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Use of clay in the manufacture of lightweight aggregate

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Abstract

Clay is used as a raw material for the production of lightweight aggregates because it is readily processed into suitable granules and forms low-density but high strength aggregate particles when sintered at relatively low temperatures. The use of waste clay generated by major infrastructure development projects to make lightweight aggregate has a positive environmental impact and contributes towards a more circular economy. This paper reviews the manufacturing process used to produce lightweight aggregates from clay and the influence of processing conditions on properties. It also reviews secondary materials that have been incorporated into clays to produce lightweight aggregates. Additional research is required to improve understanding of the effects of composition and production parameters on the pore structure, density, water adsorption and strength of clay derived lightweight aggregates.

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1. Introduction

Managing clay containing mineral wastes has become a major challenge for many major civil infrastructure construction projects [1]. Problems are often associated with transporting high volumes of clay mineral wastes, which causes pollution, increased road congestion and risks to public safety. The disposal options for excavated materials are also an issue. For example, construction of the Crossrail 1 underground tunnel in London UK, resulted in more than 4 million tonnes of waste London clay which was primarily used in land reclamation [2]. Additional future and ongoing tunnelling projects in London include the Thames Tideway Tunnel and the High Speed Two (HS2) rail link, and these two infrastructure development projects will produce an estimated 18.8 million tonnes of excavated materials that predominantly contain clay minerals [3].

There are often significant economic and environmental drivers to use recycled materials in construction as this can contribute to a circular economy in which waste materials remain part of the economic cycle [4]. The use of clay in the manufacture of lightweight aggregates (LWA) is a potential recycling option for clay wastes. LWA has previously been produced from London clay generated by Crossrail at pilot plant scale, and it was estimated that 2.8 million tonnes of LWA could have been manufactured from Crossrail excavated clay. This would have produced more than 9.0 million cubic metres of low-carbon lightweight structural concrete [2].

The market for LWA is expected to increase as the demand for lightweight and thermally insulating concrete increases. Of the alternative secondary raw materials available to produce LWA, clay remains the most viable, due to consistent properties and availability close to urban areas from excavations and tunnelling. Table 1 contains journal publications on the use of clay in manufacturing LWA over the past two decades. In addition to excavation wastes,

clay minerals are extracted from harbours, river beds and reservoirs where they cause serious sedimentation problems. Research on recycling these materials into LWA also provides a potential reuse option [5-13].

This paper reviews the properties of LWA produced from clays and discusses the role of chemical composition, processing conditions and microstructure in manufacturing optimum products. In addition, the review provides guidance on the types of secondary materials that have been incorporated into clay to produce LWA and identifies areas where further research is required.

2. LWA manufactured from clay

LWA was first manufactured commercially in the UK during the 1950s using clay and shale from the mining and slate industries. Additional types of LWA were developed to meet increasing national demand. Lytag is a LWA manufactured from pulverized fuel ash (PFA), a by-product from coal-fired power stations [14,15]. It was first manufactured in 1958 and remains a commercially leading LWA for structural lightweight concrete [16]. The availability of PFA in the UK and EU will decline in the future due to the move away from coal fired power stations [17]. In addition, PFA can be used as a supplementary cementitious material (SCM) and this is an alternative reuse application that may limit PFA availability for LWA production [18-20].

Figure 1 shows a typical manufacturing process for producing LWA from clay. The two main stages are granule formation and sintering [21]. The raw materials are finely ground and mixed in specific proportions. Unfired 'green' granules are formed by extrusion or agglomeration using an appropriate water addition. The physical properties of 'green' granules are important during handling and stockpiling and these depend on the granulation

process, the properties of the clay and the moisture content [22,23]. The influence of granulation parameters on the properties of 'green' granules is complex and a comprehensive model that predicts granulation behaviour and performance is not currently available. After granulation, the 'green' granules are normally dried and then sintered. The sintering temperatures for clay LWA are typically between 1050 °C and 1250 °C, using typical dwell times in the kiln of between 3 and 20 minutes.

The production of clay derived LWA requires processing in a temperature range where pyro-plastic deformation, gas generation and gas retention occur simultaneously. The main sources of gas generation in clay-containing minerals at high temperatures are dissociation or reduction of ferric oxides, combustion of organic matter, release of interlayer water molecules and thermal decomposition of carbonates [24,25]. The temperatures at which gases are generated vary and this influences/controls the bloating behaviour [26]. It is not normally possible to identify the critical components that cause bloating from bulk chemical composition data, but the ratio of silica and alumina content to the flux content is normally considered to be an important parameter [27]. However, the proportion of alumina-iron oxides-alkaline earths, regardless of silica content is also reported to control bloating [28]. As a result, it is difficult to predict whether or not a material will bloat based only on chemical composition data, and normally firing trials need to be completed [29].

