

The Direct Recycling of Electric Arc Furnace Stainless Steelmaking Dust

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This paper focuses on the pilot-scale investigation of direct recycling of electric arc furnace (EAF) stainless steelmaking dust. The direct recycling of EAF dust is to make pellets with the mixture of the dust and the reducing agent carbon, then introduce the pellets to the EAF. The valuable metals in the dust are reduced and get into the steel as the alloying elements. Experiments simulating direct recycling in an EAF were performed using an induction furnace. But it seems difficult to reduce all metal oxides in the dust so that some metal reducing agents are added in the late stage of reduction process. The valuable metals in the dust were reduced partly by carbon and partly by metal reducing agent for the economical concern. The recovery of iron, chromium and nickel from the flue dust and the amount of metal oxides in the slag were measured. The results showed that the direct recycling of EAF stainless steelmaking dust is practicable. It was also found that direct recycling of flue EAF stainless steelmaking dusts does not affect the chemistry and quality of stainless steel produced in the EAF. It is benefit not only for the environmental protection but also for the recovery of valuable metal resources in this way.

Key Words: EAF dust, waste recycling, induction furnace, direct reduction

Introduction

It was known that 1-2 weight percent of the scrap charge to an Electric Arc Furnace (EAF) is converted to dust in the steelmaking. And further more dust is generated in the Argon Oxygen Decarburization (AOD) or in the Vacuum Oxygen Decarburization (VOD) vessel in the electric stainless steelmaking. When stockpiled or landfilled, the flue dust is an environmental concern as it can leach heavy metals to the groundwater. Electric steelmaking flue dust has been classified a hazardous waste due to higher than acceptable heavy metal leachabilities such as zinc, lead, cadmium and chromium so that it has been banned from landfills in many countries. In addition to being an environmental hazard, the flue dust is also an economical concern to stainless steelmakers as it contains large amounts of valuable alloying elements, especially for iron, chromium and nickel.

The direct recycling of EAF stainless steelmaking dust is a remediation option which aims not only to protect the environmental but also to recover the metallic elements present in flue dusts directly to the steel bath of EAF in the steelmaking process. The dusts are mixed with carbon as reducing agent and dolomite or lignosulphonate as the binder and then formed into pellets that are subsequently fed to the EAF. Under the conditions that prevail in the EAF, the reducing agent is expected to reduce the metal oxides in the dust. In other words, the pellet is expected to self-reduced. The operation above is inexpensive to apply. The costs involved are the purchase of the reducing agent, the binder and the pelletization. Carbon, dolomite and lignosulphonate are cheap and easily available.

A number of research steps were completed prior to this study. The characteristics of the flue dust generated from EAF stainless steelmaking were checked^[1]. The laboratory-scale experiments for EAF dust-carbon pellets established the isothermal^[2] and non-isothermal kinetics model of the reduction using a Thermo-Gravimetric Furnace (TGF)^[3]. The results showed that reduction with carbon was not sufficient to recover chromium, which made to add some metal reducing agents as that of US Bureau of Mines^{[4][5]} and J&L Specialty Steels and Dereco^[6]. On the other hand, the pre-reduction for the pellets hinders metals recovery from the dust^{[7][8]}.

In this study, the possibility of using carbon and pure aluminum or calcium-silicon as a reducing agent was investigated. This would be desirable in practice since carbon is the least expensive of reducing agents. In order to increase the recovery of metals from the dust, the introduction of a metal reducing agent pure aluminum or calcium-silicon in the direct recycling process was investigated.

Experimental

Dust Characteristics

The EAF dust used in this study was from a stockpile located in the open atmosphere of stainless steelmaking site. As exposed to the air, the dust contains a large amount of moisture, which caused the particles in the dust samples to agglomerate over time. Scanning Electron Microscope (SEM) showed that the agglomerations represent both homogeneously nucleated particles and entrained fugitive particles that have experienced heterogeneous condensation. The

particles seem to have a fibrous appearance at the surface. This is caused by the adhesion of fine dust and fume particles that have agglomerated onto the surface of the larger particles thus providing a fibrous appearance. As a result, the range of particle sizes is very wide. The diameter of the particles ranged from less than 38 μm to more than 800 μm .

