

Valorization of Steelmaking Slag for Circular Economy Applications: Adsorptive Removal and Recovery of Ni(II) and Cu(II) from Aqueous Systems

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Abstract

The transition toward a circular economy requires innovative strategies for valorizing industrial by-products. This study investigates the potential of steelmaking furnace slag (SFS) as a low-cost adsorbent for the removal and recovery of nickel and copper ions from aqueous systems. The slag was characterized using XRF, XRD, SEM, FTIR, and thermal analyses, confirming the presence of reactive phases such as lime, periclase, and calcium silicates. Batch adsorption experiments revealed high sorption capacities (up to 147 mg·g^{−1}) and were best described by the Langmuir isotherm and pseudo-second-order kinetic model, indicating chemisorption as the rate-limiting step. FTIR and SEM analyses demonstrated the formation of nickel and copper hydroxide/oxide phases, confirming surface precipitation mechanisms. Subsequent thermal treatment produced NiO- and CuO-enriched oxide systems with photocatalytic and antibacterial potential, while hydrometallurgical recovery using ammonia solutions achieved desorption efficiencies of 90–97%. The results highlight the dual role of SFS as an efficient sorbent for wastewater pre-treatment and as a secondary source of valuable metals, contributing to sustainable materials management and circular economy goals.

Keywords: circular economy; steelmaking slag; heavy metals; adsorption; nickel; copper; metal recovery



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1. Introduction

The global transition toward a circular economy (CE) demands innovative methods to convert industrial by-products into valuable secondary raw materials. Within this framework, the metallurgical sector plays a key role because it generates vast volumes of solid residues that can be recirculated into production cycles. Steelmaking slag, a heterogeneous by-product produced during refining of molten steel, represents one of the most abundant inorganic wastes globally [1–3]. Annual generation of more than 200 million tons of slag worldwide introduces both challenges and opportunities; improper disposal may cause soil alkalization and heavy-metal leaching, yet the same material possesses mineralogical and chemical properties suitable for environmental remediation [4].

The valorization of steelmaking slag supports multiple sustainable development goals (SDGs), especially those targeting responsible consumption, clean water, and innovation.

Its use as a reactive sorbent fits well within industrial symbiosis models, where waste from one process becomes feedstock for another [5]. In the water-treatment context, the increasing load of toxic metals—nickel and copper among them—has spurred the search for inexpensive sorbents with high removal efficiencies [6,7]. Both Ni and Cu occur in effluents from electroplating, mining, alloy manufacturing, and fertilizer industries, posing ecological and health risks even at trace concentrations.

Traditional removal technologies such as precipitation, ion exchange, and membrane filtration often require costly reagents and generate secondary sludge [8–10]. Consequently, adsorption using industrial residues has become an attractive, sustainable solution. Various sorbents have been explored, including biochars [11,12], clays [13], zeolites, and synthetic minerals [14]. Among these, steelmaking slag (SFS) offers distinct advantages: strong alkalinity, abundance of Ca–Fe–Si–Mg oxides, high surface heterogeneity, and negligible cost.

Recent studies have confirmed the efficacy of steelmaking furnace (SFS) and electric-arc-furnace slags for immobilizing Cd, Pb, Cr, and Ni through surface complexation and precipitation [15–20]. However, despite numerous investigations, comparative analysis of Ni(II) and Cu(II) adsorption mechanisms on SFS remains limited, particularly under conditions representative of real industrial wastewaters. Moreover, integrating this process into circular-economy frameworks—where spent slag enriched with heavy metals could be reprocessed into value-added oxide materials—has received less attention.

This study, therefore, explores the dual role of SFS: first, as a low-cost sorbent for Ni(II) and Cu(II) removal from aqueous solutions; second, as a precursor for secondary materials usable in photocatalytic or antibacterial applications after metal recovery. By linking wastewater treatment with resource recovery, the work illustrates how metallurgical by-products can contribute to closed-loop material flows and reduced environmental burdens [21,22].

Beyond demonstrating adsorption capacity, the research seeks to elucidate the fundamental interaction mechanisms, model kinetic and isotherm parameters, and evaluate the recyclability of slag as a regenerable sorbent. Through this, the study advances the concept of “waste-to-resource valorization,” providing a bridge between environmental engineering and metallurgical process design [23–25].

