Mine tailings and bottom ash from waste incineration as alternative fine aggregates for controlled low strength materials

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Abstract

Solid waste generation and its sustainable disposal are big concerns in the present decade. Mine tailings and incinerated bottom ash are one of the greatest solid waste contributors to the environment. To promote sustainable development, material reuse and recycling are important. In the present study, a feasibility study was conducted by utilizing such tailings and bottom ash for the production of controlled low-strength material (CLSM). The amount of cement was utilized less and by controlling the design mix with a targeted flow, low-strength material was produced. Three different cement content (60, 80 and 100 kg/m^3) and 100% mine tailings or incinerated bottom ash were used as fine aggregate material. Fresh properties of different CLSM mixes, mechanical performance, and leaching behaviour were conducted to confirm the final product. It was observed that both the mine tailings and bottom ashes are suitable as an aggregate material for CLSM mixes. The compressive strength values of different CLSM mixes at 28 days was observed between 0.13 MPa to 1.88 MPa for various aggregate materials (mine tailings and bottom ash) and cement content. Moreover, there is an influence in the reactivity of cement hydration by the aggregate (tailings and ash); which was confirmed by the calorimetry study. The selected mine tailings and bottom ashes are not inert (like natural aggregate) within the CLSM mixes and the chemical composition of the raw materials affects their fresh and hardened properties. The leaching test further shows that the final product could leach different heavy metals beyond the limit for inert materials, but within the nonhazardous limits by international standards. The present work deals with CLSM mixes with locally available industrial side streams; some important properties like hardening time, penetration resistance, shrinkage, excavatability, and other durability properties (i.e., acid attack, freezing and thawing resistance) were not considered in the study and can be studied further.

Keywords: Controlled low-strength material; mine tailings; bottom ash; fine aggregate; waste recycling.

1 Introduction

Reduce, Reuse, and Recycle are the 3R of sustainability. The prime goal is to conserve natural resources and maximization of reuse or recycling of waste materials. Similarly, for the construction industry, it is a big challenge to adopt the sustainability concept. Although there is research to promote less carbon emission materials by adopting comparatively sustainable binders, aggregates also have a tremendous influence on the sustainability of the concrete industry (Alexander and Mindess 2005; Mehta and Monteiro 2015; Mistri et al. 2019; Priyadharshini et al. 2018; Xiao 2018). The construction industry has a massive dependency on the availability of suitable aggregate. In the present decade, the demand for construction aggregate is huge (Mistri et al. 2020). It was reported that the demand will reach around 47.5 billion metric tonnes by 2023 (The Freedonia Group 2019). Therefore, there should be a sustainable supply chain of aggregate materials (both coarse and fine) for smooth operation in the construction industry.

Some construction materials that are used primarily as backfill are known as controlled low-strength materials (CLSM) (ACI Committee 229 2013; Katz and Kovler 2004; Ling et al. 2018; Nataraja and Nalanda 2008). CLSM is self-consolidated cementitious material with a maximum and minimum compressive strength of 8.3 MPa and 0.7 MPa at 28 days, respectively (ACI Committee 229 2013). Table 1 shows some of the features and specifications of CLSM. Moreover, CLSM poses several advantages such as self-compacted material (so no vibration is required), possibility for future excavation, economical, low cement requirement, etc (Du et al. 2002; Kaliyavaradhan et al. 2022; Ling et al. 2018). Along with these advantages, there are wide ranges of applications of CLSM: backfill material, anticorrosion filling, low permeability filling, pavement base, foundation support, waste stabilization, etc (ACI Committee 229 2013). There is also flexibility in the selection of raw materials for CLSM mixes (ACI Committee 229 2013; Ibrahim et al. 2022). Apart from conventional materials and natural resources, some other waste materials can be used for CLSM. Firstly, binder content can be replaced by blast furnace slag, fly ash or other supplementary cementitious material to make the design mixes more sustainable (Lachemi et al. 2010). Secondly, the natural aggregate material can be replaced by the utilization of waste materials from different industries or processes. For instance, mine tailings from the mining industry (Bouzalakos et al. 2013), bottom ash from municipal waste recycling facilities (Razak et al. 2009; Yan et al. 2014), recycled fine aggregate from construction and demolition waste recycling facility (Achtemichuk et al. 2009; Ali et al. 2022; Etxeberria et al. 2013), excavated soil as fine aggregate (Chittoori et al. 2014; Qian et al. 2019; Wang et al. 2022; Zhu et al. 2022), etc can be utilized as an aggregate material.

