Multidiszciplináris Tudományok, 15. kötet, 2. sz. (2025) pp. 93–102. **https://doi.org/10.35925/j.multi.2025.2.6**

ANALYSIS OF RECYCLABILITY OF STEELMAKING SECONDARY METALLURGICAL SLAG IN THE ELECTRIC ARC FURNACE

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Abstract
In the steel-making process, the use of slag-forming materials is crucial. However, these materials become wasted after completing their metallurgical task, leading to environmental burdens and costs. Our investigation focuses on the first research phase of finding ways to utilize slags from the secondary treatment of steel production. Our findings reveal that this method could significantly reduce the amount of burnt lime used, thereby reducing greenhouse gas emissions both in the steel plant and the supplier's end. This result contributes to the green steel production technology development implemented so far at the Max Aicher group of companies, including Ózdi Acélművek Kft.

Keywords: *Electric Arc Furnace (EAF), Green steel production, Secondary metallurgical slag (SMS), Cleaner environment, Energy conservation*

1. Introduction

The steel industry plays a vital role in the development of global infrastructure. However, it also produces a significant amount of secondary metallurgical slag as a byproduct. This slag, mostly consisting of oxides, impurities, and alloying elements (Yildirim and Prezzi, 2011), has traditionally been disposed of in landfills or stockpiled. This has caused environmental issues and inefficiency in resource utilization (Danilov, 2003). As steel demand continues to rise worldwide, reducing its environmental impact becomes increasingly urgent. Fortunately, there is a promising solution - the recycling of secondary metallurgical slag in the EAF. This process not only cuts down on waste but also helps conserve precious resources. By reintegrating slag into the steelmaking process, we can reap various benefits, from reducing energy consumption and raw material usage to lowering greenhouse gas emissions.

The sustainable utilization of steelmaking slag has been made possible thanks to prior research. In particular, the ground-breaking work by Yu Zhang et al. (Zhang et al., 2022) has set the stage for further studies into this important field that have probed into the chemical composition and reactivity of slag. On the other hand, it is worth noting that many have recognized the immense potential of utilizing it as a primary ingredient for the production of cement (Shen and Forssberg, 2003; Geiseler, 1996). Modeling was used to assess valorization opportunities for secondary metallurgy slag, taking into account its impact on the economy and the environment (Wen et al., 2017).

There are two companies producing steel raw materials in Hungary, one of which is Ózdi Acélművek Kft., which belongs to the Max Aicher Group of Companies. The company has been operating continuously since 2000. Nowadays, with a production capacity of about 400,000 t/year, its products are used in bar and roll form, primarily as reinforcing steel in the construction industry. An essential element of the strategy of the corporate group, including Ózdi Acélművek Kft., is the environmentally friendly implementation of the applied technological elements. The electric arc furnace steel production technology used at the Ózd Steel Works uses 100 percent steel waste as raw material, which basically satisfies the concept of "green steel". The production of liquid steel takes place in two technological steps; the first phase includes the melting of the steel waste and the basic oxidation processes; during the second, so-called secondary treatment, the final adjustment of the chemical composition takes place in order to ensure the achievable mechanical properties of the finished product. The most important and largest amount of slag-forming material of the applied technology is burnt lime in both phases, in the amount corresponding to the metallurgical goal to be achieved.

Understanding slag chemistry, mineralogy, and phase transformations is crucial for optimizing steel-making processes. Through this study, we aim to provide valuable insights into the behavior of secondary slag within the EAF environment. Additionally, our aim in this investigation is that reusing SMS can be a viable solution for reducing the usage of lime during the early stages of steelmaking. This has the potential to not only improve the efficiency of the process but also contribute towards more sustainable and eco-friendly steel production.

2. Methodology and experimental work

Sampling is carried out from 20 steel batches of arc furnace (primary) and secondary furnace final slags, of the 20 portions, sampling is carried out in several parts for several steel qualities, with particular regard to the fact that the sampling takes place in one shift at the same time, the cycles do not have to follow each other; they are assigned to the normal operation of the steelworks to provide a broad insight, and the selected portions cover the entire quality range.

The number of samples from the primary slag is the usual number of daily routine, and from the secondary slag, due to the wider tests, it is twice the usual number.