This study introduces a novel circular-economy approach to the valorization of SFS. Unlike previous works focusing only on heavy-metal removal, this research integrates adsorption, metal recovery, and material reuse into a single process. The dual-function strategy enables both the purification of Ni(II)- and Cu(II)-containing wastewaters and the generation of reusable oxide products. The findings provide new insight into sorption mechanisms and demonstrate a practical pathway for transforming metallurgical waste into valuable secondary resources.

2. Materials and Methods

Stock solutions of Ni(II) and Cu(II) ions with concentrations of $1000 \text{ mg} \cdot \text{dm}^{-3}$ were prepared by dissolving $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ and $\text{Cu}(\text{NO}_3)_2 \cdot 3\text{H}_2\text{O}$ in deionized water. Solutions of varying concentrations ($50\text{--}900 \text{ mg} \cdot \text{dm}^{-3}$) were obtained through dilution.

In this work, steelmaking furnace slag (SFS) originating from the Ostrava industrial agglomeration was used. The slag was ground using a RETSCH epicyclic ball mill and sieved below 0.1 mm , and used as an adsorbent. For adsorption experiments, $0.1\text{--}1 \text{ g}$ of SFS was placed into plastic bottles with 50 cm^3 of metal ion solution with the initial concentrations of 500 mg dm^{-3} and stirred for $0.5\text{--}24 \text{ h}$. After filtration, the products were referred to as SFS + Ni and SFS + Cu. For desorption experiments, 0.1 g of SFS-Ni or SFS-Cu sample was mixed with various amounts ($10\text{--}100 \text{ cm}^3$) of aqueous ammonia solution. The resulting suspensions were stirred for 24 h and then filtered.

Metal concentrations in slag and leachates were analyzed using AAS (Varian AA280FS, Varian Australia Pty Ltd., Mulgrave, Victoria, Australia). Surface area was measured by nitrogen physisorption on a Thermo Finnigan Sorptomatic 1990 Series (Thermo Fisher, Waltham, MA, USA).

Thermal analyses were conducted on a TA Instruments Discovery SDT 650 (TA Instruments, New Castle, DE, USA) with autosampler, heating rate 5 °C/min, in an air atmosphere, and a sample weight of 20 mg.

FTIR spectra of all samples were recorded on a ThermoScientific Nicolet iS50 spectrometer (Thermo Fisher, Waltham, MA, USA) with a DTGS detector in the 4000–400 cm^{−1} region using KBr pellets.

X-ray diffraction (XRD) analysis was performed using a MiniFlex600 diffractometer equipped with a Co tube (Rigaku Corp., Tokyo, Japan) and D/teX Ultra 250 detectors (Rigaku Corporation, Tokyo, Japan). The powder samples were pressed in a rotational holder, and the patterns were recorded in the range of 10–90° 2Theta, with a step of 0.2° and a scan speed of 3° min^{−1}. The registered patterns were evaluated using SmartLab Studio II, version 2019, PDF2 database, release 2019 (ICSD, USA), which was used for phase identification.

The chemical composition of slag was determined with a Rigaku SuperMini200 wave (Rigaku Corporation, Tokyo, Japan) dispersive fluorescence spectrometer equipped with a 200 W Pd X-ray tube (Rigaku Corporation, City: Tokyo, Country: Japan) as the source. The powdered samples (4 g) were homogenized with wax (0.9 g) and subsequently pressed into pellets of diameter 32 mm using a manual hydraulic press with an applied load of 10 tons. The standard less method was used for quantification of the elements' content.

For morphological and microstructural analyses of slag samples, a Thermo Fisher Scientific FEI Quanta-650 FEG autoemission electron microscope (Thermo Fisher, Waltham, MA, USA) was used, equipped with analyzers: energy dispersive analyzer (EDS)-EDAX Octane plus (EDAX Inc., Mahwah, NJ, USA), wave dispersive analyzer (WDA)-EDAX LEXS (EDAX Inc., Mahwah, NJ, USA). The microscope worked under the following conditions: voltage 20 kV, current 8–10 nA, beam diameter 6 µm, reduced vacuum with chamber pressure 50 Pa, and samples without plating.

Adsorption isotherms were fitted using the Freundlich and Langmuir models, while kinetic data were analyzed by pseudo-first- and pseudo-second-order equations.

3. Results and Discussion

3.1. Chemical and Structural Characteristics of the Slag

Elemental analysis (Table 1) confirmed that the slag consisted mainly of CaO (21.8%), Fe₂O₃ (28.3%), SiO₂ (6.7%), and MgO (4.1%), with minor constituents of Al₂O₃ and MnO. Such a composition is consistent with SFSs reported elsewhere [2,3,19].

