Title: PRIMUS®, a new process for the recycling of steelmaking by-products and the prereduction of iron ore

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Abstract:

For years, iron and steel industry has been in search for low cost alternative to the conventional blast furnace production of iron as well as new routes for recycling of by-product which are generated during production and treatment of iron and steel. Considering these objectives, PAUL WURTH S.A. has developed a process using the multiple-hearth furnace, and coal fines as the reductant and main energy source. A trial plant with a capacity of about 2 t/h was built and has been operated for about one year at the ProfilArbed Belval site. This paper reports on the trial campaigns carried out for recycling by-products such as steelmaking sludges, dusts and scale, as well as for reducing iron ore fines, both of which have given most promising results.

Keywords:
Coal based direct reduction, Waste recycling, DRI, Zn and Pb removal, environment
1. Introduction

Two strong incentives are driving most of the new developments in iron and steelmaking:
- the search for alternative routes to the conventional blast furnace for producing low cost virgin iron, at the mini-mill scale
- the need for a cost-effective treatment for steelmaking by-products

The by-products considered are mainly:
- EAF dust (with medium to high contents of Zn, Pb, Cl)
- BF and BOF gas cleaning sludges (low to medium Zn, Pb, medium to high Cl, K, Na)
- Rolling mill scale and sludges (mainly iron oxides, with variable oil content)

Fig. 1 depicts the requirement of virgin iron in a mini-mill producing 1 million tons of raw steel per year and requiring a proportion of 25 % low residual charge to ensure the targeted steel quality. The substitution of imported low residual scrap or pig iron, which is becoming scarce and expensive, by direct reduced iron requires about 300 000 t of DRI per year.

For typical Western European conditions, prerequisites for low cost virgin iron are
- use of a low cost iron source, i.e. preferably ore fines;
- low cost reductant and energy source, preferably supplied by coal fines;
- minimized preparation, i.e. direct use of ore fines and coal fines in the reduction reactor
- low investment cost.

Fig. 2 shows a flowsheet for the treatment of steelmaking by-products, with complete recycling of all metallics and minimized transport of waste or low value materials.
Figure 2: Treatment of steelmaking by-products

In order to have a cost-effective treatment of steelmaking by-products, important features are once again minimized preparation of the by-product, the use of a low cost reductant and energy source and low investment costs; but here the economics will also be strongly influenced by the efficiency of zinc and lead separation and by the recovery of an iron concentrate in a form best suited to internal recycling.

To meet the above mentioned objectives, PW developed the PRIMUS process using the multiple-hearth furnace, and coal fines as the reductant and main energy source. Specific process and gas treatment features have been developed for treating the by-products, in order to maximize the recovery of the heavy metals and to fulfil the most severe environmental constraints.

This paper presents:
- a description of the process;
- an outline of the results obtained on laboratory and pilot installations for both DRI production from ore and treatment of steel making by-products;
- the prospects for industrial applications.
2. Process description

Furnace technology

As shown in fig. 3 the furnace is of a multi-hearth design, consisting of a cylindrical chamber on a vertical axis, with a number of superimposed or stacked annular compartments. The hearth floors/roofs between the different compartments are constructed of refractory bricks which form self-supporting levels. These levels are provided with openings, enabling the material to drop from one hearth level to the level located directly below. The material is charged at the upper level and transported by rabble arms which are driven by a rotating, air cooled axial column. This axial column supports several arms with angled scraper blades at each level. The material is alternately moved at the various levels from the furnace center to the walls and back to the center. On the lowest level of the furnace the material is then discharged.

The furnace is heated by means of burners installed in the furnace sidewall. The (reducing) gases cross the furnace upstream, but other gas evacuation systems are also conceivable.

Use of multiple-hearth furnaces

The multiple-hearth furnace is especially appropriate for the thermal treatment of fine-grained charging materials because of its operating mode. The charged materials are actively stirred, as they cross each hearth which enables a fast and homogenous heating
of the thick material layers. The temperatures inside the material layer and the furnace chamber are relatively uniform after passing a few levels with a temperature variation of less than 30K. The multiple-hearth furnace is especially suited to the drying and deoiling of sludges. Due to the continuous material motion, the drying process takes place efficiently, while avoiding the formation of agglomerates. Even difficult materials, such as oily mill sludge, can be dried and deoiled in the multiple-hearth furnace.

Suitability for metallurgical processes

The principal characteristic of the **PRIMUS** process consists in using the multiple-hearth furnace for reducing in addition to drying and calcination. The furnace specifically developed by Paul Wurth achieves operating temperatures up to 1100°C. Thus, the multiple-hearth furnace can also be used for metallurgical processes.

