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Influence of Fly Ash-Based Geopolymers on Treated-Contaminated Soil Properties

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***Abstract*—Using natural soil in landfill sites for landfill construction is common in developing countries such as Indonesia. However, the risk of contaminated soil being used as material needs to be reduced by soil improvement to meet the landfill base requirement. Geopolymer is one stabilization material with high durability towards the contaminants, in which fly ash is one of the best geopolymer raw materials due to its readily great supplies and noble properties. The present study investigates fly ash-based geopolymer (FAG) influence on the treated-contaminated soil properties specifically for landfill liner purposes. The studied soils were collected from Ngipik Landfill, an indiscriminate municipal landfill in Gresik, East Java-Indonesia. The samples were obtained from three different locations around the waste mound and stabilized with FAG at 5, 10, and 15 wt.% of unstabilized soil at its optimum water content compared to the natural soil. The laboratory-scale tests were conducted, including the unconfined compressive strength test, standard compaction proctor, consolidation test, and the permeability test to understand the treated-soil characteristics better. The Scanning Electron Microscope (SEM) analysis was gathered to examine the treated soil's micro-properties. The results denoted that the FAG addition increases the soil's compressive strength up to 36% and reduces the compressibility to up to 60%. The permeability shows a slight decrement in 5% and 10% of FAG addition. The SEM image indicates that the FAG made bonds and fills the void. This study concludes the FAG enhances the mechanical properties of Ngipik Landfill soil despite the contamination.**

***Keywords*—Treated-contaminated soil; fly ash-based geopolymer; landfill liner; standard compaction proctor; consolidation; unconfined compression strength.**

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I. INTRODUCTION

Open dumping is a hazardous yet prevalent landfilling method in Indonesia, as in some other developing countries. In Indonesia, there are still 47% of open dumping landfills in 355 cities [1]. This landfilling method is the leading cause of leachate percolation through the surrounding soil and groundwater [2], [3]. Hence, the natural soil of the open dumping landfill is at risk of contamination.

The surface and subsurface water in the Ngipik Landfill area are contaminated, as reported in the previous studies [4, 5]. About 15 years after the landfill started operating, leachate has seeped in up to 15 meters depth. Around the landfill, ammonium is the highest contamination substance on the surface water. The concentration is 1024 mg/l, while the regulation limit is 5 mg/l [4]. As the leachate seeps in, the subsurface material of the landfill is contaminated (Table 1). However, in the area of about six hectares, the thick material substitution is arduous. The necessity to get the material for landfill liner is substantial in landfill construction. Despite its methodological merit, the idea of using the natural soil of the landfill site is evaded due to the contamination risk.

One of the solutions to enhance the landfill's natural soil properties is soil stabilization. However, the stabilization material is reluctant to be contaminated. One of the stabilization materials which has the durability to the contaminated environment is a geopolymer. Geopolymer is sulfate resistance [6]–[8], acid resistance [9], [10], and even used to immobilize materials such as ammonium [11], [12], and heavy metals [13]–[15]. Geopolymer induces a reaction between the aluminosilicates and the basic activating solution called geopolymerization [16]. Geopolymerization then forms a new material, aluminosilicate, which simultaneously has noble properties and low carbon emission [17]. It is why the geopolymer has been researched for decades to be the cement substitution in concrete, the immobilization materials, and soil stabilization material.

TABLE I
THE AMMONIUM CONTENT OF THE SOILS

Sample	Constituent	Unit	Value
BH-1	Ammonium	% NH_4-N	1.06
		ppm	10600
BH-2	Ammonium	% NH_4-N	1.28
		ppm	12800
BH-3	Ammonium	% NH_4-N	0.41
		ppm	4100

However, research about the effects of the contamination area on the geopolymer and vice versa is still limited. Thus, this study is meant to analyze the influence of FAG to improve the contaminated natural soil from the landfill site by assessing three soil samples taken from three different locations with various distances from the waste mound. The three soil samples were then stabilized with 5, 10, and 15 wt.% of FAG. The permeability, compaction, and compressive strength of the natural and treated soil were assessed.

