Recovery of value-added materials from iron ore waste and steel processing slags with zero-waste approach and life cycle assessment

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1 Introduction

A large amount of waste such as mine tailings, sludge, dust and slag is generated as a waste material or by-product from iron and steel industries. Give the high concentrations of alkalinity and toxic metals, these waste materials may pose the serious environmental hazards and cause ecological imbalance (Kumar et al., 2014). A number of sources in both iron making and steelmaking processes produce wastewater containing the pollutants such as suspended and dissolved solids, chemical oxygen demand, zinc, lead, chromium, cadmium, zinc, cyanide, fluoride, oil and grease, organic pollutants (phenol) and ammonia. However, over 90% of the wastewater generated can be reused. The large volume of solid waste, tailing and slags generated from steelmaking and iron making processes has created great concern for their sustainable and economic management, because it is becoming increasingly difficult to find space for dumping these huge volumes (Chen et al., 2014). The treatment and sustainable management of fine-grain metallurgical wastes has ecological and financial limits (Vereš et al., 2011). Therefore, it is important to find out the ways of recycling of these mine wastes (Yellishetty et al., 2008). In 1995, strict government policy, regulations and compliance with environmental requirements contributed up to 20-30% of the capital costs in steel industries (Chatterjee, 1995). However, the steelmaking industries are facing more competition to reduce expenses and increase quality. Therefore, re-use and recycling of these waste and sustainable waste management is of high importance nowadays.

The different manufacturing routes of steel production emit the estimated global CO_2 emissions of 3169 Mt from approximately 1781 Mt of steel production by 2020 with the estimated specific energy consumption up to 14.43 GJ/tcs (Yellishetty et al., 2010). The major environmental benefits of increased scrap usage come from the very fact that production of one tonne of steel through the EAF route consumes only 9–12.5 GJ/tcs, whereas the BOF steel consumes 28–31 GJ/tcs and consequently enormous reduction in CO_2 emissions (Yellishetty et al., 2011). The steel slag in a pile exposed to air undergoes a pH change due to weathering and carbonation, whereas the slag buried underneath the road did not show any remarkable pH change (Suer et al., 2009; Arm et al., 2011). Renforth et al. (2009) found the extent of carbonation and a variable CaCO3 content of an industrial soil up to 20 cm near a former steelmaking plant, and below this depth it was negligible. The accelerated carbonation as an ex situ CO2 storage option, of alkaline steel slag that is rich in alkaline earth metal oxides and/or silicates at optimized operating conditions (i.e. temperature, pressure and pH) can form thermodynamically and chemically stable carbonate phases (Lackner et al., 1995) by several reaction routes (e.g. indirect, direct gas–solid or direct aqueous) (Doucet, 2010; Bobicki et al., 2012; Capobianco et al., 2014).

Over the past 20 years, the innovative technology developments and synergies with other industries have increased the recovery and use of steel industry by-products with a material efficiency rate of 97% worldwide and brought the steel industry ever closer to its goal of zero-waste. Future goal is 100% efficiency, or zero-waste. The recycling during the steelmaking process of the same production unit or sale for use by other industries generates revenues for steel producers and supports the economic sustainability of the steel industry, but also protect the environment (Das et al., 2007). There are many patents developed to re-use and recycle the iron and steel making slags, mine tailings, dust and sludges. However, in some developing countries, its majority part is dumped due to its dangerous properties (high pH value, strong alkalinity, increased content of heavy metals, etc.). The low percentage of use in these countries is presumed due to (1) the negative attitudes of the end-users about toxic effects, (2) sustainability in respective of technological development for recovery, and (3) profit and economic outcomes. Many iron and steel making industries and researchers try to solve above mentioned problem more or less successfully. Therefore, it is necessary to provide a brief overview of the most significant possibilities of slag exploitation with zero-harm and zero-waste policy. The recovery of the valuable components, and transformation of them into valuable products and the processing and utilization of these waste in construction material manufacturing can mitigate the high environmental risk related to the storage, prevent landfill waste, reduce CO_2 emissions and help preserve natural resources. Lobato et al. (2015) present an updated review of the management of slags, sludges, dusts, and mill scales generated by the steel industry, including precipitating sludges generated by galvanizing processes. Current knowledge and studies on the development of alternative management plans were also examined, bearing in mind the importance of sustainable development and the responsible consumption of natural resources. The objectives of the current review described in this book chapter is (1) to boost the innovative and sustainable technologies and re-use of iron and steel making slag, mine tailing and sludges with respect to secondary raw materials recovery, (2) to promote socially acceptable, environmentally responsible and economically viable technologies, (3) to maximize the potential re-use of these materials to existing industrial inputs to production processes, and (4) to review comprehensively the detailed assessment of the work carried until now for the utilization of slag and mine tailing in construction and civil engineering works, wastewater treatment, metal recovery including rare earths, CO_2 storage and mitigating climate change and in soil remediation. The key focus areas of this review are to develop technically and economically sound options for the productive utilization of iron and steel making slag, mine tailing and sludges in various end-uses, prevent landfill waste, and preserve natural resources.

2 Global iron and steel production and approximate amount of waste

There are many countries in the world which are producing and exporting iron ore and steel.
The historical analysis shows that the world iron ore production increased from 274 million tons (Mt) in 1950 to 1554 Mt in 2005, whereas the steel production increased from 207 to 1259 Mt (Yellishetty et al., 2010). The ultra-fines waste/tailings with diameter less than 150 μm are not useful and hence are discarded. In the world a vast amount of mining waste, approximately ----- million tons of such mined ore are lost as tailings, dust, iron and steel slags. More than 400

million tonnes of iron and steel slag is produced each year (WSA, 2010). In future, in order to meet the increasing demand worldwide, the iron and steel production is increasing leading to the increasing generation of iron ore wastes and steel slags in different countries. The iron ore and steel industries are facing the biggest challenges to safely dispose or recycle such vast mineral wastes worldwide.