Mine tailings is a by-product of the mining industry and could be used as aggregate material in construction. Economic and industrial growth in many countries has created a huge demand for raw materials. It is expected that about 30% increase in demand for raw material in the mining industry can be seen by 2050 (International Energy Agency 2022; World Economic Forum 2015). With the rise in demand for different minerals, the proportional accumulation of mine tailings is being seen in the recent past. From the mining and quarrying industry in Finland, approximately 86.7 million tonnes of waste were generated in 2019 (Statistics Finland 2021a; b). Accumulation of tailings and related environmental pollution is one of the major concerns in the mining sector. For any mining company, tailings is a big problem as it needs suitable disposal solution. It is considered as a zero (or negative) value material and land filled or stored in tailings storage facility that add to the cost of the mine operations. Moreover, the mining industry needs to have a sustainable and effective solution to handle this issue. There is very limited research on the utilization of mine tailings in CLSM (Kim et al. 2016). Strength performance along with a leaching test was performed to confirm the applicability of mine tailings as aggregate in CLSM. Study with arsenic rich mine tailings reported that with the increase in cement content (10 to 20% of mine tailings), the strength of CLSM increases (1.2 MPa to 4.1 MPa at 28 days) (Kim et al. 2016). In another study, iron ore tailings were used for the production of CLSM, and it was observed that 100% replacement of soft soil using iron ore tailings showed lower water demand with higher mechanical performance (Li et al. 2023). Moreover, a more compact microstructure with low porosity was observed when soft soil was replaced by iron ore tailings completely to produce CLSM. As mine tailings may have different chemical compositions based on their source (or type of mine), further research is needed to confirm the recyclability of different mine tailings as an aggregate material.

Similarly, incinerated bottom ash can be another potential material that may replace natural fine aggregate. The disposal of municipal solid waste (MSW) incinerated bottom ash is one of the big challenges in the recent past. In a statistic from the USA, it was mentioned that about 34% of the MSW is treated to produce bottom ash by waste-to-energy transformation (Psomopoulos et al. 2009). Approximately 23-30% of the incinerated waste converts into bottom ash in Finland (LHJ Group 2022). The average annual rate of bottom ash production is about 300,000 tonnes in Finland. The most common method for the disposal of this huge quantity of incinerated bottom ash is landfilling, which is not a sustainable approach (Bertolini et al. 2004). Although the physical properties and chemical composition of bottom ash may be similar to the natural fine aggregates, bottom ash can have some heavy metals (Bertolini et al. 2004; Forteza et al. 2004; Liu et al. 2022). It was reported that the water absorption of incinerated bottom ash (11.5%) was similar to fine recycled concrete aggregate (10.8%) with almost the same particle size (<5 mm) (Ali et al. 2022). The specific gravity of incinerated bottom ash was reported 1.83 and compacted bulk density 964 kg/m^3 (Naganathan et al. 2012; Razak et al. 2009). Moreover, the bottom ash may have a huge variation in properties in different batches based on their initial source MSW characteristics (Ali et al. 2022). According to the leaching test results by Razak et al., (Razak et al. 2009) the CLSM with incinerated bottom ash was suitable for landfill disposal. Research data shows that bottom ash may be used for CLSM production (Razak et al. 2009; Wu et al. 2016; Yan et al. 2014). It was also concluded by Ali et al. (Ali et al. 2022) that there is a potential for using incinerated bottom ash to produce sustainable CLSM. Moreover, the leaching test results confirmed that the bottom ash that was used was not a hazardous material. When bottom ash was used for CLSM material, the overall leaching of heavy metals was less compared to the raw incinerated bottom ash because of the cementing binder system.

There are many challenges while utilising such industrial side stream (mine tailings and incinerated bottom ash) materials in CLSM (Ling et al. 2018). The cement content over the total solid content plays a significant role in order to bind all the particles within the matrix. Therefore, the amount of cement content is the key to regulating the strength performance of CLSM by having an option for possible future excavation (ACI Committee 229 2013; Wu et al. 2016). The physical properties like particle size (also overall gradation) and water absorption influence the flow properties of CLSM mixes. Therefore, it is a trial process to make the CLSM mixes that have the required flow (normally high) with low strength (maximum strength at 28 days 8.3MPa) (ACI Committee 229 2013). Finally, the concentration (leaching) of heavy metals from the CLSM should be within the permissible limit by the international standard (or the regulatory authority) if such industrial side stream is used in construction (Kim et al. 2016; Yan et al. 2014). Therefore, more research is needed in this area to confirm the maximum utilization of such industrial side stream in the construction industry by maintaining a sustainable environment. The durability performance of CLSM was lower than conventional concrete (Parhi et al. 2023; Wang et al. 2023). It was reported that the compressive strength of CLSM was significantly lower after freeze and thaw cycles. Moreover, the freeze and thaw durability performance of CLSM depends on the cement-aggregate ratio and binder content; the more binder content shows better resistance to freeze and thaw cycle. It is prudent to mention that the resistance to the freeze and thaw cycle of CLSM is related to compressive strength (Achtemichuk et al. 2009). Moreover, the higher compressive strength showed lower mass loss after freeze and thaw cycle as reported by Achtemichuk et al. (Achtemichuk et al. 2009).

