

Review Research

A REVIEW OF SOLIDIFICATION/STABILIZATION OF HEAVY METAL CONTAMINATED SOIL

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Received 11/4/2022

Accepted in revised form 6/7/2022

Published 1/3/2023

Abstract: Heavy metal contamination has occurred as a result of industrial civilization. Zinc, copper, chromium, and lead are the most prevalent heavy metal pollutants. Heavy metal contamination has arisen as a significant environmental issue on a global scale. Human and environmental health is at risk when soils are contaminated. as well as having poor engineering qualities. Solidification/Stabilization is a critical remediation strategy for polluted soils which is both efficient and cost-effective. The solidification/stabilization approach has been frequently used to rehabilitate heavy metal-contaminated areas. First, The use of gypsum to strengthen and leach polluted soils was reviewed. Also, cement/fly ash-solidified/stabilized soils have better engineering qualities. On the other hand, the global output of phosphogypsum surpasses 300 million tons, raising disposal and environmental problems every year. The efficiency of the phosphogypsum-based stabilization/solidification technique was investigated, and the methods employed biochar and chemical agents such as citric acid and FeCl₃. This review examines various remediation options as well as innovative soil amendments.

Keywords: *Sustainable remediation; solidification; stabilization; heavy metals; industrial civilization.*

1. Introduction

Stabilization/solidification (S/S) is frequently used to treat soils contaminated with heavy metals through various techniques. The treatment reduces the amount of toxic compounds released

from hazardous wastes by solidifying and stabilizing them. This was initially designed for sludge management in the late 1950s but has been modified for soil remediation[1].

Due to the industrial revolution, heavy metal contamination was found in various locations. This contamination arises from various factors, including inadequate waste management, chemical leaching, and exposure to air contaminants.

Zinc, copper, chromium, and lead are the most prevalent heavy metal pollutants[2]. Polluted soils are not only hazardous to the environment and public health, but they also have worse technical characteristics[3]. Therefore, When dealing with heavy metal-contaminated soils, it is imperative to use cost-effective and efficient remediation methods to reduce environmental consequences and increase engineering qualities.

Heavy metals may be removed or stabilized using a variety of methods. Conventional soil remediation methods, such as Portland cement stabilization, thermal desorption, and chemical oxidation/reduction have proven effective in

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treating soil [4]. However, they have been admonished for their high energy consumption, greenhouse gas emissions, long-term metal leaching dangers, pollution, and economic and social challenges such as low customer acceptability. The solidification and stabilization method (chemical treatment) significantly enhances the soil's mechanical properties and can be used as a remedial method for soil degradation[5-9]. The solidification and stabilization technique has been losing market share in recent years, particularly at Superfund sites, where it has been used less often .

In addition to enhancing soil structure, stabilizing agents (e.g., biochar) may supply nutrients and reduce acidity induced by mineral fertilizers. For long-term cleanup, stabilization materials might include controlled-release reactants or microorganisms. However, in the field, monitoring is frequently overlooked when it comes to long-term S/S performance. Solidification and stabilization are more cost-effective than other upgrade options [10, 11]. Metal leakage is reduced when dangerous materials are encircled and protected from corrosion[12, 13].

Building complex advanced ageing strategies that combine environmental pressures will take more work. In recent years, data mining, enormous data, and sensor technology advancements might serve as a source of inspiration for researchers working across disciplines .

This paper presents a review focused on soil remediation processes and soil treatment techniques using various solidification and stabilization technologies.

2. Heavy Metal Solidification–Stabilization Using Gypsum:

OPC (Ordinary Portland cement) is often used in the S/S method. The presence of copper oxides and zinc oxides in polluted soil inhibits cement hydration. When polluted soils are treated with cement, contaminants may significantly weaken their structural integrity[12, 14]

Even though producing OPC requires enormous energy, it contributes to climate change by releasing greenhouse gases into the environment[15-18]. About 1 ton of CO₂ is released for every ton of OPC consumed[19, 20]. As a result, it is critical to study new environmentally favorable solidification agents that might result in cost-effective but environmentally friendly solutions. According to recent studies, low-carbon alternatives to OPC with equivalent soil stabilizing technical qualities include Portland limestones cements (PLC), lime kiln dust, cement kilns dusts, and geopolymer[21-26].