According to the USGS's estimate, the world's total economic reserves are estimated at 160 billion tonnes (Gt) crude ore containing 77 Gt of iron (Table 1). In 2009, Australia had about 12.5% of world's reserves of iron ore and was ranked third after Ukraine (19%) and Russia (16%) (Table 2). Australia has about 13% of the world's reserves and is ranked second behind Russia (14%). Australia produces around 15% of the world's iron ore and is ranked third behind China (35%) and Brazil (18%) (Table 1). The most important iron ore resources of the world are located in Australia, Brazil, China, India, Russia and Ukraine (Yellishetty et al., 2012).

2.1 Iron making blast furnace slag/waste and tailing

The Fe-mineral ores contain impurities of phosphorus, sulphur and high alkali as well as impregnations of waste rock. The magnetic separation of iron minerals and washing iron ores leaves the tailings and wastewater consisting mostly of silicate rock and clay that are not expected to be hazardous (U.S. EPA, 1988). During iron making, iron ore, coke, heated air and limestone or other fluxes are fed into a blast furnace. Blast furnace slag contains oxides of iron, silicon, aluminum, calcium, magnesium and manganese, along with other trace elements. There are three types of blast furnace slag: air-cooled, granulated, and pelletised (or expanded) (US EPA, 1990). Blast furnace slag should normally be generated at a rate of less than 320 kg/t of iron, with a target of 180 kg/t. The impurities in the feed materials control the generation of blast furnace slag. Cokeless ironmaking procedures are currently being studied and, in some places, implemented such as Japanese Direct Iron Ore Smelting (DIOS) process that produces molten iron from coal and previously melted ores (USEPA, 1995) cutting the costs of molten iron production by about 10%, reducing emissions of carbon dioxide by 5 - 10% (Furukawa, 1994).

2.2 Steel making waste and slag

Steel slag, a by-product of steel making, is produced during the separation of the molten steel from impurities in steel-making furnaces (FHWA, 2012). Quicklime--both high calcium and dolomitic is extensively used as a flux in purifying steel, particularly removing phosphorus, sulfur, and silica, and to a lesser extent, manganese in the electric arc furnace (EAF) and basic oxygen furnace (BOF). Slags consist of a mixture of silica, calcium oxide, magnesium oxide, and aluminium and iron oxides. There are different types of steel slag produced during the steel-making process such as tap slag, raker slag, synthetic or ladle slags, and pit or cleanout slag. These materials should be separated by source-oriented and well-defined handling practices to avoid any contamination of the steel slag aggregate. The solid wastes generated from the different routes of steel making process, including furnace slag and collected dust, range from an average rate of 300 kg/t of steel manufactured to 500 kg/t. Out of which 30 kg may be considered hazardous depending on the concentration of heavy metals present. The by-products containing slags, dusts, sludges and other materials are generated at an average rate of 200 kg/t (EAF) to 400 kg/t (BF/BOF) of steel manufactured.

2.3 Physical, chemical, mechanical and thermal properties of slag

Steel slag aggregates are highly angular in shape and have rough surface texture, high bulk specific gravity and moderate water absorption (less than 3 percent). Table 2 exhibits some typical characteristics and applications of steel slag. Steel slag is generally mildly alkaline with a pH range of 8 to 10. However, the pH of leachate from steel slag can exceed 11, a level that can be corrosive to aluminum or galvanized steel pipes (Chiang et al., 2014). Chemical composition of steel slag consists primarily of calcium silicates, ferrites and oxides of iron, aluminum, manganese, calcium, and magnesium. The mineralogical form of the slag is more important, which is highly dependent on the rate of slag cooling in the steelmaking process. Due to favorable mechanical properties such as good abrasion resistance, good soundness characteristics, and high bearing strength, the steel slags are highly appropriate for aggregate use. The steel slag aggregates have longer heat retention capacity than conventional natural aggregates; these characteristics are advantageous in hot mix asphalt repair work in cold weather.

The composition of ferrous slag is dominated by Ca and Si. Steel slag may contain significant Fe, whereas Mg and Al may be significant in Fe slag. Calcium-rich olivine-group silicates, melilite-group silicates that contain Al or Mg, Ca-rich glass, and oxides are the most commonly reported major mineral phases (Table 3) in ferrous slag (Piatak et al., 2015). Calcite and trace amounts of a variety of sulfides, intermetallic compounds, and pure metals are typically also present. In general, non-ferrous slag may have a higher potential to negatively impact the environment compared to ferrous slag. Because of its characteristics, ferrous slag is commonly used for construction and environmental applications, whereas both non-ferrous and ferrous slag may be reprocessed for secondary metal recovery.

3 Different types and uses of slag

The iron blast furnace slag is processed by granulating, crushing, sizing, etc. and sold for use as aggregate. In some countries, up to 80% of the cement contains granulated BF slag (Cotsworth, 1981) that prevents slag going to landfill as waste, saves energy and natural resources, and significantly reduces CO_2 emissions in cement production. By sintering (heating without melting), iron ore fines are agglomerated and thus recycle iron-rich material such as mill scale, liquid furnace slag and ladle slags. The high content of free lime (alkalinity) and heavy metal (zinc and lead) creates concern and hindrance to use steelmaking slags, which is not ideal for construction applications. Carbonation of slags and recovering the zinc, lead, vanadium and other heavy metals is currently under development to improve lime separation, carbon sequestration and storage and mitigating climate change and meta recovery. The worldwide average recovery rate for slag varies from over 80% for steelmaking slag to nearly 100% for ironmaking slag. In order to maximize the environmental and economic benefits, there is still much potential to increase the recovery and use of slags in many countries.

