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Literature Review: Iron Extraction From Copper Smelting Slag And Direct Reduced Iron (Dri/Hbi) Production

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Abstract: Studies on anode slime processing further highlight the potential of pressure leaching for near-complete nickel extraction under optimized acidic and oxidizing conditions. Overall, the literature indicates that well-designed hybrid systems offer the most effective balance of selectivity, energy efficiency, and environmental sustainability for nickel recovery. Copper smelting slag represents a significant environmental challenge, with accumulated quantities exceeding 1.8 billion tonnes globally. This slag contains 35–45 wt% iron primarily in the form of fayalite (Fe_2SiO_4) and magnetite (Fe_3O_4), making it an attractive secondary source for iron recovery. The present literature review synthesizes recent research (2013–2025) from Scopus and Web of Science indexed journals on iron extraction from copper smelting slag and its subsequent conversion to direct reduced iron (DRI) and hot briquetted iron (HBI) for steelmaking applications. Major iron recovery methods include coal-based direct reduction combined with magnetic separation (achieving 91–98% iron recovery), hydrogen-based reduction, oxidation-roasting magnetic separation, carbothermal reduction, and hydrometallurgical processing. Current findings demonstrate that reduction temperature (1200–1300°C), reductant type, slag basicity adjustment, and additive incorporation critically influence recovery efficiency. Hydrogen-based direct reduction emerges as the most promising technology for sustainable iron recovery, achieving superior metallization degrees (94–95%) while enabling near-zero CO_2 emissions when coupled with renewable electricity. Global DRI/HBI production is projected to increase from 115 Mt/year (2019) to 212.6 Mt/year (2050), with hydrogen-based production dominating future production. This review synthesizes optimal process parameters, kinetic mechanisms, thermodynamic principles, and technological innovations essential for advancing sustainable iron recovery and low-carbon steelmaking.

Keywords — copper slag, iron extraction, direct reduction, DRI, HBI, fayalite reduction, magnetic separation, steelmaking

1. INTRODUCTION

The production of copper through pyrometallurgical smelting generates substantial quantities of slag as a co-product. Globally, the copper smelting industry produces approximately 2.2–3.0 tonnes of slag per tonne of copper produced, resulting in accumulated slag exceeding 1.8 billion tonnes, particularly in China [1]. This slag represents both an environmental challenge and an underutilized resource, as it contains significant iron content (35–45 wt% total iron), copper residues (0.3–0.7 wt%), and potentially hazardous elements (lead, zinc, arsenic) [6].

The mineralogical composition of copper smelting slag differs fundamentally from other industrial slags. Unlike blast furnace or electric arc furnace slags dominated by calcium silicates, copper slag exhibits high silica content (30–40 wt% SiO_2) and contains iron predominantly as fayalite (Fe_2SiO_4), a complex iron-silicate phase, alongside magnetite (Fe_3O_4) and hematite (Fe_2O_3) [50]. This unique mineralogy necessitates specialized reduction technologies distinct from conventional iron ore processing [2].

The global steel industry faces mounting pressure to reduce carbon emissions and transition toward sustainable, low-carbon production pathways. Direct reduced iron (DRI), produced through controlled reduction of iron oxides at temperatures below the iron melting point (1538°C), has emerged as a critical intermediate product enabling this transition. DRI is subsequently processed into hot briquetted iron (HBI), a compacted form facilitating transportation and storage. Global DRI production reached approximately 115 million tonnes in 2019, with projections indicating growth to 157.3 million tonnes by 2030 and 212.6 million tonnes by 2050, predominantly through hydrogen-based reduction technologies [13].

The valorization of copper slag through iron recovery offers strategic advantages including: (1) secondary iron resource development; (2) environmental remediation and waste minimization; (3) reduction of dependency on virgin iron ore extraction; and (4) potential recovery of valuable secondary metals (copper, lead, zinc) through integrated processing routes [136]. Recent technological advances in coal-based reduction, hydrogen-based reduction, magnetic separation, and hydrometallurgical processing have substantially improved iron recovery efficiency and product quality [3][12].

This literature review synthesizes current understanding of copper slag iron extraction mechanisms, optimal process parameters, reduction kinetics, thermodynamic principles, and industrial applications derived from approximately 50 peer-reviewed articles indexed in Scopus and Web of Science databases [4].

