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Characterization of Freshwater Sludge and its Potential Application in Geotechnical Engineering

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ABSTRACT

Freshwater sludge (FS) is a by-product generated from the water treatment process using aluminium salts, lime, and polyelectrolyte to precipitate clay, colloidal particles, algae, and humic substances from water resources. There are more than 10,000 tons of FS produced worldwide per day, which is largely dumped in landfill, causing both economic and environmental issues. Therefore, it is critical to reuse or recycle FS into different applications to achieve economical and sustainable goals. In this context, this study introduced the characterization of FS generated worldwide from current literature, as well as its applications in geotechnical engineering. The results showed that the main oxides of FS were aluminium oxide (Al_2O_3), sulfur trioxide (SO_3), and silicon dioxide (SiO_2). The main minerals of FS were Al-(SO_4) compound, kaolinite ($Al_2Si_2O_5(OH)_4$), and quartz (SiO_2). Based on the chemical and mineral compositions of FS, the possible applications of FS in geotechnical engineering include using FS as a binder for strength improvement in soils and stabilization of dispersive clay with high content of sodium ions, and as an adsorbent or to enhance the effectiveness of other adsorbents for treatment of heavy metal-contaminated soils.

Keywords: Freshwater sludge, Characterization, Geotechnical Engineering, Literature review

1 INTRODUCTION

Freshwater sludge (FS) is a by-product generated from the water treatment process. A typical water treatment plant includes screening, coagulation and flocculation, sedimentation, filtration, and disinfection, as shown in Figure 1. Firstly, raw water from reservoirs or rivers is pumped through self-cleaning screens to remove particles. Then, coagulants and coagulant-aids like alum, aluminium sulphate ($Al_2(SO_4)_3$), aluminium chlorohydrate ($Al_2(OH)_5Cl$), lime, polyelectrolyte, or Na hypochlorite are added to bind or flocculate small suspended matter and particles, such as silt and sand, to form larger and heavier clumps (Ling et al. 2017; Stackelberg et al. 2004). In the sedimentation process, particles combine into larger clumped particles that settle to the bottom of the tank and are removed. The generated sludge in this process is called freshwater sludge (FS). In some waterworks, to meet the handling and trafficability requirements for landfill disposal, the sludge is dewatered at the waterworks by mechanical techniques (such as belt filter presses, recessed-plate filter presses, or centrifuge dewatering systems) or by air drying the material onsite in shallow drying beds or sludge lagoons (O'Kelly, 2008). Figure 2 shows a photo of typical filtered and pressed sludge cakes.

The production of FS per day is more than 10,000 tons worldwide and its generation has increased drastically due to rapid growth in urbanization and industrialization (Zhang et al. 2021). Currently, most of this FS is underutilized and is filtered to form sludge cakes that are then disposed in landfills, causing both economic and environmental issues. Hence, sustainable and economical recycling options are urgently required. Some studies attempted to reuse the FS as part of the building materials due to its potential cementitious properties, including using FS as a partial substitution for fine aggregate (Hoppen et al. 2005) and cement (Gastaldini et al. 2015), as one of the raw materials for the production of cement (Chen et al. 2010), partially replacing mineral clay for brick and ceramic (Gastaldini et al. 2015; Kizinievič

et al. 2013), and as an additive in concrete and mortar (Rabie et al. 2019). In addition to its application in building materials, FS may also be of use in geotechnical engineering.

This study summarized the characterization of FS generated worldwide from current literature, including chemical composition, mineral composition, and physical properties of FS. The potential applications of FS in geotechnical engineering were also reviewed and suggested.

