

## Aggregate production from burnt oil shale and CO<sub>2</sub> – an Estonian perspective

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**Abstract.** Estonia's main calcareous industrial waste output, such as burnt oil shale (BOS) from Narva Power Plants (PPs), cement kiln dust (CKD) from AS Kunda Nordic Tsement and municipal solid waste incineration (MSWI) residues from Iru Power Plant (PP), were all tested in a novel mixer-type granulator-carbonator for the production of constructional aggregates, with a focus being placed on the mechanical properties, the CO<sub>2</sub> binding ability, and leaching behaviour. The mixtures of wastes after the addition of cement and quarry fines gave aggregates which exhibited satisfactory mechanical and leaching properties for further utilisation. The results of this study could serve as a basic research step towards closing the waste cycle in Estonian energy and cement sectors.

**Keywords:** burnt oil shale, municipal solid waste incineration ash, cement kiln dust, mineral carbonisation, granulation.

### 1. Introduction

The largest proportion of all industrial waste in Estonia is generated by the oil shale-based energy sector in the form of oil shale ash [1]. In 2016 approximately seven million tons of ash, according to Eesti Energia AS, was deposited in ash fields [2]. However, the target is to increase the utilisation of ash in the nearest future [3]. The main application areas for burnt oil shale (BOS) include the production of building materials (as an aggregate in cement, or autoclaved aerated concrete blocks, or as an additive in concrete and dry construction mixtures (EVS 927:2017 [4])), road and port construction, neutralisation of acidic soils, and as a binder of CO<sub>2</sub> and sulfur that are released during energy production [5]. The ash that is produced by the cement industry and the municipal solid waste incineration (MSWI) field generate another significant

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waste stream [1]. Berber et al. [6] showed that accelerated carbonation technology could be an economically sound solution for stabilising the MSWI fly ash from Iru Power Plant (PP). However, currently the harmful ash from Iru PP cannot be directly disposed of in the landfill but must be transported to a hazardous waste management company [7]. Thermal power plants (PPs) and cement plants are also amongst the biggest CO<sub>2</sub> emitters [2, 8, 9]. Closing the waste loop requires the use of millions of tons of ash in which BOS constitutes the major proportion. By applying the agglomeration process, such waste could be turned into a product to be used either in the construction or agricultural industry. Previous practice has shown that BOS and cement kiln dust (CKD) could be utilised as liming agents for acidic soils [10–12]. However, in practice, the utilisation of these forms of waste is limited in Estonia.

Alternatively, the above waste could be utilised to produce construction aggregates by using the accelerated carbonation-based solidification [13]. Specifically, Gunning et al. [13] showed that the key properties of the carbonated aggregates that were being produced by the above-mentioned technology were similar to those of lightweight aggregates (LWA) which were available in the UK in 2009. The production of artificial carbonated aggregates has been demonstrated by Carbon8, a spin-off company of the University of Greenwich in England which uses mineral carbonation treatment to produce artificial aggregates from several residues, such as MSWI ash, quarry fines, etc., for further utilisation in the manufacturing of carbon neutral building blocks (CO<sub>2</sub>, sand, and cement) by Lignacite in the UK [14].

The current study was carried out in light of the targets set in the Estonian National Waste Management Plan 2014–2020 [15] and the Estonian Environmental Strategy 2030 [3]: 1) to promote waste recovery; 2) to reduce by 2030 waste dumping by 30% in addition to alleviating the environmental hazard posed by generated waste; 3) to abate greenhouse gas emissions and save resources. Therefore, this study could be a crucial step in the process of achieving all of the main targets by promoting waste recovery and abating CO<sub>2</sub> emissions from the power and cement sectors, as well as from MSWI plants, and, by this means spare raw materials and endorse the waste-to-product (WtP) approach.

All in all, this work could be heralded as the first study in Estonia of the variety of industrial waste, including BOS waste from Narva PPs and shale oil production (Enefit 280), CKD from AS Kunda Nordic Tsement and MSWI air pollution control (APC) residue from Iru PP, for the production of construction aggregates. This study focused on a novel mixer-type granulator-based technology, which permitted the carrying out of hydration, carbonation and granulation steps in the same reactor. The mechanical properties of produced aggregates, the CO<sub>2</sub> binding ability of the above wastes and reduction of the environmental hazard caused by them were covered as well. The specific aim was to study the applicability of this method.

