Water treatment sludge conversion to biochar as cementitious material in cement composite

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Abstract

Water treatment sludge was successfully thermally converted to obtain biochar as a stable material with resource potential. This research explored the application of sludge biochar as a supplementary cementitious material. The cement paste samples incorporating different amounts of sludge biochar were prepared, hardened, and analyzed for performance. The results show an improvement in hydration kinetics and mechanical properties of cement paste incorporating biochar, compared to raw sewage sludge. The mineralogical, thermal and microscopic analyses show evidence of pozzolanic activity of the biochar. The samples with 2% and 5% biochar showed higher heat release than the reference material. Specimens with 1%, 2% and 5% biochar showed a slightly higher compressive strength at 28 days compared to the reference material. Sludge conversion to biochar will incur an estimated cost of US$398.23/ton, which is likely to be offset by the substantial benefits from avoiding landfill and saving valuable cementitious materials. Therefore, this research has demonstrated that through conversion to biochar, water treatment sludge can be promoted as a sustainable and alternative cementitious material for cement with minimum environmental impacts, hence contributing to circular economy.

Keywords: Water treatment sludge; Biochar; Cement composites; Hydration kinetics; Structural performance

1. Introduction

Water treatment plants are playing a critically important role worldwide to purify raw water for human consumption (Chang et al., 2020). During the water treatment processes, chemical reagents such as coagulants and flocculants are added for the removal of fine particles and dissolved organic matter such as humic substances (Ahmed et al., 2018; Huang et al., 2020; Van Truong et al., 2021), leading to the formation of particle residues that will be removed in later stages by screening, sedimentation and filtration (Manda et al., 2016; Shamaki et al., 2021). These solid residues are commonly known as water treatment sludge, which is produced in enormous quantity e.g. 43500 ton/y in Australia (Gomes et al., 2020) and causes major environmental pollution if discharged directly to rivers (Hussein et al., 2021). Therefore, water treatment sludge is usually dewatered and sent to landfill sites (Gomes et al., 2019). However, with increasingly stringent legislation and high disposal costs, as well as the potential environmental impacts associated with improper disposal and landfill leachate (Yadav et al., 2020), both water treatment companies and local governments are seeking more economic and sustainable solutions for the long-term management of water treatment sludge (Teodosiu et al., 2018).

Currently, the search for solutions to minimize the adverse environmental impacts of solid waste and landfill practices has led to the development of research into the use of water treatment sludge as a potential resource. Many studies have been carried out to recover the chemical coagulants used during the water purification process and to directly use such sludge as coagulants in parts of the wastewater treatment process (Taheriyoun et al., 2020; Letshwenyo and Mokgosi, 2021). More recently, another area that has been extensively explored is the incorporation of such sludge in different construction materials such as bricks (Erdogmus et al., 2021), concrete paving blocks (Liu et al., 2020), lightweight aggregates (Lee et al., 2021; Mañosa et al., 2021b), mortar (Li et al., 2021), alkali-activated cements (Mañosa et al., 2021a), and geopolymers (Gomes et al., 2019; Santos et al., 2019), as clay and sludge have a similar mineralogical composition. As construction materials are in high demand globally, the potential use of water treatment sludge in construction industry will greatly reduce material cost, improve their environmental credentials, and make their contributions towards circular economy (Shamaki et al., 2021).

Up to now, most research has used raw sludge which is mixed with other materials (Gomes et al., 2020), or in some cases the sludge has been calcined before its incorporation (Benlalla et al., 2015; de Godoy et al., 2019). More recently, there has been research work investigating the possibility of recycling and valorizing water treatment sludge into a rich-carbon based material called biochar through pyrolysis (Lee et al., 2020; Hung et al., 2020). Biochar is a carbon-rich material obtained from carbonized biomass under a low oxygen atmosphere, with half of the carbon content from the original organic compounds (Ahmed et al., 2016; Mulabagal et al. 2017). Biochar commonly presents more stabilized carbon concentration than the original raw material; hence, the carbon contained in biochar is less likely to be released back into the atmosphere as carbon dioxide. Therefore, the motivation for biochar use stems from its unique properties (e.g. porosity, surface area, surface charge) and diverse applications in fields such as waste management, energy production, climate change mitigation and soil improvement, which individually or in combination can have very positive environmental and socio-economic effects (Lehmann, 2007; Steiner et al., 2008; Beesley et al., 2010). Recent research shows that the incorporation of only 1% biochar substitution could sequester approximately 0.5 Gt of CO_2 annually by the concrete industry, approximately 20% of the entire annual CO_2 released by cement industry (Di Tommaso and Bordonzotti, 2016). Similarly, Gupta and Kua (2017) suggested that by using biochar in construction materials to capture and then lock atmospheric carbon dioxide in buildings and structures can potentially reduce 25% of greenhouse gas emissions from the construction industry.

There has been a growing interest in the use of biochar as cementitious materials to improve its mechanical performance. Akhtar and Sarmah (2018) investigated the substitution of cement by 1% biochar produced from different raw materials (poultry waste, rice hulls and sludge from pulp and paper mills). Their results revealed that the biochar of pulp-paper sludge obtained compressive strength similar to the control sample. Interestingly, the replacement of only 0.1% of rice husk biochar increased the tensile and flexural strengths of concrete by 20% comparing with the control samples. Using biochar made from wood and sawdust at 300 °C and 500 °C, Gupta et al. (2018) showed that the addition of 2% biochar in the mortar mixture offered a significant improvement in the compressive and flexural strength of the composite. In addition, the permeability of the composite was significantly reduced due to the addition of biochar, due to the effect of biochar as a micro-filler in the mortar.

