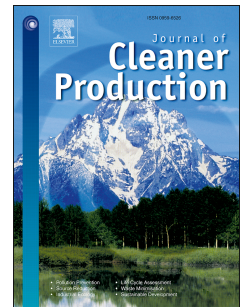


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Manufacture and performance of lightweight aggregate from waste drill cuttings

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Abstract

This research investigated the technical feasibility of transforming waste drill cuttings into lightweight aggregate. Drill cuttings produced from the North Sea oil field were dried, ball milled, formed into pellets and fired at temperatures between 1160 and 1190°C. Physical properties of the manufactured lightweight aggregate, including particle density, water absorption and compressive strength, were determined. The drill cuttings had a typical evaporite composition containing high concentrations of chloride salts. This limits the potential for using the as-received drill cutting samples in lightweight aggregate production as the products formed show high levels of leaching. The addition of a washing pre-treatment to reduce the leaching of chloride ions was necessary. Washing also reduced the initial sintering temperature and improved lightweight aggregate properties. Sintering at 1180°C produced lightweight aggregate with particle density of 1.29 g/cm³, water absorption of 3.6% and compressive strengths of 4.4 MPa. The research showed that lightweight aggregate manufacturing represents a resource efficient option for the reuse of waste drill cuttings and provides significant material saving and landfill diversion.

Keywords: drill cuttings; lightweight aggregate; resource efficiency; hazardous waste; leaching.

1. Introduction

Drill cuttings consist of a fine mix of rock particles produced by drilling for oil and gas during either exploration or production. They are contaminated with drilling fluids which is often a synthetic oil containing several additives to enhance the drilling operation. They may also contain high concentrations of Cl^- salts if the underlying formation is marine evaporite, an impermeable sedimentary rock formed in normal marine environments by evaporation of water that is often associated with oil and gas hydrocarbons (Bjørlykke, 2015).

Oil-containing drilling mud and wastes are classified as hazardous in the EU Waste Catalogue (01 05 05*). As a result, they require onshore waste disposal, adding further pressure on the hazardous waste landfill capacity. In 2016, 40,300 tonnes of drill cuttings produced from the UK Continental Shelf (UKCS) were discharged into the sea and 30,100 tonnes were returned to shore for treatment and disposal (Oil and Gas UK, 2017). The environmental impact of landfill is related to greenhouse gases (GHG) emissions, the transport of waste to the landfill, landfill operation, potential landfill leachate contamination of water and soil, and the production of noise, odour and visual intrusion (Eshet et al., 2006).

Managing drill cuttings as waste contributes significantly to the overall cost of drilling operations. North Sea operators allocate more than half of the expenditure on drilling fluids to waste management costs (Minton and McGlaughlin, 2003). Options for managing waste drill cuttings include offshore-based re-injection or discharge and onshore-based landfill (de Almeida et al., 2017), bioremediation (Yan et al., 2011), solvent extraction (Chen et al., 2017), solidification/stabilization with binders such as Portland cement (CEM I) (Leonard and Stegemann, 2010), use as Portland cement replacement in concrete products (Mostavi et al., 2015) and grouts (Aboutabikh et al., 2016) and thermal desorption treatments with microwave heating (Petri Junior et al., 2015). Offshore discharge poses a serious risk to the marine environment and the majority of the proposed onshore management options fail to reduce the hazardous properties when drill cuttings consist of high concentration of Cl^- salts. There have been only limited investigations into ways to transform waste drill cuttings into products of value and as a result there is a lack of commercially viable recycling options for this waste.

Lightweight aggregate (LWA) is a granular material with loose bulk density not exceeding 1.2 g/cm^3 or a particle density not exceeding 2.0 g/cm^3 (BS EN 13055-1, 2002). An ideal

LWA for use in concrete products should have low water absorption capacity, it should be roughly spherical (4 – 14 mm in diameter to be in the range of normal coarse aggregate), strong, porous, with a sintered core and an impermeable rough surface to enhance the cement-aggregate bond (Cheeseman and Viridi, 2005). Using LWA in concrete products improves thermal insulation properties and reduces the structural dead load, allowing the construction of larger buildings with the same foundation size, all of which are associated with reduced CO₂ emissions (Ayati et al., 2018).

Mineral wastes including excavated clay (Ayati et al., 2018), zeolitic rocks (Volland and Brötz, 2015), municipal solid waste incinerator bottom ash (MSWI BA) (Cheeseman et al., 2005), recycled glass (Arulrajah et al., 2015), high-carbon fly ash (Lo et al., 2015) and bauxite residues from aluminium production and pulverised fuel ash (PFA) (Molineux et al., 2016), have been extensively used to produce building materials including sintered porous products such as LWA. The abundance of research on the use of waste material in the manufacture of recycled aggregates is due to the environmental concerns associated with the extraction of natural aggregates. These include damage to top soil and natural plant life, pollution of groundwater due to the use of hazardous chemical such as Hg and As, landscape degradation by forming geometrical features that replace natural topographic relief, and nuisance effects caused by the mining operational equipment and the movement of trucks over quarry and public roads generating noise and dust (Papagiannis et al., 2014). It has been calculated that for every 1 tonne of recycled aggregate produced, the landfill and transportation of more than 1.5 tonne of waste and the extraction of the same amount of natural aggregate is avoided (Wijayasundara et al., 2018).

LWA are normally manufactured using high-temperature firing treatment. This provides an opportunity to use mineral wastes as raw materials because the process reduces the leachability of problematic constituents through physical encapsulation within a silica/aluminosilicate-based matrix. The aim of this research was to investigate using drill cuttings containing high concentration of Cl⁻ salts to manufacture LWA for structural concrete products.

2. Materials and methods

A sample of drill cuttings containing synthetic based mud (SBM) was obtained from a specialized waste management company responsible for managing drilling waste produced in

the North Sea (Augean plc, UK). The chemical composition of the as-received sample was determined using x-ray fluorescence spectrometry (XRF, PANalytical Axios with 4kW Rh-anode x-ray tube). The loss on ignition (LOI) was determined on dried samples by heating to 550°C for 4 hours following (BS EN 12879, 2000). Total petroleum hydrocarbons (TPH) was extracted according to (BS EN ISO 16703, 2011) and analysed using gas chromatography with a flame ionisation detector (CG-FID, Agilent 6890 Network Gas Chromatograph). Total metal content was extracted by acid digestion using Aqua Regia, (concentrated HNO₃ and HCl at a volumetric ratio of 1:3), followed by analysis using inductively coupled plasma optical emission spectroscopy (ICP-OES, Perkin Elmer Optima 8300).

The concentration of water soluble constituents in waste materials with a granular nature is normally determined by leach testing with the leachate composition controlled to mimic landfill conditions. However, in this research the manufactured LWA are used in construction scenarios where the materials own/natural pH governs the release of constituents (the leachate pH is determined by the released/water soluble constituents). Therefore, the as-received drill cuttings and manufactured LWA were subjected to leaching testing without acid or base addition as described by the British standard method (BS EN 12457-2, 2002) with leachate composition determined by ICP-OES. The leachable concentrations of Cl⁻ were analysed with a Dionex ion chromatograph (BS EN 6068-2, 1995). Dissolved organic carbon (DOC) in the leachate was determined by oxidation of organic carbon to CO₂ by supercritical water oxidation according to (BS EN 1484, 1997). The gas was then separated and analysed with a non-dispersive infrared (NDIR) module.

