

# SOLIDIFICATION/STABILISATION OF SOIL CONTAMINATED WITH METAL: A REVIEW

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## ABSTRACT

*Solidification/stabilisation (S/S) is generically defined as a chemical and physical alteration technique of reducing the mobility as well as solubility of contaminants in wastes in order to convert them into chemically inert form. The technique is specifically developed to confine the movement of contaminants in wastes so that their concentrations in the surrounding environment (e.g. subsurface soil matrices and groundwater) will not exceed stipulated environmental regulatory levels. This technique necessitates application of cementitious materials such as cement which also provides a favorable solidification effect on the stabilised wastes so that the end product can be easily transported to disposal sites or reused as construction materials. This paper reviews the S/S technology as applied to contaminated soil treatment with emphasis on its chemical binder systems, mechanisms, interferences and post-treatment leaching tests. S/S is an important soil contamination remediation technology as evident by its simplicity, technical and cost-effectiveness.*

**Keywords:** Contaminated Soil, Metal, Solidification/Stabilisation

## 1.0 INTRODUCTION

Solidification/stabilisation (S/S) technology as applied to wastes uses physical and chemical processes to produce chemically stable solids with improved contaminant containment and handling characteristics [1]. Solidification refers to a process whereby wastes in the form of sludges or soils, are solidified to produce free-standing and monolithic masses with enhanced physical integrity [1,2] whereas stabilisation is a chemical alteration technique of reducing the mobility and solubility of contaminants in wastes or soil [3,4]. Since metals are considered relatively immobile, methods for metal decontamination have focused on solid-phase processes such as S/S [5]. In the United States, S/S has been identified as the Best Demonstrated Available Technology (BDAT) for a variety of Resource Conservation and Recovery Act (RCRA) non-wastewater wastes, especially metal-based contaminants [6]. While S/S is a well-established technology for treating industrial sludges around the world, its application in remediation of contaminated soils is still in its infancy with exception of the United States and Canada where contaminated land remediation processes incorporating S/S are widespread. A review of such technology for treating contaminated soil is therefore, desirable to provide concise information of such application for the benefit of practitioners. This paper reviews the S/S technology as applied to contaminated soil treatment with emphasis on its chemical binder systems, mechanisms, interferences and post-treatment leaching tests.

## 2.0 SOLIDIFICATION/STABILISATION TECHNIQUES

S/S systems are classified according to the application of main stabilising agents which are based on cement/lime, pozzolana, silicate, thermoplastic or polymer systems [7]. S/S is evidently best suited for treatment of metal and radioactive wastes as indicated in Table 1 which shows the compatibility of selected

**Table 1: Compatibility of selected waste categories with different solidification/solidification techniques [8]**

Waste Component	Treatment Type			
	Cement-based	Pozzolan-based	Thermoplastic	Surface Encapsulation
Organics				
Organic solvents and oils	May impede setting, may escape as vapor	May impede setting, may escape as vapour	Organics may vaporise on heating	Must first be absorbed on solid matrix
Solid organics (e.g., plastics, resins, tars)	Good. Often increases durability	Good. Often increases durability	Possible use as binding agent in this system	Compatible. Many encapsulation materials are plastic
Inorganics				
Acid wastes	Cement will neutralise acids	Compatible. Will neutralise acids	Can be neutralised before incorporation	Can be neutralised before incorporation
Oxidisers	Compatible	Compatible	May cause matrix breakdown, fire	May cause deterioration of encapsulation materials
Sulfates	May retard setting and cause spalling	Compatible	May dehydrate and rehydrate causing splitting	Compatible
Halides	Easily leached from cement, may retard setting	May retard set, most are easily leached	May dehydrate and rehydrate	Compatible
Heavy metals	Compatible	Compatible	Compatible	Compatible
Radioactive materials	Compatible	Compatible	Compatible	Compatible

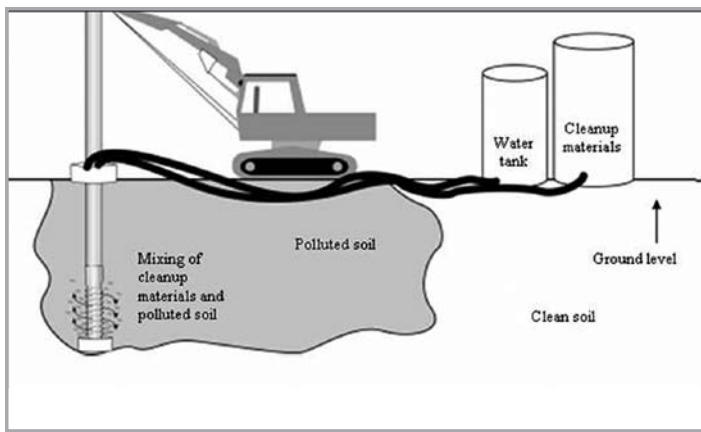


Figure 1: In-situ solidification/stabilisation of polluted soil [9]

waste categories with different S/S techniques. Conversely, certain metals such as chromium (VI) and mercury are generally not suitable for S/S since they do not form hydroxides that are highly soluble [5].

