

RECYCLING OF STEELMAKING DUSTS – THE RADUST CONCEPT –

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Abstract

Recycling of dusts and other wastes of steelmaking is becoming to a necessity of two reasons: due to high contents of iron oxides dusts are valuable raw material for steelmaking and tightening environmental legislation makes the landfill disposal of wastes more expensive. Fine dust fractions from various stages of steelmaking route contain besides iron and carbon heavy metals, especially zinc and lead and heavy hydrocarbons that are acceptable neither for landfill disposal nor for recycling back to processes without any special treatments. Some theoretical and practical aspects concerning high temperature treatments of steelmaking dusts for removal of hazardous components and production of clean high iron raw material for recycling is discussed in this paper. The Radust technology developed at Koverhar steelwork in Finland for treatment of the most problematic fine fractions of blast furnace and oxygen converter dusts is shortly presented and discussed.

Keywords: recycling of steelmaking dusts, removal of harmful components, high iron slag production, the Radust technology

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1. Introduction

Steel production dusts and sludges from blast furnace (BF), oxygen converter (BOF) and electric arc furnaces (EAF) have high contents of iron oxides. Due to heavy metals, like zinc and lead as well as alkaline oxides, halides and heavy hydrocarbons, both the recycling and final deposition of fine fractions is problematic. The amount of coarse and fine (sludge, electrostatic precipitator dust) fractions of BF-dust are reported to vary from 6 to 17 and 3 to 5 kg / ton of hot metal in European blast furnaces and corresponding values for BOF dust range from 3 to 12 and from 9 to 15 kg / ton of steel [1]. Total amounts of dust formed in BF and BOF processes in the Finnish steel plants (Ovako Koverhar and Raabe Steelworks of Ruukki Group) are roughly on the same level [2,3,4,5].

Dusts from BF- and BOF-gases are separated in two stages. The coarse fractions are collected in cyclones and fine fractions in a wet scrubber or electrostatic precipitator. Highly volatile components like zinc, lead, chlorides, fluorides, high hydrocarbons and cyanides mostly pass the cyclone and separate in the scrubber or electrostatic precipitator. Due to low contents of harmful constituents the coarse fractions can be returned back to sintering plant without any special treatments or even disposed as such to landfills. The fine fractions (see table 1) high in iron oxides and carbon (BF-dust) contain high amounts of harmful constituents and are unacceptable for landfill and cannot either be recycled back to the processes (sintering plant, BF or BOF) without special treatments for separation or decomposition of those harmful constituents. The amounts of the minor constituents both neutral (slag formers, Ca-, Mg- and alkaline oxides as well as manganese oxide) and harmful components vary greatly depending on the composition of primary (iron concentrate) and secondary (scrap) raw materials.

Technically it is possible to extract the non-ferrous metals from dust and sludge and destruct the hazardous organic compounds through high- or low temperature chemical treatments and and recycle the valuable iron-containing "cleaned" solids into the steelmaking processes. The fraction with high contents of environmentally harmful constituents should be treated elsewhere in special plants. Economy of the whole chain of processes necessary for treating the steelmaking wastes is the key question, not discussed in this paper.

Conditions for landfill disposal tightening in the European Union lead to search of new methods for treating all metallurgical wastes but also affect the economical conditions of waste treatment technology.

Hydrometallurgical alternatives to treat the iron- and steelmaking wastes are not considered here.

Table 1. Average compositions of some BF- and BOF-dusts reported in different sources; wt-%