2.1. The slag inspection procedure

The slag samples were transported to the laboratory and prepared. The chemical composition of the primary slag is determined by XRF examination according to ÓAM technology procedures followed. Examination of the secondary slag samples in accordance with the objective is as follows:

- The approximate 500 g sample is divided into four parts.
- Three parts were placed in a desiccator filled with a CO_2 binder and stored.
- The chemical composition of one part is examined by XRF examination.

3. Results and Discussion

The compound analysis of the first five batches of the operational experiment was conducted as follows.

3.1. Examination of the residual reactivity of secondary slags

As part of our research objective, we analyzed five batches of primary and secondary slags. To determine the reactivity of the slag, we examined the chemical composition of the eluates, which can be judged based on the proportion of free CaO. The Analytical Laboratory of Furol Kft conducted the tests on the premises of ÓAM Kft.

The slag samples were broken and divided into sample parts in strict accordance with the regulations of the relevant standard. Eluates were prepared from both the primary and secondary slag samples using standard procedures involving distilled water and hydrochloric acid solutions. The complete elemental composition of the eluates was determined by ICP-OES instrumental analysis. The elements deemed crucial for the study were highlighted based on their concentration, and the relevant compound composition was determined through stoichiometric calculations. The final results are presented in *Table 1*.

Table 1

Eluate compositions of the slags of experimental portions %

	Sample No.		Al_2O_3	CaO	SiO_2	MgO	$B1 = \frac{CaO}{SiO_2}$	$B2 = CaO + \frac{CaO}{SiO_2}$
EAF	ED-77	Distilled water	0.15	1.58	0.0139	0.0001	X	X
	ED-78		0.05	1.31	0.0413	0.0001	X	X
	ED-79		0.02	1.11	0.1009	0.0001	X	X
	ED-80		0.01	0.76	0.1007	0.0001	X	X
	ED-81		0	0.9	0.1979	0.0001	X	X
	ES-77	Acid	5.67	22.6	7.54	5.77	3	3.76
	ES-78		9.27	26.2	6.78	5.8	3.87	4.73
	ES-79		7.31	27.7	9.38	6.48	2.96	3.65
	ES-80		7.02	30.9	11.04	6.1	2.8	3.35
	ES-81		7.5 1	28	10.8	6.65	2.59	3.21
LF	UD-77	Distilled water	0.64	4.91	0.0094	0.0001	X	X
	UD-78		0.65	4.83	0.0074	0.0001	X	X
	UD-79		0.48	4.09	0.0073	0.0001	X	X
	UD-80		0.33	3.96	0.0072	0.0001	X	X
	UD-81		0.74	5.36	0.0064	0.0001	X	X
	US-77	Acid	23.28	53.7	13.5	8	3.98	4.57
	US-78		18.11	52.2	14.12	6.22	3.7	4.14
	US-79		18.71	46.8	13.29	7.85	3.52	4.11
	US-80		16.08	55.7	13.31	6.77	4.18	4.69
	US-81		13.98	50.8	14.27	7.7	3.56	4.1

Table 1 contains the simple $B1 = (\frac{MgO}{SiO_2})$ and complex $B_2 = (CaO + \frac{MgO}{SiO_2})$ values calculated from the composition of the acid eluate for both primary and secondary slags.

Table 2 summarizes the parameters derived from the analysis results. It also contains the sulfur content of the steel samples taken according to the production technology and the analyzed slag sulfur content supplemented with the derived parameters.

Table 2

Derived data on desulphurization

Sample No.	Sulfur %					ΔS $(EAF - CC)$	Desulfurization efficiency	Sulfur distribution
	EAF	LF 1	LF 2	CC	Sec. Slag			
77	0.035	0.031	0.030	0.017	0.856	0.018	51.43%	24.45
78	0.034	0.030	0.025	0.021	0.961	0.013	38.24%	28.27
79	0.045	0.037	0.028	0.021	1.353	0.024	53.33%	30.07
80	0.044	0.037	0.031	0.020	1.896	0.024	54.55%	43.08
81	0.045	0.043	0.039	0.029	1.238	0.016	35.56%	27.50

The CaO values calculated from the Ca analysis appearing in *Table 1* are derived from the concentration of all dissolved Ca ions in the distilled water eluates, so the values also include the amount of Ca from the ion content of other dissolved Ca salts in addition to the free CaO content. In order to eliminate this disturbing effect, the free CaO content of the secondary slags is regulated by EN 459-2:2021/ Building lime – Part 2: Test methods, 4.7. was also determined according to the hydrochloric acid titration. The results are shown in *Table 3*, with the eluate test results repeated for comparability.

