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Recycling of the Steelmaking by-products into the Oxygen Converter Charge

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INTRODUCTION

Oxygen converter slag

Converter slag is by-products and utilizable production residues arising from the steel production process in oxygen converter. Is formed by the oxidation of pig iron, the dissolution of slag-forming additives and partial melting of the refractory lining. The final properties of solidified steelmaking slags are given by chemical composition, ratio of crystalline and glassy phase, mineralogical composition and crystal size, porosity induced by gas dissolved in slag. Table 1 described relationship between the characteristics and applications of steelmaking slag.

Table 1 Characteristics and applications of steel slag

Characteristics	Applications
Hard, wear-resistant, adhesive, rough	Aggregates for road and hydraulic construction
Porous, alkaline	Waste water treatment
FeOx, Fe components	Iron reclamation
CaO, MgO, FeO, MgO, MnO components	Fluxing agent
Cementitious components (C3S, C2S and C4AF)	Cement and concrete production
CaO, MgO components	CO_2 capture and flue gas desulfurization
FeO, CaO, SiO_2 components	Raw material for cement clinker
Fertilizer components (CaO, SiO_2, MgO, FeO)	Fertilizer and soil improvement

Source: (Huang, Y. et al., 2012)

The amount of oxygen converter slag is about 100-150 kg per tonne of crude steel produced (Lan et al., 2017). The amount of produced slag is primarily dependent on the ratio of scrap to pig iron in the batch, the grades of the steel being produced, the initial chemical composition of the pig iron and the amount of slag additives added. At present, there is an effort to reduce the amount of slag arising, which is possible by pretreatment of pig iron, secondary metallurgy of molten steel (Socha et al., 2012) and careful selection of batch materials. Despite all efforts, the amount of slag produced represents about 13% of the total batch. Reducing, treatment and utilization of waste is required by international law (Kardas et al., 2013). The current utilization rate of steel slag is only 22% in China (Huang et al., 2012).

The basic constituents of slags are CaO, originating from slag-forming additives, SiO_2 and FeO from the metal bath oxidation reactions. These three major components account for about 80% of the total. The chemical and mineralogical composition of steelmaking slags is variable and depends to a large extent on the production aggregate as well as on the grades of steel produced (Podorska & Wypartowicz, 2018). Table 2 describes the average chemical composition of oxygen converter slag, it will be the most significant factor affecting its further potential use.

Table 2 The average chemical composition of oxygen converter slag

Fe_{total}	CaO	MgO	SiO_2	MnO	Al_2O_3	P_2O_5	S
[%]							
14-30	25-55	2.5-15.0	9-19	1.5-10.0	0.9-7.0	$\leq$ 0.9	$\leq$ 0.07

Source: (Huiting and Forssberg, 2003)

From the mineralogical point of view, the structure of the steelmaking slag consists of four main phases: dicalcium silicate $2CaO \cdot SiO_2$, stabilized tricalcium phosphate $3CaO \cdot P_2O_5$, solution of bivalent metals oxides, e.g. Ca, Fe, Mn, Mg, and solid solutions of calcium and aluminium ferrites $2CaO \cdot Fe_2O_3$ – $2CaO \cdot 2Al_2O_3 \cdot Fe_2O_3$. In some slags with high lime content, there is also tricalcium silicate $3CaO.SiO_2$. The steelmaking slag also contains free lime. Free lime in slag is actually a solid solution CaO – FeO or CaO – MnO containing up to 20% FeO or $FeO \cdot MnO$.

Slag from steel production is usually tapped from a steel furnace into a slag ladle at a temperature of approximately 1600°C. The slag progresses to the heap, where it is spilled to solidify in crystalline form. The process of solidification can be accelerated by the addition of a limited amount of water. The steelmaking slag has an appearance of igneous rock. These slags contain a large proportion of metallic iron, which can be separated from steelmaking slag using suitable methods. The simplest treatment of steelmaking slags appears to be the multi-stage crushing, grinding, and electromagnetic separation processes, in which metallic parts are separated from the slag. The principle of magnetic separation of steelmaking slag is the utilization of electromagnetic properties of metallic components.

By this type of treatment it is possible to obtain:

- high-quality metal lumps suitable for direct use in the metallurgical plant (agglomeration, blast furnace, steelworks),
- fine-grained concentrate of different fractions, which high calcium and magnesium content makes it suitable for reuse. This fine-grained concentrate is referred to as demetallized steelmaking slag.