2.1 Properties of LWA manufactured using clay

Standards for concrete, mortar and grout define LWA as a granular material with a loose bulk density below 1.2 g/cm^3 or a particle density not exceeding 2.0 g/cm^3 [30]. An ideal clay LWA for use in concrete would be roughly spherical, 4 to 14 mm in diameter, with a strong, porous, sintered core and an impermeable rough surface to enhance the hydrated cement-aggregate bond [31]. The pore structure of a clay LWA is a major factor determining particle

density, water absorption and strength. Raw material characteristics and production parameters also influence the properties of LWA through the pore structure. The pore structure is therefore an intermediate variable that allows construction of causal pathways, as shown in Figure 2, between the composition, processing and key properties. The influence of chemical composition and processing parameters on pore structure and the influence of pore structure on physical properties, marked as (i), (ii) and (iii), are explained as follows:

(i)   Effect of chemical compositions on pore structure of clay LWA

The bloating mechanism in clay minerals occurs when a viscous silicate-containing phase captures gases released at high temperature. The flux content determines the temperature range over which pyro-plasticity occurs, while a number factors, including the silicate content, control the viscosity [32]. An important mineralogical feature for clay-bearing raw materials is the SiO_2/Σ flux (i.e. Fe_2O_3, Na_2O, K_2O, CaO and MgO) ratio [27]. This has been widely used to predict bloating of clay bodies and some work reports a relationship between this ratio and the total porosity in manufactured LWAs. In addition, effective bloating requires the formation of a non-porous vitrified surface layer (shell), to capture the released gases [33]. The degree of shell vitrification is also related to the SiO_2/Σ flux [34].

(ii)   Effect of production parameters on pore structure of clay LWA

The schematic pore structure of clay LWA is shown in Figure 3 (left). It consists of pores evenly distributed across the core matrix, surrounded by a non-porous surface. The black core observed in LWA shown in Figure 3 (right) indicates that reducing condition occur during firing.

    Figure 4 shows scanning electron microscope (SEM) images of a typical clay LWA granule showing the ideal microstructural features and pore structure. Figure 4a shows the

high total porosity in the core. Figure 4b shows approximately spherical pores ranging in size from 1 μm to 50μm. Figure 4c and 4d show reducing pore size towards the surface where the non-porous vitrified regions form due to the temperature gradient between the core and the surface.

If the temperature is excessively high during sintering, excess gas may be generated, increasing total porosity and producing continuous pores [35]. In addition, further increases in temperature above the pyro-plasticity range can result in viscous flow, reducing the porosity and pore size [36]. The heating rate also influences the microstructure of clay LWAs. If the heating rate is low the material may be incompletely vitrified allowing any gases generated to escape without causing bloating.

Rapid cooling may also produce micro-cracks between the core and the surface of the LWA. Under these conditions the LWA shell and core may experience different thermal contractions which can induce stress in the shell, producing micro-cracks that reduce strength. It is reported that this effect can be avoided in LWAs manufactured from clay using slow cooling (0.7 °C/min) and this resulted in 114% increase in strength [37]. The influence of cooling on the strength of LWA manufactured from clay and ferrochrome sludge has been reported and a relationship found between the pore structure and cooling rate [38]. The total porosity and average pore size were found to be higher in rapidly cooled LWAs compared to the same composition subjected to slow cooling. The reduced average pore size allowed slow-cooled granules to form a continuous framework structure that had higher compressive strength compared to rapidly cooled LWA in which cracks were observed to develop from large pores.

The dwell time at high temperature also influences the LWA pore structure. Increasing the dwell time has been reported to increase the total porosity but decreases the number of pores [39]. This change in pore structure is due to increasing pore connectivity. A uniform pore size

distribution would characterise an ideal clay LWA pore structure. This can be achieved by homogenization of raw materials with techniques such as milling and sieving. However, the effect of processing parameters on the pore structure of LWAs manufactured from clays has not been extensively studied.

(iii)   Effect of pore structure on mechanical properties of clay LWA

The degree of connected porosity linked to the surface determines the water absorption capacity of LWA [11]. Poor sintering is the main cause of high open porosity and high water absorption capacity in LWA [40]. High total porosity and large pores reduced the compressive strength of LWA made of silt-clay waste [41]. The reduction in strength due to increasing porosity is based on the relationship between strength and density. A uniform pore size can increase the compressive strength of clay LWAs because it promotes a more homogenous stress distribution throughout the microstructure [31,42]. However, research to date has not adequately investigated the effect of pore structure on the strength of clay LWAs.