Chemical analysis on each EAF dust samples for the experiments performed through X-Ray Fluorescence (XRF) for Al, Ca, Cr, Fe, K, Mg, Mn, Na, P, Si, Ti and Inductively Coupled Plasma (ICP) for Ni, Pb and Zn. The compositions of the samples are given in Table .

Table 1 Composition of the flue dusts used in this study (%)

| Elements | Sample 1 | Sample 2 | Sample 3 |
|----------|----------|----------|----------|
| Al | 0.43 | 0.46 | 0.35 |
| Ca | 6.02 | 5.83 | 6.53 |
| Cr | 11.29 | 10.19 | 10.33 |
| Fe | 39.38 | 37.14 | 40.57 |
| K | 0.08 | 0.12 | 0.26 |
| Mg | 2.01 | 1.85 | 2.10 |
| Mn | 2.80 | 2.61 | 2.95 |
| Na | 0.24 | 0.20 | <0.07 |
| Ni | 2.82 | 3.52 | 5.42 |
| P | 0.02 | 0.02 | 0.02 |
| Pb | 0.07 | 0.13 | 0.16 |
| Si | 2.71 | 2.67 | 2.55 |
| Ti | 0.15 | 0.14 | 0.12 |
| Zn | 0.24 | 0.48 | 1.06 |

Most of the elements within the EAF dust are oxidized since it was formed in air at high temperature. Typical X-Ray Diffraction (XRD) pattern for the dust shows that the main phases detected in the dust were mainly metal oxides. The greatest peaks within the diffraction patterns contained iron and chromium in the oxide forms. Most iron was found to be magnetite, Fe_3O_4 , or hematite, Fe_2O_3 . Chromium was mostly present as CrO , however, it was also present as FeCr_2O_4 . Nickel was primarily grouped with manganese to form complex compounds such as NiMn_2O_4 and MnNi_2O_4 . In addition it was present in the simple oxide form as NiO . Copper is found in complex oxide forms along with iron and manganese principally as CuMn_2O_4 , CuFe_2O_4 and CuFeMnO_4 . Zn was primarily found coupled with chromium to form ZnCr_2O_4 .

Pellets Preparation

The EAF dust was screened using a 35 mesh screen for pelletization. 87% of the activated carbon particles were finer than 400 mesh and the dolomite (composition showed in Table 2), a spent refractory, was crushed in a ball miller for 20 minutes and then screened to below 35 mesh. The fines with 5% of dolomite and 15% of carbon were thoroughly mixed. A disc pelletizer was used to agglomerate the mixture. In order to increase the strength of the pellets, lignosulphonate (composition showed in Table 3) was introduced sometimes.

Table 2 Composition of dolomite used in this study (%)

| SiO_2 | TiO_2 | Al_2O_3 | Fe_2O_3 | MnO | MgO | CaO | Na_2O |
|----------------|----------------|-------------------------|-------------------------|----------|-------|-------|-----------------------|
| 4.27 | 0.32 | 2.64 | 6.99 | 0.52 | 36.57 | 33.46 | 0.26 |
| Ce (ppm) | Cu (ppm) | Ni (ppm) | V (ppm) | Zn (ppm) | | | |
| 1908 | 628 | 3631 | 244 | 190 | | | |

Table 3 Composition of lignosulphonate used in this study (%)

| Lignosol | Reducing Sugars | S | Ca | Na | N | pH | Ash | Moisture | Specific Gravity (lbs/cft) |
|----------|-----------------|-----|----|-----|-----|-----|-----|----------|----------------------------|
| 80 | 7 | 6.6 | 5 | 0.2 | 0.1 | 4.5 | 20 | 5 | 23 |

The pellets produced were screened to be larger than 7/16 inch in diameter. The pellets were dried in air at room temperature for 4 days. The cross-section of a typical pellet was checked by a Jeol-800C SEM. The different particles in the pellet cross-section were identified by energy-dispersive X-ray spectrometer. Measurements were conducted at 20 keV accelerating voltage and 100 nA beam current. Since the carbon particles are very fine, they act as a matrix for the dust and dolomite particles. Each dust particle seems to be surrounded by carbon so that the area of contact between dust and carbon is relatively high.