Table 3

Free CaO content of slags by hydrochloric acid titration

Sample No.	Free CaO %					ΔA*	ΔE**
	Measured	Average	Heated up	Average	Eluate		
77	1.34	1.35	1.62	1.44	4.91	0.09	3.47
	1.35		1.35				
	1.35		1.34				
78	3.22	3.4	1.86	1.78	4.83	–1.61	3.05
	3.76		1.88				
	3.21		1.61				
79	0.8	0.98	0.8	1.4	4.09	0.42	2.69
	1.07		2.06				
	1.07		1.33				
80	4.01	4.03	3.21	3.32	3.96	–0.71	0.64
	4.04		3.49				
	4.04		3.25				
81	1.61	1.97	1.87	1.8	5.36	–0.17	3.56
	2.15		2.17				
	2.16		1.36				

* (ΔA): The difference in the averages between heated-up and measured.

** (ΔE): The difference between the eluate and the average of heated-up.

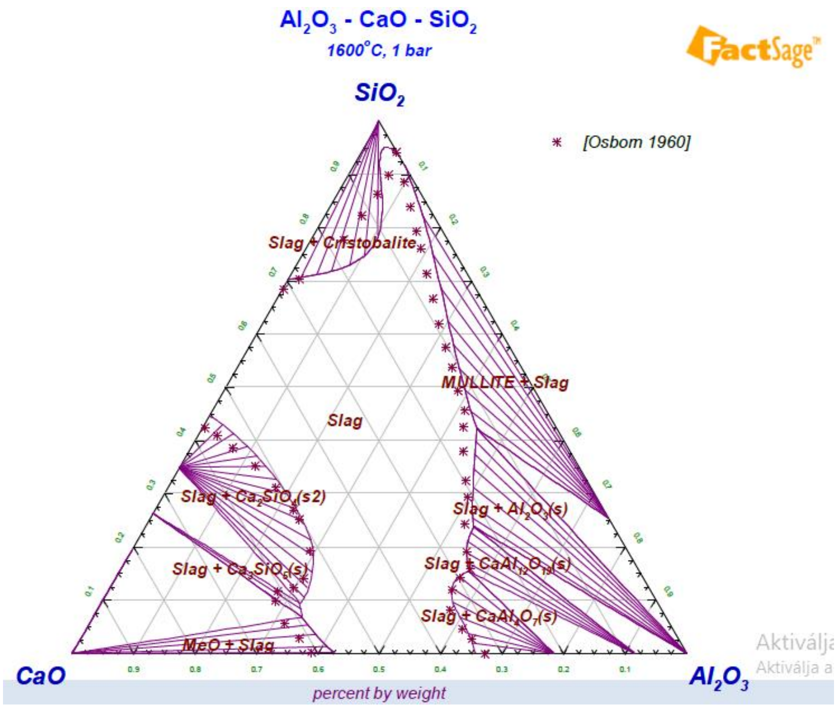
From the analytical results and the comparison of the results of both test methods, it can be concluded that the secondary slags contain free calcium oxide; their concentration varies between 1.35 and 5.36% in the comparison of the five slag samples.

3.2. Theoretical approximation of the active CaO content of slag melt

The data obtained from the analysis of solidified slags may not be sufficient to justify the intended application. However, given that a slag melt is produced during the primary process as part of the planned recycling, it is recommended to investigate the structure of these solid secondary slags in the slag melt systems formed within the temperature range typical of steel production. Examining phase diagrams of multi-component metal oxide melt is suitable for this purpose (gtt-technologies.de).

