

THE POTENTIAL OF MARINE CLAY USED FOR LANDFILL LINER: A GEOTECHNICAL STUDY

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ABSTRACT: The increase in excessive solid waste due to the rapid growth of the world's population is considered a severe environmental problem. The landfill leachate will contaminate groundwater, putting all living things at risk. Natural clay is a common liner material used to keep landfill leachate from polluting the environment. This study investigates the geotechnical properties of natural marine clay and its potential to be used as a landfill liner. To investigate the soil properties, the Sungai Besar marine clay (SBMC) was subjected to physico-chemical, morphology and mineralogy properties including particle size distribution, specific gravity, atterberg limits, compaction, permeability, pH, organic content, cation exchange capacity (CEC), specific surface area (SSA), X-Ray Diffraction (XRD) and scanning electron microscope (SEM). Batch Equilibrium Test was conducted to determine the effectiveness of SBMC in adsorbing heavy metals (Pb²⁺, Cu²⁺, Co²⁺, Cd²⁺, Ni²⁺, Zn²⁺). Results showed SBMC has a high percentage of fine grain size (silt 73-87% +clay 12-19 %), lower value of specific gravity (2.14-2.29), high liquid limit (79.50-84.00%), plasticity limit (49.18-59.35 %), plasticity index (20.15-34.22 %) and was categorized at very high plasticity in plasticity indexed chart. The SBMC also has maximum dry density value (1.36-1.37 g/cm³), optimum moisture content, w_{opt} (34.55-37.97 %) and average hydraulic conductivity (6.35 x 10⁻⁷ - 6.88 x 10⁻⁷ m/s). The chemical properties of the SBMC1 showed it has a high pH value (6.95-7.42), organic matter (5.31-6.06 %), CEC (91.25-92.32 meq/100g), and SSA value (60.28-62.38 m²/g). The XRD results showed that kaolinite, and illite were the most prevalent clay minerals, with quartz as the non-clay mineral. SEM analysis also revealed that kaolinite and microfossils were within the SBMC. The Batch Equilibrium test also showed that SBMC in single solution exhibited higher sorption for Cu (K_d= 0.4499 L/g, R²=0.98), followed by Pb (K_d= 0.3701 L/g, R²=0.85), Co (K_d= 0.3232 L/g, R²= 0.88), Ni (K_d= 0.1483 L/g, R²=0.98), Zn (K_d= 0.0711 L/g, R²= 0.93) and Cd (K_d= 0.0627 L/g, R²= 0.98). Based on physico-chemical, mineralogy and morphology results, SBMC is an excellent choice acting as natural clay-based energy material to be used as an engineered clay liner in a landfill area.

Keywords: Marine Clay, Heavy Metals, Geotechnical Properties, Clay-based energy material, Adsorption

1. INTRODUCTION

The efficient and long-term disposal of municipal solid waste (MSW) is one of the critical issues confronting urban populations [1]. Malaysia's population has grown at an average of 2.4% per year, or around 600,000 people yearly, since 1994. At the same time, building new landfill sites is becoming increasingly challenging due to land availability, rising land prices, and increased demand, particularly in urban areas due to population growth. As the population grows, so does the production of MSW, making MSW management critical [2]. This is because Malaysian's preferred disposal method is through landfilling, and most sites are open dumping places [3].

Landfills are waste containment systems designed to decrease the impact of solid waste on the environment and human health [4]. Compacted

clayey soils are frequently used as landfill liners to isolate hazardous and other waste items from their surroundings and to prevent heavy metals found in landfill leachates from migrating into groundwater [5]. The compaction of the liner is a crucial factor in affecting hydraulic conductivity. Low hydraulic conductivity is achieved when the soil is compacted at a high, dry density with optimal wet water content. Hydraulic conductivity for compacted liners must generally be less than or equal to 1 x 10⁻⁹ m/s [6]. According to [2], landfilling is difficult because landfill sites fill up quickly.

As stated by [7], the application of marine clay as a liner material is one of the most practical and environmentally friendly, where it requires an assessment of its physical qualities, chemical compounds, and mineral composition. Therefore, this study aimed to investigate the geotechnical properties of marine clay in Sungai Besar, Selangor, and its potential to be used as a landfill liner.

This study discussed the capability of marine clay to retard the leachate containing high heavy metals. Hence, this extensive study would help the engineer and policymakers understand the engineering characteristics and behavior of marine clay, especially for those looking for the suitability of marine clay function as an engineered clay liner for landfill application.