**PRIMUS** process

![Process metallurgy](image)

A solid reducing agent e. g. coal fines is charged with the metal oxide containing material (e.g. iron ore fines) into the furnace. The mixing of both materials is achieved in the furnace without any prior mixing being required. Inside the furnace, the charged materials are submitted to the different process steps, including drying (100°C), if necessary deoiling (400°C), and heating (1050°C). Thanks to intensive stirring and heating, the reduction of the metal oxides can take place inside the material layer in the final process step. The excess CO gas, generated during the reduction process escapes from the material layer and can be burned by air injection directly above the solid material. As shown in fig. 4, inside the furnace two zones develop with different reducing potentials: one zone with a high reducing potential inside the material layer and a second zone with oxidizing
conditions in the gas area. The energy generated during the postcombustion of CO, volatilized oil and volatile components of coal is sufficient to maintain the required process temperatures of 1000-1100° C. In a stationary state, the PRIMUS process does not require any additional energy supply. A complete postcombustion of the process gases inside the furnace is possible during the reduction of iron oxides (ore, scale). This high degree of postcombustion and the relatively low process temperatures (1100°C), make the PRIMUS process very energy efficient.

When the PRIMUS process is aimed at treating zinc oxide and lead containing by-products, such as sludge or dust from oxygen steelmaking or EAF dust, the contained heavy metal oxides are reduced in addition to the iron oxide in the material layer. Unlike Fe, Zn and Pb evaporate and leave the material layer as metallic fumes. The oxidizing gas atmosphere of the furnace then favours reoxidization. ZnO and PbO are entrained by the fumes in the form of dust particles and are separated by filtration in a bag filter. In addition to Zn and Pb, alkaline compounds are also evaporated and collected.

Thus the PRIMUS process enables the separation of the iron fraction from the heavy metals Zn and Pb and the alkali metals Na and K as well as Cl. The separation of the volatile elements from the charged material leads to a highly metallized iron concentrate which is valuable for recycling in the steel production process. At the same time a filtered dust is produced with a concentration of volatile elements higher than in the initial material. The zinc content in the recovered dust achieves levels of 50 – 60% which can then be sold as a value added product to the zinc industry.

Figure 5: Process flowsheet - general

Fig. 5 shows the basic flowsheet of the PRIMUS process adapted to the treatment of ore, mill scale, blast furnace sludge and scale. The multiple-hearth furnace is the core
equipment of the process. The furnace volume and the number of hearth levels is variable and adapted to the requirements of the material to be processed. In general all materials are charged on the top level. Coal or other additions can also be added on any other level, if it is required by the process. The final product (DRI or iron concentrate) is discharged, cooled and available for further process steps. It has a fine-grained consistency, which enables pneumatic conveying or magnetic separation.

The off-gases flow in the reverse direction of the solids and are evacuated at the top level. Dust particles conveyed by the gases are separated in a cyclone and returned to the furnace. The subsequent gas cleaning has to be adapted to suit the process and environmental requirements. For example, this flowsheet shows postcombustion, heat recovery, cooling and filtration.

Figure 6: Process flowsheet for EAF dust

Fig. 6 shows a flowsheet representing the further development of the PRIMUS process, specifically adapted to the treatment of EAF-dust. The main objective is to increase the purity of the separated zinc oxide to a value of > 90%. This can be achieved by separating the evaporation process of lead, alkalis and chlorine from the zinc evaporation.

The dusts are heated under oxidizing conditions to 950 - 1050° C prior to adding the reducing agent. In this phase, the above mentioned elements evaporate and are extracted selectively and separated in a bag filter. Once this process step is finished, the reducing agents are added and the reduction/evaporation of Zn occurs in the lower section of the furnace. The zinc-oxide containing off-gas of the reducing phase is collected and removed in a second off-gas line and a ZnO-concentrate is obtained.
3. Laboratory and pilot trials

To develop the **PRIMUS** process PAUL WURTH S.A. has built two test units:
- a laboratory reactor enabling extensive metallurgical testing;
- a pilot plant including all components required for an industrial installation.

**Laboratory unit**

![Laboratory furnace](image)

Figure 7: Laboratory furnace

This installation is shown on fig. 7. The furnace is a one hearth design for batch operation. The inner diameter is about 0.5 m. The shaft is equipped with two arms, one with forward rabbles and one with backward rabbles. The material (10 - 15 kg) is charged as one batch or in several steps for each trial. All parameters required for evaluation are recorded and periodic samples are taken to analyze the process over the time of the test. Each test thus results in curves showing the behaviour of the product in relation to the process time.

