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# ENVIRONMENTAL ASPECT OF INDUSTRIAL TECHNOLOGIES FOR RECYCLING OF FE – AND ZN-CONTAINING SLUDGE AND DUST[1]

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**Abstract**
We performed the analysis of bleed-through $CO_2$ emission during the recycling of BOF and BF sludges by means of their briquetting and using such briquettes in ironmaking in line with different process routes: using in blast furnace raw briquettes; using in blast furnace briquettes after prereduction them in a rotary hearth furnace or after prereduction them in a tube rotary kiln by Waeltz process; melting of raw briquettes in an OxyCup cupola. Minimal $CO_2$ emission can be achieved by using in a blast furnace raw briquettes or briquettes after prereduction them in a rotary hearth furnace.
**Key words:** $CO_2$ emission; Fe- and Zn-containing sludges; Recycling; Blast furnace process.

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## 1 INTRODUCTION

The production of 1 ton of steel requires 18-20 GJ of fuel energy and is accompanied by the emission of 1.7 t of $CO_2$ into the atmosphere which in addition generates 20-25 kg of sludges and dust containing up to 60% of iron and up to 2.5% of zinc, which interferes with their recycling in blast furnaces [1]. The situation is the same with blast furnace sludges, which also contain up to 1.5% of zinc. The rapid growth of the world steel production in the end of the twentieth and in the beginning of the twenty first centuries made the environmentalists, the metallurgists and the governments of industrial countries focus on the impact of iron and steel industry upon the environment, firstly due to the greenhouse gases' emission, generation and accumulation of the specified wastes which are hard to recycle and hazardous to deposit. Due to that the development of economically efficient process for recycling of these technogeneous raw materials, which is in line with environmental requirements is an important objective for ferrous metallurgy first of all for integrated steel plants.

## 2 DISCUSSION

At present there are rather efficient industrial processes developed, used at some of the plants, for recycling of Fe- and Zn-containing sludges and dusts, the aim of which is to extract from these wastes not only iron as a prereduced semi-product or as a pig iron, but also to extract zinc oxide. Such basic processes are as follows:

- special blast furnace melting of Zn-containing wastes in lump form (DK Recycling technology);

– prereduction in rotary hearth chamber furnaces of pellets and briquettes from Fe- and Zn-containing wastes (Fastmet, RedIron processes);

– OXYCUP process – melting of Fe- and Zn-containing wastes in oxygen cupola;

– FASTMELT process – prereduction in a rotary hearth furnace of pellets and briquettes from Fe- and Zn-containing wastes with subsequent melting of pig iron from them in an electrical arc iron melting furnace;

- Waelz-process – prereduction of Fe- and Zn-containing wastes in rotary kilns.

The selection of a reasonable process for recycling of Fe- and Zn-containing wastes for the conditions of particular plant shall be done taking into account both economical and environmental criteria, as well as the specifics of the plant's production structure. The exclusive attention of the global community to the green-house effect and the measures taken to counteract it makes it necessary to use as an ecological criterion the total (summarized) $CO_2$ emission for 1 t of pig iron, made with the use of recycled wastes [2].

With the use of this criterion for the conditions of NLMK the following routes of Fe- and Zn-containing wastes recycling have been analyzed:

Option 1 – wastes briquetting + melting of briquettes in one blast furnace (BF);

Option 2 – wastes briquetting + prereduction of briquettes in a rotary hearth furnace + melting of DRI in a blast furnace (BF);

Option 3 – wastes briquetting + melting of briquettes in OXYCUP cupola

Option 4 – wastes briquetting + prereduction of briquettes by Waelz process + melting of DRI in a blast furnace

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The degree of zinc removal in all processes is assumed as 95%. The degree of prereduction of iron in a rotary hearth furnace or in a tubular rotary kiln (Waelz process) is 85%.