2. MATERIALS AND METHODS

2.1 Sample Collection and Preparation

The marine clay used in this study was obtained in Sungai Besar, Selangor, at latitude 03° 42' 45.2" and longitude 100° 55' 30.5". Marine clay was sampled from a 10 cm depth below the ground surface. Figure 1 shows the sampling location in Sungai Besar, Selangor. The obtained samples were also bagged, labeled, and transported the soil to the laboratory. The samples were air-dried before being crushed and sieved to sizes 125mm and 63 mm before testing.



Fig 1 Marine clay in Sungai Besar, Selangor (SBMC)

2.2 Experimental Procedures

The physical properties of marine clay used in the tests such as particle size distribution and falling head permeability, were determined by ASTM D-2487 and ASTM D-2434; respectively. The specific gravity, Atterberg limit, and compaction conducted according to standard BS 1377. The chemical properties of marine clay were evaluated by using pH value (BS 1377), organic matter (BS 1377), specific surface area (Geotechnical Testing Journal by Cerato & Lutenegeger, 2002) and cation exchange capacity (ASTM D-4319). The mineralogy and morphology of marine clay were also performed through X-Ray Diffraction (XRD) using D8-Advance, Bruker AXS Co. Ltd, Germany, while SEM was measured using Q150RS Quorum equipment with micrograph obtained at x5000 magnification.

2.3 Batch Test

The Batch Equilibrium test was also conducted using USEPA (1992) [8]. To perform this Batch Equilibrium test via initial concentration, the nitrate solution of lead, Pb, copper, Cu, cobalt, Co, cadmium, Cd and nickel, Ni and zinc, Zn were prepared with 10 different concentrations 20 mg/L, 40 mg/L, 60 mg/L, 80 mg/L, 100 mg/L, 150 mg/L, 200 mg/L, 250 mg/L, 300 mg/L and 400 mg/L. Nitrate salts were chosen because nitrate has a poor ability to complex with metallic cations [9]. To perform Batch Equilibrium test, 4g of soil sample (absorbent) was mixed with 40 ml of aqueous metal solutions (1:10 soil/solution ratio). The equilibrium was achieved by shaking the samples at 100 RPM for 24 hours [10]. After shaking, materials were centrifuged for 10 minutes at 1500 RPM before filtered through 45 µm nitrocellulose membranes. The solutions were analyzed using coupled plasma mass spectroscopy (ICP-MS). The concentration of heavy metals absorbed by the solution, q_e was calculated using the formula below;

$$q_e = \frac{(C_o - C_f)V}{M} \quad (1)$$

Where; C_o and C_f are initial concentration and equilibrium concentration, respectively (mg/L), V is volume of solution added (ml), M is mass of air-dried soil (g)

$$\text{Removal percentage (\%)} = \frac{(C_o - C_f)}{C_o} \times 100 \quad (2)$$

The K_d values can be determined by following equation $q_e = K_d \times C_f$ where K_d is the partition coefficient (L/kg).

3. RESULT AND DISCUSSIONS

3.1 Geotechnical Properties of Marine Clay, SBMC

The geotechnical properties of marine clay are summarized in Table 1, while Figure 2 shows the particle size distribution curve for SBMC with five replications. Results show a high percentage of clay and silt, ranging between 12-19 % and 73-87 %, respectively. A high percentage of clay content raises the effectiveness of CEC, thus promoting the retardation of heavy metals [11]. Based on USCS classification, this marine clay was classified as sandy elastic silt. The specific gravity of marine clay was also defined as the ratio of soil's mass to water's mass and ranged from 2.14-2.29. This value is acceptable within the range studied by [12].

Marine clay is highly plastic (plasticity index ranged between 20.15%-34.22%), indicating higher

absorption capability [13]. In the plasticity chart (Figure 3), SBMC was located under the A-line with the symbol of MH +OH. This shows the SBMC has 50% higher or greater percentage of silt and clay. The symbol of MH also means that SBMC originates from inorganic silts, micaceous or diatomaceous fine sandy or silty soils, elastic silts, while the symbol of OH shows SBMC originates from organic clays of medium to high plasticity, and organic silts. The compaction curves also show in Figure 4. The maximum dry density, ρ_{dmax} values ranged from 1.36-1.37 g/cm³ with the values of optimum moisture content, w_{opt} ranged between 34.55-37.97 % to achieve permeability 6.35×10^{-7} to 6.88×10^{-7} m/s. The low permeability value was due to the higher content of the finer fractions of silt and clay. Based on EPA (1990) [14], soils used as liners should contain at least 20% fines (fine silt and clay size particles) to achieve hydraulic conductivity values of or less than 1×10^{-7} m/s. Thus, the samples of SBMC were acceptable for liner material.