Currently, researchers are examining low-carbon alternatives to binders. Various by-products are created when calcium carbonate and sulfuric acid combine to make sulfuric acid in an alkaline solution. One of these materials is gypsum, produced during sulfidic oxide oxidation[27]. Since ancient times, gypsum plaster has been used in construction [28-30]. Researchers are looking for alternatives to traditional binders with less carbon content. Aside from gypsum, made from the sulfide oxidation reaction's by-products, several alternatives are made from the pyrite oxidation process, in which sulfuric acid is formed and combined with calcium carbonate. Adding gypsum improves bentonite's mechanical properties[27]. Bromwell and Carrier Inc[31] proved that treating phosphate soil with gypsum increased its strength. Due to its low cost

and water solubility, Bell and Maud[32] and Ameta[33] proved the efficacy of a very fine gypsum powder as a binder.

Polluted soils containing heavy metals are a concern in various locations next to industrial zones.

Heavy metals like zinc, lead, nickel, and chromiums have been the subject of much research. Even though the consequences of soil pollution with these metals have been extensively studied, a lack of data on the impact of copper pollution on clays is a significant issue by Aziz et al. [34] and Kumpiene et al.[35]

Waste copper (Cu) disposal generates a substantial amount of Cu-containing pollutants.

Copper overload in soil may be deleterious to human health, biota, environment, groundwater, and agricultural production[36, 37]. In polluted places, soils are frequently varied and can contaminate many pollutants.

Latifi et al.[20] examine the possibility of using gypsum to increase the engineering qualities of copper-contaminated clays. According to compaction experiments, adding gypsum improved the ideal moisture content while lowering the maximum dry density. However, there are data on the mechanical and physicochemical properties of heavy metal-contaminated clay soils treated with gypsum. As a consequence, more investigation is necessary.

3. Stabilization/Solidification of Heavy Metal Using Chemical Interactions and Encapsulation, Binders

Heavy metal pollution of soils is becoming one of the most critical environmental problems[37, 38]. In contaminated soils, heavy metals such as nickel (Ni), zinc (Zn), copper (Cu), manganese (Mn), lead (Pb), and cadmium (Cd) are frequently detected.[39]

Heavy metal-polluted soils are routinely remedied using the S/S method [39-42]. Chemical interactions and encapsulation are utilized to stabilize the soil's chemical composition by immobilizing heavy metal components prevalent in polluted soils.

Lime (CaO or Ca(OH)₂) and Portland cement are the most often utilized binders in the S/S process (OPC) [43-48]. Due to their alkaline nature, these binders can precipitate (i.e., hydroxides or insoluble complex compounds) [49]. Calcium silicate hydrate (CSH) and calcium aluminate hydrate (CAH) are examples of compounds that may absorb heavy metals by hydrolysis processes, such as calcium silicate hydrates (CSH) [50]. Lime or OPC production, on the other hand, requires a large amount of energy and CO₂ emissions [51]. It is also possible to reduce the immobilization of heavy metals such as Zn, Pb, and Cu by creating high alkaline conditions (pH N 11) using OPC or CaO[52-55].

Due to its low production temperature, capacity to consume CO₂ [53, 54], and around 10 pH, reactive magnesia (MgO) has garnered considerable interest. However, most previous studies [55-57] remedied contaminated soils using a combination of OPC, MgO, and other industrial by-products (slag or fly ash). It is difficult to isolate the S/S efficiency of MgO from other mixed compounds when it comes to heavy metal immobilization. This is because, in addition to MgO, other mixed minerals create hydration products such as calcium silicate hydrate (CSH)-like compounds [58-61], which may help immobilize heavy metals.

Combining slag, MgO, fly ash, and/or OPC can assist minimize heavy metal leachability. It is associated with forming heavy metal carbonates [62]. MgO is an effective heavy metal immobilizer[60]. Therefore, additional study is

needed to understand how MgO-treated contaminated soils immobilize contaminants. The impact of MgO alone on the S/S of heavy metal-contaminated soils has only been the subject of a small number of research[59-63]. The findings demonstrate that although MgO generates a weaker soil than OPC or CaO, it is more successful at immobilizing heavy metals[59]. The effectiveness of MgO in lowering S/S in soils polluted with a single heavy metal is unclear since these investigations involved a mix of heavy metals. The S/S efficacy of CaO, MgO, and OPC on contaminated soils cannot be compared.

