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Recycled smelter slag as an engineering material - opportunity and sustainability

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Abstract. Slags obtained from the Vale Copper Cliff smelter in Sudbury, Ontario, Canada, were investigated as sustainable engineering materials in this study. The recycled smelter waste products can remove toxic contaminants from the aqueous environment as well as be used in the construction industry (as aggregates, cement admixtures, filling materials), soil improvement for agricultural purposes, and other value-added applications and products. The removal mechanisms of the heavy metals (such as Zn, Pb, and Cu, etc.) from aqueous solutions could be physical or chemical adsorption, ion exchange, oxidation-reduction, etc. At the same time, using recycled smelter slags in various engineering applications can help with waste reduction, disposal cost reduction, resource recovery, and increased reused activities. The present study helps explore the scope of using recycled materials in the treatment or construction industry. Using industrial smelter slag as a recycling or renewable resource rather than a waste product has environmental and economic benefits. The study also specifically discusses Ni smelter slag's composition, application, treatment efficiency, opportunity, economic benefits, and circularity for sustainable management.

Introduction

Slags, waste products generated from metal mining activities or the metallurgical industry, can be treated and recycled for other applications [1-2]. Water and wastewater treatment using low-cost by-products from the agricultural, domestic, and industrial sectors has been acknowledged as a viable option [3]. They make it possible to remove pollutants from aqueous solutions while also contributing to waste minimization, recovery, and reuse. The current study investigated the removal efficiency of various contaminants using low-cost waste products as well as other engineering applications from Ni smelter slags. It considered recycled smelter slags consisting of iron oxides, iron silicates, and different oxide minerals. Slags are a potential adsorbent or reactive media in water and wastewater treatment [1–3]. Other applications for recycled smelter slags include aggregates, cement admixtures, filling materials, fertilizers, and other value-added applications and products [4]. Vale, a major nickel producer in the world, discards 1.2 million tonnes of slag annually, whereas Inco's entire slag inventory in the Sudbury region is 115 million tonnes after about 90 years of business operations [5]. Huge amounts of abandoned slag result in leaching of metals and the waste of valuable metals, as well as environmental contamination from the oxidation of entrained sulfur to sulphuric acid [5].

To provide justification, a lab-based experimental investigation using recycled Ni smelter slags was conducted to explore the physical and chemical properties as well as the adsorption capacity and kinetics of toxic Zn, Cu, and Pb from water solutions. Furthermore, the primary motivation was to evaluate the applicability options of recycled Ni smelter slags in different engineering applications in order to assess the circularity of the materials in the sustainable development process. The study explored the benefits of using discarded waste products in the development of sustainable green technology.

Physical and chemical characteristics of Ni smelter slags
In the present study, the Ni smelter slags obtained from the Vale Copper Cliff smelter in Sudbury, Ontario, Canada were stored in the Environmental Engineering Lab at Western University in Canada. These slag particles were crushed into smaller fragments and later more finely ground into finer particles with a pestle and mortar. It was kept in airtight containers for experimental purposes. The composition of the bulk smelter slag was also measured using PAN-analytical PW-2400 Wavelength and Dispersive XRF (X-Ray Fluorescence). Moreover, the Surpass Anton Analyzer was used to determine surface charge and charge changes at different pH values. Figure 1 shows the change of surface charges with different pHs. Slag particles larger than 25 µm were used to conduct the experiment. The operational conditions for the Anton analyzer were 115 to 230 V, 50 to 60 Hz, and 200 VA. From XRF tests, the results showed that the major compositions of the slag were iron oxides and silicon dioxides, at 59% and 39-41%, respectively.