There are many types and uses of slag commercially available from iron and steel making processes (NSA, 2013). There is no risk of alkali-aggregate reaction. Steelmaking slag (BOF and EAF) is cooled similarly to air-cooled BF slag and is used for most of the same purposes (WSA, 2010). Principle uses of iron blast furnace and steel making slag, dust and sludge include re-use of slag in the steelmaking furnace or sinter plant (Fig. 1), fill and embankments, fertiliser, cement, bricks and concrete production, waste water treatment, coastal marine blocks, working platforms on difficult sites pavements, concrete aggregates, concrete sand, glass insulation wool, mineral wool, filter medium, clinker raw material, railroad ballast, roofing, trace elements in agriculture, sporting field sub-base (for drainage), mine backfilling, lightweight aggregate, road base course material, an aggregate for asphalt concrete and material for civil engineering works and as a ground improvement material (i.e., material for sand compaction piles, soil conditioner). In the BOF route, cleaning the coke oven gas creates valuable raw materials for other industries including ammonium sulphate (fertiliser), BTX (benzene, toluene and xylene – used to make plastic products), and tar and napthalene (used to make pencil pitch, electrodes for the aluminium industry, plastics and paints) (WSA, 2010).

3.1 Re-use of iron ore mineral wastes/tailings in constructions

Tang et al. (2019) developed the experimental process flowsheet to recover reduced iron, cement raw meal and concrete admixture from the iron ore tailing (Fig. 2). The new innovations and patents can find out the better utilization of iron and steel making slags without compromising the quality, environmental and economic factors (Kumar et al., 2014). It is estimated that between 7.0 and 7.5 million metric tons (7.7 to 8.3 million tons) of steel slag is re-used and recycled each year in the United States. The mean values of uniaxial compressive strength (UCS) of concrete cubes after 28 days of curing was found to be of the order of 21.93 and 19.91 MPa with mine aggregate and granite aggregate, respectively, and the metal mobility from these wastes was negligible (Yellishetty et al., 2008). The safe disposal or utilization of iron ore tailings in the form of ultra-fines or slimes, having diameter less than 150 μm has remained a major unsolved and challenging task for the iron ore industries. In India approximately 10 – 12 million tons of such mined ore is lost as tailings (Skanda Kumar et al., 2014). Skanda Kumar et al. (2014) used iron ore tailings as partial replacement to fine aggregates and at 40 percent replacement level after 28 days, the compressive strength was more than the reference mix and other replacement percentage mixes. The use of steel slag as an aggregate is considered in granular base, embankments, engineered fill, highway shoulders, and hot mix asphalt pavement after it is crushed and screened to meet the specified gradation and moisture content requirements for the particular application. Wang et al. (2010) produced iron nuggets to recycle the iron and carbon from steel making wastes. The iron in the nuggets is above 91%, the sulfur content is below 0.04%, and the carbon content is about 4.0%. These nuggets can be used as prime material for electric steelmaking and converter. Due to the presence of free lime and magnesium oxides that can hydrate and expand in humid environments, using slags is not suitable in Portland cement concrete or as compacted fill beneath concrete slabs. Therefore, steel slag should be stockpiled outdoors for several months and watered by natural precipitation and/or application of water by spraying for in-situ carbonation process.

In order to reduce adverse impact on nature created by indiscriminately extraction of natural resources for construction works, the industrial and mine solid wastes can be used for the manufacture of construction products (Venkatarama Reddy, 2009). Masonry is widely used to construct both small and large structures because of its structural versatility and attractive appearance (AL-TALAL et al., 2005; Ullas and Venkatarama Reddy, 2009). Compressive strength of masonry greatly depends on strength of the masonry units. Compressed earth blocks (CEB) or stabilised mud blocks (SMB) are widely accepted as energy efficient alternatives to burnt clay bricks (Venkatarama Reddy, 2009). Based on the block characteristics like wet compressive strength, water absorption, initial rate of absorption and linear elongation, it is found that considerable amount of sand can be replaced by iron ore tailings at different percentages (25%, 50% and 100%) without compromising desirable characteristics of SMB used for masonry (Ullas et al., 2010; Santamaria-Vicario et al., 2016). Water absorption increases with increase in iron ore tailings content but it is within limits. The wet compressive strength of SMB is about 7MPa when 7% cement was used.

Huang et al. (2016) developed the green artificial reef concrete (GARC) using granulated blast furnace slag (GBFS), steel slag (SS) and flue gas desulfurization gypsum as the major raw materials without considerable amount of portlandite. The developed GARC shows a 28-day compressive strength of 71.4 MPa, a density of 2765.5 kg/m^3, and the major hydration products of ettringite and C–S–H gel with a very dense microstructure. The successful development of GARC in the sea preliminarily demonstrates that GARC is suitable for the attachment growth of algae.

The bonding behaviour between slags and asphalt binder is one of the most important properties to ensure a durable BOF slag based asphalt pavement. The BOF slag, firmly coved with hydration products, has the potential to protect the asphalt binder from being stripped by boiling water and had better bonding strength than the values of basalt and granite aggregates (Chen et al., 2014). The reuse of steel slag in bituminous paving mixtures showed satisfactory physical and mechanical properties and a release of pollutants generally below the limits set by the Italian code (Sorlini et al., 2012). Tests on volume stability of fresh materials confirmed that a period of 2–3 months is necessary to reduce effects of oxides hydration. Arribas et al. (2015) examined the long-term aging reactions of EAF slag and its volumetric stability, to gain further knowledge of this by-product, its behaviour as a construction material, and its inherent risk of swelling. The appearance of calcium carbonate enhances the cohesiveness, stiffness and strength of this zone and, as a consequence, of the hydraulic concrete. The clinkers produced with 10% desulfurization slag had a high level of alite and good grindability (Chen et al., 2010). Generally, the improvements in clinkerization and clinker grindability are beneficial to energy conservation in cement manufacture. Quaranta et al. (2015) investigated the influence of using 50% different feedstock steel discards (steel slag, white powders, blast furnace sludge and post-mortem aluminosilicate refractories) added to clays to obtain construction ceramics. The plasticity of the mixtures is greatly influenced by the wastes addition, being the steel slag and post-mortem aluminosilicate refractories, those which most lowered the index, affecting negatively the extrusion forming of the products.

