

**Review Research**

## POZZOLANIC MATERIALS FOR STABILIZATION /SOLIDIFICATION OF SOIL CONTAMINATED BY HEAVY METALS - A REVIEW

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**Abstract:** Soil contamination with heavy metals significantly threatens human health and the ecosystem. Due to the complexity of heavy metal interactions in soils, the mobility, bioavailability, and toxicity of metals in the soil fractions are impacted by several parameters. These parameters include the qualities of both the metal and the soil. However, several remedial methods have been used in immobilization techniques. One of the best techniques is the Stabilization/Solidification(S/S) approach, which is often used to remediate contaminated sites and combines contaminants with binders to reduce the quantity of contaminant leachability through soil matrix and groundwater pollution. As well as to minimize the risks to human health and the environment, alter the metals in the soil to make them less soluble, toxic, or bioavailability. Stabilization aims to change the contaminated material's physical and chemical characteristics to decrease its chemical reactivity or solubility. In contrast, solidification aims to turn contaminants into solids that can be handled easily and contain a few dangerous materials. This review's primary goal is to examine the pozzolanic materials used in the Stabilization/Solidification process and their potential for remediating soil contamination, mainly where heavy metals are present.

**Keywords:** Contaminated soil; metals; pozzolanic materials; stabilization; solidification

### 1. Introduction

Heavy metals are metals with relatively high densities, atomic weights, or atomic numbers. It can enter the soil due to both pedogenic and anthropogenic processes. Soil contamination

occurs due to various agricultural and mechanical activities, such as burning petroleum products, using manures and pesticides in farming, mining waste, and landfill filtering [1-3]. Heavy metals, radioactive materials, pesticides, solid waste, and sewage water are the most critical soil pollutants. Both pedogenic and anthropogenic activities have the potential to result in heavy metals entering the soil environment. Heavy metal soil pollution has rapidly increased in recent decades, becoming a global environmental issue that has drawn widespread public attention[4, 5]. The risk of heavy metal contamination in soil is that these metals can be carried to other sites by wind and runoff water, resulting in an accumulation of these metals[6]. The degree of contamination is determined by the chemical compositions of the contaminated soil and the soil characteristics. So understanding the geotechnical characteristics and behavior of contaminated soils is necessary for all prospective applications of contaminated soils. In this situation, the impact of crude contaminants on existing facilities must be determined[7]. The movement of heavy metals in soil is intricate. Thus, it is necessary to devise

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means of addressing these issues. In the past, many immobilization technologies were used to perform remediation strategies. Stabilization/Solidification is one of the most successful approaches [8].

Treatment procedures for solidification and stabilization include combining or injecting treatment materials into the contaminated soils. In Stabilization/Solidification techniques, both in-situ and ex-situ processing are possible [9, 10]. Immobilization aims to stabilize metals by reducing the soil matrix's leaching ability. It prevents metals from transforming into a toxic, less soluble, or bioavailable state. It minimizes the risks to the environment and people's health [11].

This paper reviews the remediation of contaminated soil using additives such as lime, cement, Metakaolin (MK), fly ash, and RHA (rice husk ash) by applying the Stabilization/Solidification technique. The methods of Stabilization/Solidification used chemicals and emulsions as supplementary soil compactors, binders, and water repellents that change soil behavior and are more suited and efficient in immobilizing contaminants in the soil.

## 2. Stabilization and Agents' Admixtures

Soil stabilization is a technique for altering soil properties and improving its performance for engineering purposes. This method utilizes admixtures, compaction, and soil densification. Chemical binders, industrial wastes, cement, and fly ash can all be used as additives.

### 2.1. Cement

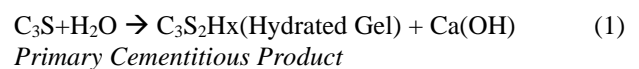
Portland cement is a traditional high-quality material. It can be considered the best material for stabilization, which has high strength and is widely available. Furthermore, cement is the most common binding substance in this

engineering field due to its easy handling and quality control features. Some of the most common cement varieties include blast furnace cement, ordinary Portland cement, cement that is sulfate resistant, and cement that is high in alumina. As well as manganese oxides (MgO) are also reactive materials that are used to stabilize heavy metals in the soil. It is more resistant and has significantly higher durability [12]. Shen et al. (2018) [13] observed that MgO significantly reduced the mobility of lead Pb(II) in the soil after just one day.