The option of melting briquettes in a BF is possible to realize in iron and steel making plants, which have excessive facilities for pig iron production, where a BF of minimal volume is possible to use for recycling of such wastes. In such case the investments into the recycling route are limited by the reconstruction of a BF in connection with its new purpose.

$CO_2$ emission in the recycling of current and accumulated Zn-containing wastes (table 1) by different routes under conditions of NLMK was estimated for the volume of wastes 600 thousand t per year.

**Table 1.** The composition of the mixture of current sludges and the ones extracted from the sludge accumulators

| Components | $Fe_{total}$ | FeO | CaO | $SiO_2$ | MgO | $Al_2O_3$ | ZnO | C |
|---|---|---|---|---|---|---|---|---|
| Content, % | 46,5 | 25 | 10,4 | 6 | 2 | 2 | 2 | 13 |

Such approach is caused by the established objective – to liquidate within 10-15 years the sludge stocks in the form of existing sludge fields accumulated during the years of the company operation with the subsequent reclaiming of the grounds, occupied by them.

In all the specified technologies the first stage of wastes recycling is the pelletizing of wastes. The method of hard extrusion was chosen as a technology for pelletizing. The products of extrusion – cylindrical briquettes 20-30 mm in diameter and 40-80 mm in length, by their size and strength characteristics are suitable both for melting in blast furnaces and in OXYCUP cupola, as well as for prereduction in a rotary hearth furnace and in rotary kilns.

The fuel consumption for prereduction processes in rotary kilns and in rotary hearth furnaces, as well as in OXYCUP process were assumed on the basis of the published data. Coke, oxygen and natural gas consumption rates at melting the briquettes and prereduced materials in a blast furnace and in were received as a result of the computer simulation of the blast furnace process. The fuel consumption at the production of sinter and high-temperature hardening of pellets, used in the BF at melting of the said briquettes and metalized materials, have also been taken into account.

DK Recycling process consists of pelletizing by the way of sintering the mixture of steel making and other zinc containing wastes and melting casting iron out of this sinter (300 thousand t/year) in the BF (working volume 580 m3) using special technology, providing efficient zinc oxide removal with the top gas. Of all the zinc, which goes to the blast furnace shop together with the burden (38 kg/t of the pig iron) 96% is removed from the BF together with the gas and is caught in the gas cleaning system together with the top gas and the sludge (17000 t/year), which contain up to 40 % and 65-68 % of zinc, respectively. Only 2 % of zinc is removed with pig iron and slag and 4 % of zinc is caught together with dust [3].

The concept for recycling of accrued and current Fe- and Zn-containing sludges, accepted at NLMK[4] provides for briquetting and melting of Zn-contain sludge briquettes in the BF with the working volume 870 $m^3$ and based on the appearance of excessive pig iron production facilities in the company after the commissioning of the new BF. The design of the process route for Fe- and Zn-containing sludges recycling is given on figure 1.

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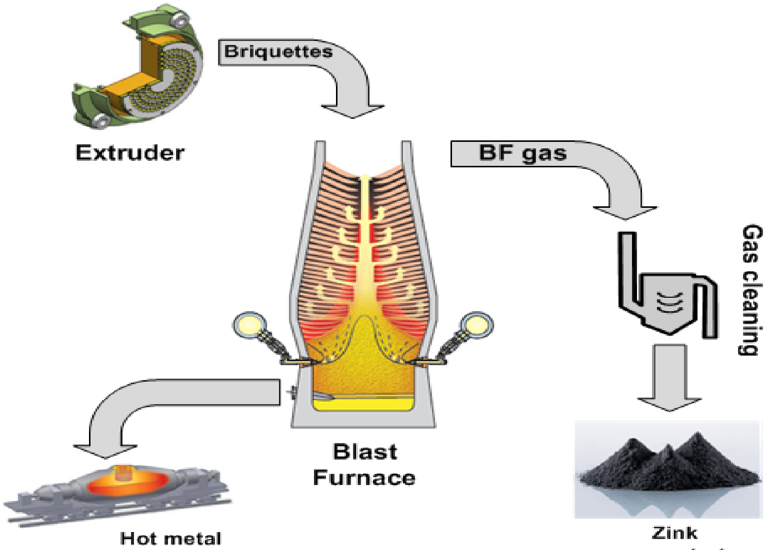