The presence of CaO, MgO and MnO can be a positive factor in the steelmaking process, as they can replace part of limestone, dolomite and manganese ore and, consequently, reduce iron and steelmaking production costs (Fernandéz-Gonzáles et al., 2019). The main emphasis must therefore be placed on economic and technological aspects of the use of secondary products (Brožová et al., 2014). In the next part we will describe the possibilities of using this part of the steelmaking slag.

Oxygen converter dust

Oxygen converter process produces a large amount of dust that is emitted with the evolved converter gases. This dust is a major pollutant in steel plants using the oxygen converter. Elaborate dust abatement systems are required to separate the dust from the flue gases before the gas is allowed. These by-products contain high iron and carbon content which encourages the idea of recycling such products. However, certain compounds may form a challenge to recycle such products. These compounds include PbO, ZnO, Na_2O, K_2O, sulphates and chlorides.

Flue dusts, by-products of oxygen converter process, are together with mill scales the most valuable secondary raw materials produced in steelmaking (Gritzan & Neuschütz, 2001). Economical evaluation of reducibility of compacted metallurgical dust with high ratio of iron is very high (Jursová et al. 2016). Recycling of metals from secondary sources is a growing industry (Pustějovská et al., 2014).

The amount of generated dust is dependent on the blowing practice, the bath chemistry, and the composition and granulometry of the input materials. Nevertheless, a fairly good estimation can be made for a given set of operating parameters. When the heats are cooled with ore, dust generation varies between 13 and 25 kg/ton of steel, but when heats are cooled with scrap, the amount is between 21 and 32 kg/ton (Pribulová et al., 2018).

It is believed that such information will be useful in finding some better utilization of the dust, rather than recycling it back to the sintering plant. The chemical composition of the dust can vary widely depending on whether or not the gas evolved, which contains 70-95% carbon monoxide (CO), is subsequently burnt to carbon dioxide (CO_2) by adding secondary air. The former process is known as total combustion, while the latter is termed suppressed combustion (Santanu et al., 1997).

The insertion of charging materials in the beginning of the blow results in production of the majority of the flue dust. Slag-forming materials are typically charged in the first minute of the blow when the bath surface is not covered with the emulsion. If lime and dolomite lime are insufficiently sorted, a large amount of particles are too small and hence easily drawn into the exhaust burnt gases. Bigger particles get onto the bath surface where they dissolve nevertheless, they can still get into the burnt gases before the slag-metal emulsion is formed. The flue dust produced in the first stages of the blow contains partially melted particles of lime and dolomite lime. This indicates that these particles were in contact with the bath surface.

A large portion of the flue dust is formed by evaporation. On average, 17% of the flue dust in the first stage of the blow, and 32% of the flue dust in the second stage (Yamamoto et al., 1994). Fast evaporating elements, such as Zn, Pb and Cl, occur mostly in the flue dust in the beginning of the blow, and slower evaporating elements, such as Mn and Cu, were observed in the flue dust at the end of the blow, as shown in Fig. 1 (Mihok, 1994).

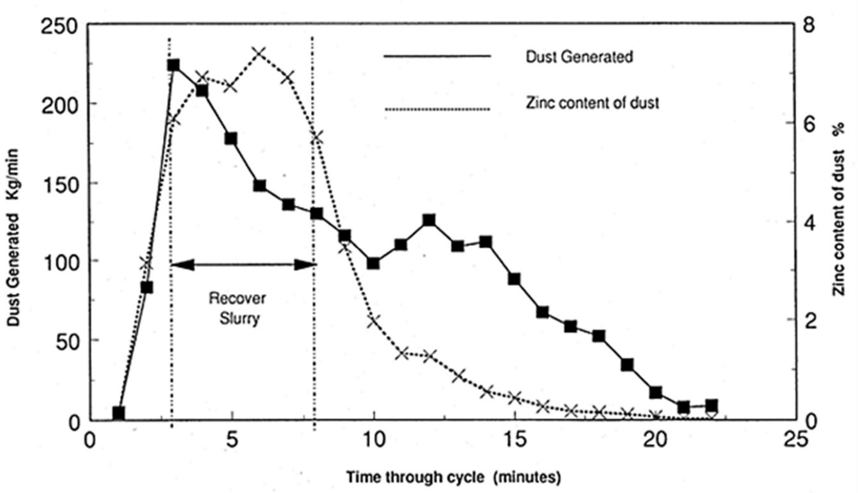

Fig. 1 Typical development of converter flue dust in the BOF process including the regeneration of zinc-rich sludge