II. MATERIALS AND METHOD

A. Material Characterization

The studied soil was taken from a municipal landfill in Gresik regency, East Java-Indonesia. The landfill has been operating for disposal since 2003. From previous studies, the soil is considered as contaminated soil based on the leachate plume findings on the landfill area [5] and adjacent monitoring wells [18]. Therefore, the soil was used to assess the effect of FAG on the contaminated soil. The studied soil was gathered from three locations at various distances from the waste mound. All the soils belong to CH or clay with high plasticity soil (CH) based on the Unified Soil Classification System (USCS). The plasticity index and liquid limit (ASTM D4318-00) of the soil from the three boreholes are summarized in Table 2.

TABLE II
SOIL PROPERTIES OF STUDIED SOIL

Soil Properties	BH-1	BH-2	BH-3
Liquid limit	73.7%	74.4%	89.9%
Plasticity index	48.9%	51.9%	60.7%
Particle specific gravity	2.72	2.72	2.70
Fine fraction (sieve no.200)	88.7%	85.8%	92.6%
Void ratio	0.915	0.865	1.020
Soil density (gr/cc)	1.451	1.474	1.360
Distance to waste mound (meters)	50	15	200

The three borehole soils have nearly similar characteristics by their color, liquid limit, specific gravity, and fine fraction. However, BH-2 is closer to the waste mound and the most contaminated among the three boreholes [4]. Therefore, this current research compared the three soil samples from three boreholes with BH-1 and BH-2 as the mildly contaminated and contaminated soil, respectively, and BH-3 as the control soil sample for the uncontaminated soil.

The fly ash utilized in this research was a Class F fly ash with the low calcium content. It was gathered from Tanjung Jati coal-fired Power Plant units 3 & 4, Central Java Province, Indonesia. Its characterization was carried out by x-ray fluorescence (XRF), with the result presented in Table 3.

TABLE III
CHEMICAL COMPOSITION OF THE FLY ASH

Component	Percentage	Component	Percentage
MgO	0.35	MnO	0.13
Al2O3	15.5	Fe2O3	22.85
SiO2	43.6	CuO	0.05
K2O	2.47	ZnO	0.06
CaO	7.99	Rb2O	1.5
TiO2	1.8	MoO3	3.32
V2O5	0.08	BaO	0.3
Cr2O3	0.03	Na2O	-

B. Geopolymer Mixing Methods

8 molar sodium hydroxides synthesized the geopolymer used in this research. The ratio of sodium hydroxide to sodium silicate was 1:2.5, and the ratio of the alkali to fly ash was 30:70. First, the 8 molar sodium silicates were prepared, then left for 24 hours at ambient temperature before mixing with sodium silicate to release the heat. After mixing sodium hydroxide and silicate, the solution was then left until the temperature drop. The low temperature reduces the geopolymer's reactivity [19], [20]. The high energy will accelerate the chemical reaction so that the geopolymerization process will be faster, and the paste will be hardened in a short time. It was not expected because the mixing of geopolymer and soil needs a longer time of a low viscosity paste to optimize the mixing process. The paste was then poured into the dry soil shortly and mixed well. The FAG-treated soil was then poured into the unconfined compression mold (tube shape with diameter 3.5 and 7.0 cm height); consolidation test (tube shape with diameter of 6.5 cm and 1.6 cm height), and the rest of them were used for standard compaction proctor and the physical properties test. The optimum water content took the water content used for the remolded sample based on Standard Compaction Proctor (ASTM D698) because geopolymer does not explicitly change the treated soil's compaction properties [21].