Table 1. Chemical composition of the slag expressed in the form of oxides (wt %).

Ca	Mg	Fe	Mn	Al	Si	Zn	Cr	Pb	Cd
21.8	4.1	28.3	4.4	1.4	6.7	0.10	0.28	0.028	0.0014

The materials' alkaline character (pH 11.6) arises from free lime and periclase phases, which enhance their reactivity in aqueous media. The measured specific surface area of 6 m²/g, though modest, is compensated by abundant reactive sites and the porous microtexture revealed by SEM micrographs.

XRD patterns (Figure 1) confirmed the presence of larnite (Ca₂SiO₄), mayenite (Ca₁₂Al₁₄O₃₃), magnetite (Fe₃O₄), and wüstite (FeO), indicating a heterogeneous crystalline

matrix favourable for both ion exchange and surface precipitation. These mineral phases can undergo partial dissolution and reprecipitation under aqueous conditions, producing active hydroxylated surfaces that strongly interact with transition-metal ions [26,27].

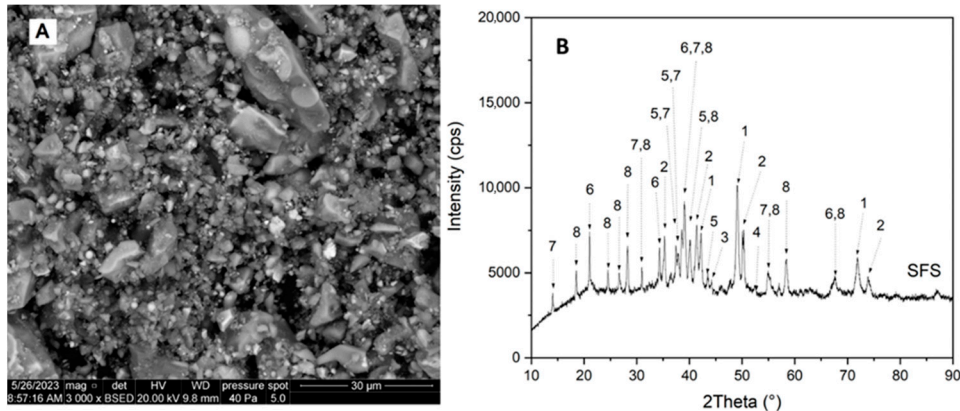


Figure 1. SEM analysis (A) and XRD pattern (B) of the SFS. 1—FeO Wustite, 2—Fe₃O₄ Magnetite, 3—CaO Lime, 4—MgO Periclase, 5—Ca₂(SiO₄) Larnite, 6—Ca₁₂Al₁₄O₃₃ Mayenite, 7—Ca₂FeAlO₅ Brownmillerite, 8—Ca(Fe_{0.77}Mg_{0.22})(SiO₄) Kirschsteinite.

3.2. Adsorption Capacity and Modelling

The maximum sorption capacities achieved—approximately 147 mg·g^{−1} for both Ni(II) and Cu(II)—are competitive with those of other low-cost adsorbents such as modified biochar or synthetic hydroxyapatites [11–13]. The results of adsorption measurements are summarized in Table 2. The equilibrium data fitted the Langmuir isotherm most accurately, suggesting a monolayer coverage of uniform adsorption sites. This observation aligns with the hypothesis of chemisorptive bonding involving hydroxyl and carbonate groups on the slag surface.

Table 2. Parameter values of Langmuir and Freundlich isotherms for Cu(II) and Ni(II) adsorption.

Ion	Langmuir Isotherm			Freundlich Isotherm		
	q _{max} [mg·g ^{−1}]	b [dm ³ ·mg ^{−1}]	R ²	K	n	R ²
Cu(II)	146.57	0.0385	0.9903	15.001	2.594	0.6582
Ni(II)	147.23	0.0249	0.9989	63.156	7.519	0.5341

The results of kinetic measurements are summarized in Table 3. The kinetic behaviour followed the pseudo-second-order model, consistent with chemical interaction being the rate-determining step rather than external mass transfer (Figure 2).

Table 3. Results of kinetic data analyses.

