Vertical Barriers for Land Contamination Containment

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Soil pollution is one of the major threats to the environment and jeopardizes the provision of key soil ecosystem services. Vertical barriers, including slurry trench walls and walls constructed with soil mix technology, have been employed for decades to control groundwater flow and subsurface contaminant transport.

Keywords: in-ground barrier ; cut-off wall ; land contamination ; soil pollution

1. Introduction

Industrial activities have increasingly been causing severe environmental impacts on air, water, and soil. Toxic chemicals, including heavy metals, herbicides, pesticides, and other organic contaminants, can be absorbed by human beings and lead to health problems when they are released into the environment [1][2][3]. The Food and Agriculture Organization of the United Nations (FAO) and United Nations Environment Programme (UNEP) have recently reported that soil pollution is one of the major threats to the environment and jeopardizes the provision of key soil ecosystem services, including the provision of safe and nutritious food, the availability of clean water, and the existence and conservation of soil biodiversity [4].

Since the industrial revolution, land contamination has posed a serious challenge to developed countries such as the UK, which has over 200,000 potential contaminated sites [5]. Land contamination can be even more severe in developing countries. In China, for instance, a government report showed that 16.1% of all tested soil samples were polluted, covering 6.3 million square kilometers [6]. It is, therefore, important to effectively manage contaminated sites and mitigate the threat posed to public health and the environment.

Land contamination risk management has grown from a niche field into a booming business, becoming a multi-billion dollar industry in many developed countries and a flourishing market in developing countries [7][8][9]. There are three primary risk management approaches by which the ‘source–pathway–receptor’ pollutant linkages can be broken: (1) source removal or soil cleaning via treatment; (2) pathway management; (3) modification of exposure of the receptor [10][11][12]. Generally, contaminated sites are treated by combining these three approaches. For example, source removal can be achieved by flushing out the contaminants or by treating the soil chemically, thermally, or biologically; pathway management involves containment technologies that can encapsulate the contaminants and prevent their further spread; modifying the exposure of the receptor can be achieved by choosing a use for the land in the future where exposure will be reduced.

Vertical barriers have been employed for decades to control groundwater flow and subsurface contaminant transport [13][14]. There are two types of in-ground barriers, namely active and passive. Active barriers are alternately called permeable reactive barriers (PRBs). They are subsurface structures made up of reactive and hydraulically permeable materials. As the contaminated groundwater flows through the barrier, it immobilizes the contaminants through degradation, sorption, or precipitation [15][16]. In contrast, passive in-ground barriers are impermeable (cut-off) walls, which are mostly constructed using geotechnical engineering techniques and employed to redirect the groundwater flow and to isolate the contaminated site [17][18][19]. There are many types of impermeable barriers, and they can generally be categorized based on the construction methods and materials. In the USA, soil–bentonite barriers are widely used for the containment of polluted sites [20]. In the UK, the most common type of vertical barriers are cement–bentonite slurry trench walls, and the past three decades have witnessed the increasing application of soil mix technology to barrier construction [21]. In China, barriers constructed with soil mix technology have also been adopted in many land contamination risk management projects.

2. Mix Design and Engineering Properties

2.1. Soil–Bentonite Slurry Trench Barriers

Soil–bentonite slurry trench barriers have been extensively used in the USA since the 1970s. The first field trial of a 15 m-deep soil–bentonite slurry trench barrier as a diaphragm wall was conducted by the U.S. Army Corps of Engineers at Terminal Island in California [22]. The first slurry trench barrier for the containment of possible contamination was constructed by the Bachy Company in 1966 in France. It is very interesting to note that this slurry trench barrier was installed as a precaution in case there were spills from a refinery rather than in response to existing contamination, and in today's terminology this would be described as a preventative measure for land contamination risk management in an active industrial site.

The typical bentonite slurry comprises 4 to 7% dry sodium bentonite and the remaining 93 to 97% water, the density reported is between 1.03 and 1.12 g/cm^3 [23]. It should be noted that the solid content in the fresh bentonite slurry depends on the bentonite quality. For example, for bentonite with a higher liquid limit (or swell index), the target bentonite content could be relatively low. The fresh properties of the bentonite slurry can be examined using the indicator parameters of Marshall viscosity, density, and filtrate loss. Both sodium and calcium bentonite are hydrophilic and absorbent, although, generally, sodium bentonite expands more than calcium bentonite after the absorption of water. The hydraulic conductivity of a soil–bentonite slurry barrier is dependent on both soil gradation and the quantity of bentonite used in blending. Typical permeabilities of soil–bentonite barriers range from over 10^{-7} m/s in backfill composed primarily of coarse soils, to less than 10^{-10} m/s in backfill containing over 60% clay. In practice, hydraulic conductivity higher than 10^{-9} m/s is not recommended for soil–bentonite barrier applications. Recently, backfills consisting of polymer- or biopolymer-amended bentonite and soil have received global attention, as conventional bentonite has poor chemical compatibility, yielding hydraulic conductivity higher than 10^{-9} m/s when exposed to cation-laden or heavy metal-laden groundwater [24]. Because of its low hydraulic conductivity, the barrier can be used to severely restrict downgradient groundwater movement. This causes the water level on the upgradient side of the barrier to rise significantly compared to the downgradient side. Therefore, the soil–bentonite barrier should be designed to withstand the great hydraulic gradients.

