of contaminated soil by attapulgite clay [25]. The results showed that adding moderate attapulgite clay could make the Cd concentration reduce 46% in soil and the soil quality and productivity of the crops were not affected. Zhang et al [26] found that the chemical fixation efficiency of phosphate rock, furfural dreg and weathered coal on the contaminated soil. The results showed that three conditioning agents could reduce the concentration of Cu, Zn, Pb and Cd at some degrees. The chemical fixation could remediate the soil with low concentration contaminant; however, the bioavailability of fixed heavy metals may be changed with the changing environmental condition [27]. In addition, the use of conditioning agents could change the soil structure at some degrees and have effects on the microbes in soil.

2.3 Electrokinetic remediation

Electrokinetic remediation is a new remediation technology [28], in which voltage is applied at the two sides of soil and then electric field gradient is formed. The pollutant was carried to two poles treatment room via electro migration, electro osmotic flow or electrophoresis and then further treated [29, 30]. It is suitable for low permeable soil, and has advantages of easily install and operate, low cost [31, 32] and not destroy the original nature environment [33-35], so can attain the environmental remediation and protect the original ecotope [36]. However, the direct electrokinetic remediation cannot control the pH value of soil system well, and the treatment efficiency was almost low. The main improved methods include adding buffer solution in cathode and anode to control pH value by using ion exchange membrane to control pH value, to add complexant to improve migration, etc. [37]. Besides, there is combing other methods to remove the heavy metals, such as electrokinetic remediation combined with iron PRB [38], electrokinetic-oxidation/reduction combined remediation [39], and electrokinetic-microbe combined remediation [40].

2.4 Vitrify technology

Vitrify technology is to heat the soil at temperature of 1400~2000°C, the process in which organic matters volatilize or decompose. The steam produced and pyrolysis product was collected by off-gas treatment system. The melt after cooling forms rock shape vitreous, sieges the heavy metals and make it lose migration. It was reported that the strength of the vitreous is high 10 times than concrete. For ex-situ remediation, the energy can be supplied by fossil fuel burning or electrode directly heating, and then through arc, plasma and microwave transferring energy. For in-situ remediation, the heat can be through electrodes inserted into the contaminated soil. In short, this technology can remove the heavy metals and the efficiency was high. However, it is complicated and need lots of energy in the melting, which makes it cost a lot and limited in application [41].

3. Biological remediation

The biological remediation includes phytoremediation, bioremediation and the combining remediation.

3.1 Phytoremediation

The phytoremediation is to use living green plants to fix or adsorb contaminants, and cleaning the contaminants or making their risk reduction or loss. The phytostabilization, phytovolatilization and phytoextraction are the main three types of phytoremediation [42]. Phytostabilization is fixing heavy metals by plants through the adsorption, precipitation and reduction of root, and thus reducing their migration and bioavailability and preventing them migrating into the groundwater and food chain [43]. Phytovolatilization is transferring the heavy metals into volatile state or adsorbing the metals and transferring into gaseous matter, using special matters secreted by root [44]. Mercury is the most studied heavy metals. To explore the potential of plants to extract and detoxify mercury, Bizily et al [45] engineered a model plant, *Arabidopsis thaliana*, to express a modified bacterial gene, merBpe, encoding organomercurial lyase (MerB) under control of a plant promoter. MerB catalyzed the protonolysis of the carbon-mercury bond, removing the organic ligand and releasing Hg(II), a less mobile mercury species. Transgenic plants expressing merBpe grew dynamically on a extensive range of concentrations of monomethylmercuric chloride and phenylmercuric acetate. Plants lacking the merBpe gene were strictly reserved or died at the same organomercurial concentrations. This work suggested that native macrophytes (e.g. trees, shrubs, grasses) engineered to express merBpe may be used to degrade methylmercury at polluted sites and sequester Hg(II) for later removal. However, this technology is only suitable for volatile contaminants, and the application is limited [46]. Phytoextraction is adsorbing the heavy metals using tolerant and accumulating plants, and then transferring, storing at the over ground parts. Studying the adsorption characterization of different plants and screening high uptake plants is the key of this technology. According to the rules of U.S. department of energy, the high uptake plants screened should have the following characterizations: 1) Have high accumulating efficiency under the low contaminants concentration; 2) Accumulate high concentrations of the contaminants; 3) Accumulate many different kinds of heavy metals; 4) Grow fast and with large biomass; 5) Have pest and disease resistance ability [46].

3.2 Biological remediation

The microorganisms cannot degrade and destroy the heavy metals, but can affect the migration and transformation through changing their physical and chemical characterizations. The remediation mechanisms include extracellular complexation, precipitation, oxidation-reduction reaction and intracellular accumulation. Microbial leaching is a simple and effective technology for extracting precious metals from low-grade ores and mineral concentrates. Besides the industrial application for raw materials supply, microbial leaching has some potential for remediation of mining sites, treatment of mineral industrial waste products, detoxification of sewage sludge and for remediation of soils and sediments contaminated with heavy metals [47]. Lamber et al [48] studied the effects of sludge on mycorrhizal (MR) uptake of P, Cu, and Zn, and confirm MR supression of Cu and Zn uptake by P. Sludge reduced P uptake at 150 mg/kg P or higher in nonmycorrhizal (NMR) plants with little difference in plant growth among sludges. In MR treatments, growth and P-uptake responses to sludge ranged from very beneficial with two sludges to a complete inhibition of the MR response with another sludge. Mycorrhizae substantially increased shoot Cu and Zn uptake only at low soil-P levels. Abdel-Aziz et al [49] evaluated the role of VA mycorrhizae as a biological agent in reducing the toxicity of heavy metals. Inoculation with VA mycorrhizae induced significant increase in these parameters as compared with the uninoculated treatments. In the sewage sludge treated soil where the heavy metals were present in high concentrations, inoculation with VA mycorrhizae reduced the concentration of heavy metals. This indicated the role of VA mycorrhizae in reducing the hazardous effect of heavy metals when present in high levels in the media of growing plants. The study of Jones et al evidanced that, uptake of Cd from hyphal compartments was higher in mycorrhizal than in non-mycorrhizal plants, corresponding to 96, 127 and 131% of that in non-mycorrhizal plants when 1, 10 and 100 mg Cd kg-1 was added, respectively. A large proportion of the increased Cd content of mycorrhizal plants was sequestered in the roots. It is concluded that extraradical hyphae of AM fungi can transport Cd from soil to plants, but that transfer from fungus to plant is restricted due to fungal immobilization [50]. However, the biological remediation is susceptible to affected by different kinds of factors, such as temperatures, oxygen, moisture, pH value. It is also limited in applications, such as some microorganisms can only degrade special contaminants, microbes/zymin maybe incur secondary pollution.

3.3 Animal remediation

Animal remediation is according to the characterization of some lower animals adsorbing heavy metals, degrading, migrating the heavy metals and thus removing and inhibiting their toxity. The studies showed that, the treatment of the earthworm-straw mulching combinations enhanced plant Cu concentration, and the amount increased by it was lower than that of the earthworm treatment but higher than that of straw mulching treatment [51]. Kou et al [52] studied the Pb accumulation of earthworm through testing the Pb concentrations in soils. The results showed that, the earthworm could accumulate Pd effectively. The accumulation amount increased with the Pb concentrations increasing.

4. Conclusion

The research of remediation technologies is still in individual and experimental stage. The development strategy of future remediation technologies is researching green, environmental-friendly biological remediation, combining remediation, in-situ remediation, based on equipped completely quick remediation, and supplying technical supporting for agricultural soil contamination, industrial enterprises brownfield, mining sites, etc.

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