The use of certain portion of the converter flue dust by returning it into the charge for an oxygen converter began in 1980s in trial operations. At present, flue dust is collected in an electrostatic precipitator, it is subsequently hot-briquetted in an inert atmosphere and dosed into a convertor as a partial replacement for scrap. The use of briquettes has also other benefits such as (Cecca, et al., 2017):

- high bulk density, if compared to the usual morphologies of the steel scrap, thus implying savings in the volume taken up by the scrap,
- known and homogeneous chemical composition,
- minimum amount of undesired elements difficult to be controlled in the scrap charge, i.e. Cu, Ni, Cr, Mo, Sn, and Pb,
- high thermal conductivity allowing a fast dissolution within the converter,
- low tendency for reoxidation in contact with fresh and/or salted water.

MATERIALS AND METODS

Recycling of the demetallized steelmaking slag into the oxygen converter charge

As mentioned above, demetallized steelmaking slag contains valuable basic components and bound iron, so there goes on a search for the possibility of its use in the steelmaking process itself. The team of authors, investigated the possibility of recovering part of the slag by return dosing into the oxygen converter. Recycling of slag in the refining process in oxygen converter can serve as an initiator of slag formation, as a medium providing the beginning of the dissolution of lime at the beginning of the refining process. Demetallized slag recycling can reduce the cost of slag-forming additives (lime). The chemical composition of the dosed demetallized slag, and lime is presented in Table 3.

Table 3 Chemical composition of the dosed demetallized slag and lime [%]

	Demetallized slag [%]	Lime [%]
SiO_2	11.88	0.44
$Fe_{tot.}$	21.78	0.83
CaO	45.36	97.40
MgO	4.00	0
Al_2O_3	1.02	0
FeO	14.22	-
P	0.11	-
S	0.26	-
MnO	5.30	-

The part of experiments consisted of 12 melting processes in the same converter. The first three melting processes were carried out without the addition of demetallized slag. The next 3 melting processes with 1.000 kg of demetallized slag, and last 6 melting processes with the addition of 4.000 kg of demetallized slag. Three of them were prepared with a reduced amount of charged lime (2.000 kg).

Recycling of the flue dust briquettes into the oxygen converter charge

The experiments were carried out using the briquettes made from oxygen converter flue dust. The used briquettes were of an elliptical shape with the following dimensions: the length of 74-75 mm, the width of 43-45 mm and the thickness of 25-27 mm.

The briquettes were transported by a belt conveyance system to the charging box above the convertor. Within the thermal planning of melting processes to be carried out using the briquettes. It was necessary to count with a cooling effect which is as much as 2-2.5 times higher than the cooling effect of steel scrap. The briquettes are not used in the production of steel with the target sulphur content below 0.008% and nitrogen content below 0.007%. The chemical composition of the dosed briquettes is presented in Table 4.

Table 4 Chemical composition of used briquettes [%]

Fe met.	Fe tot.	FeO	Fe_2O_3	SiO_2
36.750	68.750	30.320	12.150	1.420
Al_2O_3	**Mn**	**P**	**S**	**C**
0.140	0.530	0.064	0.047	2.350
Na	**K**	**Cu**	**Ni**	**Cr**
0.500	0.364	0.026	0.001	0.028
MgO	**Zn**	**Cd**	**Pb**	**CaO**
2.500	0.585	0.001	0.131	7.750

The experiments were carried out in 15 processes of steel melting in an oxygen converter. Out of them five melting processes included the addition of 1.000 kg of briquettes at the beginning of the refinement with the first dose of lime, other five processes included the addition of 2.000 kg of briquettes, and the last five processes included the addition of 3.000 kg of briquettes.

ANALYZIS OF EXPERIMENTS

Recycling demetallized steelmaking slag into the oxygen converter charge

Table 5 contains the basic parameters of the performed melting processes in an oxygen converter.