Induction Furnace Melting

The pilot-scale direct recycling trials were conducted in a 50 lb capacity induction furnace. Pure iron was melt first. Alumina crucibles were used to hold the liquid metal. The iron bath was intended to simulate the metal bath of the EAF. As soon as the iron had melted, pellets were added to the furnace in batches of approximately 1 lb each. After having fed all the pellets, a period of time was allowed for the reduction in the bath to reach completion. When visual observation indicated there is no more reactions taking place within the steel bath, aluminum or calcium-silicon was added to further reduce the metal oxides in the slag. Throughout an experiment, the slag and the metal were stirred using an iron rod. Temperature was measured periodically using an R-type thermocouple.

A heat lasted more than one hour. Then the furnace was turned off and the steel and the slag were allowed to freeze in the crucible. Once it had cooled down to room temperature, the crucible was broken to recover and weigh the ingot and the slag. The composition of the ingot and the slag was analyzed using Optic Emission Spectroscopy (OES) and X-Ray Fluorescence (XRF). The parameters for the experiments were controlled as Table 4 and the typical photos of crucible and ingot are showed in Figure 1.

Table 4 Parameters for the experiments

| No. | Starting Iron (lb) | Pellets (lb) | Lime (lb) | Reducing Agent Type | Amount (lb) |
|-----|--------------------|--------------|-----------|---------------------|-------------|
| A1 | 8 | 16.04 | 0 | | 0.00 |
| A2 | 5.25 | 10.30 | 0 | Al | 0.97 |
| A3 | 5 | 13.89 | 0 | Al | 0.50 |
| B1 | 8 | 2.50 | 0 | Al | 0.80 |
| B2 | 8 | 2.61 | 0 | Al | 0.13 |
| B3 | 10 | 2.61 | 0 | Al | 0.37 |
| B4 | 10 | 1.30 | 0 | | 0.00 |
| B5 | 10 | 1.00 | 0 | Al | 0.05 |
| B6 | 10.02 | 1.96 | 0 | Al | 0.20 |
| C1 | 20.04 | 4 | 0 | Al | 0.28 |
| C2 | 20.205 | 4 | 0 | CaSi | 0.38 |
| C3 | 19.795 | 4 | 1.15256 | CaSi | 0.38 |
| C4 | 29.9 | 1.92 | 0.55313 | CaSi | 0.18 |
| D1 | 20.1 | 4 | 1.15 | CaSi | 0.38 |
| D2 | 20.15 | 4 | 1.15067 | CaSi | 0.38 |
| D3 | 20.05 | 3.95 | 1.15 | CaSi | 0.38 |
| D4 | 20 | 3.35 | 1.15029 | CaSi | 0.38 |

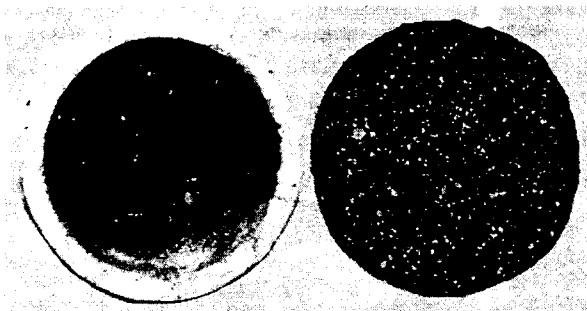


Fig. 1 Photos of Ingot and Crucible after Experiments

Results and Discussion

Recovery of Metals

The recovery of metals seems to be affected by the technological parameters such as the ratio of metal reducing agent to the pellets, pellets to start iron, lime to pellets and the basicity of the slag. The experimental results are listed in Table 5.

