Solidification/stabilization for soil remediation: an old technology with new vitality

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Solidification/stabilization for soil remediation: An old technology with new vitality

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Solidification and Stabilization (S/S) treatments limit the release of harmful chemicals from hazardous wastes. This approach was developed in the late 1950s for the management of sludge, and later adapted to soil remediation. It became the number two soil remedy in the U.S. Superfund (CERLA) program, next only to physical separation,[1] and subsequently gained popularity in Canada and the U.K. in the 1990s, and in France and the Netherlands in the 2000s. In recent years, S/S has been losing its market, with significantly reduced usage at Superfund sites (Fig 1.). Concerns over long-term effectiveness coupled with an overall decline in remediation in North America and Europe have hit this technology severely. Moreover, its use in countries like South Korea and Denmark is prohibited because of requirements to remove contaminants from soils. There is, however, one country where S/S is burgeoning, China. In the last year (2017-2018), the Chinese remediation market doubled (to US$2.9 billion), with S/S massively leading the field (48.5% adoption rate).[2]

The popularity of S/S among remediation practitioners owes to its ability to achieve remediation objectives rapidly and at relatively low cost. It is also versatile, being applicable *in situ* or *ex situ*, and effective for a wide range of common inorganic and organic contaminants. However, too much focus has been placed on practicality and short-term effectiveness, while long-term effectiveness and sustainability concerns are overlooked. For example, sulfate and acidic rain erode Portland cement (PC) based treatments, and certain contaminants, such as Pb, react with calcium hydroxide and form complex mixtures (e.g., $Pb(OH)_2$ and $PbOPb(OH)_2$) that impede PC hydration.[3] Moreover, such durability concerns are linked to sustainability because life cycle impacts multiply if S/S treatments fail and need to be reapplied. The extensive use of PC as a binding agent also aggravates carbon footprints. Each metric ton of PC mixed into soil is associated with the release of more than 900 kg of CO_2 (e.g., as part of the production of PC), and globally PC production accounts for 10% of anthropogenic CO_2 emissions.[4] It is not uncommon for excessive amounts of PC to be used to achieve unreasonably high strength and unnecessarily low contaminant leachability levels. This "over-dose" problem could be averted by the use of high-performance S/S materials developed for both sustainability and long-term performance and applied under appropriate guidelines.

Recently, greener cement binders for S/S purposes have been subject to increased levels of research (Fig. 1). These cements consist of low-carbon and low-cost materials. Magnesia (MgO) based cements not only offers these benefits but are also resistant to acid and sulfate erosion. Recent developments in self-healing cementitious materials suggest potential for durable and resilient cement-soil systems. Cracks could be self-repaired, for instance, by the presence of microcapsules incorporated into cements, which release healing agents on demand. Cement binders that incorporate waste additives, such as slags, fly ash, or phosphogypsum align with the circular economy concept of using waste to treat waste. The use of slags is also beneficial for the generation of insoluble (C-S-H) gels during cement hydration, which lessen pH increases and simultaneously enhance metal stabilization and strength.

The stabilization aspect of S/S involves chemical reactions that immobilize contaminants. Stabilization without solidification promises a way to deal with the vast areas of Chinese agricultural land with elevated heavy metals or organic pollutants (~135 million ha).[5] A wide range of novel stabilization materials, e.g., biochar, ferrous sulfate, layered double hydroxides (LDHs), apatite, clay minerals and their modified products, are currently being researched for this purpose (Fig. 1). Some stabilizing agents (e.g., biochar) may improve soil health, by improving soil structure, adding nutrients (N, P and K), mitigating acidification caused by mineral fertilizers, and increasing water holding capacity. Stabilization materials could also incorporate controlled release reactants/microbes for enhanced remediation in the long-term.

The ability to accurately predict long-term S/S effectiveness is necessary for accurate design and the selection of optimal binders and stabilization materials. There are various bench-scale accelerated ageing tests that can be used for this purpose, but limited field data to validate these simplified models. In addition, most ageing methods do not provide temporal performance data. Quantitative temporal simulations have proved challenging because of the ever-changing and heterogeneous dynamics of the natural environment. Therefore, most physical, chemical, or biological ageing tests only consider single environmental factors. More research effort is needed to develop multifaceted advanced ageing methods that couple different environmental stresses. These could be supported by artificial intelligence (AI) to help determine critical environmental stresses and their impact, e.g. wet-dry and freeze-thaw tests with variable temperatures, rainfall frequencies, precipitation levels, and freeze period factors. Climate change predictions could also be factored in.

Ensuring S/S long-term performance requires in-the-field monitoring, but this is often neglected. This may change as long-term consequences/failures are further realized. Technological innovations are key to providing refined and timely monitoring data. Researchers could look to interdisciplinary breakthroughs in data mining, big data, and sensor technology for inspiration. For instance, robust wireless sensors could provide real-time strength data to indicate treatment integrity. X-ray fluorescence (XRF) spectrometry could accurately provide soil metal concentrations within minutes - a decreasing trend would indicate a stability issue. Advanced microscopic, spectroscopic and mineralogical analyses could help us to understand S/S modes of alteration, alteration pathways, and influencing factors. Such analyses are not only valuable for directly evaluating S/S permanence at a given time, but also for providing accurate data for predictive modelling (e.g., thermodynamic and geochemical modelling). Such studies on S/S treated soils are currently limited and require effective collaboration among environmental chemists, material scientists and engineers.

In conclusion, S/S technology can be much improved by adopting more efficient and sustainable remediation materials, lowering their dosages to achieve reasonable remediation goals, and enhancing the predictability of the S/S systems. This old technology will be revitalized for better soil remediation.

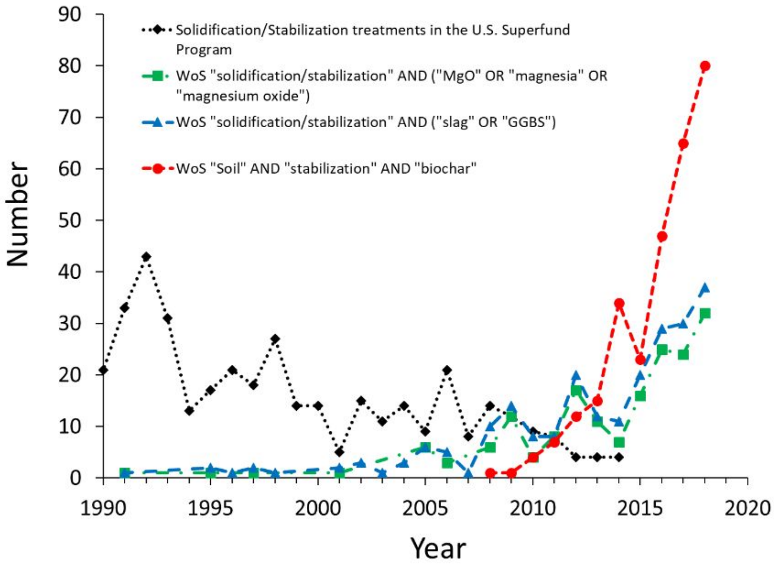

Figure 1 Number of solidification/stabilization treatments in the U.S. Superfund program (CERLA) and research articles in Web of Science (WoS) by year. The Superfund data is extracted from US EPA (2017),[1] WoS was searched on 02 September 2019.

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