There is some recent research on the application of biochar from different sources in Portland cement, however so far, there is still no published study, to our knowledge, on the application of biochar prepared from water treatment sludge as an ingredient of cement composites. Therefore, the application of water treatment sludge biochar in cement paste is the novelty of this study. The comparison of raw sludge and sludge biochar in their structural performance as a cement paste is a further novelty. In order to improve the valorization of water treatment sludge as an ingredient to the construction industry, this research aimed to prepare sludge biochar, characterize biochar for key physicochemical properties, determine the mechanical strengths of fresh and hardened cement paste incorporating different contents of sludge biochar, and analyze the impacts and interaction of sludge biochar on the hydration kinetics of cement products.

2. Materials and methods

2.1. Raw materials

The water treatment sludge was collected from the Cascade Water Filtration Plant at Katoomba, NSW, Australia, where ferric chloride ($FeCl_3$) was used as the coagulant in its conventional water treatment process. The dewatered sludge was collected from the drying bed at four different points. The sludge was dried in an oven at 105 °C for 24 h to remove the moisture, following procedures recommended by the American Society for Testing and Materials (ASTM, 2020). The dried sludge was then crushed, sieved (300 μm) and stored in an airtight plastic bag before further characterization (**Fig. A1a, Supplementary Information**).

The cement used in the study was a general purpose (GP) cement from Cement Australia, which fully complies with the Australian Standard AS 3972 (2010). The cement has a density of 3.0-3.2 g/cm^3, with a median particle size of 9.4 mm.

2.2. Biochar preparation from water treatment sludge

The naturally dried and sieved sludge was used to prepare biochar. In brief, the sludge was placed inside a fixed bed reactor (**Fig. A1b, Supplementary Information**) which was then inserted into a furnace (Labec). Pyrolysis was operated in a furnace at 700 ^{o}C for 2 h, with the temperature increase rate of 17 ^{o}C/min. To maintain a low oxygen atmosphere and prevent oxidation during the process, the nitrogen gas was injected to the reactor at a flow rate of 220 mL/min. The biochar samples were then cooled at room temperature until reaching a constant weight (**Fig. A1c, Supplementary Information**).

2.3. Cement composite testing

To examine the feasibility of sludge and sludge biochar application as supplementary cementitious material, sludge or sludge biochar was mixed with cement to prepare cement composites. Briefly, GP cement was mixed with different amount of sludge or sludge biochar (1%, 2%, 5%, 10%) based on the weight of the cement (**Table 1**). In addition, reference cement paste was prepared with no biochar addition. In order to ensure full hydration, cement paste was cast with water to cement ratio of 0.4, according to the Standard C305-14 (ASTM, 2014).

For the compressive strength test, twelve specimens of cubic moulds (50 mm × 50 mm × 50 mm) were cast for each mixture design. After demolding, the blended pastes were cured at

20 °C under 95% relative humidity and tested at 7, 14, and 28 days respectively. Compressive strength testing was conducted in accordance with the Standard C109-16 (ASTM, 2016), which provides a means of determining the compressive strength of hydraulic cement and other mortars. The compression testing was conducted on a Universal Hydraulic Test Frame (UH-500kN XR) with a REH50 load frame (Shimadzu).

The heat of hydration of the composite pastes was determined using an I-Cal 4000 isothermal calorimeter (Calmetrix), in accordance with the Standard C1679 (ASTM, 2009). All the mixtures were prepared by hand for 30 s, from which 25 g of the paste was used for each cup mixture test. All measurements were performed during the first 48 h of hydration at a controlled temperature of 20 °C.

Table 1. The experimental design of different cement paste mixtures.

Mixture	**Cement (g)**	**Sludge or biochar (g)**	**Water (g)**	**Water/binder ratio**
Reference	2800	0	1120	0.4
Sludge (1%)	2800	28	1120	0.4
Sludge (2%)	2800	56	1120	0.4
Sludge (5%)	2800	140	1120	0.4
Sludge (10%)	2800	280	1120	0.4
Biochar (1%)	2800	28	1120	0.4
Biochar (2%)	2800	56	1120	0.4
Biochar (5%)	2800	140	1120	0.4
Biochar (10%)	2800	280	1120	0.4

2.4. Characterization of materials and cement paste

The composite samples containing different amounts of sludge or the sludge-derived biochar were subjected to extensive characterization. The thermal gravimetric-differential scanning calorimetry (TG-DSC) analysis was performed using a Simultaneous Thermal Analyzer (model

STA 449 F5 Jupiter). The powder from the composite pastes of 28 days curing were collected by cracking blended pastes and immediately immersed into ethyl alcohol for 24 h and dried at 40 °C overnight. The simultaneous thermogravimetry used the nitrogen gas at a gas velocity of 20 mL/min. The 28-day powder samples were heated from 30 °C to 900 °C at a rate of 20 °C/min.

X-ray diffraction (XRD) analysis was used to identify the mineralogical characterization of sludge biochar and the hydration evolution of the composite at early-age hydration, the XRD analyses were performed by using a Bruker D8 Discover diffractometer. The fine powder from 28 days curing was subjected to the XRD apparatus at 20 mA and 40 kV, scanned within a 2θ range of 5° to 55°.

The morphology of the sludge biochar and hydration products of composite was observed by scanning electron microscope (SEM-EDS and Mapping) to examine the surface structure of composite past samples. The fractured cement paste cubes (28 days) from the compressive strength test were used. Prior to the analyses, the samples were dried at 40 °C for 24 h, then coated with gold before examination in a Zeiss Supra 55VP SEM operated at 20 kV.

