Developing a new process to agglomerate secondary raw material fines for recycling in the electric arc furnace - the fines2EAF project

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Recent years have seen a worldwide change in the environmental policy towards circular economy approaches. It is estimated that steel-making activities in Europe produce about 80 million tonnes annually of by-products and waste, equivalent to half of the European steel production, of which more than 10 million tonnes is waste for disposal. This waste of resources and land area is not sustainable and has to be decreased in the future.

The Fines2EAF project aims to increase the value of steelmaking residues by internal recycling and (re)use in the form of agglomerates. The benefit of this strategy is threefold: improved utilization of residues, internal recovery of valuable materials and reduction of the amount of dumped materials. The approach followed is the development of an innovative process to produce cement-free agglomerates based on primary and secondary raw material fines, alternative binder systems and a hydraulic stamp press. In addition, a new pre-treatment process for fines based on microwave heating is investigated.

The first results of the lab-scale investigation of the fines pre-treatment to reduce the amount of zinc, volatiles and alkalis are presented. Six materials from two steel plants have been tested in a laboratory microwave furnace. Also presented are first results of the agglomeration of fines using a laboratory press.

KEYWORDS: STEELMAKING – EAF – AGGLOMERATION – RECYCLING – SECONDARY RAW MATERIALS

INTRODUCTION

The steel industry is critical to the worldwide economy, providing the backbone for construction, transportation and manufacturing. In addition, steel has become the material of choice for a variety of consumer products, and markets for steel are expanding. Steel, already widely regarded as a high performance contemporary engineering material, is continuously being improved to meet new market demands.

The production process for manufacturing steel is energy-intensive and requires a large amount of natural resources. Steel production apart from steel as the main product leads also to the production of numerous by-products like slags or waste materials like dusts, sludges or scales. Additional fines are produced in the iron and steel industry and their supplying industry in general. The fines produced in steel industry include for example primary raw materials like iron ore fines or sieved undersize of alloying materials like FeSi or the sieved undersize of lime or dolomitic lime etc. and the already mentioned secondary raw materials like dusts, sludges, scales and slags. It is estimated that steelmaking activities in Europe produce annually about 80 million tonnes of by-products and waste, equivalent to half of the European steel production, of which more than 10 million tonnes is waste for disposal. This waste of resources and land area is not sustainable and has to be decreased in the future. The analysis of waste for disposal shows that 80% consists of slag, dusts and sludges, which can be transformed into raw materials for other users or usable products.

A direct recycling of the fines is in most cases not suitable. Therefore, agglomeration processes to produce briquettes, pellets or bricks are used to enable the handling and charging of fine materials into melting units like cupola and shaft furnaces,

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submerged arc furnaces (SAF) or electric arc furnaces to use or recycle the resources available in the raw materials. A special form of agglomerates are self-reducing bricks, which are e.g. used to utilise iron ore fines in the pig iron production in cupola furnaces or to recover the metal content (Fe, Zn, Cr, Ni, Mo, etc.) of dusts and sludges. Currently the recyclability of many process by-products and residues like disintegrated ladle furnace slags, dusts or sludges is still limited by their fine particle size and/or the low quantity of the material arising at a single steel plant. Fine dust fractions from various stages of steelmaking route contain besides iron and carbon, heavy metals and hydrocarbons that are acceptable neither for landfill disposal nor for recycling back to processes without any treatment.
Recent years have seen a worldwide change in the environmental policy towards integrated pollution prevention and control, taking into account all environmental media. Environmental regulations in the EU e.g. regarding slags become more and more restrictive, constricting the possible applications outside steel plants and prohibiting the landfilling. The integrated assessment of production processes under ecological, but also under technical and economic aspects requires specific methods (1, 2).