Mineral phases were analysed by x-ray diffraction (XRD, Philips with PW 1830 generator and PW 3020 goniometer) using Cu k-alpha radiation, a step size of 0.05° and a time for each step of 1 second. Peak identification used the programme 'Traces' in conjunction with the International Centre for Diffraction Data (ICDD) database powder diffraction file (PDF). Thermal analysis was performed using heating microscopy (EM201/HT163, Hesse Instruments Germany) with a maximum operating temperature of 1400°C ± 3°C.

The as-received drill cuttings (DC) were air-dried and ball milled in a 6-litre planetary ball mill (DECO-PBM) rotating at 300 rpm for 6 hours and sieved with a mesh size of 250 µm. Drill cuttings were subjected to a single-stage washing pre-treatment (W-DC) with de-ionised water at L/S ratio of 10 l/kg for 1 hour.

Milled samples of DC and W-DC were pelletized by adding approximately 25 ± 2 wt.% water and formed into roughly spherical pellets of between 7 and 14 mm in diameter (the size range of coarse aggregate) by hand rolling. These were dried to constant weight and the ‘green pellets formed were then fired in a muffle furnace (SNOL 6.7/1300) using a dwell time of 10 minutes and a heating rate of $10^{\circ}\text{C}/\text{min}$. The firing temperature range was determined from heating microscopy data. The minimum temperature at which the material showed sintering was defined as the lower firing limit and the temperature at which melting started was the higher firing limit. The ‘green’ pellets were then fired at four temperatures within the sintering range. Fired pellets made from as-received drill cuttings are identified as DCLWA and those made from drill cuttings subjected to a washing pre-treatment are W-DCLWA.

Particle density (ρ_{dr}) on an oven-dried basis for fired pellets was measured using Archimedes principle as described in (BS EN 1097-6, 2013) and calculated using:

$$\rho_{\text{dr}} = (\rho_{\text{w}} \times m_{\text{dry}}) / (m_{\text{sat}} - m_{\text{imm}}) \quad (1)$$

where ρ_{w} is the density of water in g/cm^3 , m_{dry} is the mass in g of oven-dried LWA, m_{sat} is the mass in g of saturated and surface-dried LWA (24 hours in water) and m_{imm} is the apparent mass (immersed mass) in g of LWA.

Water absorption capacity (WA_{24}) after immersion in water for 24 hours was calculated as a percentage of the dry mass:

$$\text{WA}_{24} = 100 \times (m_{\text{sat}} - m_{\text{dry}}) / m_{\text{dry}} \quad (2)$$

Compressive strength (S) of individual pellets was calculated by loading the pellets to fracture between two parallel rigid surfaces and using the following equation (Yashima et al., 1987):

$$S = (2.8P_{\text{c}}) / (\pi X^2) \quad (3)$$

where P_{c} is the fracture load in N and X is the distance between the loading points in mm.

The fracture load was determined using a load ring with 4.55 kN capacity (Wykeham-France) at the speed of 0.05 mm/s. The microstructure of LWA fracture and outer surface were analysed using scanning electron microscopy (SEM, JOEL JSM 6460LV).

3. Results

3.1 Material characterization

Table 1 shows XRF data for major oxides, minor elements and total metal content determined after acid digestion of as-received drill cuttings. Table 1 also includes leach testing results and LOI and TPH data. XRF analysis showed concentrations of SiO_2 and Al_2O_3 were 41.18 wt.% and 12.75 wt.%. These are in the range associated with a suitable high-temperature viscosity which is required for bloating to produce low-density LWA (Dondi et al., 2016). TPH content is due to the presence of drilling fluids and was measured at 5.6 wt.% which is above the limit of Toxic for Reproduction (HP 10). Minor elemental analysis showed Ba was present at 32,019 mg/kg which is associated with BaSO_4 which is added to drilling fluids to increase the density of the mud system and control the well pressure. The acid and water-soluble Ba concentrations were 258.0 mg/kg and 0.43 mg/kg due to the stability of BaSO_4 . The leachable concentrations of most of the heavy metals in drill cuttings were below the ICP-OES detection limit, except Cu (1.07 mg/kg) and Ni (0.42 mg/kg). This indicates chemical fixation, as the acid-soluble concentrations were 41.3 mg/kg and 41.8 mg/kg respectively. The concentration of water soluble Cl^- was 85,712 mg/kg. This was problematic in term of leaching and required the use of a washing pre-treatment.

Figure 1 shows XRD traces for DC and W-DC. The DC sample contained quartz (SiO_2), calcite (CaCO_3), dolomite ($\text{CaMg}(\text{CO}_3)_2$), anhydrite (CaSO_4), barite (BaSO_4), halite (NaCl), muscovite ($\text{KAl}_2(\text{Si}_3\text{AlO}_{10})(\text{OH})_2$) and sanidine (KAlSi_3O_8). The mineral phases identified indicate that the drill cuttings are marine evaporite (Harff et al., 2016). The same phases were identified in W-DC except the NaCl was removed, as analysis showed all the peaks associated with NaCl disappeared after washing for one hour.

Figure 2a shows heating microscope (HM) images obtained at the characteristic temperatures for DC. The initial sintering temperature was 1175°C as the outline area reduced to 95% of the original area at this temperature. As sintering continued the cube outline remained relatively uniform but the dimensions reduced at temperatures up to 1190°C which was identified as the higher firing limit. Above 1190°C a liquid phase formed on the surface and the material begins to flow. Similar thermal behaviour was observed for W-DC sample as shown in Figure 2b. However, the initial sintering temperature was reduced to 1160°C , extending the sintering temperature range, which is advantageous for large-scale LWA manufacturing.

3.2 Properties of manufactured LWA

Figure 3a shows that the surface and internal structure of DCLWA were heterogeneous in terms of smoothness, colour and porosity, despite the relatively narrow firing range used. This was possibly due to the high level of salts in evaporite minerals which can build-up a layer on green pellets and deteriorate the surface. LWA showed bloating and larger spherical pores developed in the internal structure as the temperature increased. Figure 3b shows the improved quality of the W-DCLWA surface. W-DCLWA also exhibited bloating, with cellular pore formation appearing at 1160°C. W-DCLWA showed an ideal pore structure at 1170°C but as the temperature increased the LWA pellets showed excessive bloating. This was problematic because it resulted in an inconsistent internal structure and a non-uniform pore size distribution.

Figure 4 shows the effect of firing temperature on the physical properties of DCLWA and W-DCLWA, including data on particle density, water absorption and compressive strength. The results are compared to the physical properties of a commercially manufactured LWA (Lytag) determined using identical test methods. The average particle density of DCLWA decreased from 1.44 to 1.25 g/cm³ as the firing temperature increased from 1175 to 1185°C due to the formation of larger pores. The range of particle densities obtained was comparable to Lytag. Water absorption also decreased from 9.2 to 7.4% over the same temperature range, showing that new pores formed during bloating were less accessible to water. Compressive strength remained stable at approximately 4 MPa over the temperature range from 1180 to 1190°C and this was also comparable to Lytag.