2.5. Biochar cost analysis

The full cost-benefit analysis of biochar production is highly important to support business strategy and exploitation of biochar as a valuable product, which is not the focus of this study. To prove the concept, the biochar production cost from sewage sludge is estimated, by considering both capital investment cost and operating cost, following approaches reported by Ahmed et al. (2016) and Nematian et al. (2021). The following cost analysis is based on a pilot plant of biochar, with one ton per day of biochar production.

$$C_P = C_{CI} + C_{OC} \quad (1)$$

where C_P is the overall production cost, C_{CI} is the total capital investment, C_{OC} is the total operating cost, all in US$/ton. The detailed breakdown of each cost has been described by

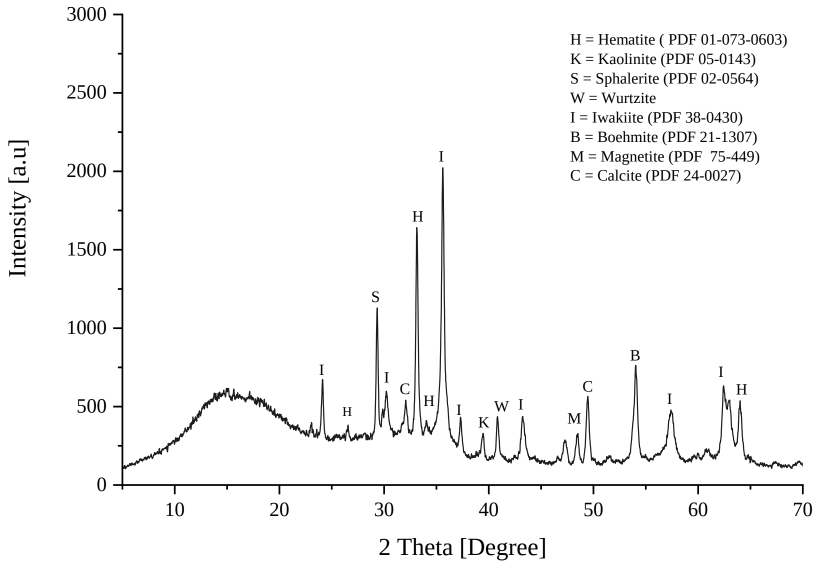
Ahmed et al. (2016). In addition, the calculation of total biochar production was estimated using a biochar budget tool proposed by Nematian et al. (2021).

3. Results and discussion

3.1. Characterization of sludge biochar

The XRD analysis of sludge biochar (**Fig. 1a**) shows a material with more crystalline peaks than the raw sludge (Gomes et al., 2020). Most of the peaks detected were formed by the dehydroxylation and conversion of iron oxides (goethite, ferrihydrite) in hematite and magnetite (Teixeira et al., 2011; Kizinievič et al., 2013; Özdemir and Dunlop, 2000). The XRD pattern shows the presence of wurtzite (ZnO), which was the convention of natural sphalerite presented on the raw water treatment sludge (Hamza et al., 2017). Furthermore, the presence of iwakiite ($MnFe_2O_4$) type is evident in the XRD profile, which is probably due to transformation reaction between manganese and iron oxides, as described by Rzepa et al. (2016). Boehmite has been detected due to partial dehydration of amorphous aluminum oxides (Tantawy et al., 2015). The single peak of kaolinite is indicative of the dehydroxylation transformation of the crystalline phase into amorphous metakaolin (Kakali et al., 2001; de Godoy et al., 2019).

a



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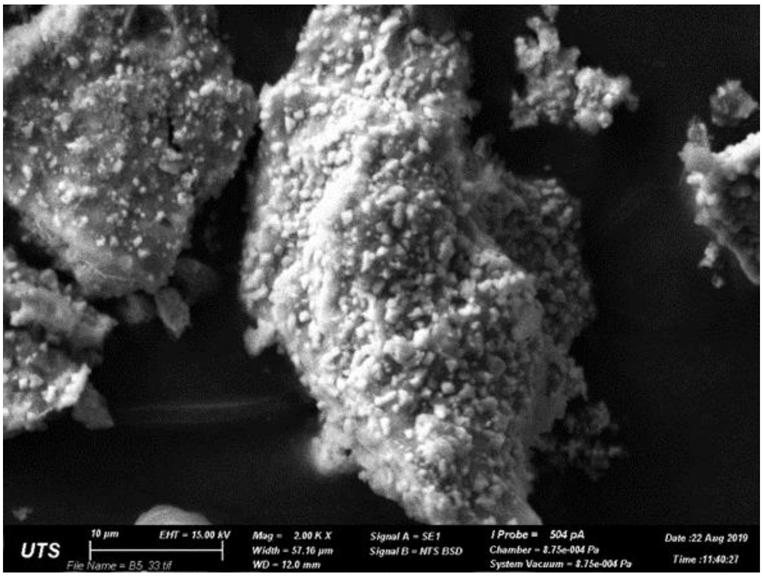


Figure 1. (a) The mineralogical composition of biochar from XRD analysis, and (b) SEM of biochar (2000 ×).

The morphology of biochar from sludge preparation was examined by SEM **(Fig. 1b**). As can be shown, after the thermal pyrolysis process, the sludge grains underwent a significant transformation in their structure. In general, non-homogeneous particles with development rough texture and small clusters on the surface can be observed (San Nicolas et al., 2013; de Godoy et al., 2019).