	Average in Radust campaign 22.3-0.4.2005		Reported in [6] (OXYFINES)		Collection from European steel plants [1]			
	BF-dust*	BOF-dust*	BF	BOF	BF dry	BF sludge	BOF dry	BOF sludge
Fe	28.6	47.9	26.9	64.5	15-40	7-35	30-85	54-70
SiO₂	5.2	1.4	7.5	1.1	4-8	3-9	-	-
MnO	4.3	0.35	0.20	1.4	0.1-0.5	< 0.2	-	-
P₂O₅	0.05	0.03	0.10	0.12	0.04-0.26	0.1-0.44	-	-
Al₂O₃	2.4	0.2	2.5	0.33	0.2-3.7	0.8-4.6	-	-
CaO	4.2	13.0	3.4	4.3	2-8	3.5-18	8-21	3-11
MgO	4.1	0.49	1.7	0.84	0.3-2	3.7-17	-	-
TiO₂	-	-	-	-	< 0.2	-	-	-
V₂O₅	-	0.16	0.01	0.02	-	-	-	-
K₂O	3 – 5	0.64	0.28	0.54	0.2-1.0	0.1-0.36	-	-
Na₂O	1 – 3	1.4	0.10	0.69	0.03-0.6	0.15-0.24	-	-
C	32.7	0.4	49.0	0.38	25-40	15-47	1.4	0.7
S	1.2	0.05	0.40	0.18	0.2-1.3	2.4-2.5	< 0.06	< 0.12
Zn	0.23	4.0	0.13	6.7	0.1-0.5	1-10	< 0.2	1.4-3.2
Pb	0.1	-	0.03	0.53	< 0.07	0.8-2.0	< 0.04	0.2-1.0

*electrostatic precipitator dusts

2. Recycling of iron and steelmaking dusts

The main tasks of special treatments of iron and steelmaking dusts before recycling or disposal for landfill are the separation of zinc and lead (and other heavy metals if present in remarkable amounts) from iron bearing residue and decomposition of heavy hydrocarbons. The are two main processing routes for dusts, selective leaching of zinc, lead and other heavy metals by

hydrometallurgical treatments and high temperature processing aiming to vaporization of zinc and lead and decomposition of heavy hydrocarbons and cyanides. Pyrotreatments are aiming either to reduction of iron oxides to molten iron and production of hot metal or smelting of high iron slag. In the first case the product is charged directly to BOF or cast and used as substitute of iron pigs or scrap, in the latter case the high iron slag is cast and recycled to sintering plant, BF or BOF.

Several industrial and pilot technologies of pyro- or hydrometallurgical as well as hybrid nature have been developed for treatment of steelmaking wastes. The aim of all methods is to separate volatile and hazardous metals and compounds from waste materials before recycling or disposal for landfill. Daido Special Method of slag melting (DSM) [8], CONTOP [7], and RADUST [9,10,11,20] are aiming to production of high-iron oxide slag, OXYFINES [6,12], RMF process (Recycling Melt Furnace) [13] as well as FASTMET/FASTMELT [13], Kawasaki recycling of EAF dust [15] and ROMELT [16] to production of molten iron. In addition to the above mentioned technologies there are a series of other methods for steelmaking waste treatment that are in different stages of development [17].

3. Thermochemical considerations on high-iron slag smelting from iron- and steelmaking dusts

The most important properties of dusts aimed for slag smelting are:

- 1) iron (oxide) content,
- 2) content of volatile/non-volatile heavy metals,
- 3) contents of carbon and hydrocarbons,

4) the morphology of feed mixture and necessity of mechanical pre-treatments for feeding the mixture into the reactor. Iron content of iron and steelmaking dusts is high without exception. The main heavy metals present in fine fractions of BF and BOF-dusts are zinc and lead, both volatile at relatively low temperatures under moderately reducing conditions. Carbon is besides the iron oxides the main constituent in fine fractions of BF-dust and, accordingly, decreases fuel requirements depending on the oxidation degree of combustion.

Evaporation of heavy metals, especially zinc the main harmful metallic constituent in iron- and steelmaking dusts is enhanced by an increase in temperature and decrease in oxidation potential in the process. The utilization of carbon in the feed mixture as fuel is increased by an increase in the degree of oxidation which, however leads to an increase in oxygen potential in the reaction zone and has, accordingly, negative effect to the zinc vaporization that takes place in the form of elemental zinc vapour. So, there might be some optimum in the oxidation level in the process, measured by CO_2/CO -ratio (and $\text{H}_2\text{O}/\text{H}_2$ -ratio) or by oxygen potential, $\lg(p_{\text{O}_2}/\text{bar})$ (or $RT\ln(p_{\text{O}_2}/\text{bar})$). Figure 1 shows the conditions for metallic iron formation in the presence of high iron oxide slag. In principle the process conditions prevailing above the slag-iron equilibrium line in the figure towards more oxidising conditions will lead to formation of only a high-iron slag.