III. RESULTS AND DISCUSSION

The BH-1, BH-2, and BH-3 treated soil had typical compaction curves regardless of the contamination rate (Fig. 1-3). The 5% addition of FAG had low dry density and decreased more in 10% of FAG. Meanwhile, 15% of FAG addition increased the dry density. The geopolymerization process caused it. The FAG material has more specific gravity than the fly ash itself. While the geopolymerization process reaches the optimum phase, the reaction increases the specific gravity of the fly ash by forming silicate crystals resulting from the reaction on the surface of fly ash particles [22], [23].

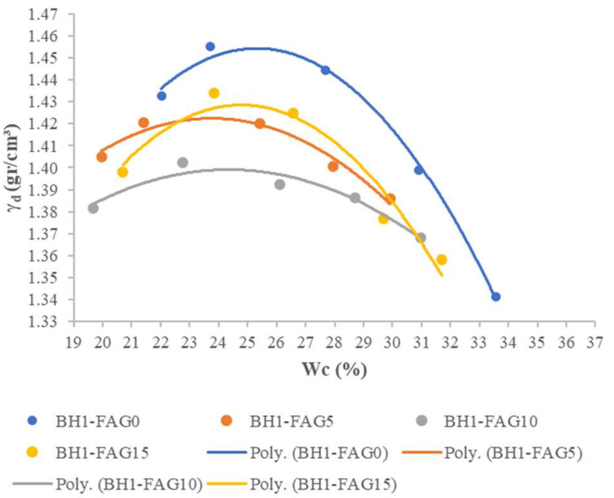

Fig. 1 Compaction Curve of BH1 soil at different FAG additions

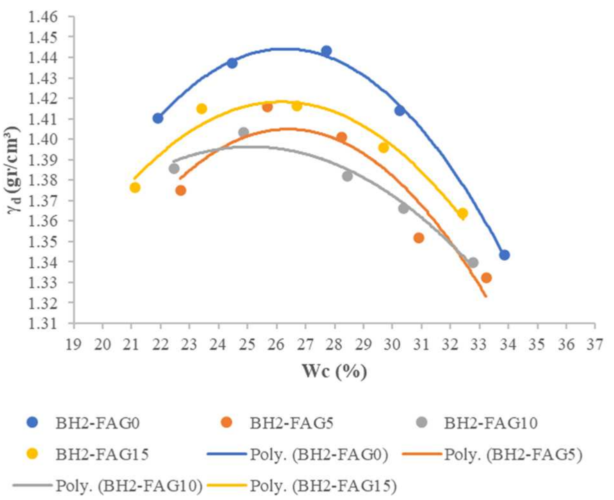

Fig. 2 Compaction Curve of BH2 soil at different FAG additions

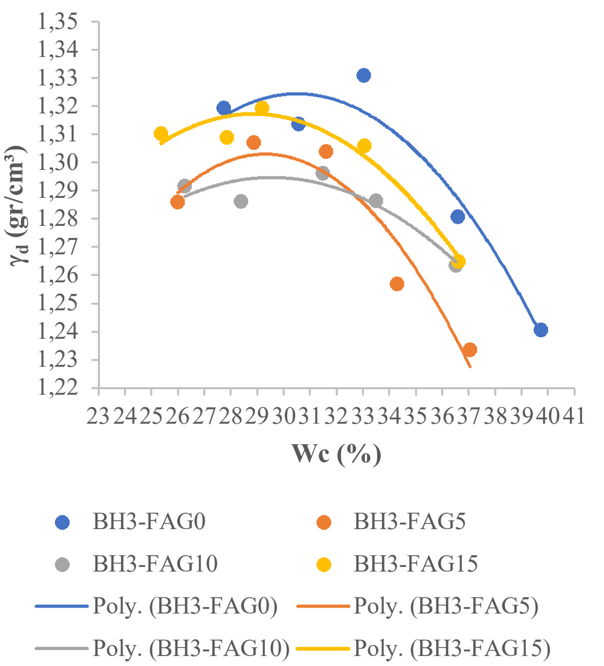

Fig. 3 Compaction Curve of BH3 soil at different FAG additions

In accordance with the compaction curve, Fig.4 shows that the dry density of FAG-treated soil was lower than the natural soil. It was because the specific gravity of the fly ash is lower than the specific gravity of clay [23]. The addition of more FAG could increase the dry density of the FAG-treated soil related to the material produced by the geopolymerization. However, the addition of more than 30% FAG was inefficient, and lots of factors should be reconsidered aside from the dry density.

