



ASSESSMENT OF BLAST FURNACE SLAG TRANSFORMATION INTO VALUE ADDED BY-PRODUCTS ON BASIS ON KNOWLEDGE OF SLAG CHARACTERISTICS

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ABSTRACT

Slags are the main by-products generated during iron and crude steel production and the steel industry is committed to increasing and improving their recycling. Today the quantity of slag produced and its utilization are one of the important indicators of sustainable steel industry. There are different forms of BF slag by-products depending on the method used to cool the molten slag. The by-products can be an excellent source of constructional materials, finding applications in road building, concrete aggregate, as thermal insulation (mineral wool), and as a clinker substitute in cement production, biological filter media, glass making etc. In addition to products obtained from fresh slag, the possibility to utilize the old and the stockpiled blast furnace slags was studied. A key role in their utilization plays the knowledge of the chemical, mineralogical, and morphological properties of BF slag. This paper summarizes the characteristics of BF slag and its possible application.

KEYWORDS: blast furnace slag, by-products, morphology, mineralogy, chemical and physical properties

1. Introduction

The target of the current metallurgical industry is to recycle and utilize all their by-products, so as to close the sustainable production loop. Wastes, particularly solid wastes generated unavoidably, should be converted into useful, value added by-products.

Slags are the main waste generated during iron and crude steel production and the steel industry is committed to increasing and improving their recycling. These are generated in a parallel route of the main processes of hot metal production in ironmaking and steelmaking and therefore the slag generation process is considered as a part of the whole steel production process [1].

Today the quantity of slag produced and its utilization are one of the important indicators of sustainable steel industry. An average of about 400 kg of solid by-products is generated in the steel industry per tone of crude steel, out of which (70-80%) consists of blast furnace slag and basic oxygen furnace slag. Due to the large slag quantities and the stricter environmental regulations, recycling and

utilization of slag are an attractive alternative in order to reduce and eventually to eliminate the disposal cost, to minimize the related environmental pollution, and to save the resource conservation.

The sustainable use of slag contributes to saving natural resources, to CO₂ emissions reduction, to energy consumption reduction, to the formation of a society founded on the recycling practice (as landfilling is avoided) and to the promotion of the iron and steel industry sustainability. Therefore potential economic and environmental benefits make slag by-products that can be further recovered and used. For all these reasons, the effective utilisation of slag and its turning into high value added products improves the competitiveness of the iron and steel industry [2].

Based on the information in literature, this paper summarizes the characteristics of BF slag and its possible application.

Its generation process is correlated with the environmental impact. In respect to slag characteristics, the possibility to transform blast furnace slag into value added by-products for various domains is evaluated.

2. Blast furnace slag, environmental and economic considerations

A blast furnace is a closed system into which iron-bearing materials (iron ore lump, sinter and/or pellets), additives (slag formers such as limestone) and reducing agents (i.e. coke) are continuously fed from the top of the furnace shaft through a charging system that prevents escape of blast furnace gas (BF gas). Iron ore processed nowadays contains a large content of hematite (Fe_2O_3) and sometimes small amounts of magnetite (Fe_3O_4).

In the blast furnace, these components become increasingly reduced, producing iron oxide (FeO) then a partially reduced and carburised form of solid iron. Finally, the iron charge melts, the reactions are completed and liquid hot metal and slag are collected at the bottom. The reducing carbons react to form CO and CO_2 . Fluxes and additives are added to lower the melting point of the gangue, improve sulphur uptake by slag, provide the required liquid hot metal quality and allow for further processing of the slag.

The slag floats on the surface of the molten iron and is subsequently drawn off and allowed to cool to produce a semi-dense porous crystalline material (light weight aggregate) known as air cooled blast furnace slag. From an environmental point of view, the blast furnace has a significant role in the ore based steel production. The manufacture of iron by chemical reduction of iron ores by coke (carbon) in a blast furnace is accompanied by production of large waste quantities, the most important of which the slag. About 70 years ago, the slag generation in the blast furnace was 980kg/t.

Recently, due to a better understanding of the slag formation mechanisms and of the overall BF process, it is also currently possible to control, optimize and minimize slag production. The specific quantity of slag generally lies in the range of 175 – 350 kg/t hot metal produced [3, 4].

The slag amount depends very much on the charging material: for example, the grade of iron ore, its sources are the gangue content of iron ore and lime content added to adjust the chemical composition of molten slag. The slag quantity has decreased today by using enriched iron ore and coke with low ash content. From the viewpoint of preservation and protection of the global environment, the main issue concerning the use of ferrous slag is the question whether it is a waste or a by-product. In order to market them the better way is to consider them by-products because the term "waste" indicates a material to be deposited instead of to be used [2, 5].

The driving force for the valorization of the slag is the stringent legislation for environmental protection concerning the use of slag that includes other laws, as follows: the Kyoto protocol, the Reference Document of Best Available Techniques, Harmonization Committees TC 351 Dangerous Substances and TC 154 Aggregates, the REACH directive. The Waste Framework Directive (WFD) (2006/12/EC) is the most important document governing the use of slag. [2].

