Sustainable approach to valorise ashes from MSWI

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ABSTRACT: Municipal solid waste incineration (MSWI) is a solution adopted by several European countries to reduce the amount of waste landfilled. After the incineration process, the inorganic fraction mainly recovered as bottom ash (BA) is generally landfilled. Therefore, in order to apply this residue safely as a building material, metals were extracted by mechanical separation from the bottom ash, which was then vitrified by electrical arc furnace. The vitrified BA was later converted into highly porous glass-ceramics by using a novel technique. After activation of the glassy slag in a weakly alkaline solution, foams were obtained by mechanical stirring with the help of a surfactant, and stabilized by gelification. Finally, the resulting open-celled structure was 'frozen' by a sinter-crystallization treatment. The Life Cycle Assessment carried out; also demonstrated the environmental benefits of upcycling MSWI BA into valuable building materials compared with the common disposal route.

1 INTRODUCTION

Nowadays in Europe, only 24 wt.% of Municipal Solid Waste (MSW) generated is landfilled (Eurostat, 2016). This achievement in waste reduction has been influenced by the use of municipal incinerators to decrease waste volumes and produce energy (IPPC Bureau. 2016). Incineration reduces the amount of waste weight to around 60-70%. By the end of the process, two types of residues are generated: bottom ash and fly ash. The first one represents around 80% of the total and contains metals such as Fe, Cu or Al (Kahle et al. 2015). In most cases, ashes are usually dried and mechanically treated to extracts metals before being landfilled.

In this work, three different scenarios have been evaluated to analyse the quality of the recovered metals from bottom ash (BA) and the potential valorisation of a post-treated vitrified bottom ash (VBA). In these new approaches, BA is first cleaned and vitrified using an electric arc furnace (EAF). Thereafter, the produced VBA is upcycled into porous glass-ceramics by an 'inorganic gel casting' technique: after the partial dissolution of fine glass powders in a weak alkaline activating solution, the suspension undergoes hardening during curing. A gel with pseudoplastic behaviour is obtained, which can be easily foamed at high shear rates with the support of a surfactant. When stirring stops, the increase of viscosity prevents the collapse of the foamed structure. Finally, the foam is dried and sinter by viscous flow (Rincon et al. 2017). Life cycle assessment (LCA) is then used as tool to compare the environmental impacts and benefits of the resource recovery processes in comparison with the more common disposal route, residual landfill.

2 MATERIALS AND METHODS

2.1 MSWI BA Upcycling

For this study, 50 kg of wet MSWI BA, provided by the company AVR in the Netherlands were consumed. Firstly, BA was dried at 200°C for 24 hours.

Magnet and eddy-current separators (ECS) where used to extract the ferrous and non-ferrous metals from MSWI BA, respectively. In order to evaluate the quality of the recovery metals in function of particle size, sieves between 45 µm and 10 mm were used. After screening, metals were deagglomerated using a jaw crusher and then washed. Metal were dried, weighted and chemically analysed with a portable XRF analyser (Thermo Fisher NITON XL3t 600).

The BA (up to 3 kg by trial) was added to a graphite crucible and smelted in a lab-scale EAF operating in DC at around 1450 °C. A graphite electrode of 50 mm was used on the top. After the smelting, the slag was quenched in water, dried and milled firstly with a jaw crusher and then with a planetary ball mill, until the particle size was below 75 μ m. The milled VBA was added to an alkali activating solution of 1M NaOH. The overall solid loading was 70 wt% and the suspension was mechanically stirred at 400 rpm for 3h. Thereafter, 4 wt.% of surfactant (Triton X-100, Sigma-Aldrich) was added to the suspension which was submitted to an intensive mechanical stirring at 2000 RPM. The foamed suspension was subsequently dried at 40 °C for 48h, demoulded and fired at 1000 °C, with heating rate of 10 °C/min and a holding time of 1h.