Table 1. Geotechnical properties of marine clay, SBMC

Geotechnical properties	Results
Particle size distribution;	
Sand (%)	1-8
Silt (%)	73-87
Clay (%)	12-19
USCS classification	Sandy elastic silt
Specific gravity	2.14-2.29
Atterberg Limit;	
Plastic limit (%)	49.18-59.35
Liquid limit (%)	79.50-84.00
Plasticity Index (%)	20.15-34.22
USCS symbol	MH
Plasticity chart (Murray et al. 1996)	Very high
Compaction;	
Max Dry Density (g/cm ³)	1.36-1.37
Moisture Content w_{opt} (%)	34.55-37.97
Permeability (m/s)	6.35×10^{-7} – 6.88×10^{-7}
pH	
Organic matter (%)	6.95-7.42
Specific surface area (m ² /g)	5.31-6.06
Cation exchange capacity (meq/100g)	60.28-62.38
	91.25-92.32
X-Ray Diffraction (XRD)	Quartz, kaolinite, illite
Scanning Electron Microscope (SEM)	Kaolinite, microfossils

The pH values for the SBMC were at pH 6.95-7.42, which is slightly acidic to slightly alkali, similar to the study by [15]. The alkalinity of marine clay is due to the alkalinity of seawater. In addition, the presence of calcium carbonate from seashells also affects the pH of marine clay. The organic matter for SBMC ranged between 5.31-6.06%. The fine-grained size and organic matter in marine clay also will lead to higher CEC and SSA ranging

between 91.25-92.32 meq/100g and 460.28-62.38%, respectively.

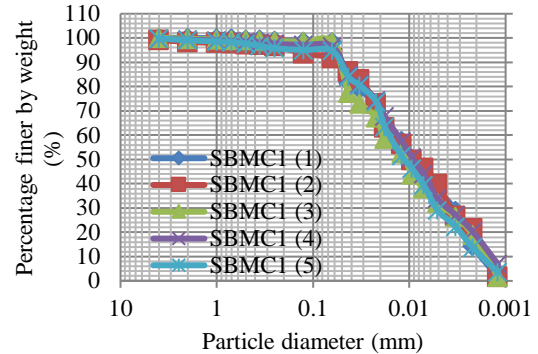


Fig.2 Particle distribution curves for SBMC sample

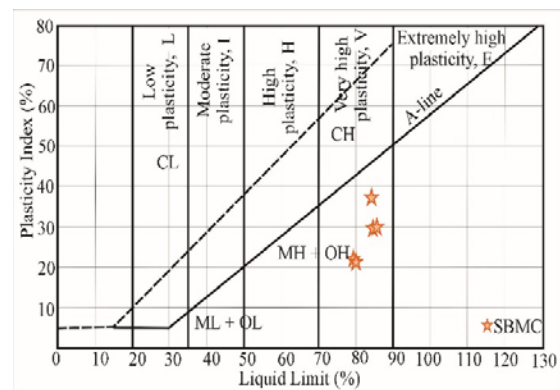


Fig.3 Plasticity chart analysis (Murray et al. 1996) for marine clay (SBMC) in Sungai Besar, Selangor

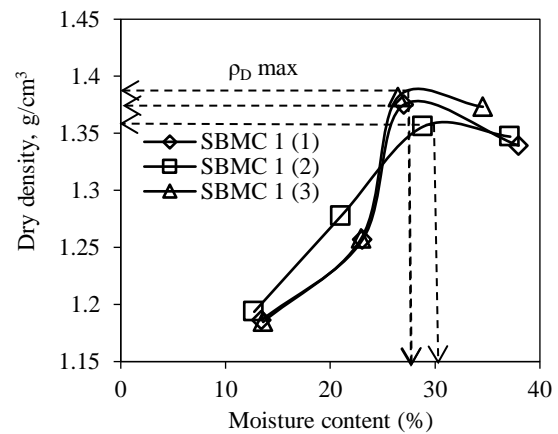


Fig.4 Compaction curves SBMC sample

According to [16], CEC affects the adsorption of heavy metals. The higher the CEC value, the higher the adsorption of heavy metals [17]. Cation exchange also occurs on the surface of negatively charged clay minerals and soil organics [5]. Increasing organic matter will increase the CEC value at neutral pH compared to acid conditions. A high value of SSA also facilitates strong physical and chemical interactions with liquids and dissolved species that were subjected to electrostatic repulsion, sorption, or specific cation

