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Process Technology of the Direct Separation and Recovery of Iron and Zinc Metals Contained in High Temperature EAF Exhaust Gas

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The innovatory process, that is the direct separation and recovery of the iron and zinc metals contained in the high temperature exhaust gas generated from the electric arc furnace for the iron scrap melting and/or the dust treatment, has been proposed. This proposed process consists of the moving coke bed filter that is directly connected to the electric furnace, and the following heavy metal condenser. The exhaust gas passes through the filter and the condenser right after exhausting from the electric furnace. The moving coke bed filter is being controlled at about 1000℃ and collects iron and slag components contained in the high temperature exhaust gas. Heavy metals such as zinc and lead pass through the filter as vapor. Based on the thermodynamic considerations, the iron oxide and the zinc oxide are reduced in the filter. The solution loss reaction rate is comparatively low at about 1000 °C in the coke bed filter by the analysis using the mathematical simulation model. The heavy metal condenser is installed in the position after the coke bed filter, and rapidly cools the gas from about 1000℃ to 450℃ by a fall of the cooling medium like the solid ceramic ball in addition to the cooling from the wall. The zinc and lead vapor condense and separate from the gas in a liquid state.

The investigation of the characteristics of the exhaust gas of the commercial electric arc furnace, the fundamental experiments of the laboratory scale and the bench scale ensured the formation of this proposed process. A small-scale pilot plant examination is carrying out at present to confirm the formation of the process. It is certain that the dust generation of the electric arc furnace is extremely decreased, and it can save the energy consumption of usual dust treatment processes by the realization of this process.

Keywords: electric arc furnace, dust, zinc, coke bed filter, heavy metal condenser

Introduction

The development of new technology for electric arc furnace dust treatment is one of the most demanded subjects for the construction of cleaner production system. In Japan, about 0.5 million tons of dust are annually generated from the electric arc furnaces for steel making. A large quantity of the metals such as zinc and iron is contained in the dust, so the dust is collected, and refined by several processes in the world. These recovery processes are usually of complexity, and reduction requires large amounts of energy. Because a treatment cost is high, it is demanded for the development of the low cost treatment process of the dust. Researches on processes for the recovery of zinc in dust are currently underway throughout the world. However all have yet to be perfected.

Since 1994 at the Japan Research and Development Center for Metals (JRCM), new technology has been developing for collecting iron and zinc components in high temperature exhaust gas generated from electric arc furnaces of the steel making process. The process is that the dust generation from electric arc furnace process is almost zero. The process of this development, which consists of a moving coke bed filter and a heavy metal condenser, is a direct separation and collection process of iron and zinc contained in high temperature exhaust gas from the electric arc furnace. The project of this development has started in 1998 as a government-subsidized project for 5 years with the plan. The Ministry of Economy, Trade and Industry of Japan, and the New Energy and Industrial Technology Development Organization (NEDO) have financially supported this development. JRCM was entrusted with the development of the process from NEDO.

This paper introduces the process, and reports on investigations of actual furnace off gas, and the results of the bench scale experiments.

Conception of the Process

The amount of exhaust gas from the electric arc furnace is estimated 75 - 100Nm^3/t, temperature about 900 °C, dust concentration 100 - 130g/Nm^3 from the analysis of the present electric arc furnace operation for steel making. As for the composition of typical dust collected from exhaust gas, iron is about 30%, zinc about 20%, lead about 4%, slag about 16%, respectively. That is, the exhaust gas contains the high vapor pressure component represented by zinc and the low vapor pressure component represented by iron. If the temperature and the oxygen potential of exhaust gas are controlled properly, it is thought to be possible that these components are separately collected.

Figure 1 shows the schematic drawing of the direct separation and recovery process of iron and zinc from EAF exhaust gas, which is proposed by JRCM. The exhaust gas from electric arc furnace, which is strengthened a seal, passes through a moving coke bed filter, which is controlled at about 1000℃. Dust and fume in exhaust gas are collected in the filter. Gas passing through the filter contains zinc and lead vapors. Next, exhaust gas passing through the filter passes through a heavy metal condenser, which rapidly cools the gas from 1000℃ to 450℃. Ceramics balls are made to fall as cooling and condensation medium from the top of the condenser. Zinc and lead vapors are condensed and separated in a condenser.

Flow and functions of this process is shown in Figure 2.