On the other hand, the recycling and utilization of the slag is an attractive alternative in order to reduce and eventually to eliminate the disposal cost, to minimize the related environmental pollution, and to save the material resources and energy. Slag recycling has been successful in different fields of application, in a variety of industries, starting with ironmaking and steelmaking. Ironmaking slag is used in different ways with high added value.

2.1. Utilization of BF slag

Different forms of slag product are produced depending on the method used to cool the molten slag. These products include air-cooled blast furnace slag (ACBFS), expanded or foamed slag, pelletized slag, and granulated blast furnace slag (GBFS), Figure 1.

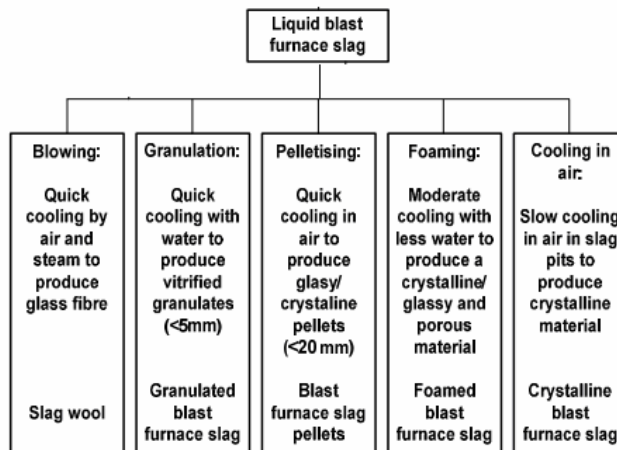


Fig. 1. Methods to processing the liquid BF slag

The utilization of ironmaking slag has a long history. The use of by-products from steel industry goes back to many centuries ago. The first appearance of slag was recorded as early as the year 700 B.C. In 350 BC Aristotle already stated "When iron is purified by fire, there forms a stone known as iron slag. It is wonderfully effective in drying out wounds and results in other benefits". The history of slag use in road building dates back to the time of Roman empire, some 2000 years ago, when broken slag from the crude iron-making forges of that area were used in base construction. The cast iron slag stones were used for masonry work in Europe of the 18th century. As early as 1589, the Germans were making cannon balls cast from iron slag [4]. In later centuries slag has been used as construction material.

The discovery of the hydraulic properties of granulated BF slag gave birth to a new era in slag exploitation: slag has been used as binding agent and/or addition for concrete. The first modern roads in the building of which slag was utilized were built

in England, in 1813 [8], and after that the use of slag spread fast to the American continent as well. The use of slag in road building was recorded there for the first time in 1830. Granulated blast furnace slag was first developed in Germany in 1853. Ground slag has been used as a cementitious material in concrete since the beginning of the 1900s.

The traditional use of slag as landfill material, after the increase of steel production since the mid-1970's, has reached its limit and the pressure for natural resources and energy saving have driven steel industry to increase the recycling of this material, by facing other important challenges (such as technologies development, production facilities maintenance and ferrous slag products certification) in order to improve their application in different sectors [2, 7].

To transform blast furnace slag into value added by-products some measures must be applied in accordance with its chemical characteristics, Figure 2.