The construction industry is facing challenges related to the scarcity of river sand. Moreover, transportation of river sand from a long distance sometimes becomes uneconomical. According to the United Nations Environment Programme (UNEP 2014), the river sand is not only for construction but also has an important role in the environment to maintain some ecological aspects (like erosion, biodiversity, water supply, food production, fisheries, etc.). Thus, there should be an alternative for river sand in future construction projects. Recently, mine tailings and bottom ash is seen as potential alternatives for this purpose. One of the main problems that hinder the recycling of mine tailings and bottom ash is the presence of heavy metals. However, immobilization of heavy metals from hazardous tailings and sustainable disposal solutions are important aspects and has become an emerging research topic of interest. Moreover, the development of guidelines to promote sustainable waste management (e.g., mine tailings and bottom ash) needs scrupulous research. However, there is ongoing research on their potential in the construction sector as a fine aggregate material for high-volume applications. The present study aims to find an effective upcycling solution for mine tailings and bottom ash as construction materials. It was found that mine tailings and bottom ash can be used as fine aggregate to produce CLSM by supporting a sustainable disposal solution. Moreover, the leaching test results confirmed that the CLSM with both the mine tailings and bottom ash was not a hazardous material.

2 Materials and Methods

The research work presented in the paper follows four steps to assess the applicability of different mine tailings and bottom ashes as fine aggregates for CLSM. Firstly, the materials were characterized to understand their possible performance and design mixes are set accordingly. Moreover, the recommendation of ASTM was followed to choose the constituents of the CLSM mixes. Secondly, the influence of different fine aggregate materials on the fresh and hardened properties of CLSM is assessed. Complete (100%) replacement of natural fine aggregate was considered for different mixes. Thirdly, the new aggregate from the mining industry and MSW recycling facility may not be inert. Therefore, an investigation was made to assess the reaction between the aggregate (mine tailings and bottom ash) and cement using a calorimetry study. Fourthly, a leaching test was performed to confirm the final product is safe as per the environmental compliances.

2.1 Characterisation of materials

In this study, four materials were used as fine aggregates for CLSM specimen. Two bottom ash and two mine tailings were used. The first bottom ash (BA1) was collected from a municipal solid waste recycling facility in Sweden and the second bottom ash (BA2) was collected from Finland. Similarly, the first mine tailings (MT3) was collected from the Ni-Cu mine (Boliden, Sweden). The second mine tailings (MT4) was collected from the quartz mine (Sibelco, Finland) (Perumal et al. 2020; Ramanathan et al. 2021). Portland Cement (CEM I 52.5R) manufactured by Finnsementti (Finland) was used as a binder material. No supplementary cementitious material was used to replace cement. The oxide composition of the materials was analysed by X-ray fluorescence spectroscopy. The chemical composition of all the materials is given in Table 2. The materials are mainly composed of alumina, silica, calcium oxide, and iron oxide. However, for MT3, the concentration of magnesium oxide (20.2%) is significantly higher. The loss on ignition (LOI) for most of the materials is less than 5%. However, BA1 has a higher LOI (10.8%). According to ASTM standards, the LOI of the aggregate materials should be less than 5%. Therefore, most of the materials can be considered aggregate except BA1 (ASTM C33 2018). However, there could be some flexible rules for such utilization of waste material in future. Although the aggregate material is normally considered an inert construction material, however, for fine aggregate there is a possibility to react some of the components with cement.

The particle sizes were 0 to 2 mm for all the aggregate materials. Mine tailings are normally fine particles from the mining industry, and it is generated during the processing of ore. Figure 1 shows the grading curve of four different aggregate materials. For comparison purposes, a grading curve of standard sand (confirming to EN 196-1) was included. The bottom ash (BA1 and BA2) and mine tailings (MT3 and MT4) are significantly finer than standard sand. BA1 and BA2 have almost similar grading curves. The possible reason behind this could be similarity in the municipal waste incineration process in Finland and Sweden. Among all the materials, MT3 is the finest one.

The specific gravity of BA1, and BA2 were 2.28 and 2.48, respectively (ref. Fig. 2). In contrast, the specific gravity of mine tailings was 3.16 (MT3) and 2.86 (MT4) which is significantly higher as compared to bottom ashes (BA1 or BA2). The specific gravity of the standard sand (2.60) lies in a mid-range between these two materials. Figure 3 shows the optical microscopic image of bottom ash and mine tailings particles. From the figure, it can be observed that the bottom ash is more heterogeneous compared to mine tailings.