The strength of soil–bentonite barriers is not usually of primary concern for contamination containment applications. These barriers are normally designed to be comparable in strength to the surrounding ground [25]. Evans and Ryan (2005) reported the undrained shear strength of the order of 5 to 20 kPa by conducting laboratory and in situ tests [26]. They confirmed that the shear strength continues to increase with time because of secondary consolidation or creep and the thixotropic nature of the bentonite.

A primary requirement for backfill material is that it contains suitable particle size distribution, preferably with between 20 and 40% of fine particles able to pass through a number 200 sieve. Some researchers advocate the use of well-graded soil requiring lesser bentonite content to achieve the targeted low hydraulic conductivity [17]. The well-graded soil contains fewer pores because the voids are filled with progressively finer material, resulting in low hydraulic conductivity. Thereby, it requires lesser bentonite content to reduce the hydraulic conductivity. Moreover, the components of a well-graded soil are relatively stable against chemical change. Hence, the well-graded backfill with less bentonite is more preferred than poorly graded backfill with high bentonite content. Such a need for good backfill material could, however, limit the application of soil–bentonite barriers.

2.2. Cement–Bentonite Slurry Trench Barriers

During construction, a trench is first excavated under a head of cement–bentonite slurry. The cement–bentonite slurry is prepared in situ by mixing cement with a pre-hydrated bentonite slurry just before its discharge into the trench [27]. The composition and mix design of the cement–bentonite slurry was frequently varied until the role of each ingredient was understood [22][28]. Unlike in the soil–bentonite slurry, bentonite minerals in cement–bentonite slurries dissolve and are basically undetectable in the hardened barrier. Moreover, carbonation and pozzolanic chemical reactions take place during the period of cement hydration and curing, resulting in the formation of secondary hydration products. It is argued by Evans et al. (2021) that the compatibility criteria traditionally applied to soil–bentonite barriers might fail to work for cement–bentonite barriers [28]. It is proposed that because of the onset of carbonation, the cement–bentonite material could deteriorate in the event of an acidic pollutant attack; in addition, the formation of pozzolanic hydration products could increase the sorption capacity of the barrier to contaminants, particularly heavy metals. In addition to the role of bentonite, the proper cement content also plays a critical part in the quality of the mixed slurry. An extremely low cement content is not able to deliver the required self-hardening nature, and a too-high cement content cannot achieve the required flowability and workability [22]. The variation of one ingredient can affect the properties such as hydraulic conductivity, strength, deformability, and chemical compatibility.

In traditional practice for cement–bentonite barriers, Portland cement (PC) has been the primary cementitious material. Recently, ground-granulated blast-furnace slag (GGBS), manufactured from a by-product of the iron-making industry, has become a more environmentally friendly cement substitute. Using one ton of GGBS reduces the embodied CO_2 by approximately 900 kg, compared to using one ton of PC [29]. Opdyke and Evans (2005) investigated the effects of the addition of GGBS as a cement replacement on the hydraulic conductivity and mechanical properties of cement–bentonite mixes [30]. Without GGBS replacement, or even with GGBS content of up to 70%, the hydraulic conductivity values are in the range from 1×10^{-7} to 1×10^{-8} m/s, which is the typical value of cement–bentonite walls traditionally formulated without slag [31]. Although the construction method of this type of barrier is similar to that of soil–bentonite, the hydraulic conductivity of cement–bentonite backfills (at 28 days curing time) is usually higher than that of soil–bentonite barriers. The strength improves with increasing GGBS replacement, with a maximum strength achieved at 80% GGBS replacement. Therefore, it was proposed that the optimum GGBS replacement range would be from 70–90% to achieve the best performance of cement–bentonite walls in terms of strength, strain at failure, and hydraulic conductivity. More recently, novel supplementary cementitious materials, particularly alkali-activated slag, are often used to replace ordinary PC in large quantities to enhance mechanical properties and durability [32][33]. For example, a recent study has shown that MgO activated slag and bentonite slurry has a higher strength and lower hydraulic conductivity because an abundant formation of expansive hydration products (hydrotalcite phases) could occupy the voids in the cementitious matrix, resulting in a denser microstructure [14]. The proportions of the different ingredients were changed by different researchers and their ratios are shown in **Table 1**. The reported cement content by weight varied from 1 to 30%, bentonite content varied from 2.8 to 7.0%, and water content varied from 65 to 85.5%.

