Zinc and Iron Recovery from Filter Dust by Melt Bath Injection into an Induction Furnace

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For simultaneous recovery of iron and zinc from filter dust, a new melt bath injection process has been tested in industrial environment. The test plant was built, implemented and operated by a consortium consisting of the recycling company DK Recycling und Roheisen, the plant manufacturer VELCO and the research institute VDEh-Betriebsforschungsinstitut (BFI). Main aspect of the new technology is the injection of pneumatically conveyable, Zn- and Fe-bearing filter dust via a submerged lance into the iron melt bath of an induction furnace. By reduction with carbon, metallic iron is formed, which is used as cast iron product. Main product is a high-grade zinc oxide product, which leaves the furnace via exhaust gas and is precipitated in a filter plant. For testing the process, the new injection plant technology has been built and implemented at an industrial 30 t-induction furnace at DK Recycling und Roheisen.

The research work carried out includes the definition of optimised operational parameters by operational trials and the development of a material- and energy balance process model for the melt bath injection used for supporting the process optimisation and for prediction of effects due to varied process parameters and the usage of different input materials. The operational trials resulted in a very good zinc and iron recovery especially producing a high-quality zinc oxide product with an average zinc content of 61 %. Process model calculations based on different input scenarios indicate different heat energy demands of the melt bath injection process depending on metallisation degree.

Keywords:

Melt bath – Hot metal – Induction furnace – Injection – Submerged lance – Zinc bearing residues – Zinc oxide

Rückgewinnung von Eisen und Zink aus Filterstaub durch Schmelzbad-Injektion in einem Induktionsofen

Zur gleichzeitigen Rückgewinnung von Eisen und Zink aus Filterstaub wurde ein neues Schmelzbad-Injektionsverfahren in industrieller Einsatzumgebung getestet. Die Testanlage wurde von einem Konsortium mit dem Recyclingunternehmen DK Recycling und Roheisen, dem Anlagenbauer VELCO und dem Forschungsinstitut VDEh-Betriebsforschungsinstitut (BFI) gebaut und betrieben. Die neue Technologie basiert auf der Injektion pneumatisch förderbarer zink- und eisenhaltiger Filterstäube mit einer Tauchlanze in das Eisen-Schmelzbad eines Induktionsofens. Durch die Reduktion mit Kohlenstoff wird metallisches Eisen gebildet, welches als Gusseisenprodukt genutzt wird. Hauptprodukt ist ein hochwertiges Zinkoxid-Produkt, welches den Ofen über den Abgasstrom verlässt und in einer Filteranlage abgeschieden wird. Die neue Schmelzbad-Injektionsanlage wurde an einem industriellen 30-t-Induktionsofen bei DK Recycling und Roheisen implementiert. Die durchgeführten Forschungsarbeiten

umfassten Betriebsversuche zur Bestimmung optimierter Prozessparameter, sowie die Entwicklung eines Stoff- und Energiebilanz-Prozessmodells für das Schmelzbad-Injektionsverfahren. Dieses unterstützt die Prozessoptimierung und prognostiziert die Auswirkungen variierter Prozessparameter sowie des Einsatzes verschiedener Einsatzstoffe. Die Betriebsversuche führten zu einer sehr guten Zink- und Eisenrückgewinnung, wobei insbesondere ein hochwertiges Zinkoxid-Produkt mit einem durchschnittlichen Zinkanteil von 61 % erzeugt wurde. Prozessmodellrechnungen mit verschiedenen Einsatz-Szenarien zeigen den jeweils unterschiedlichen Wärmeenergiebedarf des Schmelzbad-Injektionsverfahrens in Abhängigkeit des Metallisierungsgrads.

Schlüsselwörter:

Schmelzebad – Heißmetall – Induktionsofen – Injektion – Tauchlanze – Zinkhaltige Rückstände – Zinkoxid

Récupération de zinc et de fer sur la base de la poussière de filtrage et par injection dans le bain de fusion dans un four à induction

La recuperación de zinc y hierro desde filtro de polvo por inyección en baños de fusión dentro de un horno de inducción Paper presented on the ocassion of the Lead-Zinc Conference Pb-Zn 2015, June 14 to 17, 2015, in Düsseldorf, Germany This is a peer-reviewed article.