4. Phosphogypsum in soil stabilization /solidification

The widespread use of cement stabilization/solidification in the remediation of heavy metal (HM) contaminated soils may not always be successful. For this reason, there is a need to examine further methods to stabilize the soil[2, 64]. A mix of techniques is necessary to remediate polluted soil cost-effectively and sustainably by limiting contaminant movement [65]. Solidification/stabilization is the most effective and widely utilized approach for remediating areas polluted with high levels of dangerous metals. It entails converting a hazardous component into a new non-toxic product or encapsulating the pollution in a solid matrix that inhibits leaching and is chemically stable. The leaching and toxicity of heavy metals are minimized, as is the environmental load associated with waste management and resource recycling [12, 66-69].

There are vast stockpiles of the fertilizer industry by-product, phosphogypsum, that pose environmental and human health problems since they are not used as quickly as they are created. Chavali [70] investigate phosphogypsum as a

soil stabilizer, agricultural fertilizer, and controller in cement manufacturing, road construction, and building materials [71, 72]. Phosphogypsum is a stabilizer that may be applied alone or in combination with other compounds to enhance the design behavior of soils [71, 73-75]. For example, De Rezende [76] observed that when coupled with cement and lime, hemihydrate phosphogypsum rather than di-hydrate phosphogypsum swelling enhanced lateritic soil's strength. To a large extent, the stabilizing ability of phosphogypsum depends on soil mineral composition, except kaolinite, which improved more than silicate minerals. Dihydrate phosphogypsum content must be maintained below 20% when stabilizing lateritic soils, as higher amounts have a negative impact on mechanical conduct and may pollute groundwater [77]. Fig. 1 illustrates the difference in phosphogypsum's maximum dry density and optimal moisture content. As it turned out, stabilizing 20 soils from various sites with a maximum California Bearing Ratio (CBR) value of 20% required a phosphogypsum concentration of 20%.

The quantity of phosphogypsum used in pavement applications should be maintained within a suitable range, as mentioned by Ding et al.[78]. Mashifana et al.[79] found that stabilizing expansive soil with phosphogypsum-lime fly ash-basic oxygen furnace slag paste improved engineering behavior by producing various calcium magnesium silicates and coating them with calcium silicate and calcium anhydrite hydrates. Similarly, calcium silicate hydrate gel production enhanced soil behavior when phosphogypsum was coupled with other chemicals [77-80]. Excessive use of phosphogypsum may lead to heaving, and structural disturbance, weakening cement stabilized dredged soil, according to Zang et

al.[81]. It has also been used to counteract the effects of phosphoric acid on soil volume[70].

Esawy and Nasser [82] found that phosphogypsum reduced leachability more effectively than a phosphogypsum-rice straw binder when used to immobilize heavy metal-contaminated soils. In acidic soil, heavy metals such as Cd, Cu, and Pb may be reduced in leachability by phosphogypsum, according to Illera et al.[83] Wang et al.[84] proved the efficiency of phosphogypsum in terms of reduced Pb leaching and higher early strength in soils polluted with Pb. As long as phosphogypsum has been widely used to improve soil behavior, nothing is known about how it affects the solidification and stability of heavy metal-contaminated soil.

Phosphogypsum is an effective treatment for soils polluted with copper [85]. Copper-contaminated soil was shown to be weaker than soil treated with phosphogypsum. Sulfate copper minerals with high soil retention and low soil copper leaching demonstrate the efficacy of phosphogypsum.

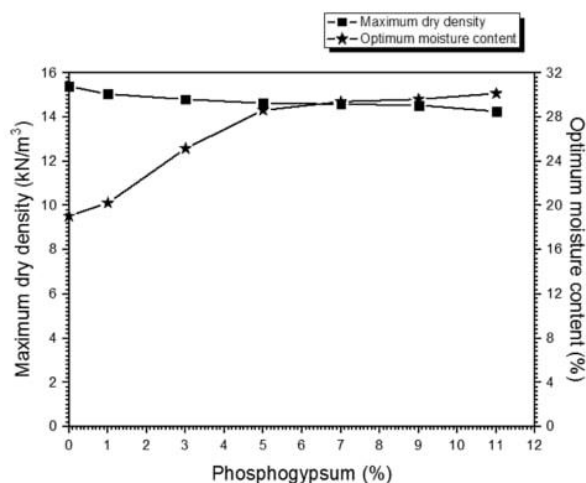


Figure 1. Soil compaction characteristics with varying phosphogypsum levels[85].