Table 5 Basic parameters analysed melting processes in an oxygen converter

Melt	Total metallic charge [kg]	Demetall. slag [kg]	Lime [kg]	Temp. of steel [°C]	Time of blowing [min.]	Time of after blowing [m³]	Yield of liquid steel [%]
A1	190 470	0	13 526	1 660	27.16	1.27	97.0
B1	190 870	0	13 534	1 670	28.09	0	91.1
C1	192 000	0	12 266	1 665	27.02	0	96.7
D1	192 300	1 200	9 578	1 665	28.14	0	90.1
E1	192 100	1 303	9 536	1 665	26.51	0	93.4
F1	192 100	1 428	11 720	1 665	28.30	0	94.2
G1	191 600	3 934	10 544	1 665	26.32	0	91.3
H1	193 490	4 029	10 520	1 700	25.36	1.34	93.2
I1	194 490	4 001	11 348	1 670	28.28	0	86.6
J1	191 900	3 922	8 630	1 675	29.35	0	93.5
K1	188 600	3 926	10 042	1 590	28.31	1.51	92.5
L1	190 880	3 924	11 564	1 700	28.02	1.50	93.2

The sampled melts were identical or similar steel grade. Samples were collected from each melting process during an interim break and after the blow completion. In melting process A1, H1, K1, L1 included afterblow after which the sampling was carried out. The development of the contents of phosphorus and sulphur in steel presented by partition ratios for sulphur and phosphorus since the beginning of the refinement in an oxygen converter is presented in Table 6.

Table 6 Partition ratios for sulphur and phosphorus

Melt	Partition ratios for sulphur (S)/[S]			Partition ratios for phosphorus (P)/[P]		
	Po(S)/Po[S]	P1(S)/P1[S]	Po(S)/Po[S]	P1(S)/P1[S]	Po(S)/Po[S]	P1(S)/P1[S]
A1	10.714	9.000	10.333	22.50	36.97	32.75
B1	10.143	10.000	-	41.04	34.57	-
C1	4.158	5.813	-	9.85	30.56	-
D1	7.462	7.364	-	9.98	30.56	-
E1	4.684	6.733	-	25.78	46.57	-
F1	4.000	5.375	-	8.83	28.53	-
G1	5.118	6.231	-	50.81	40.98	-
H1	2.690	3.680	4.261	9.78	20.38	26.49
I1	2.676	3.448	-	31.44	27.29	-
J1	4.471	5.200	-	8.73	20.04	-
K1	2.258	3.423	3.840	15.28	32.38	37.28
L1	3.100	4.520	9.364	9.63	20.88	23.77

Table 7 contains chemical composition of slag generated during the melting processes. No dependence was found between the content of the individual structural components and the addition of demetallized slag. On the other hand. with the increasing addition of demetallized slag the content of free lime in the slag decreased.