1 Introduction

For corrosion protection of steel, hot-dip or electrolytic galvanizing is a widely used application. About 50 % of the worldwide zinc production is used for galvanizing of steel as presented in Figure 1 [1]. Since several decades, the utilisation of zinc for galvanizing purposes is increasing steadily. Even in the last years, the worldwide zinc metal consumption has increased from 10.9 mill. t in 2009 to 13.0 mill. t in 2013 [1].

After end of use, galvanized steel products in large part are recycled as scrap to the EAF or BOF steelmaking process. A lesser part of the scrap is molten in foundries. The steel cycle is closed with the production of new (semi-finished) steel products, which may be galvanized again. During steelmaking or in the foundry melting furnaces the zinc coating on the galvanized steel evaporates and oxidises and leaves the process together with other particulate matter via the filter dust or sludge (depending on the dedusting process). That dust has to be processed for zinc enrichment and purification in order to obtain a secondary zinc oxide product, which is used for hydrometallurgical production of refined zinc. Most common large-scale technology for zinc enrichment e.g. from EAF dust with elevated zinc

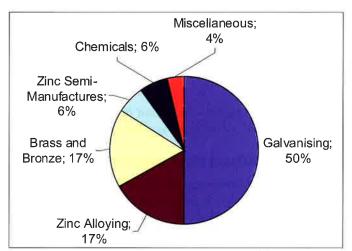


Fig. 1: Percentage of the worldwide end uses of zinc [1]

content is the Waelz process [2]. The refined zinc is used in hot dip galvanizing or electrolytic galvanizing for production of galvanized steel products. In this way, the zinc cycle is closed. In Figure 2 the simplified iron and zinc material cycle with regard to galvanized steel products is illustrated.

Currently, for processing of zinc containing steelworks' by-products (e.g. filter dust) in Europe, the following processes are – or have been – operated in industrial scale: for by-products with lower content of zinc the DK-process, the OxyCup Process or the RedIron process are used. Filter dust with a higher content of zinc may be processed besides the Waelz process also by the Primus process [3].

A gap still exists in the area of recycling filter dust with low and intermediate zinc content from small and medium melting plants like foundries. Due to the relatively low amount of filter dust at the particular plants, zinc (and iron) recovery within the above mentioned centralized industrial processes is too expensive, so a large part of these filter dusts are still landfilled.

This gap can be closed by the new melt bath injection process into an induction furnace, which is capable for inhouse operation for example at foundries. The investment costs are low, especially when using an existing induction furnace. This new technology can process filter dust without prior agglomeration and is able to recover the contained zinc and iron simultaneously. By using the new melt bath injection technology, small and medium melting plants are able to reduce disposal costs and become more independent from external disposal of waste. Further, they become more flexible to use a larger amount of cheaper, zinc coated scrap, which results in further cost savings.

2 Melt bath injection into the induction furnace

2.1 Process description

The new melt bath injection process into the induction furnace has been developed initially by BFI and DK based on operational trials using experimental equipment [4, 5].

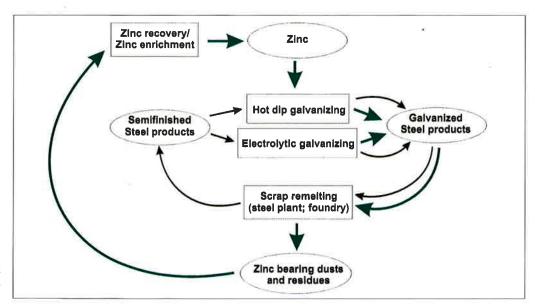


Fig. 2: Simplified iron and zinc material cycle with regard to galvanized steel products

Main point of the new developed recovery process is the pneumatic injection of fine-grained iron- and zinc bearing by-products into the hot-metal bath via a submerged injection lance. The injection is performed in an induction furnace, so that additional heating of the melting bath is possible when required. By reduction with carbon from the hot metal and/or the by-products, metallic iron is produced from the oxidic by-product, which passes on to the iron bath. Further, zinc oxide is reduced by the carbon forming metallic zinc gas and CO gas. If zinc is present in metallic state, it will evaporate from the melt bath. The zinc and CO gas leave the iron bath and combust to ZnO and CO₂ at the furnace atmosphere due to the access of air.