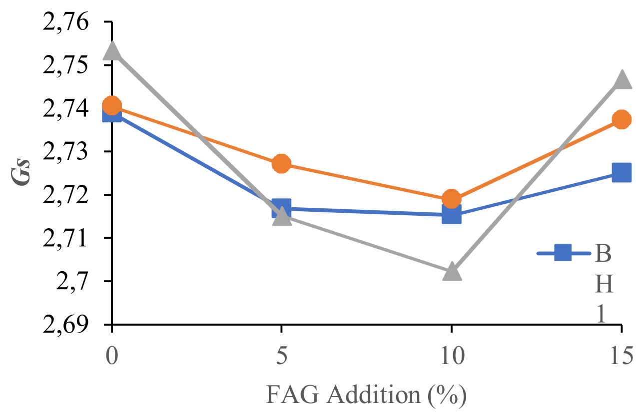

Fig. 4 The specific gravity of natural soils under study at different FAG addition

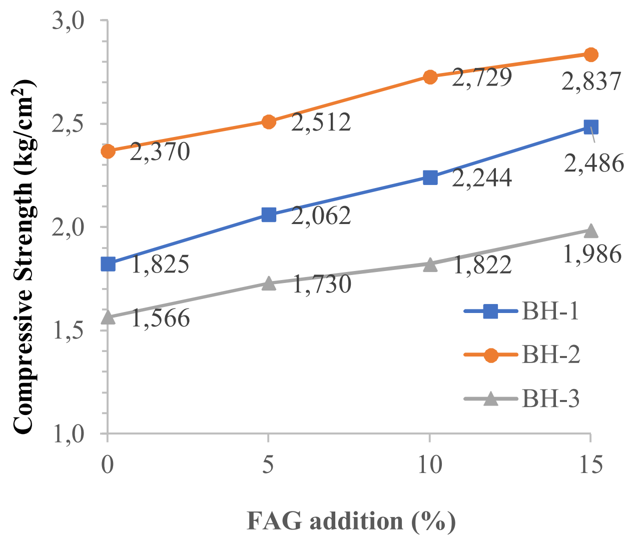

Fig. 5 The compressive strength of natural soils under study at different FAG addition

The specific gravity of the particles decreased with the FAG addition. The specific gravity, by definition, is the ratio of the mass of a given volume of solid soils to the mass of an equal volume of distilled water at the same temperature [24]. Hence, for the same volume and temperature, Fig.4 indicates that geopolymer addition decreased the mass of the solid as the fly ash substituted the clay. In 15% FAG addition, the solid mass increased due to the geopolymerization. The geopolymerization resulted in the increasing mass of the solid soils.

The compressive strength of the soils increased as the FAG was added (Fig.5). The strength of the FAG depends on the Na_2SiO_3/NaOH [25] and the molarity of NaOH [26]. The higher content of Na_2SiO_3, the molarity of NaOH, and more amount of fly ash resulted in the higher strength of the geopolymer until a specific optimum value, and then the strength will continue decreasing [27]. However, landfill subsurface unnecessarily needs a very high strength. While the FAG-soil increases the strength by about 5.9% to 36.2%. Hence, the FAG-soil suits the slope material to increase the strength of the slope soil. It is advantageous since the rain and dry season could result in slope failure in landfill facilities [28], [29].

The compressive strength of the contaminated soil, BH2, increased along with other BH soils. The contamination did not significantly affect the strength of FAG-treated soil. The compressive strength of FAG-treated soil increases despite the contamination [30]. However, Hoai and Mukunoki [31] found that a particular contamination rate affected the soil's swelling properties.

