Abstract

Hugo Lucas, Jürgen Maier and Bernd Friedrich

The greatest challenge in the 21st century is to transform a linear economy into a circular and environmentally friendly economy. Landfilling mineral wastes, mainly those caused by mining and metallurgical industry, are one of the most considerable problems to confront. The submerged arc furnace, a technology extensively used for metal extraction, is a technology capable of treating and cleaning countless mineral wastes, thereby permitting elaborate valuable mineral precursors for the construction industry. Whether conditioned slags can through high-temperature treatments fulfil local and national regulations in terms of content and leachability of pollutants, several near zero-waste upcycling strategies can be carried out for the building sector. This article is a review of the present and past work made by IME Process Metallurgy and Metal Recycling, RWTH Aachen University in collaboration with industry and others universities showing how Submerged Arc Furnaces can successfully treat a wide range of residues from tailing ores such as bauxites residues, ashes from Waste to Energy plants to hazardous metallurgical slags, i.e. copper and lead slags. Upcycling solutions include their use as aggregates for road paving and concrete, additives for cement, and finally bricks, tiles, and acoustic and thermal isolation materials via inorganic polymers or glass-ceramics.

The Use of Submerged Arc Furnace (SAF) as a Robust Technology for Upcycling Waste into Standard Mineral Products for Construction Industry

Hugo Lucas, Jürgen Maier and Bernd Friedrich

1. Introduction

The social context in the 21st century is deeply marked by clime change and a linear economy based on an unsustainable creation of waste. Reshapes our consumption habits for creating a philosophy of zero waste is perhaps one of the biggest challenges of our time. Europe is pushing for new policies regarding the avoidance, reuse and recycling of waste. Several EU indicators show that the avoidance of waste reaches values close to 8 % for some west countries (Figure 1). Nevertheless, mineral wastes are segregated from this data, presenting a picture far beyond reality.

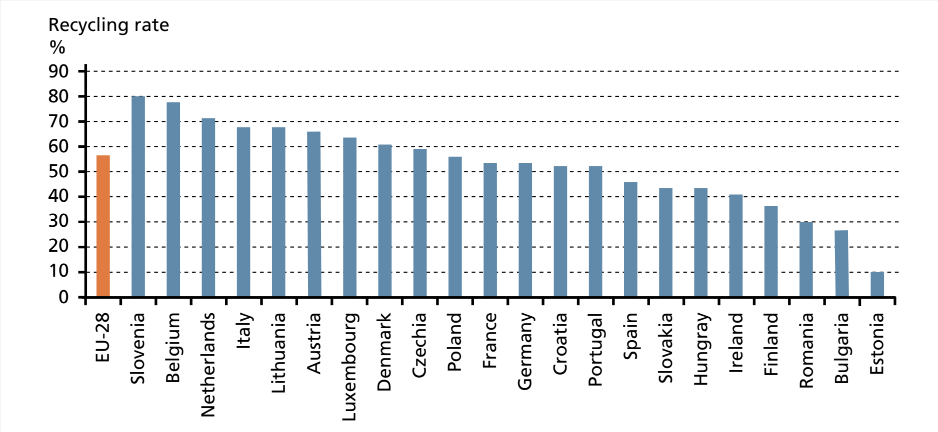

Figure 1: Recycling rate of all waste, excluding major mineral waste in 2016

Source: Eurostat. statistics: https://ec.europa.eu/eurostat/

No reliable data exist in Europe about the type and amount of mineral waste produced. In the USA, industrial waste related to the extraction and refining of metal represent almost 60 % landfilled waste, and thus a green industrial revolution has to be capable of revalorise these residues. Eliminating the environmental burden posed and turning them into mineral by-products suitable for its use in construction is only possible through high-temperatures processes such as Electric Arc Furnaces (EAFs).

Building industry is always growing due to the rising global population. According to Eurostat in 2017, Europe consumes 2.13 billion tons of construction materials per year [7], and consequently, several mineral wastes may be used as raw material whether fulfilling the environmental standard imposed by legislations.

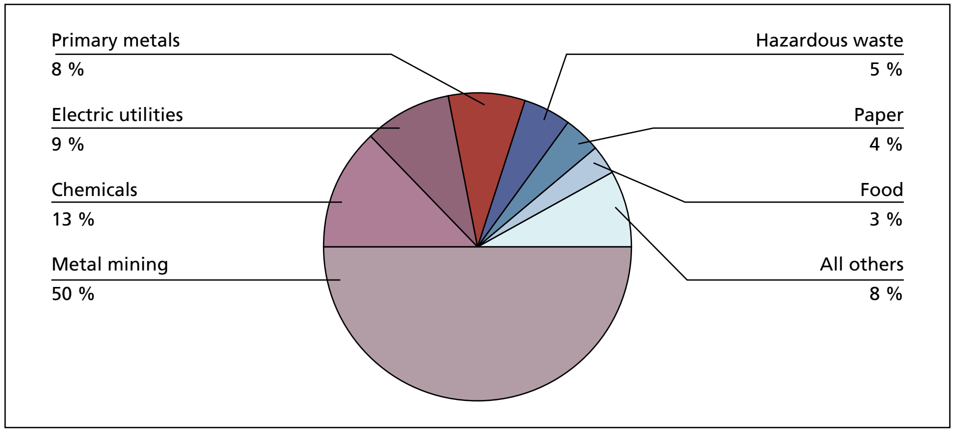

Figure 2: Share of on-site land disposed of production-related waste in the United States in 2017 by sector

Source: Statista, Share of U.S. land disposed industrial waste by sector 2017. Available: https://www.statista.com/statistics/506381/share-industrial-waste-onsite-land-disposed-in-the-us-by-sector/

2. Submerged Arc Furnaces

The utilisation of Submerged Arc Furnaces (SAF), which is a particular type of EAF, has proven a versatile unit in countless metallurgical applications and is perhaps the only robust technology capable of treating mineral wastes. This technology was initially developed for pure metallurgical applications where optimising metal production was the primary goal. During the last 20 years, in the area of nonferrous metallurgy several slag-cleaning furnaces connected to other smelting units has been installed to not only enhance metal recovery but besides conditioning the mineral phase to be reused afterwards as aggregates for the construction industry [9].