2.2 Mix design for CLSM

Different mix compositions were studied with different types and amounts of fine aggregates (Table 3). It can be seen that the fine aggregate varies for different mixes. Considering C60BA1, the amount of cement, fine aggregate (BA1) and water is 60 kg/m^3, 1187 kg/m^3 and 430 kg/m^3, respectively. Similarly, for C80 and C100 series, the cement content is 80 and 100 kg/m^3, respectively. In addition to this, an air-entraining admixture was used for better flowability, reduction in bleeding and segregation (Mehta and Monteiro 2015). It is prudent to mention that Pujadas et al. (Pujadas et al. 2015) reported a systematic approach to calculate an efficient CLSM mix with maximum packing of the fine aggregates. Different trial on aggregate mixing was conducted to achieve maximum packing density. However, the present study deals with the raw materials obtained from different industries (such as mine) and MSW recycling facilities. The prime reason behind this is to deal with the real scenario and minimizing of any additional process that involves embodied energy.

For the mix proportion, ACI 229R-13 (ACI Committee 229 2013) was followed. According to the standard, the cement content could be in the range of 15-119 kg/m^3. Moreover, the content of fine aggregate is between 1483-2076 kg/m^3. There is a flexible rule for the mix proportion of CLSM. The main important consideration is the flow of the material and strength. CLSM has high flow (200-300 mm as per ASTM D6103) and low strength (maximum strength at 28 days is 8.3MPa) requirements. For the present study, three different cement content varying from 60, 80, and 100 kg/m^3 was considered for the mix design. The minimum flow requirement was kept at 220 mm. A basic trial was conducted on the mixing of material to investigate the flowability of the mix and the mix design was adjusted accordingly to achieve the final mix proportion. Air entraining admixture (SikaAir PRO 5 (FI)) was used for all the mixes. Based on the manufacturer information (and material safety data), the admixture produces a fine-grained system of very small air pores in the mixes. Based on the trials in the laboratory, the final mix proportion for CLSM in the study is given in Table 3. All the predefined materials were mixed by following a similar method. Firstly, all the fine aggregates and cement were mixed well for 1 min. Secondly, the admixture was mixed with water and applied to the mixer. After one minute mixing period, the mixture was stopped for 30 sec. Thirdly, all the materials were again mixed for 3 minutes and ready for casting specimens.

2.3 Flow table test

For CLSM, flow is an important parameter along with strength. As most of the time the CLSM is used as backfilling materials, good or high flow is a demand. In this study, the flow table was used to measure the flowability of the fresh mix. The ASTM standard D6103/D6103M (ASTM D6103/D6103M 2017) deals with the flow consistency of CLSM, and the flow cylinder is suggested. Moreover, the procedure was for a maximum aggregate size of 19 mm. However, for the present study, the maximum particle size of the aggregate is 2 mm. Therefore, a flow table test was conducted for the CLSM in this study as per ASTM C230/C230M (ASTM C230/C230M 2020). Four measurements were noted for each material on the flow table and an average value is reported.

2.4 Hardened properties of CLSM

For CLSM, along with the flow, strength measurements are one of the key assessments. As mentioned earlier, the requirement or target strength of CLSM is low (maximum 8.7 MPa at 28 days). Cube specimens with a maximum dimension of 50 mm were considered for both the density and strength measurements (ACI Committee 229 2013). Samples were prepared as per the given mixes and kept in a humidity room for 24h. The CLSM samples were demolded after 7 days of production and then kept in 90 ± 5% moisture condition at 25^{o}C. At 7, and 28 days the cubes are tested for strength performance. Figure 4 shows the compression test setup with CLSM specimen. A minimum of three specimens were prepared for both the 7th and 28th-day testing. The density of all CLSM was measured with a different set of specimens with the same mix and curing condition.

2.5 Change in hydration kinetics

Both the coarse and fine aggregates are generally considered inert materials (Alexander and Mindess 2005; Mehta and Monteiro 2015). However, for the present study, a different type of fine aggregate is used: bottom ash and mine tailings (0 to 2 mm). The changes in hydration kinetics because of the new type of aggregate materials were studied by isothermal calorimetry. The mix design as given in Table 3 was considered for the calorimetry study. A total of 50 g material mix was considered, and the mixing ratio was as per Table 3. In this context, the chemical admixture was not considered for the present investigation and only the C80 series (cement content 80 kg/m^3) was adopted for the calorimetry study. All the material (cement, aggregate, and water) was mixed by low-speed hand mixing. Approximately, 9 g material of the mortar mix was placed inside a plastic ampoule and then sealed with a cover. It is prudent to mention that the mixing operation and the precondition of the isothermal calorimetry were at 23 ºC. The heat release from the mix was measured for up to 7 days.