However, the engineering features of cement-stabilized soils were influenced by the number of additives, curing procedure, period, and soil type [12, 13]. Constantly changing water/solid ratios, temperature, size, particle, and other elements that impact the settings, strength properties, and long-term durability of solidified waste forms may increase the efficacy of cement-based solidification and stabilization [14].

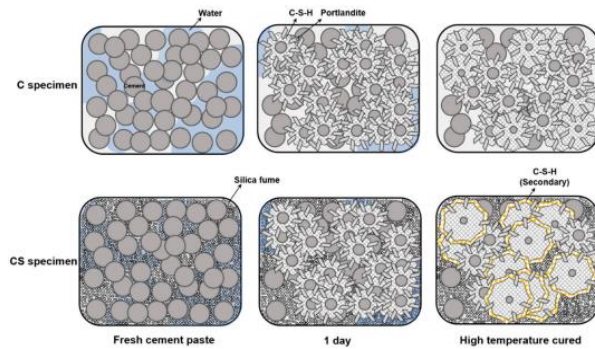
It is also known that the major cementitious components that are responsible for strong growth are tricalcium silicate (C<sub>3</sub>S) and dicalcium silicate (C<sub>2</sub>S) [15].

The reactions that occur during soil–cement stabilization is shown. The processes indicated in the following equations only apply to tricalcium silicate (C<sub>3</sub>S), one of the most crucial components of Portland cement.



The hydration of cement refers to the chemical process that occurs when cement and water are

combined. An exothermic reaction occurs during hydration [18], as shown in Fig. 1.



**Figure 1.** The cement hydration and pozzolanic reaction [18]

Contessi et al. [16] investigated the efficiency of using three different binders to stabilize lead in contaminated soil which are: Ordinary Portland Cement (OPC), Calcium Aluminate Cement CAC, and Metakaolin (MK). The efficiency of the suggested binders in the Stabilization/Solidification of the contaminated soil was investigated using leaching experiments, and lead release was assessed for every binding system. In the case of cement, lead was revealed to be linked to C-S-H. Furthermore, the CAC binder significantly contributed to the retention of this pollutant; utilizing a NaOH-activated MK produced lead retention rates of nearly 100%.

In addition, the experimental results show that stabilizing the soft clay soil with cement and fly ash enhances several significant features. The values are primarily comparable to those achieved by applying cement only.

Saeed et al. (2012) [20] studied the efficiency of cement-based Solidification/Stabilization for contaminated soil by heavy metals. The result appears that using heavy metals to mark the start of a cement hydration reaction is typical. Heavy metal hydrolysis decreases pH and hastens cement hydration, for example. Heavy metals may impact the protective hydrated layer's

structure, permeability, and characteristics and the generation and development of reaction products.

## 2.2 Lime

The other typical and traditional stabilizer is the lime used in roadway, railroad, and airport facilities to raise the maximum capacity of the layers. Additionally, it is used in tunnels and excavating as side support and a retaining wall fill material. Lime stabilization can also be employed to immobilize pollutant movement in contaminated soil due to its flexibility. The most frequent lime sources utilized in such applications are Quicklime, hydrated lime, and burned lime products (oxides and hydroxides, respectively). Lime improves through two major chemical processes, which are as follows:

### 2.2.1. Short-term reactions (modification or flocculation)

Hydrated lime calcium ions ( $\text{Ca}^{++}$ ) travel to clay particles' surfaces and absorb water and other ions throughout a short-term period. Consequently, the pore water's pH increased because of this reaction and the flocculation of soil aggregates. The soil becomes grainy and brittle, making it compact and easier to handle. This category of processes included cation exchange and carbonation.