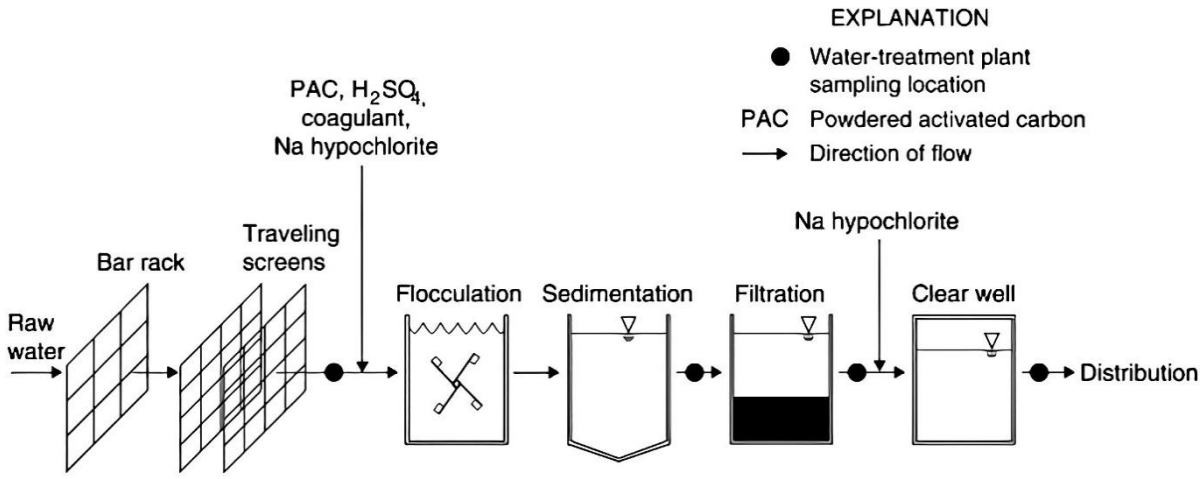


Figure 1. *Schematic diagram of a typical freshwater treatment plant* (Stackelberg et al. 2004)



Figure 2. *Photo of a typical freshwater sludge cake (Watanabe et al. 2014)*

2 CHARACTERIZATION OF FS FROM CURRENT LITERATURE

2.1 Chemical composition

Table 1 shows the main chemical composition of FS from different countries, detected by X-ray fluorescence (XRF). The main types of oxides of FS are similar in different countries, including aluminium oxide (Al_2O_3), sulfur trioxide (SO_3), and silicon dioxide (SiO_2). Al_2O_3 and SO_3 are associated with coagulants (e.g., $Al_2(SO_4)_3$) applied in the water treatment process, accounting for 15.8~31.98% and 0.02~3.39%, respectively. There is also 4.87~40.33% of SiO_2 detected in FS. This large variability of SiO_2 is due to different raw water sources containing different soil or sand. Additionally, most studies reported the loss of ignition in the XRF results of FS, ranging from 6.15~42.06%. This is mainly caused by the loss of organic matter, dehydroxylation of phyllosilicates, decarbonization of carbonates, and so on (Rodríguez et al. 2010). The presence of organic matter is attributed to the presence of algae and bacteria, humus-like material derived from the decay of plants, and a small quantity of extracellular polymeric substances produced by bacteria (Zhou et al. 2018).

Table 1. *The main chemical composition (% by weight) of FS detected by XRF*

Reference	Location	Al_2O_3	SO_3	SiO_2	CaO	MgO	Fe_2O_3	Loss of ignition (LOI)
Zhang et al. (2021)	Singapore	34.77	9.65	8.31	1.11	NM	2.31	42.06
Liu et al. (2021)	Australia	31.11	3.39	31.11	4.32	0.96	4.94	29.5
Liu et al. (2020)	Australia	28.27	0.48	26.43	5.36	1.11	6.66	6.15
Ling et al. (2017)	Malaysia	31.84	0.14	40.33	0.09	0.48	6.43	18.5
Espejel-Ayala et al. (2014)	Mexico	31.98	ND	33.2	0.64	0.63	4.94	26.1
Abo-El-Enein et al. (2017)	Egypt	22.21	ND	36.51	2.66	1.34	5.65	28.1
Cremades et al. (2018)	Spain	15.8	ND	53.7	14.4	3.6	5.00	NM
Montalvan & Boscov (2016)	Brazil	16.5	0.02	68.1	0.02	0.04	ND	22
Boscov et al. (2021)	Taiaçupeba	29.6	ND	4.87	0.49	<0.1	12.3	22~52.6