In the study, the major components of the slag particles were found to be Fe_2O_3, SiO_2, Al_2O_3, CaO, and MgO. Previous researchers have used SiO_2, Al_2O_3, Fe_2O_3, MgO, CaO, and TiO_2 for metal removal [2, 6-9]. According to Liu et al. [9] and Thakur et al. [2] steel slag containing 10 to 40% of Fe_2O_3, SiO_2 and CaO was capable of reducing 99% of Pb from aqueous solution. Perederiy [5] reported that the composition of iron and silicon are the main elements (e.g., 57.5% wt) of the Ni smelter slag. In addition to iron and silicon, Ni and Cu are found to be around 1% of the slag mass, together with some cobalt (0.24%wt). Chowdhury et al. [10] found the presence of adsorbed Cu (1.6% wt) and Ni (0.2% wt) in the recycled slag materials, indicating the affinity for Cu adsorption. The Surpass Anton Analyzer was used to determine the variations in surface charge on Ni smelting slag as a function of pH. The findings revealed that before a pH value of 3.1, recycled Ni smelting slag had a positive charge. However, it had a negative surface charge after pH 3.1 (as shown in Figure 1), indicating the positive cation affinity. From three sets of experiments, the standard deviation of each point for Figure 1 was found to be ±1 to ± 3.

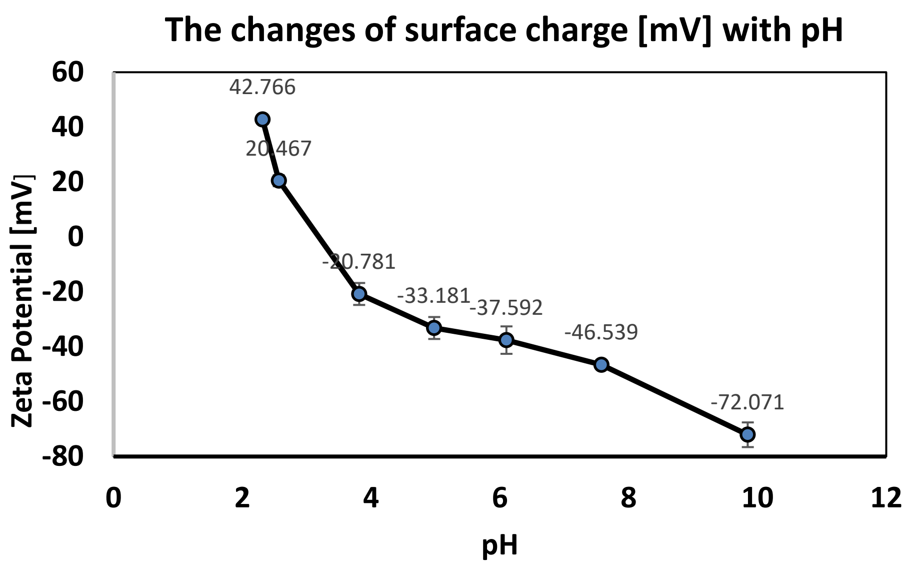

Figure 1: *The change of surface charges of Ni smelter slag with different pHs*

The slag was further analyzed by using X-ray based techniques, such as X-ray photoelectron and Raman spectroscopies. As reported by Chowdhury et al. [10] the XPS Fe mutiplet analysis showed that the Ni smelter slag surface would consist of a mixture of iron oxides such as magnetite, maghemite, and goethite, etc. More specifically, it contains 50% magnetite, 28% maghemite, and 22% goethite. Besides, 21% of SiO_4 was detected, indicating the presence of fayalite ($FeSiO_4$) compound. Thus, in conclusion, the major composition would be mixed iron oxides and silicon dioxide. Table 1 shows the characteristics and possible applications of Ni smelter slag in different engineering fields. The preliminary characterization reveals that it can be used as aggregates for concrete, an admixture for cement, fertilizer and soil improvement, raw material for cement, possible CO_2 capture, flue gas desulfurization, and an adsorbent for waste water treatment.