The particle density of W-DCLWA decreased from 1.88 to 1.29 g/cm³ as the firing temperature increased from 1160 to 1180°C (Figure 4). The lowest water absorption of 2.7% was achieved by firing at 1170°C. This increased to 5.7% at 1190°C due to increased connectivity of the pores. However, the range of water absorption values obtained remained lower than for DCLWA. The average compressive strength of individual pellets of W-DCLWA decreased from 21.0 to 2.7 MPa as the temperature increased from 1160 to 1190°C. The initial strength development of the material confirms the change in the sintering range to lower temperatures. However, as the temperature increased to 1180°C, pellets showed excessive bloating which resulted in an 80% reduction in compressive strength. Overall, W-DCLWA manufactured at temperatures between 1170 and 1180°C were comparable with Lytag in terms of density and strength, although they had lower water absorption.

Figure 5a and Figure 5b show the outer surface and fracture surface of DCLWA produced at 1180°C. A crystallised outer surface showing efflorescence caused by migration and crystallization of soluble salts formed on the outer surface during the drying stage. The fracture surface consists of a continuous phase composed of sintered drill cutting particles containing roughly spherical 10 - 50µm pores which indicates the occurrence of bloating at a micro level. There was also evidence of open porosity in the continuous phase. This is caused by the remaining hydrocarbons in drill cuttings which produce open cavity channels during firing which keep particles apart, reducing sintering efficiency. This is similar to the adverse effect caused by adding excessive water during pellet formation.

Figure 5c and 5d show SEM micrographs of fracture surface for W-DCLWA manufactured at 1160°C and 1190°C. LWA pellets fired at 1160°C developed an ideal pore structure comprising of uniform closed spherical pores with sizes decreasing towards the surface. The low-temperature bloating observed generated micro-pores with a wide size distribution ranging from 10 to 500 µm in diameter. The different microstructure of W-DCLWA is thought to be due to the removal of soluble salts which results in a more coherent matrix with a suitable viscosity necessary for bloating. At 1190°C very large voids formed in the middle of the pellet due to excessive bloating caused by simultaneous occurrence of gas release and low pyro-plastic viscosity of the matrix.

Figure 5e and Figure 5f show two distinct regions on the outer surface of the W-DCLWA manufactured at 1170°C. A lower magnification image is also attached to both images showing the locations where the images were captured. The crystallized surface texture in Figure 5e is due to efflorescence from the accumulation of soluble compounds on the sample surface. This layer is incapable of expansion and cracked during core bloating. Figure 5f shows a very different microstructure, predominantly composed of a melted/densified phase in regions free from any deposits. It is likely that these phases were formed on the surface during the bloating stage and solidified in place. The observed rough surface microstructure can be beneficial for the drill cuttings LWA performance in concrete products as it can improve the aggregate/cement paste bond.

Figure 6 shows XRD traces of DCLWA and W-DCLWA. The major neo-formed phase in both fired samples was diopside ($\text{CaMgSi}_2\text{O}_6$). Anorthite ($\text{CaAl}_2\text{Si}_2\text{O}_8$) and forsterite (Mg_2SiO_4) were also neo-formed in fired pellets. These new phases are formed by solid-state reactions of CaO and MgO liberated from CaCO_3 and $\text{CaMg}(\text{CO}_3)_2$, with

silica/aluminosilicate phases made available from quartz and $\text{KAl}_2(\text{Si}_3\text{AlO}_{10})(\text{OH})_2$. The neo-formation of diopside is favourable because it improves the high-temperature viscosity and the encapsulation properties of the LWA. Furthermore, the interlocking crystal structure of diopside can enhance compressive strength (Donatello and Cheeseman, 2013). NaCl peaks present in the DCLWA XRD trace disappeared in W-DCLWA. The intensity of CaSO_4 peaks in DCLEA and W-DCLWA reduced due to decomposition effects. Gypsum peaks were detected in fired samples and this is probably due to hydration of anhydrite.

Table 2 shows the water-soluble concentration of the majority of heavy metals in DCLWA and W-DCLWA remained below the instrument detection limits. Ba was measured at 1.1 and 0.8 mg/kg and Mo was 1.2 and 1.5 mg/kg in DCLWA and W-DCLWA respectively. The increase in Mo is possibly due the formation of phases that are unstable under the atmospheric conditions at high temperatures (González-Corrochano et al., 2012). The concentrations of Cl^- decreased to 20,365.8 and 1463.5 mg/kg in DCLWA and W-DCLWA fired at 1180°C and this demonstrates the rapid kinetics of Cl^- release in water during relatively short washing durations. DOC was below the detection limit in both DCLWA and W-DCLWA.

4. Discussion

Pelletizing and firing waste drill cuttings can produce LWA with physical properties comparable to commercial products. Drill cuttings showed bloating because they contain minerals such as muscovite, feldspars and quartz which enhance sintering and gas entrapment (Huang and Wang, 2013). Sintering transformed drill cuttings mineralogy producing LWA containing $\text{CaMgSi}_2\text{O}_6$ as the major neo-formed phase. BaSO_4 remained unreacted in the manufactured LWA due to its thermal stability over the range of firing temperatures used.

Washing extended the sintering range, improved the bloating mechanism and the microstructure of sintered pellets and also resulted in optimal physical properties being achieved within a narrow temperature range. Two batches of W-DCLWA with physical properties comparable to commercial LWA products were produced. Firing at 1170°C gave samples with a particle density of 1.54 g/cm³, 2.7% water absorption and 13.1 MPa compressive strength. 1180°C was determined to be the optimal firing temperature producing a particle density of 1.29 g/cm³, 3.6% water absorption and compressive strengths of 4.4 MPa.

The LWA manufactured in this research is expected to have comparable performance to other waste derived LWA. The very low water absorptions of drill cuttings derived LWA is beneficial during concrete mixing. This is because properties of hardened concrete are highly sensitive to variations in water-to-cement (w/c) ratios that are caused by LWA with high water absorbency (Nadesan and Dinakar, 2017). In addition, $\text{CaMgSi}_2\text{O}_6$, the major neo-formed phase in LWA, can be effective in inhibiting the expansive nature of alkali-silica gel forming reactions between cement and the amorphous silica found in many other waste derived LWA (Tajuelo Rodriguez et al., 2018). It must be noted that a comprehensive evaluation of drill cuttings LWA performance in concrete products is required. This can be crucial for durability of concrete since the manufactured LWA contained BaSO_4 which can decompose over long periods of time and cause sulphate attack.

The inherent concentrations of heavy metals in evaporite drill cuttings were acceptable compared to other problematic hazardous mineral wastes (Allegrini et al., 2014), alleviating concerns over release in various reuse applications. This is a long-term environmental advantage for drill cuttings LWA over other LWA derived from wastes such as MSWI bottom ash and fly ash that contain relatively high contents of heavy metals (Bethanis et al., 2002; Lu et al., 2015). However, the high concentration of leachable Cl^- in the raw materials made manufactured LWA unviable for concrete applications due to the criticality of the Cl^- -induced corrosion of steel in reinforced concrete structures. The 76.3wt.% reduction of water-soluble Cl^- in DCLWA, compared to the as-received drill cuttings, showed that firing was effective at mitigating the Cl^- ions. This was probably caused by volatilization of Cl^- at the firing temperatures used, although Cl^- release may also be reduced by decreasing the surface area exposed to leachate and/or isolating the ions from the environment due to the impermeable properties of the silicate based matrix of LWA (Chandler et al., 1997). The effectiveness of firing in mitigation of Cl^- was only partially successful and a single-stage washing was therefore necessary, which removed up to 98.3% of Cl^- from W-DCLWA.