Some of the apparent advantages of selecting S/S over other land remediation techniques are its cost effectiveness, comparatively rapid means and the option of utilising this technology via in-situ or ex-situ. The former option is generally favoured since it minimises wastes transport. Figure 1 illustrates in-situ S/S of polluted soil. Nonetheless, stabilisation may have possible adverse effects such as impairment of biological activity and destruction of soil chemical properties and physical structure [3].

### 3.0 CHEMICAL BINDERS

#### 3.1 Ordinary Portland Cement

The term binder is defined as a reagent that contributes to the strength gain associated with stabilisation. Portland cement is the most widely applied binder in chemical fixation and solidification systems and is generally produced by heating together limestone and sources of silica such as clay at about 2700°F, forming a mass called clinker [3]. Portland cement is essentially a calcium silicate mixture consisting predominantly tricalcium and dicalcium silicates with minor portions of tricalcium aluminate, calcium aluminoferrite and other metal oxides. This type of cement is the most widely used in S/S technology due to its relatively low cost and other advantages (which outweigh its disadvantages) as specified in Table 2.

The cementation mechanism of the waste/cement reaction which may be directly related to S/S of contaminated soil is explained in the following [3,6]. Cementation of the waste/binder mixture begins when water is added, either directly or as part of the waste. Once the cement powder contacts water, tricalcium aluminate immediately hydrates, causing the rapid setting which produces a rigid structure. In an idealised setting, the water hydrates the calcium silicates and aluminates in the cement to form calcium-silicate-hydrate (CSH). Thin, densely-packed fibrils of silicate grow out from the cement grains and interlace to harden the mixture entrapping inert materials and unreacted grain. Hydration of tricalcium and dicalcium silicates results in the formation of tobermorite and crystalline CSH. These compounds account for strength development after the initial setting of the mixture.

Table 2: Advantages and disadvantages of cement-based stabilisation techniques [6]

Advantages	Disadvantages
<ul style="list-style-type: none"> <li>• Availability of materials locally on a worldwide basis.</li> <li>• Relatively low cost of materials and mixing equipments.</li> <li>• Ability to create a strong physicochemical barrier under adverse conditions such as acid leaching and applied compressive loads.</li> <li>• Low variability in composition.</li> <li>• Availability of numerous existing data on cement-based reactions and immobilisation of metals.</li> </ul>	<ul style="list-style-type: none"> <li>• Sensitivity of product quality to presence of impurities such as non-polar organics at high enough concentrations.</li> <li>• Waste volume typically increases due to binder addition, although not necessarily more than with other inorganic binders.</li> </ul>

#### 3.2 Pulverised Fly Ash

Pulverised fly ash (PFA) is a by-product generated from burning coal during the generation of electricity in coal-fired electric power plants. Physically, PFA consists of finely divided spheroids of siliceous glass, about 1 to 50 microns in diameter, plus minor fractions of larger, irregular shaped particles [3]. PFA is extensively used in the past decades as a cement replacement material in concrete as well as chemical binder in S/S treatment since it has similar binding and stabilising properties as cement. PFA can be added to such coarse-grained wastes to augment the pozzolanic surface area and hence improve the properties of the waste mixture such as strength, workability, buffering capacity to resist pH changes and heavy metal leachability [10]. Adding PFA to treated contaminated media would be a cost-effective method of waste disposal since PFA, itself is considered a waste. A distinct advantage associated with PFA is that unburned carbon in PFA may facilitate absorption of organics from wastes [11] and thus indicates its suitability in treating both inorganic and organic wastes. Nonetheless, one problem identified with the use of PFA is the growth of the mineral ettringite in the presence of sulfates that results in destabilisation of solidified matrix.