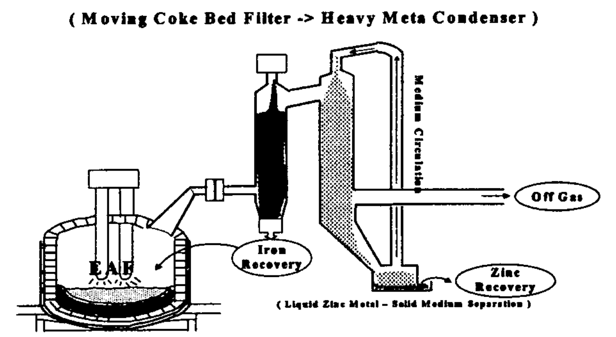


Figure 1 Schematic drawing of the proposed process

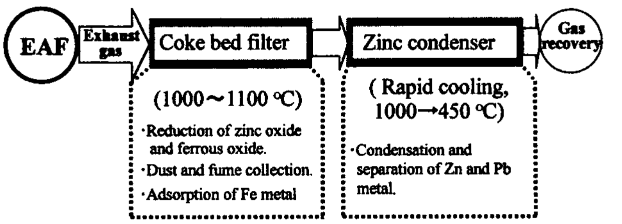


Figure 2 Process flow and function of the proposed process

As for the technical key points of the process, a reducing atmosphere is maintained by the suitable sealing up of the electric furnace, a moving coke bed filter is kept at about 1000℃ and adequate rapid cooling of gas is secured in a heavy metal condenser with a ceramics ball falling as a cooling and condensation medium in addition to cooling from the wall.

Theoretical Consideration of the Process

Results of the theoretical analysis about the exhaust gas condition to realize the above process are described in the following.

EAF exhaust gas and coke bed filter conditions

Figure 3 shows thermodynamic analysis of an electric furnace exhaust gas conditions. The figure shows the oxygen potential of the reduction of ZnO as a function of zinc vapor pressure and FeO. In the figure, CO-O_2-CO_2 equilibrium is also shown as a function of CO/CO_2 ratio. The oxygen potential under the coke existence is also shown in the figure as a function of (P_{CO}+P_{CO_2}) partial pressure. Zinc vapor pressure in the actual electric furnace exhaust gas is estimated at about 2%. The appropriate temperature value of electric furnace exhaust gas is 1000-1100℃, based on this analytical result. The appropriate gas composition is estimated at $CO/CO_2>2$. In Figure 3, the top gas operating condition of the ISP (Imperial Smelting Process) that contains zinc vapor was also shown.

The lower limit temperature of the coke bed filter depends on the reduction condition of iron oxide and zinc oxide by carbon. When zinc exists as zinc oxide (ZnO) in the electric arc furnace exhaust gas, it is necessary to reduce and to produce zinc vapor in the coke bed filter. At 900-1000°C, the reduction of ZnO depends on the vapor pressure of zinc and the partial pressure of (P_{CO}+P_{CO_2}). Furthermore, it is better to exceed the boiling point (907°C) of zinc metal for the passage through the coke bed filter. So the lower limit of the temperature of the coke bed filter is about 1000℃. The upper limit temperature of the coke bed filter depends on the melt formation. It is important to prevent the filter function obstructed due to melting of the collected iron and slag, and also not to enlarge the thermal load of the following zinc condenser. Iron component in the coke bed filter is reduced and becomes metallic iron at 1000°C. So the Fe-C alloy is formed by the reaction with the coke. Therefore it is necessary to maintain the coke bed filter under the temperature of 1153℃ which is the eutectic temperature of Fe-C alloy. So the upper limit of temperature of the coke bed filter is about 1100°C. From above reasons, it is desirable to control the temperature of the coke bed filter in the range of 1000 - 1100℃.