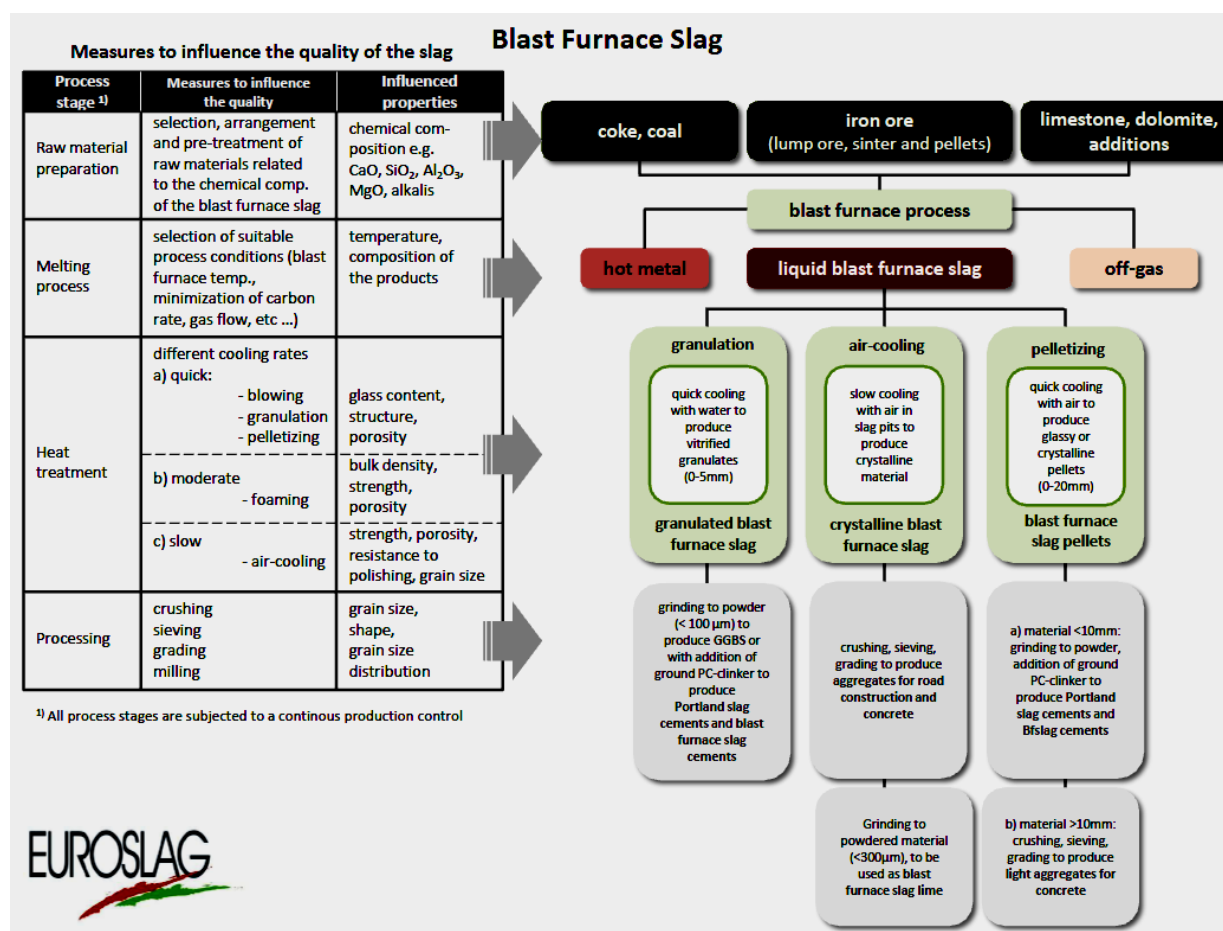


Fig. 2. Measures to influence the quality of the slag



2.2. Characteristics of the BF slag in accordance with its utilization domains

Blast furnace slag can be an excellent source of construction materials, finding applications in road building, concrete aggregate, as thermal insulation (mineral wool), and as a clinker substitute in cement production, biological filter media, glass making etc. In addition to fresh slag based products, has developed the possibility to utilise old, stockpiled blast furnace slags. Such can be re-utilised the several million tones of old slags, previously considered unusable. Availability and areas of applications of BF slag are:

- *environmental products*: production of Portland slag cement using BF slag; production of roadmaking aggregate and soil conditioner using BF slag and BOF slag; use of BOF slag as rail ballast; use of fly ash as partial replacement of cement, in manufacture of cellular concrete, bricks and sintered light-weight aggregate, and in road construction.

- *altogether new products*: glass-forming material; prime western grade zinc; high-purity zinc oxide crystal; glass-ceramic family of materials; Portland cement; iron oxide powder for use in manufacture of ferrites, pigments, etc.; ceramic floor and wall tiles; synthetic granite tiles; wear-resistant ceramic products; heat and sound insulation sandwich panels; natural organic fiber reinforced door panel; sisal fiber cement corrugated roofing sheet.

Knowledge of the chemical, mineralogical, and morphological properties of BF slag is essential because these play a key role in their utilization. The chemical, mineralogical, and morphological characteristics of BF slag are determined by the processes that generate this material. The slag

formation is the result of a complex series of physical and chemical reactions between iron bearing raw materials (iron ores, concentrates, sinter, and pellets), the non-metallic charge (limestone), the energy sources (coke, injected coal, etc.), refractory lining etc. at temperature range from 1450 to 1550°C. Also the chemical composition of the slag depends on the feeding raw materials, and the cooling rate. The amount of slag produced depends upon the gangue content of the blast furnace, the ferrous burden, the coke ash and ash from the injection material, e.g. coal, and the amount of flux required to achieve the necessary hot metal quality. In order for blast furnace slag to be acceptable for use outside the iron and steel works, it is necessary to take additional measures to ensure that the slag chemistry and/or physical properties are requisite.

BF slag is made of silicates and aluminosilicates of calcium and magnesium together with other compounds of sulphur, iron, manganese and other trace elements. Their iron content is usually below 0.5% since they result from a reduction process [8-10]. The four major constituents of blast-furnace slag - silica, alumina, lime and magnesia - constitute about 95% of the total composition. In the operation of a blast-furnace the composition of the slag must be closely controlled so that the amount of silica plus alumina balances (quantity wise) with the total of lime and magnesia to produce a composition of slag which will readily melt and flow from the furnace at the temperature of molten iron.

These limitations on the chemistry of blast-furnace slag to permit efficient furnace operation provide quite a narrow band in the chemical composition of all blast furnace slag, Table 1.