The main chemical reactions involved are as follows:

$$\begin{aligned} & \text{Fe}_2\text{O}_3 + 3\text{C} \rightarrow 2\text{Fe} + 3\text{CO(g)} & \text{(in induction furnace -} \\ & \text{hot metal)} \end{aligned} \end{aligned} \tag{1} \\ & \text{ZnO} + \text{C} \rightarrow \text{Zn(g)} + \text{CO(g)} & \text{(in induction furnace -} \\ & \text{hot metal)} \end{aligned}$$

$$2\text{Zn}(g) + O_2(g) \rightarrow 2\text{ZnO}$$
 (in furnace atmosphere) (3)
 $2\text{CO}(g) + O_2(g) \rightarrow 2\text{CO}_2(g)$ (in furnace atmosphere) (4)

For the combustion reaction a sufficient air or oxygen supply must be guaranteed by the suction of the filter plant or the infiltration or injection of secondary air or oxygen. The ZnO – as the main product of the process – is discharged via the exhaust gas and separated in the filter plant. Figure 3 shows a scheme of the new melt bath injection process. The zinc bearing by-product is conveyed with an inert conveying gas via the submerged lance into the iron bath. The inert conveying gas such as nitrogen or argon is necessary for preventing iron oxidation and decarburisation of the hot metal.

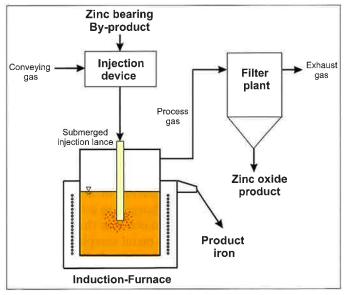


Fig. 3: Scheme of the new melt bath injection process

The main aims of the new process are the production of a high-quality zinc oxide product from the processed byproducts for use in the primary zinc metallurgy as well as the recovery of iron within the iron melting process. The carbon of the by-product acts as reductant for zinc- and iron oxide. If required, additional carbon carrier can be added or injected as reductant.

2.2 Potential input by-products

Potential zinc- and iron bearing by-products to be processed by the new melt bath injection process are mainly dry filter dusts of shaft furnaces (e.g. foundry cupola furnace, recycling shaft furnace) or foundry induction furnaces, which are produced by small and medium melting plants. In Table 1 typical analytical data of these dusts are listed (mainly based on operational data). From technical point of view many other zinc bearing by-products from the steel industry or nonferrous metallurgy may also be suitable to be used within the melt bath injection process.

Especially both shaft furnace dusts have high carbon contents, originated from the coke. The contained carbon during melt bath injection is used as a reductant for the contained zinc- and iron oxide. Due to the lack of carbon within the filter dust from foundry induction furnaces, here additional carbon has to be added as reductant when processing the dust within the melt bath injection process. The typical mass percentage of zinc within the foundry dusts (Cupola furnace, induction furnace) strongly depends on the zinc content within the used input scrap (zinc coated steel) and may vary in a broad range between 0 and 30 %. The specific amount of these filter dusts is in the range of up to 20 kg per tonne of hot metal respectively steel.

2.3 Melt bath injection test plant

In the course of a research project, DK Recycling and Roheisen in cooperation with the plant manufacturer VEL-CO have built a melt bath injection test plant based on an existing induction furnace (crucible type) with a maximum capacity of around 30 t of hot metal.

The melt bath injection test plant is designed to process the own dust from the recycling blast furnace at DK with an injection rate of around 50 kg/min.

In Figure 4 the components of the melt bath injection system built by VELCO are illustrated.

