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Feasibility study on the application of coal gangue as landfill liner material

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ABSTRACT

Coal gangue is one of the largest industrial solid waste all over the world, and many methods have been proposed for the recycling of coal gangue. In the present study, the feasibility of using coal gangue as landfill liner material is studied through a series of laboratory tests in terms of hydraulic conductivity, sorption characteristics and leaching behavior. The results indicated that the hydraulic conductivity of coal gangue could be smaller than the regulatory requirement 1×10^{-7} cm/s with a void ratio less than 0.60. The batch sorption experiments performed on Pb^{2+} and Zn^{2+} illustrated that the coal gangue showed remarkable sorption capacity for the two heavy metals, and the sorption capacity for Pb^{2+} was larger than that for Zn^{2+}. Both the pseudo first-order and pseudo second-order models fitted well with the sorption kinetics data of the Pb^{2+} and Zn^{2+} on the coal gangue, and the Langmuir model was found to best-fit the sorption isotherms. The sorption capacity decreased in presence of multiple heavy metals, both for Pb^{2+} and Zn^{2+}. Concentrations of heavy metals leached from the coal gangue were all below the regulatory limits from China MEP and U.S. EPA. These desirable characteristics indicated that the coal gangue has potential to be used as landfill liner materials.

1. Introduction

The exploitation and utilization of mineral resources produce large amounts of industrial solid wastes, including tailings, coal gangue, fly ash, slag, waste glass, red mud and calcium carbide residue (Polley et al., 1998; Cokca and Yilmaz, 2004; Fourie et al., 2007; Zha et al., 2008; Herrmann et al., 2009; Mohan and Gandhimathi, 2009; Zhang et al., 2011; Du et al., 2016a; Wang et al., 2016). According to statistics, in 2011, the production of industrial solid wastes is 47.5 million tons in UK, 385 million tons in Japan, 15 million tons in Australia, and the utilization rate of these wastes is about 50–60%. The production of industrial solid wastes in China has been increased by 230% from 1.0 billion tons/year in 2003 to 3.3 billion tons/year in 2013, while the utilization rate of these wastes only changed from 54.8% to 62.2% (China Statistical Yearbook on Environment, 2014). The inappropriate storage and disposal of these industrial solid wastes may cause severe environmental contamination to surrounding soil and groundwater, and pose serious threats to human health and ecosystem. Therefore, recycling of these wastes has been a challenging task facing geotechnical and environmental engineers. Among these industrial solid wastes, coal gangue is one of the largest and most harmful wastes generated from the coal production process. The total accumulative stockpiles of coal gangue in China have already reached 4.5 billion tons, and the disposal of such a large amount of coal gangue occupies a lot of land resources and causes many ecological and environmental problems such as lifting the groundwater level, causing soil salinization, contaminating surrounding soil and groundwater (Li et al., 2010; China Statistical Yearbook on Environment, 2014; Li et al., 2015). Currently, the coal gangue is mainly recycled and utilized for power generation, agricultural fertilizer, highway roadbeds, brick production, cement production and concrete production (Li et al., 2006; Zhang et al., 2011; Zhou et al., 2014; Wang et al., 2016). During the past decades, although the utilization rate of coal gangue has been increased from about 41% in 1998 to about 64% in 2013, the large accumulated amount and the high increasing rate of coal gangue still require alternative utilization method. The use of coal gangue as landfill liner or clay replacement materials in landfill liner can be a potential utilization method to conserve land resources and reduce environmental contamination.

About 180–200 million tons of municipal solid waste (MSW) was produced every year in China, and landfill has been one of the most important ways for the management of the large amount

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of MSW. During the operation of landfills, leachate containing different heavy metals such as Pb^{2+}, Zn^{2+}, Cu^{2+}, Cd^{2+} is gradually generated within the landfill and may move out of the landfill by advection and diffusion processes (Kaya and Durukan, 2004; Rowe, 2005; Du and Hayashi, 2006; Kasassi et al., 2008). Therefore, the anti-seepage and contaminant sorption capability of liners is crucial for the design of a landfill. To confine waste in the landfill and prevent the leachate from leaking out of the landfill, the technical code for geotechnical engineering of municipal solid waste sanitary landfill (CJJ 176 – 2012, China) established the requirement of a compacted clay liner of at least 2 m, with a hydraulic conductivity of less than 1×10^{-7} cm/s. Because of the high sorption capacity, long-term structural stability, and low permeability, clay minerals have been used as an important ingredient in landfill liners. Previous studies investigated the performance of compacted clay liner (CCL) using different clay or clay modified materials including bentonite, zeolite, bentonite modified sand, bentonite modified loess, bentonite modified zeolite, bentonite modified fly ash, lime modified clay, red-mud modified clay, marine clay, and composite liner such as a geomembrane (GMB) over a compacted clay liner (CCL) or a GMB over a geosynthetic clay liner (GCL) (Mitchell et al., 1965; Fernandez and Quigley, 1985; Foreman and Daniel, 1986; Quigley et al., 1988; Gleason et al., 1997; Tsai and Vesilind, 1998; Abichou et al., 2000; Tuncan et al., 2003; Kaya and Durukan, 2004; Rowe, 2005; Bozbey and Guler, 2006; Du and Hayashi, 2006; Kalkan, 2006; Roberts and Shimaoka, 2008; Chalermyanont et al., 2009; Cuevas et al., 2012; Rowe, 2012; Liu and Hu, 2012). However, since clay minerals are natural unrenewable resources and the liner for a landfill always consumes large amount of clay minerals to meet the requirements of anti-seepage and contaminant blocking capability, many studies have been conducted to analyze the characteristics of possible alternative materials such as compacted compost, natural clay-shredded tire mixtures, bottom ash, fly ash, red mud and sewage (Benson and Othman, 1993; Nhan et al., 1996; Al-Tabbaa and Aravinthan, 1998; Hettiaratchi et al., 1999; Kumar and Stewart, 2003; Cokca and Yilmaz, 2004; Herrmann et al., 2009; Yang et al., 2012, 2013; Rubinos et al., 2015).