3.2. Heat evolution of cement paste

During the mixing of cement, aggregate and water, significant amounts of heat are released due to the exothermic nature of chemical reactions (i.e. hydration), which increase the temperature and maturing of concrete. **Figure 2** shows the heat evolution pattern by cement paste samples with different substitution of sludge biochar. The samples with 2% and 5% of sludge biochar released more heat than both the reference sample and 10% of sludge biochar addition. The initial peak (first h) is associated with the hydration of C_3A, the dissolution of free lime and the wetting of Portland cement, and all mixtures had similar results. However, the mixture with 10% sludge biochar showed slightly more heat than the reference. The dormant stage (phase 2) occurred during the first 2-3 hours of reaction for the reference samples (cement only). As the sludge biochar was added to the cement paste, this dormant period increased, reaching 5 hours concerning the samples with 10% sludge biochar. Similar to the reference sample, the curve with 1% sludge biochar showed two peaks, with the same intensity. These two peaks correspond to the rapid hydration of C_3S and C_3A respectively, followed by a deceleration period of heating. The mixtures with 2% and 5% of sludge biochar also showed two peaks but both with higher intensity compared to the reference and 1% biochar batches. This phenomenon is possibly associated with the filler surface effect, providing additional nucleation sites for calcium silicate hydrate (C-S-H) (Scrivener et al., 2015). The addition of 10% sludge biochar shows almost the same hydration heat compared to the reference, but with around 2 hours of delay in the reaction. It is important to note that the sludge biochar has shown improved hydration heat properties of the cement composites compared to the water treatment sludge itself (Gomes et al., 2020). For example, even the sample with the addition of 10% biochar had a similar heat peak compared to the reference sample, which did not happen with samples with the same proportion of raw sludge even at 48 h. In addition, another substantial improvement is the time delay in the maximum peak (formation of inner C-S-H) which has been reduced to 12.5 h with 10% biochar addition, compared to 38 h for 10% raw sludge addition. In other words, the addition of 10% sludge biochar caused only 2 h of delay for the maximum peak compared to the reference sample.

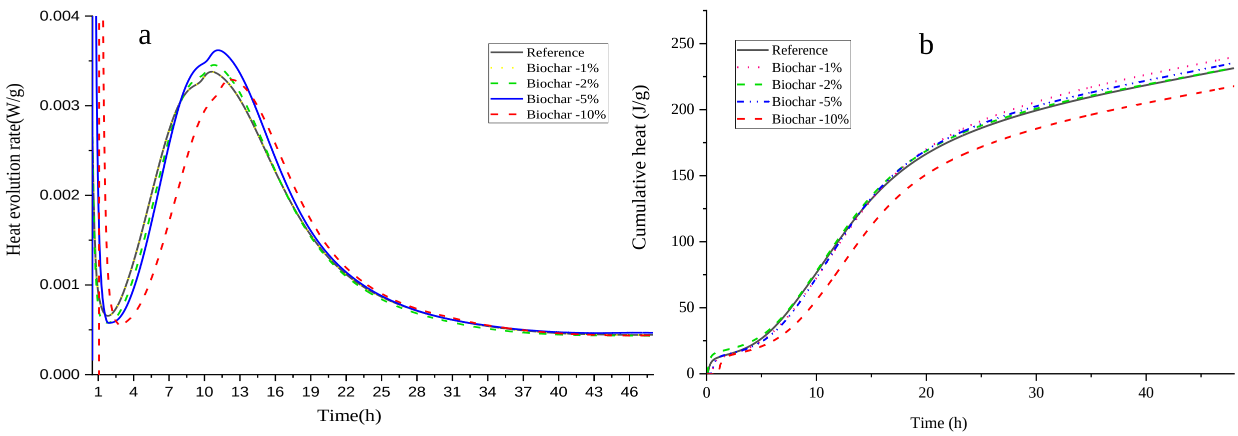
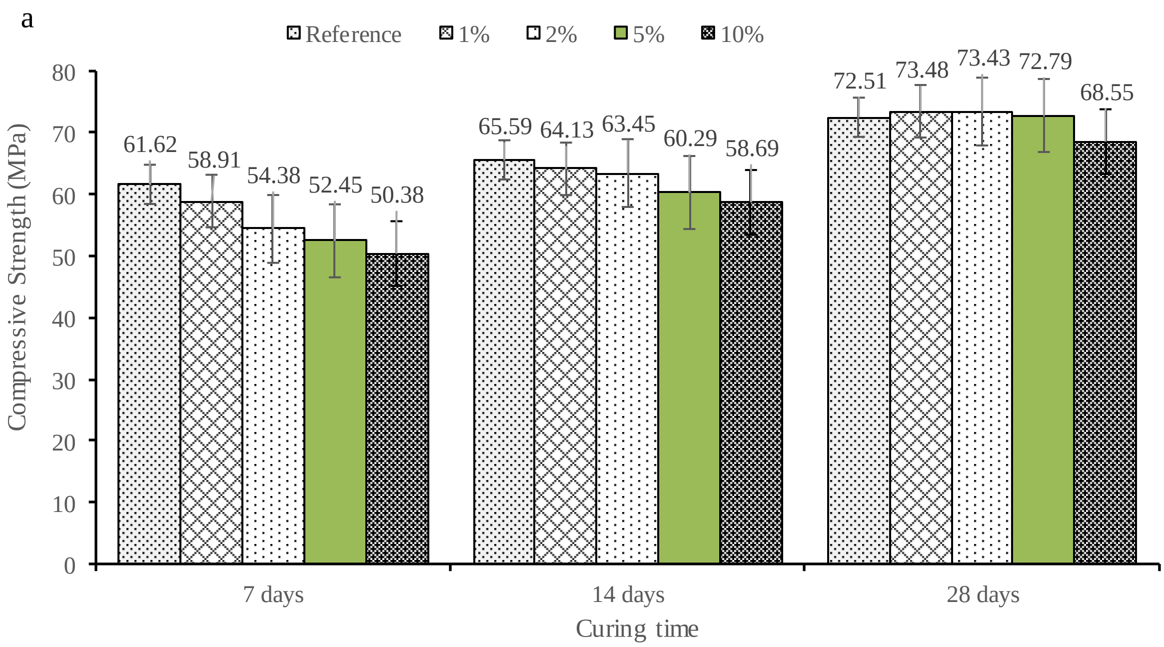


Figure 2. (a) Hydration heat evolution flow, and (b) cumulative heat evolution with the additions of different amounts of sludge biochar.