NM, Not mentioned; ND, Not detected

2.2 Mineral composition

The main minerals of FS reported by different studies using X-ray Powder Diffraction (XRD) are summarized in Table 2. The types of mineralogical phases of FS from different countries vary widely due to different raw water sources and water treatment methods, but the major minerals are similar, including Al-(SO_4) compound, kaolinite ($Al_2Si_2O_5(OH)_4$), and quartz (SiO_2). The Al-SO_4 compound such as $Al(OH)_3$ and $Al_2(SO_4)_3 \cdot 16H_2O$ are mainly derived from the coagulants (e.g., $Al_2(SO_4)_3$) used in the water treatment process. Kaolinite and quartz are associated with soil or sand particles from raw water sources.

Table 2. *Mineralogical phases of FS (detected by XRD)*

Reference	Location	Mineralogical phase
Zhang et al. (2021)	Singapore	Gibbsite ($Al(OH)_3$), Kaolinite ($Al_2Si_2O_5(OH)_4$), muscovite ($KAl_3Si_3O_{10}(OH)_2$), quartz (SiO_2), Alunogen $(Al(H_2O)_6)_2(SO_4)_3(H_2O)_5$
Liu et al. (2021)	Australia	SiO_2, $CaSO_4$, $KAlSi_3O_8$, $Mg_3AlSi_3O_5(OH)_4$, $(Al_{13}O_4(OH)_{25}(H_2O)_{10}(SO_4)_4(SO_4)_8 \cdot 20H_2O$
Roque & Carvalho, (2006)	Portugal	$CaCO_3$, $3CaO \cdot Al_2O_3 \cdot CaCO_3 \cdot 11H_2O$, $Ca(OH)_2$, $MgCO_3$, $Al_2(SO_4)_3 \cdot 16H_2O$, $CaSO_4 \cdot 2H_2O$
Ling et al. (2017)	Malaysia	kaolinite ($Al_2Si_2O_5$), quartz (SiO_2), cubic-zeolite (SiO_2) and hematite (Fe_2O_3)
Montalvan & Boscov, (2016)	Brazil	Quartz (SiO_2), kaolinite ($Al_2Si_2O_5(OH)_4$), goethite (FeO(IH)), muscovite ($KAl_3Si_3O_{10}(OH)_2$)

Abo-El-Enein et al. (2017)	Egypt	Quartz (SiO_2), illite ($(K,H_3O)(Al,Mg,Fe)_2(Si,Al)_4O_{10}[(OH)_2,(H_2O)]$), albite ($NaAlSi_3O_8$)
Espejel-Ayala et al. (2014)	Mexico	Quartz (SiO_2), kaolinite ($Al_2Si_2O_5$), albite ($NaAlSi_3O_8$)
Cremades et al. (2018)	Spain	Albite ($NaAlSi_3O_8$), anorthite ($CaAl_2Si_2O_8$), calcite ($CaCO_3$), chlorite ($(Mg,Fe,Al)_3(Si,Al)_4O_{10}(OH)_2$), dolomite ($CaMg(CO_3)_2$), diopside ($CaMgSi_2O_6$), gehlenite ($Ca_2Al(AlSiO_7)$), quartz ($SiO_2$), illite ($(K,H_3O)(Al,Mg,Fe)_2(Si,Al)_4O_{10}[(OH)_2,(H_2O)]$, microcline ($(KAlSi_3O_8)$), wollastonite ($CaSiO_3$), montmorillonite ($(Na,Ca)_{0.33}(Al,Mg)_2(Si_4O_{10})(OH)_2 \cdot nH_2O$)

2.3 Physical properties of FS

As the FS has some soil-like characteristics, several soil mechanics tests were used to analyze its physical properties in different studies. Table 3 shows some physical properties of FS from limited literature. The pH of FS is around 5.74~12.6. The water content, defined as the ratio of the mass of the pore water to the mass of the dry solids, varies from 86.18~714% in different studies. The high water content (> 200%) is due to FS not being pressed into cakes but in slurry form. The forms of FS in cakes or slurry lead to very variable values of liquid limits (65~550%) and plastic limits (30~325%). Additionally, the presence of organic matter usually results in the high plasticity of the soil (Mitchell and Soga 2005).