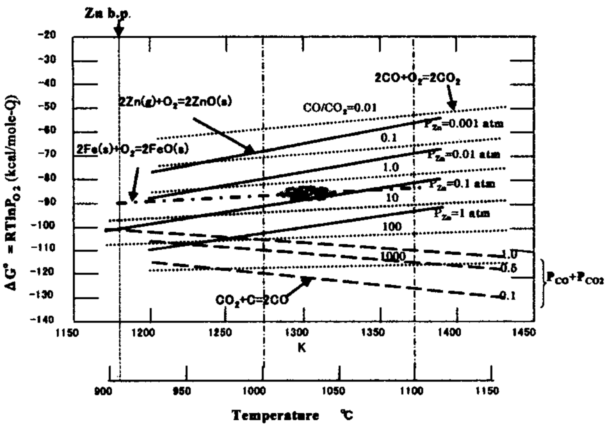
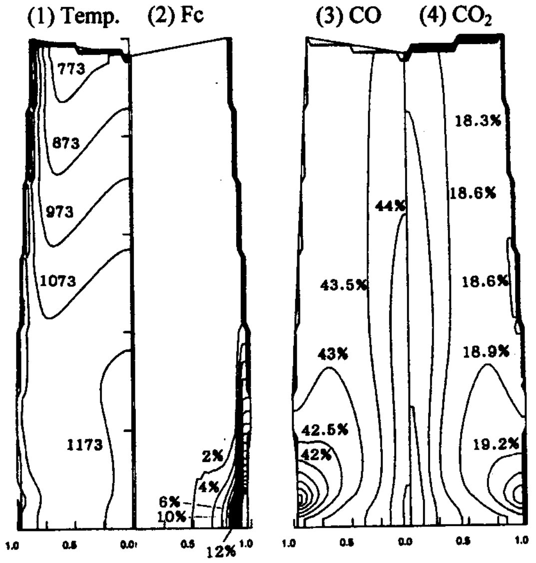


Figure 3 Thermodynamic analysis of the oxygen potential of the reduction of ZnO, FeO, CO-O_2-CO_2 and CO-C-CO_2 equilibrium

A mathematical simulation model of the coke bed filter has been constructed in order to estimate the behavior of coke gasification. Figure 4 shows an example of the simulation results concerning the coke bed filter that is supposed to install to the 50t/h EAF commercial plant. The inside diameter of this coke bed filter is 1.8m and the height is 7.0m. Coke is charged into the upper part of the coke bed filter at the feeding rate of 550kg/h and temperature of 250℃, and is discharged continuously from the bottom. High temperature gas (1000 ℃, 600Nm^3/h, CO:40%, CO_2:20%, H_2:5%, H_2O:5%, N_2:30%) is introduced into the lower part of the coke bed filter and flows up in the coke bed. The coke bed is heated from the wall and the wall surface is maintained at 1000°C. From these simulation results, H_2O almost changes into H_2. However CO_2 concentration of 20% in the inlet gas decreases to about 18% at the top of the coke bed filter. It was clarified CO_2 gas to slightly react in the coke bed filter because CO_2 solution loss reaction rate is comparatively low at this temperature.



(1) Temperature of gas (K)
(2) Coke gasification ratio, Fc (%)
(3) CO concentration (%)
(4) CO2 concentration (%)

Figure 4 Simulation results of the distributions of gas temperature, coke gasification ratio, CO and CO_2

Heavy Metal Condenser

Equilibrium vapor pressure of zinc, lead and the iron, and the analytical results of the partial vapor pressure, temperature and condensation ratio are shown in Figure 5. When zinc vapor pressure in exhaust gas from the coke bed filter is about 2%, the theoretical condensation ratio of zinc is estimated at about 96% by cooling to 450℃. In the figure, the top gas condition of lead splash condenser of the ISP was also shown. It is desirable to collect zinc under the liquid state from the problem of the handling of the condensed zinc and the heat load of condenser as a heat exchanger. Gas retention time is 1-2 second with another type condenser process. So it is objective that the cooling rate is about 600-300℃/s.

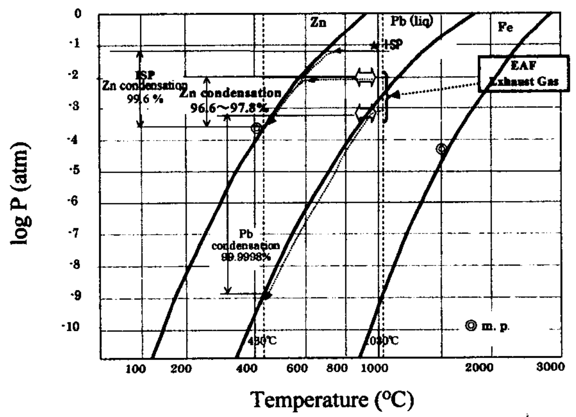


Figure 5 Relation between temperature, equilibrium vapor pressure and condensation

Figure 6 shows an example of the heat exchange simulation result when alumina ball of a diameter 0.5mm is made to fall. It is understood that the function of the condenser can be secured when the wall cooling condition of the condenser and the falling condition of alumina ball are set up properly.