Chemical and mineralogical composition of MSWI BA, vitrified slags and glass-ceramics were analysed with a PANanalytical WDXRF spectrometer and a PANanalytical X'Pert³ x-ray. The bulk density of the porous glass-ceramics was calculated by the weight-to-volume ratio of fired foams cut into cubes of 1cm³. The apparent and true densities of the glass-ceramics were assessed by using a He gas pycnometer (Micromeritics AccuPyc 1330, Norcross) with the foams and fine powder, respectively. The compressive strength of at least 7 fired foams was determined by using an Instron 1121 UTM (Instron Danvers, MA), with cross-head speed of 1 mm/min. The morphological structure of the porous glass-ceramics ash was assessed by optical stereomicroscopy (AxioCam ERc 5s Microscope Camera, Carl Zeiss Microscopy).

2.2 Life Cycle Assessment (LCA)

The LCA study on the treatment and management of MSWI BA, focused on the quality assessment of the materials recovered (see also Allegrini et al. 2015), their marketability and on the further valorisation of the treated BA as glass ceramics. Three scenarios were evaluated for this purpose. In the first scenario (S1), after the extraction of Ferrous (Fe) and Non-ferrous (NFe) metals by mechanical separation, the treated BA was landfilled. In the second scenario (S2), instead of landfilling, the BA was melted using an EAF obtaining a vitrified slag. A metal alloy rich in Fe and Cu was also recovered by this procedure. The last strategy (S3) aimed at considering the low quality of Al by avoiding the use of the ECS compared to S2. For the last two scenarios, BA was upcycled into highly porous materials for thermal and acoustic insulation.

A unitary functional unit (FU) was chosen for the study and defined as the treatment of 1 kg of MSWI BA in Germany and the time horizon was set to 100 years. The LCA was performed with GaBi 8.0 and the ecoinvent database was also used for the LCI of technologies and processes. To address the multifunctionality of the resource recovery processes system expansion was applied.

Process inputs and outputs were obtained from lab data and upscale based on Piccinno et al. (2016). When not available, data from literature, calculated or estimated was also included. For the background processes, such as the production of primary materials and secondary materials, energy data and transport data, the inventory data available in the Ecoinvent v.3.2 Database was used.

Based on previous studies on the recovery of MSWI BA and on the production of ceramics (Allegrini et al. 2015; Birgisdóttir et al. 2007; Edirisinghe. 2013; Nicoletti et al. 2002), the chosen impact categories for this study were Global Warming Potential (GWP). Acidification Potential (AP), Resource Depletion (RD), Human Toxicity (HT), and Ecotoxicity (ET). The impact assessment method chosen was the ILCD method as it considers the results from recommended methods for different impact categories.

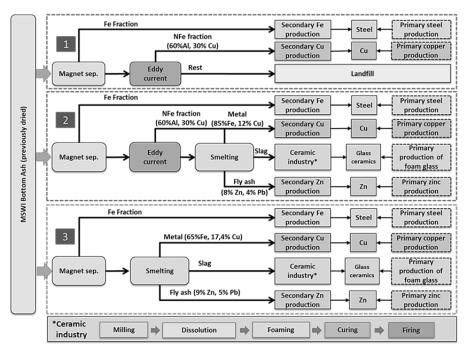


Fig.1: Evaluated scenarios for MSWI BA. (1) Common process; (2) & (3) BA upcycling

3 RESULTS

3.1 Quality Assessment of Metals

Upcycling of BA requires different steps. The most common are drying and metal recovery using magnets and eddy-current separators. Several authors show those ferrous and nonferrous fractions are in the order of 7 and 2% of BA, respectively (Kahle et al. 2015; Gisbetz et al. 2013; Muchova & Rem 2016). However, there is no information about the quality of these metals. In the current study, the fraction of ferrous and nonferrous particles measured during mechanical separation were 8 and 2.5 wt.%, respectively. Results from the screening by size and then cleaning showed that the quality of metals is inversely proportional to the particle size. Figure 2 summarises these results. In the case of magnetic separation, magnetic particles below 2 mm usually remain in BA when conventional suspension-magnet-separators are used. Magnetic pieces bellow 8 mm are strongly oxidised and polluted with inorganics. Larger size pieces have in general good quality.