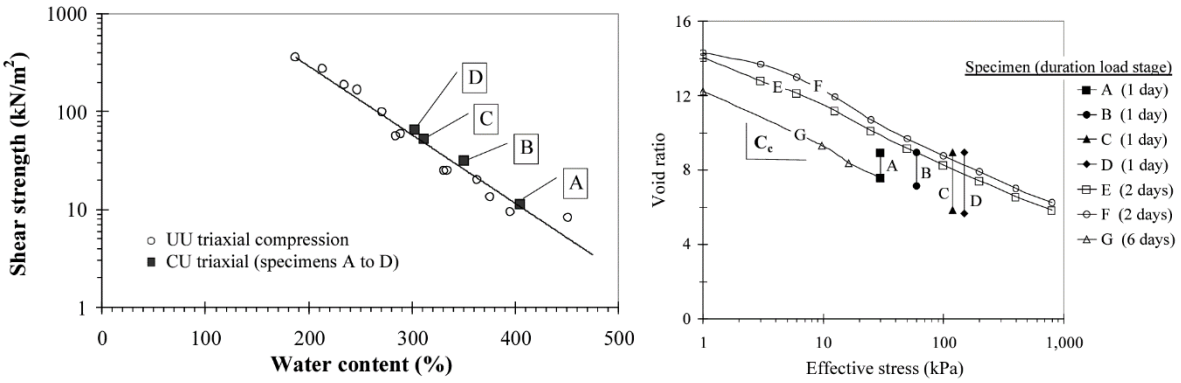
The FS has a specific gravity in the range of 1.83~2.72 in different studies. The specific gravity of water treatment sludge measured by Boscov et al. (2021) is close to that of general mineral soils (around 2.5~2.7). For the low specific gravity measured in some studies, one possible reason is that the organic matter in the soil usually decreases the specific gravity (Mitchell & Soga 2005). The dry density was found to decrease with increasing water content since pore water occupies an increasing proportion of the sludge volume (O'Kelly 2008). However, not all studies could successfully achieve the maximum compacted dry density due to different test methods, FS forms, drying methods, and organic contents.

Table 3. *Physical properties of FS*

Parameter	Roque & Carvalho (2006)	O'Kelly (2008)	O'Kelly et al. (2009)	Wang et al. (1992)	Boscov et al. (2021)	Espejel-Ayala et al. (2014)	Montalvan & Boscov, (2016)
Water content (%)	478.2	475~625	300~700	200~714	NM	86.18	NM
Specific gravity	2.27	1.86	1.83~1.99	2.72	2.42~2.95	NM	2.85~2.95
Liquid limit (%)	107.5	490	430~550	108	237~536	NM	239
Plastic limit (%)	81.7	240	240~325	47	30~138	NM	81
Max dry unit weight (kN/m^3)	6.27	NM	NM	2.83	NM	NM	NM
Optimum water content (%)	84	NM	NM	NM	NM	NM	NM
pH (in H_2O)	12.6	8.6	7.1~8.6	NM	6.4~7	5.94	7.2
Compression index C_c	NM	2.5~3.7	2.1~3.1	2.0~5.3	NM	NM	NM

NM, Not mentioned

Figure 3(a) shows the results of undrained shear strength versus water content and the coefficient of consolidation of FS, measured by O'Kelly (2008). The shear strength to mean effective confining pressure was between 1.6 and 2.0 for FS. The enhanced shear resistance of FS is attributed to the alum and polyelectrolyte applied in the water treatment process, which combines the constituent clay particles into flocs and binds them into clusters (O'Kelly 2008). As shown in Figure 3(b), the compression index C_c is reduced from 3.7 to 2.5 with the increase of effective stress from 3 to 800 kPa. However, this range of C_c value is still higher than that of most natural clays (generally less than 1.0), indicating that FS has a high compressibility. Similar results are also presented by Wang et al. (1992), where FS also has a higher C_c value (1.99~5.29) than that of general soils.