2. MATERIALS AND METHODS

2.1 Copper Slag Mineralogical Composition

Copper smelting slag exhibits distinctive mineralogical features reflecting the copper extraction process. The dominant iron-bearing phases include fayalite (Fe_2SiO_4), which typically accounts for 35–50 wt% of slag composition; magnetite (Fe_3O_4), present at 5–15 wt%; and minor hematite (Fe_2O_3). Secondary phases include amorphous silica, calcium silicates, and trace sulfide phases containing residual copper and precious metals [15].

X-ray diffraction (XRD) analysis characteristically reveals strong diffraction peaks corresponding to fayalite (d-spacing: 2.43 Å, 1.57 Å) and magnetite (d-spacing: 2.96 Å, 1.61 Å). Scanning electron microscopy with energy-dispersive spectroscopy (SEM-EDS) reveals fayalite as a continuous silicate phase matrix hosting metallic copper particles, copper sulfides, and magnetite crystals. The glassy or crystalline nature of slag depends on cooling conditions: rapidly water-cooled slag exhibits predominantly amorphous structure with dispersed crystalline phases, while slow-cooled slag develops well-crystallized mineral phases [5].

2.2 Chemical Composition

Typical copper slag contains 35–45 wt% total iron (distributed as FeO in fayalite: ~32–40 wt% and magnetite/hematite: ~3–5 wt%), 30–38 wt% SiO_2, 0.2–2 wt% CaO, 0–5 wt% Al_2O_3, 0–8 wt% MgO, and trace elements including copper (0.3–0.7 wt%), lead (0.5–1.2 wt%), zinc (1.5–2.5 wt%), and potentially hazardous arsenic and antimony [1][6][12]. The silicate-rich composition and low basicity (CaO/SiO_2 ratio typically <0.1) distinguish copper slag from iron- and steelmaking slags [6].

2.3 Environmental Significance

The accumulation of copper slag represents a significant environmental challenge [7]. Leaching studies demonstrate that copper slag exhibits potential for environmental contamination through release of copper (0.1–5 mg/L), lead (0.01–2 mg/L), zinc (0.5–15 mg/L), and arsenic under acidic conditions. Consequently, sustainable valorization of copper slag through iron recovery presents an opportunity for simultaneous environmental remediation and resource conservation.

3. RESULTS

3.1 Iron Extraction Methods from Copper Slag

3.1.1 Coal-Based Direct Reduction with Magnetic Separation

Coal-based direct reduction represents the most extensively studied and industrially implemented iron recovery technology. The process comprises three sequential stages: (1) slag preparation and pelletization; (2) carbothermal reduction roasting at 1200–1350°C; and (3) magnetic separation of reduced iron [1].

During carbothermal reduction, carbon and carbon monoxide reduce iron oxides according to:

$Fe_2SiO_4 + 4C \rightarrow 2Fe + SiO_2 + 4CO$ (Prim. fayalite reduct.)

$Fe_3O_4 + 4C \rightarrow 3Fe + 4CO$ $FeO + C \rightarrow Fe + CO$

Thermodynamic analysis indicates that fayalite reduction becomes favorable above approximately 900°C, with equilibrium conversion approaching 95% at 1200°C and exceeding 98% at 1300°C [8]. The addition of CaO flux (15–25 wt%) modifies slag basicity, promoting slag fluidity and metallic iron grain aggregation through formation of low-melting phases including dicalcium silicate ($2CaO\cdot SiO_2$) [50].

Under optimized conditions (1300°C, 30 min reduction, 35 wt% coal, 20 wt% CaO addition), coal-based direct reduction achieves iron recovery rates of 91–98.13%, with iron grades in the magnetic concentrate reaching 90–96% [1]. The recovered metallic iron exhibits particle sizes predominantly >50 μm, enabling efficient subsequent magnetic separation at field intensities of 60–80 $kA\cdot m^{-1}$ [9].

3.1.2 Oxidation-Roasting Followed by Magnetic Separation

An alternative processing route employs sequential oxidation and reduction roasting to enhance iron phase crystallinity and magnetic properties [10]. Oxidation roasting at 600–800°C converts fayalite to hematite and amorphous silica, promoting phase segregation. Subsequent reduction roasting converts iron oxides predominantly to magnetite (Fe_3O_4), which exhibits significantly enhanced magnetic susceptibility compared to hematite [11][15].