### 2.2.2. Long-term reactions (solidification /stabilization)

The pozzolanic reactions are created by these processes[17]. Developing various cementing substances will bring the particles together and improve the durability of clay soils. Most previous research, it is important to note, [18, 19] concentrated primarily on the physical qualities of soil parameters. As a result, extensive investigation into the reactions of lime clay based on the microstructure was not very extensive.

Jaber, 2013[20] investigated how lime affected soft soil's geotechnical characteristics. In experiments for compaction, lime was applied at rates of 2%, 4%, and 6 % by weight of dry soil. liquid limit, specific gravity, and plasticity index. The findings show that increasing the lime content reduces the liquid limit, plasticity index, and specific gravity.

### **2.3. Fly Ash**

It is one of the waste materials that is used to improve soil quality. It is a very fine waste material produced by the smoke produced when coal is burned in coal-fired boilers at power plants. It is hazardous because of its multiple adverse health effects, such as lung sickness and pollution[21].

Fly ash is one of these extensively utilized to improve soil qualities. Fly ash represents approximately 80% of all coal ashes produced worldwide[22]. Various power plants generate around 38% of electric energy by producing damaging byproducts [23].

Calcium oxide has been the most critical factor in classifying fly ashes (CaO). According to Previous studies, fly ash class C contains a high percentage of CaO and has thus been used in various civil engineering applications and remediation of contaminated soil. Because it has cementations properties when combined with appropriate water[24].

Kadhim, H.J [25] studied the effect of using pozzolanic materials such as fly ash for remediation of contaminated soil lead using an activator. The results indicated that solid soil morphology showed no lead elements, demonstrating that lead compounds can precipitate and be encapsulated inside a rich silica cementation framework matrix.

### **2.4. Rice Husk Ash**

A pozzolanic material is RHA (rice husk ash). That can be used to stabilize Contaminated and weak, soft soil. If rice husk is incinerated at a controlled temperature, ash is generated, representing 17% to 25% of the rice husk's weight.

Koteswara et al. [26] studied the effect of adding rice husk ash, lime, and gypsum to the expansive soil, and the strength properties of the enormous soil were significantly improved. It was discovered that RHA, alone or in combination with gypsum and lime, might be able to regulate soil expansion.

With increased rice husk content for the soil, the unconfined compressive strength significantly increases and reaches its maximum at RHA between 6 - 8%. [31]

Utilizing industrial wastes like lime, RHA, and gypsum in rural locations may help reduce road construction costs.

Kadhim et al. used RHA (rice husk ash) as a stabilizer for contaminating the soil with lead after mixing it with an activator (NaOH + Na<sub>2</sub>SiO<sub>3</sub>) and using it as a geopolymer material. Results showed that the reduction in lead concentrations for leaching rate and efficiency was 96.68 %.

Eberemu [27] found that the rice husk ash improved the consolidation properties of the soil by up to 16 %. Increased rice husk ash concentration resulted in higher liquid and plastic limits and a lower plasticity index. The swell and compression indexes are reduced when the RHA (rice husk ash) contents are increased. As the ash content of rice husks rises, so does the coefficient of volume compressibility.

## 2.5. Silica Fume

It is a pozzolanic material with a high percentage of amorphous silicon dioxide and fine spherical particles. It is a fine powder produced as a byproduct of the manufacture of silicon and ferrosilicon metals, with an average particle diameter of 150 nm. The most common use in high-performance concrete is as a pozzolanic substance, as shown in Fig. 2.

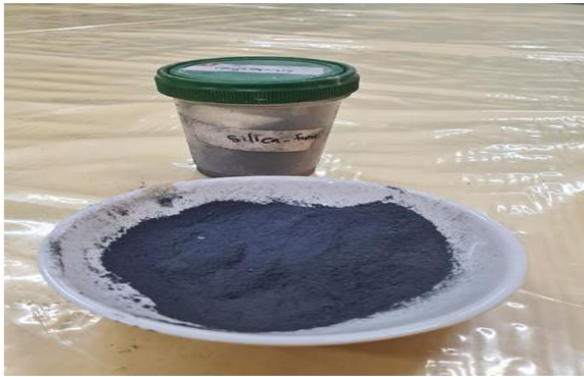


Figure 2. Silica fume.

