Feasibility study on the application of alum sludge (AS) as alternative landfill liner material in sustainable landfill infrastructure model: XRD and SEM analysis

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Abstract. The increasing number of Alum Sludge (AS) in water treatment plant (WTP) from water treatment process has been directed to the reduction of storage capacity for keeping of AS waste itself in the landfill. Other than disposed in the landfill site, this accumulated AS can be commercialized into a useful product such as landfill barrier. Conventionally, clay is used as the protection layer in the engineered sanitary landfill to reduce the leachate intrusion into groundwater. However, due to higher usage and demand on the clay (C) material caused its depletion and higher in cost. Therefore, accumulated AS can be recycled and reused to reduce the disposal problem in landfill and financial burden facing by the landfill operator. In this study, AS was investigated and compared with the kaolin clay (KC) in terms of XRD and SEM characteristics. The index properties and hydraulic conductivity, k test were also conducted to classify the AS and KC composition. Based on the testing, AS can be classified as High Plasticity Clay and the value of k for AS is higher than KC. Based on the results from XRD, both of KC and AS samples was having a similar crystalline phase which is Quartz. Based on the summary of compounds elements, the chemical elements of Oxygen, Aluminium, Silicon and Iron presence in AS have a quite similar percentage of weight (%) with KC. From all of these results, it can be concluded that the AS is mostly having similar materials to KC and amorphous in nature.

1. Introduction

Conventionally, landfill system can be divided into two types namely: open dumping landfill and sanitary landfill [1]. The infrastructure system for sanitary landfill systems are solid waste open area, leachate collection and treatment facilities (piping system and ponding system for treating the leachate before it is being released to the receiving environment) and landfill liner system (clay soil as a final impoundment, geotextile, geomembrane and others) [2]. The landfill system itself are either single (waste and clay liner), double (waste, clay liner and supported by geomembrane or geotextile) or composite liners (waste, clay liner and supported by geomembrane and geotextile or others synthetic materials. Function of Landfill liner is to support the natural material from leachate leaking into soil and water sources (surface and ground). Higher demand on the clay usage in construction especially building and landfill are among of the reasons why researchers is searching for alternative liner
material for clay replacement. Study done by Wu [3] on the potential application of coal gangue (CG) in landfill application and found that CG has good absorption capacity to heavy metal (Pb$^{2+}$ and Zn$^{2+}$). In addition, the permeability of the CG was smaller than the regulatory requirement for a monolayer landfill liner. The potential of bauxite residue (red mud - RM) as a landfill liner material was studied by Rubinos [4] and this RM has an ability to absorb As$^V$ in leachate (higher sorption capacity for leachate with acidic condition).

Landfill liner materials shall durable and flexible enough to sustain any received loading during the installation of municipal waste throughout its service life. There are some requirements should be highlighted in the selection of suitable materials as a landfill liner material. The highlighted points are smaller hydraulic conductivity value, maximum shear strength, minimum shrinkage strength, the suitability of the materials with landfill site condition and able to sustain maximum compaction characteristics. These important points should be highlighted in order to maximize cost and come up with a suitable material without failure during its service life (to avoid any redesign and reconstruction of landfill site or leaking of leachate into environmental system due to failure during operation).

Over the year, production of waste by products from Water Treatment Plant (WTP) namely Alum sludge (AS) caused problems to WTP operator due to financial burden for AS management and disposal. AS is usually used for coagulation process in water treatment process. The presence of AS can give bad effect to the environment such as soil system and surface water source such as sorption of nutrients and organic matter, create micro-locations in soil [5]. According to Gopalakrishnan and Velkennedy [6] the characteristic of sludge being generated is depended on the quality and quantity of the chemical used for water treatment. AS is considered as one of the major adsorbents for pollutant removal and has an ability to remove a large amount of heavy metals and organic constituents due to small particulate materials which provide available surface area for inter-particulate bonding [7]. Furthermore, AS is a cost-effective adsorbent in arsenic contaminated water systems [8]. It has similar characteristic with clay, which known to have higher absorption capacity. The rate of absorption is very important as a liner for heavy metal adsorption.