3.2 Recovery of value-added metals and resources from waste

Several possible treatment processes for the recycling and recovery of blast furnace sludge are under investigation, e.g., pyrometallurgical processes (Asadi Zeydabadi et al., 1997, Das et al., 2007) that is however very expensive; only large quantities of sludge with a relatively high Zn concentration can be processed economically. Many steel sludges have Zn content of only a few percent (0.7 %). Hydrometallurgy is presumed to be a more suitable alternative, offering the advantage: it can be used for small quantity of sludge with a low Zn concentration. Vereš et al. (2011) described a hydrometallurgical process by using a microwave furnace to remove the Zn from sludge in order to recycle the Fe and C in the blast furnace and to recover the Zn (Vereš et al., 2012). Harsher chemical conditions involving a higher dose of acid, high temperatures and use of an appropriate catalyst would probably cause dissolution of greater amounts of zinc. However, this will probably be at the expense of greater loss of iron units. Microwave treatment of BFS resulted in very rapid dissolution of zinc phase within less time and electric energy compared to conventional leaching. The higher dissolution rate and higher zinc recovery in the microwave leaching process could be attributed to one or more factors: superheating of the liquid, interaction of the microwaves with the BFS particles in the solution.

Min (2000) extracted zinc and lead from steelmaking dusts using alkaline leaching process and subsequently separated zinc and lead using sulfide precipitation method. It was found that only about 53% of zinc and over 70% of the lead could be leached out of the dusts, while the other 47% of zinc and 30% of lead were left in the leaching residues.

Salt roasting-basic leaching was applied to separate and extract vanadium (more than 80%) from steel making slag (Mahdavian et al., 2006; Li and Xie, 2012). The optimum temperature, time and sodium carbonate content in the roasting process were found to be 1000°C, 45 min and 10%, respectively. The most suitable conditions for leaching process was found to be 80°C, 60 min, sodium carbonate to sodium hydroxide mass ratio of 40-50:10 and particle size between 100 and 120 mesh. Song et al. (2014) extracted vanadium from molten vanadium bearing slag by oxidation with pure oxygen in the presence of CaO. The vanadium could be effectively removed and recovered from steel slag leachates with anionic exchange resins in both batch and column tests (Baxter et al., 2016).

3.3 Effective utilization of blast furnace flue dust from integrated steel plants

Integrated steel plants in general, produce large amounts of solid wastes during iron and steel making process. The blast furnace flue dust obtained from the integrated steel plants contained unburnt coke and iron rich grains as the value-added products (Das et al., 2002). The column flotation technique recovered around 80% carbon with more than 90% recovery from the flue dust samples using diesel oil and MIBC as the collector and frother, respectively. Iron rich grains (magnetite, hematite, wustite and Fe metals) were recovered by low intensity magnetic separation (LIMS) technique. Magnetic separation of flotation tailings gave an iron concentrate of 61-64% Fe with 50-56% over all recovery. For the effective utilization of whole flue dust samples in sinter making, the harmful components such as alkalis present in the flue dust sample should be removed. The simple scrubbing, washing and classifying the products at a finer range removed □85% Na and 60% K values.

3.4 Recovery of REEs including neodymium from steelmaking slag

Rare earth elements (REEs)—particularly samarium, neodymium, and dysprosium— are widely used in energy technologies (hybrid/electrical vehicles, wind/tidal turbines, energy-efficient lighting phosphors, lasers) (Du and Graedel, 2011). Therefore, their supply is critical for the manufacturing of modern materials and future energy technologies (Alonso et al., 2012). The associated rare earth elements in flue dust are La, Ce, Nd, Pr, Y, Er, Dy, etc. Concentration of La and Ce was higher in comparison to other elements (Das et al., 2002). After reaction with oxygen, the resulting rare earth oxides accumulate in the electric arc furnace slag (Nakajima et al., 2009; UNEP, 2013), which is a common waste product from steel production. The slag samples contained considerable amounts of neodymium with an average content of 0.032- 0.014 wt% Nd with the highest amount of 0.058 wt% (Apfel, 2014; Nucor Corporation, 2014) indicating that a significant amount of neodymium can be present in the steelmaking slag (Dhammika Bandara et al., 2015); this concentration is comparable to other REE mines. For example, the richest ore from the Pea Ridge mine contains 12 wt% rare earths overall, but only 1.1–2.1 wt% neodymium (USGS, 2014). Other mines (Mountain Pass, Bear Lodge, Bokan Mountain, Iron Hill) contain between 0.40 and 8.2 wt% rare earths and between 0.034 and 0.78 wt% neodymium (Gupta and Krishnamurthy, 2005; USGS, 2014).

3.5 Recovery of fine iron minerals from iron ore processing tailing

Fine valuable minerals are usually difficult to recover from their ores by processes other than floatation, leaching and bio-recovery since a good number of the common mineral concentration processes employ size and density differences which are more effective if the minerals are liberated at coarse size (Aldennan, 2002; Barratt and Sherman, 2002; Brierly and Briggs, 2002; Chudacek *et al.*, 1997; Fandrich *et al.*, 1997). An analysis of ore properties and the existing processing plant revealed that the current flowsheet can not recover the fine grain minerals leaving them in the processing stream due to their either natural fine-grain size or process of comminution, stacking and reclamation from the blending yard (Burt, 2002; Barratt and Sherman, 2002; DeMull *et al.,* 2002; Srivastava *et al.*, 2001; Anthony, 1993; Barnes, 1988; Ajaka, 2009). For example, Ajaokuta steel plant and the Delta steel plant, Aladja, in Nigeria produces tailing product having between 20% to 22% fine-grained iron minerals (Soframines, 1987). Therefore, Ajaka (2009) analyzed the existing circuit and undertook specific recovery tests on the tailing material using simple hindered settling and floatation process for the recovery of fine iron minerals in the tailings. He recommended that the flowsheet used for the recovery process be integrated with the flowsheet of the existing plant in order to improve recovery of the fine iron minerals lost to the waste.