Table 1: *Characteristics and possible applications of Ni smelter slag*

Slags Properties	Possible Engineering Applications
Porous, alkaline	Waste water treatment
Al_2O_3, FeO, SiO_2	Adsorbents
Hard, wear-resistant,	Aggregates for concrete
Adhesive, rough, Cementitious components (e.g. dicalcium silicate, tricalcium silicate)	Aggregates, admixture for cement
FeOx, $FeSiO_2$ and Fe components	Adsorbent in Water Treatment, Iron reclamation
CaO, SiO_2, MgO, FeO	Fertilizer and soil improvement, raw material for cement
CaO, MgO components	Possible CO_2 capture and flue gas desulfurization

In this study, Laser Tests were used to determine the concentrations of crystalline, trace, and bulk substances. After crushing, a Malvern Laser 2000 was used to assess the fresh slag's specific surface area and particle size distribution. The average size and specific surface area of the slag materials were found to be 55 µm and 0.6 m^2/g, respectively. It was found that the slag grain size distribution ranged from 0.3 to 155 µm.

Utilization of Ni smelter Slag materials

Water treatment

Slag has a porous structure, large surface area, and is quite dense, making it simple to separate from water, alkaline and consisting of FeO, Al_2O_3, FeO, SiO_2 etc. As a result, the use of steel slag in the treatment of industrial waste water has garnered a lot of interest recently.

Different Metals Removal by Ni smelter slags

Batch experimental methods

A batch experiment was conducted by using known amounts of nickel slag with 50 mL metal solutions in the 125 to 300 mL Erlenmeyer flask at 23ºC ± 2ºC. This test measured the adsorption efficiency of smelter slag. All batch experiments were conducted three times on the rotary shaker in certain environmental conditions. A speed of 170 rpm was used during batch experiments since good contact between the adsorbent and adsorbate was achieved. In all experiments, control blanks were also used for quality control and assurance purposes. In this study, the batch tests were conducted in different experimental conditions. Solution samples were collected after the completion of adsorption and then filtered with 0.2 µm filters. Metals' concentrations were measured using the Varian Vista Pro inductively coupled plasma optical emission spectroscopy (ICP-OES). The filtrates were acidified with 2 % HNO_3 for the determination of metal concentration. In this study, the batch adsorption experiment was performed at 0,1 to 10 mg/L of Pb As, Cu solution with 10 g/L of slag particles for 10 hours at pH 2–11 in order to find out the adsorption capacity.

Experimental results
The extent of adsorption and the effect of pH on Cu and Pb removal by Ni smelter slag were determined in this study. Figure 2 shows the results of the possible removal of Pb and Cu from the series of experiments in the different pH ranges. The experimental results were found to be 98% of Pb removal from water in the pH range of 4 to 9 and more than 90% of Cu removal was achieved in the same pH range. In Fig. 2, the range of standard deviations for the removal efficiency (%) of Pb and Cu was found to be small, that is, ±0.08 to ± 2.5.

The effect of pH on cations' removal could depend on the point of zero charge [11]. The point of zero charge of the nickel smelting slag was around 3.1 in this study (as shown in Figure 1). The surface of the adsorbent is negative below the point of zero charge. Thus, cations (Me^{2+}, $Me(OH)^{+}$) would be attached to the surface through ion exchange with H^{+} [12]. This explains why metal removal rates at pH 5 and higher are so high in the case of Cu or Pb removal by Ni smelter slag.