## 2. Materials and methods

### 2.1. Materials

Different solid wastes from power and cement plants were utilised in order to produce construction aggregates via the carbonation-based solidification technique. The BOS1 and BOS2 markers represent the ash that is produced in electrostatic precipitators and BOS5 stands for total ash silo in the circulating fluidised bed boilers of Narva PPs. The BOS3 sample was collected from an SO<sub>2</sub> removal system (Novel Integrated Desulphurisation, Alstom Power) that is part of a pulverised firing boiler. The BOS4 sample was collected from Enefit280 units used in shale oil production. The solid waste from other industries, i.e. MSWI APC residues from Iru PP and CKD from Kunda Nordic Tsement, were also tested. The additives – Portland cement (CEM I, 42.5R) and quarry fines – were provided by AS Kunda Nordic Tsement.

### 2.2. Methods

The materials under study were analysed using X-ray fluorescence spectroscopy (XRF, Rigaku Primus II) and quantitative X-ray diffraction (XRD, Bruker D8 Advanced) at the Institute of Geology, University of Tartu (UT). The contents of free lime (fCaO) (the ethylene glycol method [16]), sulfate [17] and total carbon (TC) (Electra CS – 580 Carbon/Sulfur Determinator) were determined. In addition, the BET specific surface area (SSA) and the particle size distribution were determined using a Kelvin 1042 sorptometer and a Horiba laser scattering particle size distribution analyser LA-950, respectively.

Figure 1 shows the experimental setup. The hydration, carbonation and mixing with the additives (step 1), followed by granulation (step 2), were all carried out by Eirich GmbH in a Co EL1-type intensive mixer/granulator operated under different regimes. In the first step, the rotational speed of the rotor tool was set at 3000 RPM for intensive mixing, in a clockwise direction, whereas it was set at 610 RPM in an anti-clockwise direction throughout the second step of granulation. The mixing pan was operated at slow speed (85 RPM) throughout the experiment. Granulation is the agglomeration of powdered material particles into granules of a specific size and spherical shape. The granules structure is formed either by physical forces or through

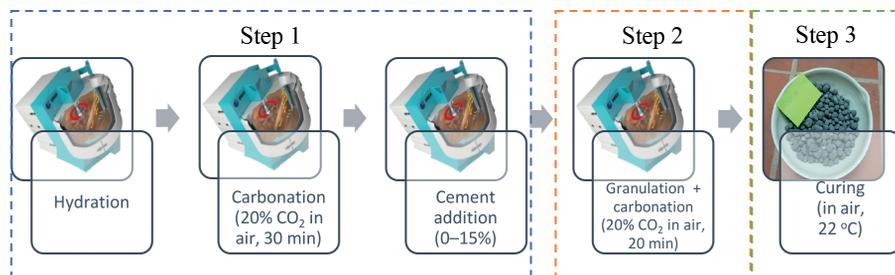


Fig. 1. Experimental setup.

the use of binders. The granulation process is controlled by the physico-chemical properties of the powdered material, and by moisture content, particle size, operating conditions, and process parameters such as the rotary speed of the granulator [18, 19]. The waste was treated individually as well as in the composition of mixtures (using 0–15% cement and 0–30% quarry fines as additives), under different operating regimes, by varying the solid-to-liquid ratio, the rotation speed, the CO<sub>2</sub> content in model gas, residence time, curing conditions, etc. It is important to note here that the CO<sub>2</sub> concentration in the model gas was determined as being at 20% since in the cement industry's typical flue gas stream, it varies between 14 and 33% [20]. The experiment plan and process parameters are given in Table 1.

