



Kinetics of electric arc furnace slag leaching in alkaline solutions



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HIGHLIGHTS

- The leaching of Al and Si from steel slag in alkaline solutions was investigated.
- Leaching is controlled by the bulk diffusion through the product layer crystals.
- C–A–S–H gel is reaction product of slag leaching in alkaline environment.
- Choice of alkali solution greatly influences the Si and Al dissolution.

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ABSTRACT

The dissolution of electric arc furnace steel slag (EAFSS) in highly alkaline MOH solution (M = alkali metal Na or K) has been investigated with emphases on the influence of the initial alkali concentration, solid to liquid (S/L) ratio and temperature on the kinetics of Si and Al dissolution from EAFSS. The shrinking core model (SCM) was used to analyze experimental data and the results have shown that the Si and Al dissolution from EAFSS was greatly influenced by MOH concentration, alkaline ion type and temperature. The dissolution process was controlled by the bulk diffusion through the product layer crystals. The activation energy for Si and Al dissolution was 55.27 kJ/mol and 48.05 kJ/mol in NaOH solution and 90.68 kJ/mol and 33.62 kJ/mol in KOH solution, respectively.

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1. Introduction

Metallurgical slag is a by-product from the metal processing industry. During the metal extraction and refining, liquid slag covers the molten metal preventing its oxidation and removing undesirable impurities. As 50 million tons of steel slag is produced per year worldwide [1], 12 million of which only in Europe [2], special attention is paid to its valorization. Usually steel slag has found an application in civil engineering but nevertheless considerable amounts of this waste material are disposed of at landfills. Electric arc furnace steel slag (EAFSS) is a by-product from iron and steel scrap conversion into the steel in electric arc furnaces. In this process, the scrap is melted using heat generated by an arc, created by

an electric current. An important part of the steelmaking process is the addition of limestone, in order to form the slag which contains oxides of calcium, magnesium, silicon, iron, and aluminum. The molten slag floats on top of the molten bath of steel, before it is removed from the furnace and allows to solidify. This process generates anywhere between 10% [3] and 15–20% of slag per ton of steel [4]. The chemical composition of EAFSS is highly variable and depends significantly on the compositions of scrap charged into the furnaces as well as the processing conditions. Since the feed into the furnaces varies, the main chemical constituents of EAFSS can vary widely. Typically, The EAFSS comprises of 10–40% FeO, 22–60% CaO, 6–34% SiO₂, 3–14% Al₂O₃, and 3–13% MgO [5]. Mineralogical composition of EAFSS also varies. Larnite (also known as belite, β -Ca₂SiO₄) and wustite (FeO) are the main crystalline phases observed in EAFSS but different calcium aluminosilicates

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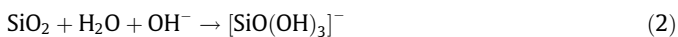
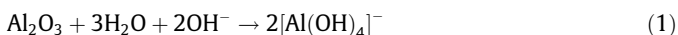
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silicates such as brownmillerite ($\text{Ca}_2(\text{Al,Fe})_2\text{O}_5$), gehlenite ($\text{Ca}_2\text{Al}(\text{AlSiO}_7)$) and mayenite ($\text{Ca}_{12}\text{Al}_{14}\text{O}_{33}$) are also observed [6–9].

Today, steel slag is used for a different application. EAFSS is considered as a raw material for the production of typical Portland cement clinkers [7] and low energy belite (C_2S) cements [9]. However, comprehensive research, with respect to the possibility of EAFSS exploitation as recycled aggregates in concrete, was carried out. The results have shown that steel slag aggregate concrete exhibits the same or even improved mechanical properties, in comparison to a natural aggregate concrete [10–12]. This fact aside, the application of steel slags in construction has some limitations with respect to their possible volume instability due to the presence of CaO and MgO, which are sensitive to volume expansion [13,14]. Moreover, EAFSS is also considered as a potential adsorbent for phosphorous and heavy metal removal from waste waters [15,16] and as a neutralizing agent in bioleaching operations [17].

In the past decade, special attention was paid to the use of alkali activated steel slag (inorganic polymer) as a replacement material for cement binders. Blast furnace slag was used mainly as a source material for the alkali activation process, but a recent shift was observed towards the steel slag valorization by the alkali activation process [18,19]. Due to fast-growing environmental concerns of the cement industry, like its carbon footprint, the alkaline activation of various by-products is considered as an alternative cement technology.