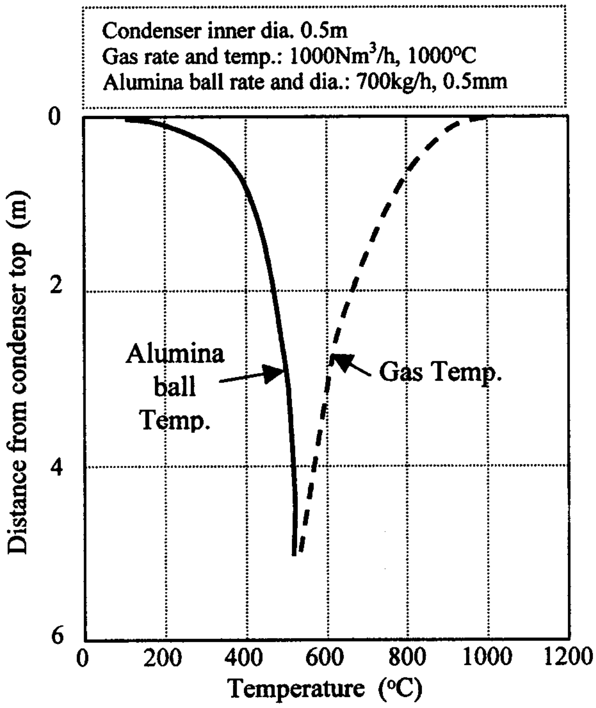


Figure 6 A heat exchange simulation result of the heavy metal condenser

Experimental Results and Discussion

In order to confirm the realization of the process, actual electric arc furnace investigations, laboratory scale experiments and bench scale experiments were done.

The results are followings.

Analyses of actual EAF exhaust gas

150-ton electric furnace of Aichi Steel Corp. was used. The volume and composition of its exhaust gas were analyzed. As shown in Table 1, it was estimated that about $88Nm^3/t$ gas was generated in the furnace and dust concentration was $147g/Nm^3$.

Table 1 Volume and dust concentration of gas generated from the 150-t electric arc furnace

Gas volume	88 Nm^3/t
Dust concentration	147 g/Nm^3

The dust samples collected by rapid cooling were put to X-ray diffraction analysis, and substances contained in them were quantified from the diffraction peaks. Analysis showed that dust from inside the furnace contained metallic alpha-iron, zinc, and lead. The substances contained in the dust are shown in Figure 7. Particle size ranges of the dust were from several hundred nanometers to about 5 microns. Zinc particles are extremely small, approximately 500 nanometer. As for the gas composition inside the furnace, $CO/CO_2>2$ could be attained at the melt down and refining period.

From these analyses, it was confirmed that strengthening seal of the furnace could attain the exhaust gas condition that satisfied the performance of the coke bed filter and the heavy metal condenser.