3.3. Compressive strength of sludge biochar composite

Concrete is used as a structure material; thus, its mechanical strengths should be fully tested. The changes in comprehensive strength of cement paste samples over different curing time are examined (**Fig. 3a)**. At seven days of curing, mixtures with biochar showed slightly lower compressive strength than the reference material, with 18.3% less compressive strengths when 10% biochar was added. However, there was a significant increase in the compressive strength of the biochar composite over 28 days of curing. The mixtures with 1%, 2% and 5% sludge biochar showed a slightly higher compressive strength at 28 days compared to the reference material. Even with the addition of 10% sludge biochar, the cement paste presented a comprehensive strength value close to that of the reference material, with only 5.5% less strength than the reference material with 100% cement. The analysis of variances (ANOVA) indicates that there is no statistical difference between the reference sample and any biochar composite samples, at the end of 28 days of curing. The findings therefore confirm that up to 10% biochar can be safely used to maintain the structural performance of cement paste.



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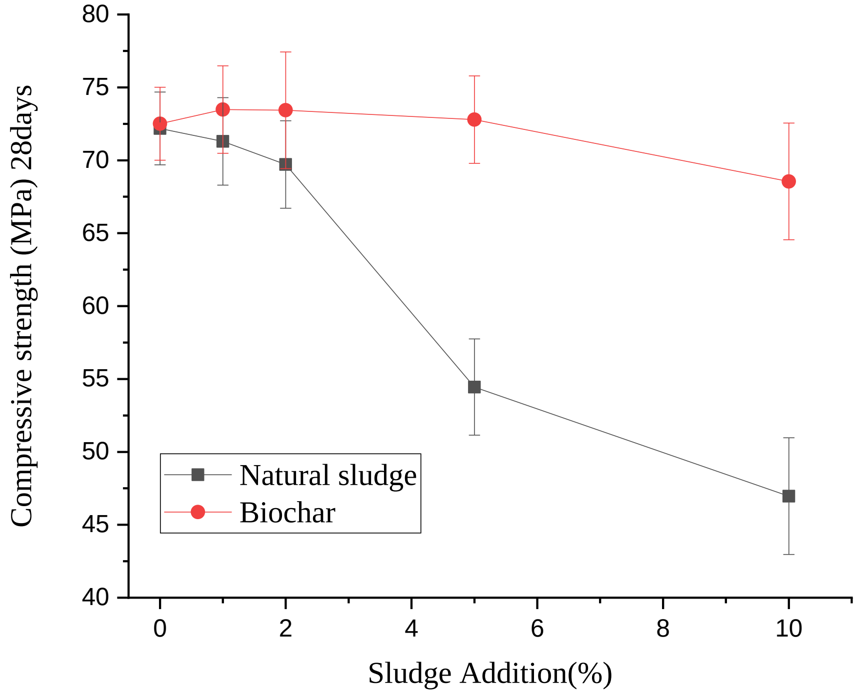

Figure 3. (a) Compressive strength of cement paste with different amounts of sludge biochar. (b) Comparison of compressive strength of cement paste at 28 days between water treatment sludge and sludge biochar.

Regarding the results at 28 days of curing, **Fig. 3b** compares the compressive strength between the composites of natural sludge and sludge biochar. For the additions of 5% and 10% sludge biochar, there was a significant increase in the compressive strength of 37% and 46% respectively, compared to the composite with natural sludge. This improvement in the behaviour of the biochar material was mainly due to the removal of organic matter in raw sludge through the thermal treatment, and also possibly the pozzolanic activity of biochar, even in small proportion at 28 days (Kaish et al., 2018; de Godoy et al., 2019).

3.4. Mineralogy of hydration products from sludge biochar composite

The XRD analysis of sludge biochar composite cement paste mixture at 28 days is shown in **Fig. 4a**. Very similar to mixtures with raw sludge, the incorporation of the sludge biochar did not create significant new crystalline phases to the composite. All mixtures presented the main crystalline phases, i.e. calcium silicate hydrate (CSH), ettringite (Aft) and portlandite (CH). A single peak of calcite was detected in all mixtures. It is important to note that there was a slight decrease in the peak intensity of calcite in mixtures with biochar compared to blends with raw sludge (Gomes et al., 2020).

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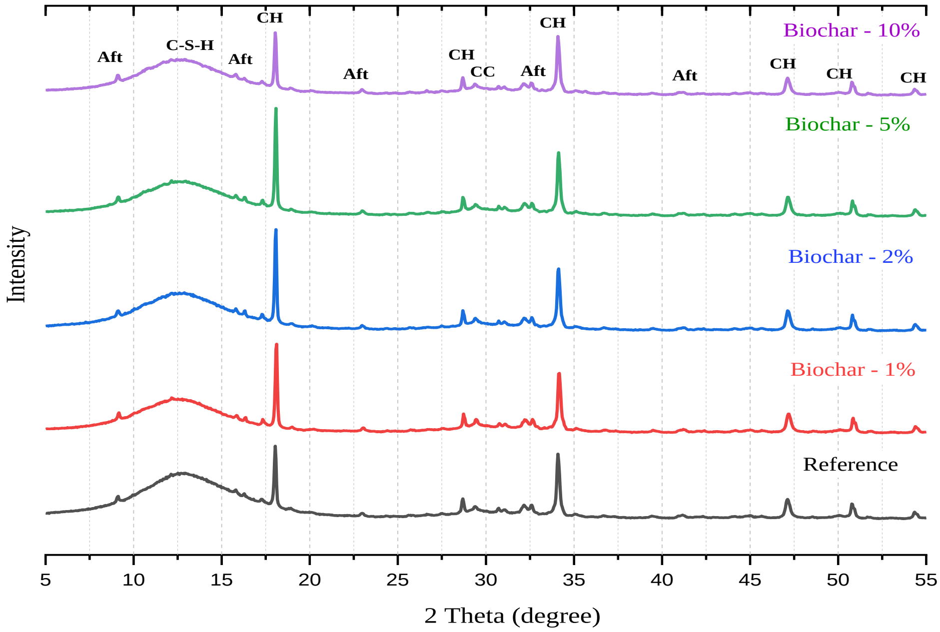