Alkali-activated binders are produced by mixing aluminosilicate or calcium aluminosilicate powders with an alkaline activator (mainly highly alkali or alkali silicate solutions) which yields materials with properties similar or superior to those reported for Portland cement. Alkaline activation of slag is a complex process involving dissolution of raw materials and the destruction of chemical bonds Ca–O, Mg–O, Si–O–Si, Al–O–Al and Al–O–Si, [20] as well as the formation of unstable structural units which accumulate during the induction period. Under highly alkaline conditions, Al exists in the form of $[\text{Al}(\text{OH})_4]^-$ while Si predominately exists in the form of $[\text{SiO}(\text{OH})_3]^-$ and/or $[\text{SiO}_2(\text{OH})_2]^{2-}$ depending on the pH of alkaline solution [21–23]. Formation of these structural units can be presented by the following reaction:



The next step is the condensation of these Al and Si structural units and the formation of aluminosilicate oligomers which react with Ca and develop a poorly ordered structure which exhibits high mechanical strength [21,24]. It is reported that calcium (aluminosilicate) hydrate C–(A)–S–H gel is the main reaction product of the alkali activation of slag [25] which is similar to the calcium silicate hydrate (C–S–H) gel, the reaction product of the cement hydration process. Numerous investigations have been performed in the field of alkali activation of metallurgical slag, but only a few of them have reported on slag dissolution in alkali environment [26] in terms of its use in the production of alkali activated binder.

The purpose of this research was to obtain essential information on the kinetics of dissolution of EAFSS in a highly alkaline MOH solution. Attention was paid to the dissolution of Al and Si from the EAFSS in NaOH and KOH solutions depending on the various leaching factors. The shrinking-core model was used to express the dissolution kinetics of Si and Al from EAFSS. This may provide relevant theoretical guidance for understanding the leaching process of EAFSS in a different alkaline solution, which is of essential importance for the synthesis of alkali activated slag binder.

2. Experimental

Powdered EAFSS was supplied from the Steel Mill Nikšić in Montenegro and its chemical composition is given in Table 1. The slag was grounded down and the mean diameter particle size was $d_{50} = 24 \mu\text{m}$. Leaching of EAFSS was carried out by mixing the slag with NaOH and KOH solutions with concentrations of 5, 7 and 10 M, in S/L ratios of 0.05, 0.1 and 0.3 g/ml for 120 min. The dissolution process was carried out in a 500 ml glass reactor and the slurry was agitated by a magnetic stirrer with a constant stirring speed of 200 rpm. The leach slurry was extracted at regular intervals and filtered. Leachates were analyzed for Si and Al concentration by inductively coupled plasma emission spectroscopy (ICP-OES).

The calculation of Si and Al extraction was performed with respect to their amounts in the leachate. In order to investigate the influence of temperature on dissolution kinetics, experiments were carried at 25, 40, 50 and 60 °C at a constant stirring speed, S/L ratio and MOH concentration. For this purpose, the alkali solutions were poured into the reactor, heated to the desired temperatures at which the slag was added. To prevent liquid loss by evaporation, the reactor was fitted with a reflux condenser.

Scanning electron microscopy (SEM) analysis of EAFSS before and after the dissolution in MOH (M = Na or K) solution was examined using the FEI Strata 235 DB focused ion beam system, equipped with an EDAX (Energy-dispersive X-ray) unit.

The phase analysis of EAFSS and leaching solid residues was performed by X-ray powder diffraction (XRPD) technique with a PHILIPS PW 1710 diffractometer using monochromatised $\text{CuK}\alpha$ radiation ($\lambda = 1.54178 \text{ \AA}$) and the step-scan mode (2θ range was $4\text{--}90^\circ$ 2θ , step 0.02° 2θ , time 0.8 s).

3. Results and discussion

3.1. Effect of alkali hydroxide concentration and alkali ion

The effect of initial alkali concentrations and alkali ion type on the dissolution of Si and Al from the EAFSS was investigated for MOH concentrations of 5, 7 and 10 M and the results are given in Fig. 1(a) and (b). The results have shown that initial concentrations of alkali solutions greatly influenced the efficiency of EAFSS dissolution and that the highest release of Si and Al was observed in the first 15 min of the slag dissolution process. After this time, slag dissolution increase was less rapid. The Si and Al leaching from EAFSS increased as the MOH concentration was raised to the range of 5–10 M. As the concentration of MOH increased, the increasing OH^- concentration led to the faster breaking of a greater amount of Al–O and Si–O bonds, which accelerated the dissolution of Al and Si. The higher quantity of Al was leached out in comparison to the Si under the same conditions because Al can be easier to dissolve due to weaker Al–O bonds, compared to the Si–O bonds [27]. The highest efficiency of Si and Al dissolution was obtained using 10 M MOH. These elements are slightly released in alkali environment, which is in agreement with a previous study [28]. In this work, the release of Si was in the range of 1.1–7.78% while the Al release was in the range of 3.03–18.73%, at ambient temperature after 120 min.

The rate-limiting step in Si dissolution in alkali solutions is thought to be the breaking of siloxane bonds at the surface which proceeds via a nucleophilic attack of H_2O or OH^- and forming silanol groups which may exist as $\equiv\text{Si}-\text{OH}_2^+$, $\equiv\text{SiOH}$ and $\equiv\text{SiO}^-$, where \equiv denotes a surface functional group [29,30]. Dissolution rate of Si is greatly dependent on pH value and can be explained

Table 1
Chemical composition of EAFSS.

Component	%
CaO	46.5
FeO	23.5
SiO ₂	12.2
Fe ₂ O ₃	0.9
MgO	6.5
MnO	1.3
Cr ₂ O ₃	0.8
Al ₂ O ₃	7.24
TiO ₂	1.06

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