Fig. 8 shows a typical development of the degree of metallization for different materials. Similar curves are derived for different process parameters in order to evaluate the influence of the parameters such as temperature, granulometry, quality of reductant a.s.o. Fig. 9 depicts a typical diagram for the treatment of EAF dust. In this particular case, the material has first been heated and calcified. The coal for the reduction has been added after calcination. This type of diagram is used to determine the required time and parameters necessary for a separate extraction of the chlorine, the alkalis and the lead in the calcining stage, and of the zinc in the reducing stage. Systematic simulations may further reveal other relations; for example, the relationship between the degree of dezincification and the metallization.
Figure 8: Metallization

Figure 9: Zinc removal
Pilot installation

In co-operation with ProfilARBED, Paul Wurth has built a pilot plant in the works of Esch-Belval in Luxembourg. The plant is designed for a throughput of 1-2 t/h. The layout of the plant is shown in Fig. 10. The core equipment is a multiple-hearth furnace with an inner diameter of about 2 m and 11 hearths. The rotating centre shaft and the four arms per hearth are air cooled. The feed material is transported to the top of the furnace inside large bags and individual materials are stored in separate hoppers. Each material from these hoppers is accurately metered and directly fed into the furnace. With the material handling equipment charging is possible through the roof onto the top hearth as well as directly into the lower hearths. This possibility is mainly used to add the coal at the appropriate location of the process. Each hearth level is equipped with ports to allow material feed, sampling, measuring, inspection etc.. In addition each level also has burners and postcombustion air inlets. The burners are only used for furnace preheating and calcining, when high zinc containing material is treated.

The iron concentrate is discharged from the bottom hearth directly into a water cooled screw conveyor. The product falls from the screw into a weighing hopper. Measuring of all incoming and outgoing solids and gases gives continuous material and energy balances. As explained above, the furnace would normally be operated with one off-gas outlet and one waste gas treatment. Fig. 11 shows the plant as it has been extended with a second waste gas treatment for the selective extraction of zinc. The off-gas from the upper calcifying zone of the furnace first passes through a cyclone. The collected fines are continuously recycled into the furnace. With EAF dust treatment, the top part of the
furnace is operated at a temperature and an oxygen level high enough to avoid a separate postcombustion chamber. Thus the off-gases can immediately be quenched to between 150 and 180° C after the cyclone, before being dedusted in a baghouse. The off-gas from the bottom reducing zone also passes through a cyclone with continuous dust recycling. The off-gas is then measured and analysed to allow optimum adjustment of the subsequent postcombustion chamber. Part of the waste heat is reused in a recuperator to preheat the combustion air of the multiple-hearth furnace. The cooling is again achieved with water spray quenching, and the dedusting in a baghouse. Fig. 12 is a photo of the pilot plant.

Trials have been run on this pilot plant since April 1999. The first campaign has been performed with iron ore, mill scale and oily mill sludge. These trials performed with a single off-gas line have successfully demonstrated the feasibility of the process on a continuous basis. Results and conclusions are available on the productivity, the energy consumption and on all other figures required to evaluate the process for different iron oxide bearing materials. A short trial period with EAF dust with one off-gas line has proven that a typical filter dust containing zinc and lead oxides together with chlorine and alkalis can be collected. This first trial campaign between April and November 1999, has resulted in more than 145 tests with a total duration in excess of 900 hours.

Paul Wurth has since equipped the pilot plant with a second off-gas treatment line in order to collect zinc oxide with a high zinc concentration. Since February 2000 trials have been achieved with selective extraction of the off-gas from the top calcining zone containing mainly the lead, chlorine and alkalis, and the gas from the bottom reducing zone containing the zinc.
4. Results and outlook

The PRIMUS technology developed by PAUL WURTH S.A. at a pilot scale of 1~2 t / h fully meets the initial objectives:

- Excellent quality DRI is produced using ore fines and low cost coal fines as the single energy source; no preparation of the raw materials is required, and metallization levels exceeding 95 % can be easily achieved. The carbon content can be adjusted to the requirements of further processing steps.

- By-products including EAF dust, BF sludge, and rolling mill scale and sludge have been successfully treated, resulting in a highly metallized (>90%) iron concentrate which is easily recoverable in the melting unit (EAF or BF).

More than 95 % of Zinc and lead are removed. With high zinc contents a specific process configuration enables the possibility to selectively extract the zinc in the form of highly concentrated zinc oxide.

The trial program is currently continuing, with extensive tests on EAF dusts and mixtures of different types of by products.

The observed consumption and productivity figures of the pilot plant shows that the new technology is economically attractive for both steelmakers and by products recyclers.

Figure 13: Pilot installation