Table 1. Summary of reported values of the cement, bentonite, water, and GGBS proportions in cement–bentonite barriers.

Reference	Cement (%)	Bentonite (%)	GGBS (%)	Water (%)	Water-to-Binder Ratio
Evans (1993) [17]	**15–30**	**4–7**	**-**	**65–80**	**2.2–5.4**
Manassero et al. (1995) [34]	**7.7**	**4**	**11.5**	**76.8**	**4.0**
Philips (2001) [35]	**3.5**	**3.5**	**13**	**80**	**4.8**
Opdyke and Evans (2005) [30]	**1–20**	**4–4.5**	**0–18**	**76–85.5**	**3.8**
Joshi et al. (2010) [18]	**2.5**	**3.4**	**10.1**	**84**	**6.6**
Carreto et al. (2016) [36]	**12.6–16.2**	**2.8–2.9**	**-**	**81.0–84.3**	**5.0–6.6**
Royal et al. (2017) [37]	**3.2**	**3.2**	**12.9**	**80.7**	**5.0**

2.3. Soil Mix Technology Constructed Barriers

Vertical barriers constructed via deep soil mixing employ a soil treatment methodology by which in situ soil is blended and mixed with cementitious and other agents to create a barrier. Its potential in geo-environmental applications became evident in the 1980s in the USA when the Bureau of Reclamation first used soil mixing to construct an upstream cut-off wall at Jackson Lake Dam in Wyoming [38]. In 1995, soil mixing was first introduced to the UK for the remediation of contaminated land [39]. Based on a cement content in soil between 100 and 300 kg/m^3 (dry weight), barriers constructed using this method typically have predesign values of unconfined compression strength (UCS) of more than 1 MPa and hydraulic conductivity of 1×10^{-8} m/s [38][40].

The grout mixtures normally consist of cement and bentonite as with a cement–bentonite slurry trench wall, although, with much lower water content. GGBS and fly ash are two main replacements that have been added to cement grout to reduce cement usage and improve durability. The addition of GGBS improves the hydrated cement product because it reduces the weak portlandite content and increases the quantity of stronger calcium silicate hydrate (CSH) [41]. Pulverized fuel ash (PFA) is a synthetic pozzolan created by the combustion of pulverized coal in power stations. The cementation effect of PFA relies on the formation of CSH which slowly hardens to form a stable material that may be similar to those of PC [42].

As discussed earlier, conventional barrier designs are primarily based upon achieving a low hydraulic conductivity to inhibit contaminated groundwater advective flow, without consideration of diffusive transport. However, low hydraulic conductivity, in itself, may not be sufficient to ensure that a barrier wall will effectively inhibit contaminant transport over a long period (e.g., decades) as the barriers typically have only a limited exchange/adsorption capacity for contaminants [43]. Some recent studies have incorporated reactive additives to deal with diffusive contaminant transport in low-hydraulic conductivity barriers while maintaining their desirable characteristic of low hydraulic conductivity. These barrier systems have been termed impermeable reactive barriers [44].

Because of their hydrophilic character, unmodified bentonites have only a limited ability to adsorb organic contaminants [45][46]. However, bentonites are receptive to be modified by exchanging inorganic cations with various types of organic cation [47]. Organoclay (OC) is a clay that has been modified to make it organophilic so that organic contaminants will sorb to it, and, therefore, be immobilized. Many organic cations may be used to modify bentonite, the most commonly used substances are quaternary ammonium cations (QACs). The QACs are surfactants consisting of an ammonium center with four branches for the attachment of functional groups [48]. Zeolites are naturally occurring or engineered aluminosilicates with an open, rigid, three-dimensional cage-like structure that contains channels and cavities. Contaminant metal ions that pass through the structure are trapped by ion-exchange reactions, in particular, zeolite is suited to adsorption of ammonia and heavy metal contaminants. Their cavity sizes enable the selective adsorption of some molecules into the porous structure while rejecting others on the basis of their size, giving zeolites their 'molecular sieve' title [49]. However, their efficiency toward organic contaminants is thought to be low due to their low organic carbon content [50]. Research has been conducted on the effect of zeolite in cement-based grouts for the deep mixing of clays, including on the mechanical performance, hydraulic conductivity, and durability for ground improvement purposes [21]. Zeolites have been blended with cement as they have been shown to offer strength as well as durability advantages over cement alone.