Silica fume is one of the nanomaterials, while the density of nano-silica is  $1.16 \text{ g/cm}^3$ , and its specific surface area is  $80 \text{ m}^2/\text{g}$ . The additions were nano-silica particles with 99.9%  $\text{SiO}_2$  in nanoscale and silica fume  $\text{SiO}_2$  in the 0.5-1 micron range, employed as efficient pozzolanic materials. For use in construction, silica fume may fortify soil that is otherwise too weak or contaminated to be safe. It can also lower the presence of potentially dangerous heavy metals in soils that have already been contaminated. [33].

Al-Azzawi et al., 2012[28] It was researched how silica fume affected the technical properties of silty clay. Much laboratory research has been done on naturally occurring silty clay soil, and silica fume mixes made from silty clayey soil compacted at the proper soil wetness.

Saeed et al. (2013) [29] studied the effects of calcium-based stabilizers like lime on the

characteristics of clay soil investigated. It was shown that the strength of the samples treated with lime increased with time. On the other hand, cementitious products were noted to be present. These studies showed lime's ability to stabilize kaolin clay.

In order to stabilize artificially lead-contaminated silty clay soil, Kadhim et al. utilized silica fume with an activator. They came to the conclusion that the lead leachability through the matrix of solidified soil is proportional to the strength of the soil. During the TCLP leaching test, a significant amount of leaching was achieved. A crucial part of monitoring the chemical processes occurring in the soil during stabilization is the leaching test. Lead leachability is reduced to a minimum after 28 days, and As can be seen in Fig. 3, the UCS made with silica fume and alkaline activator demonstrated the greatest strength growth over time, as well as rising in strength with an increase in the quantity of silica fume.

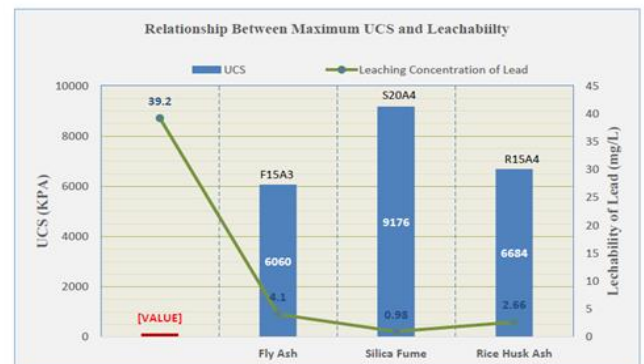


Figure 3. Maximum unbound components of soil and soil leachability after treatment [25].

## 2.6. Metakaolin

As shown in Fig. 4 below, MK is a species of calcined clay prepared by calcining kaolin clay. In recent years, considerable interest has been in using MK [30]. MK is generated by calcining (heating) kaolin clays at 600 to 800°C [31]. Calcination is an essential step in making highly

reactive pozzolanic material. In addition, the capacity of MK to react with hydroxide sodium in the presence of water to create hydrated silicate gel hydrate products is the essential sign of MK[32].



Figure 4. Meta kaolin.

MK has garnered much attention in recent years [30]. The effectiveness and method of stabilization/solidification of heavy metals in polluted soil using a metakaolin-based geopolymer were investigated by El-Eswed et al. [33]. The results showed that Metakaolin-based geopolymer (MKG) is very effective in stabilizing heavy metal ions during the leaching reduction.

Zhang et al. 2013[34] found that all specimens were created according to Optimum Moisture Content(OMC) and cured for 7 and 28 days after stabilizing clay with (MKG) at varying percentages (i.e., 3 to 15%) by weight of dry soil. The experiments demonstrated that the UCS value increases over time, but the rise is insignificant between days 7 and 28. Shrinking values were decreasing. The microstructure of the developing gels in the treated soil was also examined, as shown in Fig. 5.