3.6 Manufacture of cement from furnace slag

The ground-granulated iron blast furnace slag (GGBFS), a by-product of iron and steel-making, can replace 30-50% of Portland Cement in 'normal' concrete as a supplementary cementitious material, but can replace up to 70% in specialist applications such as marine concrete because of the powerful latent hydraulic property, low heating speed when reacting with water, and high chemical durability (Lessing, 1911). It has a glassy structure and can be used as a fine aggregate or binder. GBFS is commonly used in the manufacture of blended cements where it is inter-ground (lower performance) or blended separately (better performance) with cement (Wood, 1981) usually at a 20% - 40% proportion. It reduces the heat of hydration in mass concrete pours, the permeability of concrete, life-cycle costs and maintenance costs, but improves the durability properties of concrete in resistance to aggressive environments, makes concrete more sustainable and enhances the performance characteristics of concrete. The combinations of ground granulated slag and lime were the earliest cements made from slag (Lewis, 1981). Their properties were tested and used in the USA and different European countries as PuzzoIan and Portland cement. But their production ceased in most countries due to its sensitivity to deterioration in storage and the low strength in comparison to present-day Portland cements (Lea, 1971).

The optimal mixing ratio for steel/iron slag blended mortar (SISBM) compressive strength ranged from ground granulated basic oxygen furnace slag (GGBOS) (steel slag): ground granulated blast furnace slag (GGBS) (iron slag) = 3:7 to 5:5 (by weight). At the age of 91 days, the compressive strength of SISBM reached 80–90% compared with that of the ordinary Portland mortar (Tsai et al., 2014). The cementitious characteristics were mainly generated because the GGBOS increased the free-CaO or $Ca(OH)_2$ concentrations in the SISBM curing water and provided alkaline environments for $Ca(OH)_2$ to engage in the pozzolanic reaction with the SiO_2 and Al_2O_3 in GGBS, forming crystals such as calcium aluminum silicate hydrate, (C–A–S–H), calcium silicate hydrate (C–S–H), and calcium–magnesium–alumina–silicate (C–M–A–S), which generated strength and strengthened microstructure.

3.7 Types of blast-furnace slag and uses in cements

The blast-furnace slags, basic-oxygen furnace steel slags and magnesium slags are used, or are under investigation for potential applications, in cements, and that both copper and nickel slags can be utilized with cements as pozzolanic materials (Smolczyk, 1980; Lewis, 1981). The air-cooled slag has little; or no cementitious properties. The granulated slag glass consisting of the same major oxides and suitable activator sets as does Portland cement, has excellent hydraulic properties. Expanded slag is produced by treating the molten blast-furnace slag with controlled quantities of water, usually less than that used for granulation. Physical properties and the cementitious characteristics of the expanded slags vary with extent of crystallization, as does the appearance, from those of air-cooled slags to those of granulated slags. Blast-furnace slag uses in cements, where it maintains its separate identity and is a cementitious component, are divided by Lea (1971) into three categories: (a) ground glassy (granulated) slag is mixed with hydrated lime, (b) super-sulfated cements, and (c) Portland blast-furnace slag cements.

3.7.1 Super-sulfated cement

The cement is usually made by grinding a mixture of 80-85% granulated slag, 10-15% gypsum or anhydrite and about 5% Portland cement. It possesses outstanding resistance to a variety of aggressive agents: sea water, sulfates, weak acids, chlorides, alkali hydroxides, etc. It has been used very successfully in sea water work, concrete pipe exposed to aggressive ground waters, in chemical plants, etc., which is better than that of other cement types. It has excellent strength characteristics and a low heat of hydration. Compared to Portland cements, they are more susceptible to carbonation during storage, and require extra care during the initial curing period to keep the surface moist, if a friable, dusty layer is to be avoided.

3.7.2 Portland blast-furnace slag cements

Portland blast-furnace slag cements constitute the most successful and widely used variety of "slag cements" throughout the world. Consisting of a mixture of Portland cement and ground granulated slag, they are subject to many different specifications with respect to strengths and slag contents in the various countries, but intended to be the equivalent of, and meet the same requirements as, ordinary (or Type I) Portland. In the U.S. these cements are currently covered by C595. The blast furnace slag cement is widely produced in many countries in recent years: 24% in West Germany, 32% in Belgium, 42% in France, 55% in the Netherlands, etc. (Smolczyk, 1980). However, the tonnage of slag ground for use as a cement is equal to about 10% of the total cement tonnage in Germany, France and South Africa. Similar use in the U.S.

would take about 8,000,000 tons of slag annually - nearly 1/3 of the recent production. The performance of Portland blast-furnace slag cement depends on (a) chemical composition of the slag (more basic materials may be more cementitious), (b) high glass content, (c) more fineness of grind, and (d) the amount of slag used in the blend. Determination of its properties, blended with any given cement, is best determined by tests with that cement. There is no other cement component by which more energy and natural resources can be saved than it is with the granulated blast-furnace slag. Therefore, countries with suitable blast-furnace slags and sufficient experience with slag cements can use blast-furnace slag cements in the same or better way as other cements (Smolczyk, 1980).

3.8 Environmental application of iron ore waste and steel slag

Remediation of gold and base metal mining waste

Phytostabilisation of a metal contaminated soil using beringite and *Agrostis capillaris* L. (Vangronsveld and Cunningham, 1998) decreased metal uptake by plants and leaching of metals to groundwater (Knox et al., 2001). Application of steel industry waste such as steelshots to soil decreased Cd and As exposure and uptake through the formation of Fe and Mn oxides in soil. Beringite can change soil pH and immobilise Zn and Cd (Vangronsveld and Cunningham, 1998, Vangronsveld et al., 1999). Mench et al. (2003) used iron and steel industrial waste and revegetation to rehabilitate the metal mining contaminated soil by *in situ* inactivation of trace elements by C (5% compost), CB (5% compost + 5% beringite), CSS (5% compost + 1% steelshots), and CBSS (5% compost + 5% beringite and 1% steelshots) treatments. After 3 years, revegetation by native plant species was excellent in the CSS treatment, and successful for the CBSS. The CBSS and CSS treatments promoted rehabilitation of mining wastes by revegetation via limiting water-soluble As and decreasing long-term metal leaching due to iron oxide minerals produced from iron and steel waste.