The equilibrium conditions for metallic iron separation in dust smelting as well as evaporation and distribution of charge mixture species between the slag and gas phase (forming dust in cooling) were calculated using HSC 5.1 software [21]. In calculations for iron separation from high iron oxide slag solution thermodynamics of molten iron was roughly modelled using CONSIM 5 oxygen converter simulator [22]. Calculations suggests, that no significant dissolution of carbon or other constituents, present in the dust mixtures treated in the Radust process (presented in the next Chapter), will take place under the relative reducing conditions of iron precipitation. The situation is completely different if the process is aiming to low melting high carbon hot metal production that leads to high carbon activity and highly reducing processing conditions.

As can be seen by comparing the information presented in Figure 1 and Table 3, the processing conditions in Radust pilot campaigns (CO_2/CO -ratio, Table 3) are far more oxidising than those proposed for iron precipitation (Figure 1). Any (equilibrium) formation of solid or molten iron could not be, accordingly, expected and were neither observed in the Radust test runs (reported in the next Chapter). The process aiming to iron oxide slag formation could be run in remarkably more reducing mode without iron formation. However, this would lead to lower degree of carbon (monoxide) utilization and higher fuel (propane) consumption for obtaining the equal temperature of combustion gases and slag as well as higher gas volume per treated amount of dust mixture.

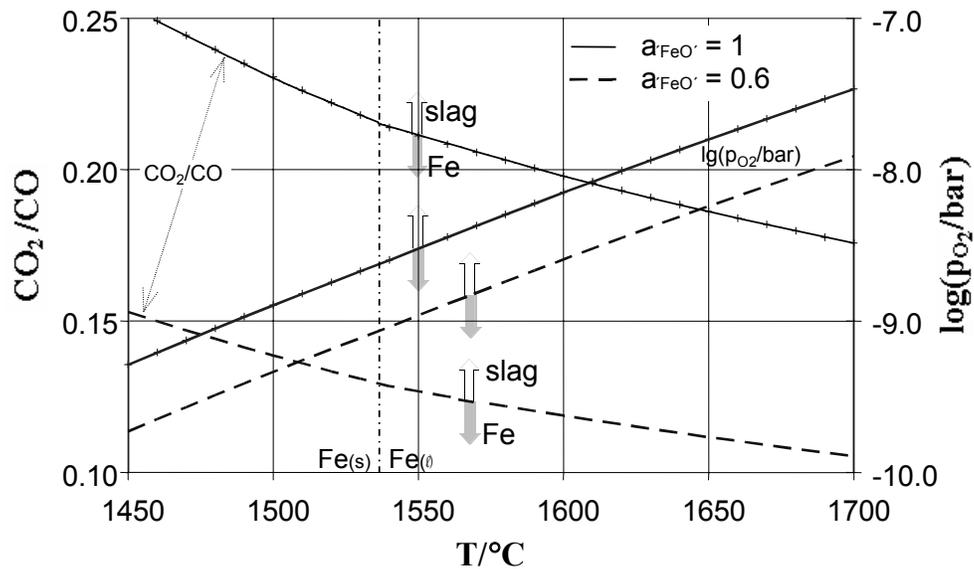


Fig. 1. Limits for formation of iron in smelting of high iron oxide dusts measured in the CO_2/CO -ratio and oxygen potential (logarithm of oxygen pressure). Equilibrium solubility of carbon and other dust constituents in iron seemed to be negligible (for carbon < 0.03 wt-%) at both wustite activities. Stoichiometry for wustite in calculations was $\text{Fe}_{0.947}\text{O}$. Calculations were carried out using HSC Chemistry 5 software [21].

Equilibrium calculations corresponding to the typical conditions of test runs in Radust reactor are presented in Tables 2 and 3. Equilibrium distribution of charge mixture elements between the slag and gas phase presented in Table 3 are calculated using HSC-Chemistry 5 software [21]. As any reliable solution thermodynamic data for Radust-type high iron slag was not available, slag phase was assumed to be ideal solution of monometallic oxide species ($a_{\text{FeO}} = x_{\text{FeO}}$, $a_{\text{FeO}_{2/3}} = x_{\text{FeO}_{2/3}}$ etc.; a = activity, x = mole fraction). Slag melt is surely not ideal especially not for zinc oxide and alkaline oxides (K_2O , $\text{Na}_2\text{O} \rightarrow \text{KO}_{0.5}$, $\text{NaO}_{0.5}$) and the results presented in Table 3 are only qualitative. The calculations suggest anyway, that thermodynamic prerequisites for evaporation of zinc and alkaline elements are

good and high evaporation degree of those components under the conditions of Radust test runs can be expected, even when the reduction degree in the flame and reactor is only moderate.