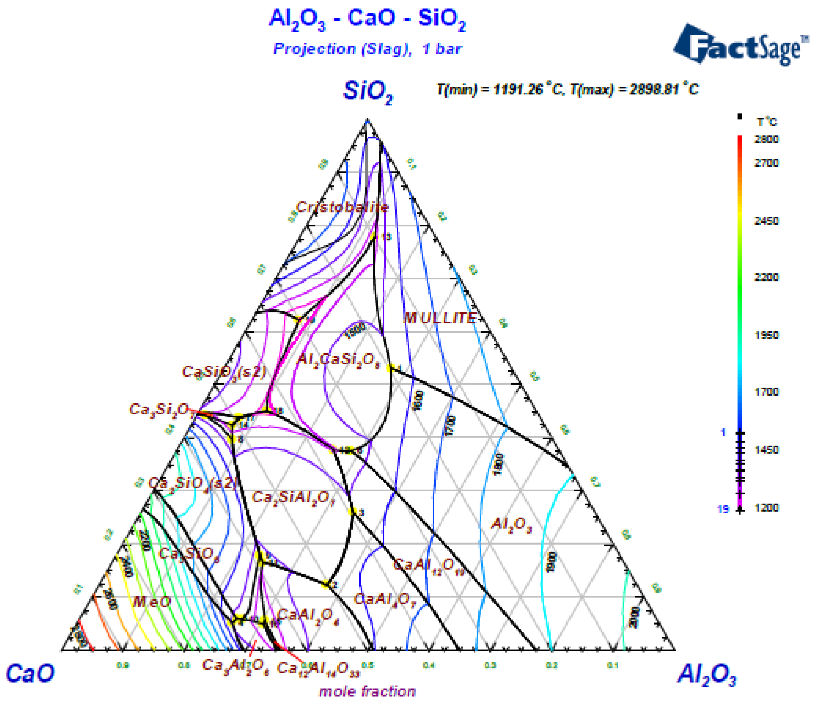
Steel production slags contain various metal oxides, which means we encounter oxide systems with highly diverse properties, considering individual oxides' tendency to form complex compounds. The phase diagrams examine the structure of slags in two-, three- and four-component systems. Among the three-component phase diagrams available, the secondary, deoxidized (desulfurization) slags are best characterized by the $CaO - SiO2 - Al2O3$ system, the phase diagram of which is shown in *Figures 1 (a) (b)*.

Figure 1(a) shows the same slag system, highlighting the critical factors for desulphurization and indicating the compositional range of melts in the liquid state at each temperature interval. Using these figures will bring an opportunity to examine the characteristics of the molten state of the examined secondary slags.



(a)

Figure 1. *Ternary phase diagram for* $SiO2 - Al2O3 - CaO$ (gtt-technologies.de)



(b)

Figure 1. *Ternary phase diagram for* $SiO2 - Al2O3 - CaO$ (gtt-technologies.de)

Figure 1(b) shows the same slag system, highlighting the critical factors for desulphurization and indicating the compositional range of melts in the liquid state at each temperature interval. This figure allows us to examine the obtained secondary slags' molten characteristics.

Since the charts show the conditions in a three-component system, we averaged the chemical composition of the five investigated plant slags, from which we allocated the slag composition to the three characteristic components shown in *Table 4*.

Table 4

Allocated composition of the five examined slags

Allocated composition %		
Al_2O_3	CaO	SiO_2
21.58	62.03	16.39

Figure 3 shows that the secondary slags are close to the ideal composition in the literature; they are located in the same melt temperature interval (gtt-technologies.de). Based on the stability codes that characterize complex compounds, the typical compounds are the $2C_aO \times SiO_2$ and $3CaO \times Al_2O_2$ complexes. Knowing the number of moles, the amount of CaO bound by the two complexing compounds can be determined, to which we must add the amount of CaO bound in the form of CaS, calculated on the basis of the decrease in sulfur content. The mole numbers that form the basis of the calculation are listed in *Table 5*.