**Table 1. Experimental plan and process parameters**

Granule	Initial waste material	OPC, %	Quarry fines, %	L/S, w/w
gBOS1	BOS1	5	0	0.45
gMIX1-0	4BOS1+APC+CKD mixture	0	0	0.30
gMIX1-9		9	0	nd
gMIX1-10-q		10	30	0.22
gBOS2	BOS2	5		0.42
gMIX2-0	4BOS2+APC+CKD mixture	0	0	0.38
gMIX2-5		5	0	0.32
gMIX2-10		10	0	0.30
gBOS3	BOS3	5		0.23
gMIX3-15	4BOS3+APC+CKD mixture	15	0	0.17
gMIX3-5-q		15	30	0.16
gMIX3-10-q		10	30	0.15
gMIX4-5	4BOS4+APC+CKD mixture	5	0	0.25
gMIX4-10		10	0	0.25
gMIX4-10-q		10	30	0.18
gBOS5	BOS5	5		0.34

OPC – ordinary Portland cement; nd – not determined; 4BOS+APC+CKD stands for 4:1:1 proportion.

The granules were dried in a thermostat for three hours at 105 °C and analysed for TC and free lime content as the main indicators of the carbonation process. The efficiency of the carbonation process was described by the CO<sub>2</sub> binding degree (BD<sub>CO<sub>2</sub></sub>). BD<sub>CO<sub>2</sub></sub> shows how much of the theoretical CO<sub>2</sub> binding capacity is being utilised (Eq. (1)):

$$BD_{CO_2} = \frac{CO_2 \text{ granules}}{CO_{2 \text{ max ash}} + CO_{2 \text{ max cement}} + CO_{2 \text{ max quarry fines}}} \times 100, \quad (1)$$

where CO<sub>2 granules</sub> is the analytically determined CO<sub>2</sub> content of the granules, CO<sub>2 max</sub> is the maximum possible CO<sub>2</sub> content of the granulated mixture of ashes, cement and quarry fines in percentage terms, according to Equation (2):

$$CO_{2 \text{ max}} = \frac{X + CO_2^i}{100 + X} \times 100, \quad (2)$$

where  $X$  is the CO<sub>2</sub> uptake capacity of the initial material when considering free lime, wollastonite, belite, merwinite, alite, and Ca-aluminate, and CO<sub>2</sub><sup>*i*</sup> is the carbon dioxide content of the initial materials.

After one week of curing at room temperature (about 22 °C) the granules were tested for mechanical properties such as fractional composition and compressive strength by using the Brookfield CT3 25K texture analyser according to CEN-EN 1097-11:2013 [21]. The apparent density was measured using a Mettler Toledo density kit AX204 according to EVS-EN 1097-6:2016 [22].

The leaching tests were carried out according to European standard EN 12457-2:2002 [23]. The leachate pH was determined using a Mettler Toledo SevenGo pH meter and the electrical conductivity was determined employing a Mettler Toledo SevenGo Duo Pro. The composition of ash leachates was determined according to DIN EN ISO 10304-1/-2(D19/20, E29) [24, 25].

Environmental analyses were carried out using the methods of DIN EN 16171 [26], DIN EN ISO 10523 [27], DIN EN 27888 (C8) [28], DIN EN ISO 17294-2 (E29) [29] and DIN EN ISO 10304-1 D20 [24] by GBA Gesellschaft für Bioanalytik mbH.

### 3. Results and discussions

Burnt oil shale (BOS1–BOS3) and APC residue contained lime and/or portlandite as the main phases that reacted with CO<sub>2</sub>, to form calcium carbonate (Fig. 2). In addition, APC residue contained water soluble chloride compounds, such as halite, sylvite and calcium hydroxide chloride, which, due to their expansive behaviour that causes the development of both pores and cracks in the microstructure, might be of concern for aggregates produced [30]. Calcium appeared mainly as calcite in CKD and BOS4 samples. Silica was found in samples as quartz and K-feldspar, as well as secondary silicates, which could also bind CO<sub>2</sub>. The selected physico-chemical characteristics of the materials are provided in Table 2.