Fig. 6 displays that the permeability of BH1, BH2, and BH3 was very low because the three soil samples were classified as clay ($1.5 – 3.2 \times 10^{-11}$ m/s). The addition of 5% FAG increased the permeability of the soil. Then, 10% FAG addition decreased the natural soil permeability to $1 – 2.5\ 10^{-11}$ m/s. For BH-2, which had the highest contamination, the permeability decreased along with the FAG addition (5 and 10% wt.).

The Scanning Electron Microscope (SEM) image of 5%, 10%, and 15% wt. in Fig 6 depicts the interparticle condition in the soil matrix. The 5% of FAG addition showed that the geopolymer crystal was not formed yet (Fig. 6a), and the 10% wt. of FAG addition made bonds between the soil layer and fly ash, and filled the void (Fig. 6b). The 15% wt. of FAG addition made an excessive FAG shown by the alkaline crystal in FA surface (Fig. 6c). However, the excessive FAG made a bigger pore as the alkaline crystal is also larger and harder.

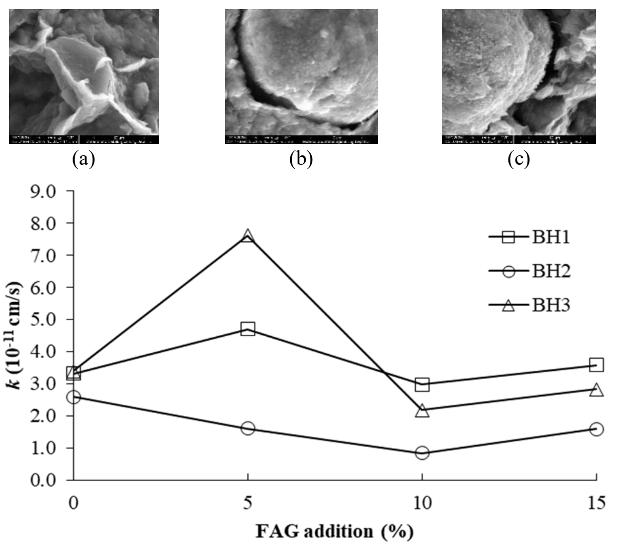

Fig. 6 The permeability and SEM images of natural soils in the study at (a) 5% wt. (b) 10% wt. and (c) 15% of FAG addition