exchange reactions [18]. The XRD analysis showed the presence of quartz, kaolinite, and illite minerals. SEM analysis confirmed the presence of flaky rolled and rough-edged shapes of kaolinite in SBMC. The microfossils are also found in marine clay, contributing to high Ca in SBMC. This was due to microfossils are rich in calcium carbonate.

2.2 Batch Equilibrium Test

The adsorption of heavy metals and removal percentage by marine clay, SBMC are shown in Figure 5. Figure 5(a) shows heavy metal adsorption increased with increasing equilibrium concentration (0-500 mg/L). Table 2 also shows the K_d values of all heavy metals, and the selectivity sequence of K_d is $Cu > Pb > Co > Ni > Zn > Cd$. The SBMC appeared to be the best natural adsorbent of Pb, Cu, Co, Cd, Ni and Zn metal solutions due to having more than 90% removal percentage (Figure 5(b)). This is because marine clay relatively has a large amount of small particle size, high organic matter, and SSA and CEC values which give marine clay greater adsorption capacity.

Figure 6(a) shows the effect of pH on heavy metals' adsorption capacity while Figure 6(b) shows their removal percentage. Note that the initial pH of each element is at the ranged between pH 5 to pH 6. For all elements except Cd, the adsorption curve are the highest ($q_e=5.15$ mg/g, removal percentage are 100%) occurs over a wide pH range (pH 2 to pH12) and not just at one pH value. For Cd, the adsorption of Cd onto SBMC are low at the lowest pH (acidic condition). However, the adsorption of Cd kept increasing with increasing pH value.

According to [19], [20], the ion exchange site in the soil becomes positive at low pH levels, where heavy metal ions must compete with H^+ ions in the solution for the active site, resulting in lower adsorption. The soil surface turns negative at high pH levels, reducing competition and enhancing heavy metals' adsorption. Previous researcher [21] claimed that as the pH rises (alkaline), adsorption reduces. This was due to the competition between excess OH^- ions and negative ions from the soil surface area.

Table 2. K_d values and removal percentage for all elements in a single solution

Element	K_d (L/g)	R^2	Removal percentage (%)
Pb	0.3701	0.85	99.03
Cu	0.4499	0.98	98.19
Co	0.3232	0.88	98.27
Cd	0.0627	0.98	90.70
Ni	0.1483	0.98	95.52
Zn	0.0711	0.93	94.00

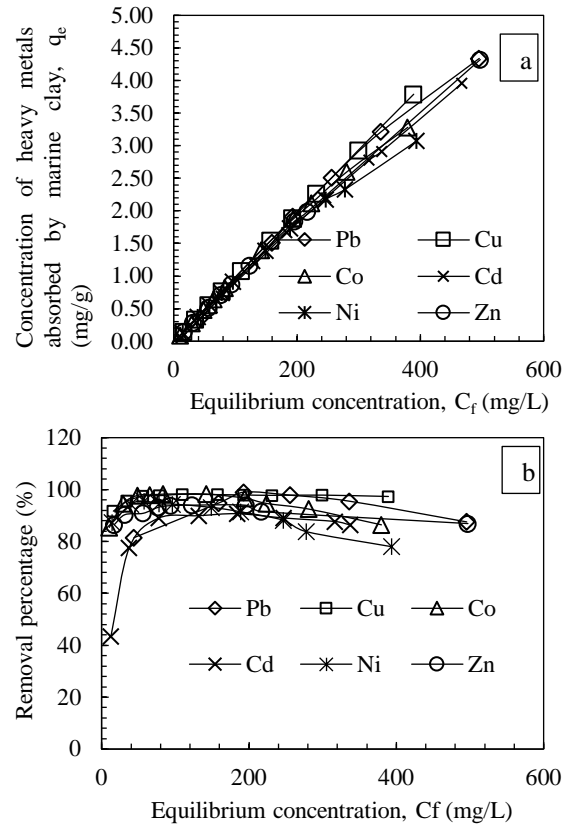


Fig.5 Batch test results, (a) adsorbed number of heavy metals in single solutions and (b) removal percentage by SBMC.