Many researches have been conducted on the commercialization of AS into a useful product. Wang, [9] comes up with an innovative research on the recycling of AS (as a fine aggregate) in producing eco-friendly construction building material (controlled low strength material - CLSM). By adding AS (silt/clay rich contents) into the products, the water demand is dramatically increased, resulting in long stiffening time and low compressive strength [9]. Other researcher [10] had done study on the application of AS in producing high-strength self-compacting concrete (HSSCC), where the treated AS used in this research contains silicon dioxide and aluminium trioxide which are the main constituents of cement. In this study, AS is proposed as a replacement to clay material. This is due to the depletion of clay material in landfill (major construction material especially landfill). This study is conducted to investigate the XRD and SEM characteristics of AS as an alternative liner material and compare with the characteristics of conventional material: KC.

2. Materials and Methods
The collected materials in this study is Alum Sludge (AS) and Kaolin Clay (KC). The preparation of each of the materials and methods used for samples testing is describe in the following section. AS and KC were tested for X-Ray Diffraction (XRD) and Scanning Electron Microscopy (SEM).

2.1. Collection and Preparation of Alum Sludge
Alum Sludge (AS) used in this study was obtained from Kulim Hi-Tech Water Treatment Plant (KH-TWTP), Pinang Tunggal, Sg. Petani, Kedah, Malaysia. The sample was collected using grab sampling method and in the form of dry sample. The alum sludge sample was oven dried for 24 hours to eliminate the moisture content in the collected sample. The sample was then sieved passes through the 0.002 mm sieve pan to satisfy the requirement for particle size distribution of clay. Figure 1 shows the water treatment processes and process involve in AS production from KH-TWTP.
2.2. Collection and Preparation of Kaolin Clay
The Kaolin Clay (KC) sample used in this study is KC is white in colour and in soft powdered form. It is mostly constituted of kaolinite minerals derived from the weathering of orthoclase (potash) feldspar which is an essential mineral of granite [11]. The clay (C) sample should be collected in site with the consideration of depth more than 3-meter depth. Due to the absence of suitable clay at the existing site, KC is generally better alternatives [12].

2.3. Characterization of Alum Sludge and Kaolin Clay Samples (Index Properties)
The value of soil properties is obtained from the Index Properties Test (Atterberg Limit Test) and Compaction Testing. Table 1 shows the summary of soil properties value for Kaolin Clay (KC) and Alum Sludge (AS).

Table 1. Summary of soil properties value for KC and AS samples.

| Parameter                  | Unit          | KC     | AS    |
|----------------------------|---------------|--------|-------|
| Optimum Moisture Content   | %             | 16.46  | 43.57 |
| (OMC)                      |               |        |       |
| Maximum Dry Density (MDD)  | Mg/m³         | 1.69   | 1.2   |
| Liquid Limit               | %             | 35     | 50    |
| Plastic Limit              | %             | 15     | 26    |
| Plasticity Index           | %             | 20     | 24    |
| Characteristic             |               | Intermediate Plasticity Clay | High Plasticity Clay |

Figure 1. Water Treatment Processes and Production of Alum Sludge (AS) from Water Treatment Plant (WTP)
Based on Table 1., KC can be categorized as Intermediate Plasticity Clay (MDD is 1.69 Mg/m³ at 16.46% of its OMC). AS have 1.2 Mg/m³ of MDD while the OMC obtained is 43.57 % and can be classified as a High Plasticity Clay.