(a) (b)

Figure 3. *(a) Undrained shear strength versus water content (O'Kelly 2008), (b) void ratio versus effective stress plots. Note: solid symbols denote single-increment load tests (O'Kelly 2008)*

3 POTENTIAL APPLICATIONS OF FS IN GEOTECHNICAL ENGINEERING

3.1 Soil improvement

Currently, only very limited studies directly use FS for soil improvement. However, as FS contains a high amount of alum, it can find potential applications based on other similar alum-based sludges. Table 4 shows some applications of reusing alum-based sludges as additives or binders in soil improvement. Kamon & GU (2001) reported that adding alum sludge to ferrum lime significantly improved the early strength of stabilized soils, as shown in the left figure in Figure 3. This is primarily because the addition of alum contributes to the formation of calcium alumnae hydrate (CAH), and these plate-like products can combine with other hydration products in stabilized soil to form honeycomb-like structures. Aamir et al. (2019) also presented similar strength improvement in soil by using alum sludge as the stabilizer. Shah et al. (2020) conducted compaction tests by adding alum sludge as a soil stabilizer and found that the addition of alum sludge could improve soil strength even at a low compaction energy level of 600kN-m/m^3. Furthermore, FS may be also applied in the stabilization of dispersive clay (with high content of sodium ion (Na^+)), because the trivalent cations of Al in FS may replace Na^+, changing the soil structure from a dispersive state to a condensed state.

Table 4. *Potential application of FS in soil improvement*

Reference	**Additives/binders**	**Improvement in soils**
Kamon & GU (2001)	Alum sludge, ferrum lime	Enhance the early strength or flexural strength of stabilized soils
Aamir et al. (2019)	Alum sludge	Improve the strength and California Bearing Ratio (CBR) of weak soil with a very low bearing capacity
Shah et al. (2020)	Alum sludge	Improve the strength of soils even at a low compaction energy

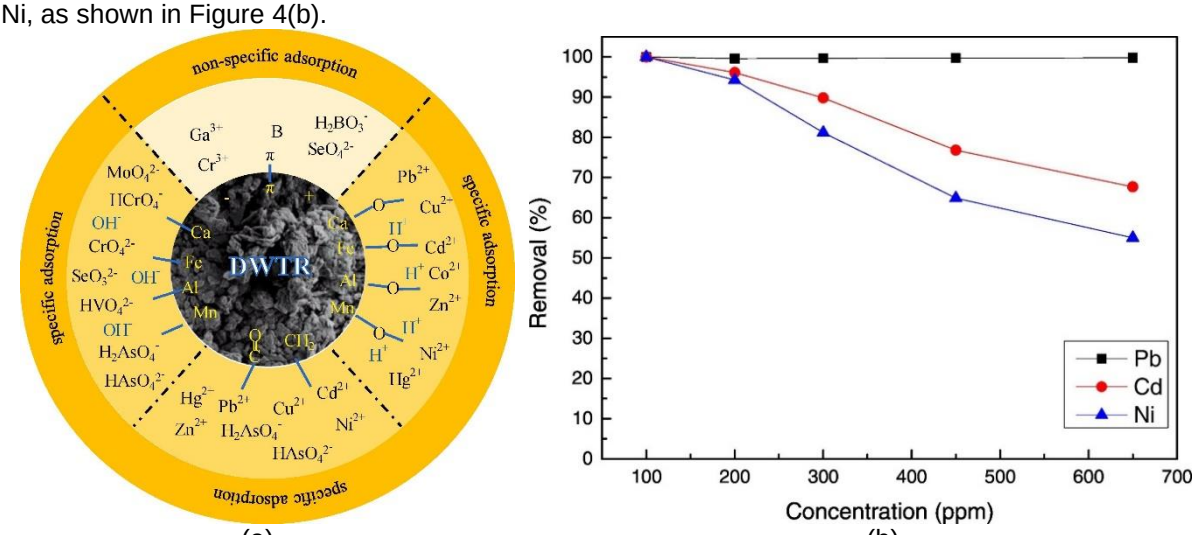
Jafari & Hassanlou (2012)	White natural alum	Stabilization of dispersive clay with high content of sodium ion