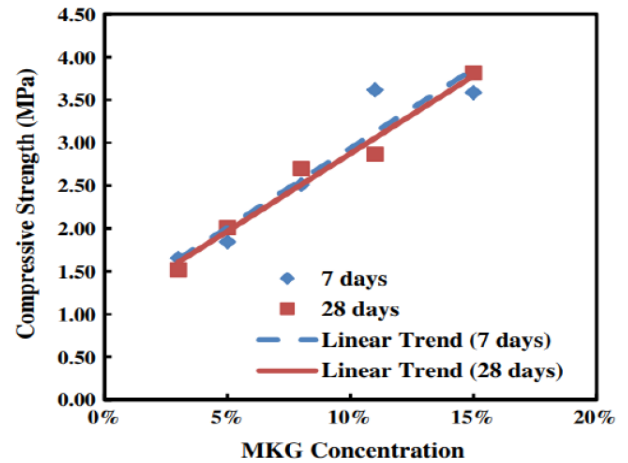


Figure 5. The effect of MKG on UCS[34].

In order to stabilize expansive clay soil and increase soil strength, Samuel et al. 2020 employed MK with varying concentrations and stabilization durations. Experiments show that geopolymers significantly enhance expansive soils' strength, stiffness, and volume-change characteristics. Improvements in properties are demonstrated in Fig. 6 as being considerably more pronounced now that content and cure durations have been increased [35].

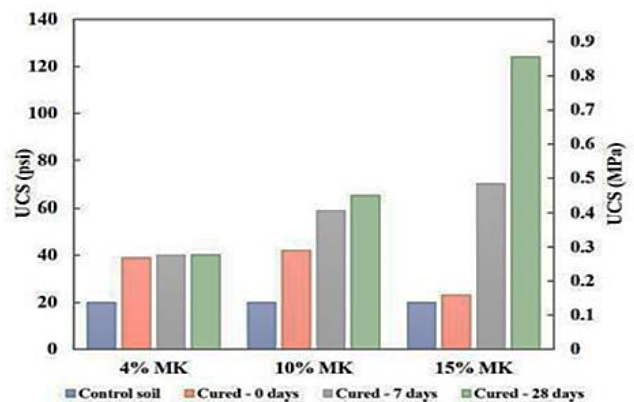


Figure 6. The effect of MK on UCS[35].

### 3. Mechanism of Stabilization /Solidification

Stabilization/Solidification is a procedure that includes mixing waste with agents to reduce the number of contaminants. These contaminants can leach through physical and chemical qualities to transform waste in the environment intended for landfill or other potential routes,

despite several past immobilization technologies that have been used to implement remediation methods. The Stabilization/Solidification process is one of the most optimistic.

Depending on the construction process, mechanical and chemical stabilization are the two main soil stabilization methods. Mechanical stabilization aims to fill any holes in the soil, compared to chemical stabilization, which increases strength while reducing permeability.

Nevertheless, chemical stabilization happens when the heavy metals change into metal carrier phases that are more stable and less soluble. Chemical stabilization alters the chemical mobility of waste components to a condition with lesser aqueous solubility, morphological alterations, and contamination in the soil [36].

When waste undergoes physical stabilization, sometimes called solidification or encapsulation, the physical structure is altered without forming chemical bonds between the constituents. Furthermore, adjustments were made to the mechanical properties.

Chemical stabilization of inorganic contamination in clays may occur through various reaction routes, with consequential effects on binder modification and solidification processes. The cation exchange reaction considers the first bonding between metal pollutants and clays.

Heavy metals might also be solidified by precipitation, which occurs when certain metals precipitate in alkaline environments.

The Environmental Protection Agency's Fig.7 illustrates the metal fixation methods involved in stabilization and solidification [37].