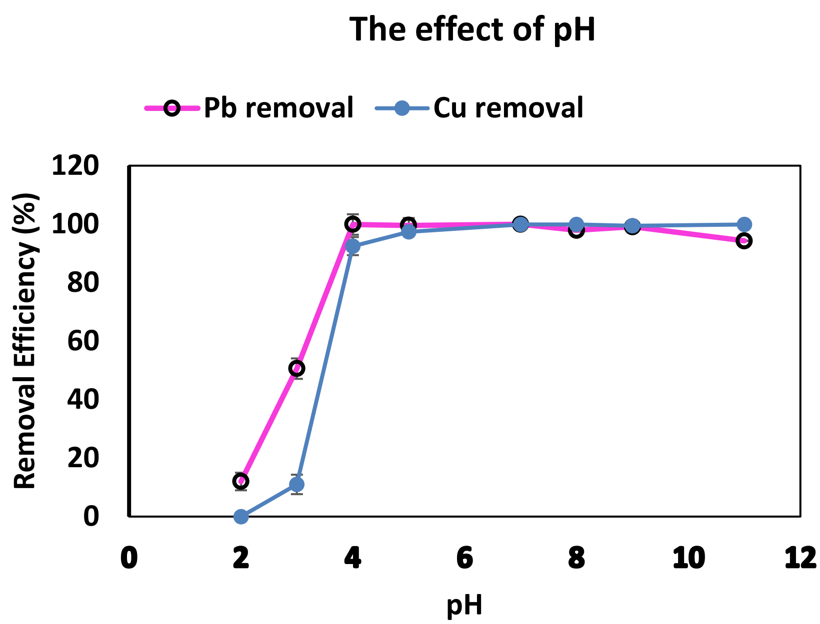

Figure 2: *The removal of Pb and Cu by Ni smelter slag with different pH*

The pseudo first order and second order kinetic models were used to investigate the sorption kinetics of Pb and Cu on Ni smelting slag. Figure 3 shows the fitted lines obtained from the second order kinetic rate models for Pb and Cu removal from aqueous solution. From the results, it can be concluded that the pseudo second order model was better fitted to the experimental data than the pseudo first order model. An R^2 value of 1 for both Pb and Cu was found.

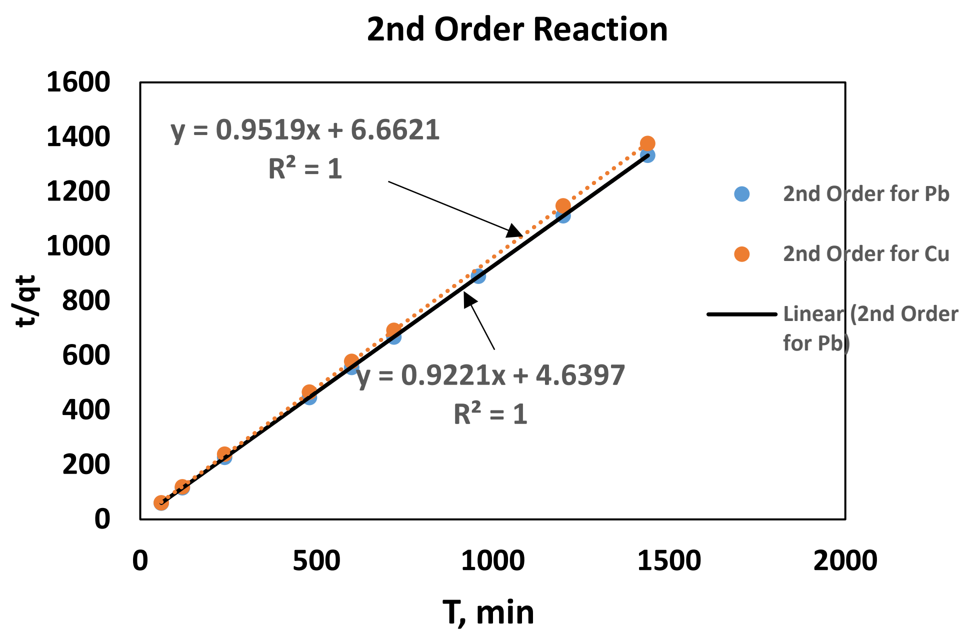

Figure 3: *Pseudo second order plot for Cu and Pb removal by Smelter Slag*

The pseudo-second order describes chemisorption, which includes both ion exchange and covalent interaction between an adsorbate and an adsorbent [7,12-13]. The results from this study showed that the adsorption of Pb and Cu by the Ni smelter slag was due to chemical interaction between the adsorbate and the adsorbent (e.g., reactive Ni smelter slag). Thus, it is clear that the removal mechanism of Pb and Cu could be physio-chemisorption.