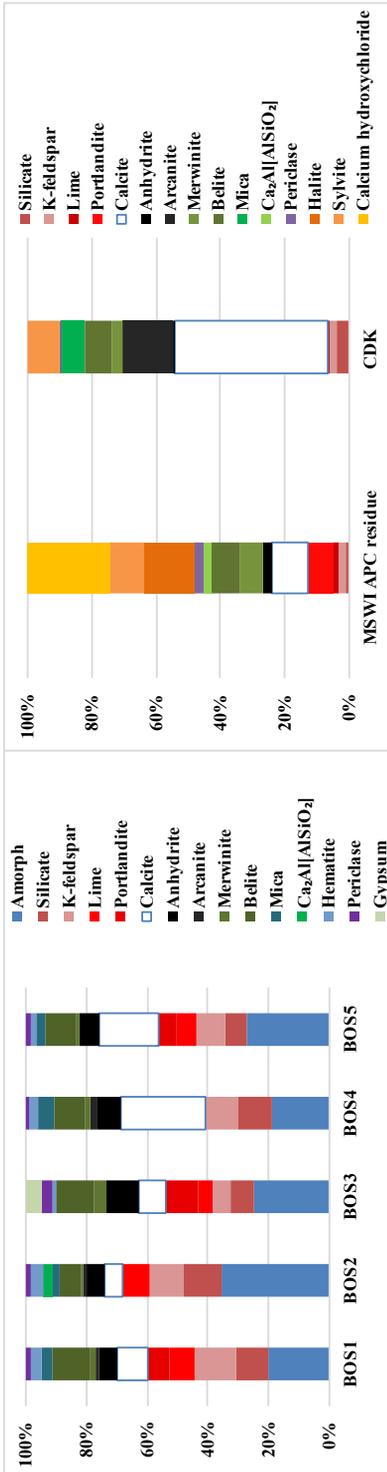


Fig. 2. The phase composition of BOS, MSWI APC residue and CKD.

**Table 2. Physico-chemical characteristics of industrial waste materials**

Oil shale ashes from Narva PPs and Enefit280					
Initial sample	d <sub>mean</sub> , μm	SSA, m <sup>2</sup> /g	fCaO, %	CO <sub>2</sub> , %	S <sub>so4</sub> , %
BOS1	20.99	4.75	12.00	5.31	1.93
BOS2	43.58	3.08	9.76	3.79	2.20
BOS3	14.86	1.27	8.56	4.26	2.42
BOS4	36.48	4.24	0.36	16.83	2.26
BOS5	39.51	3.74	11.63	12.95	2.22
Cement kiln dust and OPC from AS Kunda Nordic Tsement					
CKD	34.33	4.16	2.01	20.30	2.49
OPC	16.76	1.76	2.54	1.80	1.41
Quarry fines	–	4.26	0.03	41.12	–
MSWI APC residue from Iru PP					
APC residue	40.42	7.88	10.02	6.35	2.49

OPC – ordinary Portland cement; “–” represents no data.

The results of the leaching analyses for the initial waste samples are given in Table 3. In the case of APC residue from Iru PP as the most hazardous sample in this study, the leachability of chlorides, Pb and total dissolved solids (TDS) exceeded the limits for hazardous waste, and Hg, Cr, Zn, Cu, Mo and Ba surpassed the target values [31]. The CKD from Kunda Nordic Tsement was characterized by the high leachability of chlorides, sulphates, fluorides, Pb, Cr and Mo. The contents and mobility of heavy metals in other BOS samples were found to be similar.

The experiments indicated that CO<sub>2</sub> uptake as well as the mechanical properties of BOS types, CKD and MSWI APC residue-based aggregates could differ across a very wide range. The CO<sub>2</sub> uptake was mainly attributed to the relatively high content of free lime (10–15%) in BOS1, BOS2, BOS3 and BOS5 ashes from Narva PPs and the MSWI APC residue from Iru PP. At the same time, the fCaO content was negligible in the CKD from Kunda Nordic Tsement and BOS4 ash from shale oil production, which did not favour CO<sub>2</sub> uptake (Table 2).

Tests indicated that the free lime content was exhausted within thirty minutes under conditions that included a small-range water-to-solid ratio (0.2–0.5 w/w) and a gas flow of 20% CO<sub>2</sub> in air (Fig. 3). The fCaO content