Table 7 Chemical composition of slag generated during the melting processes

Sample	Fe tot. [%]	MnO [%]	SiO_2 [%]	Al_2O_3 [%]	CaO [%]	MgO [%]	P_2O_5 [%]	S [%]	Free lime [%]
$A1_0$	7.55	5.45	15.21	0.98	59.43	5.08	1.34	0.075	0.070
$A1_1$	14.16	6.22	11.48	0.67	55.05	3.31	1.27	0.063	0.035
$A1_2$	17.35	5.99	10.82	0.70	51.98	3.43	1.05	0.062	0.014
$B1_0$	15.42	8.24	14.28	1.17	47.03	4.47	1.41	0.071	0.012
$B1_1$	12.56	5.14	10.41	0.81	59.70	3.57	0.95	0.060	0.070
$C1_0$	7.40	7.66	20.87	2.00	47.28	8.19	0.88	0.079	0.005
$C1_1$	13.95	7.43	13.22	2.08	49.72	5.13	1.05	0.093	0.004
$D1_0$	7.25	6.05	18.60	1.96	52.32	7.99	0.96	0.097	0.024
$D1_1$	7.76	7.54	20.40	3.17	47.28	7.96	0.84	0.081	0.004
$E1_0$	13.66	8.4	14.85	1.58	46.58	6.39	1.24	0.089	0.013
$E1_1$	11.94	8.58	15.84	1.98	47.40	6.31	1.28	0.101	0.004
$F1_0$	8.51	8.61	21.92	2.34	44.83	7.55	0.91	0.072	0.005
$F1_1$	14.69	8.14	13.69	1.75	45.07	7.99	0.98	0.086	0.048
$G1_0$	11.25	7.61	17.38	1.78	49.59	4.72	1.28	0.087	0.008
$G1_1$	10.61	7.39	16.75	1.99	50.63	5.28	1.22	0.081	0.006
$H1_0$	7.91	7.64	22.91	1.85	47.43	6.14	1.03	0.078	0.006
$H1_1$	19.82	6.37	12.90	1.13	45.24	3.89	0.98	0.092	0.026
$H1_2$	20.22	6.24	12.59	1.06	45.39	3.77	0.91	0.098	0.005
$I1_0$	13.83	6.19	18.11	3.31	45.28	4.84	1.08	0.091	0.002
$I1_1$	20.17	5.01	12.14	2.03	45.0	5.09	0.75	0.100	0.007
$J1_0$	7.59	8.70	21.63	2.75	47.73	5.68	0.96	0.076	0.004
$J1_1$	9.59	8.30	19.10	2.5	48.27	5.41	1.01	0.078	0.004
$K1_0$	8.44	9.88	23.64	3.03	44.78	3.87	1.05	0.070	0.003
$K1_1$	23.39	7.00	14.40	1.77	37.79	3.74	0.89	0.089	-
$K1_2$	16.16	7.08	13.01	1.52	47.99	4.85	1.11	0.096	0.003
$L1_0$	8.29	7.39	22.01	1.92	48.01	6.15	0.97	0.093	0.004
$L1_1$	13.13	6.32	16.54	1.36	49.70	4.73	1.10	0.113	0.013
$L1_2$	13.86	5.24	12.54	0.92	55.06	3.99	0.98	0.103	0.038

Recycling of flue dust briquettes into the oxygen converter charge

Table 8 shows the basic parameters of the performed melting processes in an oxygen converter and the sampled melts were identical or similar steel grade.

Table 8 Basic parameters of individual melting processes in an oxygen converter

Melt	Total metallic charge [kg]	Briquettes [kg]	Temp. of steel [°C]	Time of blowing [min.]	Time of after blowing [m³]	Yield of liquid steel [%]
A2	194 871	1 000	1 666	27.23	0	87.7
B2	191 473	1 000	1 662	27.23	0	83.9
C2	194 853	1 000	1 689	25.57	0	89.1
D2	194 406	1 000	1 657	27.24	0	89.8
E2	195 751	1 000	1 656	32.59	0	88.8
F2	196 504	2 000	1 641	25.15	0	90.1
G2	193 910	2 000	1 644	26.22	0	86.9
H2	191 897	2 000	1 673	18.42	0	96.6
I2	194 747	2 000	1 674	24.42	0	94.6
J2	193 563	2 000	1 665	26.38	0	90.2
K2	197 743	3 000	1 671	31.12	0	88.6
L2	197 436	3 000	1 675	26.34	0	92.9
M2	197 742	3 000	1 678	28.35	0	88.9
N2	197 476	3 000	1 660	26.12	0	89.4
O2	495 896	3 000	1 649	25.57	1.11	86.3

Sampes were collected from each melting process during an interim break and after the blow completion. Only melting process O_2 included afterblow after which the sampling was carried out.

The development of the contents of phosphorus and sulphur in steel presented by partition ratios for sulphur and phosphorus since the beginning of the refinement in an oxygen converter is presented in Table 9. The development of desulphurization and dephosphorization was absolutely regular, typical for the oxygen converter process.

Table 9 Partition ratios for sulphur and phosphorus

Melt	Partition ratios for sulphur (S)/[S]			Partition ratios for phosphorus (P)/[P]		
	Po(S)/Po[S]	P1(S)/P1[S]	P2(S)/P2[S]	Po(P)/Po[P]	P1(P)/P1[P]	P2(P)/P2[P]
A2	3.46	4.71	-	13.00	30.15	-
B2	2.70	4.17	-	23.07	34.07	-
C2	2.65	4.14	-	20.38	32.23	-
D2	5.05	4.14	-	20.81	45.40	-
E2	3.00	6.31	-	44.67	63.33	-
F2	4.86	6.19	-	72.33	39.31	-
G2	2.19	4.52	-	20.52	41.50	-
H2	4.75	4.95	-	55.78	51.25	-
I2	3.44	5.18	-	21.26	35.21	-
J2	2.50	3.04	-	23.26	61.13	-
K2	3.36	5.00	-	59.50	74.20	-
L2	4.62	6.17	-	12.65	30.06	-
M2	4.00	6.42	-	42.14	66.00	-
N2	4.76	5.87	-	55.18	52.40	-
O2	6.00	7.20	8.38	69.88	97.80	101.50