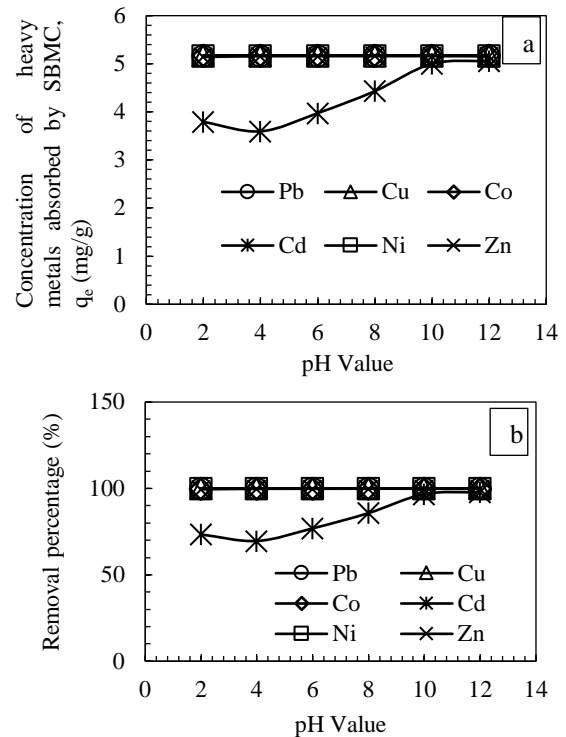


Fig.6 The effect of pH on (a) heavy metals adsorption onto SBMC and their (b) removal percentage.

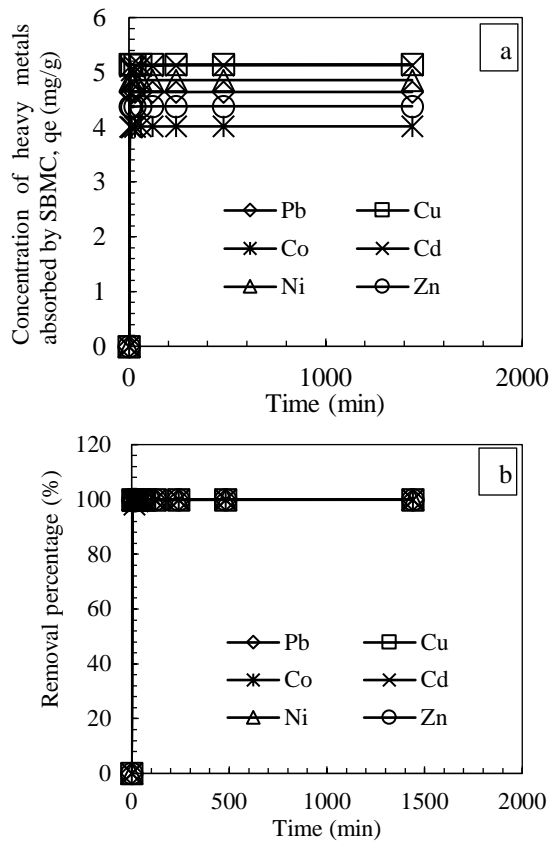


Fig.7 The effect of kinetic on (a) heavy metals adsorption onto SBMC and their (b) removal percentage.

The kinetic effect of heavy metal adsorption onto SBMC and their removal percentage is depicted in Figure 7. According to Figure 7(a), the adsorption curve is linearly proportional (adsorption at the greatest rate) up until the 5th minute, after which the adsorption progressively reached equilibrium. At this point, the removal percentage are 100% and no more adsorption occurred. According to [22], heavy metal molecules initially bind to the active site, where the adsorption rate is at its highest. And as time passes, the remaining vacant surface sites become difficult to occupy due to repulsive forces between the solute molecules on the solid phase and in the bulk liquid phase [23]. This study also considered the adsorption of heavy metals for up to 24 hours (1440 minutes). Based on previous study, marine clay in breakthrough curves will slowly decreased with increasing of influent/ initial heavy metals. According to [24], leachate contains a variety of chemical compounds that can influence its electrical conductivity and thus its interaction with soil. Leachate contamination of soil often focuses on chemical properties rather than mechanical alteration. When leachate is present, it may react differently with soil than typical water. The study by [24] also found that, leachate in the soil has a negative impact on the geotechnical qualities of

sandy-clayey soil. This due to increase of leachate in soil will decrease the values of plastic limit, liquid limit, plastic index, soil shear strength, maximum dry density and optimum moisture content. So, before any development can begin, soil-leachate contamination must be removed. It can be treated using the stabilization approach.