**Table 3. The content and mobility of the main ash components**

Component	Content						Mobility, %		
	Limit <sup>1</sup>	Limit <sup>2</sup>	Unit, wt	APC residue	CKD	BOS1	APC residue	CKD	BOS1
SO <sub>4</sub>			g/kg	74.7	73.8	57.9	19.5	72.10	0.0
Ca			g/kg	271.4	252.3	238.5	27.2	10.5	4.6
K			g/kg	50.6	111.7	15.2	92.1	87.6	18.4
Na			g/kg	40.0	2.1	1.4	91.0	81.0	7.1
Mg			mg/kg	14580	12530	20820	–	–	0.01
Mn			mg/kg	571	696	344	–	–	–
Hg	0.5	10	mg/kg	20	0.18	< 0.1	1.1	–	–
Cd	1	20	mg/kg	70	12	0.45	–	–	–
Pb	50	600	mg/kg	1900	1870	70	36.8	1.2	0.3
Cr	100	800	mg/kg	209	31	37	1.2	12.9	0.2
Ni	50	500	mg/kg	80	18	31	–	–	–
Zn	200	1000	mg/kg	8880	773	< 0.1	1.5	0.1	–
Cu	100	500	mg/kg	681	31	16	0.6	–	–
As	20	50	mg/kg	39	13	25	–	0.1	–
Mo	10	200	mg/kg	22	4,3	14	20.5	58.1	2.1
Ba	500	2000	mg/kg	751	76	176	3.9	7	15.3

Limit<sup>1</sup> – target value for industrial land; Limit<sup>2</sup> – limit value for industrial land (both [31]); “–” represents no data.

remained at a higher level in BOS3, which is a pulverised firing ash that is characterised by lower BET SSA levels (Table 2).

In BOS1, BOS2 and BOS5-based aggregates, a CO<sub>2</sub> binding degree of up to 78–81% was realised with a remarkable reduction in free lime content, whereas the reactivity of gBOS3 towards CO<sub>2</sub> was moderate (52% of BD<sub>CO<sub>2</sub></sub>).

Adding cement to the BOS mixture caused a decrease in BD<sub>CO<sub>2</sub></sub> as the calculation (Eq. (1)) also included the cement minerals as CO<sub>2</sub> binders (Fig. 4). The reason behind this finding could be the following. First, the outer layers that were formed by the addition of cement might have ceased the further

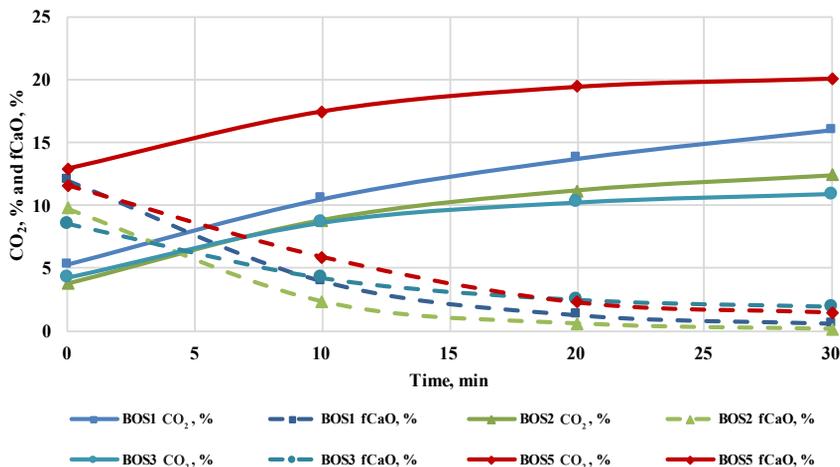


Fig. 3. Changes in CO<sub>2</sub> and fCaO contents in different ashes during the carbonation step.

carbonation of the agglomerated particles in the second step (Fig. 1). Second, the carbonatable compounds inside the cement might not have been exhausted by the 20th minute of carbonation. In addition, the formation of granules decreased further the mobility of reactive species (Ca-silicate phases) owing to that they might have been trapped inside the layers.

The hazardous APC residue and CKD (gMIX1-gMIX4) were included in the mixtures in order to remedy them by capsuling soluble salts (calcium hydroxychloride, halite, sylvite, arcanite (Fig. 1)) and heavy metals into carbonated aggregates. The addition of these forms of waste did not affect the BD<sub>CO<sub>2</sub></sub> (Fig. 4).