THE FINES2EAF PROJECT

The project aims at the increased use of low quality/low volume primary and secondary raw material fines, reducing costs of raw materials and dumping. A flexible, validated and cheap agglomeration technology, which can work "easily" inside the steel plant, still needs to be developed, validated and applied continuously. The installation of a treatment plant inside the steel shop reduces costs of transportation and also the need of special authorisation from local authorities.
The basic idea of this proposal is to develop an innovative process, which can be managed directly in the steel plant, reducing the amount of plant investments and reducing also handling, storage and transportation cost and management. This project is aimed at maximising the internal recovery of secondary raw materials from steelmaking wastes, with relatively small plant modifications and investments, saving production costs and reducing at minimum the landfill disposal of waste materials. Only the development of an easy technology, to be applied directly inside the steel plant will ensure the recycling of wastes materials, strongly contributing to a zero waste steel production.
To reuse the several types of metal or slag former bearing wastes and other residues of the steel plants, they have to be mixed with binders and possibly reducing agents and transformed to be reused directly in the EAF. On the experience of previous projects (1, 3), apart from fossil carbon based reducing agents, also biogenic reducing agents like biomass or char coal can be used. Within an inter-sectoral recycling approach it can even be suitable to use e.g. SiC abrasive dusts or other materials suitable as reducing agents to recover metals like Iron or also Cr, Ni, Mo or others for alloying purposes from residues and by-products. Even low value primary raw material fines like iron ore fines, sieved undersize of alloying materials like FeSi or the sieved undersize of lime or dolomitic lime can be valuable ingredients in agglomerates flexibly tailor-made to the needs of each specific steel plant. So apart from basic self-reducing agglomerates for iron recovery also plant-specific slag former or alloying agglomerates without self-reducing characteristics are possible.
The approach followed within the project is the development of a process to produce cement-free bricks on the basis of primary and secondary raw material fines (Fig. 1), alternative binder systems and a hydraulic stamp press. The bricks have to possess sufficient cold compression strength for low-abrasion handling and, for self-reducing bricks, a sufficient reduction behaviour and metallurgical performance (metal yield). To achieve these goals the fundamental understanding of the bricks, their manufacturing and their subsequent use in the EAF is necessary. Important factors for the brick itself are granulometry, morphology and chemistry of the raw materials, their interaction with slag components within the brick, their behaviour during heat-up, their sinter behaviour, porosity of the brick, thermodynamics and kinetics of the processes within the brick, developing slag and metal phases etc. Therefore, defined residues from the EAF steelmaking are characterized in detail. Well-known and also custom developed methods are applied to these various materials. Regarding the EAF steelmaking process the influence of the bricks on slag (e.g. composition, viscosity etc.), metal and the processes energy and mass balance have to be investigated. In addition, the quality of slag after solidification concerning environmental behaviour and technical properties has to be analysed.