2.6 Leaching test

The mine tailings and bottom ash may contain several heavy metals with different concentrations. Therefore, in the present study, a leaching test was conducted as per EN 12457 (CEN - EN 12457 2002) for both the material level and CLSM samples after 28 days of curing. For CLSM mixes C80 series was chosen for the leaching test and the test results were recorded. According to the Finnish Ministry of the Environment (Ministry of the Environment 2016), waste should be characterized properly to assess a batch composition of a material, leaching behaviour, and pH. Moreover, the document also reports the criteria for the acceptable limits of different inert, nonhazardous, and hazardous elements within landfill waste (ref. Supplementary file Table S1).

3 Results and discussion

3.1 Flowability

The workability of CLSM was determined by following the ASTM C1437 standard (ASTM C 1437 2001). According to the standard, the increase in flow compared to the initial base diameter of the mortar mass is represented in a percentage. Figure 5 shows the flow table test observation of CLSM mixes. Figure 6 shows the flow results for all the CLSM mixes as given in Table 3. All the CSLM mixes had high flow (more than 100 %). Moreover, the MT3 shows comparatively higher flow (overflowed from the flow table with a diameter of 300 mm). This can be justified by using the particle size distribution curve (ref. Fig. 1). Although the mine tailings (MT3) have a finer fraction compared to the other aggregates (MT4), the flow is comparatively higher than other mixes. The primary reason is linked to the chemical composition of the material. MT3 has a high content of MgO and based on the literature (Aiken et al. 2022; Polat et al. 2017), high content of MgO increases the flow of the cement paste and decreases the rate of reaction. Furthermore, the influence of MgO was confirmed by another study in the upcoming section of the paper. From the overall flow table test results, it can be concluded that the fine aggregate particles are also equally suitable for CLSM with high workability. The influence of admixture was similar for all the material mixes as it varies (fixed 3% wt. of cement) with the cement content (60-100 kg/m^3). The doses of admixture were fixed based on laboratory trials of the mixes. Initially, the mixes were prepared without admixture and then 1, 2 and 3% admixture doses were tried to achieve the flow of 250 mm (flow table test). Finally, 3% was the most suitable one and hereby suggested in this study.

3.2 Density and strength performance

The density of the specimen with mine tailings is comparatively higher than the specimen with bottom ash (Fig. 7). The mixes with MT3 show the highest density (approximately 2300 g/cm^3) which is because of the specific gravity of the material (3.16). In contrast, the CLSM mixes with bottom ash show lower density. Therefore, it can be said that based on the fine aggregate material properties, the density of the hardened specimen may vary. From the present study, the range of density value of different mixes is 1240-2390 g/cm^3. The relation between the curing age and density is marginal. However, during the measurement of the dimension of each specimen, it was noticed that the height of the specimen is lower (by 2±1 mm) as compared to the other dimensions (length or width). Even, for MT3 series the reduction in height (and corresponding volume) was noticeable. The initial dimension of the fresh specimen was 50×50×50 mm^3, however, after 24h the height was measured as 46-47 mm. The primary reason behind this is the high-water content within the mix that results in change in volume with time. Therefore, the CLSM has high shrinkage (ACI Committee 229 2013; Das 2021) in the vertical direction (gravitational force direction) of the application. Study by Kim et al. (Kim et al. 2016) reported that there is an initial settlement (0 to 0.38 mm/mm) after the casting of CLSM over time when arsenic rich mine tailing was used for CLSM. It was reported that first 3-4 h is the time for initial settlement and this issue can be minimized by using higher cement content in the CLSM mix. This consideration should be kept in mind when similar material will be used for practical application. Moreover, the CLSM is sometimes considered for re-excavation material (ACI Committee 229 2013; Devaraj et al. 2022).

The strength performance of all the CLSM mixes was not the same (ref. Fig. 8). Although the flow and density were comparable within the mixes, however, the compressive strength was different. In the present study, the density of different CLSM mixes and corresponding strength has no direct correlation. Similar observation was made by Razak et al. (Razak et al. 2009). The requirement of the compressive strength at 28 days is 0.7 MPa as per the standard (ACI Committee 229 2013; ASTM D4832 2016). Overall, with the increase in cement content of CLSM mixes, strength increases. Similar common observations were reported in literature (ACI Committee 229 2013; Kim et al. 2016; Liu et al. 2022). The C80 series mixes with BA1, BA2, and MT3 is applicable as the strength at 28 days is higher than 0.7 MPa. Similarly, the C100 series for BA1, BA2, and MT3 can be used in practice. The highest strength of the MT4 series is for C80MT4 and that is 0.42 MPa at 28 days and which can be used with an aim for future excavation (ACI Committee 229 2013). It can be observed that the particle size of MT4 is almost similar to BA1. Therefore, the reason behind the reduction in strength is not the particle size. Interestingly, the reason behind the decrease in compressive strength of MT4 mixes could be the presence of high alumina within the aggregate (ref. Table 2). MT4 has high alumina content (23.43%) and is the highest among the other aggregate material considered in the study. Further research can be done on this aspect to confirm the same effect.