#### 3.3 Lime

Lime is a generic term usually used to cover the various chemical and physical forms of quicklime ( $C_aO$ ), hydrated lime [ $C_a(OH)_2$ ] and hydraulic lime according to Conner [3]. The reaction product formed as a result of combining lime and PFA with water is initially a noncrystalline gel that eventually forms calcium-silicate-hydrate (CSH), a compound found in hydrated Portland cements. Lime is generally used to enhance engineering physical properties of soils for foundation purposes but its application has been extended to stabilise heavy metals in soils, particularly of clayey nature as evident in studies conducted by Boardman [7] and Musta *et al* [12].

#### 3.4 Rice Husk Ash

Recent researches on S/S of metal contaminated soils were focused primarily on standalone Portland cement systems or incorporation of other established pozzolans such as pulverised

fly ash (PFA) and lime as reflected in studies conducted by Boardman [7], Musta *et al* [12], Dermatas *et al* [10] and Wang and Vipulanandan [13]. While these S/S systems exhibit excellent treatment effectiveness, their applications in Asian countries such as Malaysia have drawbacks which include relatively high costs of cement and lime as well as inavailability of mass amount of fly ashes since there is only a comparatively small number of coal-fired power plants within the region. Concurrently, Asian countries are experiencing predicament in disposal of rice husk heaps due to their abundance.

Rice husks are the discarded external layers of rice grains where the husks' silica content can be enriched by burning the husks and converting them into ashes. Rice husks are the largest milling by-products of paddy, constituting about 15 % of paddy by weight [14]. Rice husk ash (RHA) does not readily exhibit self-cementing characteristics but upon addition of Portland cement and water, pozzolanic reactions will be initiated, leading to cementitious product formation. This is due to the high amount of SiO<sub>2</sub> present in RHA. Concrete technologists are gradually finding applications in RHA as an additive for producing high-strength concrete but there is no identified usage of RHA in solidification/stabilisation of contaminated soils. Therefore, the usage of RHA in the field of land remediation would represent a two-prong approach in solving its disposal dilemma as well as providing an inexpensive cement replacement material.

#### 4.0 RECENT RESEARCH ON S/S OF METAL CONTAMINATED SOILS

The aforementioned chemical binders have been used by researchers as additive for S/S technology. Table 3 lists the summary of research on S/S of metal contaminated soil as conducted by various researchers.

**Table 3: Research on S/S of metal contaminated soil based on binder system used**

Researcher(s)	Nature of Study	Binder System
Bennett and Al-Tabbaa [15]	Investigated bench-scale chemical immobilisation of sand (particle sizes 300 – 600 µm) contaminated with 660 mg of copper sulphate per liter of water and 10 g/kg of vegetable oil. Results indicated that treatment was effective for copper sulfate but unsuccessful for oil. The authors suggested that the oil was retained in the matrix by physical encapsulation.	Cement and Hydrofoam (cementitious foam)
Boardman [7]	Investigated stabilisation of lead (II) nitrate and iron (III) nitrate contaminated clay via removal of the contaminants from aqueous phase. High pH environment resulted in the contaminants being removed from the aqueous phase by precipitation of the ions as solid hydroxides.	Lime

Dermatas and Meng [10]	Studied the use of fly ash in addition to quicklime and sulfatesaltstosolidify/stabilise lead (7,000 mg/kg soil) and chromium (4,000 mg/kg soil) contaminated clayey sand soil. The treatment resulted in a high strength and swell-resistant monolithic solid that effectively immobilised lead.	Fly ash, quicklime and sulfate salts
Dutre <i>et al</i> [16]	Studied S/S of contaminated soil and rock-like material from an arsenic contaminated site. The soil material contained 2.5 – 3.5 dry wt. % of arsenic while the rock-like material contained up to 20 %. Concentration of arsenic in leachate of stabilised material was less than 1 mg/L.	Cement and Lime
Garcia <i>et al</i> [17]	Assessed the effectiveness of low grade-MgO as a stabiliser used to remove heavy metals from heavily contaminated soils. Regardless of the quantity of stabiliser employed (greater than 10%), low grade-MgO provides an alkali reservoir that allows guaranteeing long-term stabilisation without varying the pH conditions.	Magnesium oxide
Jing <i>et al</i> [18]	Investigated S/S of Cr(III) contaminated soil. Modeling results indicated that the release of Cr(III) was controlled by adsorption on iron oxides at pH < 10.5, and by precipitation of Ca <sub>2</sub> Cr <sub>2</sub> O <sub>5</sub> ·6H <sub>2</sub> O at pH > 10.5.	Cement, lime and fly ash
Kostarelos <i>et al</i> [19]	Determined the optimum dose of lime and fly ash required for treatment of hexavalent chromium contaminated soil. Adequate treatment was obtained after 1 day of curing with just 25% fly ash and 10% quicklime.	Lime and fly ash
Kumpiene <i>et al</i> [20]	Evaluated the changes in Cu and Pb mobility and bioavailability in soil induced by the addition of coal fly ash and natural organic matter (peat). Results indicated that this method can be a promising technique for in situ remediation of Cu and Pb contaminated soil.	Fly ash and peat
Mahabadi <i>et al</i> [21]	Evaluated the effect of zeolite to stabilise cadmium in a variety of soil textures in Gilan (northern Iran) with different pH values. The effect of preventing heavy metals from leaching was found to be more pronounced when zeolite was applied to clay soils.	Clinoptilolite (natural zeolite)