**Figure 1.** Melting briquettes in a blast furnace (DK Recycling process).

The prereduction of pellets and briquettes from Fe- & Zn-containing wastes in rotary hearth furnaces, as a technology, was developed by Midland-Ross in USA in 1960. Nowadays it is used in the plants of Nippon Steel Co in Japan (Fastmet process) [5] and in Lucchini-Piombino plant in Italy (RedIron process) [6,7]. Due to the high rate of heating of the thin layer of carbon containing pellets (1–2 pellets in height) up to 1100–1200 °C the process of iron and zinc reduction in the pellets is finished in the rotary hearth furnace within 10–15 minutes during one rotation of the hearth. After the reduction the zinc is sublimed, it comes out of the furnace with waste gases and is caught in the form of dust in the gas cleaning unit. The prereduction rate for pellets makes 60–85%, and the rate for removal of zinc from them – not less than 80–95% [5,6]. Hot pellets go to the roll press, producing hot briquetted iron (HBI) [6,7]. At prereduction of pellets in Fastmet process from iron ore concentrate the fuel consumption per one t of prereduced product makes: Coal – 0.3-0.4 t, fuel to be burned in the burners, - 70-100 kg of equiv. fuel .The consumption of fuel at prereduction of pellets from carbon containing sludges makes 100-130 kg of equiv. fuel [8]. The process diagram of sludges recycling according to this method is given at figure 2.

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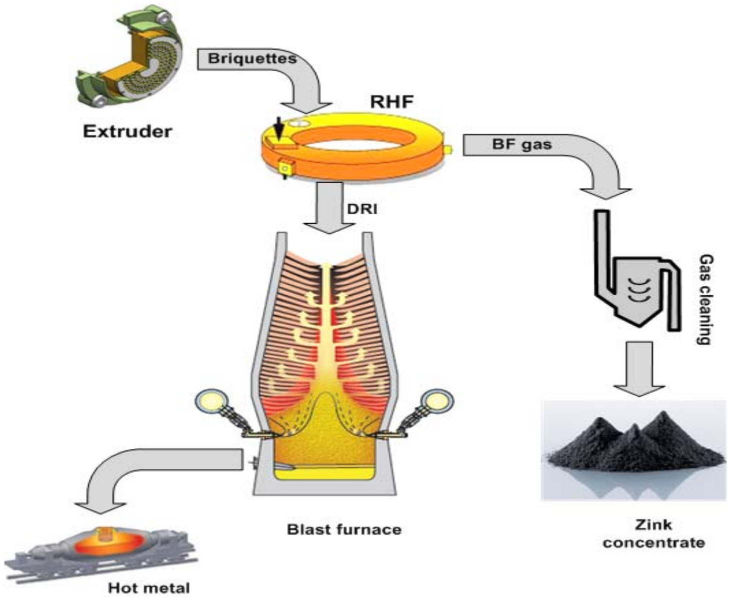


**Figure 2.** Metallization (prereduction) of briquettes in a ring chamber furnace with rotating floor (Fastmet, Rediron) and the use of the received DRI in blast furnace process.