The particle density and water absorption data for the LWAs of Table 1 are plotted in Figure 5. The water absorption for most manufactured LWA is below 20% and the particle density ranges between 0.50 and 2.00 g/cm³. Very low particle density (<0.5 g/cm³) and more than 50% water absorption were observed in LWA prepared using smectite-rich clay [43]. Clay LWAs with high water absorption (>50%) and particle density around 1.00 g/cm³ were manufactured from batches containing high quantities (up to 75% by weight) of sewage sludge in which open porosity was formed as the sludge combustion gas formed open channels that were connected to the granule surface [44].

The particle density and water absorption of three commercial LWAs, Lytag (raw material: PFA), Trefoil (raw material: PFA, sewage sludge and clay) and LECA (raw

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material: clay) are included in Figure 5. In many cases, water absorption of clay LWA were lower than for Lytag and Trefoil. This indicates that high-quality LWA with low particle density and water absorption comparable to commercial products can be manufactured from various types of clay containing materials.

2.2 Mineralogy of clay LWA

Table 2 summarises X-ray diffraction (XRD) data showing the crystalline phases present in clay and clay derived LWA products, indicating the mineralogical changes that occur during sintering. Natural clay deposits typically contain smectite, kaolinite, chlorite and illite together with some non-clay components such as quartz, feldspars and calcite. Other phases associated with clay mineral such as muscovite and vermiculate have also been identified. Most clay minerals have demonstrated bloating and can be used to form LWA [8,12]. Illitic clays are reported to be more effective at trapping CO_2 from carbonate decomposition than kaolinitic clays [29].

The neo-formed minerals in Table 2 show that most of the initial phases transform as a result of thermal treatment. The amorphous phase in clay LWAs can have different origins in addition to the clay component, such as the opal phase in clay-rich diatomite [45] or the amorphous phase added to the mix as a secondary material [46]. The amount of amorphous phase has also been reported to increase at higher temperatures as demonstrated when reservoir sediments were used as the raw material [7].

Among the three main end members of feldspars, anorthite has been attributed to high content of CaO that remains after decomposition of carbonates. Ca^{2+} ions are capable of substitution into the aluminosilicate matrix and this allows the formation of anorthite [47]. This was observed in LWA manufactured from water reservoir sediments when CaO was

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added to the mix [7]. Albite, the Na end-member of the plagioclase division, has also been reported to form in LWA manufactured from reservoir sediment as a result of the addition of NaOH [13].

Research results have also linked the presence of certain minerals to the physical properties of LWAs and compressive strength, in particular. For example, in LWAs manufactured from high carbon ferrochrome slag, the forsterite phase that belongs to the olivine group of nesosilicates, influences strength [38]. Forsterite has a poor resistance to thermal shock and if this phase is neo-formed, the internal stresses during cooling can lead to micro-cracks that reduces compressive strength. Conversely, neo-formed mullite, the main product of illitic and kaolinitic clay thermal decomposition, has been associated with strength development [48]. However, the effect of neo-formed phases on the physical properties of LWA have not been extensively studied.

3.   Compatibility of clay with secondary materials

An important economic aspect of LWA production is the potential to incorporate secondary materials, which are often wastes, as partial replacements for clay. Leachable heavy metals and other soluble constituents present in the waste can be encapsulated in a silicate-based matrix or substitute other ions in the crystal structure during sintering at high temperature, making them non-leachable [49]. This method has been presented as a recycling option for a number of secondary materials including sewage sludge, waste polishing residues and waste glass. In some cases, hazardous waste such as energy-from-waste (EfW) residues have been used, as shown in Table 1.

Despite the success in manufacturing LWAs from clay and wastes that meet regulatory requirements, most of the reviewed studies have not investigated the long-term risk

associated with this approach. Many studies have used the toxicity characteristic leaching procedure (TCLP) or the single batch BS EN 12457 leaching test which are not appropriate for characterising long-term leaching. These tests fail to take into account the conditions of different applications and therefore they should be used with other test methods such as the pH dependence leaching tests [50]. For instance, synthetic aggregates may have a poor buffering capacity which makes them sensitive if they are exposed to, or used in, acidic or alkaline conditions [51].