The fuel requirements for the Radust process were as well evaluated for 1.5 ton per hour (maximum capacity of the pilot reactor) of 1/1-mixture of BF- and BOF-dusts on the base of average dust composition and process parameters of the Radust test campaigns. The results are presented in Figure 2. The relationship is qualitative corresponding to average operational

Table 2. Input data for calculation of equilibrium conditions and heat balance for Radust process: basis the spring campaign 2004

Temp. 25°C	Amount kmol	Amount kg	wt-%	Volume, Nm ³
C ₃ H ₈ (g)	0.446			10
O ₂ (g)	16.646			379.375
N ₂ (g)	5.354			122.037
C	18	216.2	16.7	0.083
SiO ₂	0.896	53.9	4.2	0.021
Fe			38.3	
Fe ₃ O ₄	0.986	228.3	17.7	0.044
Al ₂ O ₃	0.277	28.2	2.2	0.007
MnO	0.031	2.22	0.17	0
CaO	1.977	110.9	8.6	0.033
MgO	0.867	35.0	2.7	0.01
TiO ₂	0.013	1.02	0.08	0
V ₂ O ₅	0.006	1.08	0.08	0
K ₂ O	0.487	45.9	3.6	0.02
Na ₂ O	0.401	24.8	1.9	0.011
ZnO	0.44	35.8	2.8	0.006
S	0.377	12.1	0.94	0.006
Pb	0.003	0.6	0.05	0
P ₂ O ₅	0.008	1.1	0.09	0
Fe ₂ O ₃	2.946	470.4	36.42	0.09
	Σ	1267		

conditions without any serious technical problems. It suggests, however, that with technical improvements of the reactor under the conditions of smooth, continuous operations it is possible to smelt BF-BOF-dust mixture practically without extra fuel.

Table 3. Equilibrium and heat balance consideration for the Radust process

Temp. 1500°C	Amount, kmol	Volume, Nm ³	vol-%		Amount kmol	kg	wt-%
Gas				Slag			
CO ₂ (g)	14.90	339.589	51.4	FeO(l)	4.2800	294.834	32.7
N ₂ (g)	5.35	121.946	18.5	CaO(l)	1.9800	111.037	12.3
CO(g)	4.40	100.281	15.2	Fe ₃ O ₄ (l)	1.6000	370.462	41.1
H ₂ O(g)	1.66	37.207	5.7	SiO ₂ (l)	0.8960	53.836	6.0
K(g)	0.97	21.831	3.4	MgO(l)	0.8670	34.944	3.9
Na(g)	0.79	17.752	2.7	Al ₂ O ₃ (l)	0.2770	28.243	3.13
Zn(g)	0.39	8.674	1.3	ZnO(l)	0.0532	4.329	0.48
SO ₂ (g)	0.38	8.591	1.3	MnO(l)	0.0313	2.22	0.25
H ₂ (g)	0.13	2.932	0.44	TiO ₂ (l)	0.0128	1.023	0.11
Pb(g)	0.0028	0.062	0.01	P ₂ O ₅ (l)	0.0076	1.075	0.12
Na ₂ (g)	0.0002	0.004	0	V ₂ O ₅ (l)	0.0059	1.08	0.12
K ₂ (g)	0.0001	0.003	0	Na ₂ O(l)	0.0041	0.256	0.028
K ₂ O(g)	0	0.001	0	PbO(l)	0.0001	0.029	0.003
Na ₂ O(g)	0	0	0	K ₂ O(l)	0.0001	0.01	0.001
SiO(g)	0	0	0		Σ	901.2	

Adiabatic flame temperature 2285°C

Heat loss: 20.2 % (from input enthalpy)

$$\eta_{\text{CO}} = \frac{\text{CO}_2}{\text{CO}_2 + \text{CO}} = 0.77; \eta_{\text{CO+H}_2} = \frac{\text{CO}_2 + \text{H}_2\text{O}}{\text{CO}_2 + \text{CO} + \text{H}_2\text{O} + \text{H}_2} = 0.74; \text{CO}_2/\text{CO} = 3.4$$

4. Short description of the Radust process

Rautaruukki Company (at the present Ruukki Group) initiated the Radust Project in year 2000 to develop an economically and environmentally feasible dust processing technology. The equipment for smelting high-iron slag from BF- BOF-dust mixtures reached the test run phase in 2002. Since then five test campaigns have been executed [11,20]. The Radust technology is now in the

stage of reconstruction based on the test campaign experiences.