Table 1. Typical composition of blast furnace slag

Constituent	Percent							
	1949 ^a		1957 ^a		1968 ^a		1985 ^a	
	Mean	Range	Mean	Range	Mean	Range	Mean	Range
Calcium Oxide (CaO)	41	34-48	41	31-47	39	32-44	39	34-43
Silicon Dioxide (SiO ₂)	36	31-45	36	31-44	36	32-40	36	27-38
Aluminum Oxide (Al ₂ O ₃)	13	10-17	13	8-18	12	8-20	10	7-12
Magnesium Oxide (MgO)	7	1-15	7	2-16	11	2-19	12	7-15
Iron (FeO or Fe ₂ O ₃)	0.5	0.1-1.0	0.5	0.2-0.9	0.4	0.2-0.9	0.5	0.2-1.6
Manganese Oxide (MnO)	0.8	0.1-1.4	0.8	0.2-2.3	0.5	0.2-2.0	0.44	0.15-0.76
Sulfur (S)	1.5	0.9-2.3	1.6	0.7-2.3	1.4	0.6-2.3	1.4	1.0-1.9

a. Data source is the National Slag Association data: 1949 (22 sources); 1957 (29 sources); 1968 (30 sources) and 1985 (18 sources).

There may be a limited variation between the silica plus alumina and lime plus magnesia constituents, however, the ratio of the composition fluctuates only a small amount. The chemistry usually has a narrow ratio of basic to acid components. The most BF slags are characterized by CaO:SiO₂ ratio of 1.0-1.3. Calcium and magnesium oxides content, reported as silica and alumina, normally results in a

0.85 to 1.20 basicity ratio. The pig iron for the steelmaking purposes has 1.1-1.3 CaO:SiO₂ ratio and (CaO+MgO):(SiO₂+Al₂O₃)=1-1.2. For the high Al₂O₃ content (SiO₂:Al₂O₃<2.5) the (CaO+MgO):(SiO₂+Al₂O₃) ratio decreases to 0.9.

The presented data suggest that these have remained relatively consistent for all blast furnaces and over the years [6].

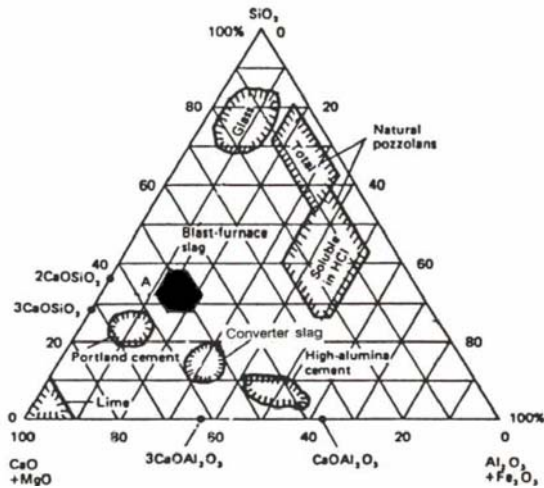


Fig. 3. Position of the BF slag on thermal diagram with respect to its chemical composition and its utilization possibility [11]

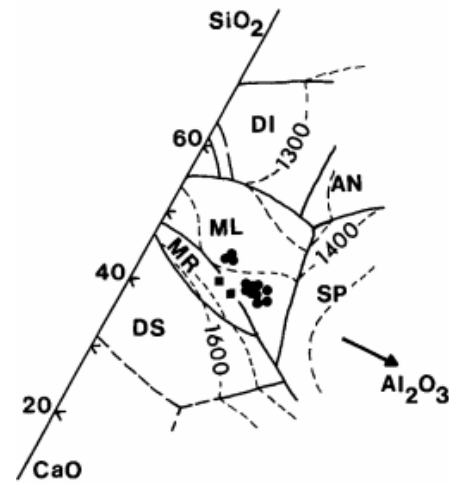


Fig. 4. Section through the CaO-SiO₂-Al₂O₃-MgO system at 10 % MgO with chemical data of slags plotted for some French slags (squares) and Scunthorpe and Redcar slags (circles): DI: diopside. AN: anorthite. ML: melilite. MR: merwinite. SP: spinel. DS: dicalcium silicate. Dashed lines are isotherms in °C [13].

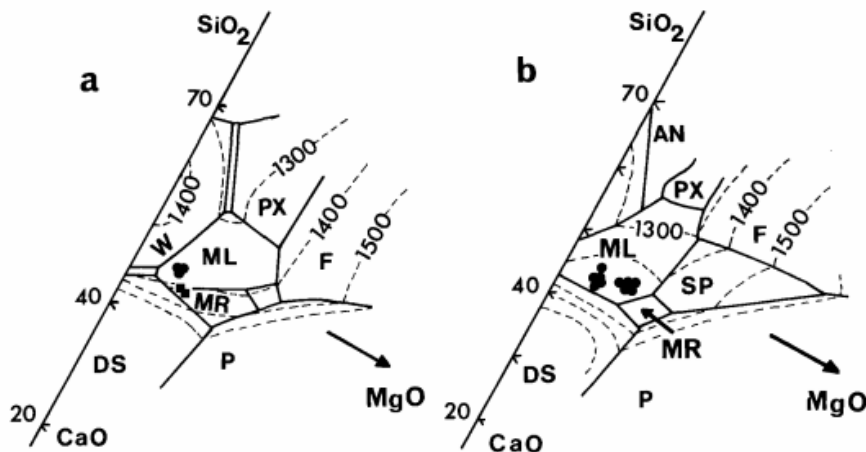


Fig. 5. Sections through the CaO-SiO₂-Al₂O₃-MgO system at 10 % Al₂O₃ (a) and 15 % Al₂O₃ (b): PX: pyroxene. F: forsterite. W: wollastonite. ML: melilite. MR: merwinite. DS: dicalcium silicate. P: periclas. AN: anorthite. SP: spinel. Chemical data for low Al (<11 % Al₂O₃) Scunthorpe (circles) and French slags (squares) plotted on a. Scunthorpe and Redcar slags with >11 % Al₂O₃ a plotted on b. [13].