EU directives require treatment of hazardous waste before landfill and the high cost of hazardous waste landfill (£130 - £150 per tonne) in the UK are strong incentives to drive waste management companies to treat the drill cuttings to achieve inert landfill criteria. Thermal desorption technologies have significant issues in terms of efficiency, mainly due to the granular nature of drilling cuttings that makes heat transfer extremely inefficient (Pereira et al., 2014). Alternatively, drill cuttings can be mixed on-site with other mineral wastes such as pulverised fuel ash (PFA) for stabilization and disposal, although this is not a sustainable

management option. It is clear that increased quantities of drilling waste will be produced in the future and with no existing reuse technology available, more drill cuttings will be landfilled. New oil reservoirs in the North Sea and the need for more complex drilling operations will also increase the volume drill cutting waste produced. Another important benefit of using waste drill cuttings as a raw material for LWA production is the remaining portion of hydrocarbon in the material. In large-scale LWA manufacturing, organic matter is normally added to the mix to provide part of the energy required for firing. Synthetic drilling fluids have a high calorific value and therefore this can supply additional energy to the firing process.

In addition, drill cuttings LWA will have lower embodied CO₂ compared to commercial products. The reduction is due to the onshore availability of waste drill cuttings which saves energy associated with the import and extraction of raw materials for LWA. Current LWA production includes PFA derived aggregates (~150,000 tonnes per year), expanded clay aggregates (~150,000 tonnes per year) and naturally occurring pumice (~100,000 tonnes per year) either imported or sourced from quarries (Cresswell, 2007). At the same time the reliance on PFA is not sustainable due to the reduction in coal-fired power generation and the supply of naturally occurring lightweight materials, such as pumice, is undesirable due to the serious environmental impacts associated with natural resource extraction. There are therefore significant drivers for reusing drill cuttings for LWA, although a comprehensive environmental impact assessment of large-scale LWA manufacture is needed.

5. Conclusions

This research demonstrates that technically viable LWA can be produced from waste drill cuttings. Evaporite drill cuttings contain high concentrations of leachable Cl⁻ and reuse requires a washing treatment. A single-step washing at L/S ratio of 10 kg/l for 1 hour mitigated Cl⁻ leaching and improved the physical properties of the LWA formed. Firing temperatures of 1180°C was optimal to achieve LWA with properties comparable to existing commercial products including particle density of 1.29 g/cm³, water absorption of 3.6% and compressive strength of 4.4 MPa. Firing changed the mineralogy of drill cuttings and produced LWA mainly composed of CaMgSi₂O₆. LWA manufacturing represents a beneficial reuse of drill cuttings that diverts this waste from landfill. No alternative viable recycling option for drilling waste currently exists and the feasibility of industrial/commercial scale-up requires further investigation.

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References

- Aboutabikh, M., Soliman, A.M., El Naggar, M.H., 2016. Properties of cementitious material incorporating treated oil sands drill cuttings waste. *Constr. Build. Mater.*, 111, 751-757.
- Allegrini, E., Maresca, A., Olsson, M.E., Holtze, M.S., Boldrin, A., Astrup, T.F., 2014. Quantification of the resource recovery potential of municipal solid waste incineration bottom ashes. *Waste. Manage.* 34(9), 1627-1636.
- Arulrajah, A., Disfani, M.M., Maghoolpilehrood, F., Horpibulsuk, S., Udonchai, A., Imteaz, M., Du, Y.J., 2015. Engineering and environmental properties of foamed recycled glass as a lightweight engineering material. *J. Clean. Prod.* 94, 369-375.
- Ayati, B., Ferrándiz-Mas, V., Newport, D., Cheeseman, C.R., 2018. Use of clay in the manufacture of lightweight aggregate. *Constr. Build. Mater.*, 162, 124-131.
- Bethanis, S., Cheeseman, C.R., Sollars, C.J., 2002. Properties and microstructure of sintered incinerator bottom ash. *Ceram. Int.* 28(8), 881-886.
- Bjørlykke, K., 2015. Introduction to Petroleum Geology. In: Bjørlykke K. (eds) *Petroleum Geoscience.*, Petroleum Geoscience. Springer, Berlin, Heidelberg, pp. 1-29.
- BS EN 1097-6, 2013. Tests for mechanical and physical properties of aggregates. Determination of particle density and water absorption.
- BS EN 1484, 1997. Water analysis. Guidelines for the determination of total organic carbon (TOC) and dissolved organic carbon (DOC).
- BS EN 6068-2, 1995. Water quality. Determination of dissolved anions by liquid chromatography of ions. Determination of fluoride, chloride, nitrite, orthophosphate, bromide, nitrate and sulfate ions. Method for water with low contamination.
- BS EN 12457-2, 2002. Characterisation of Waste-Leaching-Compliance Test for Leaching of Granular Waste Materials and Sludges: Part 2. One Stage Batch Test at a Liquid to Solid Ratio of 10 L/kg for Materials with Particle Size below 4 mm (without or with Size Reduction).
- BS EN 12879, 2000. Characterization of sludges. Determination of the loss of ignition of dry mass.
- BS EN 13055-1, 2002. Lightweight aggregates. Part 1: Lightweight aggregates for concrete, mortar and grout.
- BS EN ISO 16703, 2011. Soil quality. Determination of content of hydrocarbon in the range C10 to C40 by gas chromatography.
- Chandler, A.J., Eighmy, T.T., Hjelm, O., Kosson, D.S., Sawell, S.E., Vehlou, J., van der Sloot, H.A., Hartlén, J., 1997. *Municipal Solid Waste Incinerator Residues.* Elsevier Science, Amsterdam, The Netherlands.

- Cheeseman, C.R., Makinde, A., Bethanis, S., 2005. Properties of lightweight aggregate produced by rapid sintering of incinerator bottom ash. *Resour. Conserv. Recy.* 43(2), 147-162.
- Cheeseman, C.R., Viridi, G.S., 2005. Properties and microstructure of lightweight aggregate produced from sintered sewage sludge ash. *Resour. Conserv. Recy.* 45(1), 18-30.
- Chen, Z., Chen, Z., Yin, F., Wang, G., Chen, H., He, C., Xu, Y., 2017. Supercritical water oxidation of oil-based drill cuttings. *J. Hazard. Mater.* 333, 205-213.
- Cresswell, D., 2007. Industrial sector study on the utilisation of alternative materials in the manufacture of manufactured aggregates. DEFRA Project Code WRT_177 / WRO115. Mineral Industry Research Organisation (MIRO), Birmingham, UK.
- de Almeida, P.C., Araújo, O.d.Q.F., de Medeiros, J.L., 2017. Managing offshore drill cuttings waste for improved sustainability. *J. Clean. Prod.* 165, 143-156.
- Donatello, S., Cheeseman, C.R., 2013. Recycling and recovery routes for incinerated sewage sludge ash (ISSA): A review. *Waste. Manage.* 33(11), 2328-2340.
- Dondi, M., Cappelletti, P., D'Amore, M., de Gennaro, R., Graziano, S.F., Langella, A., Raimondo, M., Zanelli, C., 2016. Lightweight aggregates from waste materials: Reappraisal of expansion behavior and prediction schemes for bloating. *Constr. Build. Mater.* 127, 394-409.
- Eshet, T., Ayalon, O., Shechter, M., 2006. Valuation of externalities of selected waste management alternatives: A comparative review and analysis. *Resour. Conserv. Recy.* 46(4), 335-364.
- González-Corrochano, B., Alonso-Azcárate, J., Rodas, M., 2012. Chemical partitioning in lightweight aggregates manufactured from washing aggregate sludge, fly ash and used motor oil. *J. Environ. Manage.* 109, 43-53.
- Harff, J., Meschede, M., Petersen, S., Thiede, J., 2016. *Encyclopedia of Marine Geosciences*. Springer, Dordrecht.
- Huang, C.-H., Wang, S.-Y., 2013. Application of water treatment sludge in the manufacturing of lightweight aggregate. *Constr. Build. Mater.* 43, 174-183.
- Leonard, S.A., Stegemann, J.A., 2010. Stabilization/solidification of petroleum drill cuttings: Leaching studies. *J. Hazard. Mater.* 174(1-3), 484-491.
- Lo, T.Y., Cui, H., Memon, S.A., Noguchi, T., 2015. Manufacturing of sintered lightweight aggregate using high-carbon fly ash and its effect on the mechanical properties and microstructure of concrete. *J. Clean. Prod.* 112, 753-762.
- Lu, C.-H., Chen, J.-C., Chuang, K.-H., Wey, M.-Y., 2015. The different properties of lightweight aggregates with the fly ashes of fluidized-bed and mechanical incinerators. *Constr. Build. Mater.* 101, Part 1, 380-388.
- Minton, R., McGlaughlin, J., 2003. Rising drilling-waste costs promote novel technologies, approaches. *Oil Gas J.* 101(31), 51-51.