Ion	Pseudo-First Order		Pseudo-Second Order	
	K ₁ [min ^{−1}]	R ²	K ₂ [g·mg·min ^{−1}]	R ²
Cu(II)	3.00·10 ^{−4}	0.6345	3.86·10 ^{−5}	0.9836
Ni(II)	7.00·10 ^{−4}	0.9480	6.21·10 ^{−5}	0.9932

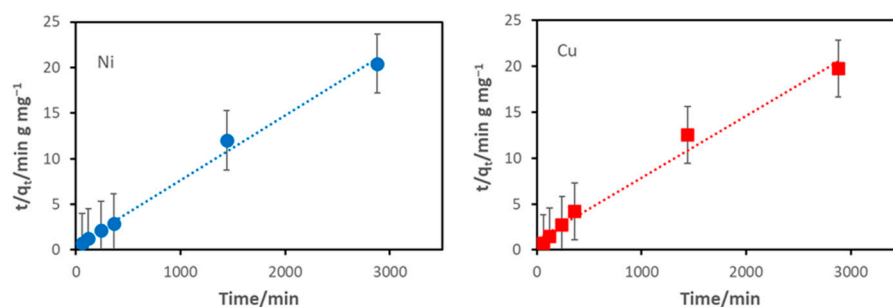


Figure 2. Application of the pseudo-second order sorption model to the kinetic data for 0.1 g of the slag and 50 cm^{−3} of solutions of Ni(II) and Cu(II) ions with the initial concentrations of 500 mg dm^{−3}.

The relatively fast adsorption (≤ 60 min to equilibrium) implies strong electrostatic attraction and surface complexation between Ni²⁺/Cu²⁺ ions and the reactive Ca–Fe–O matrix [28,29].

When compared with other metallurgical residues, SFS exhibits superior performance: for instance, blast-furnace slag typically shows 90–120 mg·g^{−1} capacity for Ni²⁺ [2,20], while untreated fly ash rarely exceeds 40 mg·g^{−1} [22]. The slightly higher uptake of Cu²⁺ relative to Ni²⁺ can be attributed to its lower hydration radius and higher affinity toward oxygen donor ligands, confirming selective adsorption behaviour [4,5].

3.3. Surface Mechanisms and Spectroscopic Evidence

FTIR spectra (Figure 3) elucidate the principal functional groups involved. The emergence of bands at 3543 cm^{−1} and 3693 cm^{−1} corresponds to –OH stretching vibrations, indicative of hydroxylated species. The broad bands between 3600 cm^{−1} and 3000 cm^{−1} in the spectra of SFS + Ni and SFS + Cu samples correspond to physically bound water [26,27]. Carbonate vibrations at 1440 cm^{−1}, 875 cm^{−1}, and 714 cm^{−1} suggest precipitation of metal carbonates on the slag surface, while strong nitrate bands around 1385 cm^{−1} confirm partial adsorption of nitrate anions associated with the original metal salts. The bands near 570 cm^{−1} and 512 cm^{−1} correspond to Fe–O lattice vibrations [30,31], implying the coexistence of iron oxides in the slag matrix before and after sorption.

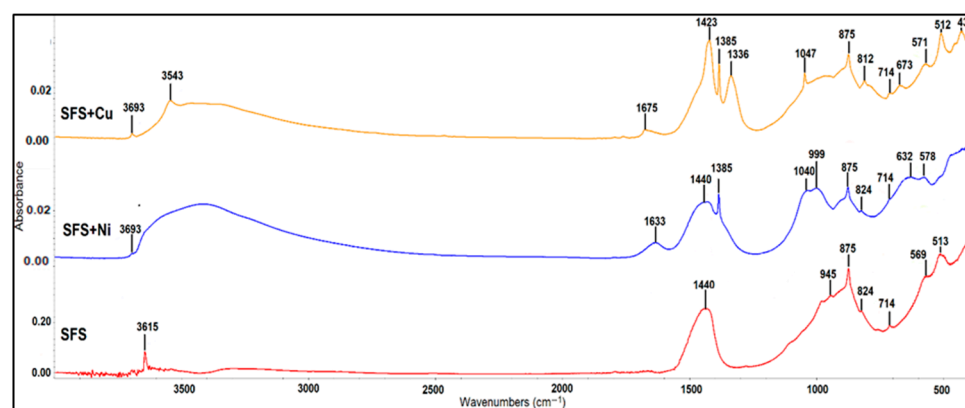


Figure 3. FTIR spectra of the slag before and after Ni(II) and Cu(II) sorption.

The spectra do not reveal any distinct shifts or new bands that would indicate surface complexation between the metal ions and functional groups of the slag. Considering the highly alkaline environment (pH > 11), it is likely that the removal of Ni(II) and Cu(II) from aqueous solutions occurs dominantly by surface precipitation of hydroxide/oxide and carbonate phases. Such precipitation-driven immobilization mechanisms are typical for strongly basic metallurgical residues and have been reported for similar alkaline systems [20,23].