2.1. Basics on SAF technology

SAF technology used is basing on immersed electrodes in a melt. Electrical energy is converted into heat through the electrical resistance of the burden or molten slags. Electric energy is supplied through an external source into the furnace via self-backing graphite electrodes serving for the melting and chemical reactions. Figure 3 shows a general overview of the main SAF components. Depending on the application, liquid or solid charging is possible. After treatment, metals and mineral by-products are tapped by different tapping holes or by tilting into ladles. In some cases, flue dust enriched in heavy metals, i.e. zinc or lead is one of the saleable product generated by this technology.

For the environmental perspective, because of the use of electrical energy, SAFs have lower CO_2 footprint compared to Gas-heated furnaces. In the future, the integration of renewable energies in metallurgical processes will make this technology even more suitable.

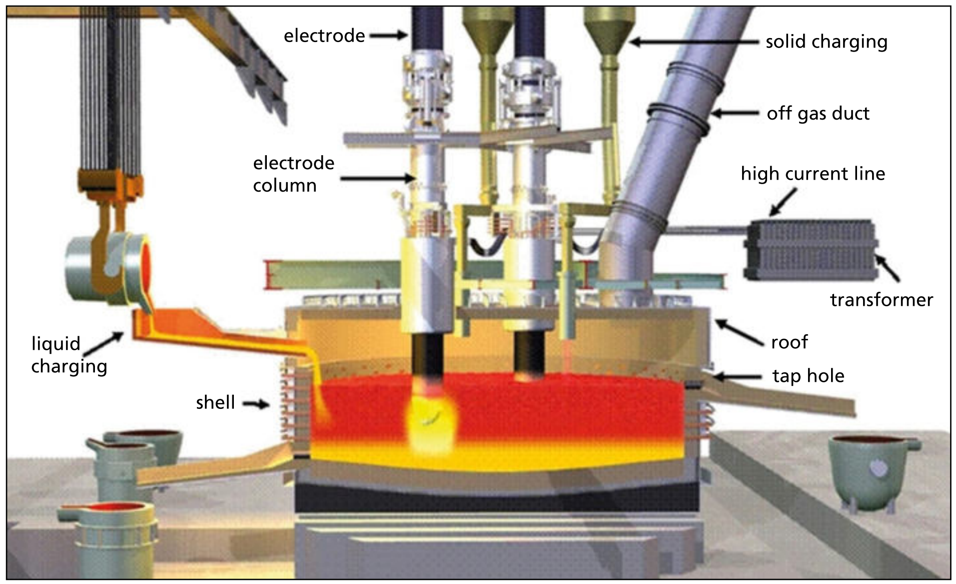

Figure 3: Layout of a submerged arc furnace

Source: Degel, R., Kunze, J., 2003. History, current status of submerged arc furnace technology for ferro alloy metals. Steel Grips 3.

2.2. Slag cleaning

Mineral waste can contain valuable metals such as Cu, Zn, Ni, Ag, Au besides others not as valuables and hazardous as Sb, Hg or As. Pyrometallurgical cleaning consists in melting the feeding material, reducing oxides, settling down metal droplets and releasing boiling metals across the exhaust conducts.

The chemistry of each mineral product influences the choice of thermal conditions, fluxes, reductants and times needed to carry out the cleaning process. Increasing the temperature and the proper use of fluxes decrease the viscosity and accelerates the kinetics and settlement of metal particles [9].

Depending on the nature of the melt, different reactions between metal, oxides, mattes and reductants could take place (Figure 4):