The optimum contact time and the kinetics of the removal of Pb and Cu with the recycled Ni smelter slag were also assessed using batch experiments. The results showed that the removal was very fast in the initial stages of the experiments and then gradually decreased at the end of the experiments. 55.85 % and 91.58 % of Pb were adsorbed in the first 5 and 20 minutes while 34.55 % and 88.65 % of Cu was removed in the experimental condition. The study was conducted at pH 5, 10 mg/L of initial concentration and 10 g/L of slag doses.

The effect of slag size on the removal of Pb or Cu by smelter slag was investigated. The batch experiment was conducted at an initial concentration of 10 mg/L for both metals, 10 hours of contact time, 170 rpm of shaking speed, and 250C. Different particle sizes of slag at a dose of 10 g/L were used to measure the effect of particle size on metal removal. The removal of toxic metals was shown to increase with a decrease in particle size (as shown in Figure 4). Removal between 10% and 100% was achieved for Cu and 16 % to 100% for Pb over the size range studied. A significant increase in adsorption was achieved when the particle size was reduced from 200 µm to 128 µm for both Cu and Pb removal. More than 99% of both toxic metals' removal was achieved when particle size was kept below 90 µm.

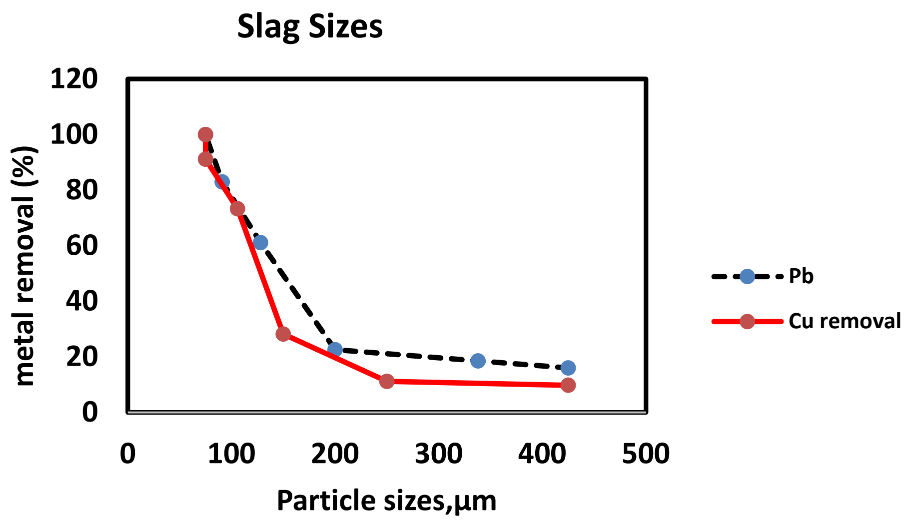

Figure 4: *Effect of slag sizes on removal of Pb or Cu by smelter slag (Initial concentration: 10 mg/L, contact time: 10 hours, shaking speed: 170 rpm, temperature: 25^{0}C)*

Utilization of Slags in Construction

The cement industry consumes a lot of energy and raw materials. Therefore, it has a major impact on the environment because it releases a lot of greenhouse gases, like carbon dioxide, during the manufacturing of cement clinker and energy use. The need to limit the emission of industrial pollutants through the utilization of industrial byproducts like ferrous and nonferrous slag materials. With an annual production of more than 3 billion tonnes, cement is the most commonly used building material [4, 19, 20]. The production of cement is an energy-intensive process and produces large amounts of CO_2 emission, which causes serious environmental problems such as global warming and climate change [20]. Therefore, it is necessary to make cement-based products more environmentally friendly [19–20]. Due to their comparable composition and morphology to cement or sand, some metallurgical slags, such as steel, copper, slags, such as steel, copper, nickel, lead-zinc, and electric furnace ferro-nickel slag, have the potential to be employed in cement-based materials [4, 9–21]. A partial substitution of metallurgical slags for cement or sand can significantly reduce CO_2 emissions, save natural resources, and minimize the discharge and pollution of waste slags due to the enormous use of building materials.