This two-stage approach achieves iron recovery rates of 78–80% with iron grades of 78.87% in the concentrate. While recovery efficiency remains lower than coal-based direct reduction, oxidation-roasting magnetic separation offers advantages including lower process temperature and reduced carbon consumption, making it suitable for slag samples where carbon-based reduction proves technically challenging [12].

3.1.3 Hydrogen-Based Direct Reduction

Hydrogen-based reduction represents the most promising future pathway for sustainable iron recovery. Pure hydrogen reduction eliminates CO_2 generation, producing only water vapor as a byproduct:

$Fe_2SiO_4 + 4H_2 \rightarrow 2Fe + SiO_2 + 4H_2O$ $Fe_3O_4 + 4H_2 \rightarrow 3Fe + 4H_2O$

Recent studies demonstrate that hydrogen reduction of copper slag achieves metallization degrees of 85–95% at reduction temperatures of 800–950°C, with activation energies ranging from 29.1–36.1 kJ/mol—substantially lower than coal-based systems [35]. The unreacted shrinking core model indicates that hydrogen-based reduction is controlled by combined effects of internal diffusion and chemical reaction, with internal diffusion becoming predominant as reduction progresses [13].

Hydrogen-based DRI exhibits superior product quality compared to natural gas-derived DRI, achieving metallization degrees of 94–95% and carbon contents typically <1.5 wt%, enabling production of high-purity HBI with density >5.0 g/cm³ meeting international maritime transport standards [13][36].

3.1.4 Carbothermal Reduction with Additives

Recent research emphasizes the importance of additive incorporation during carbothermal reduction to enhance iron grain growth, aggregation, and ultimate recovery efficiency. Addition of compounds including sodium carbonate (Na_2CO_3), calcium carbonate ($CaCO_3$), and compound additives promotes nucleation during reduction, enhances metallic iron grain migration and coalescence, and facilitates slag-metal separation [14].

The compound additive mechanism involves formation of nucleation sites promoting iron grain growth through interfacial energy minimization [13][37]. Addition of 3–5 wt% compound additive during pelletization followed by reduction at 1250°C increases iron recovery by 5–8% compared to unadditized processes [15][38].

3.1.5 Hydrometallurgical Processing

Hydrometallurgical methods employ acid or base leaching to selectively extract iron from copper slag. While less commonly employed than pyrometallurgical methods, hydrometallurgical processing offers lower temperature requirements and potential for selective metal extraction [16].

Sulfuric acid leaching with concentration 200–300 g/L at 50°C for 2–3 hours achieves copper recovery exceeding 95% but generates iron-rich residues requiring separate processing. Alkaline leaching with ammonia solution shows promise for selective copper extraction while maintaining iron in solid residue for subsequent magnetic separation [17][39]. Organic acid leaching (including L-ascorbic acid) demonstrates feasibility for simultaneous recovery of lead (68–88%), copper (up to 99.1%), and iron (99.2%) under optimized conditions, though at higher chemical cost [18].

3.2 Reduction Kinetics and Thermodynamic Analysis

3.2.1 Temperature Dependence

Temperature exerts the most significant influence on reduction efficiency and recovery rate. Coal-based copper slag reduction exhibits nonlinear temperature dependence:

- 1050°C: ~35% reduction degree
- 1200°C: ~70% reduction degree
- 1300°C: ~85% reduction degree
- 1350°C: >92% reduction degree

This temperature dependence reflects changes in rate-controlling mechanisms [19]. At lower temperatures (1050–1150°C), the reduction process is controlled by interfacial chemical reaction between carbon powder and slag and the Boudouard reaction generating CO. At intermediate temperatures (1150–1300°C), mixed control involving both interfacial reaction and gaseous diffusion occurs. At elevated temperatures (>1300°C), CO diffusion through the slag pellet becomes rate-limiting [1].

3.2.2 Kinetic Models and Activation Energies

The Arrhenius equation successfully describes temperature dependence of reduction kinetics:

$$k(T) = A\cdot\exp(-Ea/RT)$$

For coal-based copper slag reduction, apparent activation energies range from 85–220 $kJ\cdot mol^{-1}$ depending on temperature range and slag composition [40]. At lower temperatures (1050–1150°C), apparent activation energy approximates 180–220 $kJ\cdot mol^{-1}$. At elevated temperatures (1250–1350°C), apparent activation energy decreases to 85–150 $kJ\cdot mol^{-1}$, reflecting transition to diffusion-controlled kinetics [19].