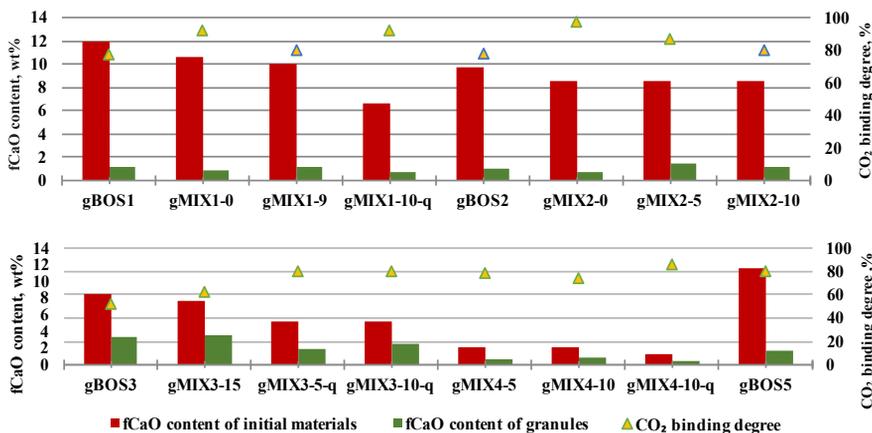


Fig. 4. Changes in free lime content and the resulting CO<sub>2</sub> binding degree of burnt oil shale-based granules.

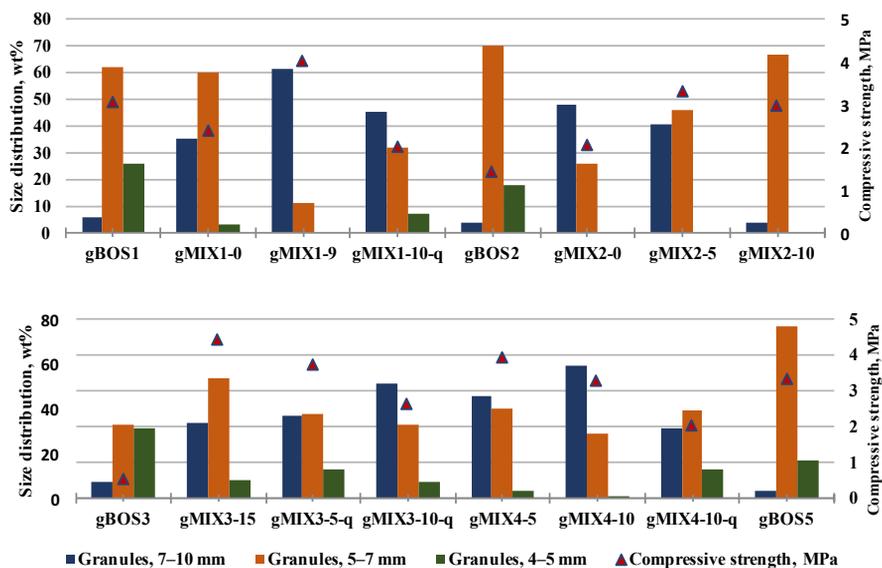


Fig. 5. Compressive strength and size distribution of burnt oil shale-based granules.

A sieve analysis was used to determine the granule size distribution. The results showed that the granules were mainly in the  $4 < x < 10$  mm size range. The compressive strength values were calculated for the  $4 < x < 10$  mm granule size range and varied from 0.5 MPa (gBOS3) to 4.4 MPa (gMIX3-15) (Fig. 5). Compared to BOS alone (gBOS1-5), the mixtures (gMIX1-gMIX4) gave stronger granules. BOS3 afforded the lowest compressive strength, which was attributed to its very slow hardening dynamics [32]. Compressive strength was enhanced by adding 5–15% cement; for example, in the case of gMIX1, adding 9% cement led to a 68% increase in compressive strength. The improvement in the compressive strength values could be the result of several mechanisms, such as the formation of a C-S-H-like gel and calcite in the accelerated carbonation conditions [33], the formation of C-S-H stemmed from the reaction of cement with the interior silicate phases of the ash samples [34], as well as pozzolanic reactions [35]. However, the addition of cement was not always proportional to the gain in the granules strength (gMIX2 and gMIX4), meaning that the effect of the mixture composition could be more complicated. The addition of quarry fines as a neutral filler generally decreased the compressive strength of the granules due to the lower amount of the binder in the mixtures [35]. Expressly, the compressive strength values of gMIX1-10-q and gMIX4-10-q were 60% lower than those observed in the same conditions with no inclusion of quarry fines (gMIX1-9 and gMIX4-10).