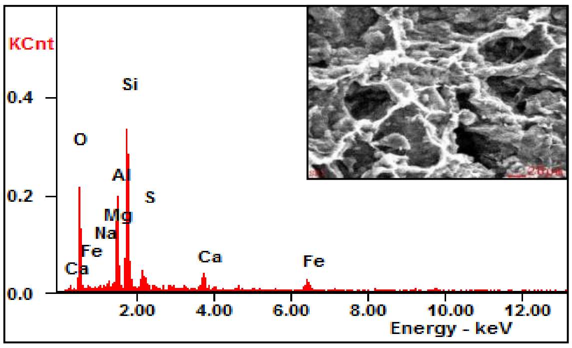

Fig. 7 The result of SEM-EDX of BH2 Soil

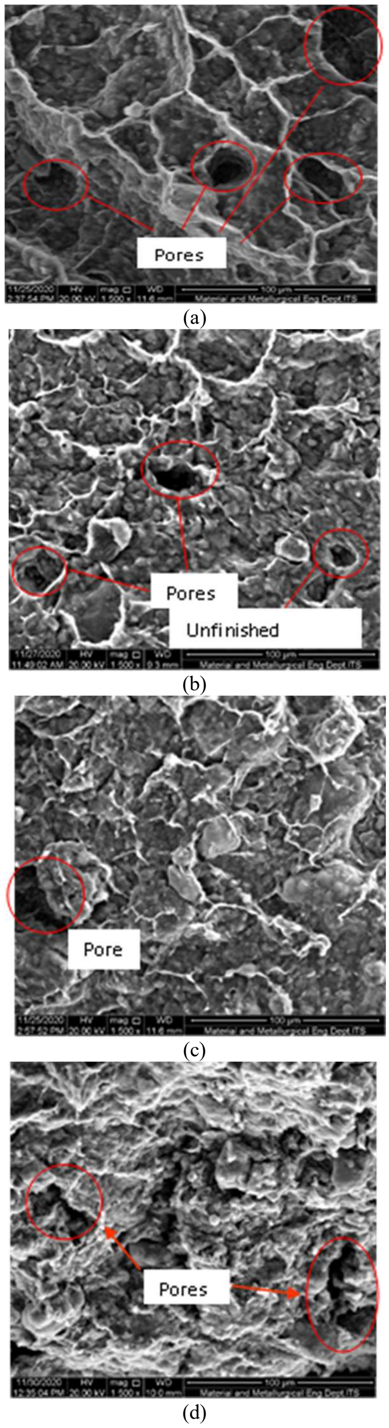

Fig. 8 The SEM images of uncontaminated soil, BH3 (a) untreated soil, (b) 5 %wt., (c) 10 %wt., and (d) 15 %wt. of FAG addition

Naturally, the permeability of soil depends on water flow paths in the soil matrices [32]. The contamination in BH2 seems to clog the paths. The Scanning Electron Microscope Energy Dispersive X-Ray (SEM-EDX) result showed the material of BH2 natural soil contained S, commonly found in landfill as Sulphate, which resulted from the metal waste (Fig.7). A high concentration of ammonium was also found in BH2, as mention in Table 1. The ammonium concentration was highest in BH2, then BH1 and BH3 corresponded to the location of BH soil sample to the waste mound, respectively. Nevertheless, the geopolymer is known resistant to acid and sulfate attacks [33], [34].

Fig. 8a shows the typical SEM images of clay soil with some pores. The pores of the natural soil were about 30-75 µm. The 5% FAG addition formed smaller pores in about 20-40 µm, and some were produced by the unfinished geopolymer reaction, as shown in Fig. 8b. The 5% addition of FAG was hypothetically not enough for the reaction of the soil, and the alkali resulted in the partially reacted soil particle (Fig.9a). The unreacted and partially dissolved FA particles induced agglomerations added voids between particles and resulted in the increased permeability [35]. The admixture of the soil and FAG reached the lowest permeability in 10% FAG addition. The particles are more compact, and the pores are filled by FAG [35], [36], as shown in Fig 8c. The 15% FAG addition had several residual FA (Fig.9b) and pores, increasing the permeability (Fig.8d).