Thermal treatment generates vitrified (glassy) phases that can provide physical encapsulation and limit the exposure of leachable constituent to the environment. However, that is not always the case, as some constituents such as Mo and As may transform into more mobile chemical forms on heating [52]. A study of the leaching behaviour of clay LWA incorporating coal fly ash showed that heavy metals such as Cr, Ni, Mn and Zn can form part of neo-formed spinel groups and feldspar crystalline structures, and Pb and Cd can react with SiO_2 (in phyllosilicates) and enter the amorphous phases [53]. This study reported that divalent and trivalent ions such as Mn and Cr can partially replace Mg^{2+} and Al^{3+} and/or Fe^{3+} in the spinel and/or magnetite series. In a similar study on stabilization of heavy metals in LWA incorporating sewage sludge and river sediments, it was shown that under oxidative conditions, Al^{3+} can replace tetrahedral Si^{4+}, producing an additional negative charge in the network that can be balanced by heavy metal cations such as Cd, Cr and Cu [54]. The study also related leaching characteristics to the degree of sintering. The authors reported that higher Fe_2O_3 content was beneficial for solidification of Cd, Cu and Pb, as Fe_2O_3 reduces the eutectic point, promoting liquid phase sintering and reducing the permeability of the alumina-silicate matrix.

4. Embodied carbon dioxide in clay derived LWA

The embodied carbon (ECO_2) in clay LWA comes mainly from the carbon dioxide associated with excavation and transportation of the raw materials and the manufacturing process. These need to be taken into account for a comprehensive and accurate estimation of the ECO_2 [55]. Although LWA manufacture uses energy and emits CO_2, the use of waste clay to manufacture LWA can be associated with significant CO_2 saving.

Transportation of waste clay to a LWA manufacturing plant (or building a plant in the vicinity of waste clay stream) will be highly site specific. The use of LWA to replace normal weight aggregate in pre-cast concrete products is associated with additional carbon savings associated with more efficient component transportation due to reduced weight [56]. However, if calculations are based on concrete volume (cubic meter), then minimal difference in ECO_2 are reported [19].

Sintering is the most energy intensive stage in the LWA manufacturing process. Clay LWA manufacture has been estimated to emit ~0.22 tonne of CO_2 per tonne of aggregate [57]. This compares favourably with the emissions from Portland cement production, where ~0.83 tonne of CO_2 is generated per tonne of cement. Energy for LWA production can be provided from biomass combustion given the relatively low sintering temperature required. A number of studies have used organic matter such as sewage sludge in the mix as an energy source [20,54,58]. In addition, the use of waste clay derived LWA improves thermal insulation properties of concrete, reduces permeability and reduces structural dead load, allowing the construction of larger buildings with the same foundation size, all of which are associated with reduced CO_2 emissions.

5.   Conclusions

The potential environmental impacts associated with reuse applications for waste clay and the high cost of landfill should ensure that infrastructure development projects increasingly consider LWA production from waste clay. This paper has reviewed the literature relevant to this reuse application. LWA production from clay can be commercially viable, but control of the manufacturing process and particularly the bloating process is essential to produce an ideal LWA pore structure. The pore structure in LWA is characterised by the total porosity, pore connectivity and the pore size distribution and these all relate to the final LWA properties of particle density, water absorption and compressive strength. Using waste clay as a raw material for LWA production can provide CO_2 savings and can allow additional secondary materials to be beneficially used in the manufacturing process. The long-term leaching implications of incorporating waste materials in LWA need to be fully investigated.

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Table 1

Lightweight aggregate research studies using clay that have been reviewed.

Raw Material	Secondary material incorporated	Reference
Clayey diatomite rock	Sawdust	[1]
Clay	CaF_2 sludge	[2]
Smectite-rich clay stone	Sand	[3]
White clay and shale	MSWI* fly ash	[4]
Clay	Sewage sludge	[5]
Reservoir Sediment	-	[6]
Clay	Granite polishing residue	[7]
Silt-clay waste	CC** fly ash	[8]
Silt-clay waste	Sewage sludge	[9]
Reservoir Sediment	MSWI fly ash	[10]
Reservoir sediment	-	[11]
Reservoir sediment	-	[12]
Harbour sediment	Waste glass	[13]
Reservoir Sediment	MSWI fly ash	[14]
Reservoir sediment	-	[15]
Clay	-	[16]
Silt-clay waste	CC fly ash	[17]
Clay	-	[18]
Clay	-	[19]
Clay	APC*** residues	[20]
Clay	APC residues	[21]
Clay	CC fly ash	[22]
Clay	FeCr slag	[23]
Clay	Granite	[24]
Silt-clay waste	Sewage sludge	[25]
Clay	Bauxite (red mud)	[26]
Clay	Sewage sludge	[27]
London clay	-	[28]

* Municipal solid waste incineration, ** Coal combustion and *** Air pollution control residues.

