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Utilization of shale-clay mixtures as a landfill liner material to retain heavy metals

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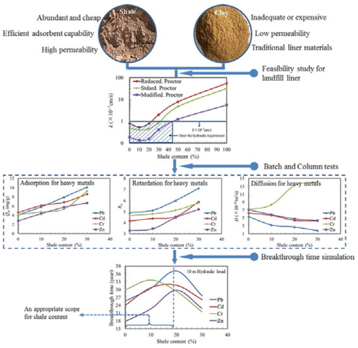
HIGHLIGHTS

- Incorporation of shale in clay liner not only improves engineering properties but also reduces the cost.
- Addition of shale in clay liner also helps to improve the adsorption and retardation for heavy metals.
- A liner with a proper mixture of shale and clay can effectively increase the breakthrough time of heavy metals.

GRAPHICAL ABSTRACT



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ABSTRACT

This study aims to research the feasibility of using shale and clay mixtures as an improved landfill liner material. Relevant laboratory tests were performed to examine basic geotechnical properties, compaction, hydraulic conductivity, batch adsorption, and transport parameters. The test results showed that the dry weight percentage of shale in shale-clay mixtures should be maintained below 32%, 48% and 23% under the standard, modified, and reduced Proctor compaction tests, respectively, to meet the minimum requirement of landfill liners for hydraulic conductivity (i.e., $\leq 1 \times 10^{-7}$ cm/s). The batch tests showed that the addition of shale improved the adsorption capacity of shale-clay mixtures for Zn, Cd, Pb, and Cr. The column tests revealed that as the shale content increased, the retardation factors of Zn, Cd, Pb, and Cr increased, while the diffusion coefficients of Zn, Cd, and Pb decreased. Based on the laboratory test results, a numerical model was constructed to simulate a typically sized landfill liner consisting of shale-clay mixtures. The modelling results showed that the proper addition of shale helped to increase the breakthrough time of Pb, Cd, and Zn.

1. Introduction

Landfills are the most popular and important method for disposing solid waste due to their simplicity, low exploitation and low capital costs [1]. The wastes disposed in landfills include household waste, commercial waste, industrial waste, and treated or untreated sludge, which usually contain many different types of heavy metals with concentrations ranging from micrograms per litre to low milligrams per litre [2]. For many landfill leachates, heavy metals are often found that are toxic to the environment and to human health [3,4]. To minimize soil and groundwater pollution from landfills, various types of hydraulic barriers are used in modern landfills to isolate the waste from the

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ambient environment [6,7]. Although the landfill technology and relevant regulations have experienced significant developments over the past decades, liner systems are still the most important structure for a modern engineered landfill [5,6].

Compacted clay liners (CCLs) are generally used in landfill construction because they meet the requirements of high contaminant attenuation and cost effectiveness. Low hydraulic conductivity ($\leq 10^{-7}$ cm/s) is required for clay or alternative liner materials [7]. Moreover, a high attenuation capacity for contaminants is required to reduce the pollution risk of landfill leachate [8,9]. However, the fractures in liner would result in the decrease of engineering and increase of permeability properties, which were caused by desiccation or unequal settlement [10–12]. To reduce usage of natural clay and improve the properties of liners, other natural or artificial materials have been considered in liner construction in the last several decades. Sand-bentonite mixtures are a common technology for impervious liner construction [13]. Chapuis and Sivapullaiah presented a method to evaluate the hydraulic conductivity of sand-bentonite mixtures [14,15]. Kaoser et al. performed a study of the adsorption of Cd, Pb and Cu on a sand-bentonite liner, indicating that Pb was the least mobile among the metal cations tested, whereas Cd was the most mobile [16]. In previous studies, it was found that the utilization of fly ashes in liner construction helped to improve the adsorption of heavy metals [17]. However, fly ash itself may carry heavy metals, posing a potential pollution risk to the environment; therefore, the leachability of heavy metals must be evaluated or controlled when using fly ash as the geotechnical material [18,19]. A comparison based on cost and environmental impact reveals that the natural local soil or mineral materials are preferable to fly ash for application as a landfill liner. Tuncan et al. found that natural zeolite and bentonite mixtures with a proper ratio were a desirable landfill liner material that possessed low hydraulic conductivity and high cation exchange capacity [20]. Furthermore, the use of a zeolite and bentonite mixture remarkably reduced the liner thickness compared with clay. Guney et al. evaluated the feasibility of mixing sepiolite with zeolite or clay for use as a landfill liner [21]. They found that the addition of sepiolite increased the strength, swelling potential and metal adsorption capacities, while it decreased the hydraulic conductivity.