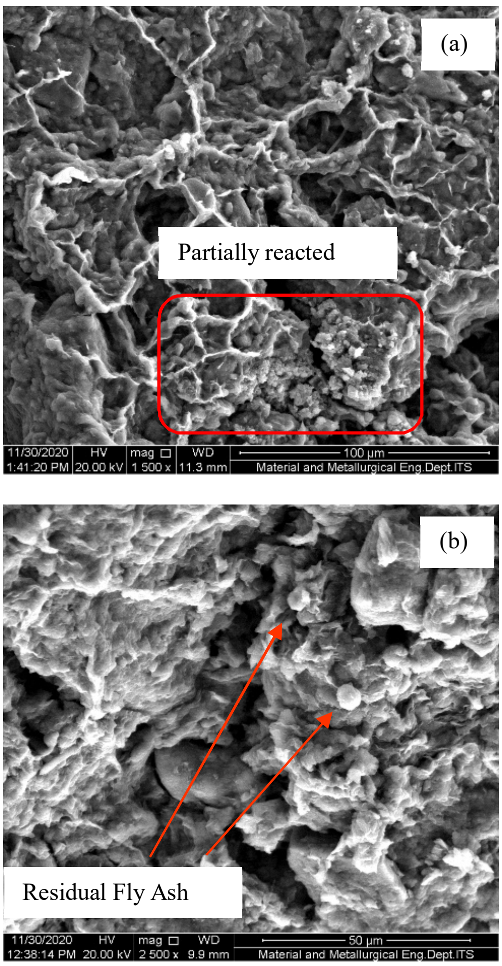

Fig. 9 The SEM images of 5% wt. (a) and 15% wt. of FAG addition

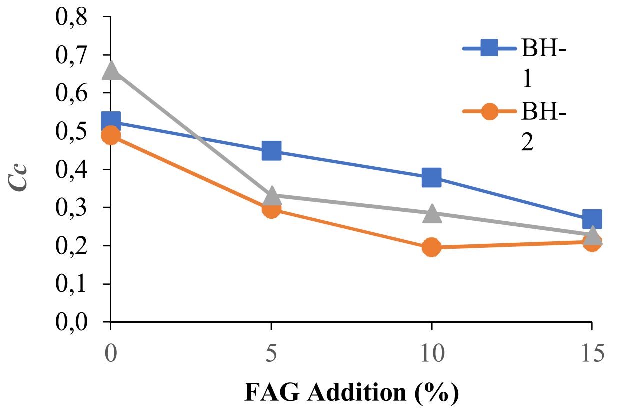

Fig. 10 The compression index of natural soils under study at different FAG addition

In liquid flows in the soil particles of the landfill subsurface, the void will be filled by the water and/or the leachate. Notably, in a tropical country like Indonesia, which has a dry and wet season, the compression index, *Cc* of soil will be affected by the cycle year to year. The ability of the water to flow between soil particles is measured by the oedometer test or known as the consolidation test. The same pressure, σ resulted in different void ratio, *e* on the soil, depends on the soil type [37]. The higher *Cc* indicates that the soil has a bigger void ratio change, Δe for the same Δσ.

The addition of FAG decreased the Cc of the soil [38] (Fig. 9). It indicates that the ability of the soil to bear the pressure increased. The bond of the particles was stronger as the FAG addition induced geopolymerization. The geopolymerization process forms bridges between particles [21]. The added polymer attaches to clay particles and constructs nanocomposites within the voids of the stabilized clay matrix [39]. The compression index of the soil decreased along with the FAG addition, meaning that the FAG stabilization resulted in a less compressible stabilized soil [32], [40].

IV. Conclusions

The research presented the standard compaction tests, oedometer tests, and unconfined compression strength tests on the natural and the FAG-treated soil. The mechanical properties of the natural soil and the FAG-treated soil were compared among the highly contaminated soil (BH-2), mildly contaminated soil (BH-1), and uncontaminated soil (BH-3). The results revealed that the contamination did not significantly affect the soil's mechanical properties. The contamination rate of BH-2 was hypothetically not high enough to affect the mechanical properties. However, the permeability of 10% FAG addition was the lowest among the stabilized soils. It indicates that 10% FAG was the optimum addition for Ngipik Landfill subsurface stabilization from a permeability perspective.

Despite the contamination, the compaction curves and compressive strength had typical results between the three soils. The dry density decreased by 5% and 10% FAG addition and increased by 15% FAG. The FAG-treated soil had a lower dry density than the natural soil since the fly ash had low specific gravity than clay. Nevertheless, more addition of FAG increased the dry density of the soil because of the geopolymerization process.

The Ngipik Landfill subsurface contamination rate did not significantly affect the mechanical properties of the FAG treated soil. However, as a contaminant, the rate of ammonium should further be investigated for how much ammonium, and not limited to any other contaminating substances, would affect the mechanical properties of the soil.

NOMENCLATURE

C_c	compression index	-
e	void ratio	-
Vv	volume of void	cc
Vs	volume of solid	cc
Gs	specific grafity	-
K	permeability	m/s
Wc	water content	%
Ww	weight of water	g
W	total weight	g

Greek letters

σ	Overburden pressure	kg/m^2
γ_d	dry density	kg/m^3

Abbreviations

FAG	fly ash based geopolymer
BH	bore hole
USCS	Unified Soil Classification System
ASTM	American Society for Testing and Materials

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