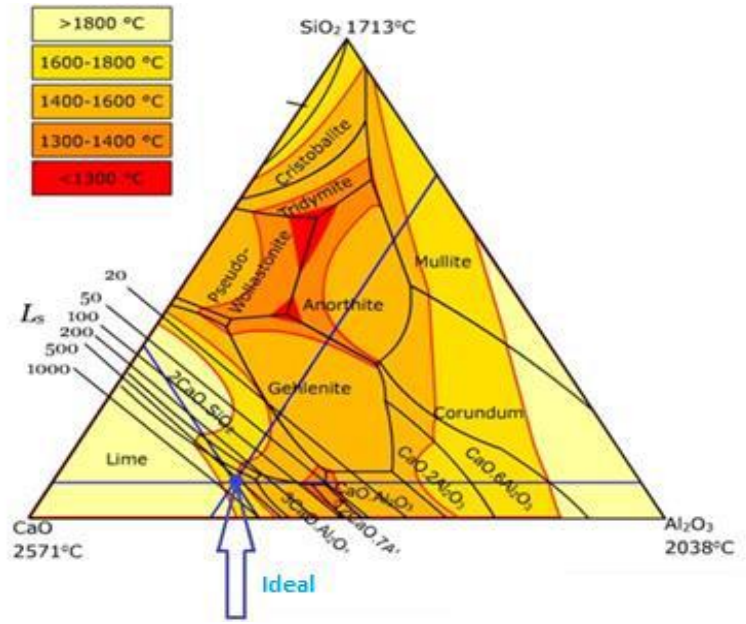


Figure 2. *Representing the converted composition according to Table 4 in the slag system, the conditions shown in Figure 3 are obtained* (gtt-technologies.de)

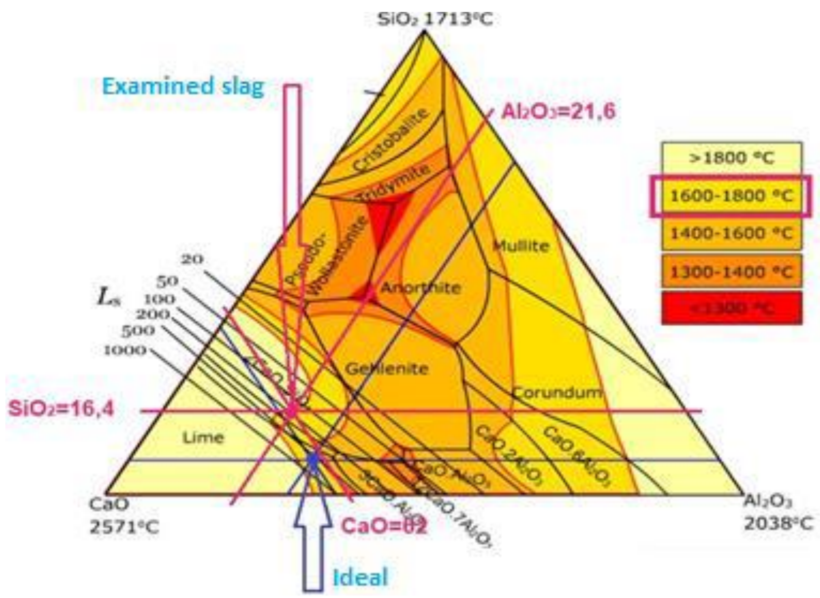


Figure 3. *Location of the examined secondary final slags a CaO-SiO2-Al2O3 in a ternary system* (gtt-technologies.de)

Table 5

Mole numbers of slag constituents

Sample No.	CaO		CaS		SiO_2		Al_2O_3	
	%	mol	%	mol	%	mol	%	mol
77	53.7	0.9588	0.86	0.0267	13.5	0.2251	23.28	0.2283
78	52.2	0.9324	0.96	0.03	14.12	0.2354	18.11	0.1775
79	46.8	0.8353	1.35	0.0423	13.29	0.2215	18.71	0.1834
80	55.7	0.9939	1.9	0.0592	13.31	0.2218	16.08	0.1577
81	50.8	0.9069	1.24	0.0387	14.27	1.2378	13.98	0.137

Calculations based on mole numbers:
Active CaO remaining after 2CaO × SiO2 formation:

$$M1CaO = 0.9855 - 2 \times 0.2251 - 0.0267 = 0.4819 \text{ converted: } 26.98\%$$

Active CaO remaining after $3CaO \times Al_2O_2$ formation:

$$M2CaO = 0.9855 - 3 \times 0.2283 - 0.0267 = 0.2472 \text{ converted: } 13.84\%$$

As can be seen from the analysis of the slag melts based on the phase diagram, the melt constituents influence each other, so their actual effect within the melt system differs from the analyzed concentration value, and as a result, the interaction is smaller than that. A diagram containing the activities of the three-component system provides an opportunity to determine the effect. The activity ternary diagram starts from the molar ratio of the constituents. Therefore, the molar ratios of the constituents of the slag system had to be determined from the available data, which is summarized in *Table 6*, with the average values in the bottom row. (Considering that the sum of the molar ratios follows from the interpretation, the number of moles of CaS was omitted from the calculation. The values corrected in this way differ only in the 2nd decimal place.)