Andreas et al. (2014) evaluated the use of steel slags (EAF slag and cementitious ladle slag) as the construction materials for a final cover of an old municipal landfill (Fig. 3). The tested electric arc furnace slags are suitable to be used in the foundation and equalisation layer, in the drainage layer and, mixed with cementitious slag (ladle slag and similar), in the low-permeability barrier layer (liner). The recipe for the liner mixture should contain at least 50 wt% of cementitious slag; the particle size of the ballast EAFS should not exceed about 3% of the liner thickness. The existing results of cover performance and stability are promising, but since they are the first and only results, they must be supplemented with monitoring and testing over a longer period before any firm conclusions can be made about long-term processes.
Slag amendment to the soil aquifer could contribute more removals of DOC and phosphate under both unsaturated and saturated conditions of soil aquifer treatment possibly due to its larger surface area (Cha et al., 2006). The steel slag greatly accelerated the formation of humic like substances. The Fe(III)-and Mn(IV)-oxides in steel slag act as oxidants and substantially enhance the polycondensation of humic precursors while composting biowastes (Qi et al., 2012). Leaching tests show that only low amounts of Cr, present at relatively high concentration in BOF steel slag reused as an aggregate for road constructions, are released while the release of V is significantly high. X-ray absorption near-edge structure (XANES) spectroscopy indicates that Cr is present in the less mobile and less toxic trivalent form and that its speciation does not evolve during leaching. On the contrary, V which is predominantly present in the 4+ oxidation state

seems to become oxidized to the pentavalent form (the most toxic form) during leaching (Chaurand et al., 2006, 2007).

Inter-industry utilization of solid residues such as combination of desulphurization slag arising within the steel for soil amendment shows promise in the replacement of commercial liming materials (neutralizing value 38.3% Ca equivalents, d.w.) (Mäkelä et al., 2012). The pseudo-total concentrations of regulated trace elements were lower than the Finnish statutory limit values for fertilizers (Decree on Fertilizer Products 24/11)) and the European limit values for awarding the Community eco-label for soil improvers (Commission Decision 2006/799/EC).

4 Carbon sequestration, resource recovery and environmental benefit

$Ca(OH)_2$ and CaO from steel slag or concrete waste can be dissolved in water on a time scale of hours, and reacted with CO_2 in ambient air to capture and store carbon safely and permanently in the form of stable carbonate minerals ($CaCO_3$) (Stolaroff et al., 2005). The EAF slag from the pavement edge showed traces of carbonation and leaching processes, whereas the road centre material was nearly identical to fresh slag, in spite of an accessible particle structure (Suer et al., 2009). Tufalike precipitates, a white powdery precipitate, consisting primarily of calcium carbonate ($CaCO_3$), result from the exposure of Ca-rich steel slag aggregates to both water and the atmosphere (CO_2). Chiang et al. (2014) developed a three-stage process to transform blast furnace slag into two valuable products such as precipitated calcium carbonate such as calcite, aragonite and vaterite and micro- and meso-porous zeolitic materials (tobermorite, sodalite, lazurite, and analcime) to simultaneously achieve sustainable CO_2 sequestration and solid waste elimination (Kunzler et al., 2011). The carbonation of EAF and argon oxygen decarburization (AOD) slag in powdered form resulted in a linear increase of compressive strength up ~30 MPa after three weeks reaching a maximum of 4.3 wt.% CO_2 uptake, which is sufficient for many construction applications (Baciocchi et al., 2015a,b).

5 Conclusions and technological development

Steel slag is generally mildly alkaline with a pH range of 8 to 10 with occasionally pH value exceeding 11. Chemical composition of steel slag consists primarily of calcium silicates, ferrites and oxides of Fe, Al, Mn, Ca, and Mg. Due to favorable mechanical properties such as good abrasion resistance, good soundness characteristics, and high bearing strength, the steel slags are highly appropriate for aggregate use. The steel slag aggregates have longer heat retention capacity than conventional natural aggregates.

In order to increase the viability of using slag, neutralization of alkaline material and recovery of metals are necessary. Carbonation of alkaline iron ore and steelmaking slags is a promising technology for carbon capture and storage as a CO_2 mitigation strategy in the context of climate change. To maximize the economic and environmental benefits, it is important to find out the new innovations and sustainable ways of carbonation and recycling of these mining wastes. Re-use and recycling of these iron ore and steel making slag, recovery of valuable metals (Zn, Pb, V, REE especially Nd) and resources can prevent landfill waste, reduce CO_2 emissions, preserve natural resources and mitigate climate change by natural and accelerated carbonation of alkaline steel slag. The iron blast furnace and steel making slag, dust and sludge can be recycled and re-used in the steelmaking furnace or sinter plant, fill and embankments, fine iron ore minerals, fertiliser, cement, bricks and concrete production, waste water treatment, remediation of metal mining contaminated soil, coastal marine blocks, pavements, concrete aggregates, mineral wool, filter medium, railroad ballast, roofing, road base course material, a ground improvement material, BTX (benzene, toluene and xylene), and tar and napthalene. Hydrometallurgy is better than pyrometallurgy to recover metals such as Zn, Pb, V, Nd, etc from steel slag and sludge. The steel slag contained considerable amounts of neodymium with an average content of 0.032-0.014 wt% Nd with the highest amount of 0.058 wt% indicating that a significant amount of neodymium (REE) can be present in the steelmaking slag. The new innovation and integration of new flowsheet with the existing one can improve recovery of the fine iron minerals lost to the waste streams.