$$2\,MeS + 3\,O_2 \longleftrightarrow 2\,MeO + 2\,SO_2 \qquad (1)$$

$$MeS + 2\,MeO \longleftrightarrow 3\,Me + SO_2 \qquad (2)$$

$$MeO + C \longleftrightarrow Me + CO \qquad (3)$$

$$MeO + CO \longleftrightarrow Me + CO_2 \qquad (4)$$

$$Me_AO\ Me_BO + C \longleftrightarrow Me_AMe_B + CO_2 \qquad (5)$$

$$Me_AO + Me_B \longleftrightarrow Me_A + Me_BO \qquad (6)$$

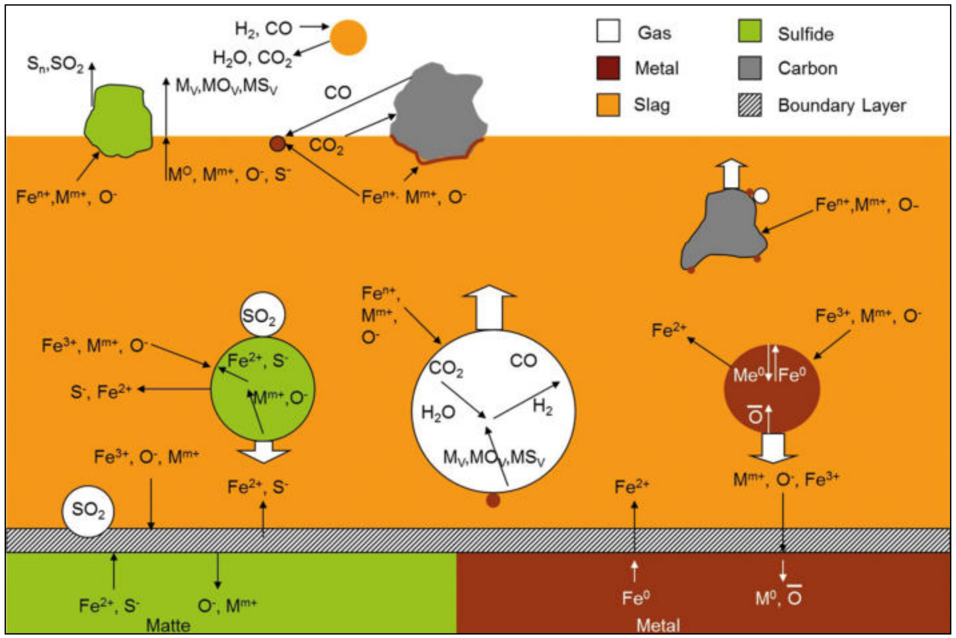

Figure 4: Reactions in a fayalitic copper slag

The settlement time of metal particles as described by the formula of Hadamard-Rybcznki (Equation 7) depends on the diameter of the metal particle, the difference of viscosity between the metal formed and the slag and its viscosity:

$$u = \frac{(\rho_M - \rho_S) \cdot g \cdot d_M^2}{18 \cdot \eta_S} \qquad (7)$$

Where d_M is the particle size of metal droplets in millimeter, g the gravitation constant in m_2/s, ρ_M and ρ_S are the density of the metal and slag respectively, η_S the viscosity of the slag in Ns/m^2, and u is the settling velocity in *m/s*.

An accurate cleaning and consolidation of mineral by-products through the use of SAF will depend on the right choice of parameters, fluxes and time to obtain a product capable of fulfilling the standards of the building industry and local legislations.

3. Conditioning mineral waste from nonferrous industry

In ferrous industry, several of its outputs are CaO or MgO rich minerals, and currently reused as aggregates for road paving, concrete or as additives for cement. The main concern in metallurgy is related to the nonferrous industry, after the extraction and concentration of ores such as those from cooper or aluminium tailings due to its chemical and physical characteristics are landfilled. Besides landfilling tailings ores, nonferrous industry uses mostly fayalitic slags in their operations. After processing, these silica-iron rich slags contain several metal oxides, mattes and metallic droplets with valuables metals, i.e. Cu, Ni, Mo, Co, Pb, Zn, Sb that in some cases are also considered hazardous and thus restricted for almost all EU regulations meaning that their reuse for construction purposes is limited.

3.1. Bauxite residues

Aluminium industry uses bauxites ores to gain alumina via the Bayer process. For every 2 – 3 Tons of bauxite, one ton of alumina is produced. According to the US Geological Survey in 2017, 179 million tons of bauxite residues were worldwide produced [17]. Due to its high alkalinity and fine particle sizes these residues are landfilled in especial reservoirs. Landfilling these reddish tailing possess a serious and real threat to the environment not only because their high alkalinity but also since they are stored in massive dykes that by different causes can collapse spreading for kilometres reddish slurries destroying everything in its path as it happens in 2019 in Brazil (Pana) or 2008 in Hungary (Figure 5).

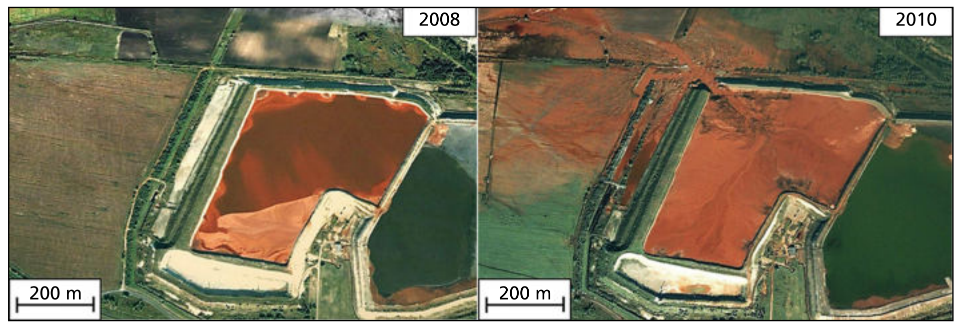

Figure 5: Aerial view of a red mud reservoir in Ajka (Hungary) in 2008 (left side) and two days after the collapse of the dam in 2010 (right side)

Source: Google Earth.

Although these tailings contain high-concentration of aluminium and iron oxides (Table 1) cannot be valorised since re-leaching of alumina is not effective, and iron oxides cannot be reduced in traditional blast furnaces owing to their high alkalinity which is not compatible with traditional lining used in the installed furnaces [11].