### 3.1. The leaching of BOS-based aggregates

Despite the fact that the addition of APC residue and CKD to the mixtures (gMIX1–gMIX4) increased their sulfate content, the leachable content of the sulfates remained well below the hazardous limit (50,000 mg/kg, see Fig. 6). As expected, the high concentration of easily leachable chlorides in the APC residue and CKD (Fig. 2) had a strongly adverse effect on the leaching characteristics of the granules (gMIX1–gMIX4), emphasising the fact that the leachable chloride contents of gBOS1, gBOS2, gBOS3 and gBOS5 were remarkably lower than the limit (20,000 mg/kg). The excessive amount of leachable chlorides and heavy metals in the mixtures could be removed by employing a washing technique [36–38]. Therefore, although the addition of CKD and APC residues rather complicated the leaching behaviour of the obtained granules, the skilful stabilisation of these initially hazardous materials by optimising the mixture composition could provide their rational use.

The addition of cement (5–15%) to the mixtures of different types of waste afforded aggregates that had satisfactory mechanical and leaching properties which were revealed by the decrease of the mobility of heavy metals, chlorides and sulphates. The leaching test indicated that the content of heavy metals such as Hg, Cd, Cr, Ni, Zn, Cu, As, Mo and Ba remained below the hazardous limit in all of the leachates. However, in the case of granules that contained APC residue and CKD, the leachate Pb content slightly exceeded the limit value set for hazardous waste.

One of the main goals in this process was to decrease the alkalinity levels in the initial materials, bringing them down to a pH value less than 11.5 to meet the requirements for inert material (Fig. 7). The granules that were produced from BOS samples (gBOS1, gBOS2, gBOS4, gBOS5) achieved a leachate pH < 11.5. Even though the incorporation of APC residue, CKD and quarry fines in the mixtures played an adverse role in water durability, not to mention the alkaline effect of granules, the leachates for gMIX1, gMIX2 and gMIX4 were characterised by a pH ≤ 11.5. However, the leachates for gMIX3 remained deeply alkaline. Previous studies have also shown that those composites that contained ash from an SO<sub>2</sub> removal system, which were characterised by very

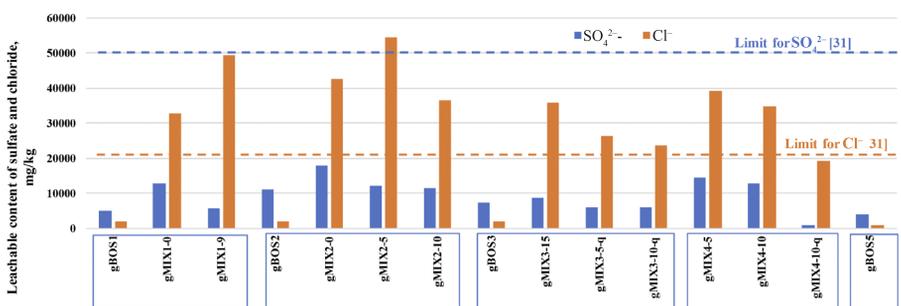


Fig. 6. Leachable contents of sulfates and chlorides.

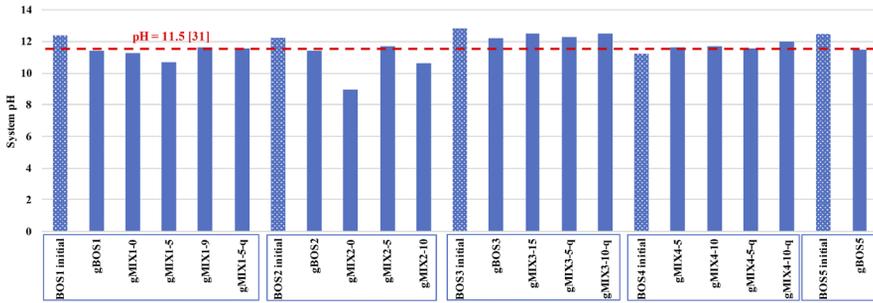


Fig. 7. Leachate pH levels in burnt oil shale samples before (initial) and after accelerated carbonation and granulation.

low compressive strength and water resistance, decomposed in water [32]. The electrical conductivity levels of all types of granules remained below the hazardous limit.