In spite of long history of using the ground-granulated iron blast furnace slag, a by-product of iron and steel-making in the manufacturing of cement and replacing the use of Portland Cement in 'normal' concrete as a supplementary cementitious material, their production ceased in most countries due to its sensitivity to deterioration in storage and the low strength in comparison to present-day Portland cements. The blast-furnace slags and their uses in cements are divided into three categories: (a) ground glassy (granulated) slag is mixed with hydrated lime, (b) super-sulfated cements, and (c) Portland blast-furnace slag cements. The blast furnace slag cement is widely produced in many countries in recent years. However, the tonnage of slag ground for use as a cement is equal to about 10% of the total cement tonnage in some countries. There is no other cement component by which more energy and natural resources can be saved than it is with the granulated blast-furnace slag. Therefore, countries with suitable blast-furnace slags and sufficient experience with slag cements can use blast-furnace slag cements in the same or better way as other cements.

Technologies to further improve by-product recovery rates and expand their potential benefits include improved material separation technologies and carbon sequestration that could dramatically reduce steel industry CO_2 emissions (WSA, 2010). One technology, in early development, uses slag to sequester carbon during steelmaking and could reduce CO_2 emissions by 85% while converting the slag and exhaust gas to potentially marketable products such as carbonates (Geological Sequestration, 2007). Together with existing technologies, new developments provide environmentally and economically sustainable solutions to bring the steel industry ever closer to its goal of zero-waste.

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Table 1: Iron ore reserves in selected countries in the world (2009 data) (Yellishetty et al., 2012, USGS, 2010a)

Country	Iron ore reserves (G)	Iron content (Gt)	Production in 2009 (Mt)		Rank in 2009	
			Iron Ore	Crude Steel	Iron Ore	Crude Steel
Australia	20	13	370	5.25	3	23
Brazil	16	8.9	380	26.51	2	9
China*	22*	7.2*	900	567.84	1	1
India	7	4.5	260	56.6	4	5
Russia	25	14	85	59.94	5	3
Ukraine	30	9	56	29.75	6	8
USA	6.9	2.1	26	58.14	10	4
World	160	77	2,300	1,220	--	--

*China is based on crude ore, not saleable ore (China has large but low grade, poor quality reserves)

Table 2: Characteristics and applications of steel slag (Yi et al., 2012)

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, SiO2 components	Raw material for cement clinker
Fertilizer components (CaO, SiO_2, MgO, FeO)	Fertilizer and soil improvement

Table 3: Most common minerals found and in the tested steel slags (Andreas et al., 2014). Open access, copyright permissions under a common creative license.

EAFS 1 and 2	Ladle slag		
Iron	Fe	Periclase	MgO
Merwinite	$Ca_3Mg(SiO_4)_2$	Calcium Silicate	γ and α-Ca_2SiO_4
Monticellite	$CaMgSiO_4$	Iron	Fe
Clinoenstatite	$MgSiO_3$	Spinel	$MgAl_2O_4$
Calcium silicate	Ca_2SiO_4	Mayenite	$Ca_{12}Al_{14}O_{33}$
Magn. alum. oxide	$MgAl_2O_4$		

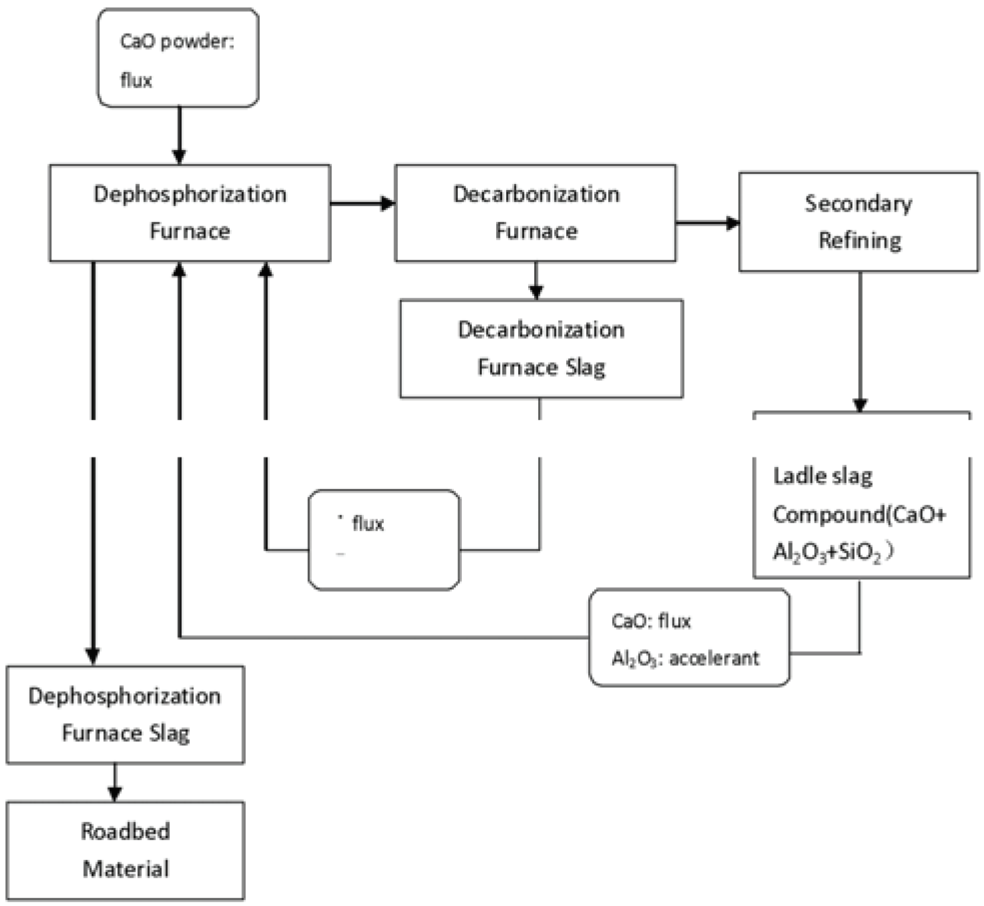

Figure 1. Recycling and application of steel slag in steelmaking plant (Yi et al., 2012).