The pneumatic conveying device has a material chamber (pressure vessel) with a volume of 2.5 m³ and a capacity of around 1600 kg dust. Nitrogen is used as conveying gas. The nitrogen consumption is at maximum 100 m³ _n/h. The conveying rate is determined by the pressure of the material chamber, which is kept constant at the preset value during

By-product C Fe Zn

Cupola furnace dust (shaft furnace) up to 30 % up to 22 % 1.5-20 % 1)

Dust from recycling blast furnace (shaft furnace) ca. 25 % ca. 20 % up to 30 %

Filter dust from foundry induction furnace low ca. 12 % [6] 0-30 % 1)2)

1) strongly dependent on Zn content in input scrap; 2) estimated

Table 1:
Typical analytical data of potential filter dusts, capable for processing by the new melt bath injection process [mass.-%]

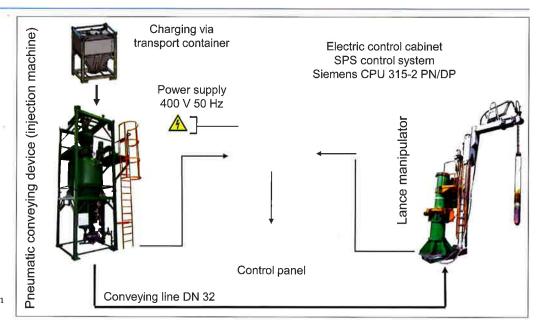


Fig. 4: Components of melt bath injection system (VELCO)

conveying. A weighing system at the material chamber is used for monitoring and control of the conveying process. The pneumatic conveying device is charged with the dust via a transport container. The dust is conveyed to the injection lance at the lance manipulator via a DN 32 conveying line. The lance manipulator holds and moves a refractory covered monolithic injection lance with an outlet port diameter of 18 mm. The lifting range of the lance manipulator is 3600 mm. The overall melt bath injection system is controlled by an SPS control system with a control panel as user interface.

The zinc oxide product is separated in a bag filter plant with a maximum volumetric flow rate of 40,000 m³/h. The filter plant has 192 filter bags with a filter area of 460 m². The filter bags consist of a glass fabric with PTFE membrane, allowing an operating temperature of up to 250 °C.

By the control system, the operation of the pneumatic conveying device and the movement of the lance manipulator are closely connected. When starting the injection sequence, first the lance manipulator moves the lance tip in a position near above the melt bath surface. Then the



Fig. 5: Induction furnace during the melt bath injection process

pneumatic conveying is started immediately before the lance is immersed into the melt bath. This prevents the hot metal from entering the injection lance, which would cause lance blocking after a short time. For security reasons the pressure in the conveying line is monitored continuously. If any stoppage of the material flow or any blocking of the conveying line should be detected, the injection operation is stopped automatically, the lance is lifted outside the melt bath and the pneumatic conveying device starts a special sequence for unblocking the conveying line. Even in case of downstream operational disruptions (e.g. in the filter plant) the injection operation is stopped automatically.

Figure 5 illustrates the running melt bath injection process at the induction furnace as viewed from the operating platform.

3 Operational trials

3.1 Performance and parameters

The operational trials concerning the new melt bath injection process have been performed at DK's industrial induction furnace. The injected shaft furnace filter dust in average contains 30 % Zn, 16 % Fe, 26 % C and a very low amount of slag forming components. One operational injection trial comprises the injection of around 200 kg filter dust. The initial hot metal charge was used for several injection trials. Usually, the hot metal was changed once a day. For material balancing, hot metal samples have been taken from the initial hot metal charge and after each injection trial. In order to keep the hot metal temperature as far as possible at the same level, the heating of the induction furnace was operated at low power during the melt bath injection trials.

As base data for material- and energy balancing (Process model), the following parameters have been gathered or measured:

- mass and temperature of hot metal,
- electric energy consumption of the induction furnace,

- mass flow of injected filter dust,
- volumetric flow rate and temperature of exhaust gas,
- · dust concentration within the exhaust gas and finally
- the chemical composition of hot metal and zinc oxide product.