3.3 Effect of bottom ash and mine tailings on cement hydration

The bottom ash influences the initial reactivity of binder material (ref. Fig. 9). Although aggregates are considered inert material (Mehta and Monteiro 2015; Mistri et al. 2019), in the present study the initial reaction for both the bottom ash (BA1 and BA2) is different than other materials (like MT). The cumulated heat and the heat flow were normalized with mixing weight and the cement content was the same for all the material (80 kg/m^3). Based on the isothermal calorimetry results, it is clear that the bottom ashes (BA1 and BA2) are comparatively reactive in the initial stage just after mixing with water. Therefore, the addition of bottom ash in CLSM accelerated the cement hydration. Moreover, the higher heat of hydration is seen at the second peak. In contrast, the mine tailings has very less significance in cement hydration. In mine tailings, MT4 has more influence on cement hydration compared to MT3. The mine tailings are finer than the bottom ashes and the number of total hydration (reaction) zone should be higher for the finer particle mixes. However, in the present study, it can be observed that the fine aggregate is not inert.

Similarly, the total heat release (ref. Fig 10) at first 24 h was significantly higher for bottom ash aggregates as compared to the mine tailings or even the natural aggregate (standard sand). Therefore, the CLSM mixes with the bottom ash indicating higher reactivity. The MT4 has a different heat release curve than MT3. Although the initial heat release for different CLSM mixes does not show an exact correlation with the strength development, the fine aggregates have participated in the overall strength development. Ideally, the aggregate should be inert when it is used in concrete or other application. Therefore, before the adoption in practice of the CLSM with bottom ash or mine tailings, the influence of aggregate in cement hydration must be taken into consideration.

3.4 Heavy metal leaching and pH

The leaching test results for both the material level and CLSM mixed after the curing period was obtained and compared with the limiting value given for the inert material as per the standard (Ministry of the Environment 2016). After comparing the test results (ref. Table 4) with the limiting values (also ref. Table S1), it can be seen that in the CLSM samples, the concentration of heavy metals has decreased. However, the values are beyond the limiting value for an inert material, but within the limit for non-hazardous materials. For instance, the limiting value for chromium is 0.50 mg/kg to be within the inert category. However, from the test results, BA2, C80BA1, and C80BA2 show higher chromium concentration than the limiting value. It is prudent to mention that the additional chromium is from cement. In contrast, the concentration of SO_3 is reduced after the waste materials (BA and MT) were used for the production of CLSM. Again, for the Cl element, at the composite level, the concentration has reduced significantly as compared to the material level. Overall, cement hydration may isolate some of the heavy metals with a certain concentration. However, complete isolation is not possible under the present condition as mentioned in the study. Therefore, before the application of such waste materials as fine aggregate for the production of CLSM, proper approval from the concerned authority is needed. In this context, Zhen et al. reported from the leaching test that the utilization of bottom ash (from Shanghai, China) in CLSM shows negligible health and environmental risks (Zhen et al. 2013). A similar observation was made by Razak et al. (Razak et al. 2009) and the materials was from Malaysia. Therefore, the test results for heavy metal concentration may vary based on the geographical locations, characterisation of waste, method of collection and processing, etc.

3.5 X-Ray Diffraction

The present study deals with very low cement content (around 5%) compared to total aggregate materials (cement to aggregate ratio is approximately 1:15). The XRD patterns of the raw materials (or aggregates, BA1, BA2, MT3, MT4) and at the composite level are almost similar (ref. Fig. 11). The main influence in the test results is because of the high content of different aggregate materials (BT or MT). Although the CLSM has isolated some of the heavy metals or other elements within the hydrated product (ref. Table 4), a marginal difference in the peaks can be seen. Some of the peaks at the raw materials level have been reduced. Otherwise, there is no such major difference in peaks. The only minor difference in peak is with the quarts (SiO_2) which could be due to the pozzolanic reaction of ash (BA) with calcium hydroxide. In the present study, the bottom ash and mine tailings were used as fine aggregate, not as cement substitute (or supplementary cementitious material (SCM). Moreover, at 28 days it is expected that within 5% cement (or 80 kg/m^3), the whole amount has not hydrated. Therefore, within this less cement hydration product, the differentiation in the hydrated product is difficult.