5. Stabilization Using Biochar

Heavy metals (HMs) have been released into the environment by human activities such mining, smelting, milling, pesticide and fertilizer use, wastewater irrigation, and sludge since the Industrial Revolution [86-88]. Soil HM pollution has been a concern for decades [89, 90], accounting for 82.8% of pollution [91, 92]. Soil microbial diversity, water quality, and agricultural production are mostly impacted by soil HMs [89-93]. Because HMs do not degrade in soils and may build up in agricultural products via the food chain, prolonged exposure to them is dangerous for human health and the environment[94-96].

Effective and environmentally friendly remediation strategy [97]. Apatite composites, phosphoric fertilizers [97], lime[98], iron and ferric salts [99]. These amendments can potentially lower the activity of heavy metals in soils and the accumulation of heavy metals in plants to some degree [100]. However, China has yet to begin a large-scale cleaning of HMs in agricultural soils owing to economic restraints, inefficiency, a lack of specificity, and the danger of secondary contamination[101, 102].

Developing innovative, highly precise, efficient, and environmentally friendly additives for soil remediation is essential.

Biochar, a fine-grained, porous, and carbon-dense substance, has been the focus of recent studies [103, 104]. Several investigations have been conducted and published [105, 106]. Biochar may help stabilize heavy metals and prevent plants from collecting them in their tissues as a soil amendment. There was a 56.5, 50.0, and 54.0 percent reduction in soil-accessible Cd, Pb, and Zn when added sugarcane straw-derived biochar [107]. For example, according to the findings of Houben et al.[86],

biochar decreased bioavailable Cd and Zn by 71% and 92%, respectively; (CaCl₂-extraction). Biochar increased biomass output by three times. Biochar has also been changed to improve cleanup effectiveness. Manganese oxide (MnO) was added to charcoal to stabilize arsenic (As) in the soil, according to [108]. Numerous rice fields saw arsenic levels fall due to arsenite oxidation to arsenate as a consequence of the usage of biochar.

It has been shown that phosphorus-loaded biochar reduces the availability of HMs in soils and transforms them into more stable components. The variables that affect the characteristics of biochar are shown in Fig. 2. Biochar that is high in phosphorus was employed by Ahmad et al. [109] to limit the availability of HMs in soils. They noticed that it made HMs into more stable components and reduced the amount of labile HMs in soils.

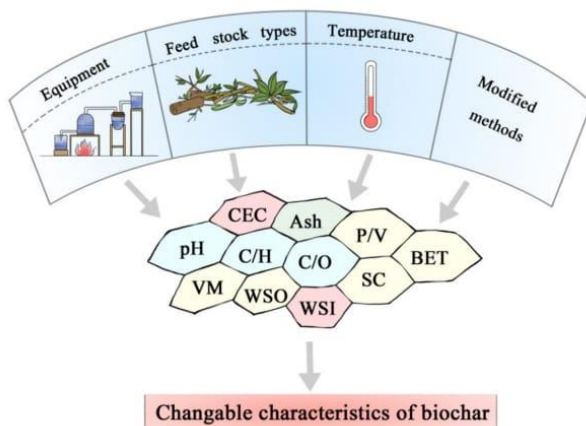


Figure 2. Factors determine the properties of biochars. “WOS: water-soluble organics; WSI: water-soluble ions; SC: surface charge; VM: volatile matter; P/V: pore-to-volume ratio”[69].

Biochar, on the other hand, may not necessarily be beneficial. In most investigations, the capacity of biochar to help remediate soils polluted with heavy metals was highlighted [108]. Few studies have examined the negative impacts of biochar

on soils or HMs, which may lead environmental engineers to believe that biochar is a one-stop solution for soil improvement. Biochar’s possible disadvantages and limits in soil remediation must be addressed to give a vital reference for biochar application [110]. Before employing biochar in soil restoration, several potential drawbacks and limitations must be considered.