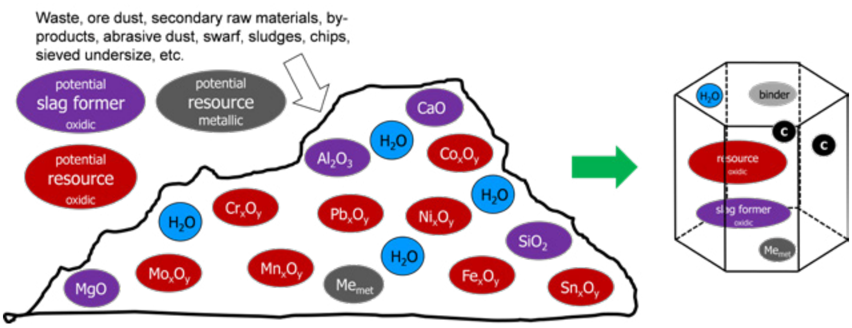

Fig. 1 – Schematic of the project approach

Another important factor in recycling raw material fines to EAF is to control the amount of volatile components in the fines. Excessive amount of volatiles causes them to enrich in EAF off-gas, which can cause problems in off-gas channel or in filter baghouse. To control the amount of volatiles in raw material fines, microwave technology is employed. Microwave allows selective heating of raw material fines. Fines with high amount of zinc oxides or alkali metals will be treated with microwave heating to reduce their amount to acceptable levels. The results from the microwave treated fines will be compared to the results from removing volatile components with conventional thermal treatment.
The use of hydraulic stamp presses instead of the vibration presses used in the cement-bonded brick production offers the advantage, that with this new process considerably finer material can be processed and that numerous other binder systems (e.g. organic binders) are feasible, which have no negative impact on the subsequent metallurgical process. Additionally, a plant concept based on hydraulic stamp presses will provide the opportunity to process relatively low amounts of raw materials (substantially less than 200,000 t/a) economically, due to the considerably smaller erection area needed and due to lower investment and maintenance costs than for briquette presses.
Currently only vibrating presses are used for the production of brick agglomerates because of the high throughput that can be realised with this kind of presses. If a lower material throughput is sufficient, as proposed in this project, stamp presses are an innovative alternative because of the increased pressing power of stamp presses. The pressing power can be increased by factor 25 or even 500 and up to 200 N/mm² going from a typical industrial vibrating press to "low tech" or "medium tech" stamp presses, leading to substantial increases in strength of the produced bricks, reducing the need for additional binder.
Both briquetting and conventional cement brick production have limits with regard to the grain size of the raw materials. The small briquette size usually strongly restricts the upper grain size limit while cement binding strongly limits the amount of fines (< 0.2 mm) allowed in a mixture. Pelletisation on the other hand usually needs a grinding treatment of the raw materials because the particle size for pelletisation has to be significantly below the pellet size of 9-16 mm. For cement-free bricks with alternative binders it is however expected, that they can consist of very fine material (< 0.02 mm) and material with a particle size > 18 mm at the same time due to different binding agents used in combination with the new pressing technology. So the proposed technology is also the most flexible with regard to different particle sizes.
For the recycling of metallic fractions in residues using self-reducing bricks, the new process offers the additional advantage, that particularly fine grained material, oxidic residue as well as reducing agent, can be processed. Because of the high surface area of the materials the reactivity and kinetics of reducing reactions are expected to be increased. Due to this, an optimised metallurgical performance even in the highly oxidising environment of a typical EAF in comparison to e.g. cement-bonded bricks or briquettes is expected.
To illustrate another innovative aspect of producing bricks cement-free, Fig. 2 shows typical compositions of cement-bound and cement-free bricks. Inherent to the use of cement as a binder in the vibrating press is the high water content of the brick. The water content also cannot be reduced as it is partly needed to reach sufficient plasticity of the mixture and partly chemically bonded water. Furthermore, the cement itself is at best neutral in the EAF process but can also have a negative impact on slag composition. Cement-free bricks usually have an increased "payload" (raw material and carbon carrier) of about 23 % directly increasing press capacity. If organic binders can be used, they can even add to e.g. the reduction capacity or heating value of the agglomerate.

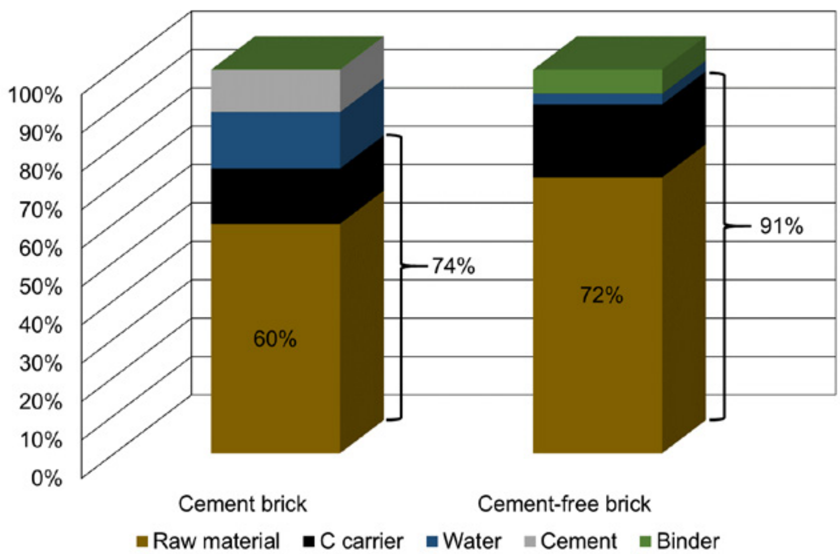

Fig. 2 – Typical composition of cement-bonded and cement-free bricks

Microwave technology is also a new promising technology, which can be applied in the processing of primary and secondary raw materials. Microwave energy has the potential to heat various kinds of metal oxides contained in iron and steelmaking dusts selectively, and in a commercial context may provide savings in both time and energy. The aim of this proposal is to develop a new method by using microwave technology to process the waste generated from iron and steel making industries. The advantages of microwave heating are: an environment friendly process, an efficient and rapid heating of minerals, and a mobile concept, so the same microwave device can be used for the treatment of wastes in different plants.
All in all the cement-free brick production technology developed in the project will contribute a new option for the closing of material loops within the EAF steelmaking route and at the same time provide new tailor-made high quality charge materials for iron input, slag forming or alloying elements for the EAF process.
As final results, following issues are expected from the project:
- treatment carried out inside the steel factory, in order to minimise expenses of transportation, also limiting the impact to the area around the steel plant,
- reduction of the amount of dumped materials,
- plant dimensioning proportional to steel plant productivity, avoiding needs of large material storage,
- the developed treatment can be applied to all the steel factories, independent of plant dimensioning,
- reducing the environmental impact and saving costs of raw materials.
All these innovative aspects represent an important upgrade of the current situation where waste and by-product fines like ladle slag, secondary dusts, sludges and other steel work residues cannot be reused in EAF steelmaking because of technologies to treat and reuse these materials are not available in small scale or not economic for a common use in steel plants.