Nitrogen content values in the steel were at a rather stabile level and were not affected by the amount of added briquettes made of converter flue dust. Also zinc, tin and lead contents in samples of refined liquid metal and steel collected from the examined melts were not affected by the amount of added briquettes made of converter flue dust.

RESULTS OF EXPERIMENTS

Results of the recycling demetallized steelmaking slag into the oxygen converter charge

No negative effects of demetallized slag on parameters of the refining process, particularly dephosphorization and desulfurization was found. Partial replacement of charged lime is recommended. Substitution ratio lime – demetallized slag was determined to 1:2. Recycling of demetallized slag had little effect on the resulting chemical composition of converter slag. Free lime contained in the final slag was reduced with increasing amount of recycled demetallized slag. Recommended recycling ratio of demetallized slag is up to 25 kg per tonne of crude steel.

Results of the recycling flue dust briquette into the oxygen converter charge

Within the research of dephosphorisation and desulphurisation processes in an oxygen converter it was observed that the highest partition ratios for phosphorus and sulphur were detected in the melts produced with the addition of 3.000 kg of briquettes. This applies to the comparison with the melts produced with the addition of 1.000 and 2.000 kg of briquettes because the partition ratios for the melts produced without the addition of briquettes were higher. Adding briquettes did not affect the steel quality. Adding briquettes was most significantly reflected in the composition of convertor flue dust in the content of phosphorus and accompanying elements. The research indicates that the addition of briquettes made of converter flue dust into the charge for an oxygen converter has no negative effects on the refinement process or the quality of the produced steel. The recommended dose of the addition is 3.000-4.000 kg of briquettes in a single melting process. However this recommendation should be supported with an examination of the amount of the produced flue dust which may be significantly affected by the amount of added briquettes.

CONCLUSION

In the case of demetallized convertor slag recycling no negative effect of demetallized slag on parameters of the refining process, particularly dephosphorization and desulfurization was found. Substitution ratio lime – demetallized slag was determined to 1:2. Also in the of case recycling of converter flue dust in form of briquettes info oxygen converter charge has had no negative effects on the refinement process and on the quality of the produced steel. The recommended dose of the addition is 3.000-4.000 kg of briquettes in a single melting process. If it is possible to use steelmaking by-products perfectly, the amount of material being deposited in landfills can be decreased by millions of tons annually. Very important is also the preservation of natural resources. Effectivity utilization of steelmaking by-products is environmentally acceptable as well as the use of natural materials and poses no increased risk to human health or the environment. Oxygen converter slags, and flue dust assist the sustainable preservation of natural landscapes and reduce CO_2 emissions. Products made from slag and flue dust replace products of natural origin and therefore help to preserve non-renewable natural resources. Recycling of by-products can reduce the cost, however it is necessary to thoroughly know their chemical composition and physical properties. This condition is determining to the possibility of utilization of by-products.

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Abstract.
In the technological process of the steelmaking plant, secondary products are produced in parallel with the production of the main product, which have the character of secondary by products or industrial waste. The major secondary products of steelmaking production include waste gases, process fluids, flue dust, sludge, slags and mill scales. The paper presents the results of research project directed to the utilization of demetallized steelmaking slag and oxygen converter flue dust in charge of top blowed oxygen converter. The influence of demetallized slag additions on slag regime in converter and chemical composition of final slag is described and discussed. Recommendations concerning amount of demetallized slag additions are also presented. Flue dust was recycled in form of briquettes. No significant effect of the recycling demetallized converter slag and flue dust briquette on process of hot metal refining and on quality of produced steel were recorded. Regarding the achieved results it can be confirmed that the use of the secondary products like demetallized slag and convertor flue dust in form briquettes, is environmentally acceptable as well as the use of natural materials and poses no increased risk to human health or the environment.

Keywords: oxygen converter, demetallized slag, flue dust, recycling