3.6 Microstructure using SEM

The SEM micrographs (Fig. 12) show that the microstructure is different within different mixes. In C80BA1, regular microstructure with the less cement-hydrated product can be seen. The lower hydrated product is obviously because of the mix design for CLSM (approximately 5% cement). The needle-like fibre component should be ettringite (AFt) and the sizes are within 5 microns (Zhen et al. 2013). In Fig. 12(b), C80BA2 shows a similar microstructure with a comparatively more cement-hydrated product than C80BA1. The corresponding improvement in compressive strength can be seen (ref. Fig. 8). The EDS mapping micrographs from the present study can be found in the supplementary file (Fig. S1 and S2). In C80MT3, the content of the Mg element is seen (confirmed by EDS elemental mapping). In this context, Kim et al., (Kim et al. 2016) studied different elemental map of mine tailings using EDS which shows the overall distribution of elements at materials level. Although the content of MgO influences the fresh properties of CLSM (ref. Fig. 6), the strength performance was within the range provided by the standard (ACI Committee 229 2013). Similarly, for the C80MT4 high alumina can be seen from the microstructure with EDS mapping (ref. Fig. S2). It is expected that the high content of alumina that is distributed is the main reason behind the lower strength performance.

4 Conclusion

This paper presents the research outcomes from the characterisation and utilization of mine tailings and bottom ash as fine aggregate for the production of CLSM with an aim for maximum utilization of such waste for a sustainable environment. Based on the information presented in this study following major conclusions are outlined:

- The materials characterisation results showed that bottom ash (BA2) and mine tailings (MT3, and MT4) can be used as fine aggregate material for the production of CLSM. Considering the flow and strength requirements together, CLSM mixes with BA1, BA2, and MT3 was suggested which has a minimum compressive strength requirement of 0.7 MPa at 28 days. Moreover, the cement content of 80 kg/m^3 was found adequate which qualifies both the strength and low cement content benefits. CLSM mix with MT4 (C80MT4) showed highest compressive strength 0.42 MPa at 28 days and which can be used with an aim for future excavation as per ACI Committee 229.
- Mine tailings and bottom ash can give a high flow for CLSM with a proper design mix. However, the materials (mine tailings and bottom ash) may have an influence on the flow properties. MT3 has a high content of MgO which resulted in a high flow of the mix. Therefore, the MT3 used in the study was not truly inert material as natural river sand. Similarly, high alumina content showed negative influence on the strength performance of the CLSM with MT4.
- Although the mine tailings and bottom ash were used as fine aggregate, the materials (BA1, BA2, MT3, and MT4) showed influence on cement hydration based on their chemical composition. From the initial calorimetry data, the heat release curve was different for various CLSM mixes which confirms that bottom ash (BA1, BA2) and mine tailings (MT3, MT4) are not completely inert.
- The leaching test confirmed that both the bottom ash and mine tailings are within the limit for non-hazardous material but beyond the inert limit. Therefore, proper approval from the governing authority may be needed to adopt these materials in construction.

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Data Availability Statement

Some or all data, models, or code that support the findings of this study are available from the corresponding author upon reasonable request.

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526 **Table 1.** Some aspects of the preparation of CLSM mixes

Subject/Criteria	Limits	Remarks	Reference
Cement	30 to 120 kg/m^3 according to ACI Committee 229	-Cement content may increase as per the increase in strength requirement.	(ACI Committee 229 2013)
Fly ash	0 to 415 kg/m^3 according to different studies as given in Table 5.2.2 (ACI Committee 229)	-Higher content of fly ash can be used to improve the strength and flow properties of CLSM -The use of high-volume fly (i.e., 360 kg/m^3) ash shows higher shrinkage of CLSM	(ACI Committee 229 2013)
Water	193 to 344 kg/m^3	-Water content should be higher in the case of finer particles	(ACI Committee 229 2013; Du et al. 2002)
Coarse aggregates	Normally, not used in CLSM		
Air-entraining admixtures	15-30% air content is also possible	-Improves workability -Reduces shrinkage, bleeding, segregation, and unit weights	(Du et al. 2002; Etxeberria et al. 2013)
Flowability	High flow (200-300 mm slump)	-Depends on water-cement (w/c) ratio and particle sizes (aggregates and binder) -Higher the w/c ratio, higher flowability -More finer particles lesser flowability	(ASTM D6103/D6103M 2017)
Unit weight	1840-2320 kg/m^3	-Lightweight aggregate can be used to make lower unit weight.	(ASTM D6103/D6103M 2017)
Compressive strength	At least 0.7 MPa at 28 days and maximum strength is 8.3 MPa at 28 days.	-May vary as per the application. -Well-compacted soil (0.3 to 0.7 MPa) -Structural filler (2 MPa and higher) -Pavement applications (bases, sub-bases, and subgrades) (at least 1.5 MPa)	(ACI Committee 229 2013; ASTM D4832 2016)

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Table 2. Chemical composition (weight %) of different materials in oxide form