- Molineux, C.J., Newport, D.J., Ayati, B., Wang, C., Connop, S.P., Green, J.E., 2016. Bauxite residue (red mud) as a pulverised fuel ash substitute in the manufacture of lightweight aggregate. *J. Clean. Prod.* 112, 401-408.
- Mostavi, E., Asadi, S., Ugochukwu, E., 2015. Feasibility Study of the Potential Use of Drill Cuttings in Concrete. *Procedia Eng.* 118, 1015-1023.
- Nadesan, M.S., Dinakar, P., 2017. Mix design and properties of fly ash waste lightweight aggregates in structural lightweight concrete. *Case Stud. Constr. Mater.* 7, 336-347.
- Oil and Gas UK, 2017. Environment Report 2017. <https://oilandgasuk.co.uk/wp-content/uploads/2017/12/Environment-Report-2017.pdf>. (Accessed 31/07/2018).
- Papagiannis, A., Roussos, D., Menegaki, M., Damigos, D., 2014. Externalities from lignite mining-related dust emissions. *Energ Policy* 74, 414-424.
- Pereira, M.S., Panisset, C.M.d.Á., Martins, A.L., Sá, C.H.M.d., Barrozo, M.A.d.S., Ataíde, C.H., 2014. Microwave treatment of drilled cuttings contaminated by synthetic drilling fluid. *Sep. Purif. Technol.* 124, 68-73.
- Petri Junior, I., Pereira, M.S., dos Santos, J.M., Duarte, C.R., Ataíde, C.H., Panisset, C.M.d.Á., 2015. Microwave remediation of oil well drill cuttings. *J. Pet. Sci. Eng.* 134, 23-29.
- Tajuelo Rodriguez, E., Anovitz, L.M., Clement, C.D., Rondinone, A.J., Cheshire, M.C., 2018. Facile emulsion mediated synthesis of phase-pure diopside nanoparticles. *Sci Rep.* 8(1), 3099.
- Volland, S., Brötz, J., 2015. Lightweight aggregates produced from sand sludge and zeolitic rocks. *Constr. Build. Mater.* 85, 22-29.
- Wijayasundara, M., Mendis, P., Crawford, R.H., 2018. Net incremental indirect external benefit of manufacturing recycled aggregate concrete. *Waste. Manage.* 78, 279-291.
- Yan, P., Lu, M., Guan, Y., Zhang, W., Zhang, Z., 2011. Remediation of oil-based drill cuttings through a biosurfactant-based washing followed by a biodegradation treatment. *Bioresour. Technol.* 102(22), 10252-10259.
- Yashima, S., Kanda, Y., Sano, S., 1987. Relationships between particle size and fracture energy or impact velocity required to fracture as estimated from single particle crushing. *Powder Technol.* 51(3), 277-282.

Table 1

Chemical composition of evaporite drill cuttings. LOI: Loss on ignition, TPH: Total petroleum hydrocarbon and DOC: Dissolved organic hydrocarbon.

Major oxides		Minor elements		Acid soluble	Water soluble *
	(wt.%)		(mg/kg)	(mg/kg)	(mg/kg)
SiO ₂	41.18	As	11.8	<DL ^{**}	<DL
Al ₂ O ₃	12.75	Ba	32,019	258.0	0.43
Fe ₂ O ₃	3.29	Cd	-	<DL	<DL
MgO	7.34	Cr	74.3	41.3	<DL
CaO	14.95	Cu	23.9	23.0	1.07
Na ₂ O	17.32	Hg	-	<DL	<DL
K ₂ O	2.53	Mo	1.1	<DL	<DL
TiO ₂	0.45	Ni	36.0	41.8	0.42
MnO	0.07	Pb	38.5	<DL	<DL
P ₂ O ₅	0.12	Sb	-	<DL	<DL
		Se	-	<DL	<DL
LOI	8.2	Zn	102.4	109.1	<DL
TPH _{C06 - C44} ^{***}	5.6	Cl ⁻	80,911		85,712
		DOC	-	-	477.1
		pH	-	-	9.9

* Batch leaching test (BS EN 12457) at L/S=10 l/kg

** Below detection limit.

*** Number of carbons in the polymer chain length.

Table 2

Results of leaching test (BS EN 12457-2) for DCLWA and W-DCLWA manufactured at 1180°C. DOC: Dissolved organic hydrocarbon.

	DCLWA	W-DCLWA
	(mg/kg)	(mg/kg)
As	<DL*	<DL
Ba	1.1	0.8
Cd	<DL	<DL
Cr	0.2	<DL
Cu	<DL	<DL
Hg	<DL	<DL
Mo	1.2	1.5
Ni	<DL	<DL
Pb	<DL	<DL
Sb	<DL	<DL
Se	<DL	<DL
Zn	<DL	<DL
Cl ⁻	20,365.8	1463.5
DOC	<DL	<DL
pH	9.4	10.6

* Below detection limit.