SAMPLING AND CHARACTERISATION ACTIVITIES

In a first step of the Fines2EAF project, an extensive inventory of materials has been created. The inventory contains residuals, which have been collected from steelmaking operations but also from suppliers and other industrial sectors. These materials have been sampled and characterized by physical and chemical methods. The characterisation includes an optical assessment and documentation by sample photos and stereo-micrographies. The physical characterisation includes determination of moisture content, bulk density and true density, grain size distribution, and determination of the melting / softening behaviour by hot stage microscope. The chemical characterisation activities include elemental analysis by XRF spectroscopy, determination of carbonate amounts, specification of fractions and phases by SEM-BSE micrographies and SEM-EDS analysis, determination of weight loss and/or volatiles and phase transformations by TG-DSC analysis, phase determination by XRD analysis and for specific samples analysis for the iron oxides and metallic iron content.
From the steelmaking plants materials like ladle furnace slags, spent refractories, wet and dry mill scale, dusts collected from the EAF, LF, floor, roof or combustion chamber, oxygen cutting fines, fines from EAF and LF additions as well as sludge from water treatment were included. To broaden the scope of investigated materials further, interesting materials from related or other industrial sectors have been included in the inventory. These include filter dusts from FeMnC and FeSiMn production, used shot material, iron containing residues from pigment production, grinding sludge and a molybdenum concentrate. Tab. 1 gives the chemical composition of some of these materials. Especially interesting are here materials with a high metallic iron content but also materials, which could partially replace alloying materials.

Tab. 1 – Chemical composition of selected samples from other sectors (wt.%)

COMPONENT [1]	FESIMN FILTER DUST	FEMNC FILTER DUST	USED SHOT	IRON PIGMENT RESIDUE	GRINDING SLUDGE
Fe_2O_3	1.93	2.67	90.85	82.71	92.09
Fe met. [2]	n.a.	n.a.	n.a.	47.87 / 11.28 [3]	59.2
Al_2O_3	5.71	1.89	0.39	0.27	1.07
CaO	9.35	5.61	1.16	0.20	0.16
MgO	4.55	3.38	0.14	0.09	0.10
SiO_2	28.26	9.00	5.30	8.84	3.58
MnO	34.67	58.81	8.84	0.96	0.32
Cl	0.45	0.99	0.03	0.49	0.01
K_2O	9.60	9.68	0.06	0.02	0.03
Na_2O	1.26	3.90	-	-	0.11
C	n.a.	n.a.	1.57	6.75	3.03

1 all oxides calculated from XRF analysis; 2 analysed according to ISO 5416; 3 coarse / fine fraction were analysed separately; n.a. not analysed

MICROWAVE PRE-TREATMENT

A new pre-treatment technology for the removal of volatile components in the fines based on microwave heating is investigated. Microwave energy is a non-ionising electromagnetic radiation with frequencies in the range of 300 MHz to 300 GHz (4, 5). Microwave heating is fundamentally different from conventional heating because microwaves take the form of electromagnetic energy and can penetrate deep into the sample. This allows sample heating to be initiated volumetrically, as opposed to conventional thermal processing, which heats the sample from the outside inward via standard heat transfer mechanisms, i.e., through convection, conduction, and radiation (6). Compared with conventional heating techniques, the main advantages of microwave heating are: non-contact heating, energy transfer rather than heat transfer, saving energy, rapid heating, material selective heating and volumetric heating (4, 6).