Main aim of the operational injection trials was the determination of optimum injection parameters with regard to maximisation of the zinc oxide product quality (maximum Zn enrichment; minimum content of undesired components) and maximisation of the process productivity (maximum injection mass flow). For this reason, the two operational parameters injection mass flow and hot metal temperature have been varied.

The injection mass flow has been varied in three steps by adjustment of the vessel pressure of the pneumatic conveying device. In this way the injection mass flow was varied in the range of around 30 to 60 kg/min. For each injection mass flow, the hot metal temperature has been varied in two steps. Two temperature levels of the hot metal have been applied by manual adjustment of the heating power of the induction furnace: The normal hot metal input temperature level was in average 1335 °C, while the high hot metal input temperature level was in average 1420 °C.

3.2 Results

The major aim of the submerged injection process is the production of a zinc oxide product with a low content of by-components and a particularly high zinc content. This aim was achieved well during the operational trials, since the average mass fraction of zinc in the zinc oxide product was 61 % for the overall injection campaign. The average chemical composition of the zinc oxide product obtained during the operational injection trials is listed in Table 2. The content of Fe, mineral compounds (CaO, SiO₂), Pb and Cl with <2 % in each case is low. The carbon content of 16 % results from unreacted carbon from the input dust (from recycling blast furnace). Further, during the injection trials a certain accumulation of unreacted carbon on top of the melt bath was observed. In the specific input dust, the carbon is supposed to have a low reactivity, which prevents a complete incineration within the short available time, leaving some residual carbon within the zinc oxide product and on top of the melt bath.

Table 2: Average chemical composition of the obtained zinc oxide product

Element/Component	Zn	Fe	С	CaO+SiO ₂	Pb	Cl
Mass fraction [%]	61	<2	16	<1	1.4	<1

In Figure 6 the visual comparison between the greyish input filter dust and the yellowish-white zinc oxide product is illustrated.

During the injection trials, also with the highest injection mass flow of around 55 to 60 kg/min, without exception, in the zinc oxide product a very high zinc content in the range of 64 % was obtained. This zinc content was amongst the highest of the overall trial campaign. This result reveals that high process productivity is achievable without any detrimental influence on the zinc oxide quality.



Fig. 6: Comparison input filter dust and zinc oxide product

In contrast to that, no clear correlation between the hot metal temperature and the absolute value of the zinc content within the zinc oxide product was observed. But there is a tendency, that even when zinc content within the injected input dust is lower, at high hot metal temperature the resulting zinc content within the zinc oxide product is on the same level as obtained at normal hot metal temperature with input dust having a higher zinc content. In other words: an increased hot metal temperature seems to promote zinc enrichment into the zinc oxide product. This can be explained by the faster zinc evaporation from the (iron) melt bath with increasing hot metal temperature.

With the relatively low amount of injected input dust during one injection trial (related to the amount of hot metal), the initial zinc-lean hot metal at first solves a significant part of the zinc and in this way acts as a zinc buffer. So, the zinc content in the hot metal with increasing amount of injected input dust (respectively increasing number of trials) will rise until it is saturated. During the melt bath injection trials the analysed zinc content of the hot metal increased from initially <0.008 % to a maximum of partly above 0.15 % after several injection trials. Nevertheless, the obtained hot metal quality with regard to the zinc content is sufficient for further processing in foundry applications.

The fact that a zinc saturation level is reached in the hot metal after some melt bath injection trials leads to the conclusion that at long term operation of the new melt bath injection process, the zinc buffering of the hot metal bath will be negligible, so that roughly 100 % of the zinc will finally be distributed to the zinc oxide product. This zinc partition is assumed as follows in the process model of the melt bath injection. For removal of zinc from the hot metal before tapping, a heating and holding treatment within the induction furnace has further been tested. Here, the zinc content within the hot metal was decreased by up to factor 4.