The chemical properties of smelter slags (e.g., Ni smelter slags) show some advantages to use as an additive, admixture or cementing substances for the cement and concrete construction. The presence of calcium, alumina and silicate endorse the Ni smelter slags cementitious properties [21]. It is generally agreed that the cementitious properties of ferrous and nonferrous slag increase with its basicity. Thus, slag materials crushed into fine materials could be used as cement additives and concrete admixtures. The potential cementitious property of slag particles considerably increases with their fineness. The fineness of the slag is another major element determining the activity [21-22].

Slags from nickel smelters can also be utilized as an aggregate in concrete for refractory and high strength applications. Because slags are hard and have a poor grind-ability index, it is important to build highly effective and energy-efficient pulverizing mill equipment for crushing [21]. According to Qasrawi, et al. [23], steel slag used as fine aggregates increased the compressive strength of concrete by 1.1 to 1.3 times. Papayianni et al. [24] created a high-strength (>70MPa) concrete from Iron slags. Zhao et al. [19] used Cu smelter slags for concrete construction and achieved maximum compressive strength to be 40 MPa (after 28 days). Therefore, it is expected that similar application in the construction sector can be achieved by Ni smelter slags. In addition, in many situations, the slag material may represent the most effective and affordable long-term solution in the waste management economy, filling the market gap in light of the depleting anthropogenic resources.

Fertilizer from Ni smelter Slags

CaO, SiO_2, and MgO are fertilizer constituents found in Ni smelter slag. Thus it can be used for a variety of agricultural uses because it comprises these three components as well as others including FeO, MnO, and P_2O_5. Its alkaline characteristics treat acidic soil [19]. Moreover, converter slags are used to create siliceous fertilizer, phosphorus fertilizer, and micronutrient fertilizer in developed nations including Germany, the USA, France, and Japan [25].

Possible application in CO_2 capture

One of the main greenhouse gases, CO_2, has a significant impact on climate change. Therefore, research on CO_2 reduction technologies has been concentrated on carbon capture and storage (CCS). At present, mineral CO_2 sequestration is one of the most widely used methods of CO_2 sequestration due to its advantages, such as being ecologically friendly, permanently trapping CO_2 in the form of carbonate, and obviating the need for post-storage surveillance for CO_2 leakage [26]. Magnesium or calcium oxides in silicate minerals are promoted to react with carbon dioxide and create carbonates in order to store CO_2 gas in minerals carbonation [27]. Ni smelter slag contains CaO, MgO as well as SiO_2. Thus, it is possible to store CO_2 in carbonates forms using Ni slag slag slurry with room temperature and CO_2 pressure [28]. With regard to reaction time, liquid-to solid ratio (L/S), temperature and initial pH, Chang, et al. [29] studied the technological conditions of CO_2 sequestration with steel slag slurry. Sun, et al. [30] in his experimental study showed that the maximum CO_2 capture capacity could reach to 211kg CO_2/ton steel slag. The results found that the consideration of the contribution $Mg(HCO_3)_2$ in capturing CO_2. The precipitate produced under ideal carbonation conditions contained a high amount of $CaCO_3$, up to 96 2 wt% in composition [30].

Technical benefits and sustainable management

Recycling is one of the options for waste reduction. This can be done by recovering products from waste outputs. It is clear that smelter slag is a promising reactive material for metal adsorption. Slags can be used as a recycling or renewable resource rather than as a waste product, which has environmental and economic benefits. The results of the present study indicate the potential use of recycled smelter slags as adsorbents. Although the maximum adsorption capacity of slag was less than many conventional adsorbents, the results indicated that the slag adsorbed As (V), Pb, and Cu from aqueous solutions and thus could be used in the treatment of metal contaminated water. They can be considered circular products by applying recycling at the source, recovering the precise compounds before final disposal, and using the products in different engineering applications. The amount of industrial smelter waste for disposal would be reduced if it is used in different engineering applications. The results from the different scientific studies suggested that the application of recycled waste materials for site treatment as well as contaminated water treatment would be sustainable and promising.