Moon and Dermatas [22]	Investigated a fly ash-based S/S technique using field soil samples contaminated with arsenic and lead. It was found that arsenic release was reduced by 98.3% while lead release was reduced by 98.5% upon addition of 25% Class C fly ash.	Fly ash
Moutsatsou and Protonotarios [23]	Investigated a S/S technique of using fly ash as a synthetic zeolite to remediate soils polluted with heavy metals and metalloids (As, Pb, Cu, Zn, Fe, Cd and Mn). Retention results showed that the yield of the process depends on a number of factors such as the metal speciation in soil and the interaction of solvent with both the soil and the substrates.	Fly ash, lime and cement
Musta and Kassim [12]	Investigated stabilisation of metal contaminated clayey soil and waste sediments. Treatment was effective for clayey soil but waste sediments required addition of pozzolans.	Hydrated Lime
Shawabkeh [24]	Tested the ability of a mixture of sand, cement and clay for immobilising cadmium ions from leaching out from sandy soil. Results indicated that a sand–cement–clay mixture with mass percentages of 25% sand, 50% cement and 25% clay was eligible for obtaining a maximum adsorption capacity toward cadmium, the metal of interest and having a good compressive strength.	Cement and clay
Wang and Vipulanandan [13]	Investigated S/S of 25,000 mg/kg potassium chromate contaminated clayey soil by pretreating the soil with Fe (II) prior to the solidification process. This technique was effective in reducing the leaching of Cr (VI) by 99%. Chromium hydroxide was one the reaction products identified in the treated soil.	Fe (II) (for pre-treatment) and cement
Yin <i>et al</i> [25]	Investigated S/S of lead-contaminated soil. Results indicated that usage of cement with RHA as an overall binder system for S/S of lead-contaminated soils is more favorable in reducing the leachability of lead from the treated samples than a binder system with standalone cement.	Cement and RHA

Yin <i>et al</i> [26]	Investigated ex-situ S/S of scrap metal yard contaminated soil. Results indicated that chemical stabilisation of metal-contaminated soils using cement was effective for prevention of metal leaching from both disintegrated samples subsequent to years of weathering (crushed block) and intact samples (whole block) into the environment.	Cement
Yukselen and Alpaslan [27]	Evaluated the efficiency of S/S to treat lead, copper and iron contained in contaminated soil samples of an old smelting facility. Results indicated that the degree of heavy metal leaching is highly dependent on pH.	Cement and lime

## 5.0 MECHANISMS IN SOLIDIFICATION/STABILISATION TREATMENTS

### 5.1 Immobilisation Mechanisms

Stabilisation of wastes or contaminated soils may involve physical mechanisms, chemical mechanisms or a combination of the two. Physical stabilisation (solidification or encapsulation) alters the physical form of the waste but does not necessarily cause chemical binding of the waste constituents. Chemical stabilisation changes the chemical states of waste constituents to forms with lower aqueous solubilities [28].

Physical mechanisms of S/S which comprise macro and micro encapsulations, operate by confining waste constituents within an area in the stabilised wastes or contaminated soils. Macroencapsulation works by physically entrapping contaminants within a large structural matrix whereas microencapsulation entraps contaminants within the crystalline structure of solidified matrix at a microscopic level [11]. These mechanisms generally relate to the treatment of organic wastes such as oil and grease, PCBs, pesticides and volatile compounds that typically occur exclusive of accompanying chemical interactions [3]. However, in the context of metal wastes, S/S treatment relies predominantly on chemical interactions between the binder(s) and the wastes rather than physical stabilisation. The distinction between immobilisation mechanisms of metal and organic wastes can be generally elucidated based on the interactions of their molecules with the binders. Metal contaminants generally consist of polarised ions that are capable of electrochemical interactions with binder molecules in the presence of water and this may result in formation of highly insoluble substances that are resistant to leaching. This is in contrast with the non-polarised molecules of organic contaminants which are incapable of chemical interactions with the binders and thus only encapsulated in the S/S matrix. Figure 2 shows the various metal fixation mechanisms associated with S/S.