4 Process model

In order to support the optimisation of the new melt bath injection process, and to be able to predict the effects of varied process parameters and the usage of different input materials, the VDEh-Betriebsforschungsinstitut GmbH has developed a flow sheet process model by use of the HSC-Chemistry® software. The process model is a material

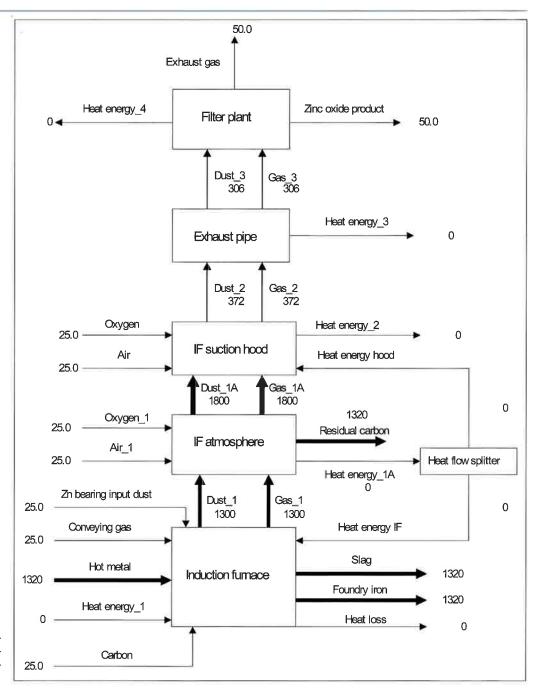


Fig. 7: Flow sheet process model of induction furnace melt bath injection (HSC Chemistry; values: temperature in °C)

and energy balance model using different process units and material and energy streams. Different units are connected with specified streams. The process model is able to calculate the amount, composition, temperature and enthalpy of all streams.

Figure 7 shows the layout of the developed flow sheet process model of the induction furnace melt bath injection. Exemplary, the temperature values of the material streams are given in the flow sheet.

The process units of the melt bath injection are the induction furnace (IF, melt bath), the IF atmosphere, the IF suction hood, the exhaust pipe and finally the filter plant. At the DK melt bath injection test plant, the furnace atmosphere is separated from the furnace suction hood by a refractory top cover. This division of reaction chambers is also part of the process model.

The main input streams of the induction furnace are the hot metal, the input dust and the (electric) heat supply. The products of this unit are cast iron and slag. Solid matter and gas streams lead from each unit to the following unit. The main air entry into the system due to the induced draught of the filter plant is within the furnace suction hood. A minor part of air enters into the furnace atmosphere. Finally, at the filter plant the zinc oxide product is discharged.

At each process unit thermal energy is released ("heat loss") for example due to convection and radiation. Only the thermal energy released in the furnace atmosphere due to the combustion of zinc (gas) and CO gas, is divided into two thermal energy streams. One minor stream heats up the induction furnace and the larger stream enters the furnace suction hood and heats up the exhaust gas.

For the three process units induction furnace, furnace atmosphere and furnace suction hood, specific chemical reactions are defined (e.g. reactions (1) to (4)) together with a coefficient of the respective reaction progress. The reaction progress coefficient is set according operational experiences.

Further, the following boundary conditions are part of the melt bath injection process model:

- The temperature of output material streams is calculated based on the precondition "Enthalpy total = 0" within the respective unit.
- The hot metal temperature is kept constant by adjusted inductive heating.
- The carbon content in hot metal product is fixed to 3.9 %.
- The zinc content in hot metal product is assumed to remain unchanged (no long-term zinc accumulation within hot metal).
- The partition of heat energy flow from furnace atmosphere to induction furnace is fixed to 25 % of the total energy flow (remaining energy to furnace suction hood respectively exhaust gas).
- The temperature within furnace atmosphere is fixed.
- The heat loss of induction furnace, furnace suction hood, exhaust gas line and filter plant fixed individually based on heat loss calculations (strongly dependent on plant layout).