The potential mineralogical composition of the BF slag is established from the chemical composition based on the phase diagram of the CaO-SiO₂-Al₂O₃-MgO quaternary system.

The place of these usual metallurgical slags on SiO₂-Al₂O₃-MgO-CaO quaternary system is delimited by the Al₂O₃ content that is lower than 20%. Also the MgO content that not higher of 20% is used to established this location. Thus established the domain of BF slag is given in Figure 3 which presents the compositional zone for the main products in which it is used. The quaternary system of SiO₂, Al₂O₃, MgO, CaO oxides for the 10% MgO plane is given in

Figure 4 and for the 10% and 15% Al₂O₃ planes in Figure 5 [12, 13].

The absence of anyone quaternary compound is certainly in this quaternary system. The oxides, binary and ternary compounds, binary, ternary and quaternary solutions were identified.

The mineralogical compounds from quaternary system CaO-SiO₂-Al₂O₃-MgO that can be found after crystallization from acid and basic blast-furnace slag are: pseudo-wollastonite (CS), rankinite (C₃S₂), dicalcium silicate (C₂S), melilite, merwinite (C₃MS₂), monticellite (CMS), diopside (CMS₂), anorthite (CAS₂), Figure 6.

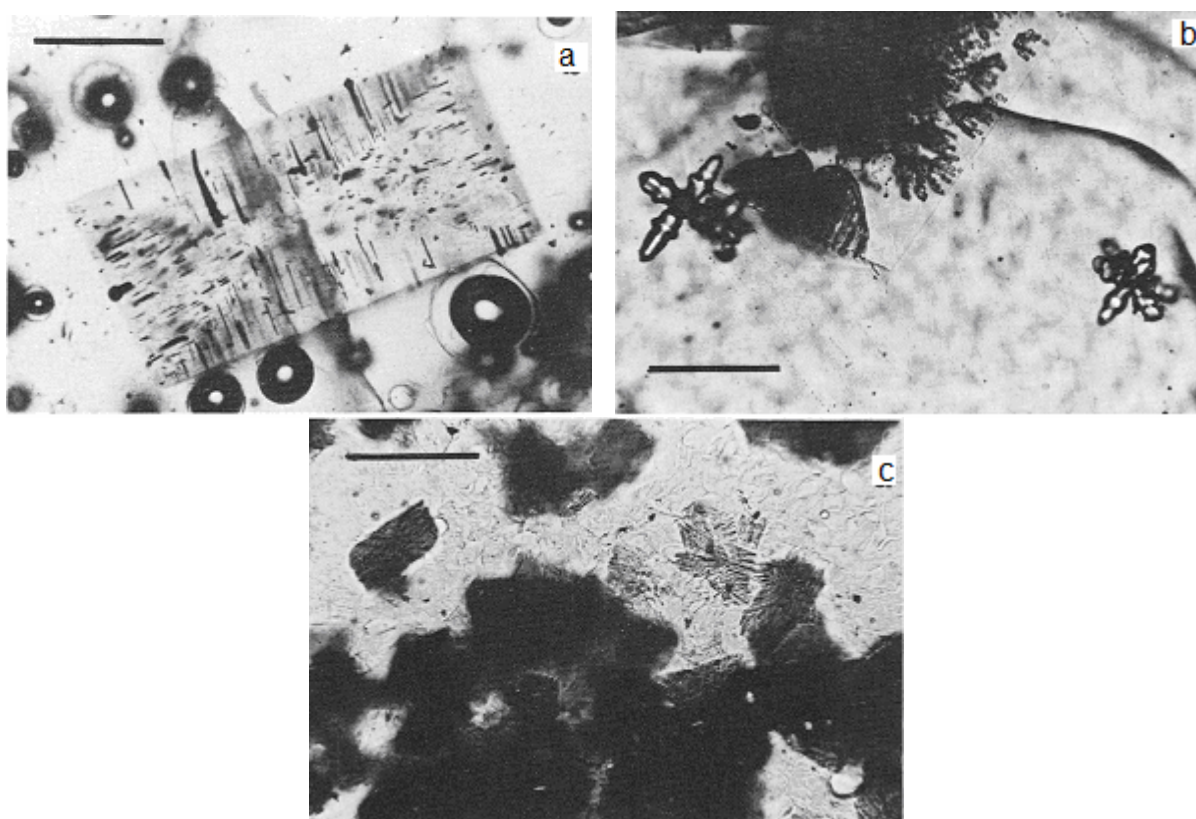


Fig. 6. Micrographs of granulated slag: (a) Sector-zoned euhedral melilite with orientated inclusions of iron. Some of the iron extends beyond the crystal forming a spherical globule attached to the surface. Sample 3. Plane polarized light. Scale bar 0.10 mm. (b) Stellate oldhamite crystals and dendritic growth of oldhamite. (c) Cluster of merwinites within glass [13]

When molten blast-furnace slag is rapidly chilled and quenched in water as granulated slag, the solid phase is amorphous, predominantly a granular and cellular glass. When such molten slag is more slowly cooled, crystallization takes place while the slag is cooling. The air-cooled BF slag is a mixture (crystalline and amorphous). The crystal phase predominate in air-cooled blast furnace slag is melilite, a name applied to any series of solid solutions extending from akermanite (2CaO·MgO·2SiO₂) to gehlenite (2CaO·Al₂O₃·SiO₂).