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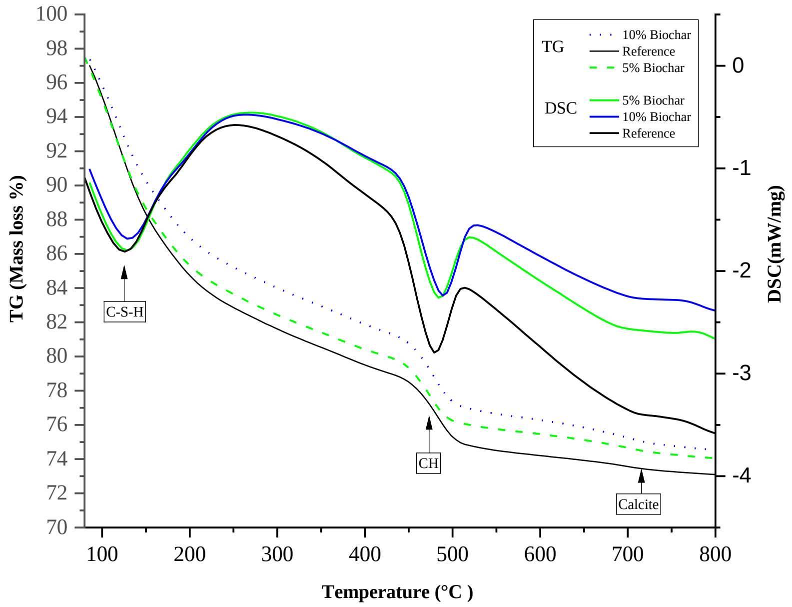


Figure 4. (a) XRD of sludge biochar hydrated mixtures. Aft =Etringita, C-S-H =Calcium silicate hydrate, CH= Portlandite, CC= Calcite. (b) TG-DSC pattern of reference, 5% and 10% of sludge biochar.

3.5. Thermogravimetry and differential scanning calorimetry analysis

To fully understand the effects of sludge biochar on cement paste samples, their chemical composition was examined using TG-DSC. **Figure 4b** shows a comparison of the intensities of portlandite, calcium silicate hydrate (or C-S-H) and calcite between the reference sample, 5% and 10% biochar. Analyzing the first endothermic peak corresponding to C-S-H demonstrated that the cement paste sample with 5% biochar showed similar peak intensity to the reference material, and the sample with 10% biochar had a slightly lower peak intensity compared to the reference sample. The second endothermic peak is related to the calcium hydroxide or portlandite (CH). The results show that all cement paste mixtures, whether with 5% and 10% biochar incorporation, or the reference material, all had a very similar peak intensity. However, when comparing the difference between the intensity of composites with biochar and those with raw sludge, it can be seen that the concentration of C-S-H and portlandite on the biochar composites is very close to the reference material, which does not happen with the samples with raw sludge (Gomes et al., 2020). This is in agreement with the results of mechanical performance, wherein the mixtures with biochar showed compressive strength close to the reference material at 28 days. Unlike the lower intensity of C-S-H and portlandite in the raw sludge composites due to the organic matter, the reason for the lower portlandite peak and compatible compressive strength with biochar is the pozzolanic reaction with the biochar. The consumption of portlandite for the production of extra calcium silicate hydrate (C-S-H) and calcium aluminium silicate hydrate (C-A-S-H) is very common in the presence of pozzolanic materials with a reasonable concentration of metakaolinite (Frias et al., 2013; Mohammed, 2017).

3.6. Microstructure analysis

The SEM analysis (**Fig. 5**) shows the microstructure of the hydrated composite with additions of 1%, 2%, 5% and 10% biochar. The cement paste mixtures containing 1% and 2% of sludge biochar (**Fig. 5a-b)** showed clearly a well-developed structure with a network of defined hydrated compounds very similar to the reference material. The samples with 5% biochar also presented a clear and dense microstructure very similar to the reference material, with visibly hydrated compounds such as portlandite spread throughout the structure (**Fig. 5c).** For the highest amount of biochar addition (10%), it was possible to observe biochar grains added to the cement paste structure (**Fig. 5d**). In addition, that the structure of the composite with 10% biochar proved to be well developed when compared to the low bond development structure of 10% raw sludge (Gomes et al., 2020).