Table 5 Recovery of metals from dust

| Reducing No. | agent/Pellet | Basicity | Pellet/Start iron | Lime/Pellet | Recovery of Fe (%) | Recovery of Cr (%) | Recovery of Ni (%) |
|--------------|--------------|----------|-------------------|-------------|--------------------|--------------------|--------------------|
| A1 | 0.000 | 2.173 | 2.005 | 0 | 93.08 | 54.42 | 100 |
| A2 | 0.094 | 0.445 | 1.962 | 0 | 96.60 | 79.60 | 100 |
| A3 | 0.036 | 2.141 | 2.778 | 0 | 80.73 | 23.61 | 100 |
| B1 | 0.320 | 0.180 | 0.313 | 0 | 100 | 46.28 | 100 |
| B2 | 0.050 | No Slag | 0.326 | 0 | 100 | 100 | 100 |
| B3 | 0.142 | 0.665 | 0.261 | 0 | 94.67 | 80.55 | 100 |
| B4 | 0.000 | No Slag | 0.130 | 0 | 100 | 100 | 100 |
| B5 | 0.054 | 0.644 | 0.100 | 0 | 100 | 6.32 | 100 |
| B6 | 0.104 | 0.445 | 0.196 | 0 | 100 | 20.40 | 100 |
| C1 | 0.070 | 0.548 | 0.200 | 0 | 96.18 | 94.63 | 100 |
| C2 | 0.094 | 0.768 | 0.198 | 0 | 87.78 | 94.49 | 100 |
| C3 | 0.094 | 0.830 | 0.202 | 0.288 | 95.40 | 93.98 | 100 |
| C4 | 0.095 | 0.759 | 0.064 | 0.288 | 100 | 72.29 | 100 |
| D1 | 0.095 | 6.323 | 0.199 | 0.288 | 100 | 65.44 | 100 |
| D2 | 0.095 | 7.965 | 0.199 | 0.288 | 100 | 58.66 | 100 |

It can be seen from table 5 that pellets exhibit the high nickel and iron recoveries regularly. All the calculated nickel recoveries are 100%. But the chromium recovery is relatively lower, especially without aluminum or calcium-silicon added to reduce the chromium oxide from the slag. Even though the addition of aluminum or calcium-silicon in a ratio of 9.4% the weight of pellets could not reduce all chromium oxide in the slag, it could increase the chromium recovery by 22.8% up to 60.4%. Thus reduction by carbon only of the pellets can recover the nickel and most of the iron from the dust, but a further reduction of the slag with at least 9.4% aluminum or calcium-silicon in weight of pellets is needed to recover chromium. The relationship between the metal recovery and the ratio of metal reducing agent to pellets is showed in Figure 2.

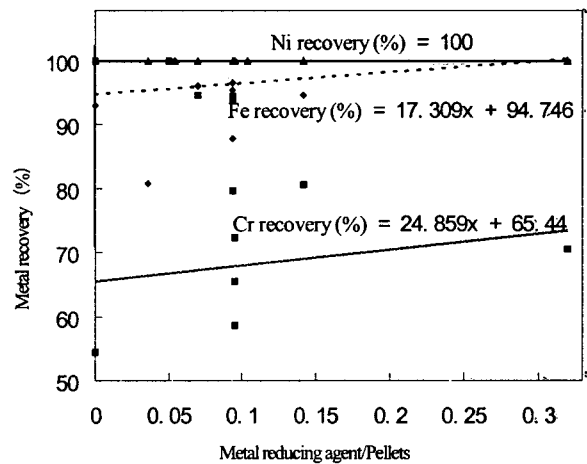


Fig 2 The relationship between metal reducing agent and pellets

On the other hand, the relationships between the metal recovery and other technological parameters are showed in Figure 3, 4 and 5. It can be seen from them that the metal recovery will increase with the increase of slag basicity and the ratio of lime to pellets and it will decrease with the increase of the ratio of pellets to start iron.