Figure 8 shows the pH value of the final heavy metals solution, which refers to soil buffering capacity. The initial pH value of the heavy metals solution ranged between pH 5 to pH 6. After the adsorption test, the pH value is between pH 7 and 8 (for all elements). SBMC is an alkaline (pH 6.95-7.42) soil. At high pH values (above the point of zero charge), marine clay's surface has a higher negative charge, resulting in higher cations (heavy metals) attraction. According to earlier studies by [25], the buffering capacity correlated positively with clay concentration, CaCO₃, pH and negatively with soil organic content. They also suggest that the soil buffering capacity is a sensitive indicator of soil changes after adding various amendments. Thus, no standard categorization scheme for soil buffering capacity makes classification challenging.

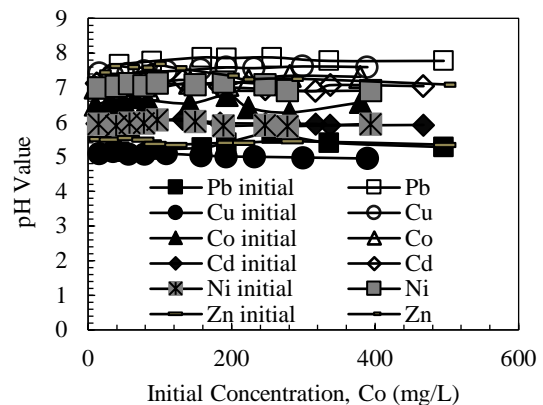


Fig.8 The SBMC soil buffering capacity in single heavy metals solution.

Figure 9 also shows the relationship between the conductivity values of heavy metals solution before and after the adsorption test. The conductivity curve shows the same trend for all elements: the conductivity increased gradually with increasing initial concentration. According to [26], this trend is called the 'second negative Wien effect.' The conductivity values also decrease with increasing ion strength of the heavy metal. From figure 9, the conductivity after adsorption tests is in the range of 2000 μ S/cm to 4500 μ S/cm, and this range is higher compared to the conductivity values before the adsorption test (0 μ S/cm to 2000 μ S/cm). This is due to the heavy metal ion concentration in the equilibrium solution decreasing (after the adsorption test); thus, the conductivity value rises. SBMC has a high conductivity value due to the ability of marine clay can adsorb more cations.

Marine clay has a high percentage of clay, rich in negative charges and can attract positive charges from pollutant/ heavy metal solutions.

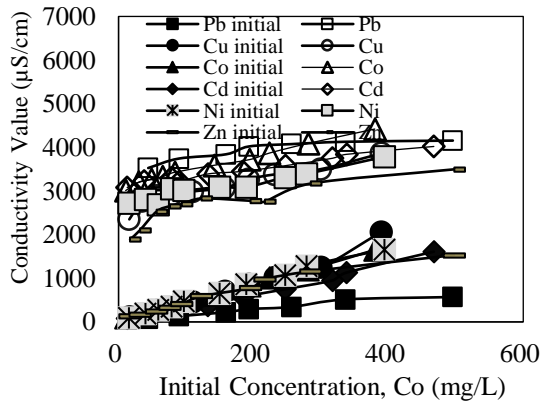


Fig.9 The conductivity value of SBMC soil before and after adsorption test

4. CONCLUSIONS

Based on geotechnical properties of marine clay, SBMC can be utilized as a landfill lining material. This was due to SBMC has achieving minimum EPA's hydraulic conductivity value (1×10^{-7} cm/s). The SBMC also showed higher adsorption capacity as it can eliminate more than 90% of the Pb, Cu, Co, Cd, Ni, and Zn metal solutions. This study also demonstrates the 100% removal percentage occurred in a wide pH range (pH 2 to pH12) except for Cd. The Cd's adsorption favours are at higher pH (alkaline condition). While the optimum condition for kinetic effect are after 5th minutes, SBMC tend to remove 100% heavy metals in aqueous solution. This study also concludes that marine clay is one of the natural clay-based materials that have a very excellent candidate to use as an engineered clay liner in a landfill area. Marine clay also proved to become a reliable barrier against leachate leakage and transport by considering the design, construction, and long-term monitoring methods.

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