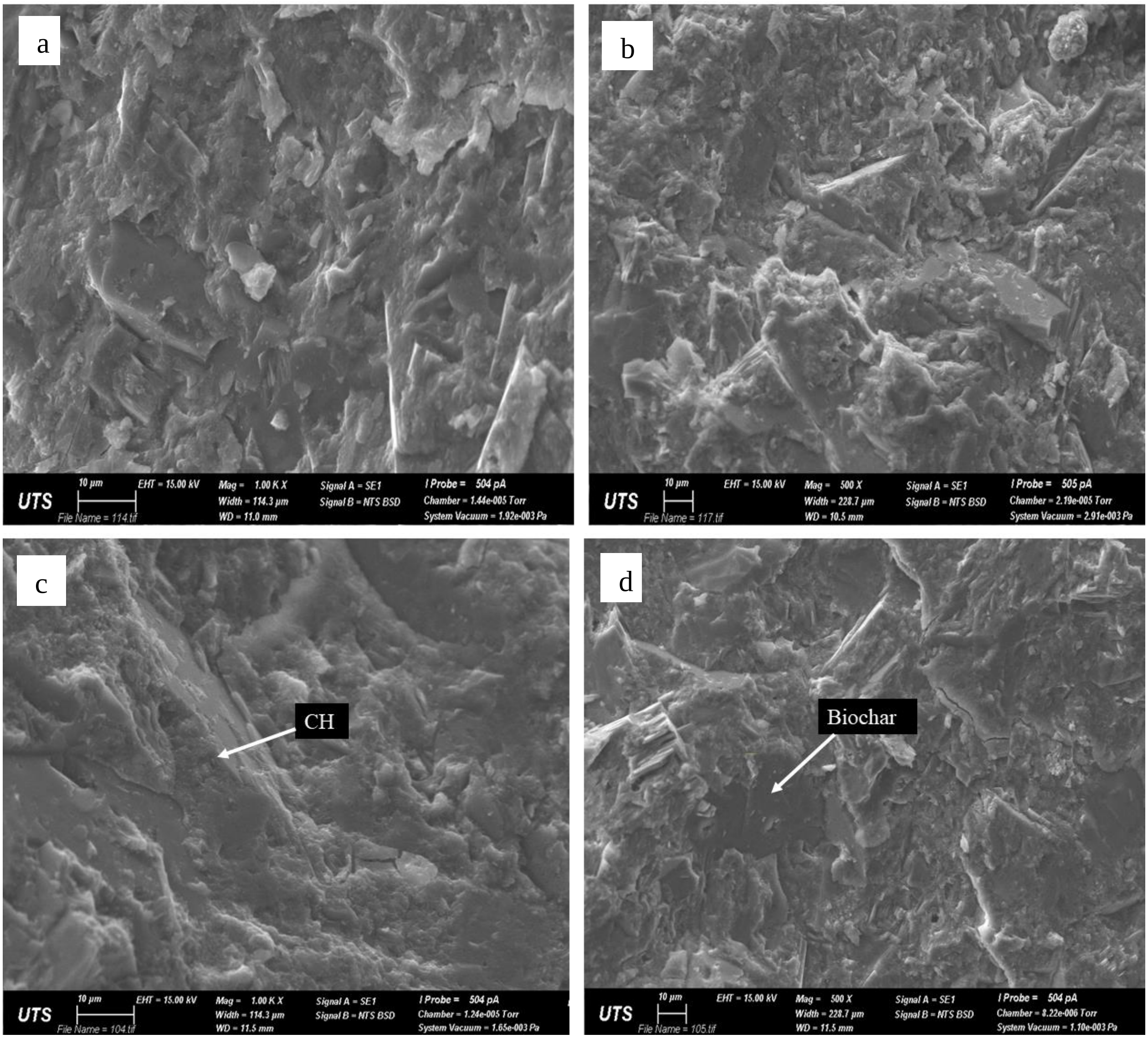
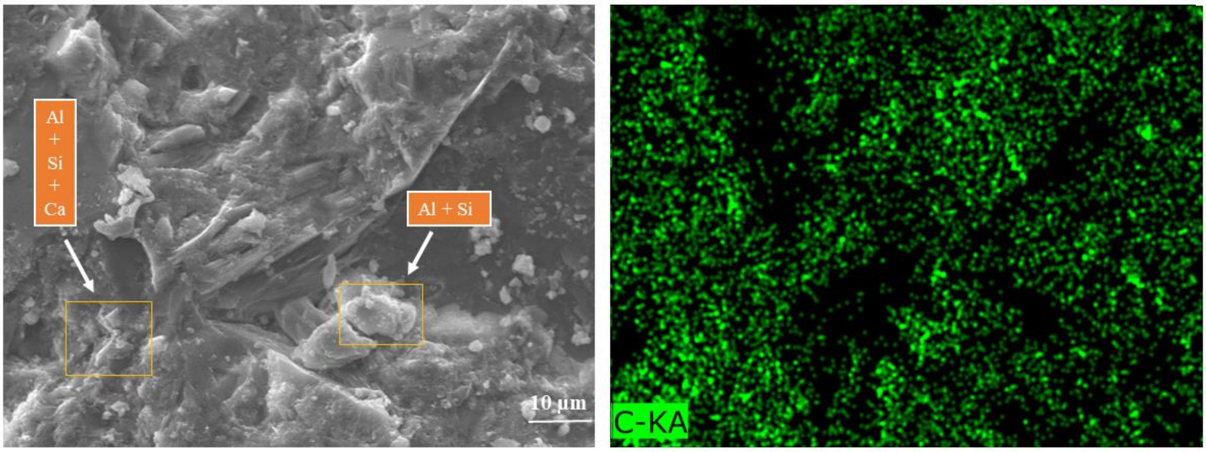


Figure 5. SEM analysis of hydrated reference cement paste with (a) 1% sludge biochar, (b) 2% sludge biochar, (c) 5% sludge biochar, and (d) 10% sludge biochar.

In order to visualize the element distribution surface of the hardened paste with biochar, elemental mapping using SEM-EDS spectroscopy was carried out. **Figure 6** shows the distribution of the elements for the cement composite samples with 5% biochar at 28 days. The predominant presence of calcium (Ca), oxygen (O) and silica (Si) can be clearly observed, confirming the main cement hydrated compounds of portlandite ($Ca(OH)_2$) and C-S-H. Another clear observation is the association of aluminium with other elements such as silica, indicating the presence of aluminosilicate particles such as metakaolin. The presence of this pozzolanic material in combination with calcium could lead to the formation of other hydrated products such as C-A-S-H (common in concretes with the addition of metakaolin) visible in the spectrum (Avet et al., 2019)