Also spinel is present. The compositions of spinel (MgO·Al₂O₃), merwinite (3CaO·MgO·2SiO₂) and periclase are close to their stoichiometry. In addition, in certain conditions other minerals found to be present in blast furnace are pseudowollastonite, monocalsiumsilicate, olivine, pyroxene, merwinite, calcium sulfide, ferrous sulfide and manganous sulfide [13, 15].

Figure 7 shows well quenched liquid slag in equilibrium with melilite (Figure 7a) and spinel (Figure 7b) respectively.

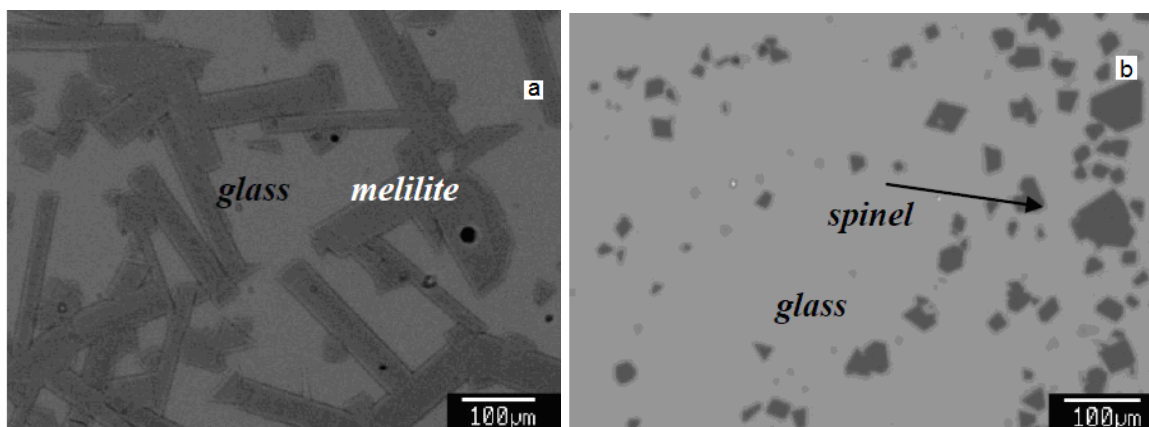


Fig. 7. Typical microstructures of quenched slags from a) melilite and b) spinel primary phase fields in air [15]

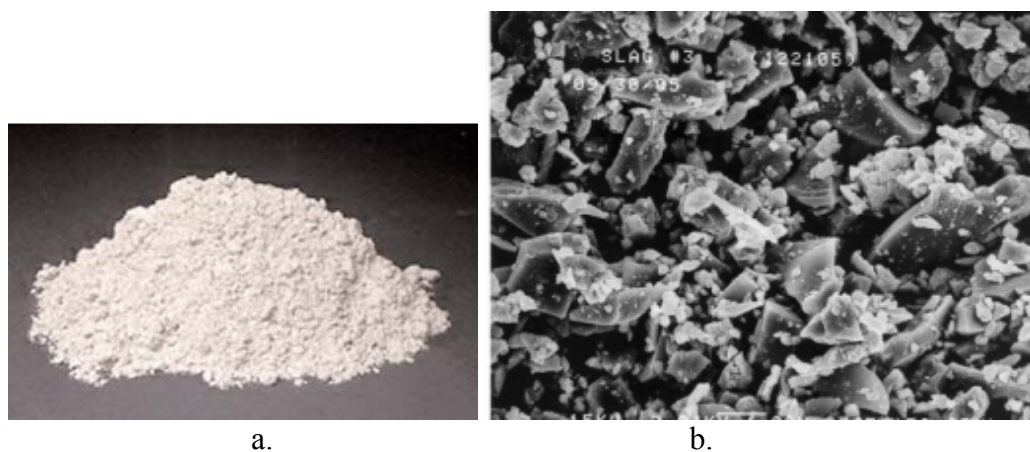


Fig. 8. Ground granulated blast-furnace slag (a) and its SEM micrograph (b) [16]

The physical appearance shows that the blast furnace slag is a dull white color, and it has rough and angular-shaped particles, Figure 8.