The main objective of this study was to investigate the feasibility of using a coal gangue as landfill liner material in terms of hydraulic conductivity, sorption capacity towards heavy metals and leaching characteristics. A flexible wall permeameter instrument was used to measure the hydraulic conductivity of the coal gangue under different consolidation pressures. A series of batch tests were performed to investigate the sorption mechanism of two heavy metals Pb^{2+} and Zn^{2+} on the coal gangue in terms of sorption kinetics, equilibrium isothermal sorption and thermodynamics, and the relevant factors affecting the sorption process were analyzed and discussed, including contact time (t), soil/solution ratio (s/s), initial solution concentration (C_0) and temperature (T). The competitive sorption behavior between the two heavy metals was also studied by comparing the equilibrium isothermal sorption for single and multiple heavy metal solutions. Finally, a toxicity characteristic leaching procedure (TCLP) experiment was conducted on the coal gangue to examine the safety for the application in a landfill liner.

2. Materials and methods

2.1. Characterization of the coal gangue

The coal gangue used in this study was obtained from a coal field in Yulin, Shanxi Province, China. Before the permeability and batch sorption tests, the coal gangue was oven-dried at 105 °C, cooled to room temperature, and ground to pass a mesh with opening size of 1 mm (Fig. 1a). Table 1 summarizes the geotechnical properties and chemical composition of the coal gangue, and Fig. 1b shows its grain size distribution. The specific surface area was measured by nitrogen adsorption method using the Brunauer-Emmett-Teller method with an automatic surface area analyzer (Quadrasorp SI, Quantachrome, USA), and the result was 8.01 m^2/g. The pH of the coal gangue was 6.14 according to ASTM standard D4972-13. The carbon in the coal gangue was considered as a main constituent for contaminants sorption, and for the tested Yulin coal gangue, the carbon content was 3.49%. Other chemical composition was analyzed with an XRF-1800 X-ray fluorescence (XRF, Shimadzu Corporation, Kyoto, Japan), and the result indicated a composition of 60.4% SiO_2, 24.7% Al_2O_3, 5.8% Fe_2O_3, 4.3% K_2O, 1.4% MgO, 1.4% TiO_2, 0.9% Na_2O, and 0.8% CaO. Fig. 2 displays the XRD pattern measured by a Bruker D8 Advance X-ray diffractometer (XRD) (Bruker, Germany). The main constituents in the coal gangue include quartz, albite, and a small amount of clay minerals such as montmorillonite and kaolinite.

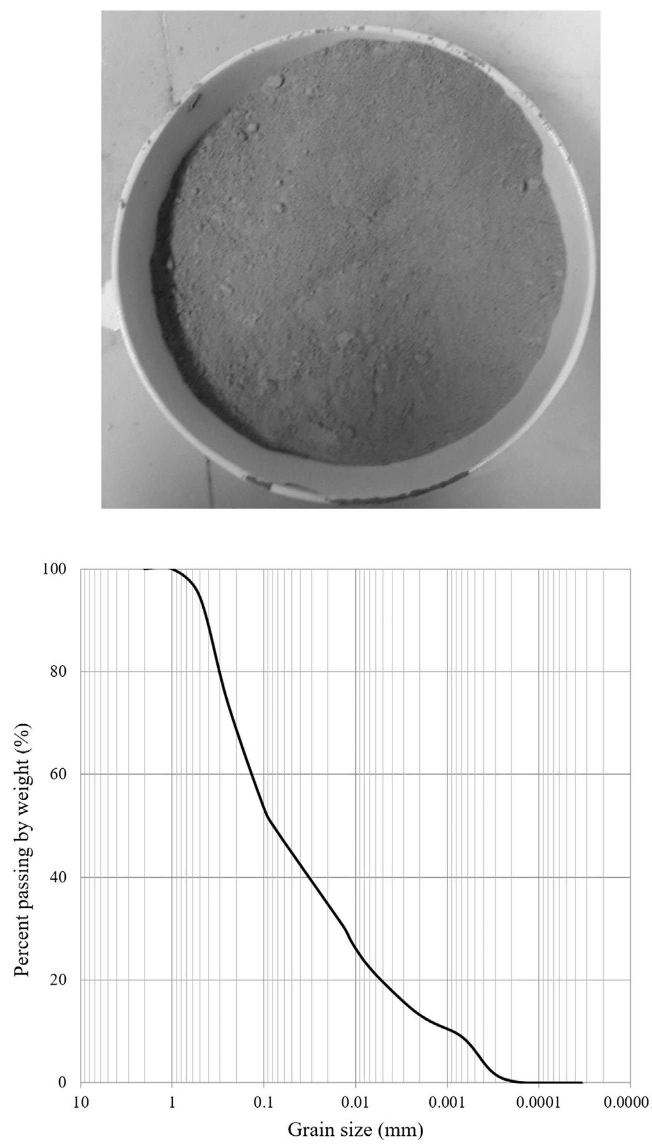

Fig. 1. Coal gangue and the grain size distribution.