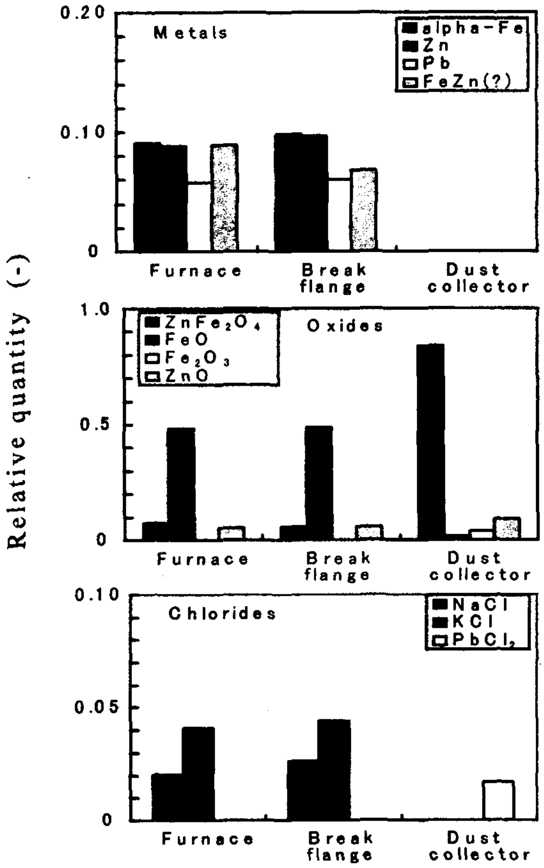


Figure 7 Analytical results of the dust composition

Bench-scale hot test results of the coke bed filter

The basic functions of the coke bed filter of this proposal are as follows: reduction of zinc oxide and passage of zinc vapor, collection of iron and slag components, and decomposition of dioxin. In order to investigate the basic performance of the coke bed filter at a high temperature, a dust collecting experiment with the bench scale system was carried out. The schematic flow of the bench scale experimental equipment is shown in Figure 8. Dust is mixed with the CO/CO_2 mixed gas which has been heated at a high temperature and is fed into the coke bed filter (Diameter: 80mm, Height: 300 and 600mm). The preheated coke is supplied and the dust is collected in the coke bed filter. The dust-collected coke is discharged continuously into the coke cooling device. Iron making coke with a particle size of 10mm or less was used, and EAF dust was used for the experiments. The standard experimental conditions are as follows: the dust concentration; 200g/Nm^3, the gas composition; $CO/CO_2=$ 80%/20%, the temperature in the packed bed; 1000°C, and the superficial velocity of gas; 1m/s.

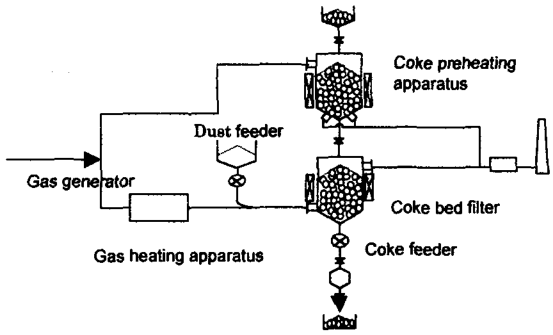


Figure 8 Schematic flow of the bench scale coke bed filter equipment

An example result of the dust collection efficiency of the coke bed filter is shown in Figure 9. A collection efficiency of approximately 94 – 98% can be obtained for the iron and slag components. Some reduction of iron and zinc oxides in the coke bed filter were also confirmed.

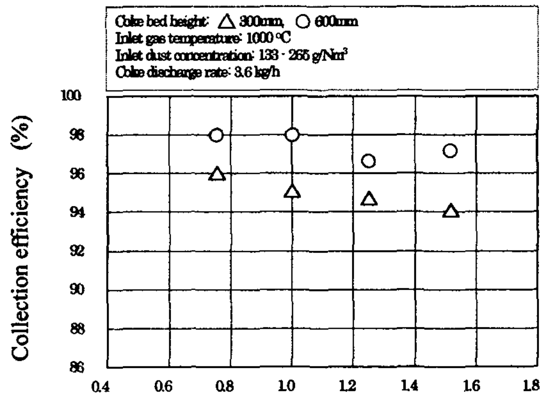


Figure 9 An example result of the dust collection efficiency of coke bed filter

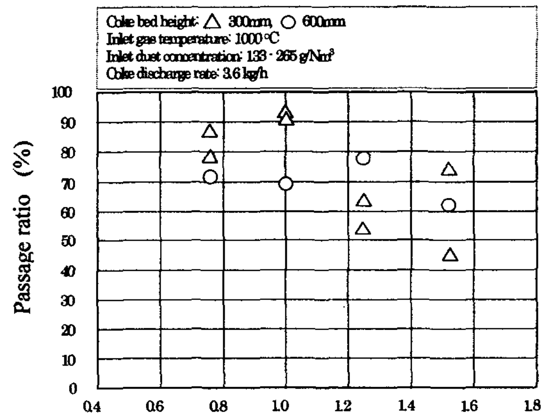


Figure 10 An example result of the zinc passage ratio of coke bed filter

The result of the zinc passage rate is shown in Figure 10. 70% and more zinc passage ratio was obtained in less than the gas superficial velocity 1.0m/s. The reason for a low passage ratio of zinc in this experiment is considered as follows: the zinc component in the feeding dust is mainly zinc oxide, the temperature of feeding dust into the coke bed is low (about 600°C), and the collected dust in the coke bed is discharged from coke bed filter in a short retention time (about 5 minutes). It was short for the ZnO in dust to be heated up and reduced. In an actual process, the zinc vapor pressure will increase because of sealing up electric arc furnace and the high temperature exhausted gas. Therefore it is considered that the passage ratio of zinc through the coke bed filter increases.