3.4. Morphological and Structural Transformations

SEM images (Figures 4 and 5) reveal a distinct morphological evolution of the steel-making slag following metal sorption. Micrographs were obtained using an auto emission scanning electron microscope at $12,000\times$ magnification. The pristine slag surface exhibits an irregular and compact granular texture (Figure 1), whereas the Ni(II)- and Cu(II)-loaded samples show finer, aggregated particles and locally developed needle-like or flaky morphologies typical of hydroxide and oxide phases. These morphological features suggest that surface precipitation occurred during the sorption process. Ion exchange at the Ca-Si-Fe reactive sites cannot be ruled out either.

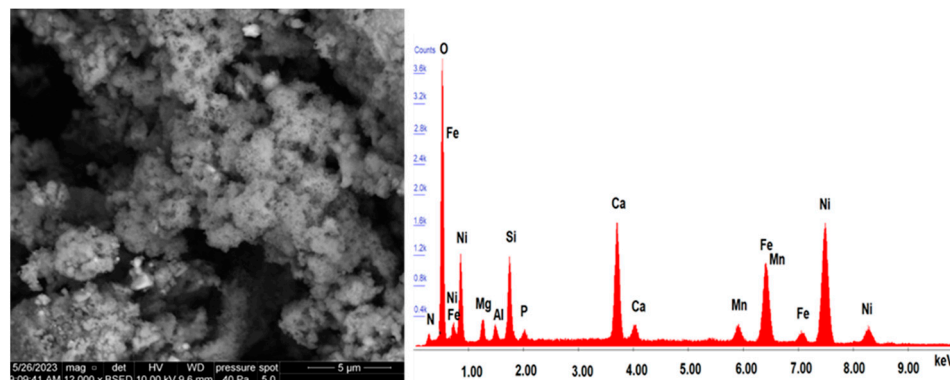


Figure 4. SEM analysis of the steelmaking slag after Ni sorption.

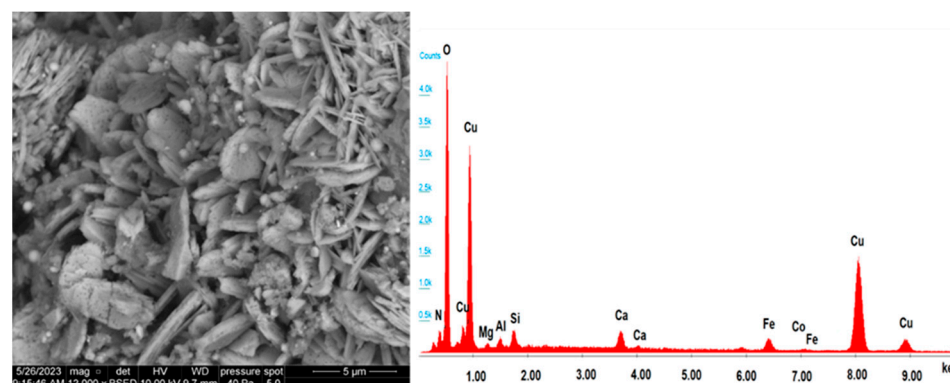


Figure 5. SEM analysis of the steelmaking slag after Cu sorption.

Comparison of XRD patterns of slags after Cu(II) and Ni(II) sorption (Figure 6) confirms structural modifications accompanying Ni and Cu incorporation. Both samples retain the dominant crystalline phases typical of steelmaking slag—wüstite (FeO), magnetite (Fe_3O_4), lime (CaO), periclase (MgO), larnite (Ca_2SiO_4), mayenite ($\text{Ca}_{12}\text{Al}_{14}\text{O}_{33}$), brownmillerite ($\text{Ca}_2\text{FeAlO}_5$), and kirschsteinite ($\text{Ca}(\text{Fe}_{0.77}\text{Mg}_{0.22})(\text{SiO}_4)$).

In addition to these dominant phases, additional reflections corresponding to $\text{Ni}_2(\text{CO}_3)(\text{OH})_2$ (nickel carbonate hydroxide) appear in the nickel-containing sample, confirming the formation of nickel-containing hydroxide and hydroxycarbonate phases. In contrast, the Cu-loaded sample exhibits weak but distinct peaks assignable to $\text{FeO}(\text{OH})$ (lepidocrocite), suggesting secondary Fe–Cu interactions during sorption.