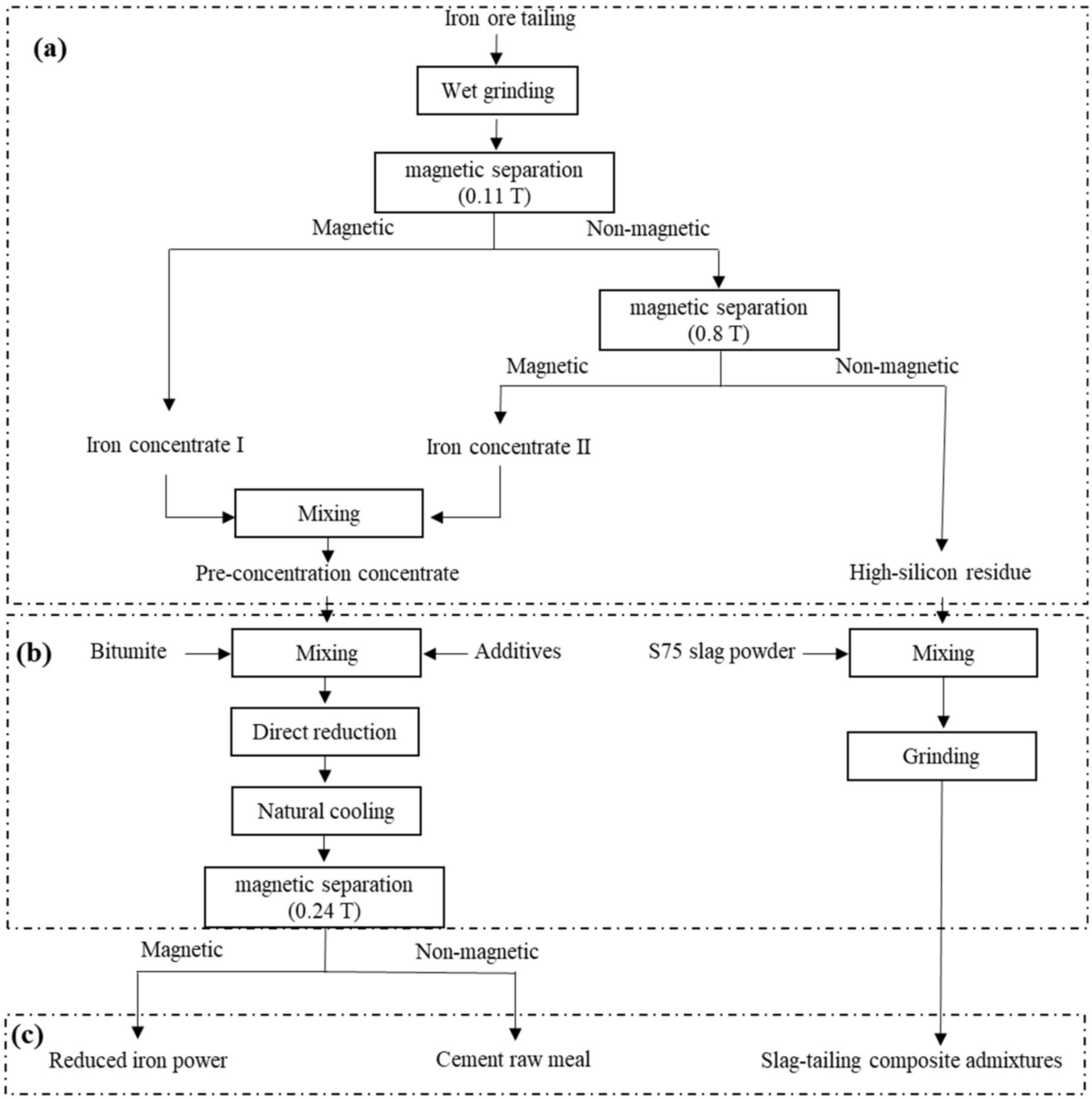

Figure 2. Experimental flowsheet: (a) The process of pre-concentration; (b) the process of iron recovery; (c) final products (Tang et al., 2019). Open access, copyright permissions under a common creative license.

	Area 1 2005	Area 2+3 2007, 2008	Area 4+5 2010, 2011	
Vegetation layer	Compost	Compost	Compost	> 0.25 m
Protection layer	80% borrow soil 10% bio ash 10% treated sludge 9.1 E- 09 m/s	100% borrow soil 2.5 E- 08 m/s	80% borrow soil 10% bio ash 10% treated sludge 3.1 E- 09 m/s	≈ 1.5 m
Geotextile Drainage layer Geotextile	EAFS 1+2 (8-60 mm)	EAFS 1+2 (35-60 mm)	EAFS 1+2 (20-60 mm)	≥ 0.3 m
Liner	>50% EAFS 1+2 (<8 mm) <50% LS (<20 mm) < 2.2 E- 11 m/s	>65% EAFS1+2 (<35 mm) <35% EAFS4+LS (<20mm) < 1.2 E- 10 m/s	50% EAFS 3 (< 20 mm) 50% EAFS 4+LS (<20 mm) < 2.2 E- 11 m/s	≈ 0.7 m
Foundation layer	Sweepings	Sweepings	EAFS 1+2 (0-150 mm)	≥ 0.3 m
	Waste	Waste	Waste	

Figure 3. Design for the test areas A1–A5; year of construction, materials used, recipes (percentage by weight) and layer thickness. Hydraulic conductivities determined in laboratory tests (Herrmann et al., 2010) are given for liner and protection layer (Andreas et al., 2014). Open access, copyright permissions under a common creative license.