Table 2
XRD analyses for the reviewed manufactured clay LWAs and the corresponding raw materials. "m" indicates minor phase detected.

| Materials	Initial phase															Final phase										
	Quartz	Kaolinite	Smectite	Illite	Vermiculite	Muscovite	Chlorite	Feldspars	Amphibole	Magnetite	Opal	Wustite	Hematite	Corundum	Calcite	Glass	Quartz	Feldspars	Mullite	Hematite	Albite	Anorthite	Hercynite	Sillimanite	Spinel	Forsterite
Clay-rich diatomite [1]	m		×	m	m		m	m			×					×	×	m	m	m				m		
Clay and granite [7]	×		×			×		×									×		×						×	
Reservoir sediment with added CaO [11]	×					×	×										×				×	×				
Harbour sediment and glass [13]	×							m				×		×			×									
Reservoir sediment with added NaOH [15]	×					×	×	×									×			×	×		×			
Clay [19]	×			×		×	×	×	×				×				×	×		×			×			
Clay and APCr [20]	×	m	m	×				m					m		m											
Clay and FeCr slag [23]	×					×										×									×	×
Clay and granite waste [24]	×	×	×					×		m						×	m	m								
Silt-clay waste and sewage sludge [25]	×	×		×												×	×		×							
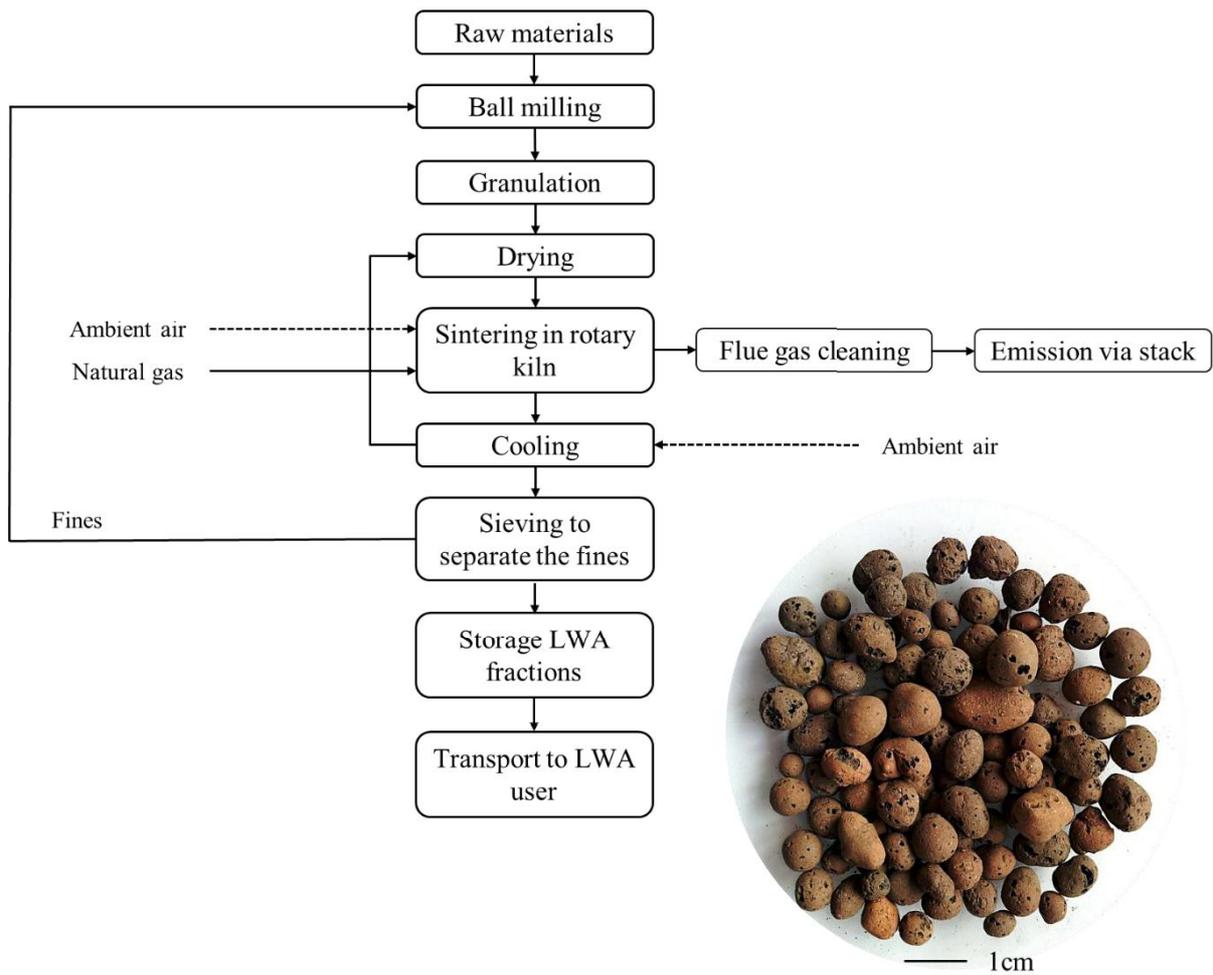