A review of the above studies suggests that different materials may be utilized to substitute for clays or to reduce the clay usage in landfill liners. Since the 1980s, China has experienced a rapid economic growth and has faced mounting environmental pressures. As an important tool to address environmental issues, landfill technology has advanced significantly, and the construction of landfills has occurred at an unprecedented pace [22]. The massive amount of landfill construction boosts the increasing consumption of clays. Furthermore, China generally has more mountainous areas than plain areas. Due to these factors, natural clays that are suitable for use as landfill liners may no longer be locally available or cost effective. Therefore, it is critical to find alternative materials that possess proper engineering properties and that are economical for landfill liner construction. Shale, a sedimentary rock formed from silt and clay under a specific temperature and pressure, is an abundant mineral resource in China. The Chinese government has proposed the replacement of clay with shale to produce fire brick, cement and other building materials because the mineral composition of shale is similar to that of clay. Published studies have shown that shale was generally a good sorbent for organic contaminants [23,24]. Pimentel et al. reported that oil shale (in which the oil volume comprises approximately 7% of the total volume) could be used as an efficient adsorbent for heavy metals; the dosage of heavy metal removal by oil shale was approximately 50 to 70% for Cr, Fe, Co, Ni, Cu, and Zn and was >90% for Hg and Pb [25]. Bartelt-Hunt et al. concluded that the mass of benzene transported per unit area was significantly reduced when shale was incorporated into the liner [26]. Mohamedzein et al. confirmed that shale used as clay liner had satisfactory geotechnical engineering properties required through a series of geotechnical tests [27].

A literature survey showed that most published studies focused on using shale as the sorbent for organic contaminants or focused on the geotechnical engineering properties of shale. However, limited research was conducted on the utilization of shale or shale-clay mixtures as a landfill liner and the associated adsorption behaviour for heavy metals from landfill leachate. In this study, pertinent geotechnical and chemical tests were performed to analyse the characteristics of the shale-clay mixtures used in landfill liners. Compaction and hydraulic conductivity tests were performed for different mixing ratios of shale and clay. The adsorption capacities of heavy metals were measured by batch tests. Column tests were conducted to obtain the diffusion coefficients and retardation factors with different heavy metal solutions. Using the parameters measured from the laboratory tests, a numerical model was constructed to estimate the breakthrough time of a typical landfill liner composed of shale-clay mixtures.

2. Materials

The materials used in this study include shale and clay collected in different areas of China. The shale is a sedimentary rock derived locally in the Zhejiang Province where mountains are a major geologic feature and clay shortage has presented a problem for the construction of landfill liners. The clay, obtained in a local area in Nanjing City, is commonly used as a liner material in this area. As indicated by Benson and Daniel [28], the hydraulic conductivity of the liners increase when large particles are present in compacted clay liners. Accordingly, the clay and shale were air-dried and crushed by passing through a 2.0-mm sieve.

The physical properties of the materials were determined according to ASTM methods, including the Liquid Limit (ASTM 4318) [29], Plastic Limit (ASTM 4318), specific gravity (ASTM D854) [30], and grain size distribution (ASTM D421, 422) [31,32].

The cation exchange capacity (CEC) was measured by the ammonium replacement method. An X-ray fluorescence spectrometer was employed to identify the mineral composition of the specimens. Scanning electron microscope (SEM) was performed on micro-structure analysis of materials.

The landfill leachates were simulated using the dissolved heavy metals, including Cd(II), Pb(II), Cr(III) and Zn(II), which are common in landfill leachates [3,4]. Salt solutions of $CdCl_2$, $PbCl_2$, $CrCl_3 \cdot 6H_2O$, and $ZnCl_2$ were used to determine the adsorption capability, diffusion coefficient and retardation factor of shale, clay and their mixtures. All the compounds were commercial products and were of analytical grade.

3. Methods

3.1. Compaction tests

Compaction tests were performed on specimens containing pure shale, pure clay and four shale-clay mixtures. The mixtures of shale and clay were prepared based on dry weights, including 10S90C (10% shale and 90% clay), 20S80C (20% shale and 80% clay), 30S70C (30% shale and 70% clay), and 50S50C (50% shale and 50% clay). Three compaction techniques, including the standard Proctor (ASTM D698) [33], the modified Proctor (ASTM D1557) [34], and the reduced Proctor, were used in this study. The reduced Proctor compaction tests use the same size hammer as that used in the standard Proctor tests but apply fewer blows per layer, such as 15 blows instead of 25 blows. In contrast, the modified Proctor compaction tests employ a heavier hammer than that used in the reduced and standard Proctor tests. The relationship between the maximum dry unit, $(\gamma_d)_{max}$, and optimum moisture content, w_{opt}, could be obtained using different compaction methods.

3.2. Hydraulic conductivity tests

To achieve minimum hydraulic conductivity, the above-mentioned specimens were prepared by adding deionized (DI) water so that the

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