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REVIEW

Research on Improvement of Carbon-Free Self Heating Smelting Technology in Copper Smelting and Synergistic Reduction of Carbon Emissions along with Flue Gas Pollutants

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ABSTRACT

Carbon-free self-heating (autothermal) smelting has emerged as a promising route to decarbonize primary copper pyrometallurgy while improving control of flue-gas pollutants. In this review, recent advances in exploring technology routes and system integrations to permit a stable autothermal regime with minimal routine fossil-carbon feeds in various stages of preparing concentrate, smelting, converting, and anode/fire-refining interfaces are synthesized. The heat-balance aspects needed to allow the self-heating of a carbon-free furnace are discussed in the context of sulfide oxidation heat release, moisture and gangue penalties, matte slag equilibria, and under conditions of variability in feed controllability. With further elaboration of these principles, the review categorizes the following oxygen-enriched and oxygen-blown intensification methods, families of flash and bath smelting, and integrated smelting-converting ideas, with greater emphasis on the fact that oxygen potential and thermal profile are key to process stability, refractory durability, slag family, and energy efficiency. The main emphasis is on the so-called synergistic co-reduction process of CO_2 with decreased off-gassing volume and composition, which influence SO_2 capture, NO_x pathways, particle formation, and the distribution of semi-volatile/toxic species (e.g., As and Hg) into dust, acid, or residues. The review indicates that the net emissions are sensitive to boundaries because of electricity and oxygen production, and this identifies significant obstacles that include heat-balance resilience, corrosion, and dust deposition in the heat recovery systems, impurity recycle loops, and cross-media burden shifting. Lastly,

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coupled modeling, long-duration demonstration with harmonized key performance indicators (KPIs), integrated impurity management, and robust measurement, reporting, and verification are suggested as the priority research requirements to enable scale-up.

Keywords: Copper Smelting; Autothermal Self-Heating; Decarbonization; Oxygen Enrichment; Multi-Pollutant Control

1. Introduction

Modern electrification and the transition to more low-carbon energy systems are based on copper[1]. The speed at which renewable power generation, expansion of grids, electric vehicles, and energy storage are being developed has added copper pressure and has put the environmental footprint of their production under strain like never before. Pyrometallurgical processes to sulfide concentrate are still the most important in primary copper production, and they commonly include the stages of concentrate drying and preparation, smelting to form matte and slag, converting matte to blister copper, and performing fire refining and electrorefining[2]. These unit operations are directly connected with multiple energy flows and multifaceted gas-solid-liquid reaction networks, which both generate greenhouse gases (GHG) emissions and a wide range of flue-gas pollutants. Accordingly, copper smelters are under a dual requirement, namely: (i) a transition to deep decarbonization in tandem with net-zero pathways and (ii) a more rigorous regulation of traditional and toxic air pollutants, such as sulfur dioxide (SO_2), nitrogen oxides (NO_x), particulate matter (PM) and trace elements, including arsenic (As), mercury (Hg), selenium (Se), and antimony (Sb), and lead (Pb). Notably, both of these goals are not independent. Pollutant formation, partitioning, and capture efficiency are also dictated by the conditions of the processes that produce carbon emissions, such as oxygen enrichment level, fuel consumption, furnace temperature, gas volume, and heat recovery[3,4]. This connection presents opportunities and threats: integrated design can lead to synergistic co-reduction of CO_2 and pollutants, but unbalanced interventions can cause subsidisation of burdens among media (air-to-solid residues, or air-to-wastewater) or cause a trade-off between pollutants.

Self-heating (autothermal) smelting is a central concept in the context of copper pyrometallurgy, which has a high ability to decarbonize[5,6]. In autothermal mode, the exothermic oxidation of sulphide minerals (e.g., $CuFeS_2$ and FeS) and associated iron oxidation reactions are the major source of heat input, not because of prolonged burning of fossil fuels. Autothermal behavior has already been used to some extent in industrial copper smelting: most smelters use oxygen-enriched air or oxygen-blown operation, and the energy of sulfide oxidation assists in melting and separating matte and slag. However, fossil carbon is still common throughout the flowsheet. Concentrate drying, furnace warm-up, temperature trimming, or stability during feed variability, converting, and anode/fire-refining processes can use natural gas, and some processes use carbon-based reductants or hydrocarbon fuels in auxiliary processes. Consequently, even the intensely smelting routes can still have considerable direct (Scope 1) combustion emissions and other indirect (Scope 2) emissions related to the production of oxygen and the subsequent consumption of electricity. To establish meaningful decarbonization, therefore, means to go beyond partially autothermal operation to a broader concept: the carbon-free self-heating smelting not just of the integrated smelting-converting-anode chain but of the complete chain, facilitated by such enabling systems as low-carbon oxygen supply, high-efficiency waste heat recovery, and high-performance off-gas cleaning.