2.4. Hydraulic Conductivity of Alum Sludge and Kaolin Clay Samples

Hydraulic conductivity is one of the important roles to meets the requirement as a barrier in the landfill. Table 2 shows the hydraulic conductivity value, $k$ for Kaolin Clay (KC) and Alum Sludge (AS).

| Sample | KC     | AS     |
|--------|--------|--------|
| Hydraulic conductivity, $k$ (m/s) | $3.94 \times 10^{-7}$ | $4.79 \times 10^{-6}$ |

Based on Table 2. The value of hydraulic conductivity, $k$ for KC and AS is $3.94 \times 10^{-7}$ m/s and $4.79 \times 10^{-6}$ m/s, respectively. The $k$ value is being affected by the index properties characteristic which is OMC. OMC value is increasing resulting in the decreasing of $k$ value.

2.5. Testing of Samples using X-Ray Diffraction (XRD) and Scanning Electron Microscopy (SEM)

The Alum Sludge (AS) and Kaolin Clay (KC) samples were tested for its chemical characteristics through X-Ray Diffraction (XRD) and Scanning Electron Microscopy (SEM) analysis.

2.5.1 X-Ray Diffraction (XRD) Analysis. The function of X-Ray Diffraction (XRD) analysis is used to determine the composition of materials (ex.: Silica and Quartz) in both alum sludge (AS) and kaolin clay (KC) samples. A one (1) gram of fine powder AS and KC were placed into the XRD sample holder plate. Then, it was placed into the XRD chamber prior analysis from 2$^\theta$ of 20° to 40°. The analysis of XRD spectrum was conducted using D2 Phasers equipped with LYNXEYE ultra-fast detector. The obtained result is the form of graph showing the peaks for each of the materials in AS and KC samples.

2.5.2 Scanning Electron Microscopy (SEM) Analysis. The function of Scanning Electron Microscopy (SEM) analysis is used to determine the crystalline phases of alum sludge (AS) and kaolin clay (KC) samples. The samples were prepared in term of powder form. The samples were placed on the stage and the system was setup to analyze both samples. The obtained result is in the form of image showing the crystalline phases for each of the materials in AS and KC samples.

3. Results and Discussion

Alum Sludge (AS) samples were tested for X-Ray Diffraction (XRD) and Scanning Electron Microscopy (SEM) to determine the materials compound of AS and KC samples and to determine the crystalline phases of AS and KC samples, respectively.

3.1. X-RAY Diffraction (XRD) Analysis

The phase of material compound can be determined by conducting XRD testing on AS and KC samples. Figure 2 shows the material Phase on AS and KC.
Based on Figure 2, KC shows four peak locations which are 21°, 27°, 36.25° and 39° (Quartz low, Quartz, Carbon Oxide, Illite). XRD analysis for AS, the pattern shows that the AS belongs to quartz at peak locations, 27°. Variability in the crystallinity of the minerals in the KC and AS bodies are inferred from XRD peak intensities; with AS exhibited the least crystallinity and KC showed the highest crystallinity. The AS had kaolinite, mica, and quartz as major mineral components [13]. The AS has a chemical composition as belong to clay with the presence of silica and alumina [14].

3.2. Scanning Electron Microscopy (SEM) Analysis
The SEM test was conducted to determine the cross section crystalline phase in the samples while the EDS test was conducted to identify the compound materials in the samples. Figure 3(a), 3(b) and Figure 4(a), 4(b) show that AS and KC samples were having a different cross section crystalline phase.
From the SEM micrograph at Figure 3(a) and Figure 3(b) the particle size of AS and KC are ranging in between less than 1 µm to more than 40 µm. The AS sample is having a combination of small and big crystalline phase compared to KC which is having even more similar pattern of crystalline phase. SEM images shows kaolinite particles of varying sizes. Figure 4(a) and Figure 4(b) show SEM analysis where it is indicates that KC consists of ultra-fine and thin platy-flaky particles which is the size less than 1 µm. The SEM micrograph of the AS did not find any well-defined crystalline structure on the AS sample. The homogeneity of the AS also depends on its particle size. The AS is a non-homogenous material due to its unclear particle shape as demonstrated by SEM analysis [7]. In addition, AS is amorphous in nature [7]. The summaries of compound elements for both samples (AS and KC) were listed in Table 3 and Table 4.