The appearance of these secondary phases indicates the partial oxidation and precipitation of sorbed metals on reactive Ca–Si–Fe–Mg sites, consistent with the FTIR and SEM findings.

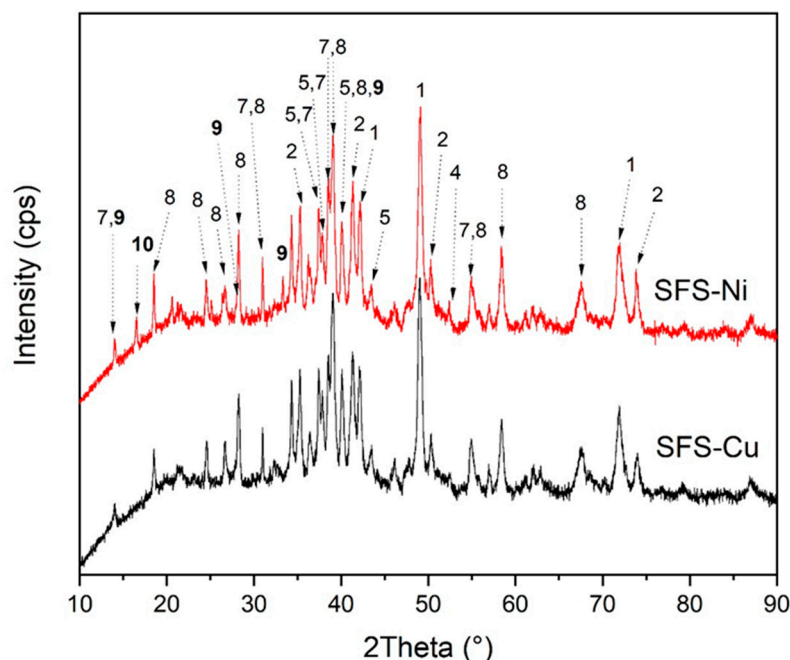
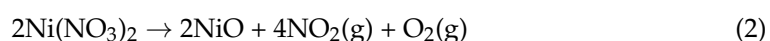
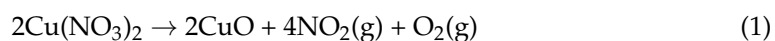


Figure 6. Comparison of X-ray diffraction patterns of SFS-Ni and SFS-Cu samples. 1—FeO Wustite, 2—Fe₃O₄ Magnetite, 3—CaO Lime, 4—MgO Periclase, 5—Ca₂(SiO₄) Larnite, 6—Ca₁₂Al₁₄O₃₃ Mayenite, 7—Ca₂FeAlO₅ Brownmillerite, 8—Ca(Fe_{0.77}Mg_{0.22})(SiO₄) Kirschsteinite, 9—Ni₂(CO₃)(OH)₂ Nickel Carbonate Hydroxide, 10—FeO(OH) Lepidocrocite.

These structural modifications not only immobilize Ni and Cu but also alter the slags' physicochemical properties, generating composite oxide–hydroxide systems with enhanced reactivity. These metal-bearing phases may serve as precursors for functional oxide materials, particularly NiO and CuO nanoparticles, known for their photocatalytic and antibacterial activities [24,32–34].

3.5. Thermal Behaviour and Metal Recovery

Thermogravimetric analysis (Figure 7) revealed distinct mass changes associated with the thermal decomposition and oxidation processes occurring in the original and metal-loaded slags. In all samples, an initial minor mass loss below 200 °C corresponds to the desorption of physically adsorbed water and dehydration of surface hydroxyl groups. Between approximately 250 °C and 400 °C, additional weight losses are observed in the Ni- and Cu-modified slags. These losses can be attributed primarily to the decomposition of nickel and copper hydroxides and hydroxycarbonates into the corresponding metal oxides. This weight loss is secondarily contributed to by the thermal decomposition of residual nitrates that remained on the surface after adsorption, and whose presence was also demonstrated by FTIR analysis. The following Equations (1) and (2) illustrate the stoichiometry of oxide formation from nitrate precursors, although the actual nitrate content in the samples is expected to be very low:



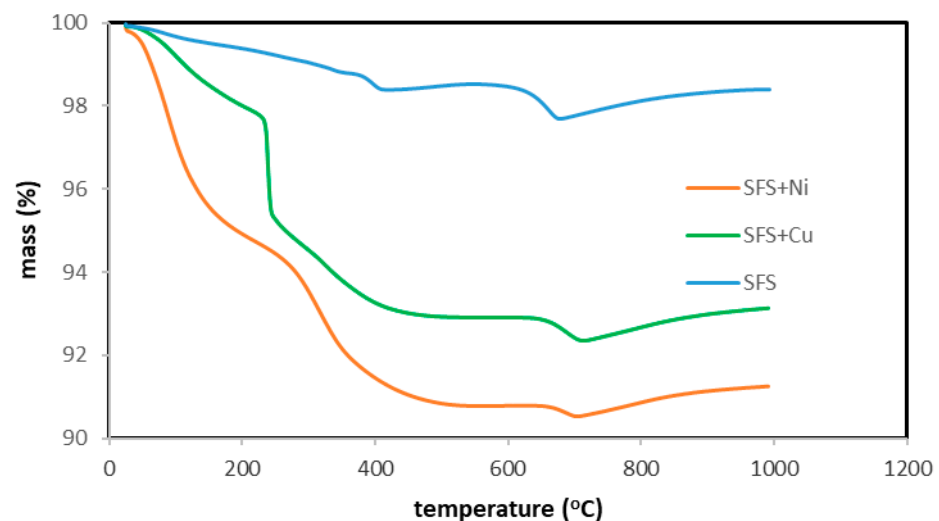


Figure 7. Comparison of TG records of original slag and slag after Ni(II) and Cu(II) sorption.

At temperatures above 600 °C, the TG curves of Ni- and Cu-loaded slags diverge significantly from that of the pristine slag. A gradual mass increase starting around 750–800 °C is evident, particularly for the Cu-enriched sample. This gain in mass can be attributed to oxidation reactions involving residual Fe^{2+} -bearing phases (such as wüstite, FeO) and Ni^{2+} species, leading to the formation of higher-valence oxides such as magnetite (Fe_3O_4), hematite (Fe_2O_3), or nickel ferrite (NiFe_2O_4). These processes consume atmospheric oxygen during heating, resulting in a net mass increase.

Additionally, partial recrystallization and structural rearrangement of existing silicate phases (e.g., larnite and kirschsteinite) may occur at elevated temperatures. These transformations reflect thermally driven stabilization of the silicate matrix rather than oxidation processes, as the relevant phases are already present in the original slag. The simultaneously occurring processes, oxidation of residual phases containing Fe^{2+} and phase stabilization of the silicate matrix, thus explain the overall upward trend of the TG curve above 800 °C.

Such thermal transformations are beneficial for pyrometallurgical valorisation, as they yield stable oxide systems—predominantly $\text{NiO-CuO-Fe}_2\text{O}_3$ composites—with potential functional properties for use in ceramics, coatings, and heterogeneous catalysts [32–34]. Moreover, the conversion of metal-loaded slags into crystalline mixed oxides facilitates their safe handling, minimizes the leachability of heavy metals, and enables integration into the circular recovery chain for advanced material production.

Hydrometallurgical recovery using 24% aqueous ammonia achieved desorption efficiencies of ~97% for Ni and ~90% for Cu (Figure 8). During the hydrometallurgical recovery stage, the dissolution of Ni(II) and Cu(II) precipitates proceeds via complexation with aqueous ammonia. Ammonia functions both as a ligand and as a weak base, forming highly stable ammine complexes such as $[\text{Ni}(\text{NH}_3)_6]^{2+}$ and $[\text{Cu}(\text{NH}_3)_4]^{2+}$, which promote effective desorption of the metals from the slag matrix. This mechanism facilitates nearly complete metal recovery but simultaneously increases the overall ammonia demand, especially in systems containing abundant hydroxide and carbonate species that buffer the solution and consume part of the reagent. The resulting ammonium-rich leachates require careful post-treatment to prevent secondary contamination. Typical approaches include air stripping or acid neutralization to remove free ammonia, followed by distillation or membrane-assisted recovery for reagent recycling [30,31]. Integration of such recovery steps into the process not only minimizes nitrogen emissions but also enhances the circularity of the system, aligning the hydrometallurgical route with sustainable resource-management principles.