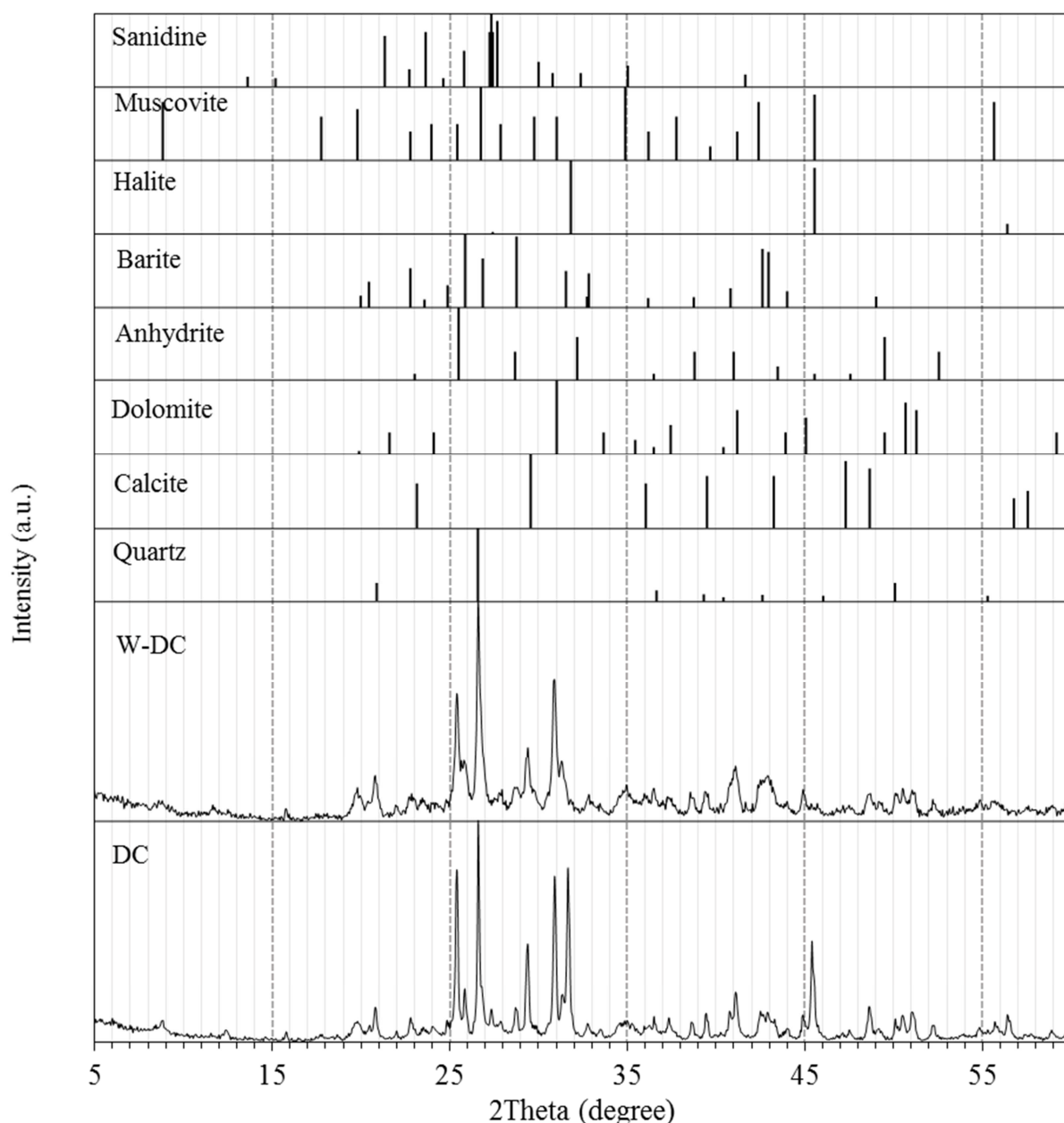
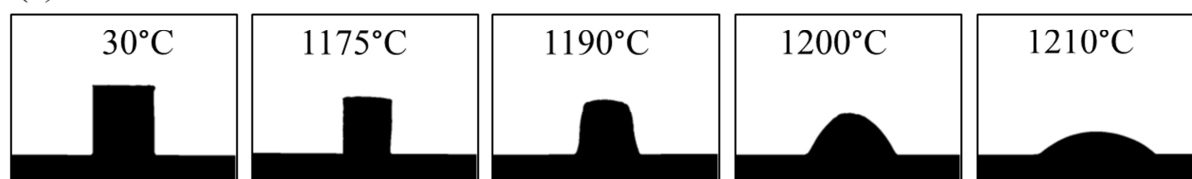


Fig. 1. XRD traces for the as-received drill cuttings (DC) showing the major crystalline phases are quartz (SiO_2), calcite (CaCO_3), dolomite ($\text{CaMg}(\text{CO}_3)_2$), anhydrite (CaSO_4), barite (BaSO_4), halite (NaCl), muscovite ($\text{KAl}_2(\text{Si}_3\text{AlO}_{10})(\text{OH})_2$) and sanidine (KAlSi_3O_8). XRD traces for washed drill cuttings (W-DC) shows the absence of NaCl peaks. The representative XRD patterns of the major phases are included with the same intensity scale.

(a) DC



(b) W-DC

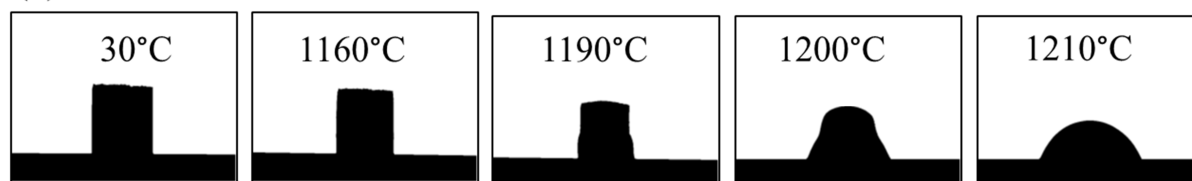


Fig. 2. Heating microscope images for a cubic sample (3 x 3 x 3 mm) of (a) as-received drill cuttings (DC) and (b) washed drill cuttings (W-DC) at characteristic temperatures.

