Kossge International Symposium on Soil & Groundwater Environment
October 27-28, 2005 Mayfield Hotel, Seoul, Korea

Lead Speciation and Leachability in Untreated and Stabilized/Solidified Soils: A Review

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ABSTRACT

Soil contamination by heavy metals is a growing concern in the industrialized world and extensive research is taking place with regard to their speciation, mobility and treatment in various media, including different forms of hazardous waste and contaminated soils. Lead (Pb) is well established as a compound of significant toxicity to humans and other life forms and is thus ranked as one of the most hazardous species in the U.S. Environmental Protection Agency priority list. According to U.S. Geological Survey, as of 2002, the main sources of Pb contamination in the soil environment are the battery industry and firing ranges. However, Pb mobility in the environment, greatly depends on its speciation, i.e. its chemical form, as well as on the properties of the contaminated soil. The type of treatment that can be applied in order to remove/extract and/or stabilize Pb also depends to a significant degree on Pb speciation. The feasibility of a physical or chemical extraction depends, for example, on lead availability and the stability of its chemical form. Alternatively, stabilization/ solidification (S/S) has been widely employed as a treatment method for contaminated soils. Both the chemical form of Pb as well as the properties of the soil matrix, are altered in order to minimize Pb mobility in the subsurface and render it unavailable to humans, animal and plant life forms.

Stabilization/solidification routinely employs agents such as cement, lime, fly ash and other cementitious products in order to produce a matrix of low permeability and high strength that is resistant to water penetration and degradation by environmental factors. The soil mineralogy and physicochemical properties are drastically changed by the addition of these highly alkaline media to the soil, depending also on the inherent properties of the contaminated soil.
The degree of contamination of a soil and the effectiveness of S/S are assessed by applying regulatory leaching tests, such as the Toxicity Characteristic Leaching Procedure (S/S) in USA. During TCLP, acetic acid solution is added to the solid, in order to simulate worst-case conditions of acid attack to the tested medium. Again, the solid matrix is altered and chemical reactions take place during the test itself, which alter the speciation of hazardous compounds and thus, their solubility. Other leaching tests, such as DI water extraction, columns tests, sequential extraction test and synthetic precipitation leaching

procedure (SPLP) have also been applied to assess Pb speciation and mobility in contaminated soils.

In all of the above cases, the possible immobilization mechanisms of Pb in a soil (untreated or treated) are the following: a) precipitation b) sorption (adsorption, absorption and ion exchange) and c) inclusion (physical encapsulation and chemical inclusion.

Various studies have been conducted on Pb speciation in untreated and treated soils, as well as on the implications on its mobility and the treatment effectiveness. The present paper attempts to effectively summarize previous findings in a comprehensive review of Pb forms encountered in soil environments, as well as in S/S-treated soils. The soil properties and environmental or testing conditions, under which the specific Pb forms are favored, are also addressed. A specific focus is granted to firing range soils, as these constitute a primary source of Pb soil contamination across the world.

Lead is introduced into the firing range soils in its pure metallic form, as it is an integral part of the ammunition due to its favorable physical characteristics such as low melting point, its malleability, high density and relatively high resistance to corrosion. Metallic Pb is, generally, unstable in soil environments and subject to weathering reactions. The main transformation product identified by previous research is lead carbonate, i.e. cerussite and hydrocerussite. It seems that the formation of lead carbonate is a primary immobilization mechanism in both untreated and treated soils and will dictate Pb leachability in terms of TCLP. The formation of lead carbonates can effectively reduce lead concentrations below regulatory limits, depending on the availability of carbonate and soil properties such as pH and Acid Neutralization Capacity.

Other major Pb precipitates reported in the literature are anglesite (lead sulfate), litharge (lead oxide) and pyromorphite (lead phosphate). The latter is the most insoluble Pb precipitate under aerobic conditions and has therefore been proposed as a treatment mechanism. However, there is limited evidence on the solubility of phosphate and the extent of pyromorphite formation in soils. Moreover, there are significant sustainability issues associated with phosphate treatment of Pb contaminated soils that would need to be addressed. Sorption on ferromanganese oxides, goethite, gibbsite and clay and other aluminosilicate minerals has also been reported as an active immobilization mechanism for Pb in firing range soils. With regard to S/S-treated soils, it has been reported that the interaction with the pozzolanic products formed, namely Calcium Alumina Hydrates and Calcium Silica Hydrates (CAH/CSH) is the mechanism for Pb immobilization. However, it is still under debate, whether physical or chemical inclusion is the active mechanism, while the behavior of Pb under TCLP conditions is still not clear.

Several studies account for Pb speciation by employing the sequential extraction test, which associates Pb with operationally defined soil fractions. However, detailed mineralogical and physiochemical analyses reported point at a dynamic behavior of Pb during the test, i.e. the testing conditions themselves alter Pb speciation.

In conclusion, it is extremely important to address the possible Pb transformations during each step of a characterization and treatability investigation and to elucidate the active immobilization mechanism and its sensitivity to changes in the environmental conditions. The choice of test and the experimental conditions have a significant impact on Pb speciation and may skew the final evaluation of the contamination form and extend as well as treatment and/or life-cycle maintenance options.