OXYCUP process, developed by Küttner GmbH & Co, was implemented at the industrial scale in 2004 in Hambourne by ThyssenKrupp Steel. The self-reduced briquettes from sludges with cement binding agent, produced by the process of vibropressing are molten in cupola furnace, working at the blast enriched with oxygen. In addition to the briquettes, acid flux and large size (up to 1 m) slag contaminated scrap are also used, which are received at desulfurization of pig iron. The resulting hot metal is mixed with the blast furnace iron and after the desulfurization is used in the BOF shop. The slag which similar in composition to the blast furnace slags is granulated, the treated top gas is used as a fuel and the top dust is recycled into briquettes. At the increase of zinc content in the top dust up to the level defining its liquidity, the dust is regularly withdrawn from the process and sold as zinc concentrate. Unlike the blast furnace process, in OXYCUP process the bigger part of iron is reduced at 900–1400 °C during some short period of time (20–30 minutes) mostly by carbon, contained in briquettes. Apart from Germany, four OXYCUP modules, constructed according to the design by Küttner GmbH & Co, are operated in Japan, Mexico and China[9,10]. The diagram of sludges recycling according to this technology is given at figure 3.

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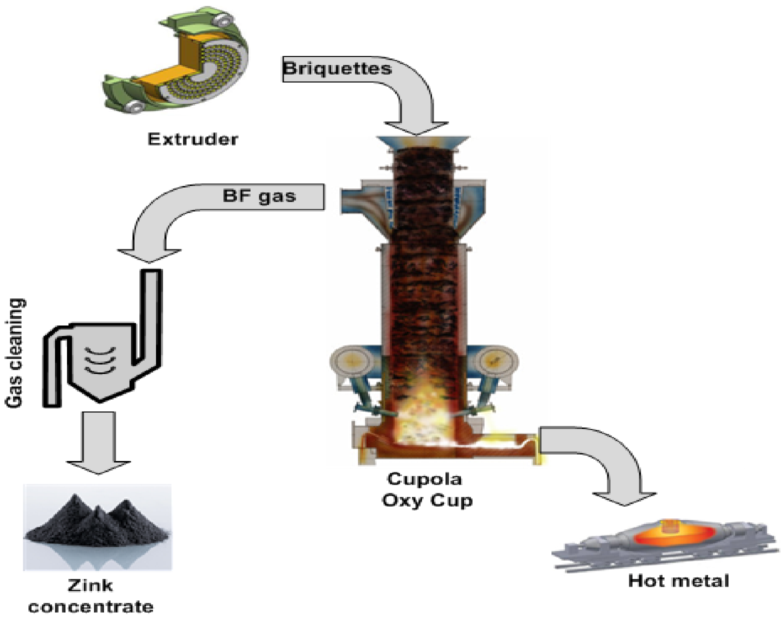


**Figure 3.** Melting of briquettes in OxyCup cupola.

Waelz process was developed especially for processing iron ores and industrial wastes for the purpose of extraction of metals from them, which are to be sublimed in the form of vapors, sulfides, oxides or chlorides. The main industrial application was in recycling of EAF dust, rich in zinc (25–35%). The minimum zinc content in wastes which makes their processing by Waelz-process economically efficient is not less than 9% [11]. The process is carried out in rotary tubular kilns 2.5-4.5 m in diameter and 40-75 m in length. Before waelzing small sized wastes shall be pelletized with the use of binding agent.

The main heat sources in the Waelz-process are the oxidation of the solid fuel carbon (coke breeze), which is charged to the furnace together with the processed material (80%), the fuel burned in the burners (app. 10%), oxidation of the sublimed zinc (app. 7%) and physical heat of the blast. Heat losses with waste gases, sublimates, dust and external losses made up to 50%. The production output varies within the range of 0.5–1.3 t/$m^3$ per day. Coke breeze consumption makes 450 kg/t of charge and natural gas consumption makes 29 $m^3$/t[11]. Even higher consumption of fuel takes place in a similar process of prereduction of iron ores which are difficult to enrich in tubular rotating kilns [8]. The route for recycling of Zn-containing sludges with the use of Waelz process is given at figure 4.

