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Lightweight aggregates produced by granulation of peat-wood fly ash with alkali activator

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article info abstract

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This study presents a new method to produce lightweight aggregates (LWAs) by granulating peat-wood ash using alkali activators. Peat-wood ash was granulated with potassium silicate and sodium aluminate in a highshear granulator to produce spherical granules. Blast furnace slag, coal fly ash and metakaolin were studied as co-binders. A suitable liquid/solid ratio, granulation growth type, microstructure, strength, and crystal structure of granules have been determined. The granulation proceeded by induction-type growth behaviour and suitable liquid/solid ratio was between 0.34 and 0.43. In the BSE images it was observed that the precursor particles were embedded in a dense alumino-silicate matrix. The X-ray diffraction analysis supported the success of the alkali activation. All co-binders increased the strength of the granules, but the blast furnace slag produced the strongest granules. The study shows that by simultaneous granulation and alkali activation it is possible to increase the utilisation of ash and produce valuable products.

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1. Introduction

The European Union has set a target to decrease $CO₂$ emissions and increase the use of renewable energy sources up to 20% by 2020 [\(European Commission Europe 2020 targets](#page--1-0)). Biomass, such as wood, is considered as carbon neutral because it binds the same amount of $CO₂$ when growing as is released in combustion. In Finland, wood is often co-combusted with peat in biomass burning facilities, such as power plants and paper mills. As a by-product, 600,000 tons of peatwood ash is generated just in Finland every year [\(Emilsson, 2006\)](#page--1-0).

One way to utilize ash and other waste materials is to produce lightweight aggregates (LWAs) that can be used in lightweight concrete or in civil engineering. Artificial LWAs made from waste by granulation, agglomeration or pelletization are ecologically sound since natural aggregates are saved and damaging activities of aggregate mining are prevented. LWAs are also cost efficient because waste materials are turned into products that can be sold. Different waste materials such as ash ([Anagnostopoulos and Stivanakis, 2009; Arslan and Baykal,](#page--1-0) [2006; Cheeseman et al., 2005a; Cheeseman and Virdi, 2005b; Geso](#page--1-0)ğlu

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[et al., 2012; González-Corrochano et al., 2009a; Huang et al., 2007](#page--1-0)), sludge ([Huang et al., 2007; González-Corrochano et al., 2009a, b\)](#page--1-0), and slag (Gesoğ[lu et al., 2012\)](#page--1-0) have been studied as LWA raw materials. In order to produce strong LWAs, either ordinary Portland cement or high sintering temperature is required. Both options increase the costs of the LWAs and cause $CO₂$ emissions.

An interesting option to bypass the usage of cement and hightemperature sintering is alkali activation. Alkali activation of coal fly ash has been intensively researched during the last few decades and has been comprehensively reviewed in a state-of-the-art report by Provis and van Deventer ([Provis and van Deventer, 2014a](#page--1-0)). Results [\(Provis and van Deventer, 2014a\)](#page--1-0) show that alkali-activated materials (AAMs) offer excellent properties compared to ordinary Portland cement, such as lower $CO₂$ emissions, excellent compressive strength, and thermal resistance. In addition, immobilisation of heavy metals in AAMs has been reported [\(Ogundiran et al., 2013; van Jaarsveld et al.,](#page--1-0) [1997, 1998; Zhang et al., 2008](#page--1-0)) which means that this way even the fly ash fractions that contain toxic elements could be utilized.

The objective of this study was to make granules from peat-wood ash using an alkali activator as the liquid binding phase to glue the primary particles together in a permanent alumino-silicate matrix. The intention was to produce spherical granules using a high shear granulator and appropriate granulation conditions. To the best of our knowledge, no previous studies have used the approach described herein to produce LWAs. We studied the effect of different alkali activators and co-

Abbreviations: AAM, alkali activated material; BSE, back-scattered electron; C, coal fly ash; KSil, POTASSIUM silicate; M, metakaolin; Na-Alu, sodium aluminate; P, peat-wood fly ash; S, ground granulated blast furnace slag.

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binders on the strength of the granules. The paper aims for a proof of principle and focuses on the granulation mechanism and the extent of the alkali activation reaction.

2. Material and methods

2.1. Materials

Peat-wood fly ash (P) was collected from a power plant that uses a circulating fluidised bed boiler. The proportion of wood in the fuel mix was approximately 30%. Ground-granulated blast furnace slag (S) (ORCEM, the Netherlands), coal fly ash (C) and metakaolin (M) were used as co-binders, and their effects on granulation and alkali activation were studied. The coal ash was from a pulverised fuel power plant. Metakaolin was obtained by thermal treatment of kaolin (Sigma Aldrich) at 750 °C for 2 h.