Table 1: Typical composition and span of bauxite residues

Components	Fe_2O_3	Al_2O_3	SiO_2	TiO_2	Na_2O	CaO	P_2O_5
	wt %						
Bauxite residue from Lünen (Germany)	29.5	27	13.1	8	6.9	3.8	0.22
Bauxite residue from Greece	43.5	24	5.5	5.6	2	10.5	0.15
Span	30 – 50	10 – 30	5 – 20	3 – 15	3 – 7	1 – 8	0.1 – 0.3

Sources: Kaußen, F.M., Friedrich, B., 2016. Methods for Alkaline Recovery of Aluminum from Bauxite Residue. Journal of Sustainable Metallurgy 2, 353 – 364. https://doi.org/10.1007/s40831-016-0059-3

Thomas, C., Rosales, J., Polanco, J.A., Agrela, F., 2019. 7 - Steel slags, in de Brito, J., Agrela, F. (Eds.), New Trends in Eco-Efficient and Recycled Concrete. Woodhead Publishing, pp. 169 – 190. https://doi.org/10.1016/B978-0-08-102480-5.00007-5

During the last decade, a near-zero waste holistic exploitation has been intensely studied to increase the sustainability of aluminium industry [2]. Among the strategies carried out, smelting and conditioning of bauxite residues through the use of SAF between 1.500 and 1.550°C offers the best technical solution for a downstream valorisation. Through smelting and carbothermic reduction, more than 93 % of iron is valorised by coalescing and settlement on the furnace bottom [23]. Depending on the utilised fluxes (CaO, MgO, SiO_2, BOF) the final slag enriched in Al_2O_3 and TiO_2 can be leached again via alkaline treatment for the recovery of alumina [10] or by acid leaching for titanium and scandium recovery besides other REEs [1]. Figure 6 shows an outline of on-going research strategies for the reuse and recycling of bauxite residues.

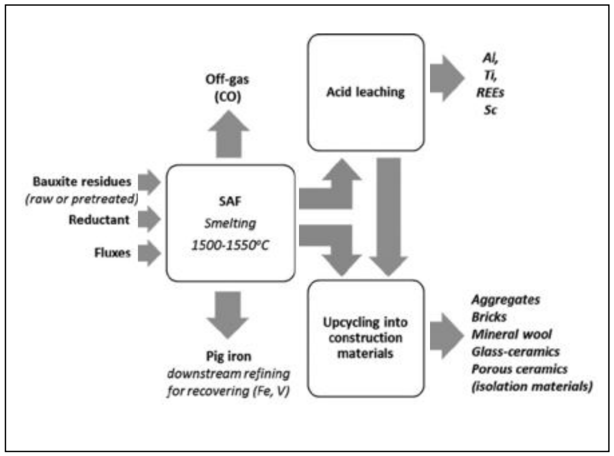

Figure 6: Flowsheet for a near-complete utilisation of bauxite residues

Table 2 shows the results of the slag and metal phases produced after a carbothermic reduction of a Greek bauxite residue using a lab-scale Direct Current (direct current) SAF (Figure 7a) and lime, silica or BOF slag as fluxes. Large-scale trials for treating these residues have also been carried out using a 1 MW Alternative Current SAF (Figure 9c).

Table 2: Slag and metal composition of conditioned bauxite residues via SAF smelting

Components	Fe_2O_3	Al_2O_3	CaO	SiO_2	TiO_2	Na_2O
	wt %					
slag + 30 % lime	1.7	33.8	49	6.9	6.9	0.2
slag + 30 % BOF slag	3.7	34.5	31.6	15.7	7.8	2.9
slag + 30 % silica	1.2	36.8	15.3	38	7.3	2.4
Components	**Fe**	**Cu**	**Ni**	**Si**	**Ti**	**C**
metal 1 (30 % lime)	95	0.0	0.3	0.1	0.1	4
metal 2 (30 % BOF slag)	99	0.3	0.5	0.1	-	-
metal 3 (20 % silica)	93	0.0	0.3	0.1	0.3	5

Sources: Thomas, C., Rosales, J., Polanco, J.A., Agrela, F., 2019. 7 - Steel slags, in de Brito, J., Agrela, F. (Eds.), New Trends in Eco-Efficient and Recycled Concrete. Woodhead Publishing, pp. 169 – 190. https://doi.org/10.1016/B978-0-08-102480-5.00007-5

Xakalashe, B., Friedrich, B., 2018. Combined carbothermic reduction of bauxite residue and basic oxygen furnace slag for enhanced recovery of Fe and slag conditioning. Presented at the 2nd International Bauxite Residue Valorisation and Best Practices Conference, Athens, Greece.

3.2. Copper slags

In the primary copper production, slags containing between 1 – 8 % of copper are produced after the treatment of mattes. These slags are treated using SAF for reducing the copper content below 1 % (Table 3).

Table 3: Typical composition of Cu slag after SAF cleaning

Compo-nents	FeO	SiO_2	CaO	Al_2O_3	Zn	Cu	S	Pb	Mo
	wt %								
Cu slag	40 – 43	31 – 34	3 – 5	2.5	1.5	0.8	0.4	0.3	0.3

Source: Zander, M., Friedrich, B., 2011. Improving Copper Recovery from Production Slags by Advanced Stirring Methods. https://doi.org/10.13140/RG.2.1.4708.9522

Under the standard regulations, the content of heavy metals restrains the use of this mineral by-product to be used as a raw material in the construction industry. Through intensive slag cleaning by using additives such as TiO_2, CaO, MgO, Coke and Coke as reductant and electromagnetic stirring methods, lead to 1.5 times better separation of metallic droplets reducing in 50 % the remaining copper from the slag showed in Table 3 (Figure 7). Figure 8 shows a stirring pilot reactor coupled with a SAF.

