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Review on remediation technologies of soil contaminated by heavy metals

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Abstract

The environmental safety of soil has become severe in China with the boost of industrialization and urbanization. In this paper, on the basis of investigating the status of soil contaminated in China, 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

Soil is the basic environmental elements constituting ecosystem, and the important material basis of human being surviving and developing. The environmental safety of soil becomes severe in China with the boost of industrialization and urbanization. It was calculated that, the contaminated soil reached about 150 million mu. The soil contaminated resulting from sewage irrigation was 32.5 million mu, and the soil stockpiled and ruined by solid waste was 2 million mu. These covered 10% of the total cultivated area. As different kinds of industrial wastewater, exhaust gas, livestock manures, sewage irrigation and sludge farm application have all become the sources of heavy metals [1], the soil contaminated by heavy metals has become one of the environmental problems that polluted widely and harm severely. It can be see from the bulletin on Chinese domestic environmental conditions of 2000 year that [2], the heavy metals in 36 thousands hectare of the soil was out of limit in the surveyed 0.3 million hectare soil and the over standard rate reached 12.1%. The data in high level Chinese food safety forum of 2009 year revealed that,

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1/6 of the cultivated land was contaminated by heavy metals and the area was more than 20 million hectare. The pollution of water and contamination of soil by heavy metals have become a threat to ecological environment, food safety and agriculture sustainable development. It was estimated by the ministry of environment protection, the grain contaminated by heavy metals reached 12 million tons and the immediate economic loss was over 20 billion yuan. The soil contaminated by heavy metals manifests as concealment, accumulation, irreversibility, and protraction. The prevention of heavy metals contaminated soil is not only needed to control the sources, but also enhance the remediation of contaminated soil [3].

The European countries have invested a lot to remediate the contaminated soils [4]. 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.

1. Physical remediation

The physical remediation mainly includes soil replacement method and thermal desorption. The soil replacement means using clean soil to replace or partly replace the contaminated soil with aim of dilute the pollutant concentration, increase the soil environmental capacity, and thus remediate the soil [4, 8]. 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 feasibly, or else it will incur the second pollution. (2) Soil spading is deeply digging the contaminated soil, making the pollutant spread 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 large in working volume, costs a lot and is suitable for soil with small area and polluted severely [3].

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 achieve the aim of removing the heavy metals [9]. 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 [10].

2. Chemical remediation

2.1 Chemical leaching

Chemical leaching is washing the contaminated soil using fresh water, reagents, and others fluids or gas [11, 12] that can leach the pollutant from the soil. Trough the ions exchange, precipitation, adsorption and chelation, the heavy metals in soil was transferred from soil to liquild phase, and then recovered from the leachate. The leachate using mainly include inorganic eluent, chelation agents, and surfactant, etc. Tokunage and Hakuta [13] investigated the effects of different concentrations of hydrogen fluoride, phosphoric acid, sulfuric acid, hydrogen chloride, nitric acid on As extraction from artifical 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. An environment-friendly and cost-effective extraction method has been studied for the removal of arsenic from contaminated soil [14]. A yellow-brown forest soil was contaminated with arsenic (V) and used as a model soil. Among various potassium and sodium salts, potassium phosphate was most effective in extracting arsenic. Arsenic was efficiently 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 [15]. Removal efficiencies for fine sediments were >95% after 1 h of washing with 0.2 M citric acid. When using 0.2 M citric acid mixed with 0.1 M potassium phosphate, the As removal efficiency increased to 100%. It is worth mentioning that, the effect is almost unsatisfactory using single extractant 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 effective 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 [16]. Ehsan et al [17] evaluated the efficiency of a washing process with cyclodextrin in combination with EDTA for the simultaneous mobilization of heavy metals and PCBs from a field contaminated soil. These studies demonstrated that PCB compounds and selected heavy metals can be coextracted efficiently from soil with three successive washes with the same washing suspension containing 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 [18]. Hong et al [19] 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 [20] 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 adding reagents or materials into the contaminated soil and using 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 achieving the remediation of soil [3]. The soil conditioning materials used include clays, metallic oxides, biomaterials, etc. Hodson et al [21] evaluated the ability of bonemeal 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 bonemeal amended soils using 0.01 M $CaCl_2$ and DTPA indicated that bonemeal additions reduced the availability of the metals in the soils. Lv [22] 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 [23]. 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 [24] evaluated 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 environmental condition changing [25]. 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 [26], which is mainly applying voltage at the two sides of soil and then forming electric field gradient. The pollutant was carried to two poles treatment room via electromigration, electroosmotic flow or electrophoresis and then treated further [27, 28]. It is suitable for low permeable soil, and has advantages of easily install and operate, low cost [29, 30] and not destroy the original nature environment [31-33], so can achieve the environmental remediation and protect the original ecotope [34]. However, the direct electrokinetic remediation can not 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, using ion exchange membrane to control pH value, adding complexant to improve migration, etc. [35]. Besides, there is combing other methods to remove the heavy metals, such as electrokinetic remediation combined with iron PRB [36], electrokinetic-oxidation/reduction combined remediation [37], and electrokinetic-microbe combined remediation [38].

2.4 Vitrify technology

Vitrify technology is heating the soil at temperature of 1400~2000°C, in which process the 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 summary, 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 [39].

3. Biological remediation

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

3.1 Phytoremediation

The phytoremediation is the use of living green plants to fix or adsorb contaminants, and cleaning the contaminants or making their risk reduction or disappearance. The phytostabilization, phytovolatilization and phytoextraction are the main three types of phytoremediation [40].

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 foodchain [41].

Phytovolatilization is transferring the heavy metals into volatile state or adsorbing the metals and transferring into gaseous matter, using special matters secreted by root [42]. Mercury is the most studied heavy metals. To explore the potential of plants to extract and detoxify mercury, Bizily et al [43] 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 vigorously on a wide range of concentrations of monomethylmercuric chloride and phenylmercuric acetate. Plants lacking the merBpe gene were severely inhibited 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 [44].

Phytoextraction is adsorbing the heavy metals using tolerant and accumulating plants, and then transferring, storing at the overground 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 [44].

3.2 Biological remediation

The microorganisms can not 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 valuable 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 [45]. Lamber et al [46] 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 [47] 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 showed 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 [48]. However, the biological remediation is vulnerable 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 [49]. Kou et al [50] 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 in China 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 agicultural soil contamination, industrial enterprises brownfield, mining sites, etc.

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