Researchers Wang et al. [69] investigated biochar application’s adverse effects and challenges. Contrary to popular belief, the main drawbacks of using biochar in soils are activating some heavy metals and the non-specific adsorption of certain heavy metals. If these difficulties are resolved, they believe biochar can be a cost-effective and environmentally friendly complement to HM-contaminated soil restoration. Biochar’s broad application has been facilitated by developing new study areas and ideas.

6. Dry Jet Mixing Method With The Hydroxyapatite-Based Binder For Solidification/Stabilization

employing a new hydroxyapatite-based binder and a dry jet mixing process. Cement was often used as a binder material in the solidification/stabilization of heavy metal-contaminated soils (such as cadmium (Cd), zinc (Zn), and lead (Pb))[111]. However, using cement in the solidification/stabilization (S/S) process has a number of disadvantages that restrict its efficacy in immobilizing heavy metals and its use in field applications:

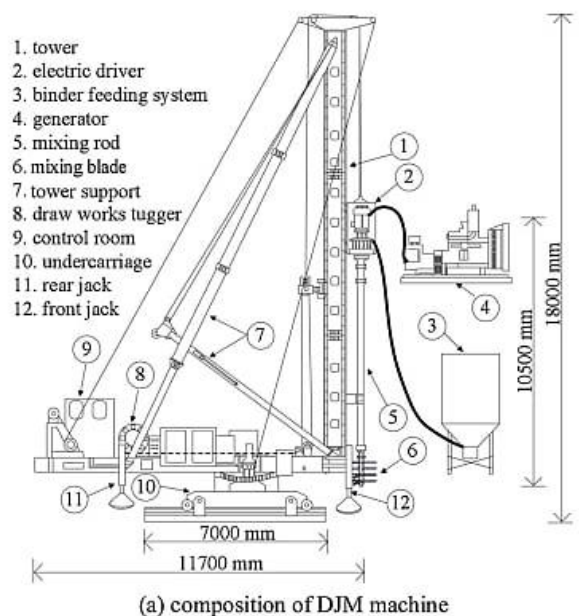
The bulk of heavy metals are found in the form of pH-sensitive metal hydroxides, such as Zn(OH)₂ and Pb(OH)₂, in soils that have been treated with cement. Metal hydroxides are usually amphoteric, and their solubility rises as the acidity or alkalinity of the solution

increases[112]. Adverse environmental impacts of cement manufacture. Due to the significant impact of cement manufacturing on air pollution (NO_x, SO₂, CO, H₂S, and particulates) and energy consumption (particularly greenhouse gas emissions), the area surrounding cement factories suffers from high levels of pollution (particularly CO₂)[113]. Challenges associated with applying cement to soils containing relatively high levels of heavy metals. Recent research by Lu et al. [114] indicates that zinc and chromium (Cr) has a detrimental effect on the performance of cement-stabilized soil (Zn 0.2 percent and Cr 1.5 percent, respectively). Cement-treated soils are highly susceptible to environmental changes. Numerous factors, including acid rain, freeze-thaw cycles, and sulfate bombardment, may negatively affect the ecosystem[115].

The authors developed new hydroxyapatite (Ca₁₀(PO₄)₆(OH)₂)-based binder for solidifying and stabilizing soil polluted with heavy metals using mixing blades. A powder binder is pneumatically delivered and blended with the soil in this field investigation, resulting in overlapping columns of soil-binder mixture. Dry Jet Machine (DJM) composition and mixing head structure are depicted schematically in Fig. 3 (a) and (b) [116]. SP stand of superphosphate, whereas C is calcium oxides (CaO). In the laboratory, stabilized contaminates soil in 4% - 10% SPC shown considerable improvements in leaching behavior and strength properties [116]. SPC converts heavy metals such as zinc, lead, and cadmiums into insoluble hydroxyapatites and phosphate-based precipitates as soon as it comes into touch with them [116].