Fig. 1. Lightweight aggregate (LWA) manufacturing process flowchart and typical clay LWA end product [1].
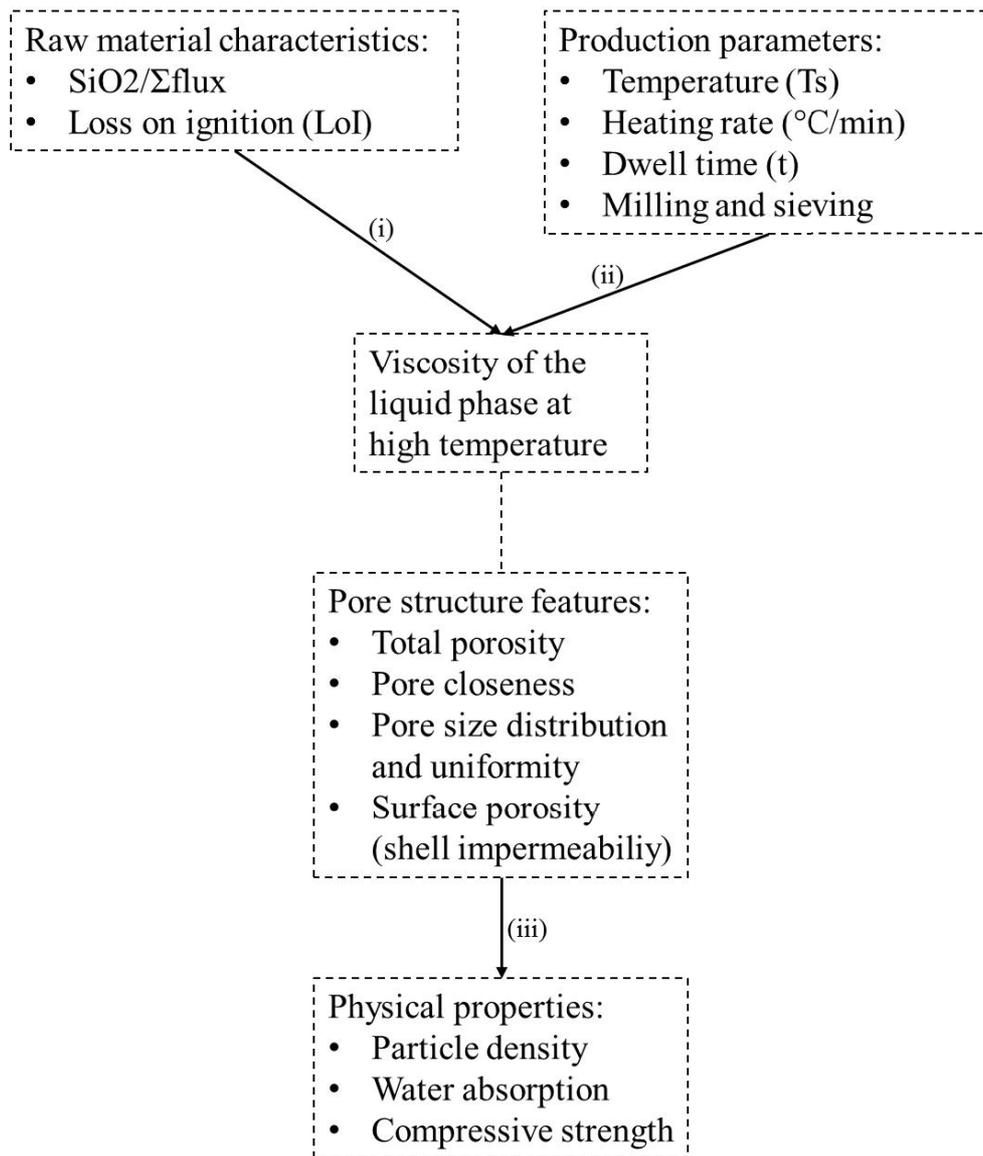
Fig. 2. Relationships between raw material characteristics, production parameters and mechanical properties, showing the pore structure as an intermediate variable.
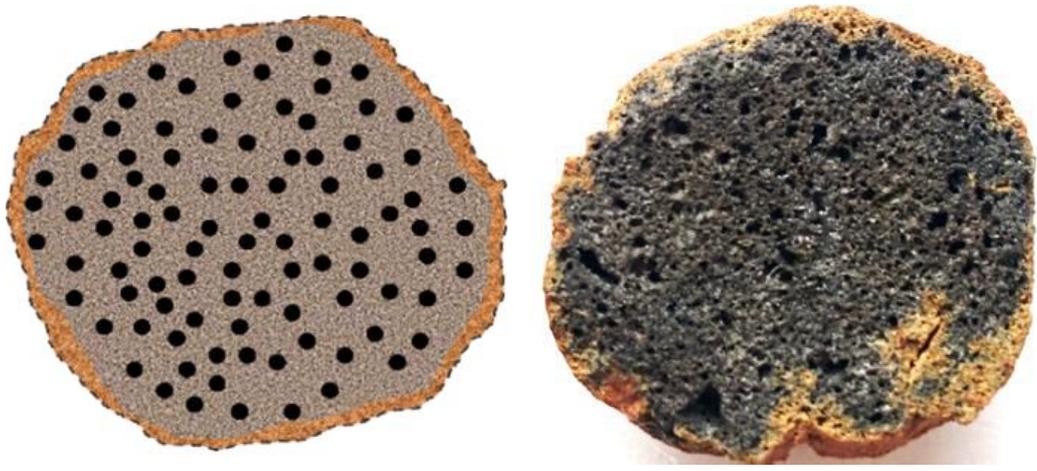
Fig. 3. Schematic of an ideal pore structure (left) and typical pore structure of clay LWA (right).
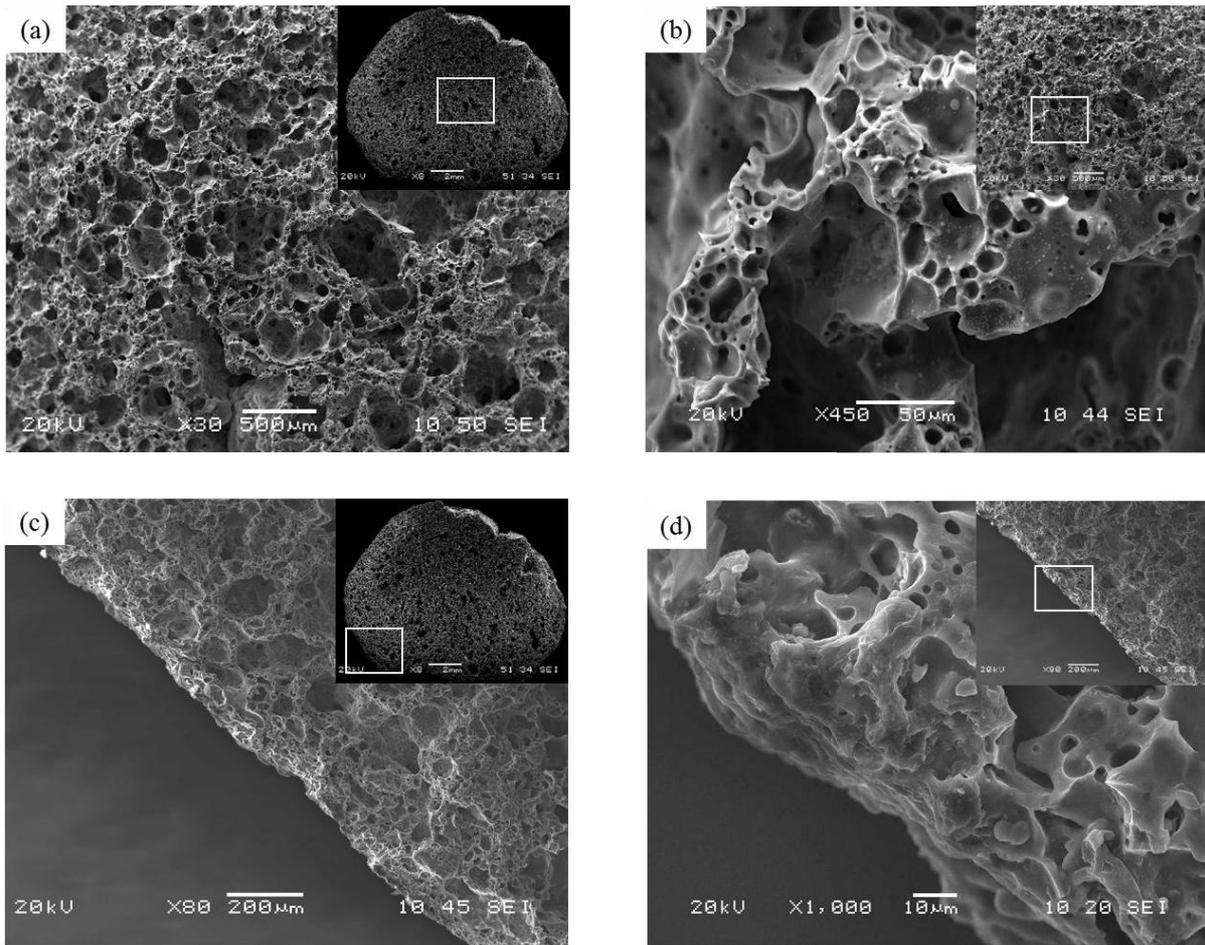