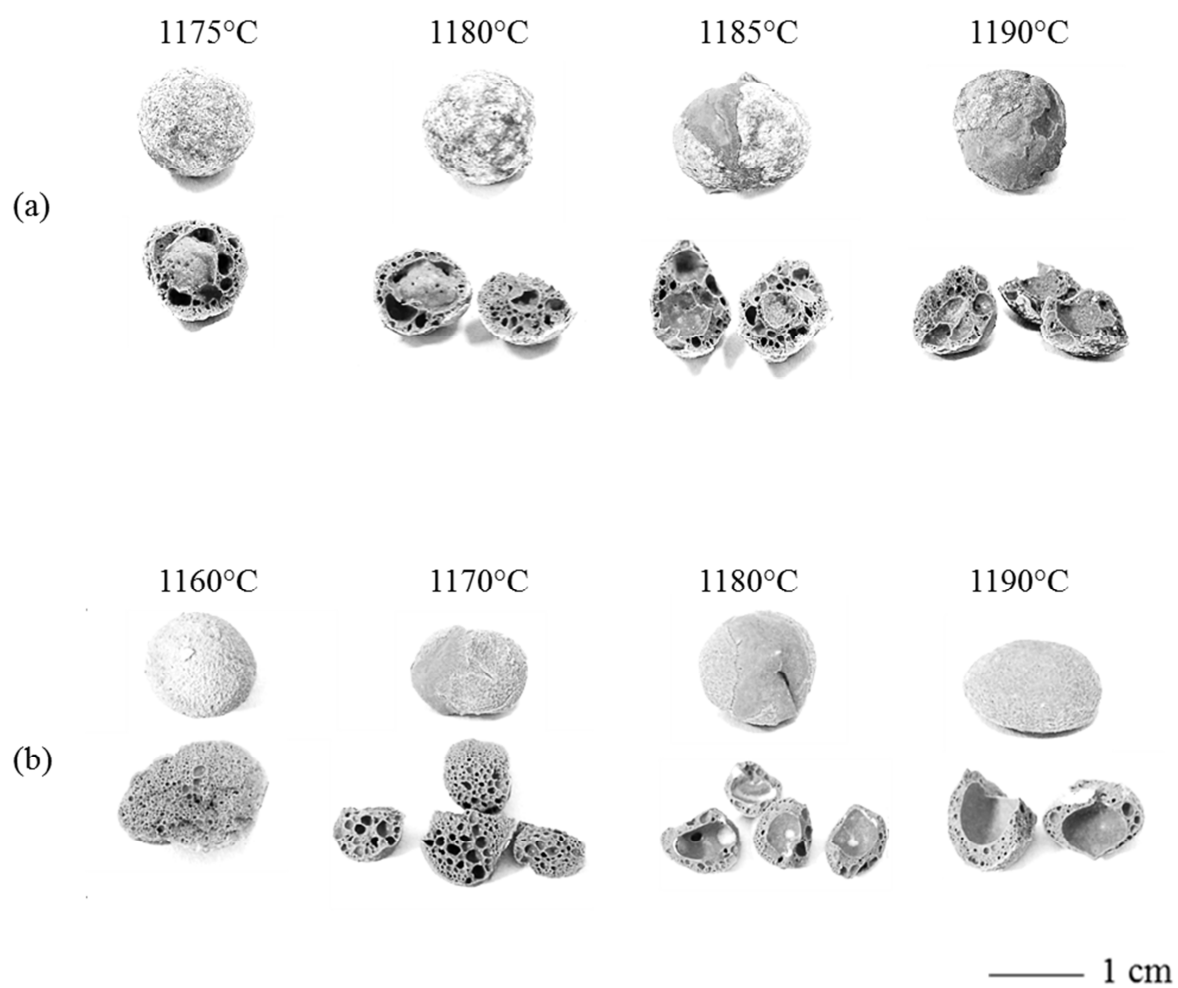


Fig. 3. Appearance and internal structure of (a) DCLWA and (b) W-DCLWA.

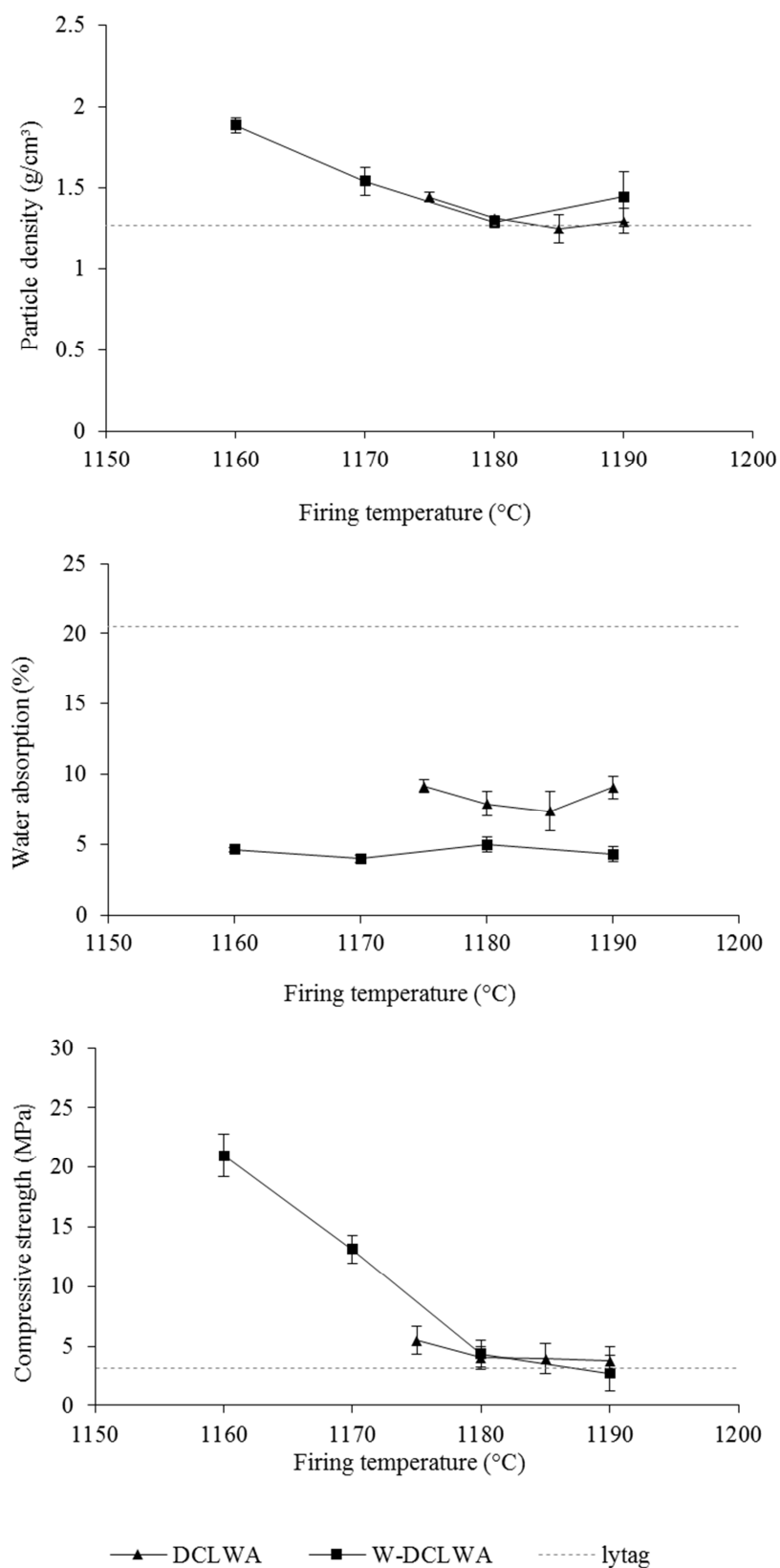


Fig. 4. Particle density, water absorption and compressive strength measured at various firing temperatures for DCLWA and W-DCLWA. Properties of Lytag are marked (dashed line). Error bars are $\pm$ one standard deviation.