The reduction of fayalite exhibits distinctive two-stage kinetic behavior [67][75]: - **First stage (0–33% conversion):** Phase boundary chemical reaction controls rate; activation energy ≈ 175–202 $kJ\cdot mol^{-1}$ - **Second stage (>33% conversion):** Two-dimensional to three-dimensional diffusion with concurrent boundary reaction; activation energy ≈ 194–248 $kJ\cdot mol^{-1}$[41].

This two-stage behavior reflects the coupled processes of fayalite decomposition and iron oxide reduction followed by metallic iron grain growth and coalescence [20][42].

3.3.3 Thermodynamic Equilibrium

Gibbs free energy calculations confirm the thermodynamic feasibility of copper slag reduction:

$\Delta G°(1200°C) = -45.3\ kJ\cdot mol^{-1}$ (favorable)

$\Delta G°(1300°C) = -52.8\ kJ\cdot mol^{-1}$ (highly favorable)

The thermodynamically more favorable reduction of Fe_3O_4 compared to Fe_2SiO_4 indicates that magnetite reduction proceeds preferentially during the initial reduction stage, with fayalite reduction occurring subsequently [43]. The addition of CaO flux stabilizes calcium silicates and calcium aluminosilicates, further enhancing iron oxide reduction thermodynamics [21].

3.4 DRI/HBI Production and Characterization

3.4.1 Direct Reduced Iron Properties

Direct reduced iron produced from copper slag exhibits distinctive characteristics. The product contains 85–96 wt% metallic iron, 1–3 wt% residual carbon (from reducing agent), 0.5–2 wt% FeO (unreacted), 2–5 wt% SiO_2, and trace elements including copper and sulfur [22][23]. Metallization degree—defined as the percentage of total iron in metallic form—typically reaches 90–95% for coal-based reduction and 94–96% for hydrogen-based processes [44].

3.4.2 Hot Briquetted Iron Manufacturing

Hot briquetted iron (HBI) represents the commercial product form enabling long-distance transport and storage. DRI particles are compressed at elevated temperature (>900°C) into briquettes with density specifications exceeding 5.0 g/cm³, meeting International Maritime Organization (IMO) requirements for safe marine transport [13][24]. HBI density directly correlates with compressive strength, dustability, and reducibility in electric arc furnaces [45].

3.4.3 Slag Formation in DRI Melting

When DRI is charged into electric arc furnaces (EAF), slag formation occurs through melting of gangue components and oxidation of impurities. Total slag volume typically reaches 100–150 kg/tonne steel for DR-grade DRI derived from high-quality ore. DRI derived from lower-grade ores, including copper slag-derived DRI, generates 200–400 kg slag/tonne steel, necessitating enhanced slag management and requiring increased fluxing [25][46][53].

The slag composition in EAF melting of copper slag-derived DRI exhibits elevated SiO_2 content (35–45 wt%), lower CaO content (20–30 wt%), and variable FeO concentrations reflecting incomplete slag-metal separation [47]. Optimal basicity (CaO/SiO_2 ratio ≈ 2.0) requires substantial flux additions, increasing operational costs and slag disposal requirements [24][48].

3.5 Process Parameters Optimization

3.5.1 Reduction Temperature

Reduction temperature optimization represents a critical challenge balancing recovery efficiency against energy consumption. Industrial and laboratory studies demonstrate:

- **1200°C:** Standard operating temperature achieving 70–75% reduction efficiency with moderate energy consumption
- **1250°C:** Industrial optimum achieving 80–85% efficiency with acceptable energy economics
- **1300°C:** Enhanced recovery (90–92%) but with proportionally increased energy costs
- **1350°C+:** Near-complete reduction (>95%) suitable only for specific applications due to extreme energy requirements and accelerated equipment degradation

Optimal industrial operation typically targets 1200–1300°C as the balance between recovery efficiency and operational economics [25].

3.5.2 Reductant Type and Dosage

Coal characteristics significantly influence reduction kinetics and product quality. Coals with fixed carbon content >85 wt% perform optimally [49], requiring dosage of 30–40 wt% (relative to slag mass) to achieve near-complete reduction [26][55]. Alternative reductants including coke, biochar, and biomass-derived chars have demonstrated feasibility with comparable recovery efficiency to fossil coal [].

Hydrogen-based reduction requires dosages of 55–70 kg H_2/tonne slag (~650 Nm³/tonne), with process efficiency exceeding 94% compared to 75–85% for carbon-based systems [27][51].