Fig. 4. SEM micrographs of fracture surface of a clay LWA granule, taken from Sustainability Research Institute SEM archive (unpublished data): (a) LWA core pore structure, (b) core at a higher magnification, (c) LWA shell cross-section and, (d) shell cross section at a higher magnification.
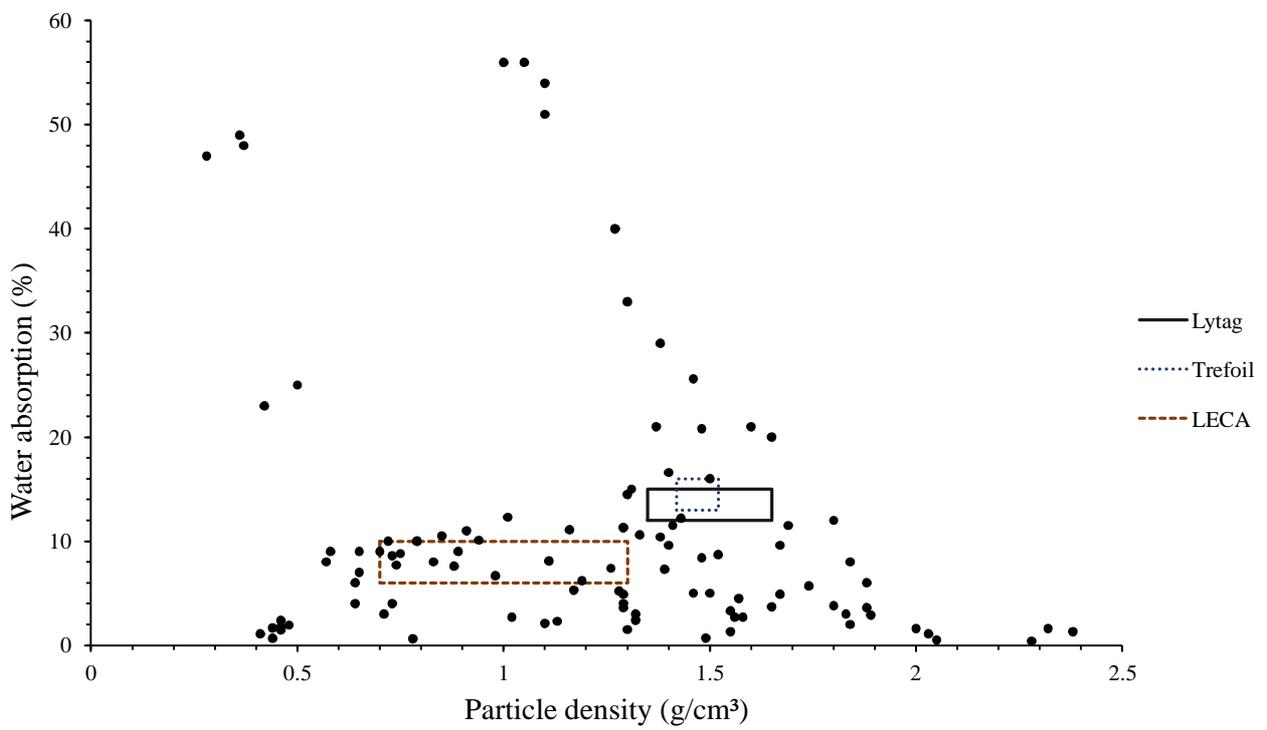
Fig. 5. Relationship between particle density and water absorption of the reviewed manufactured LWAs. Lytag: straight black line, Lightweight expanded clay aggregate (LECA): brown dashed line, Trefoil: blue dashed line.