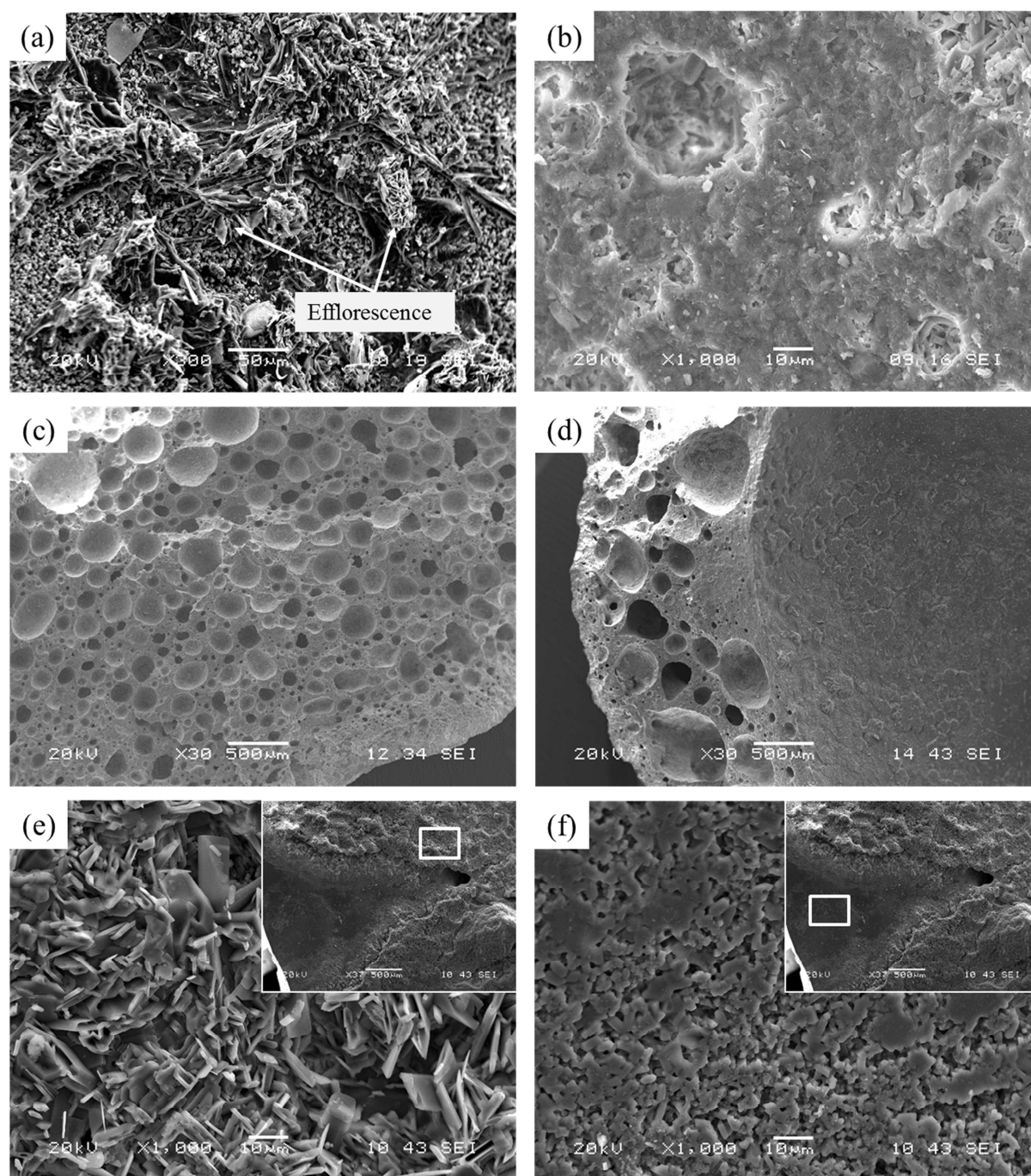


Fig. 5. SEM micrographs of (a) outer surface of DCLWA fired at 1180°C, (b) fracture surface of DCLWA fired at 1180°C (c) fracture surface of W-DCLWA fired at 1160°C (d) fracture surface of W-DCLWA fired at 1190°C (e) – (f) outer surface of W-DCLWA fired at 1170°C (the attached image shows the locations of magnified areas on the outer surface).

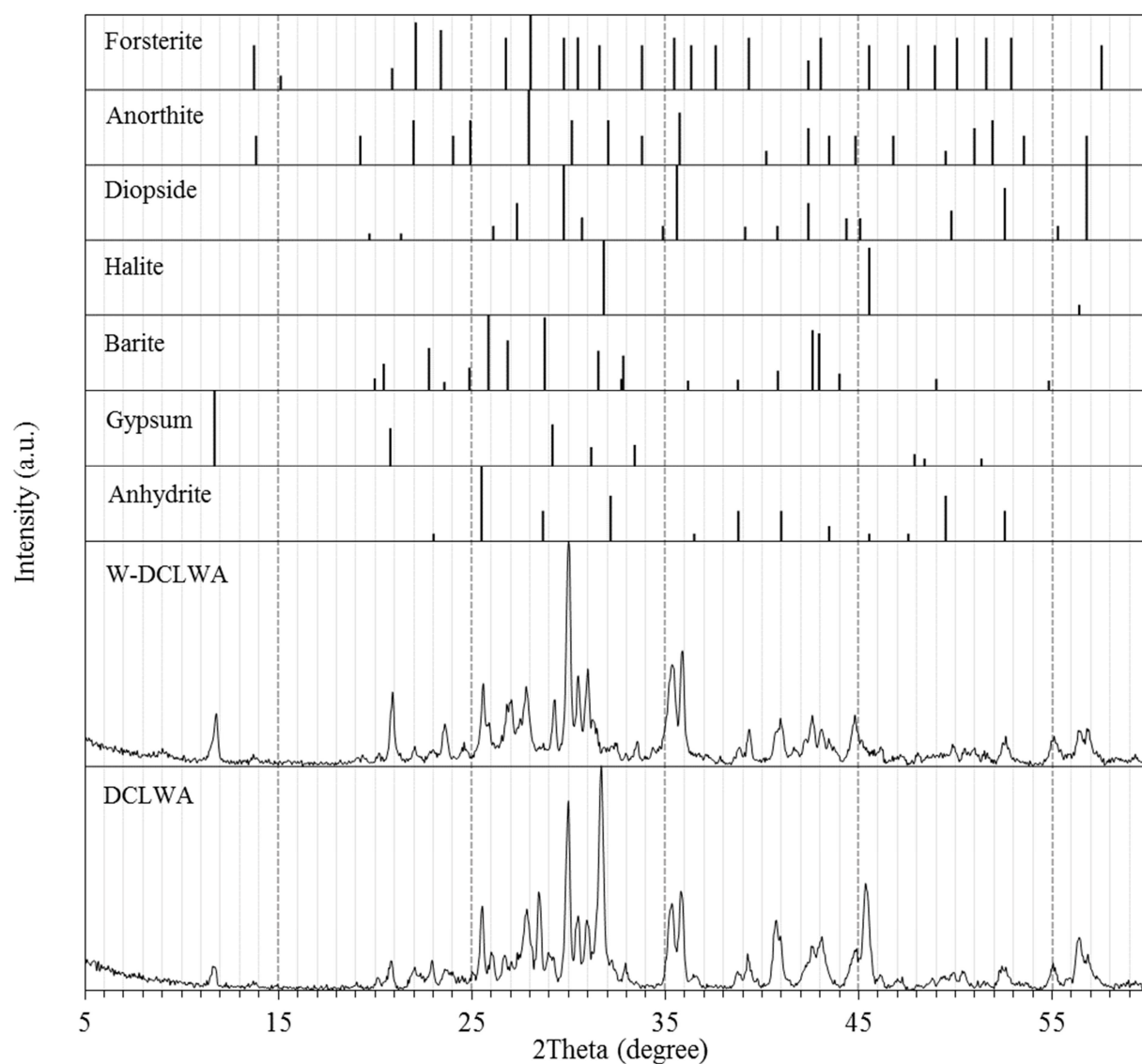


Fig. 6. XRD traces for DCLWA and W-DCLWA manufactured at 1180°C showing that diopside ($\text{CaMgSi}_2\text{O}_6$) is the major neo-formed crystalline phase present after firing. The representative XRD patterns of the major phases are included with the same intensity scale.

- Beneficial reuse of waste drill cuttings in the manufacture of LWA is presented
- LWA with properties comparable to commercial products were produced
- A washing treatment was required to mitigate Cl^- leaching from LWA
- The effect of washing on the physical properties of LWA is reported