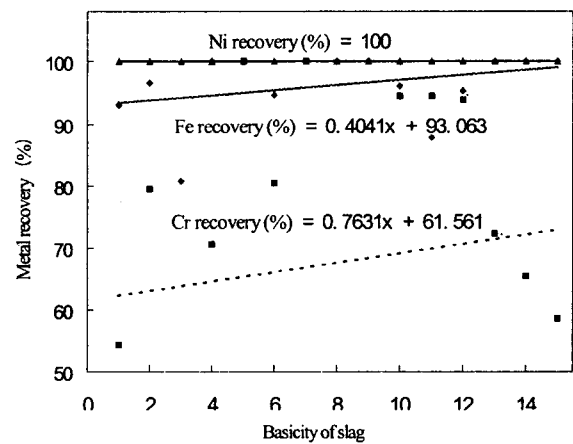


Fig. 3 Relationship between metal recoveries and basicity

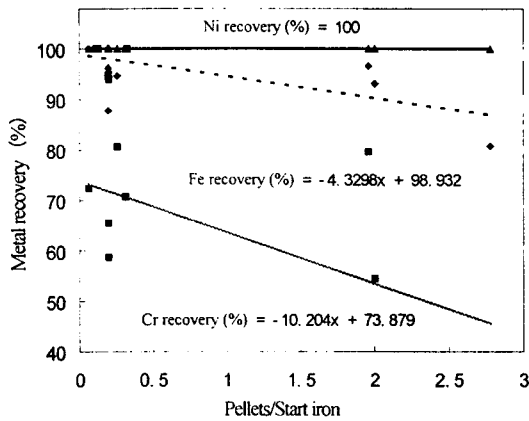


Fig.4 Relationship between metal recovery and ratio of pellets to start iron

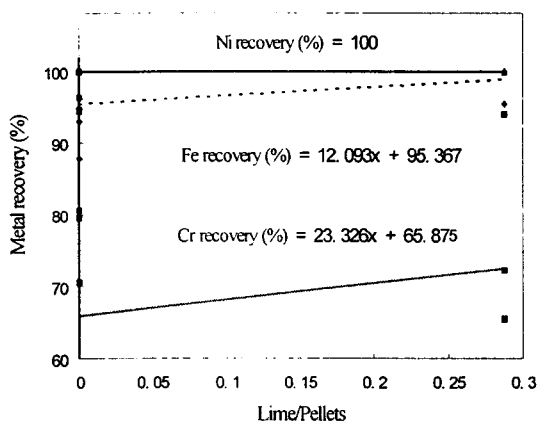


Fig. 5 Relationship between metal recovery and ratio of lime to pellets

Influence of Direct Recycling on Steel Quality

One of the concerns associated with the EAF dust direct recycling is that it may have an impact on the quality of the produced steel. As for this, the ingots obtained from the pilot-scale direct recycling experiments were analyzed for C, Cu, Co, N, P, S, Sn and Ti. The composition of the steel produced from the direct recycling of EAF dust is showed in the Table 6.

Table 6 Element increase in ingot (%)