The Ni smelter slag can be used for in-situ treatment of the mine effluent or contaminated groundwater [31] by using permeable reactive barrier treatment since the present study showed the adsorption efficiencies for As, Pb, and Cu. For every successful engineering solution to be implemented, cost is a critical consideration. Proper installation, operation, and maintenance costs should be determined for any proposed technology. Smelter slags are crushed into desired sizes prior to use as a reactive medium in any treatment process. The major cost factor associated with the use of slag particles as an adsorbent would be the crushing of the slag particle sizes [31]. Using crushed slag in water and wastewater treatment is beneficial both in terms of recycling and economics. Figure 5 shows Ni smelter slags production and the possible applications. The possible application of recycled Ni smelter slag materials suggest as potential circular engineering materials which could be used in contaminated water treatment, and site remediation engineering. It can be used in construction work as well as soil improvement purposes [4, 19].

Conclusions

In conclusion, the present study examined an overview of the adsorption process between the crushed waste product and the contaminant. The application of recycled smelter products in the treatment process can contribute to a circular economy, sustainable technology and a green environment. In addition, recycled slags can also reduce costs associated with obtaining natural aggregates and reduce the environmental impact associated with solid waste disposal. It can be used as admixture of cement and can be constructed concrete blocks or aggregate for construction projects. Recycling of waste slags could reduce the demand for virgin natural resources. By lowering the amount of new materials and products being produced, recycling and upcycling of used or discarded products have a significant potential to ease the burden on our planet. The innovation in the circular economy are crucial to the transition to more sustainable infrastructure development.

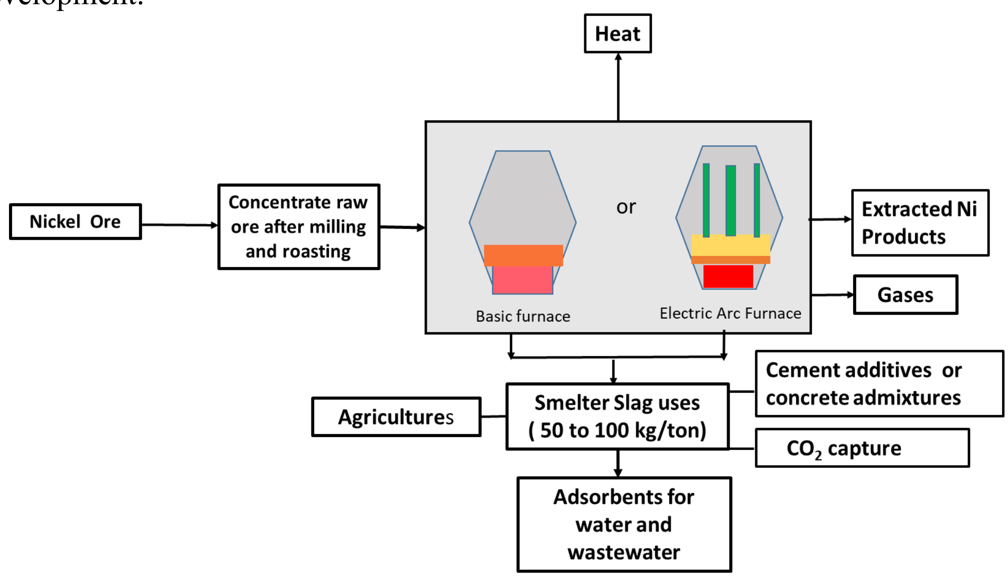

Figure 5: *Ni smelter slags production and the possible applications [32]*

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