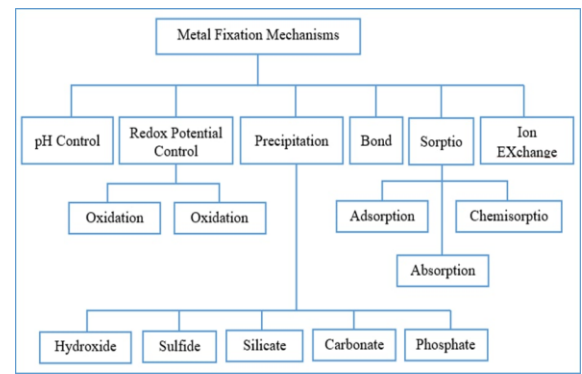
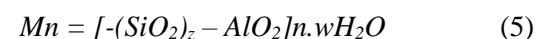


Figure 7. The Mechanisms of Metal Fixations[37].

#### 4. Principle of Geopolymers

The geopolymer hypothesis for alkali aluminosilicate binders was developed in 1976 by Joseph Davidovits. Active silicon and aluminum are present in large quantities in geopolymers. Steel slag, bauxite, waste glass, volcanic ash, diatomite, coal gangue, and high-magnesium nickel slag are all good examples of geopolymer raw materials. In addition to MK, fly ash, blast furnace slag, and biomass ash are utilized [38]. Various advantages of geopolymers include their capacity to resist fire, and chemical corrosion, high mechanical strength, and high durability [39-44].

A three-dimensional polymeric chain and ring structure formed of Si-O-Al-O linkages are produced as a result of the relatively quick chemical reaction on Si-Al minerals in a strongly alkaline environment during the polymerization process.:



Where:

M = A cation or alkaline element such as sodium, potassium, or calcium; the symbol – indicates the presence of a bond.

n = the level of polymerization or polycondensation.

The utilization of various aluminosilicate sources became necessary to understand the impact of various contaminants on binder formation,

leading to research into the effects of various additives on Geopolymer material resources and formation chemistry, as shown in Fig. 8 [51].

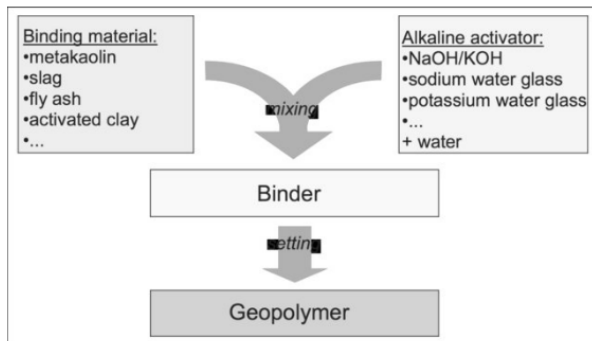


Figure 8. Production of geopolymer[45].

## 5. Conclusions

This study discusses the potential application of pozzolanic-based polymers for immobilizing the concentration of heavy metal-contaminated soil. It seems that soil Stabilization/Solidification is the best approach for the remediation of contaminated soil and the improvement of soft clay and different of type soil. Contaminants are enclosed in a monolithic matrix and formed into solid forms using pozzolanic materials. Geopolymers mainly solidify heavy metals through physical encapsulation and chemical bonding. In addition, these pozzolanic reaction products, which fill the pores, modify the pore size distribution or pore structure. The permeability of the binder is hence decreased. In addition, it is possible to polymerize alkali polysilicates and alumino-silicate oxides to produce Si-O-Al links. It can subsequently be used to make geopolymers. Source materials and alkali liquids are the two significant components of geopolymer binder. Typically, solutions of sodium or potassium make up the alkali liquids. Clays, fly ash, metakaolin, slag bottom ash, and rice husk ash are all examples of byproducts or geologically derived materials high in silicon (Si) and aluminum. Therefore, geopolymer is a more environmentally friendly binding medium than

traditional Portland cement since it produces less greenhouse gas.

## Conflict of interest

The authors declare that there are no conflicts of interest regarding the publication of this manuscript.

## Author Contribution Statement

Nidhal S. Jama developed the theory and performed the experiments. Khitam A. Saeed verified the analytical methods and suggest the research idea.

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