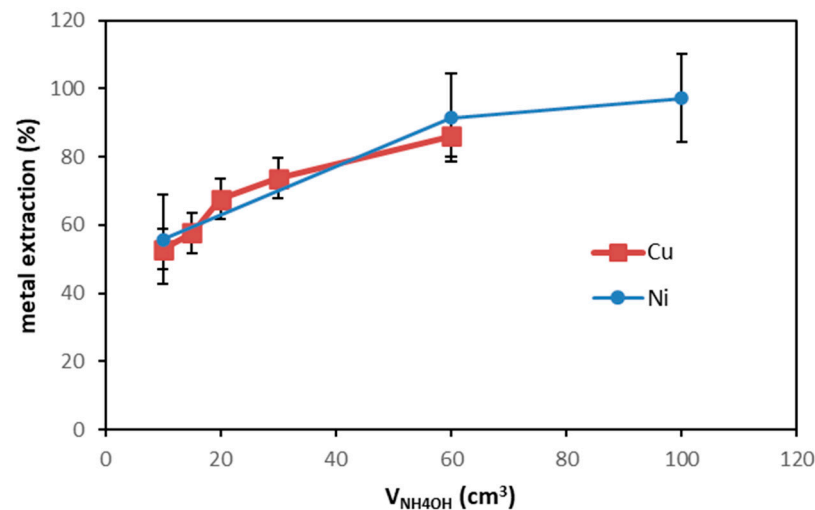


Figure 8. Recovery of copper and nickel from the slag.

The efficiency suggests that adsorption is largely reversible and that the sorbent can be reused in multiple cycles with minimal loss of capacity. Comparable findings were reported by Xue et al. [19] for multi-metal sorption on SFS. The near-complete recovery of metals confirms the feasibility of closed-loop utilization—capturing contaminants and subsequently recovering them as valuable raw materials.

The recovered Ni and Cu oxides could be integrated into the production of photocatalytic coatings, antibacterial composites, or energy materials, thereby establishing a circular materials pathway consistent with the principles of industrial ecology [5,25].

3.6. Environmental and Economic Implications

The presented process offers a dual environmental benefit: remediation of metal-contaminated waters and reduction in metallurgical waste volumes. In terms of a life-cycle perspective, valorising SFS as a sorbent offsets raw material extraction and reduces carbon emissions associated with cement or limestone production [3,21].

Economically, the near-zero cost of slag combined with its regenerability positions it as a viable material for decentralized wastewater treatment in metallurgical and electroplating industries. Future scaling may focus on granulated or pelletized slag forms to improve hydraulic performance in continuous-flow reactors. Moreover, the potential substitution of natural minerals with industrial residues supports resource efficiency and aligns with the EU Circular Economy Action Plan (2020), emphasizing waste valorisation and secondary raw-material markets [35,36].

Despite its high removal efficiency, the adsorption performance of steelmaking slag is limited by several operational and environmental factors. The strong alkalinity of the material ($\text{pH} > 11$) can elevate the final pH of treated effluents above regulatory limits, necessitating subsequent neutralization steps before discharge or reuse. Moreover, the high calcium content may cause secondary precipitation of hydroxides, reducing the sorbent's efficiency in continuous systems. From an energy and cost perspective, the thermal treatment required to convert metal-loaded slag into oxide products, as well as the aqueous regeneration with ammonia, both contribute to the overall process footprint. These steps demand additional energy input and chemical consumption, which must be balanced against the environmental benefits of waste valorisation to ensure that the process remains economically and ecologically sustainable.

4. Conclusions

This study provides comprehensive evidence that steelmaking furnace slag (SFS) can serve as an efficient and sustainable adsorbent for the removal of Ni(II) and Cu(II) ions from aqueous solutions. The materials' intrinsic alkalinity and multi-oxide composition promote surface precipitation mechanisms, yielding high sorption capacities ($\sim 147 \text{ mg}\cdot\text{g}^{-1}$) and rapid kinetics consistent with chemisorption.

Analyses (FTIR, SEM, and XRD) verified the formation of hydroxide/oxide and carbonate phases. Subsequent thermal and hydrometallurgical treatments demonstrated the feasibility of recovering Ni and Cu as oxide products, enabling their reuse in photocatalytic or antibacterial applications.

The results thus integrate wastewater purification and resource recovery into a single process, exemplifying the principles of circular economy and industrial symbiosis. Compared with conventional sorbents, steelmaking slag provides a low-cost, abundant, and easily regenerable alternative with minimal environmental burden.

From a practical perspective, some limitations remain—mainly related to high alkalinity and potential leaching of minor trace elements, which may necessitate pH adjustment or pre-treatment before field application. Future research should therefore investigate long-term stability under dynamic conditions and the potential of surface modification (e.g., carbonation, phosphate treatment) to enhance selectivity and safety.

Overall, this work highlights the valorization potential of metallurgical by-products, promoting a shift from linear waste management to circular, resource-efficient solutions consistent with modern sustainability goals.

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