Microwave pre-treatment is applied on the fines with high amount of zinc oxides or alkali metals to reduce their amount to acceptable levels. The results from the microwave treated fines will in future be compared to the results from removing volatile components with conventional thermal treatment. The aim is to produce raw materials with acceptable levels suitable for agglomeration and briquette production. The requirements for the raw materials used in agglomerate production are compared to characteristics of the available raw materials. The effect of microwave heating parameters (microwave power intensity, and exposure time, etc.) on the processing of wastes is studied on laboratory scale tests. The material treated in laboratory tests will be used in the lab-scale production of bricks.

Materials tested and experimental procedure

Tab. 2 gives the chemical analysis of material samples from steel plants. Politecnico di Milano and University of Leoben carried out the analysis.

Tab. 2 – Chemical composition of steel plant samples (wt.%)

COMPONENT	EAF DUST	COMBUSTION CHAMBER DUST	SLUDGE	FLOOR DUST	ROOF DUST	COMBUSTION RESIDUE
ZnO	39.76	7.77	14.58	2.24	0.27	4.11
Fe_2O_3	43.72	64.08	15.90	31.71	32.82	46.36
Na_2O	-	3.17	-	-	0.26	1.56
Cr_2O_3	0.84	1.28	0.60	1.14	1.90	0.80
MnO	3.57	3.45	15.16	3.42	4.52	1.74
CaO	2.90	9.30	20.97	36.87	26.18	31.06
MgO	1.40	1.95	8.77	7.66	3.65	1.58
SiO_2	2.24	4.36	9.79	8.68	22.25	8.43
C	1.07	2.57	7.12	4.65	3.89	1.29

In this preliminary investigation a conventional multimode microwave oven was used. The microwave absorption ability of the selected samples at a microwave power intensity of 1000 W were measured. The temperature of the sample was measured using a stainless steel-sheathed, K type thermocouple. The samples were placed in a microwave transparent crucible, in the centre of the microwave furnace. The microwave absorption ability of samples was measured by measuring the temperature increase of the sample using a thermocouple. In order to decrease the heat loss from the surface of the samples and to ensure efficient heating, the crucibles were insulated by alumina block, as shown in Fig. 3.

Fig. 3 – Image of the crucible insulated with alumina block.

In another set of experiments, the effects of microwave heating parameters (microwave power intensity, and heating time) on the removal of zinc from the selected wastes will be studied on laboratory scale tests. A first preliminary test was conducted with the EAF dust sample. The EAF dust was well mixed with 10 wt.% graphite in an agate mortar. The mixture of the EAF dust and graphite was then heated for 10, 15 and 20 min with a microwave power of 1000 W. The vapours from the crucible were evacuated by a pump and condensed particles were then collected by a paper filter inside a collector. At the end of the experiment, the residue remaining in the crucible was cooled to room temperature in the microwave oven (Fig. 4). Subsequently, the residue was sent for chemical analysis to determine the chemical composition and rate of zinc removal. Zinc removal (R) was calculated according to the following equation [1]:

$$R\ \% = \frac{C_0 - C}{C_0} \cdot 100 \qquad [1]$$

where C_0 is the initial Zn concentration and C is the Zn concentration in the solid residue.

Fig. 4 – Image of the crucible insulated with alumina block.

The chemical composition of the EAF dust sample is given in Tab. 2. The chemical composition of EAF dust demonstrated that iron and zinc were the dominant elements in EAF dust. The contents of Fe_2O_3 and ZnO were 43.72 and 39.76 wt.%, respectively before microwave heating. The EAF dust also contained Si, Ca, and Mn (Tab. 2).

Results of microwave pre-treatment tests

The temperature over time profiles at a microwave power of 1000 W are shown in Fig. 5. The tests indicate that EAF dust, combustion chamber dust and water treatment sludge are excellent microwave absorbing materials. The high microwave absorbing properties of EAF dust, combustion chamber dust and water treatment sludge can be attributed to the contents of carbon and iron oxides, which are classified as excellent microwave absorbers (4, 6). For example, when combustion chamber dust was heated in the microwave furnace at 1000 W for 5 minutes, the temperature of the EAF combustion chamber dust reached 850°C, the temperature of the EAF dust was 630°C and the sample of water treatment sludge under the same conditions was 723°C (Fig. 5). The measured sample temperature increases with increasing microwave heating time. The temperature increased very rapidly at first and thereafter increased slowly.