3.2 Treatment of heavy metal-contaminated soils

According to the chemical and mineral compositions of FS in different literature (Abo-El-Enein et al. 2017; Schulthess & Huang 1990), FS mainly consists of Al, Si, Fe, and organic matters (humic substances), which all have the potential ability for the adsorption of heavy metals. Hence, FS may be used as an adsorbent or to enhance the effectiveness of other adsorbents for the treatment of heavy metal-contaminated soils. Currently, no literature reports using FS for the immobilization of heavy metals in contaminated soils. Only limited studies have used drinking water sludge for the adsorption of heavy metals in water.

There are three main processes involved in the removal of heavy metals using drinking water sludge: adsorption, surface precipitation, and fixation. Because drinking water sludge is also mainly composed of aluminum, silica, and iron. A majority of the functional groups present on the surface of drinking water sludge are hydroxyl groups, which can donate protons to the solution by taking up the surrounding ions. For electron-pair acceptor metal ions (Pb, Cu, Cd, Zn, Ni, etc.), surface complexes were formed between metal and hydroxyl groups (Shen et al. 2019), as shown in Figure 4(a). It has also been shown that organic matter was also found to contribute to the adsorption of Pb, Cd, Cu, Zn, Hg, and N (Shen et al. 2019). Moreover, As(III) and As(VI) can bind to organic matter through the formation of ternary complexes with polyvalent cations (Fe, Al) to provide a bridge between As anions and organic matter (Caporale et al. 2013).

Abo-El-Enein et al. (2017) also conducted some experiments on using drinking water sludge to remove Pb, Cd, and Ni and found that drinking water sludge exhibits an adsorption efficiency towards Pb > Cd > Ni, as shown in Figure 4(b).



(a) (b)

Figure 4. *(a) Schematic of specific and non-specific adsorption mechanisms of various heavy metals by drinking water sludge* (Shen et al. 2019)*, (b) Effect of initial metal ions concentration on the removal percent of Pb, Cd, and Ni ions* (Abo-El-Enein et al. 2017)

4 CONCLUSIONS

The chemical and mineral compositions of freshwater sludge (FS) from different literature vary largely, but the major oxides and minerals are similar. The major oxides of FS are aluminium oxide (Al_2O_3), sulfur trioxide (SO_3), and silicon dioxide (SiO_2), accounting for 15.8~31.98%, 0.02~3.39%, and 4.87~40.33%, respectively. The main minerals of FS are Al-(SO_4) compound, kaolinite ($Al_2Si_2O_5(OH)_4$), and quartz (SiO_2). The organic matter was also detected in FS. The physical properties of FS in most literature reveal that FS has a high water content, high plasticity, high compressibility, and enhanced shear resistance.

The possible applications of FS in geotechnical engineering include (1) using FS as a binding material for strength enhancement in stabilized soils due to its contribution to the formation of calcium alumnae hydrate, (2) using FS as a stabilizer in the stabilization of dispersive clay with high content of sodium ion based on the cation exchange capacity of Al in FS, (3) using FS as an adsorbent or a to enhance the effectiveness of other adsorbents for the treatment of heavy metal-contaminated soils due to the potential ability of Al, Si, Fe, and organic matter for the adsorption of heavy metals. In the future, a comprehensive research on characterization analysis of FS generated in Singapore will be conducted. The goal of this research will be to identify and assess suitable applications of FS in the field of geotechnical engineering.

5 ACKNOWLEDGEMENTS

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