The in situ hydraulic conductivity is dependent on the homogeneity and quality of the mixed soil achieved by the contractor's equipment and mixing methodology. The quality and homogeneity of soil mixing are governed by many factors, including the homogeneity, plasticity, and density of the in situ soil, as well as groundwater conditions, etc. [51]. The great variability of natural soils and the actual performance of the soil mixing equipment make it difficult to predict the final in situ hydraulic conductivity. The actual hydraulic conductivity may exhibit variation spatially, both horizontally and vertically, which tends to decrease the certainty in estimating the overall hydraulic conductivity of the cut-off wall. The UCS test is the most common test for assessing the strength of cement-mixed soil. The UCS of mixed soil depends on the type of the binder used, the geotechnical and chemical properties of the in situ soil [52]. Bruce and Bruce (2003) summarized the typical ranges of some key properties of cement-mixed soil [53]. The hydraulic conductivity ranges from 1×10^{-6} to 1×10^{-9} m/s, and the 28-day UCS from 0.5 to 5 MPa for granular soils and 0.2 to 2 MPa for cohesive soils.

3. Future Perspectives

A wide range of future work needs to be conducted in order to advance the understanding and application of in-ground barriers and increase the confidence of geo-environmental engineers to adopt these approaches. Proposed future work needs to focus on two main areas: the use of geophysical methods for non-destructive monitoring and the optimization of resilient in-ground barrier materials.

Although in situ geophysical methods (e.g., electrical resistivity, electromagnetic, acoustic) provide the promise of cost-effective and non-destructive post-construction evaluation of flaws in in-ground barriers, only very limited successful case studies are available in the literature [54]. Improvements in several areas may help facilitate wider and more successful applications of geophysical methods to the monitoring of in-ground barrier systems, such as the (1) incorporation of geophysical methods into the design of monitoring plans at an early stage; (2) development of more convenient and advanced methods for data acquisition, mining, processing, and interpretation; (3) introduction of artificial intelligence algorithms into geophysics data analysis and development of data-driven solutions to improve the reliability; (4) development of novel instrumentation. For example, the interpretation of geophysical data of in-ground barriers may be more reliable and efficient if a background dataset of the in situ soil is available before the barrier construction [55]. Any changes in subsurface geophysical properties caused by contaminant transport or breakthrough could be detected through a comparison of monitoring data and background data.

The self-healing performance of SAPs, oil sorbents, MgO, and microcapsules has been investigated separately, and no combined systems of any these healing agents have been studied. Each healing agent has its own advantages and disadvantages. For example, SAPs are only responsive to water solutions, whereas oil sorbents are triggered by organic liquids. In a polluted site with both contaminated groundwater and organic contaminants, the self-healing performance of a barrier would be more effective with the combined system of SAPs and oil sorbents. Additionally, the mineral additives such as MgO pellets and microencapsulated sodium silicate can be used to complement the polymeric healing agents. SAPs and oil sorbents can swell and block cracks very quickly (usually within a few hours) when triggered by the ingress of contaminants; however, these swollen polymers are not compatible with the barrier materials and are unable to provide strength. In contrast, it takes weeks or even months for the mineral healing agents to yield enough healing products to heal cracks. These mineral healing products are compatible with the cementitious matrix, and, thus, can potentially provide strength recovery. Other novel materials such as engineered cementitious composites (ECC) and modified ECC have also been found to possess excellent self-healing performance of tensile-induced cracks upon hydration. The hydraulic conductivity values in water and target contaminant solutions of ECC and modified ECC have shown the good chemical compatibility of these materials [56][57]. The combination of these polymeric and mineral healing agents is expected to block cracks within several minutes and help the cementitious matrix regain some strength after several weeks or months.

The lack of standardized test methods for self-healing geo-environmental materials may hinder international collaboration and slow further development. Additionally, it can impede future commercialization as it is difficult to convince engineers, who are used to a strictly regulated construction methodology. Recently, six different inter-laboratory testing programs to evaluate test methods to assess the efficiency of self-healing concrete have been established within the framework of the EU COST Action SARCOS [58]. However, many of those established standardized test methods for concrete cannot be applied to barrier materials. Specialized standard geo-environmental laboratory test methods need to be established for advanced barrier materials. For example, a long-term triaxial cell hydraulic conductivity test can be used to monitor the recovery of the hydraulic conductivity of barrier materials in the long term, and a triaxial shear test can be used to measure the mechanical properties considering the effects of earth pressure, consolidation, and drained/undrained conditions. Finally, the laboratory-scale model barriers and large-scale field trials are needed to establish the efficacy of different additives and verify the proposed crack-resistant and self-healing approaches.

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