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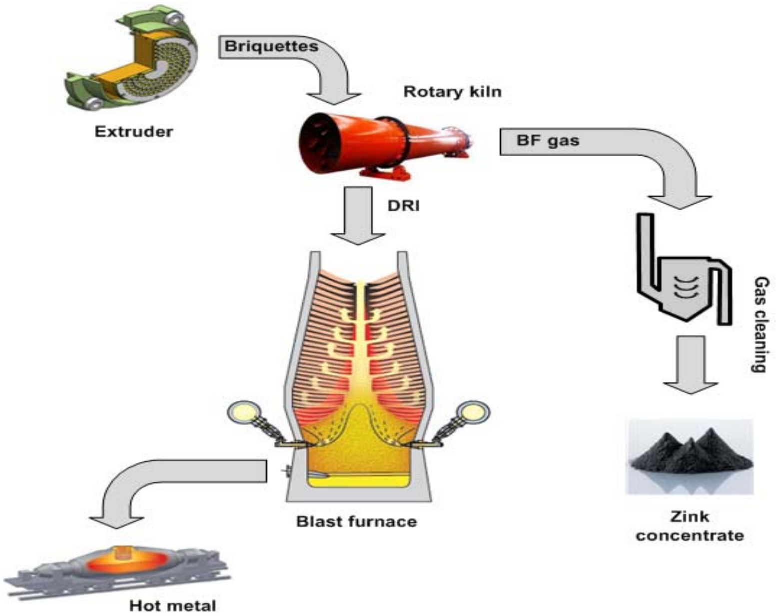


**Figure 4.** Metallization of briquettes by Waeltz process and the use of the resulting DRI in blast furnace process.

At the analysis of fuel consumption and $CO_2$ emission in the proposed options cement bonded extrusion briquettes were used (6% of briquetting mixture), prereduced materials, produced out of them (Table 2). At the accepted prereduction rate in rotary hearth furnace and in rotary kilns 85% output of prereduced materials makes 70%.

**Table 2.** Chemical composition of raw products and prereduced materials

| Components | Content, % | |
|---|---|---|
| | Briquettes | Metalized (prereduced) briquettes |
| Fe | 43,8* | 52,7** |
| FeO | 23,5 | 12,0 |
| CaO | 13,3 | 17,7 |
| $SiO_2$ | 6,8 | 9,6 |
| MgO | 2,2 | 3,1 |
| $Al_2O_3$ | 2,7 | 3,8 |
| ZnO | 1,9 | 0,1 |
| C | 12,2 | 1,0 |

*Note: * $Fe_{total}$; ** $Fe_{met}$*

In order to evaluate the fuel consumption and total emission of $CO_2$ according to the considered process routes the results of computer simulation of the blast furnace melting with the use of metalized (options 2, 3) and raw briquettes (option 1) were used, as well as the published data on the fuel consumption for prereduction [5-7,8,11]. For OXYCUP process (option 4) we used the published process values [9].

At simulation of blast furnace melting with the use of raw briquettes, the temperature (theoretical burning temperature), gas dynamical (humidity and blast temperature, top gas pressure) and slag oriented (slag basicity) modes of operation were set, which contribute to the maximum zinc removal with the top gas (table 3). The total carbon consumption (table 4) was evaluated taking into account the consumption of fuel for the production of sinter, pellets calculating, briquettes prereduction and production of electrical power, consumed for oxygen, consumed in the processes of pig iron melting calculated per 1 ton of pig iron. The carbon content in briquettes and HBI was also taken into account. The share of briquettes and HBI,

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made out of those briquettes, in blast furnace burden was given on the basis of calculation of utilization of 600 thousand t of sludges and 640 thousand t of briquettes of the composition, given above. For prereduction of such quantity of briquettes there shall be two rotary hearth furnaces with the capacity of 320 thousand t per year each[6], and there shall be two cupola furnaces for melting those briquettes in OXYCUP process[9]. For prereduction of briquettes by Waelz process one needs three rotary kilns 4 m in diameter and 60 m in length[11].