The main purpose of Radust process is to treat the non-recyclable dust fractions from BF and BOF in order to transform them to recyclable high-iron slag and minimize the amount of hazardous components containing fraction to be send for special treatments. Radust process is aiming to

- 1) separation of heavy metals, Zn, Pb, alkali compounds and halides from iron oxide and other non-volatile oxides (typically CaO, MgO, Al₂O₃, MnO) of BF- and BOF-dust,
- 2) elimination of hazardous organic compounds (hydrocarbons, cyanides etc.) by combustion,
- 3) production of high iron (oxide) slag, valuable raw material for recycling into BOF or BF,
- 4) maximum utilization of carbon of BF-dust as energy source

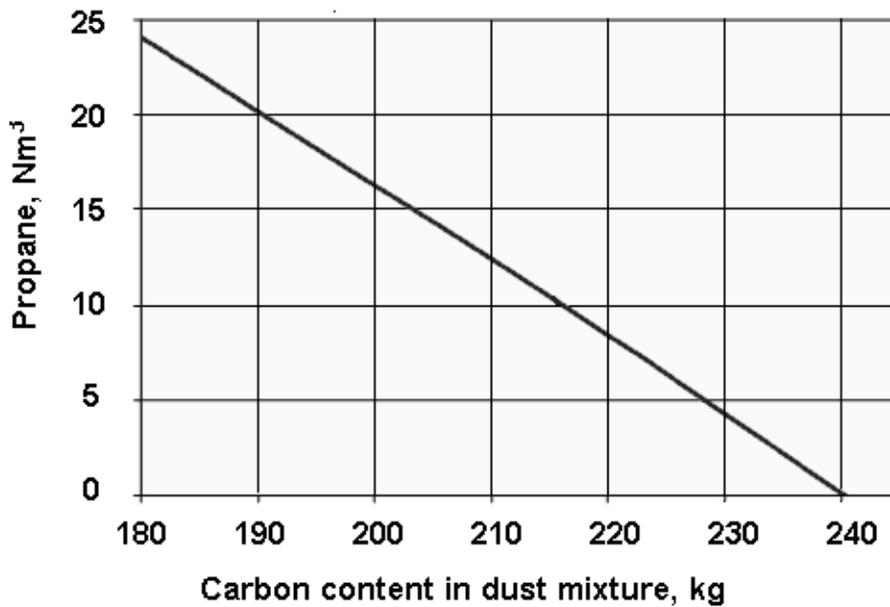


Fig. 3. Substitution of fuel (propane) by carbon content in charge. Calculated for one-to-one mixture of BF and BOF dust for feed rate 1500 kg / hour. Based on optimal continuous smooth smelting conditions without brakes.

5. Characteristics of the Radust technology

Equipment diagram of the process and the general view of the Radust plant is presented in Figures 3 and 4. The mixture of BF-dust with high carbon content (typically over 30 %) and low carbon BOF-dust (see Table 1) is burned with oxygen in propane flame. Dust mixture is pneumatically fed in the burner. Molten slag droplets are collected onto the bottom of the reactor from which the melt is time to time poured in slag pots. Hazardous organic components are burned in the flame and combustion gases with volatile heavy metals are led through the combustion chamber, in which carbon monoxide is burned and volatile heavy metals oxidised, to the dust collection units, cyclone separator and filter box.

The thermal capacity of Radust pilot reactor was estimated to be around 2.9 MW corresponding to capacity 1500 kg/h (30-33 ton /day) of dust mixture. Capacity of the equipment was enough, when the smelting was proceeding smoothly without any technical problems, to treat all BF dust from electrostatic precipitator and an equal amount of BF-dust produced at Koverhar Steelworks [11,20].