It is widely established that cement and pozzolan-based waste forms rely profoundly on pH control for metal containment. In S/S, binders such as cement are employed to elevate pH of these waste forms to the ranges of 8 to 11 which, in turn, facilitates

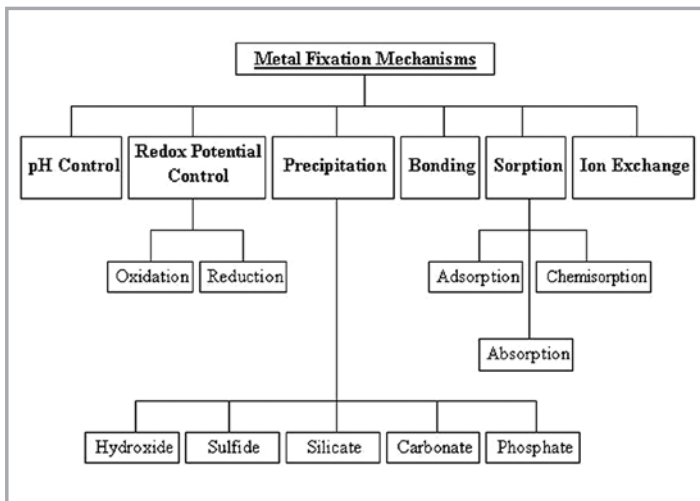


Figure 2: In-situ solidification/stabilisation of polluted soil [9]

precipitation of metal salts (metal hydroxides, metal sulfides, metal silicates). These precipitates generally exhibit minimal solubility in the aforementioned high pH range. Another crucial factor that influences the solubility of the stabilised wastes is the amphoteric nature of certain metals such as chromium, lead and zinc. These metals exhibit maximum solubility at both extremes of low and high pH. Therefore, it is vital that the most suitable and optimum pH to minimise the solubility of amphoteric metal contaminants be determined in order to increase the effectiveness of S/S treatment.

## 5.2 Leaching Mechanisms

In the context of S/S, leaching is defined as the process of gradual extraction of contaminants from treated wastes through exposure to either stagnant or flowing of leachant (in the form of water or other solvent) through and around the wastes. The following paragraph [3,28] describes the leaching mechanisms pertaining to S/S-treated waste in the presence of a leachant.

A stabilised waste is a porous solid which is at least partially saturated by with water and gas. There may be several different solid phases, each containing contaminants. Prior to introduction of leachant, this pore system is in equilibrium with the surrounding solid phase and therefore, there is no net transfer between phases. The leachant alters the composition of the system and disrupts the chemical equilibrium, resulting in the mobilisation of contaminants. The new system may evolve towards a new equilibrium if sufficient time passes with no leachant renewal. At this stage, the two basic leaching mechanisms are mobilisation and transport of the contaminant. The leachant mobilises contaminants within the pores by dissolving the contaminant while dissolution results from a combination of chemical and physical mechanisms. Factors that affect the extent of equilibrium concentrations include the solubility of constituent and chemical makeup of the pore water. As more soluble constituents are leached from a relatively insoluble solid matrix, a layer deficient in the leached constituents develops. As constituents leach, the layer may become more porous compared to the unleached solid, so that molecular diffusion in the pore water and boundary layer phenomena become the limiting factors. Transport of the mobilised contaminants occurs by bulk advective flow or diffusion. If water flows within the S/S-treated waste, advective transport causes