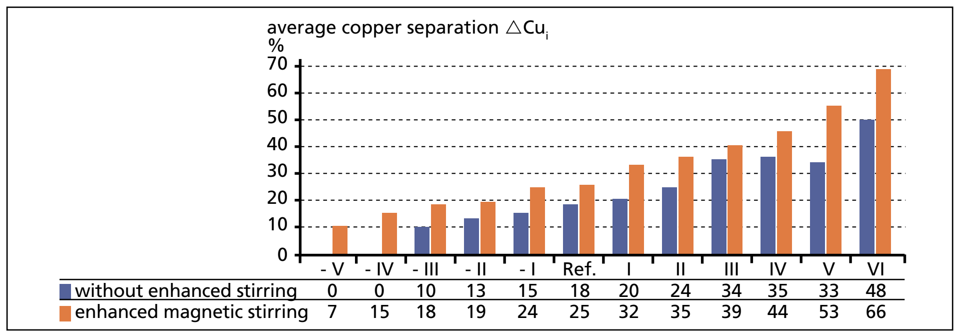

Figure 7: Cooper separation with and without enhanced magnetic stirring coupled with SAF

Source: Zander M., Friedrich B., Degel R., König R., Hoppe M., and Schmidl J., 'Efficiency of slag cleaning in a magnetically induced stirring reactor', 2013, https://doi.org/10.13140/RG.2.2.24232.78080

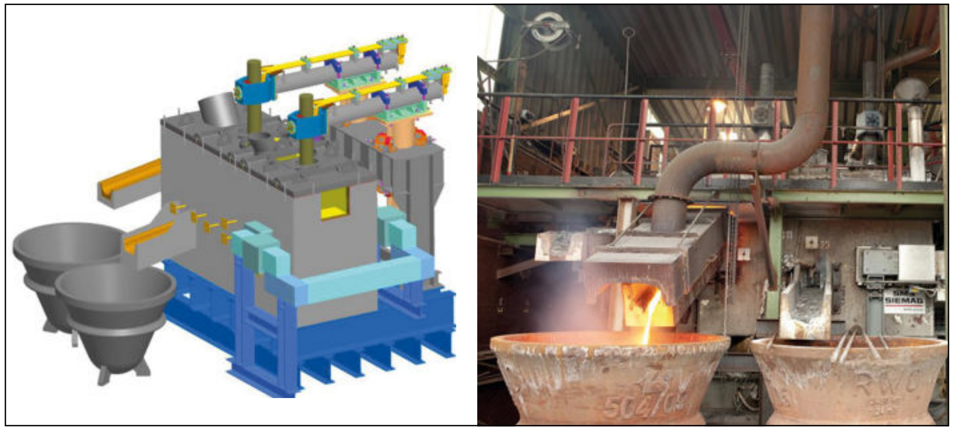

Figure 8: Magnetically induced stirring reactor of Aurubis in Hamburg

Source: Zander M., Friedrich B., Degel R., König R., Hoppe M., and Schmidl J., 'Efficiency of slag cleaning in a magnetically induced stirring reactor', 2013, https://doi.org/10.13140/RG.2.2.24232.78080

3.3. Lead and zinc slags

Lead worldwide production in 2018 amounted 11.6 million tons [18], which 65 % comes from secondary raw materials. Lead recycling uses a ratio metal scrap/slag of 3:1, leading to a worldwide slag production of 2.6 million tons. These slags usually contain up to 7 wt % lead and high amounts of zinc up to 16 wt % both as oxides (Table 4). Further accompanying hazardous compounds are arsenic and antimony. For this reason, it was attempted to treat such slags in SAF reducing the content of heavy metals to avoid landfilling this hazardous waste enabling its utilisation as aggregate for the construction industry. By-products expected from reductive slag smelting is high zinc oxide containing flue dust and a metal phase mainly composed of lead.

Table 4: Typical composition and span of lead slags

Components	Pb	ZnO	SiO_2	Fe_2O_3	CaO	Al_2O_3	MgO
	wt %						
Typical	3	12.5	20	30	20	3	3
Span	2 – 6	8.5 – 16	15 – 25	20 – 45	8 – 25	2 – 5	2 – 4

Source: Friedrich B. and Böhlke J.K, Lead and Zinc Recovery from Metallurgical Slags, 2010, https://doi.org/10.13140/RG.2.1.4770.5842

First experimental works have been carried out using first a lab-scale 100 kilowatt direct current tiltable SAF and later on a pilot-scale 250 kW direct current SAF (Figure 9a and 9b) with a melting capacity of 70 and 1.000 kg respectively. Off-gas produced during the trial is afterwards cleaned in an electrostatic precipitator and a scrubber. For emptying these furnaces, slag and metal phases are poured into a preheated mould except for pilot-scale trials (Figure 9 b) where materials are extracted through tapping holes [4].