The burning of the dust mixture is aimed to be controlled so that high evaporation degree of zinc, lead as well as potassium and sodium oxides, depending on temperature and oxygen potential, is ensured with maximum utilization of carbon. Combustion control was semi-automated (Siemens PCS7) on the characteristics of the charge mixture (feeding rate, charge mixture composition, especially its carbon content) and thermal requirements of the process and the reactor (flame and slag temperature, heat loss of the reactor). Consumption of extra fuel (propane) is dependent on the carbon content of the charge mixture (see Figure 3) and on smoothness of the operation without brakes. High flame temperature, far above 2000°C (see estimation of adiabatic flame temperature in Table 3) with moderate reduction conditions guarantees effective evaporation of zinc, lead and alkaline components. High slag temperature (average temperature of slag in test campaign has been at around 1500°C) is resulted from high flame temperature. The oxidation degree in combustion (target value for so called “carbon monoxide utilisation degree” or $\text{CO}_2/(\text{CO}_2+\text{CO})$ -ratio in combustion gas is around 0.8) is enough for effective evaporation of zinc and lead (see

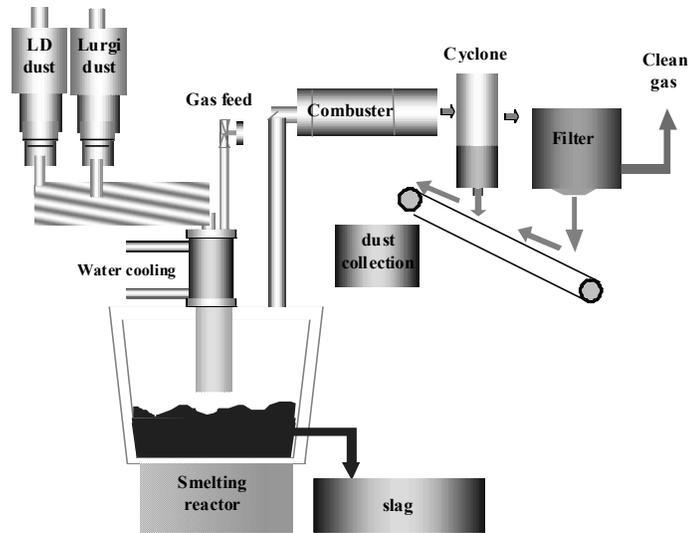


Fig. 4. Equipment diagram of Radust process



Fig. 5. General view of Radust plant in Ruukki Group Koverhar steelworks (at present Ovako Koverhar Steelworks)

Tables 2 and 4), but safely above the limit of iron reduction (compare information given in the Fig. 1 and Table 3).

6. Test campaigns of Radust process

Five test campaigns were launched since 2002. The main target values of the process are slag temperature and basicity (CaO/SiO₂-ratio) as well CO₂/(CO₂+CO)-ratio of the combustion gas. The rough target values for these three parameters have been 1500°C, 2.2 and 0.8, respectively. The feed rate in the several test periods has varied mainly within limits 1400 to 1500 kg / hour, dust mixture ratio of BF/BOF-dusts has been close to 1/1.

The results of the test campaigns met the expectations. When the process operated smoothly without any serious technical problems, the degree of zinc and alkaline elements separation to dust has been high, close to or above 90% (see the results of latest test period presented in Fig. 4 and Table 2). The average zinc content of slag in the technically successful test runs in three first campaigns was 0.28% [11], potassium oxide and sodium oxide contents in slag in the various campaigns have been varied between 0.5 to 1.7 and 0.4 to 1.6, respectively. There have been one week periods with remarkably low zinc (down to 0.04%) and alkaline oxide contents [11]. Iron content of slag is constantly close to 50% which means over 60% total content of iron oxides (FeO+Fe₂O₃). Due to that and low contents of volatile components observed in the most successful test periods, slag produced by Radust technology is appropriate for recycling back to BF and BOF. It can also be used for production of construction materials if the slag composition is modified by suitable additions (silica sand, lime).

The amount of environmentally problematic dust with heavy metals, alkaline compounds and chlorine is reduced to one fifth of the original amount. Rate of alkaline element, especially potassium removal to gas has been clearly found to be dependent on slag temperature (corresponding with the flame temperature) and basicity; increase in both of these factors enhance the removal of alkaline elements [11]. The average (analysed) values observed from in the filter dust of Radust process during the first three test periods have been 17% for iron, 24 % for potassium oxide, 4,4 % for sodium oxide, 11 %

for zinc 5,7 % for sulphur and 13 % for chlorine [11]. The maximum analyzed values for lead and fluorine in Radust dust have been 2.9 and 2.2%, respectively. Dust fractions with high zinc, lead and alkaline content collected into a dust separator unit are to be sent for special treatment.