Tab.	1: Chemical	composition	and n	nass t	palance 1	tor (different	scenarios.	

Scenario		1.2 & 3		1 & 2		2			3			
vart 0/-	wt.% BA		Mag. Sep		ECS		EAF			EAF		
WL. 70	DA	Rest	Fe	Rest	NFe	Slag	Met.	FG	Slag	Met.	FG	
Si	17.0	17.0	0.0	17.0	0.0	16.1	0.1	8.0	15.8	0.1	1.1	
Ca	10.5	10.5	0.0	10.5	0.0	10.2	0.0	0.4	10.1	0.0	0.4	
Na	2.7	2.7	0.0	2.7	0.0	1.6	0.0	1.1	2.0	0.0	0.7	
Al	6.4	6.4	0.0	5.6	8.0	5.4	0.0	0.2	6.3	0.0	0.1	
Fe	10.9	6.0	4.9	6.0	0.0	0.1	5.7	0.1	0.6	5.1	0.2	
Cu	1.5	1.5	0.0	1.1	0.4	0.1	0.9	0.0	0.2	1.3	0.0	
Zn	0.7	0.7	0.0	0.6	0.1	0.0	0.0	0.6	0.0	0.0	0.7	
Pb	0.4	0.4	0.0	0.3	0.1	0.0	0.0	0.3	0.0	0.0	0.4	
Cr	0.1	0.1	0.0	0.1	0.0	0.0	0.0	0.0	0.0	0.1	0.0	
Ti	8.0	0.8	0.0	8.0	0.0	0.7	0.0	0.1	0.6	0.1	0.2	
S	0.7	0.7	0.0	0.7	0.0	0.3	0.0	0.4	0.6	0.0	0.1	
Р	0.5	0.5	0.0	0.5	0.0	0.0	0.1	0.4	0.1	0.1	0.3	
С	4.3	4.3	0.0	4.3	0.0	0.0	0.0	4.3	0.8	0.0	3.5	
0	41.2	41.2	0.0	41.2	0.0	28.9	0.0	12.3	28.6	0.0	12.6	
Bal.	2.43	2.43	0.0	2.43	0.0	1.30	0.0	1.13	1.36	0.0	1.06	
Total	100	95.1	4.9	93.7	1.4	64.7	6.8	22.2	67.0	6.8	21.3	

BA: dried MSWI bottom ash; Fe: Iron recovery after cleaning; NFe: non-ferrous metals recovered after cleaning; Met.: metal alloy taped in the EAF; FG: Flue gas and Fly Ash; Bal.: include K, Mg, Cl.

For non-ferrous metals, ECS is efficient to extract pieces larger than 5 mm. Below this size, instead, efficiency drop exponentially. Regarding the type of metals and their quality, AI represents almost 60% of this fraction, followed by Cu with 30%. Municipal incinerators work with temperatures between 700 and 1000 °C (IPPC Bureau 2016) which favours the melting and oxidation of AI. The chemical analysis made on AI particles revealed a metal not only polluted with inorganics but also with other metals such as Cu, Zn or Fe.

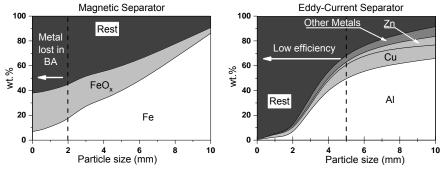


Fig. 2: Quality of metals extracted by mechanical separation in MSWI BA.

The presence of heavy metals like Cu, Zn, Pb, and Cr make these ashes hazardous for human and natural ecosystems if disposed in landfills or used as aggregate in road paving. Therefore, the smelting of BA using EAF allows cleaning it from heavy metals recovering them in the metal tapped out from the furnace or from the fly ash. The mass balances of the different step for each scenario are presented in Table 1. For the LCA, substitution ratios for metals recycling were calculated taking into account the quality (represented by the recovery efficiencies), and the market absorption potential, based on the framework developed by Vadenbo et al. (2017). The recovery efficiencies for the 3 different scenarios are summarized in Table 2. All materials were assumed to be marketable except for Al, given its low quality which was assumed not to be recycled.