Potassium silicate (KSil) solution was chosen as an alkali activator for its known ability to produce strong AAMs, especially with coal fly ash and slag [\(Nugteren et al., 2009\)](#page--1-0). The solution was prepared by mixing a commercial KSil solution (Kasil 2135; PQ Europe) with potassium hydroxide to obtain $K_2O/SiO_2 = 0.8$ (65 wt.% of H₂O). Sodium aluminate (Na-Alu) was also used as an alkali activator. Na-Alu solution is available as a waste product. If it could be used as an alkali activator, the cost of producing AAMs could decrease. The Na-Alu (Sigma-Aldrich) solution was dissolved in sodium hydroxide-solution to yield a total molar ratio of Na₂O/Al₂O₃ = 2.2 (60 wt.% of H₂O). To evaluate the success of the alkali activation, the granulation was also prepared with H_2O .

2.2. Granulation

Nine batches with different compositions were prepared (Table 1). Peat-wood fly ash was granulated using three different liquids: KSil, Na-Alu and H_2O . The effects of slag, coal fly ash and metakaolin as cobinders were studied by adding them in 20 wt.% and 40 wt.% proportions to peat-wood fly ash. KSil was used as an alkali activator with the co-binders. The materials were blended prior to granulation manually for 30 s and then inside the granulator with an impeller for 30 s.

A high-shear granulator (Eirich R-02) was chosen for this study due to its known ability to spread viscous liquids, to process sticky material and to produce more compact granules than low-shear granulators [\(Reynolds et al., 2007\)](#page--1-0). A schematic of the granulator is presented in Fig. 1. The granulator has a rotating drum with an impeller (14 cm diameter) inside that spins in the opposite direction to that of the drum. The volume of the drum was 13 dm³, the tilt angle was 33 $^{\circ}$, and the drum rotating speed was 44 rpm. Preliminary experiments were made to determine a suitable liquid/solid-ratio (w/w) for the material.

Abbreviations: peat-wood fly ash (P), blast furnace slag (S), coal fly ash (C), metakaolin (M), potassium silicate (KSil) and sodium aluminate (Na-Alu). The numbers in the codes represent weight percentages of the precursors.

Fig. 1. A schematic of the granulator. The impeller (14 cm diameter) inside spins in the opposite direction to that of the drum. The volume of the drum was 13 dm³, the tilt angle was 33°, and the drum rotating speed was 44 rpm. The binder was added by drops from a flask through a tube on to the powder bed.

The granulation process was carried out using the following method:

- 1) Dry precursors were weighed and mixed carefully prior to the granulation.
- 2) Mixed material was added to the drum, and the drum and impeller were switched on.
- 3) Liquid was added by drops on to the powder bed rotating inside the drum with a spray flask from the hatch of the granulator.
- 4) Process was continued until the granules stopped growing.
- 5) Each batch was sealed in an air-tight plastic bag and stored at room temperature until the analysis of the granules.

2.3. Analysis methods

The chemical composition of the precursors was determined with Xray fluorescence (XRF) from a melt-fused tablet. The particle size distributions were measured with a Beckman Coulter LS 13320 and reported as volumetric-based size (d_{10} , d_{50} and d_{90}).

The crushing strength of the granules after 28 days was measured with a Zwick, Z100 Roell testing machine. TestXpert II-software was used to determine the crushing force of the granules. The pre-load force was 5 N, and the compression speed was 0.01 mm/s. Similar single granule crushing tests for LWAs have been performed by ([Arslan and](#page--1-0) [Baykal, 2006; Cheeseman et al., 2005a; Cheeseman and Virdi, 2005b;](#page--1-0) Gesoğ[lu et al., 2012; González-Corrochano et al., 2009a](#page--1-0)).

Loose bulk density and the granule size distribution were determined with standards ([SFS-EN-1097-3, 1998\)](#page--1-0) and [\(SFS-EN 933-1,](#page--1-0) [2012](#page--1-0)) respectively.

A Siemens 5000 X-ray diffractometer with CuKα radiation (40 mA and 40 kV) and a graphite monochromator was used to identify the main crystalline phases of the powdered samples. The step interval, integration time and angle interval used were 0.04°/step, 2.5 s/step and 10–70°, respectively. The ICDD database was used for identification of crystalline phases ([The Powder Diffraction File, 2006](#page--1-0)).

The cross-sections of three granules (5–6 mm diameter) from each batch were analysed with a field emission electron microscope (FESEM) (Zeiss Sigma). The granules were impregnated in the epoxy resin. After curing for 24 h, 2 mm slices were cut and placed in a 25 mm diameter plastic mould. The slices were then impregnated in the epoxy resin and left to cure for 24 h. Hardened samples were then grinded, polished and coated with carbon to obtain an optimal surface

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