



## Research paper

# Evaluation and reutilization of water sludge from fresh water processing plant as a green clay substituent



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## ABSTRACT

Water sludge from a fresh water processing plant in Sungai Dua Penang was investigated from the aspects of physicochemical properties and its reutilization possibility. Morphology and size distribution of the water sludge were studied via field emission scanning electron microscopy and sieving method, respectively. The water sludge particles of irregular shapes and sizes exhibited a relatively large specific surface area of 27 m<sup>2</sup>/g, pore size of 8.8 nm and pore volume of 0.05 cm<sup>3</sup>/g. Besides, the water sludge chemical composition comprised of major components such as silica (SiO<sub>2</sub>, 40.33 wt%) and alumina (Al<sub>2</sub>O<sub>3</sub>, 31.84 wt%), which are the basic content of soil. The presence of aluminium in water sludge justified the addition of coagulant during water processing process. 97.9% of the mineral phase in the water sludge was identified as kaolinite (Al<sub>2</sub>Si<sub>2</sub>O<sub>5</sub>) and quartz (SiO<sub>2</sub>) that showed beneficial mechanical properties in the sludge-based products, e.g. pottery clay. Whereas hematite phase (Fe<sub>2</sub>O<sub>3</sub>, 1.6%) in water sludge contributes to the reddish color of the final products after firing, which makes an ideal raw material for terra cotta. Application of the water sludge as an environmentally friendlier green clay was explored through the clay reformulation. In short, reutilization of water sludge to partially replace mineral clay for ceramic products have been successful and may be useful in solving environmental issues and waste management.

## 1. Introduction

Water sludge is a by-product from fresh water processing plant and its generation increased drastically due to rapid growth in urbanization and industrialization (Xu et al., 2015; Yang et al., 2015). The usual practice for water sludge disposal in Malaysia is by discarding it back into water sources without any treatment and such activity is not in conformity with cleaner production practices. This action has also been prohibited by the Malaysian government Environmental Act 1974 and Environmental Quality Regulations (Sewage) 2009. The water processing plants in Penang state alone produced a humongous amount of ~7,000,000 m<sup>3</sup> water sludge annually based on an annual report from Malaysian National Water Services Commission in 2011 (Goto, 2013). Hence, appropriate water sludge handling is crucial as water sludge contains various toxic substances (pathogens, organic pollutants, metals) that not only poses threat to the environment but also lives of current and future human generation (Wolff et al., 2015; Xu et al., 2015).

Conventional water processing process usually involved flocculation and coagulation (Benlalla et al., 2015). The function of coagulant is to purify water by neutralizing the electrical double layer of fine colloids so as to allow agglomeration (Cheng et al., 2016). Types of coagulants mostly used were aluminium sulphate or alum [Al<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub>], iron (III) sulphate [Fe<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub>], iron (III) chloride (FeCl<sub>3</sub>) and aluminium chlorohydrate [Al<sub>2</sub>(OH)<sub>5</sub>Cl]. Cheng et al. (2016) reported using alum-generated sludge to treat ammonium-rich wastewater and to obtain ammonium alum with 93% recovery ratio. On the other hand, sludge with high mesoporosity and cation exchange capacity has been employed as an adsorbent, a cheaper alternative than activated carbon, which has efficient and extensive applications in adsorbing numerous heavy metals, organic compounds, dyes and gaseous contaminants (Hadi et al., 2015; Xu et al., 2015).

Water sludge is also a potential material for cost effective brick clay or pottery clay manufacturing since it exhibits almost similar chemical composition as in clay products. The amount of water sludge that can be incorporated into clay products and other applications is relatively

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dependent on the sludge characteristics (Benlalla et al., 2015; Wolff et al., 2015). Basically, physicochemical behavior of water sludge is determined by the properties of raw water sources, treatment procedures and total volume of water treated (Gastaldini et al., 2015). In addition, chemical composition of water sludge varies at different processing stages during treatment (Ahmad et al., 2016). As far as reported, there are various successful applications of sludge as part of the building materials, for example, cement (Chen et al., 2010; Gastaldini et al., 2015), brick, ceramic (Kizinievic et al., 2013; Benlalla et al., 2015; Wolff et al., 2015), concrete and mortar (Gastaldini et al., 2015).

The types of water sludge added into mass formation could decide the physical and mechanical characteristics of the formulated clay and the constructed ceramic bodies. Furthermore, water sludge composed of ferric oxide ( $\text{Fe}_2\text{O}_3$ ) that is a natural dye causes final ceramic to turn into red color after firing process (Kizinievic et al., 2013). Wolff et al. (2015) explained that clay products with high mechanical strength could be fabricated with an optimized clay formulation, especially referring to size distribution of sludge particles and metal oxides proportion in the mixture. Recommended composition of alumina sludge (~62.66 wt%  $\text{Al}_2\text{O}_3$ ) to fabricate a good quality brick was found to be 20% (with 30% moisture content) and fired at 900–930 °C (Benlalla et al., 2015).