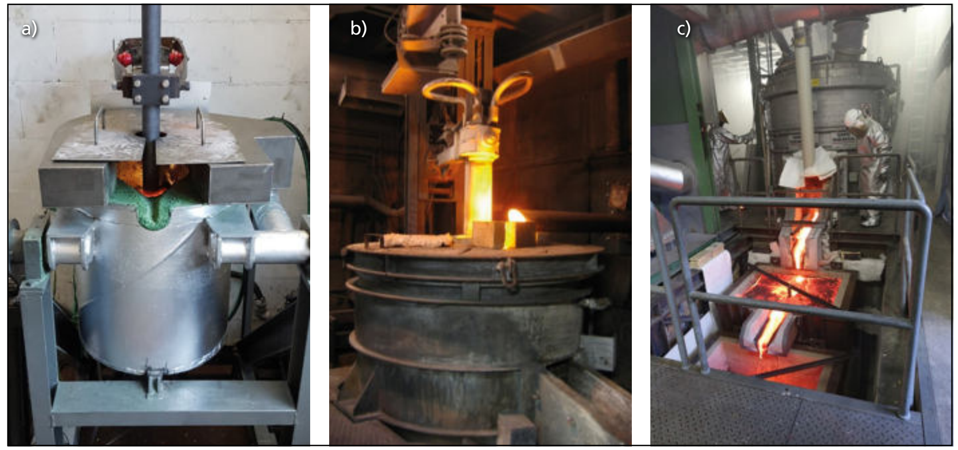

Figure 9: IME-SAF technology. (a) Lab-scale 100 kW direct current SAF, (b) pilot-scale 250 kW direct current SAF and (c) pilot-scale 1 MW AC/direct current SAF

Figure 10 compares the lead and zinc content in the final slag and flue dust after treating a lead fayalitic slag using lignite coke as reductant injected through hollow-electrodes (HE) or feed conventionally by previous premixed (PM) with the charge. Using lignite coke as reductant injected through hollow-electrodes with working temperatures between 1.450 and 1.600 °C, it was possible cleaning these slags reaching the EU requirements and terms of content and leachability of heavy metals for using these by-products in building applications. Lead and zinc were enriched in the flue gas for downstream treatment.

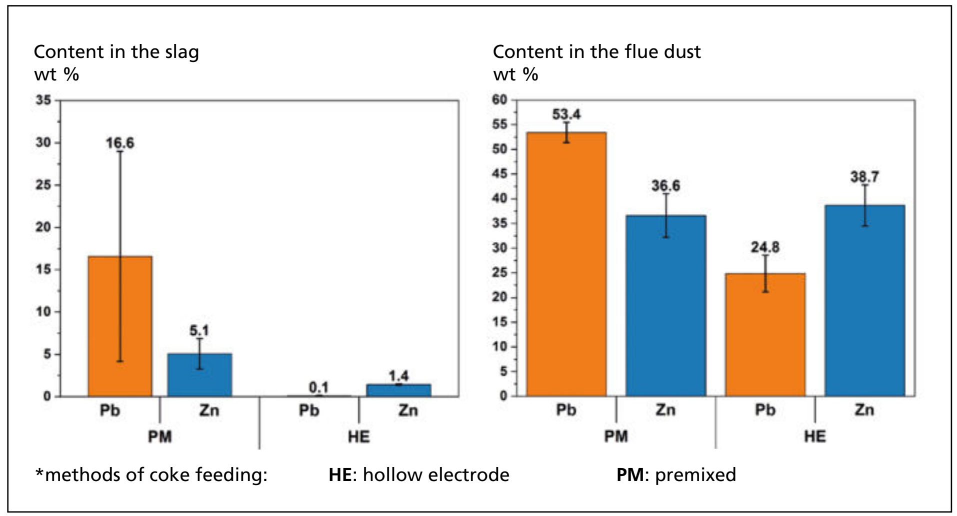

Figure 10: Lead and zinc content after treatment in a pilot-scale 250 kW DC-SAF (working temperatures 1.450 – 1.600 °C): (a) slag and (b) flue dust

Source: Böhlke J., 'Behandlung von Schlacken der Bleigewinnung im Elektrolichtbogenofen', Doctoral dissertation. Aachen, Germany, 30-May-2016.

Treating fayalitic slags at temperatures higher than 1.350 °C comes along with a partial reduction of iron which is undesirable because the metal tends to freeze on the bottom when temperatures are below 1.550 °C. Today, the leading research on cleaning lead slags focus in optimising zinc recovery via boiling the meal in the flue gas and lead settling down at the furnace bottom with working temperatures between 1.200 and 1.300 °C. After holding time of two hours of treatment using a lab-scale direct current SAF (Figure 9a), the melt is poured out (Figure 11a). Once both phases slag and metal solidifies are removed from the mould and separated from each other.