Tab. 2: Recovery efficiency of the material fractions based on the quality assessment.

$\eta_{recovery}$ [%]	Fe	Cu	Zn	Al	
Scenario 1	45	28	0	11	
Scenario 2	45	87	14	11	
Scenario 3	45	90	95	0	

3.2 Upcycling BA mineral fraction

As confirmed by the XRD patterns, the vitrified slags obtained from S2 and S3 have an amorphous structure (Fig. 3), with the typical "halo" of glasses. In addition, it can be seen that the crystallization of glasses from S2 and S3 was substantially different. Labradorite ((CaNa)(Al.Si) $_4$ O $_8$. PDF 83-1371) was the only crystal phase detected in both glass-ceramics. This phase was more crystallized in glass-ceramics from S3, in which gehlenite (Ca $_2$ Al(AlSiO $_7$), PDF 72-2128) was also detected. Regarding the glass-ceramics made with S2, it was also detected pseudowollastonite (CaSiO $_3$, PDF 89-6463) and augite ((Ca.Na)(Mg.Fe.Al.Ti)(Si.Al) $_2$ O $_6$, PDF 70-3753). The difference between the crystallization of these glasses is mainly related to the different Al2O3/SiO2 ratio of glasses S2 and S3.

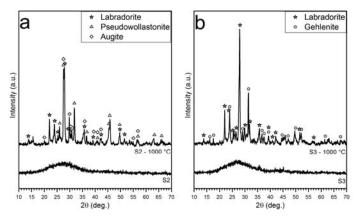


Fig. 3: XRD patterns of VBA and their derived glass-ceramics: (a) S2; (b) S3.

The density, porosity and compressive strength of the porous glass-ceramics made with VBA are shown in Table 3. The developed glass-ceramics present very low relative density with a total porosity higher than 75 vol.% (mainly open). In addition, the foams are quite strong with a compressive strength higher than 3 MPa. The foams made with S2 are stronger but one must consider the lower relative density of foams made with S3.

Tab. 3: Physical and mechanical properties of glass-ceramics made with vitrified bottom ash.

Strat.	D	ensity (g/cm	³)		Porosity (%	Compressive	
	$ ho_{geom}$	Papparent	$ ho_{true}$	Total	Open	Closed	strength (MPa)
2	0.66 ± 0.01	2.55 ± 0.01	2.64 ± 0.01	74.8	73.9	0.9	4.2 ± 0.7
3	0.54 ± 0.01	2.70 ± 0.01	2.81 ± 0.01	80.7	79.9	0.8	3.0 ± 0.5

Figure 4 presents the morphological structure of the developed foams after firing. The porosity distribution is quite heterogeneous and it is possible to observe the open-celled morphology of the foams as indicated in Table 3.

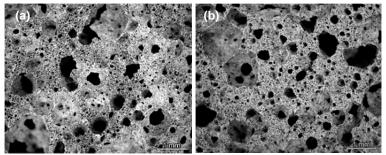


Fig. 4: Micrographs of VBA foams: a) S3 after firing; b) S2 after firing.

3.3 LCA results

The results for GWP, AP, and AD (see Fig. 5) show net environmental savings for S2 and S3. The environmental benefits resulted in being increasing with the increase in metal recovery, for Cu, Zn, and mostly influenced by the production of glass-ceramic foam and the consequent avoided use of primary resources and avoided landfilling. More specifically, S3 demonstrated higher savings given the higher amount of MSWI BA reaching the production stage of the glass ceramics and the higher % of metals (Cu, Zn) recycled.