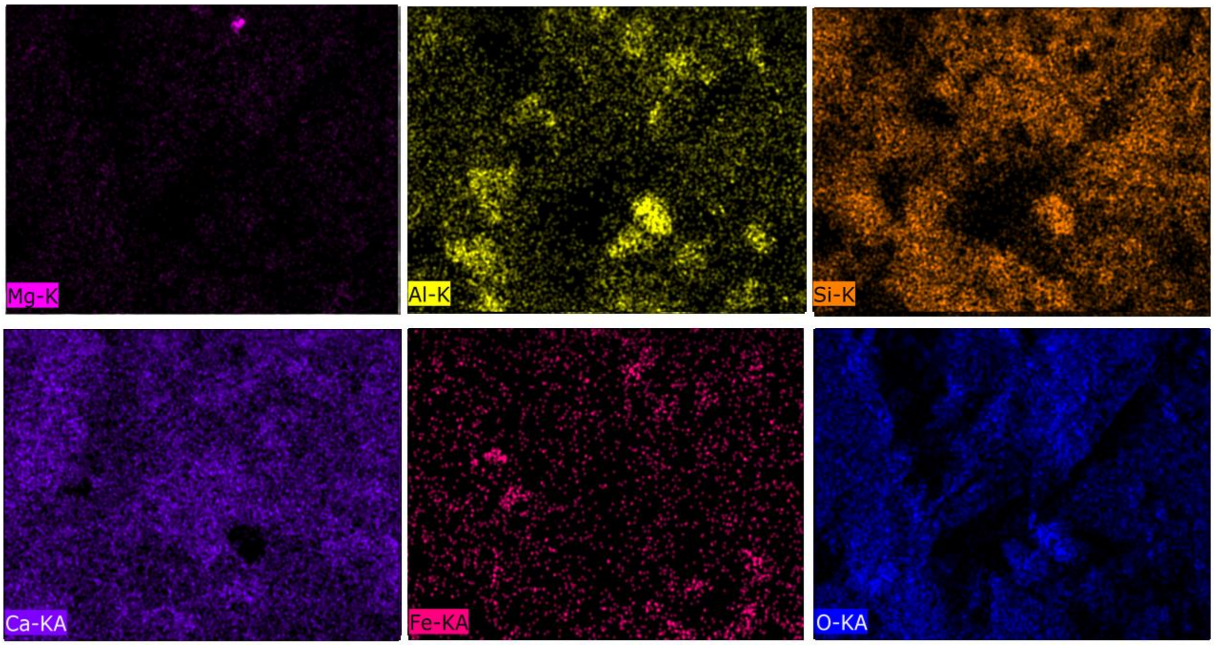


Figure 6. SEM-EDS image of the cement paste with 5% of sludge biochar, and its element distribution mapping.

3.7. Biochar production cost

The production of biochar involves several steps such as sample collection (i.e. dewatered sewage sludge), sample preparation (e.g. drying of sewage sludge), pyrolysis, and storage prior to application as a construction material. In addition, transportation is needed to move sludge to the pyrolysis plant, and to move biochar to a construction site. Each of these steps may need capital expenditure (i.e. equipment purchase, depreciation, and insurance), and operating cost such as fuel and labor. The approach by Nematian et al. (2021) was used to estimate itemized cost, on the basis of a mobile pyrolysis unit producing one ton/day of biochar. The calculations are shown in Table 2.

Table 2. Cost of biochar production with one ton/day capacity.

	Unit cost (US$)	**Cost/ton biochar (US$)**
Fixed cost		
Truck	62775	46.84

Trailer and fabrication	30000	25.21
Mobile pyrolysis unit	100000	81.09
Storage shed	20000	13.16
Portable toilet	1270	0.84
Portable septic tank	500	0.33
Fees, permits, and other payments		16.46
Sub total 1		183.93
Variable cost		
Fuel for truck		1.47
Labor		
Pre-processing		42.61
Operations and transportation		127.84
Miscellaneous		
Biochar bags		23.42
Waste disposal		23.42
Sub total 2		195.34
Administrative cost		18.96
Total cost		**398.23**

Based on the calculations shown in Table 2, the overall cost of biochar production from sewage sludge will be approximately US$398.22/ton. The cost is within the range reported for global sales price, from US$90/ton in the Philippines to US$8850/ton in the UK, although the average price is US$2650/ton (Ahmed et al., 2016). Similarly, Nematian et al. (2021) suggested that biochar cost ranged between US$571 and US$1455/ton, by converting orchard waste to biochar.

Although biochar production from sewage sludge adds some cost, both economic and environmental benefits can be made. If the biochar produced is sold on the market, the cost is likely to be recovered. In addition, biochar production will avoid sending sewage sludge to landfill, which will incur landfill tax (e.g. AU$147.10/ton in NSW, Australia, £96.70/ton in the

UK). The use of sewage sludge-derived biochar as raw materials in construction will undoubtedly bring environmental benefits, and reduce the use of precious natural materials.

4. Conclusions

The application of water treatment sludge-derived biochar in cement paste has been tested comprehensively. Water treatment sludge showed a heterogeneous morphology, with various sizes of angular particles and a semi-crystalline/amorphous structure. In comparison, XRD and SEM-EDS analyses of sludge biochar revealed a product with well-defined crystalline phases and pozzolanic material such as methakaline. The thermal process applied to the original sludge has improved hydration heat properties of the cement composites compared to the raw sludge. The composite samples with 2% and 5% of biochar release more heat than the reference samples. After 28 days of curing, the addition of 1-5% biochar in the composite produced a slightly higher compressive strength than the reference material, even 10% biochar addition showed similar compressive strength to the reference material. The results from TG-DSC and SEM-EDS analyses indicate that the reduction of portlandite in the 5% and 10% biochar specimens due to the pozzolanic reaction with biochar. The production of biochar from sewage sludge will be an expenditure, which is reasonable considering the benefits of protecting soil environment and replacing precious natural raw materials in construction industry.

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