5 Model calculations

By use of the melt bath injection process model, three scenarios regarding different metallisation state of the input dust have been calculated in order to predict the effects on temperatures and energy consumption. In each case, for the model calculations an amount of 1000 kg input dust is assumed. Further, the specific amount or volumetric flow rate of hot metal, conveying gas and exhaust gas is set according operational experience at the melt bath injection test plant.

Background of the different calculation scenarios is the unknown and varying metallisation degree of Zn and Fe in the injected DK input dust. This input dust is originated from a recycling blast furnace, so due to the reducing conditions initially a large partition of Zn and Fe is assumed to be present in metallic form. Depending on the storage conditions an unknown partition of these metals will be oxidised before injection.

For the model calculations the metallisation degree (in %) of the input dust is varied in the following three steps:

1.	Zn:0%,	Fe: 0 %,
2.	Zn: 50 %,	Fe: 100 %,

3. Zn: 100 %, Fe: 100 %.

Here, the second case (Zn: 50 %, Fe: 100 %) is assumed to be close to the operational conditions since zinc is oxidised more easily than iron. In each case one basic chemical analysis of the DK input dust has been taken into account for the calculations: Zn: 33.5 %, Fe: 14.5 %, C: 23.2 % plus several minor elements and components. According the specified metallisation degree, the given percentage of Zn and Fe is converted into the respective partition of the metallic state and the oxide (ZnO respectively Fe_2O_3) and finally the overall chemical composition is normalized to 100 %.

The following main parameters have been calculated: The amount of slag product and zinc oxide product, the resulting temperature within the induction furnace suction hood as well as the composition of C and Zn within the zinc oxide product. In Table 3 the main results of the model calculations are listed.

With increasing metallisation degree, the required heat input (electric power consumption) at the induction furnace decreases, because in these cases less (no) endothermic reduction reactions take place within the melt bath. Further, with increasing metallisation degree, the temperature at the furnace suction hood decreases, since less CO gas is formed (which combusts to CO₂ and thus releases additional heat) at the furnace suction hood due to less carbothermic reduction reactions.

A higher amount of slag product results from the calculations when iron within the input dust is 100 % oxidic (0 % metallisation degree) since more iron oxide reports to the slag in this case. The amount and the main composition of the zinc oxide product are comparable for all three calculation scenarios, so in the table only average values are reported.

6 Conclusions and outlook

By the melt bath injection process a very good zinc yield into the zinc oxide product was achieved. The zinc oxide product is a high-quality product with an average Zn mass fraction of 61 %, with only low amount of disturbing by-components.

The significant carbon content within the zinc oxide product still leaves room for improvement. Connected with the carbon in the zinc oxide product is a certain amount of residual carbon on top of the melt bath. One possible way for decreasing the carbon content of the zinc oxide product as well as the amount of residual carbon on the melt bath

Percentage within zinc Metallisation degree of Heat input Temperature Amount [kWh] oxide product [mass-%] input dust [mass-%] [°C] [kg] Zn Fe Induction Furnace Slag Zinc oxide Zn product suction hood product furnace 0 0 935 421 121 average: 562 average: 11 average: 69 100 50 673 372 92 100 100 512 348 97

Table 3: Main results of the model calculations

could be the adjusted oxygen supply into the furnace atmosphere in order to improve carbon incineration. This might be a topic for future research work.

The most important result of the operational melt bath injection trials is the fact that a high injection mass flow of 60 kg/min is applicable without restrictions in the zinc oxide product quality. This means, the melt bath injection process can be operated at (or slightly above) design capacity without any problems, confirming the high productivity of the process.

Still missing is the proof of long-term process stability during industrial operation, which exclusively can provide a sound basis for calculation of process economics. During the past injection trials, each with a restricted amount of injected dust, no reliable information for example on refractory wear, lance consumption and auxiliary process times, could be obtained. These information can only be provided by long-term industrial pilot plant trials. Further research work is also required for obtaining operational and economic data regarding the processing of other types of by-products by induction furnace melt bath injection, like for example filter dusts with lower Zn and/or C content.

Another topic for future research is the energetic optimisation of the induction furnace melt bath injection process. Here for example measures for heat recovery are an important topic.

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