When BF slag is crushed and screened it produces an aggregate with a rough surface texture and relatively high porosity. Crushed air-cooled BF Slag (ACBFS) is angular, roughly cubical, and has textures ranging from rough, vesicular (porous) surfaces to glassy (smooth) surfaces with conchoidal fractures. The 1-5 mm granulate particles are very angular with a vitreous appearance. Their colour varies from pale honeybrown to grey-brown and rarely almost black. This reflects the chemical variability, the darker slags being richer in Mn (0.98-1.65 %). Small vesicles (<1 mm) are common in the larger pieces. Occasional vitreous fibers occur. The internal structure of the granulate particles has many cracks visible in thin section, some of which are perlitic. There are abundant vesicles varying considerably in size and quantity within a single grain. These often aggregate to form linear, curved or irregular clusters of single sized vesicles. Overall,

glass dominates. In every sample it is > 80% by volume, but usually > 95% and often > 99% of the granulate. Individual granules, however, sometimes contain up to 30 % crystals [13].

The mineralogy of the solid state determines the material properties such as the durability, the solubility and the reactivity. Generally, an amorphous material is less soluble than a crystallized phase with a similar chemical composition, due to the very slow kinetics for a re-crystallization and reactions with the surrounding liquid media [17]. However, rapid cooling (quenching) and enhancing the amount of amorphous material is also a process that increases the reactivity of a material with pozzolanic properties, such as ironmaking slag [18-20]. Rapid cooling is also a means of reducing the content of unstable silicates, as well as free CaO and periclase, MgO that expand at phase transformation [21].

From the point of view of the hydraulic power of the blast-furnace slag, this depends on the one hand on their chemical composition, and on their microstructure, on the other. Most of the authors



estimate that the vitreous structure is the controlling condition because the vitreous state makes possible and explains the reactions which would not be possible from crystallized material. Blast-furnace slags entirely crystallized of normal composition do not have hydraulic property or only a very limited one. Other estimates that "vitreous-crystalline" slag has higher potential hydraulic properties than slag vitrified up to 100%. Also the size and the distribution of crystals in the vitrified blast furnace slag have an effect upon hydraulic power. In conclusion, for an identical chemical composition a perfect vitrification is not the criterion of a highest reactivity. The increase of Al₂O₃ rate in a quaternary slag glass gives an improvement of hydraulic properties. Particularly the increase of Al₂O₃ rate in a quaternary slag glass gives an improvement of hydraulic properties [22-25].

Blast furnace slag is mildly alkaline and exhibits a pH in solution in the range of 8 to 10. Although blast furnace slag contains a small component of elemental sulfur (1 to 2 percent), the leachate tends to be slightly alkaline and does not present a corrosion risk to steel in pilings or to steel embedded in concrete made with blast furnace slag cement or aggregates. Also as a matter of fact, slag is frequently used to eliminate acid conditions in industrial operations as well as being used to neutralize acid soil conditions in agriculture [14].

Relatively low bulk density of BF slag is typically 1300 kg/m³ for air-cooled and stable with no expansion tendencies. Instability problems with blast furnace slag are relatively rare. Fresh-make (i.e. new) blast furnace slag should in fact have no instability problems. However, some older sources of blast furnace slag, after a period of weathering, may contain pockets of unstable material. The most likely form of instability in this type of slag is when, as a

result of weathering, a significant proportion of the sulphur oxidises to sulphate, often present as gypsum. This, under given conditions, may take part in chemical reactions resulting in the formation of a sulphoaluminate hydrate phase, apparently similar to that taking place in sulphate attack on concrete. The result is volumetric expansion and disruption of the mass [14].

In certain situations, the leachate from blast furnace slag may be discolored (characteristic yellow/green color) and have a sulfurous odor. These properties appear to be associated with the presence of stagnant or slow moving water that has come in contact with the slag. The stagnant water generally exhibits high concentrations of calcium and sulfide, with a pH as high as 12.5 [26]. When this yellow leachate is exposed to oxygen, the sulfides present react with oxygen to precipitate white/yellow elemental sulfur and produce calcium thiosulfate, which is a clear solution. Aging of blast furnace slag can delay the formation of yellow leachate in poor drainage conditions but does not appear to be a preventative measure, since the discolored leachate can still form if stagnant water is left in contact with the slag for an extended period [27-30].

The physical properties of blast furnace slag have a considerable variability depending on the iron production process. Table 2 compares key the physical properties of air-cooled blast furnace slag and a common naturale aggregate [8]. Of all the slag types generated, air-cooled blast furnace slag (ACBFS) is the type that is most commonly used as an aggregate material. Processed ACBFS exhibits favorable mechanical properties for aggregate use including good abrasion resistance, good soundness characteristics, and high bearing strength. Table 3 lists some typical physical properties of air-cooled, expanded, and pelletized blast furnace slags [33].

Table 2. Typical properties of BF slag compared with a natural aggregate

Property	BF slag	Carboniferous limestone
Apparent density	2.55	2.72
Water absorbtion (%)	4	0.7
Impact value dry (%)	34	19
Crushing value (%)	34	21
10% fines soaked -kN	85	160
Polished stone value	53-57	38-48
Abrasion value (%)	5-7	8

Table 3. Typical physical properties of blast furnace slag

Property	Slag Type and Value		
	Air-Cooled	Expanded	Pelletized
Specific Gravity	2.0 - 2.5	—	—
Compacted Unit Weight, kg/m ³	1120 – 1360	(800 - 1040)	840
Absorption (%)	1 - 6	—	—

Granulated blast furnace slag is a glassy granular material that varies, depending on the chemical composition and method of production. Grinding reduces the particle size to cement fineness (Figure 8 a), allowing its use as a supplementary cementitious material in Portland cement concrete.