Bench-scale hot test results of the heavy metal condenser

The basic functions of the heavy metal condenser of this proposal are the rapid cooling of the passage gas, and condensation and separation of zinc and lead metals. A ceramics ball as a cooling and condensation medium is made to fall from the top of the condenser. The heavy metals are condensed on the medium and wall surfaces. After the condensed heavy metal was separated, the medium is circulated. The schematic flow of the bench scale experimental equipment is shown in Figure 11. The dimension of the condenser body is 90mm inner diameter and 2000mm effective height.

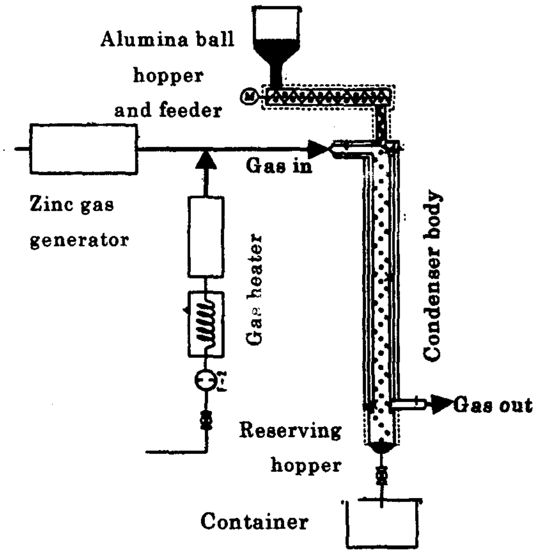


Figure 11 Schematic flow of the bench scale heavy metal condenser equipment

Photo 1 shows the alumina ball and the metal zinc, which obtained the bench scale experiment. As shown in the photograph, when the 0.5mm diameter alumina ball of the room temperature was made to fall in the nitrogen gas flow at about 800°C which contains zinc vapor, metal zinc condensed on the alumina ball surface. From the above results, it became clear to be possible to secure the expected function of the heavy metal condenser with the falling ceramic ball of this proposal.

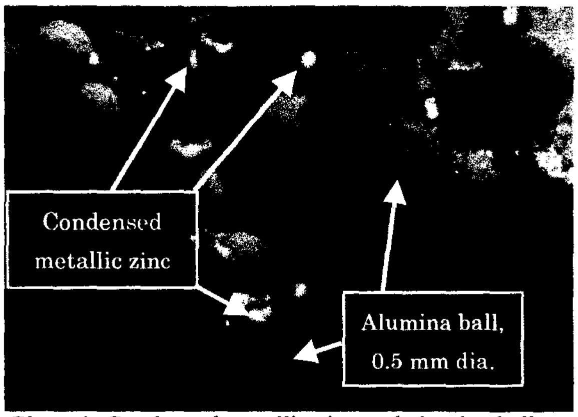


Photo 1 Condensed metallic zinc and alumina ball obtained by bench scale experiments

Small-scale pilot plant test

Based on the above research results, 1 t/h scale pilot plant was designed and constructed. At present, test operation is carrying out. Dust collection efficiency of the coke bed filter has been confirmed more than 97% in this small-scale pilot plant test.

Conclusion

A new treatment technology for high-temperature exhaust gas from electric furnaces was introduced. With respect to the proposed new process, that is directly collecting iron and zinc contained in the high temperature exhaust gas generated from an electric arc furnace for steel making, the theoretical considerations, the investigation of the exhaust gas from an actual furnace, laboratory experiments and bench scale tests were carried out.

By the theoretical and the experimental analysis, it was confirmed that the proposed process, that is the direct separation and recovery of the iron and zinc metals contained in the high temperature exhaust gas generated from the electric furnace for the iron scrap melting and/or the dust treatment, could be realized.

In order to verify this new process, experimental operation is carrying out at an integrated pilot plant combining sealing up electric arc furnace, coke bed filter and zinc condenser.

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