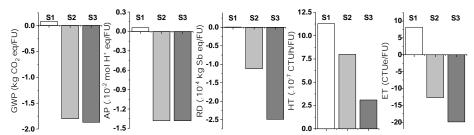


Fig. 5: Life cycle impact assessment by category and by scenario.

The toxicity categories, HT, and ET were dominated in all scenarios by the impacts from the BA disposal in landfill (S1), and of the NFe slag from the copper smelter. On the other hand, savings were associated to the recycling of Cu and Zn, and in case of ET, from the recycling of vitrified slag and the production of glass-ceramic foam (S2 and S3).

4 CONCLUSION

Mechanical separation technics are efficient to extract larger pieces of metals. However, the quality of these metallic pieces is in general low. All is the main metal in the non-ferrous fraction but the degree of pollution is too high to be used as a substitute by the aluminum industry.

The use of EAF allows treating and cleaning the incineration ashes from heavy metals. The metal tapped out of the furnace and the fly ash produced can be considered good raw materials for the copper and zinc industry respectively due to the high concentration of these metals.

The vitrified slag was further upcycled into highly porous and strong glass-ceramics by alkali activation gel-casting and sintering. Due to their high porosity, these foams could be potentially applied as panels for thermal and acoustic insulation.

Overall, the LCA results highlight the benefits of resource recovery and further valorization of vitrified slag to glass ceramics. Moreover, the increasing savings with increasing recycling rates emphasize the influence of quality and recoverability on the environmental impacts of metal recycling, as also discussed in Allegrini et al. (2015).

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REFERENCES

- Allegrini. E., Vadenbo C., Boldrin A., & Astrup T. F. Life cycle assessment of resource recovery from municipal solid waste incineration bottom ash. Journal of Environmental Management. 151, 132-143, 2015.
- Birgisdóttir H., Bhander G., Hauschild M. Z., & Christensen T. H. Life cycle assessment of disposal of residues from municipal solid waste incineration: Recycling of bottom ash in road construction or landfilling in Denmark evaluated in the ROAD-RES model. Waste Management, 27(8), 75–84, 2007.

 Editsinghe J. Life Cycle Assessment of a Ceramic Tile Produced in Sri Lanka. 3(11), 1060–
- European IPPC Bureau. "Reference Document on the Best Available Techniques for Waste In-
- cineration". http://eippcb.jrc.ec.europa.eu/reference/wi.html, 2006
 Gisbertz K., Hilgendorf S., Friedrich B., Heinrichs S., and Rüßmann D. "Maximising Metal Recovery from Incineration Ashes." Vol. 3, Weimar, Germany, 2013.
 Kahle K., Kamuk B., Kallese J., Fleck E., Lamers F., Jacobsson L., and Sahlén J. "Bottom Ash from WTE Plants: Metal Recovery and Utilization". Denmark: ISWA Working group on energy recovery, 2015.
- High recovery, 2015.

 Muchová L., and Rem P.C. "Metal Content and Recovery of MSWI Bottom Ash in Amsterdam". In WIT Transactions on Ecology and the Environment, 92:211–16, 2006.

 Nicoletti G. M., Notarnicola B., & Tassielli G. Comparative Life Cycle Assessment of flooring materials: Ceramic versus marble tiles. Journal of Cleaner Production, 10(3), 283–296,
- Rincón A., Giacomello G., Pasetto M., and Bernardo E. "Novel 'inorganic gel casting' process for the manufacturing of glass foams." Journal Eur. Ceramic Soc., vol. 37, N° 5, pp. 2227– 2234, 2017.
- Piccinno F., Hischier R., Seeger S., & Som. C. From laboratory to industrial scale: a scale-up framework for chemical processes in life cycle assessment studies. Journal of Cleaner Production, 135. 1085–1097, 2016.
 Vadenbo C., Hellweg. S., & Astrup T. F. Let's Be Clear(er) about Substitution: A Reporting Framework to Account for Product Displacement in Life Cycle Assessment, Journal of Industrial Ecology. 3(15), 1079, 1079, 2017.
- trial Ecology, 21(5), 1078-1089. 2017.