Table 6

Molar ratios of slag constituents

All mole	Mole ratio		
	CaO	SiO_2	Al_2O_3
1.41	0.68	0.16	0.16
1.35	0.69	0.17	0.13
1.24	0.67	0.18	0.15
1.37	0.72	0.16	0.11
1.28	0.71	0.19	0.11
1.33	0.7	0.17	0.13

The activity ternary diagram is shown in *Figure 4*, marked with the molar ratios of the tested slags.

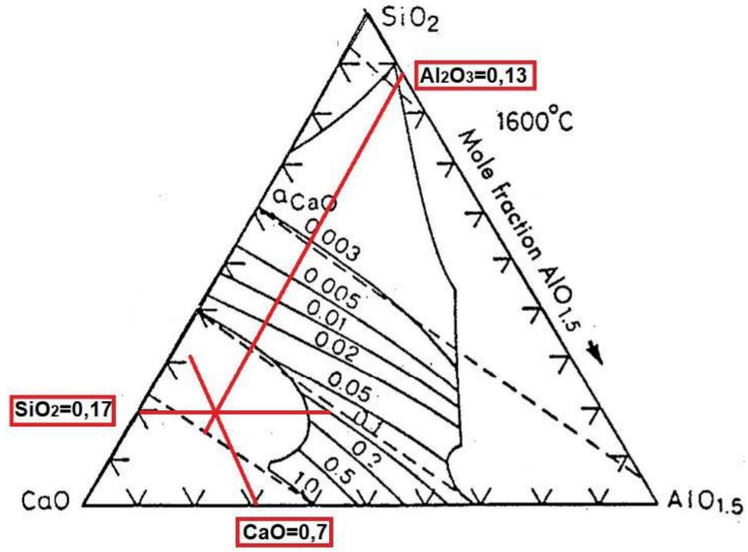


Figure 4. *CaO activity in the* $CaO - SiO2 - Al2O3$ *system* (gtt-technologies.de)

As it can be seen, the activity value appears between values 0.5...0.7. Deriving the active CaO concentration from this, the activity can be determined between 25.9 and 36.26% in the average of the five tested slags.

4. Conclusions

During the first phase of the research objective, operational experiments indicated that only a small amount of the CaO content in secondary steelmaking slags remains free. However, the concentration of "metallurgically free" CaO is at a level that makes utilizing these slags possible and even necessary. The results of utilization can be marked below:

- A portion of the extra slag can be utilized in the initial stage of steel production to substitute some of the consumed lime.
- Reduces the amount of landfilled secondary slag that would otherwise be unusable, thus reducing the ecological footprint of steel production.
- The substitution of quicklime means that it becomes necessary to purchase a smaller amount of quicklime, which results in cost savings and reduces the carbon dioxide emissions of lime works.

In summation, this study endeavors to make a meaningful contribution to the ongoing quest within the steel industry to bolster sustainability by recycling secondary metallurgical slag in the Electric Arc Furnace. Using steel production secondary slag within the technology reduces the greenhouse effect emissions of both the production plant and the lime supplier partner, in addition to cost reduction. Undertaking a thorough appraisal of the slag's composition and its behavior within the EAF context, we aspire to provide invaluable insights into the prospective benefits and challenges entailed in this recycling paradigm.

The research work confirms the strategic aspiration of the Max Aicher Group, including Ózdi Acélművek Kft., to consider the ecological aspects of service providers (purchasing green energy) and suppliers as part of its strategy in addition to its technology.

5. Acknowledgment and funding

The research is carried out in several stages, funded by the SteelTech-Center Hungary competence center, established as a collaboration between Ózdi Acélművek Kft. and the University of Miskolc.

Acknowledgements

This Research topic is supported by the SteelTech-Center Hungary project of University of Miskolc.

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