Subject	BA1	BA2	MT3	MT4	Cement
Al_2O_3	9.73	11.25	2.91	23.43	1.98
SiO_2	31.35	39.42	48.35	42.83	23.28
CaO	17.09	12.07	13.95	14.00	66.40
Fe_2O_3	11.49	16.87	9.82	6.97	0.33
MgO	1.75	2.16	20.19	5.10	0.27
Na_2O	3.94	3.27	0.59	1.22	0.27
K_2O	1.11	1.30	0.25	0.24	0.03
P_2O_5	1.39	0.98	0.02	0.07	0.32
SO_3	2.59	1.72	0.50	0.06	2.03
TiO_2	1.23	2.11	0.34	0.46	0.03
C	0.98	2.58	0.07	2.38	-
LOI (at 950°C)	10.8	4.9	2.0	4.0	1.6

Table 3. Design mix of CLSM (materials in kg/m^3)

Mixture Name	Cement	BA1	BA2	MT1	MT4	Water	Admixture
C60BA1	60	1187	-	-	-	430	1.8
C80BA1	80	1173	-	-	-	430	2.4
C100BA1	100	1158	-	-	-	430	3
C60BA2	60	-	1289	-	-	430	1.8
C80BA2	80	-	1274	-	-	430	2.4
C100BA2	100	-	1258	-	-	430	3
C60MT1	60	-	-	1649	-	430	1.8
C80MT1	80	-	-	1628	-	430	2.4
C100MT1	100	-	-	1608	-	430	3
C60MT4	60				1491	430	1.8
C80MT4	80				1473	430	2.4
C100MT4	100				1455	430	3

Table 4 Leaching test results from different materials and at CLSM mixes

Subject	BA1	BA2	MT3	MT4	C80 BA1	C80 BA2	C80 MT3	C80 MT4	Limiting values (mg/kg of dry mass) (Ministry of the Environment Finland 2016)		
									Inert	Nonhazardous	Hazardous
L/S=10 pH	9.05	9.85	9.63	9.43	11.42	11.21	12.16	12.37	-	-	-
L/S=10 conductivity [mS/cm]	6.06	3.02	0.26	0.06	5.31	2.30	3.31	5.53	-	-	-
L/S=10 Al [mg/kg]	13	42	<1	13	66	98	5.1	6.8	-	-	-
L/S=10 As [mg/kg]	0.043	0.039	0.027	0.012	0.037	0.027	<0.01	<0.01	0.5	2	25
L/S=10 Ba [mg/kg]	0.32	0.45	0.13	<0.06	0.22	0.59	0.64	2.8	20	100	300
L/S=10 Cd [mg/kg]	<0.002	<0.002	<0.002	<0.002	<0.002	<0.002	<0.002	<0.002	0.04	1	5
L/S=10 Co [mg/kg]	<0.004	<0.004	<0.004	<0.004	0.0049	<0.004	<0.004	<0.004	-	-	-
L/S=10 Cr [mg/kg]	0.17	0.78	0.019	<0.01	0.87	0.58	0.12	0.25	0.5	10	70
L/S=10 Cu [mg/kg]	4.7	0.58	<0.01	<0.01	9.5	1.4	<0.01	<0.01	2	50	100
L/S=10 Fe [mg/kg]	<0.15	0.36	<0.15	<0.15	0.32	0.36	0.19	<0.1	-	-	-
L/S=10 Mo [mg/kg]	1	3.5	0.059	0.026	1.7	5.5	0.2	0.13	0.5	10	30
L/S=10 Ni [mg/kg]	0.12	0.014	<0.01	<0.01	0.08	0.016	0.012	0.017	0.4	10	40
L/S=10 Pb [mg/kg]	<0.004	<0.004	<0.004	<0.004	<0.004	<0.004	<0.004	<0.004	0.5	10	50
L/S=10 Sb [mg/kg]	0.38	0.73	0.014	0.01	0.43	0.42	<0.01	<0.01	0.06	0.70	5
L/S=10 Se [mg/kg]	<0.04	0.16	<0.04	<0.04	<0.04	0.12	<0.04	<0.04	0.10	0.50	7
L/S=10 Ti [mg/kg]	<0.15	<0.15	<0.15	<0.15	<0.15	<0.15	<0.15	<0.15	-	-	-
L/S=10 V [mg/kg]	<0.01	0.13	0.027	<0.01	0.45	0.5	<0.01	<0.01	-	-	-
L/S=10 Zn [mg/kg]	0.052	0.081	0.046	<0.04	0.084	0.099	0.047	0.11	4	50	200
L/S=10 F [mg/kg]	<5	12	<5	<5	5.4	10	10	7.8	10	150	500
L/S=10 Cl [mg/kg]	9000	3200	460	<25	8100	2600	360	<25	800	15000	25000
L/S=10 SO_4 [mg/kg]	20000	12000	57	72	11000	4800	250	69	1000	20000	50000
L/S=10 TDS [mg/kg]	47000	26000	3400	<2000	34000	15000	8800	14000	400	60000	100000