| No. | ΔC | ΔCu | ΔCo | ΔN | ΔP | ΔS | ΔSn | ΔTi |
|-----|------------|-------------|-------------|------------|------------|------------|-------------|-------------|
| A1 | 3.5E-04 | 6.1E-05 | 1.9E-05 | 0 | 5.8E-06 | 2.8E-05 | 2.2E-06 | 3.2E-06 |
| A2 | 8.17E-05 | 6.2E-05 | 9.8E-06 | 0 | 5.9E-06 | 1.9E-05 | 0 | 3.37E-06 |
| A3 | | 4.8E-05 | 1.4E-05 | 0 | 3.9E-06 | 0 | 0 | 2.3E-06 |
| B1 | 5.5E-04 | 1.2E-04 | 2.1E-05 | 0 | 2.1E-05 | 5.9E-05 | 0 | 0 |
| B2 | 4.1E-05 | 1.2E-04 | 5.9E-05 | 0 | 1.8E-05 | 1.0E-04 | 0 | 0 |
| B3 | 4.7E-04 | 1.2E-04 | 2.5E-05 | 0 | 2.2E-05 | 5.2E-05 | 0 | 0 |
| B4 | 1.9E-04 | 1.9E-04 | 9.8E-05 | 0 | 3.9E-05 | 8.9E-05 | 0 | 0 |
| B5 | 1.8E-04 | 2.6E-04 | 2.6E-04 | 0 | 4.5E-05 | 1.2E-04 | 0 | 0 |
| B6 | 4.1E-04 | 1.6E-04 | 1.3E-04 | 0 | 2.3E-05 | 4.6E-05 | 0 | 0 |
| C1 | 2.0E-04 | 1.6E-04 | 1.3E-04 | 0 | 2.9E-05 | 6.1E-05 | 0 | 3.2E-05 |
| C2 | 3.3E-04 | 1.6E-04 | 1.3E-04 | 0 | 2.9E-05 | 6.8E-05 | 0 | 3.2E-05 |
| C3 | 5.7E-04 | 1.6E-04 | 1.3E-04 | 0 | 2.9E-05 | 3.2E-05 | 0 | 3.2E-05 |
| C4 | 4.5E-04 | 5.0E-04 | 4.0E-04 | 0 | 8.0E-05 | 1.2E-04 | 0 | 10E-05 |
| D1 | 2.4E-04 | 1.6E-04 | 1.3E-04 | 0 | 3.5E-05 | 3.5E-05 | 0 | 3.2E-05 |
| D2 | 1.8E-04 | 2.3E-04 | 3.2E-05 | 0 | 1.6E-05 | 6.5E-05 | 0 | 3.2E-05 |

Among these element increases in the steel, carbon can be easily removed in the VOD or AOD vessel to make very low carbon stainless steels. The main concern about the steel quality for the element is the increases of P and S because they are harmful to the mechanical properties of the steel. These increases are very low according to the experimental result. It seems that P came from the start iron and S came from the binder lignosulphonate. In the practical steel production, S can be removed by increasing reducing agent content and the basicity of the melting slag

Conclusion

The pilot-scale direct recycling of electric stainless steelmaking flue dust was investigated and the effect of the smelting was studied. It was found that over 90% iron and nearly 100% nickel recoveries can be achieved, but the recovery of chromium is low. A further reduction of chromium from the slag requires more aluminum or calcium-silicon and high temperature condition^[9]. It should be mentioned that chromium recoveries higher than 80% were achieved in the experiments. It was also found that direct recycling of EAF dust does not have a deleterious effect on the quality of steel produced in the EAF^[10].

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Appendix

The iron recovery in the direct recycling of EAF dust is calculated according to the following formula. It is assumed that all iron from the starter iron got into ingot in the process.

$$\% \text{Recovery}_{\text{Fe}} = \frac{\% \text{Fe}_{\text{ingot}} * W_{\text{ingot}} + W_{\text{starter}}}{\% \text{Fe}_{\text{pellets}} * W_{\text{pellets}}} * 100$$

Where $\% \text{Fe}_{\text{ingot}}$ is the concentration of Fe in the Ingot

W_{ingot} is the weight of the ingot (lb)

W_{starter} is the weight of pure iron used to start up the induction furnace experiments (lb)

$\% \text{Fe}_{\text{pellets}}$ is the concentration of Fe in the pellets

W_{pellets} is the weight of pellets (lb)

Chromium and nickel recovery in the direct recycling of EAF dust is calculated according to the following formula.

$$\% \text{Recovery}_{\text{M}} = \frac{\% \text{M}_{\text{ingot}} * W_{\text{ingot}}}{\% \text{M}_{\text{pellets}} * W_{\text{pellets}}} * 100$$

Where M is either chromium or nickel

$\% \text{M}_{\text{ingot}}$ is the concentration of M in the Ingot

$\% \text{M}_{\text{pellets}}$ is the concentration of M in the pellets