Table 3. Summary of compound elements in AS sample.

| Element   | Weight (%) | Weight % (σ) | Atomic % |
|-----------|------------|--------------|----------|
| Oxygen    | 56.9       | 0.36         | 70.88    |
| Magnesium | 0.43       | 0.07         | 0.35     |
| Aluminium | 15.86      | 0.19         | 11.72    |
| Silicon   | 20.62      | 0.23         | 14.63    |
| Potassium | 1.19       | 0.08         | 0.61     |
| Titanium  | 0.51       | 0.09         | 0.21     |
| Iron      | 4.49       | 0.21         | 1.6      |

Table 4. Summary of compound elements in KC sample.

| Element   | Weight (%) | Weight % (σ) | Atomic % |
|-----------|------------|--------------|----------|
| Oxygen    | 57.21      | 0.32         | 71.94    |
| Magnesium | 0.3        | 0.07         | 0.25     |
| Aluminium | 15.65      | 0.17         | 11.67    |
| Silicon   | 17.7       | 0.19         | 12.68    |
| Phosphorus| 1.45       | 0.09         | 0.94     |
| Potassium | 0.84       | 0.07         | 0.43     |
| Iron      | 5.1        | 0.2          | 1.84     |
| Barium    | 1.76       | 0.2          | 0.26     |
Based on Table 3, there were seven compound elements found in AS sample. The elements were Oxygen, Magnesium, Aluminium, Silicon, Potassium, Titanium and Iron. Oxygen, Aluminium and Silicon were the dominant components in the AS sample. The Oxygen element had the highest compound element weight of 56.9% while Iron Magnesium element had the lowest compound element weight of 0.43%. Based on Table 4, there were eight compound elements found in KC sample. The elements were Oxygen, Magnesium, Aluminium, Silicon, Phosphorus, Potassium, Titanium, Iron and Barium. The Oxygen element had the highest compound weight of 57.21% while Magnesium had the lowest compound element weight of 0.3%. Both AS and KC samples contains higher Oxygen and lower Magnesium levels. Based on the obtained result, it is indicated that the compounds element is affecting the different of $k$ value for AS and KC.

4. Conclusion
In this study, index properties and hydraulic conductivity, $k$ testing were conducted to further classify the type of soil depending to Optimum Moisture Content (OMC) and Maximum Dry Density (MDD) as well as other criteria such as plastic limit. These basic geotechnical testing is very important for soil analysis before further testing is required to identify the relationship between different soil classification with materials compound and crystalline phase. Based on the characterization, AS can be classified as High Plasticity Clay and the value of $k$ is higher compared to KC.

Alum Sludge (AS) and Kaolin Clay (KC) were tested for X-RAY Diffraction (XRD) and Scanning Electron Microscopy (SEM) to determine the materials compound of AS and KC samples and to determine the crystalline phases of AS and KC samples, respectively. Based on the obtained result from XRD for AS and KC samples, both samples having a similar material composition which is Quartz. Based on the SEM, it was proven that the crystalline phase in AS is different with the crystalline phase in KC. KC is having even more similar pattern of crystalline phase compared to the AS sample (a combination of small and big crystalline phase). It is indicated that the different crystalline phases of AS and KC samples is affecting the hydraulic conductivity, $k$ of AS. The tendency of AS sample to have a high hydraulic conductivity, $k$ is relatively higher compared to KC due to the larger particles or cross section of the AS sample. Based on the summary of compounds elements, the chemical elements of Oxygen, Aluminium, Silicon and Iron presence in KC have a quite similar percentage of weight (%) with AS. Based on the obtained results, it can be concluded that the AS is mostly having similar materials to KC and amorphous in nature.

5. Recommendation
In order to further propose Alum Sludge (AS) as an alternative to Kaolin Clay (KC), further testing such as adsorption capacity and leachability testing is required to detect either the proposed AS is capable to be used as a replacement to clay in landfill practically.

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