The intention of our study is to evaluate water sludge as a green replacement alternative in clay formulation and to determine the possibility of converting water sludge into functional products. Physicochemical properties of the water sludge mainly characterized by examining its morphology, size distribution, chemical composition, chemical structure, purity, thermal behavior, surface area and pore size/volume. The reutilization of water sludge, as a partial substituent in green clay has been further verified via the fabrication of pottery wares. This study does not only solve the environmental pollution issue including massive generation of hazardous sludge waste but also provide carbon neutral green clay as an alternative that reduces clay-mining activities for large consumption of natural resources especially in the field of civil construction.

## 2. Experimental methods

### 2.1. Water sludge collection and preparation

Raw water sludge sample for this study was collected from a water processing plant located in Sungai Dua Penang Malaysia. To prepare raw water sludge into powder form, its clumps were initially dried in an oven over 24 h at 100 °C. Dried water sludge were then ground and ball-milled with Pascall Pestle Mortar Mill (5 min) and 4-Stage-Pot Mill machine (7 h), respectively. The weight ratio of alumina balls media to water sludge was kept at 5:1 and the speed of milling machine was set at 180 rpm. Lastly, the fine water sludge powder sieved (5 min, 1.5 mm amplitude) using a Letsch sieve shaker with sieve opening of 425  $\mu\text{m}$  to separate out larger particles.

### 2.2. Morphology and elemental analysis

Water sludge powder surface morphology was observed via a field emission scanning electron microscopy (FESEM, Carl Zeiss Supra 35VP), at 1000 $\times$  magnification with a copper anode of 5.0 kV accelerating voltage. Simultaneously, energy dispersive spectroscopy (EDS) was conducted to study the elemental presence and their respective weight fraction found in water sludge powder.

### 2.3. Particle size distribution

To determine the particle size distribution of the water sludge powder, sieving method involving a set of sieves with different opening sizes was applied. The sieves were arranged in deck from coarsest sieve

opening (475  $\mu\text{m}$ ) on top to finest at the bottom (25  $\mu\text{m}$ ). About 200 g of premeasured water sludge powder was poured into the top sieve and then sieved through via a mechanical sieve shaker (5 min, 1.5 mm amplitude). The mass of water sludge particles in each sieve was collected, weighed and recorded as in weight percentage.

### 2.4. Chemical composition analysis

The composition of chemical substances (wt%) in water sludge powder was investigated by employing an X-ray fluorescence spectrometer (XRF, Rigaku RIX-3000). Prior to the XRF analysis, loss of ignition (LOI, wt%) was measured where excess water and volatile substances were removed from water sludge until a constant water sludge weight obtained. 1 g of water sludge powder was dried in an oven (105 °C) for 12 h and weighed. The respective water sludge powder was later heated up to 1000 °C (1 h soaking time) in a furnace (Lenton Muffle Furnace 1200) with 10 °C/min heating rate. The final weight was measured again and the value of LOI (wt%) was calculated by referring to Eq. (1).

$$\text{LOI (wt\%)} = \frac{\text{weight of loss after burning}}{\text{weight of sample before burning}} \times 100\% \quad (1)$$

### 2.5. Mineralogical phase analysis

Types of mineral phase in water sludge powder were identified through crystalline structure examination by X-ray diffraction (XRD) analysis using a Bruker Axs D8 Advance X-ray diffractometer (Cu K $\alpha$  radiation,  $\lambda = 1.5406 \text{ \AA}$ ). Scanning range (2 $\theta$ ) was fixed in the 10–75° range with 0.030° step size and 38.4 s step time.

### 2.6. Structural analysis

The chemical structural properties of water sludge powder were determined by a fourier transform infrared spectrometer (FTIR, Perkin-Elmer Spectrum One). A mixture of

1 mg water sludge powder and 99 mg potassium bromide (KBr) powder was prepared and hand-pressed into a thin disc with a Hydrotec EN837-1 press. The pressed sludge-KBr disc was scanned through a range of 400–4000  $\text{cm}^{-1}$  mid-infrared wavelength, at 8 scan cycles. A pure KBr thin disc was also made and scanned for background signal.

### 2.7. Thermal behavior

Both thermogravimetric (TG) and differential scanning calorimetry (DSC) analyses were conducted concurrently to examine the weight and enthalpy change (exothermic or endothermic) in the water sludge sample throughout the heating process. 12 g of water sludge powder was heated up to 1000 °C at a 10 °C/min heating rate. The analyses utilized TG/DSC analyzer (NETZSCH STA 409 PC/P) operated under normal atmosphere condition with an empty alumina crucible as a reference. Additionally, thermal expansion meter (Linseis dilatometer) was used to study the sintering temperature and thermal expansion/shrinkage behavior of water sludge during heating. The water sludge powder was compressed into a billet form (20  $\times$  4  $\times$  4 mm) before heating was conducted in the dilatometer.

### 2.8. Pore characteristics and surface area analysis

Surface area of the water sludge was investigated by employing Brunauer-Emmett-Teller (BET) method based on the amount of nitrogen ( $\text{N}_2$ ) gas adsorbed on its surface at specific pressure. Barrett-Joyner-Halenda (BJH) theory was applied to determine the pore characteristics (e.g. pore size, pore volume) of water sludge by referring to the  $\text{N}_2$  gas adsorption/desorption activity. For BET and BJH analyses, water sludge powder was pretreated via heating (300 °C, 24 h) in a vacuum chamber

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