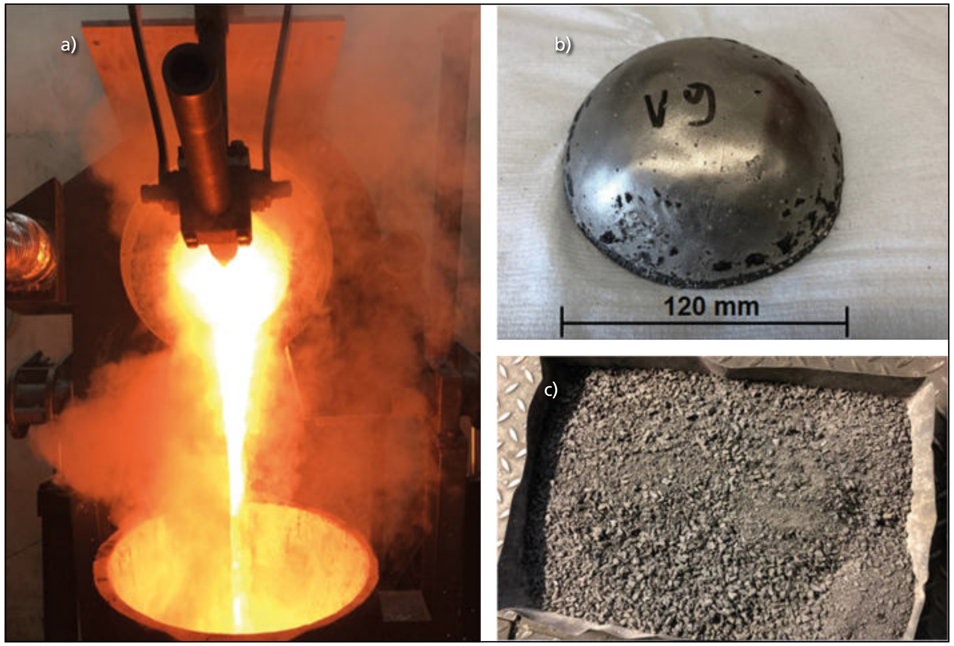

Figure 11: Emptying SAF after (a) slag cleaning, (b) metal phase and (c) slag phase

The slag phase is crushed (Figure 11b) to scope chemical and mechanical properties as well as eluate behaviour in order to check whereas fulfilling or not the requirement necessaries to be used by the construction industry. Obtained metal phase (Figure 11c) contains 90 wt % of lead and 8 wt % antimony which are valuables metals (Figure 12b) while flue dust from off-gas contains more than 85 wt % zinc oxide besides others metals such as lead, antimony and arsenic (Figure 12a). Thus, the obtained metal phase is a potential raw material for the production of antimonial lead or can be used as input material for lead refining process to win pure lead.

A possible field for further processing of generated flue dust is the zinc industry. For this purpose, some pretreatment in Waelz or clinker process is necessary to eliminate contaminants and purify zinc oxide. Afterwards, pure zinc oxide can be utilised in the electrowinning process to obtain refined metal.

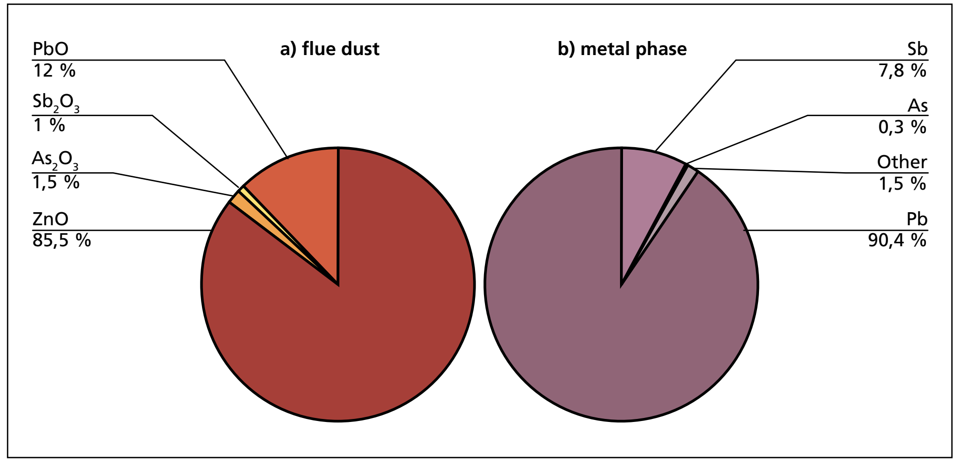

Figure 12: Chemical composition of treated lead slag adding 5.5 wt % of coke: (a) flue dust and (b) metal phase

3.4. Consolidating ashes from WtE plants

Several of the ashes produced by waste to energy plants in Europe have been partially or entirely reused for some member states as aggregates for road paving, and in rare cases for building application. Nevertheless, these ashes contain several elements such as Sb, Cd, Mo, Cu, Cl, and SO_4 partially or consistently exceeding the leachability limits imposed by European and national regulations [20]. SAF technology can help to transform these heterogeneous wastes into a clean and stable vitrified by-product (Figure 13).

Figure 13: Municipal solid waste (a) before incineration, (b) bottom ashes remaining after incineration (IBA) and (c) vitrified IBA and the recovered metal phase

During the remelting of municipal solid waste incineration bottom ash (IBA) between 1.450 – 1.500 °C, inorganic compound merge to form a stable SiO_2-CaO-Al_2O_3 based slag which is in general completely amorphous. Table 5 shows the chemical composition of IBA and main outputs generated after smelting in a direct current SAF (Figure 9a). During the thermal treatment almost all metal oxides are reduced forming by coalescing an iron-copper rich metal phase. These high temperatures are required to clean the mineral phase from heavy metals, dioxins and chlorides favouring its utilisation as a raw material for several building applications [13]. Besides copper, the metallic phase encloses several valuables metals such as Co, Mo, Nb, Nd and Ag.