3.5.3 Slag Basicity Modification

The addition of CaO flux (15–25 wt% of slag mass) fundamentally alters slag chemistry and reduction kinetics. Optimal basicity (CaO/SiO_2 molar ratio ≈ 0.3–0.75) reduces slag viscosity, promotes slag fluidity, facilitates metallic iron grain aggregation, and enhances iron-slag separation. CaO addition increases iron recovery by 5–10% while improving iron grade in the concentrate [28][54].

The mechanism involves CaO reaction with SiO_2 forming wollastonite ($CaO{\cdot}SiO_2$) and dicalcium silicate ($2CaO{\cdot}SiO_2$), which exhibit low melting temperatures facilitating slag fluidity. Excessive CaO (>25 wt%) increases slag viscosity through formation of tricalcium silicate ($3CaO{\cdot}SiO_2$), potentially reducing iron recovery efficiency [].

3.5.4 Residence Time

Reduction roasting time critically influences recovery efficiency, with optimal duration typically 30–45 minutes for coal-based processes at 1250–1300°C [50]. Shorter durations (<20 min) result in incomplete reduction; extended durations (>60 min) provide marginal efficiency gains not justifying additional energy input [51]. Hydrogen-based processes achieve comparable metallization within similar timeframes despite lower temperature operation [29].

4. DISCUSSION

4.1 Integration with Electric Arc Furnace Steelmaking

Copper slag-derived DRI/HBI provides a secondary iron source enabling EAF operators to dilute scrap-derived impurities (copper, tin, molybdenum, tungsten) while maintaining steel quality. Industrial trials demonstrate that 30–50% DRI substitution for scrap substantially reduces tramp element content, enabling production of higher-grade steel products previously accessible only via blast furnace routes [30][52].

4.2 Hydrogen-Based Green Steelmaking

The convergence of copper slag iron recovery with hydrogen-based DRI production offers a pathway toward carbon-neutral steelmaking [31]. Green hydrogen (H_2 from renewable electricity via water electrolysis) combined with copper slag-derived iron enables near-zero CO_2 steelmaking chains [53]. Current hydrogen costs (€3–6/kg) exceed natural gas equivalents (€0.5–2/kg), but cost reductions to €1–2/kg are projected by 2030 through renewable electricity scaling [31][32].

4.3 Circular Economy Integration

Advanced processing routes combining stepwise metal recovery enable comprehensive copper slag valorization [33][54]. A representative flowsheet comprises: (1) flotation recovering copper concentrate (21.5% Cu grade, 77.78% recovery); (2) reduction-magnetic separation of flotation tailings (91.34% Fe, 90.21% Fe grade, 83.41% Cu recovery); and (3) Portland cement production from non-magnetic residues [33][55].

This integrated approach achieves total iron recovery of 85–90% and enables recovery of 75–85% of residual copper, 60–70% of lead, and 50–65% of zinc through sequential leaching and precipitation [6][34][56].

5. CONSCLUSION

Recent literature systematically demonstrates that copper smelting slag represents a valuable secondary iron source with recovery potential of 85–98 wt% achievable through optimized pyrometallurgical or hydrometallurgical processing. Coal-based direct reduction combined with magnetic separation remains the most industrially established technology, achieving 91–98% iron recovery under optimized conditions (1250–1300°C, 30–45 min, 35 wt% coal, 20 wt% CaO).

Hydrogen-based direct reduction emerges as the most promising future technology, offering superior metallization degrees (94–96%), lower process temperatures (800–950°C), near-zero CO_2 emissions when powered by renewable electricity, and enabling production of premium HBI meeting rigorous international standards. Projected growth of hydrogen-based DRI from negligible current production to 212.6 million tonnes by 2050 indicates strong industrial momentum toward hydrogen-based steelmaking.

Key research priorities include: (1) pilot-scale demonstration of emerging technologies (>500 tonne/day capacity); (2) hydrogen infrastructure development enabling large-scale supply chains; (3) kinetic and thermodynamic modeling of complex slag systems; (4) integrated flowsheet development for multi-metal recovery; (5) circular economy pathway design maximizing resource and energy efficiency; and (6) techno-economic evaluation under variable commodity price scenarios.

The integration of copper slag valorization with hydrogen-based DRI production represents a critical enabling technology for achieving carbon-neutral, circular-economy steelmaking by 2050.

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