There are many factors influence the microwave heating such as: the mineralogical composition of the sample and the phase transformation during microwave heating (7, 8). For example, the composition of the materials changed during the microwave heating due to the reduction of the contained iron oxide and zinc ferrite to Fe. Iron oxide phase absorbs more energy from the microwaves as compared to reduced phase, which can be attributed to the initial rapid increase in the temperature (8).

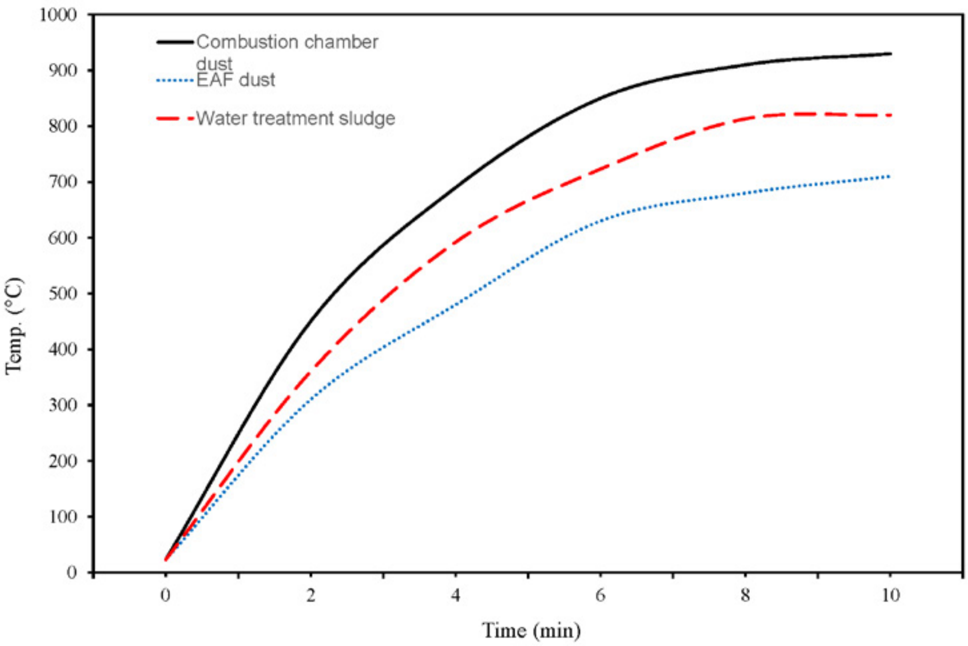

Fig. 5 – Microwave heating profile of combustion chamber dust, EAF dust, and water treatment sludge

The chemical composition of EAF dust after microwave heating indicated that, when the EAF dust was heated for 10 min at a microwave power of 1000 W, the zinc content was reduced by 53.49 %. Increasing the microwave heating time resulted in increased zinc removal rates of up to 90.43 % (Fig. 6). During the microwave heating process, zinc ferrite decomposes to ZnO and FeO as follow (9):

$$C + ZnFe_2O_4 = ZnO + 2\ FeO + CO\ (g) \quad [2]$$

$$CO\ (g) + ZnFe_2O_4 = ZnO + 2\ FeO + CO_2\ (g) \quad [3]$$

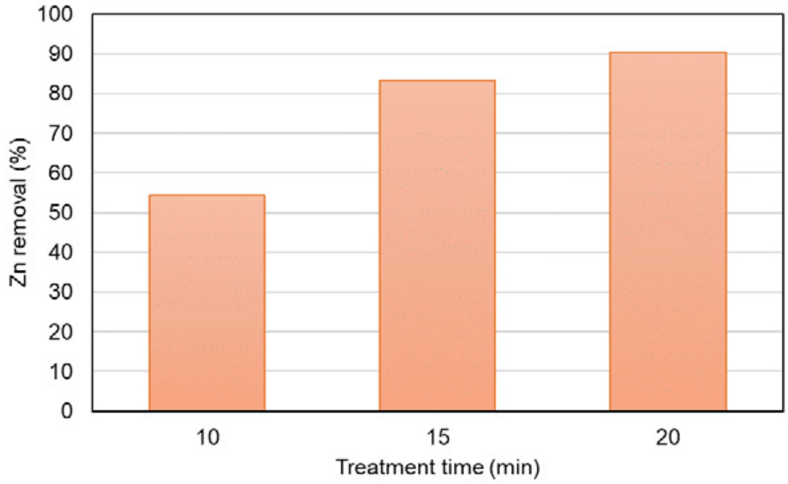

Fig. 6 – Zn removal from EAF dust

Subsequently, zinc oxide is reduced to elemental zinc vapour, according to equation [4] (9, 10). The reduction processes and the Boudouard mechanism (reaction [6]) produce a CO/CO_2 atmosphere. Gas/solid reactions between CO and ZnO reduce the zinc oxide to zinc vapour.