Most of the granules were characterised by an apparent density of nearly 2.1 g/cm<sup>3</sup> (Fig. 8). There was no significant change in apparent density after the leaching test. On the other hand, the compressive strength of the granules was enhanced by leaching. Exposure to water promoted strength development because the delayed reactions might have been inhibited due to a lack of reactive water inside the aggregate [32]. Figure 9 demonstrates the visual features of the granules regarding the shape and the outer surface before and after the leaching test. Most of the granules have a spherical shape without any "blackberry" structure. After the leaching test, the size of the granules decreased owing to the mass loss, whereas the outer surface became smoother.

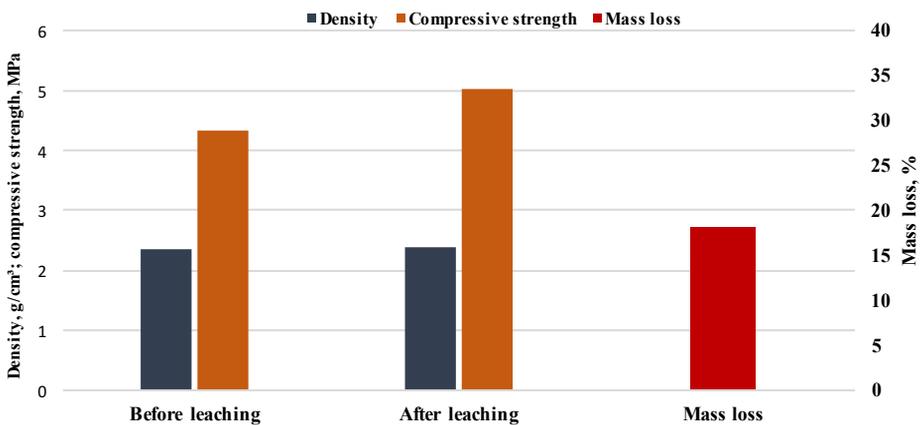


Fig. 8. The density and compressive strength of the granules before and after leaching.



Fig. 9. The granules before and after the leaching test.

#### 4. Conclusions

This paper investigates the possibility of utilising different types of industrial wastes from Estonian oil shale combustion, cement production and municipal solid waste incineration to produce light-weight aggregates as a remedy for the huge volume of CO<sub>2</sub> emissions and vast ash deposits formed in Estonia. Different types of burnt oil shale, cement kiln dust and municipal solid waste incineration air pollution control residues were tested for carbonation-based solidification in a mixer-type granulator. This technique enabled all steps – hydration, carbonation and granulation – to be handled in the same reactor, concentrating on the mechanical properties of the aggregates obtained, including the CO<sub>2</sub> binding ability, and the reduction of the environmental hazard from the respective wastes.

The results showed that the CO<sub>2</sub> uptake was attributed mainly to the relatively high content of free lime (10–15%) in BOS and MSWI APC residues. Ca-silicates such as alite, belite, merwinite, etc. endorsed solidification by also capturing CO<sub>2</sub>. Most of the ash and ash blends showed a high reactivity, with CO<sub>2</sub> up to 98% of BD<sub>CO<sub>2</sub></sub>, resulting in a significant decrease in free lime content. The aggregates were mainly in the 4 < x < 10 mm granule size range, with the compressive strength being up to 4.5 MPa and the apparent density close to 2.1g/cm<sup>3</sup>. Generally, compared to burnt oil shale alone, the mixtures of different wastes after addition of cement produced stronger granules. This indicates that better results could be achieved by the further optimisation of the mixtures. The BOS collected from the SO<sub>2</sub> removal system provided the weakest granules, which was attributed to its very slow hardening dynamics.

The granulation process under the accelerated carbonation conditions led to a significant decrease in the pH levels and electrical conductivity values of the leachate. In the case of hazardous MSWI APC residues, the mobility of chloride and heavy metals could be decreased by optimising the mixtures. The leaching test indicated that the granules suffered from some weight loss at current cement additions (5–10%). These granules shall be studied further to elucidate their potential to be used in the production of building blocks.

Finally, the results of this study could serve as the first step towards closing the waste cycle in Estonian energy and cement sectors, to achieve national targets of promoting waste recovery, reducing waste dumping and decreasing greenhouse gas emissions for a sustainable future.

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