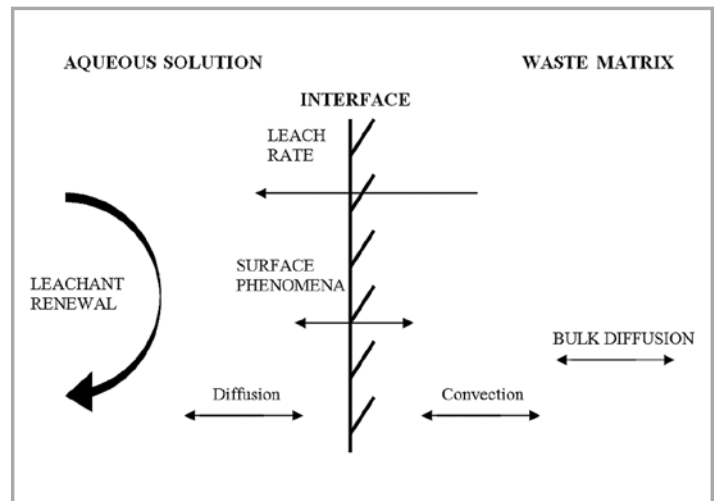


Figure 3: Leaching mechanisms [29]

contaminants that have been mobilised by reactions in the pores to flow through the waste. Figure 3 illustrates the various leaching mechanisms that pertain to S/S-treated waste.

## 6.0 INTERFERENCES IN CHEMICAL STABILISATION

Interferences in chemical stabilisation of wastes are primarily caused by presence of chemical constituents in the wastes being stabilised or inadvertently introduced into the stabilisation process. These interferences can affect either the solidification or stabilisation processes or both of them simultaneously. Unfavourable effects derived from interference with solidification include retardation of setting for the waste-binder mixture as well as reduced physical strength and integrity of the final product.

Retardation of stabilisation is postulated to occur primarily via adsorption of retarding compounds onto hydration products, principally CSH, although the aluminate phase consumes a disproportionate share of the retarder [30]. In the context of metal ion inhibitors, it is thought that insoluble metal hydroxides and sulfates form an impermeable barrier to hydration by precipitating onto cement particles. This barrier forms an impervious coating on CSH and inhibits further hydration by impeding transport of water into the cement grain [30]. In addition to interferences caused by chemical constituents, fine particulates such as silt, clay or coal dust can also coat cement particles and prevent the growth of CSH crystals from the cement grain [6]. It is this factor that provides a challenging aspect to treatment of contaminated soils as soils, in essence, consist of silt and clay particles.

## 7.0 POST-TREATMENT LEACHING TESTS

### 7.1 Toxicity Characteristic Leaching Procedure

The Toxicity Characteristic Leaching Procedure (TCLP) is used to determine the extent of toxicity characteristics of particular contaminants of hazardous wastes or soils which determines the mobility of organic and inorganic analytes of the wastes. In this test, waste samples are crushed to particle size less than 9.5 mm and extracted with an acetic acid solution with pH of either 2.88 or 4.93 depending on the alkalinity of the waste [6]. The TCLP leaching solution is designed to simulate worst-case leaching

conditions on disintegrated landfill wastes due to prolonged aging effects. Although the TCLP test is principally used to determine hazardous characteristics, it is occasionally utilised to determine the impact of a waste on groundwater even when the waste is stored or disposed in non-landfill conditions [31].

### 7.2 Synthetic Precipitation Leaching Procedure

The Synthetic Precipitation Leaching Procedure (SPLP) is similar to TCLP and differs only in the leaching solution, where the acetic acid solution is replaced by a dilute nitric/sulfuric acid mixture. SPLP simulates acid rain as opposed to simulating a leachate in a sanitary or municipal landfill. It is observed that the majority of land remediation engineers utilise TCLP instead of SPLP as the mechanism for regulatory compliance. However, a recent study conducted by Shieh [32] rebukes this practice by concluding that the two tests exhibit different effects on leaching of contaminants from different wastes. The study recommends that SPLP should be considered over TCLP in the case of disposal of wastes containing very minor organic contents while TCLP should be recommended for consideration if organic decomposition were anticipated to occur. Another noteworthy finding of this study is that for leaching of lead from wastes in an anticipated high final leachate pH (>10.5), TCLP should be selected over SPLP as the more appropriate leaching test.

### 7.3 Whole Block Leaching (Semi Dynamic Leaching)

Whole block leaching (WBL) or semi-dynamic leaching is conducted to determine the leachability of heavy metals from the monolithic solidified cubes after a stipulated period of curing. The term semi-dynamic means that the leachant is replaced periodically after intervals of static leaching [2].

## 8.0 CONCLUSIONS

S/S is an important soil contamination remediation technology as evident by its simplicity, technical and cost-effectiveness. It is the opinion of the author that practitioners should focus on usage of recyclable waste materials such as PFA and RHA as replacement for chemical binders due to their obvious benefits. Research on S/S should also be focused on ways to reduce interferences caused by organic constituents present in contaminated soils as this is one of the major disadvantages of the technology.

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## PROFILES



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