For the reasons below, in-situ treatment performance cannot be deduced directly from laboratory test findings. To guarantee reliable

findings, laboratory testing must be conducted under precisely calibrated settings, which include meticulous sample preparation and temperature and relative humidity control throughout the drying process. In contrast, in-situ field treatment often faces soil heterogeneity and temperature and relative humidity fluctuations. Zang et al. [117] conducted a laboratory-scale testing that confined to 28 days of curing, but the performance of SPC-treated soil with a longer curing duration has been unknown. Consequently, field research is required to establish the time-dependent performance of SPC-solidified/stabilized contaminated site soil. Studies on time-dependent performance characteristics of SPC binder, such as heavy metals and organics leachability, as well as the effectiveness of SPC binder in remediating extensively polluted site soils, are urgently required to close these research gaps.



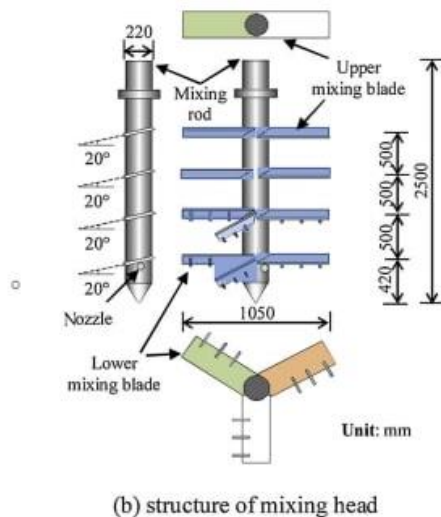


Figure 3. Schematic of the DJM used in the field trial: (a) DJM machine composition; and (b) machine mixing head construction[118].

7. Low-temperature Thermal Desorption Stabilization

Due to Hg's high volatility, thermal desorption is beneficial in reclaiming polluted soil.

However, the high temperature ($>600^{\circ}\text{C}$) required for this process indicates that it is relatively expensive and has a significant carbon footprint [119]. Additionally, The considerable changes in the treated soil's physicochemical qualities make it useless for agricultural uses[120].

Low-temperature thermal desorption solutions are required to minimize the primary and secondary issues and increase the "net environmental benefit" [121]. Citric acid and FeCl_3 were used to lower the heating temperature while maintaining Hg removal effectiveness [122]. Citric acid or the production of volatile Hg species at low temperatures may aid in Hg elimination at these temperatures (i.e., HgCl_2 and Hg_2Cl_2). Mercury may be removed via microwave-induced thermal desorption.

By employing microwave irradiation (400 W for 40 minutes) using granule-activated carbon as a

microwave receptor, Cao et al. [123] demonstrated that soil heated to 350°C could be effectively removed mercury (Hg) (i.e., 87 percent). A specific soil particle was found to have an external temperature substantially lower than its inside temperature. Because of the lowered heat transfer barrier, off-gas may travel much more readily from the soil (relative to traditional desorption procedures)[124, 125].

In contrast, using low-temperature thermal desorption procedures, the same Hg removal rate may be achieved in a much longer period. The long-term viability of the process may be threatened by applying chemical agents or GAC. Low-temperature thermal desorption must be studied to determine its cumulative impact. Using citric acid for agricultural soil thermal desorption, Hou et al.[121] demonstrated that using citric acid for low-temperature thermal desorption may reduce greenhouse gas emissions from 357 kg CO_2 to 264 kg CO_2 .

A further benefit of low-temperature thermal desorption is the ability to reuse soil right where it was extracted. Only on a bench scale has this process been shown to be practical. A complete picture of this energy-efficient technology's potential and deeper and more specific insights should be gained via more field investigations

8. Conclusions

Soil pollution with heavy metals has sparked widespread concern due to the potential damage it might do to both people and ecosystems. A chance exists to investigate and put into practice long-term, cost-effective soil remediation methods. This study's goal was to review some of these corrective treatments. Since gypsum is both cost-effective and ecologically friendly, it is a viable option for solidification/stabilization (S/S). In addition, a mixture of MgO , slag, fly ash, and/or OPC helps reduce heavy metals'

leaching. Phosphate gypsum may enhance soil behavior and reduce the leachability of heavy metals, including Cd, Cu, and Pb in acidic soil. As a soil amendment, biochar may help stabilize heavy metals and prevent plants from collecting them in their tissues. Chemical agents such as citric acid and FeCl_3 were used to lower the heating temperature to 400°C while still removing Hg. More studies on solidification/stabilization methods and strategies are required.

Conflict of interest

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

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