**Table 3.** Blast furnace performance indicators (volume 870 $m^3$) upon the results of simulation

| Blast furnace conditions and performance values | Base case | Charge | |
|---|---|---|---|
| | | with briquettes | with HBI |
| Sinter rate, kg/t | 1482 | – | 322 |
| Pellets, kg/t | 147 | 889 | 767 |
| Briquettes, kg/t | – | 836 | – |
| DRI, kg/t | – | – | 445 |
| Fe content in the burden, % | 59,5 | 54,44 | 63,3 |
| Consumption rate: | | | |
| oxygen rate, $m^3$/t | 51,5 | – | 38,6 |
| coke rate, kg/t | 418 | 375 | 384 |
| natural gas rate, $m^3$/t | 100 | 50 | 90 |
| Blast temperature, °C | 1200 | 1000 | 1200 |
| $O_2$ content in the blast, % | 26 | 21 | 25 |
| Theoretical combustion temperature, ˚C | 2140 | 2056 | 2133 |
| Top pressure (absolute value), kPa | 270 | 190 | 260 |
| Top gas output, $m^3$/t | 1622 | 1890 | 1536 |
| Top gas heat value, MJ/$m^3$ | 3,96 | 3,36 | 4,4 |
| Slag output, kg/t | 300 | 309 | 284 |
| Slag basicity $CaO/SiO_2$ | 0,995 | 0,9 | 1,0 |
| Furnace output, t/day | 2396 | 2135 | 2733 |

The total consumption of carbon per 1 ton of iron in OXYCUP process, calculated according to the composition and output of the top gas, makes 564,5 kg/t taking into account the consumption of fuel for production of oxygen, the consumption of which in the process makes 174 $m^3$/t of pig iron [8].

**Table 4.** The consumption of carbon in the options, kg/t of pig iron

| Points of carbon consumption, kg/t of hot metal | Base case | Briquettes | DRI Fastmet | DRI (Waelz) | OXYCUP process |
|---|---|---|---|---|---|
| Sinter Production output | 65 | – | 14 | 14 | – |
| Pellet production | 2,5 | 15 | 13 | 13 | – |
| Prereduction * | – | – | 71 | 393 | – |
| In briquettes | – | 108 | – | – | 262,5 |
| Content in HBI | – | – | 4,5 | 4,5 | |
| Coke** | 315 | 277 | 285 | 285 | 263 |
| Natural gas | 56 | 28 | 50 | 50 | – |
| Oxygen content in the blast*** | 11,5 | – | 9 | 9 | 39 |
| Carbon consumed | 451,0 | 428,0 | 446,5 | 768,5 | 564,5 |

*Note: * Heating up to 1300 °C, direct iron reduction, overall heat losses. ** Less carbon, which was transferred into iron. *** Assuming 1787 kkal/$m^3$ $O_2$ or 0.224 kg C/$m^3$ $O_2$.*

Estimated per 1 t of pig iron $CO_2$ emission makes according to the options, kg/t:

- Basic option (regular blast furnace heat) -1654;
- Option 1 (blast furnace heat with briquettes) – 1569;
- Option 2 (prereduction in rotary hearth furnace + blast furnace heat) – 1637;

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- Option 3 (prereduction by Waelz process + blast furnace heat) – 2817;
- Option 4 (OXYCUP process) – 2070.

## 3 CONCLUSIONS

From the applied industrial technologies for recycling of Fe and Zn containing blast furnace and BOF sludges the minimal emission of $CO_2$ and consequentially the minimal consumption rate per 1 ton of pig iron, produced with the use of them, are provided by: the blast furnace process recycling of briquettes from these sludges and blast furnace processing of DRI which was produced by the prereduction of such briquettes in rotary hearth furnaces. If compared with respect to $CO_2$ emission and fuel consumption, OXYCUP process and blast furnace melting of DRI, produced by Waelz process prereduction, are inferior to those technologies.

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