$$ZnO\ (s) + C\ (s) = Zn\ (g) + CO\ (g) \quad [4]$$

This reaction is a combination of the following reactions:

$$ZnO\ (s) + CO\ (g) = Zn\ (g) + CO_2\ (g) \quad [5]$$

$$CO_2\ (g) + C\ (s) = 2\ CO\ (g) \quad [6]$$

In addition, the overall lead, chloride and alkaline contents in the residue were reduced. The lead content in the residue was reduced from 0.85 wt.% to 0.27 wt.%. The chloride level was reduced from 1.27 wt.% to 0.28 wt.%. The K_2O concentration was significantly reduced from 0.86 wt.% to 0.08 wt.%.

AGGLOMERATION TESTS

With very fine and dry FeMnC filter dust first agglomeration tests with a carbon-based binder have been conducted. Two series of agglomerates have been produced with 4 wt.% binder (series A) and without binder (series B). Fig. 7 shows the resulting agglomerates.

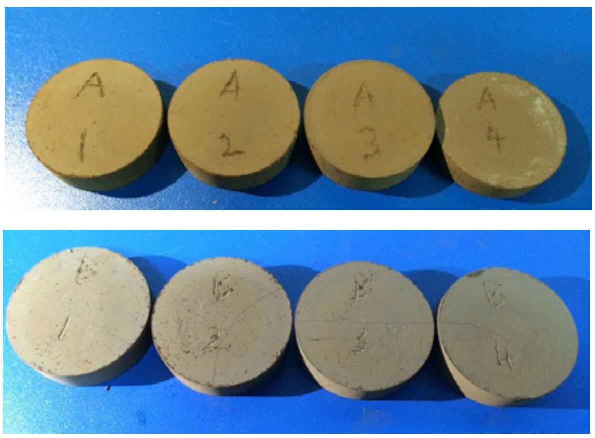

Fig. 7 – FeMnC filter dust agglomerates with (top) and without (bottom) binder (4 wt.%)

Already from Fig. 7 it can be seen that the agglomerates without binder show a number of cracks. Subsequently, a drop test was conducted by dropping the sample three times from a height of 2.5 m on the ground. This drop test let to the complete destruction of the samples of series B, while the samples of series A only showed a mean weight loss of about 1.5 % as can be seen in Fig. 8.

Fig. 8 – Two samples of series A (left) and of series B (right) after the drop test

These very first agglomeration tests will be followed up by further receipt development for single material and mixed material agglomerates according to the priorities of the steel plants participating in the Fines2EAF project. The receipts will be tested for agglomeration and resulting agglomerates will undergo comprehensive testing and characterisation.

CONCLUSION

Within the Fines2EAF project, a comprehensive inventory of residuals from the participating steel plants but also with materials from other industrial sectors has been established. The extensive physical and chemical analysis of the materials build the basis for the further work on a new microwave pretreatment technology as well as for the development of the agglomeration process based on a stamp press.

First tests of the microwave pre-treatment could already show that many of the residuals are suited for microwave heating and that it is possible to reduce the amount of zinc as well as the overall lead, chloride and alkaline content of the residues by a microwave pre-treatment. Further work in this area will investigate the energy demand and efficiency of the microwave pre-treatment in comparison to conventional methods. The process could be useful to reduce the lead, chloride and alkaline content to acceptable level for a further use in the EAF while producing a zinc-rich concentrate, which could be used in the Waelz kiln.

The initial agglomeration tests could already show, that with a stamp press very fine materials like the tested FeMnC filter dust can be agglomerated with about 4 wt.% of binder to durable agglomerate with only 1.5 % of weight loss in drop tests. In future more receipts with materials from the steel plants as well as from other sectors, pure or as blend, according to the needs of the steel plants will be tested. The created agglomerates will be evaluated extensively, including e.g. their melting behaviour. Finally, selected recipes will be used to produce agglomerates in pilot-scale and to test the agglomerates in the industrial EAFs of the project partners.

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