The results of last campaign (spring 2005) are presented in Table 4 and Figure 4 for temperature and zinc content of slag. The variation in both slag temperature and zinc content of slag emphasize the importance of control over combustion conditions, flame temperature and oxidation degree affecting the separation efficiency of volatile components.

The Radust process is laid down since the spring 2005 for technological reconstructions that are based on the experience from test campaigns.

Table 4. Mass balance and component distribution between slag and dust (Radust campaign 22.3 to 20.4.2005)

T _{slag} (average) 1488°C	Charge	Slag	Dust	Recovery in dust %
BF/BOF	49 / 51			
kg	456673	298983*	83650*	
Fe wt%	38.4	50.28	30.05	
SiO ₂ “	3.28	6.14	3.59	
MnO “	0.18	0.28	0.12	
P ₂ O ₅ “	0.04	0.05	0.10	
Al ₂ O ₃ “	1.25	1.84	1.08	
CaO “	8.72	14.08	3.12	
MgO “	1.46	1.90	1.20	
TiO ₂ “	0.02	0.11	0.03	
V ₂ O ₅ “	0.08	0.22	0.11	
K ₂ O “	(2.3)**	0.87	13.68	82***
Na ₂ O “	(1.7)**	0.40	6.44	81***
C “	16.2	-	-	
S “	0.64	0.33	1.74	
Zn “	2.14	0.29	10.49	90
B2 (CaO/SiO ₂) = 2.2				

* From iron distribution

** Rough estimation

*** Calculated from dust-slag distribution

7. Conclusions

The thermochemical consideration demonstrated the feasibility of the Radust-concept in treating the most problematic steelmaking dusts to high iron, low impurity slag under relatively highly oxidising conditions allowing high carbon utilization degree.

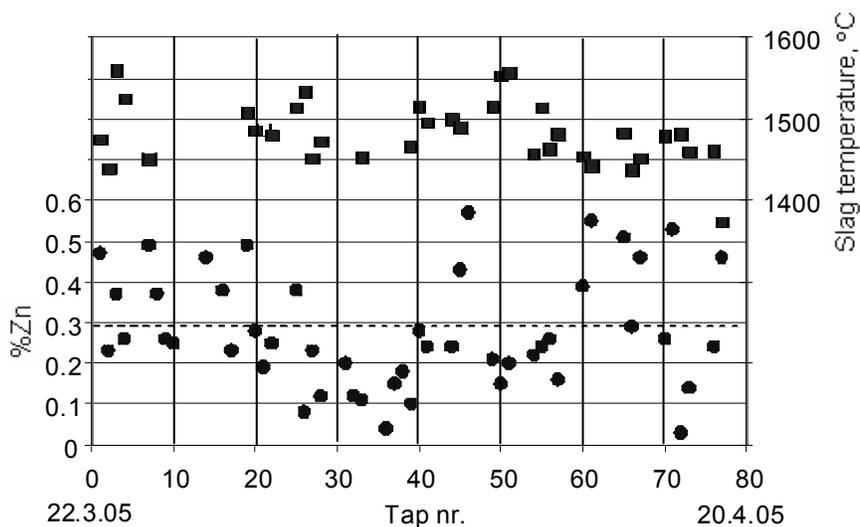


Fig. 4. Variation in temperature and zinc content of slag in the Radust spring campaign 22.3 to 20.4 2005.

The test campaigns have demonstrated the capability of the Radust concept in treating of the most problematic steelmaking dusts to “clean” high iron slag with low contents of zinc, lead and alkaline oxides acceptable both for recycling and landfill disposal. The variation in the contents of critical constituents zinc and alkaline in the slag are due to problems of smooth process control that is not a straightforward task for such a complex raw material as mixture of fine dusts with large variation in composition and morphological characteristics (particle size and specific weight).

The main technical improvements to be done in the Radust technology are related to feeding of fine dust mixture composed of particles with to the burner, smooth control of oxygen to feed mixture ratio necessitating continuous control of CO/CO₂-ratio in the reactor gas (*control based on only*

dust analysis is problematic due to relative large changes in the dust mixture composition) and off gas combustion [11].

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