Table 5: Chemical composition of the main outputs generated after smelting MSWI bottom ash (IBA) through SAF

Components	SiO_2	Al_2O_3	CaO	Na_2O	MgO	Fe_2O_3	SO_3	Cl	C
	wt %								
IBA	39.1	8.18	23.7	3.1	1.4	8	2.8	1.6	5.6
Slag	48.8	17.2	22.7	3.8	2.4	0.8	1.2	0.1	0.6
Components	**Cl**	**Na_2O**	**K_2O**	**ZnO**	**CaO**	**SiO_2**	**Fe_2O_3**	**Al_2O_3**	**PbO**
Flue dust	26.1	13.9	12.8	9.2	8.1	3.2	2.6	1.9	1.3
Components	**Fe**	**Cu**	**Si**	**P**	**Cr**	**Ni**	**Co**	**Sn**	**S**
Metal	83.4	10.5	1.4	1.4	0.9	0.6	0.4	0.2	0.1
Components	**Co**	**Mo**	**Sb**	**Zn**	**Nb**	**Ag**	**Nd**	**Cd**	**Au**
	ppm								
Minor metals	6.500 – 2.500	1.550 – 250	1.200 – 700	490 – 100	90 – 5	80 – 25	20 – 15	9 – 5	< 5

4. Applications

During the last decades, efforts have been made for the avoidance of waste. Steel industry works with CaO and MgO based slags in their process, and exist extensive bibliography and industrial examples for their use as aggregates for road paving, building, or as additives for cement [21]. The main concerns for reusing most mineral wastes are the legal restriction in terms of content and leachability of hazardous compounds. Although these residues do not fulfil common standards, several EU states have prepared especial guidelines or requirement for its reuse principally as a baseline of asphalts [20].

Cleaning these mineral by-products using high temperature is a sustainable approach for more valuable applications according to the concepts of circular economy. Balomnenos et al. demonstrated at pilot-scale that using SAF, iron can be extracted from bauxite residues, but at the same time the smelted mineral phase can be directly transformed by spinning in mineral wool which is an extensive isolation material used in almost all modern buildings [3].

State of the art for upcycling slags into valuable construction materials is based on broad engineering strategies such as inorganic polymers and glass-ceramics. Both strategies use in general alkaline solutions based on sodium and potassium hydroxides to partially dissolve the slags promoting controlled recrystallisation. While construction material based in inorganic polymers use high concentrated alkaline solutions and are cured at

room temperature like conventional cements; materials based on glass-ceramics promote the recrystallisation using weak alkali activators and temperatures between 800 – 1.000 °C for their sintering. Several authors have reported the production of bricks, tiles, large building structures, fire-resistant isolation materials (porous glass-ceramics and inorganic polymers) and light aggregates using these proceedings [2, 15, 14]. Figure 14 summarized the different strategies for valorising mineral by-products treated by SAF.

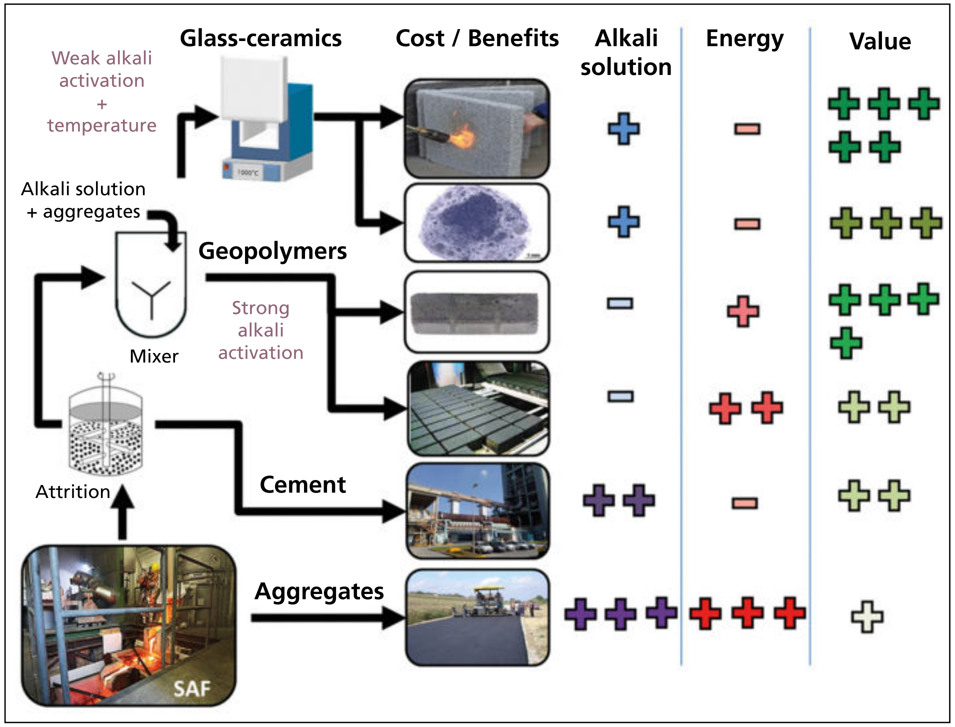

Figure 14: Strategies for upcycling slags in building industry

5. Conclusions

Avoiding industrial wastes is perhaps one of the most substantial barriers to circular economy. Landfilling these residues demonstrated to be unsustainable over time. Several environmental damages caused by industrial waste have been reported along the years. Nowadays, industries confront severe restrictions in terms of waste generation and concentration of pollutants, and these new challenges only can be addressed if mineral by-products become raw materials.

SAF proved in countless applications that is a robust technology capable of treating and cleaning a wide range of mineral waste fulfilling local and national legislations. Modern processes can transform these mineral products into different construction materials from low-value application like aggregates for road paving until valuable products such as fire resistance acoustic and thermal insulation panels.

Whatever the technology used to transform wastes into products, SAF units will probably become the missing link in the current waste management system to accomplish near-zero waste solutions.

Acknowledges

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