The GGBF slag has the lowest percentage of large particles. The fines specification requires

that <20 wt % of the material is retained by the 45 micron sieve during wet sieving.

The particle size distributions (PSDs) of the slag powder (with the chemical composition given in Table 4) in deionized water and obtained by laser light scattering indicate that many fine particles (<1 μ m) are present, Figure 9 [34].

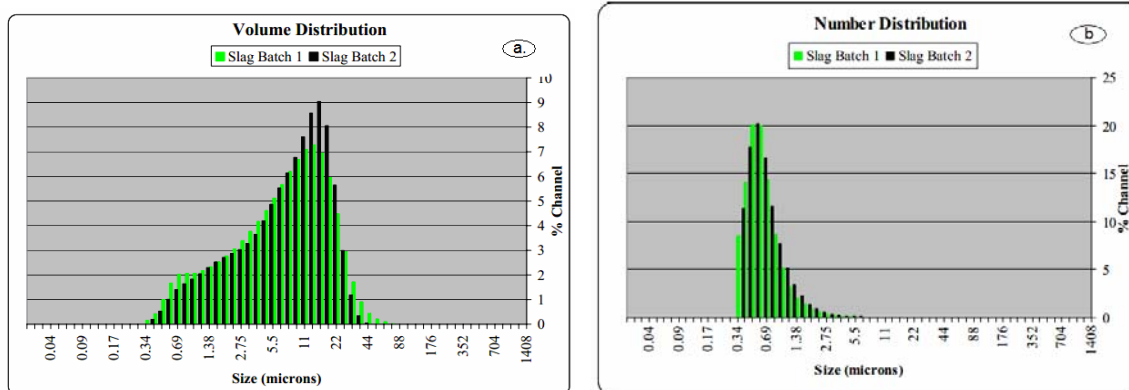


Fig. 9. PSD (volume based) (a) and PSD (number based) (b) for GGBF slag [34]

Table 4. Chemical compositions of two samples of BF slag [34]

Sample	Al ₂ O ₃	CaO	Fe ₂ O ₃	K ₂ O	MgO	Na ₂ O	SO ₄	SiO ₂	TiO ₂
1	8.4	38.5	0.4	0.3	12.9	0.3	1.0	37.9	0.4
2	6.6	35.0	0.3	0.5	13.1	0.3	2.5	40.4	0.3

Also was determined other physical properties of this slag: the mean diameter volume for two samples is 8.80, respectively 9.10; the medium diameter volume is 6.74 and 7.83; the mean diameter number is 0.63 and 0.73 as mean value, respectively 0.52 and 0.58 as medium value; the particles density is 2.70 and the aerated and tapped densities measured for slag are around of 0.80 and respectively 1.26 (tapping deaerates the powder, allowing the particles to compact within the graduated cylinder to produce a greater density than that measured for the aerated materials); the value of ratio of the two densities (tapped/aerated) that is the Hausner Ratio is about 1.56...1.59 and provides an indication of the degree of compaction that a dry powder can undergo in handling and storage.

The porosity of expanded blast furnace slag aggregates is higher than ACBFS aggregates. The bulk relative density of expanded slag is difficult to determine accurately, but it is approximately 70 percent of that of air-cooled slag. Typical compacted unit weights for expanded blast furnace slag aggregates range from 800 kg/m³ to 1040 kg/m³. Unlike air-cooled and expanded blast furnace slag, pelletized blast furnace slag has a smooth texture and rounded shape. Consequently, the porosity and water

absorption are much lower than those of ACBFS or expanded blast furnace slag. Pellet sizes range from 13 mm to 0.1 mm, with the bulk of the product in the minus 9.5 mm to plus 1.0 mm range. Pelletized blast furnace slag has a unit weight of about 840 kg/m³. Because of their more porous structure, blast furnace slag aggregates have lower thermal conductivities than conventional aggregates. Their insulating value is of particular advantage in applications such as frost tapers (transition treatments in pavement subgrades between frost susceptible and nonfrost susceptible soils) or pavement base courses over frost-susceptible soils [35].

3. Conclusions

The global emphasis on stringent legislation for environmental protection has changed the scenario of slags dumping into slags management. Because of natural drive to be cost-effective, there is a growing trend of adopting such slags management measures as would convert slags into wealth, thereby treating these waste as by-products. This has led to aiming at development of zero-waste technologies. In addition to the environmental achievements, the recycling practices produced economic benefits, by providing



sustainable solutions that can allow the steel industry to achieve its ambitious target of "zero-waste". To satisfy environmental and technical requirements of international and national standards is necessary to know the chemical, mineralogical, and morphological properties of BF slag. The chemical composition and the cooling rate determine the phases and the particle shape of slag. These are in correlation with its physical and mechanical properties. All of these BF slag characteristics are essential because they play a key role in their utilization.

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