This review refers to the process of smelting copper and associated pyrometallurgical processes as carbon-free self-heating smelting, where (i) the thermal equilibrium is maintained constant by oxidation of sulfides and heat integration, and (ii) (or to the best of its ability) no fossil carbon is added or large amounts of carbon are burned to achieve continuous operation of smelting and converting, or to the large-scale thermal processing of anode furnace/fire refining, and so on[7–10]. This definition is purposefully operational instead of absolute: the majority of industrial systems need some start-up energy and may have emergency burners to provide safety and reliability, and the net climate benefit is determined by the carbon intensity of the electricity used to make oxygen and electrified auxiliaries. In that regard, an System Carbon Intensity (SCI)-standard assessment should specifically outline the limits of the system and accounting

procedures. To be useful comparatively, the full process boundary has been included in this review (preparation of the concentrate to final products) and direct emissions (fuel combustion, use of reductants, and process off-gas) and indirect emissions (electricity to operate oxygen plants, gas handling, and acid plant operations). Carbon-free self-heating is not a single technology but a family of process strategies within this boundary, such as oxygen enrichment and oxygen-blown intensification, advanced heat recovery, electrification of auxiliary heat requirements, and integrated gas-cleaning systems, which allow stable autothermal operation and intensification and improve the ability of a process to capture pollutants.

For the purpose of this review, "carbon-free self-heating smelting" is defined as a steady-state operating mode in which the integrated pyrometallurgical chain (concentrate preparation → smelting → converting → anode/fire refining) maintains thermal autothermal balance using exothermic sulfide/iron oxidation as the dominant heat source, without routine addition of fossil carbon. Non-routine fuel use (e.g., furnace start-up, emergency burners, planned shutdowns) is not considered part of the carbon-free steady state; such events must be disclosed separately in annualized accounting. Considering the discrepancy in the use of the terms autothermal and carbon-free in the literature, the most important definitions, system boundary, and normalization values will be taken as those used in this review set in **Table 1**, which will be the foundation of all further comparisons of CO_2 and pollutant performance. To understand the point of carbon and pollutant generation and capture or transfer of the carbon and pollutants, **Figure 1** maps the system boundary between the preparation of the concentrate and off-gas treatment with oxygen supply and waste-heat recovery, which includes the anode/fire refining.

Table 1. Definitions, system boundary, and normalized metrics used to evaluate carbon-free self-heating copper smelting and co-control of flue-gas pollutants.

Term/Metric	Definition	Typical Unit/Basis	Notes for Interpretation
Self-heating (autothermal) smelting	Smelting in which exothermic oxidation of sulfides/Fe provides the dominant process heat	-	Must be demonstrated via heat balance (not just furnace type)
Carbon-free (operational)	No routine fossil-carbon combustion in steady-state smelting–converting–anode stages; start-up/emergency fuel disclosed	-	Requires explicit boundary and annualized accounting. Steady-state operation uses no fossil carbon; start-up/emergency fuel is disclosed separately and not counted toward "carbon-free" claim.
System boundary	Concentrate prep → smelting → converting → anode/fire refining → gas cooling/cleaning → acid plant → oxygen supply → waste heat recovery (WHR)	-	Specify what is counted as Scope 1 vs. Scope 2
Functional unit	Production basis for normalization	t blister Cu or t cathode	Use one consistently across the paper
Direct CO_2	On-site fuel/reductant CO_2 (Scope 1)	kg CO_2/t product	Separate from electricity-related emissions
Indirect CO_2	Electricity-related emissions (oxygen plant, fans, electrified heating)	kg CO_2e/t product	Grid intensity drives variability
SO_2 capture efficiency	Fraction of sulfur fixed as acid/solid relative to sulfur in feed	%	Must report both capture % and residual mass emissions
Gas strength	SO_2 concentration in process gas to acid plant	vol% SO_2	Higher strength generally supports better capture but affects corrosion/cooling
NO_x emission factor	Mass NO_x emitted normalized to product	kg NO_x/t product	Distinguish fuel-NO_x vs. thermal NO_x drivers
PM emission factor	Mass particulate emitted after controls	g PM/Nm^3 or kg/t	Report dust loading upstream and stack emissions downstream
Trace element indicator	Hg/As/Se/Sb partitioning and discharge	mg/Nm^3 or kg/t	Track to dust, acid, wastewater, stabilized residues

A vital reason to pay attention to carbon-free self-heating smelting is that it will be possible to redesign the volume and quality of smelter off-gas and provide a favorable environment to control the pollution [11]. The addition of oxygen ensures the reduction of nitrogen dilution that normally raises the concentration of SO_2 and lowers the overall amount of gas. The increased strength of SO_2 streams might result in higher efficiency of the capture of sulfur and a higher performance of the sulfuric acid plant; the smaller volumes of gases will also result in less fan energy and, potentially, less risk of fugitive emissions due to easy enclosure and negative pressure control. Likewise, direct cuts of CO_2 and fuel-based

NO_x can be achieved by eliminating the use of fossil burners. But it is the same intensification that intensifies SO_2 streams that can elevate flame temperatures and process temperatures and raise oxygen potentials, under some conditions, may also encourage thermal NO_x formation, and also alter the volatilization and condensation paths of trace elements. Increasing the temperature and changing the gas chemistry can enhance the volatilizations of As, Se, or Hg and relocate these species to dust or acid streams, posing new limits to dust recycling, acid quality, corrosion, and hazardous waste stabilization. Thus, the synergy pledge should be measured in multi-pollutant terms and not in one-emission terms.

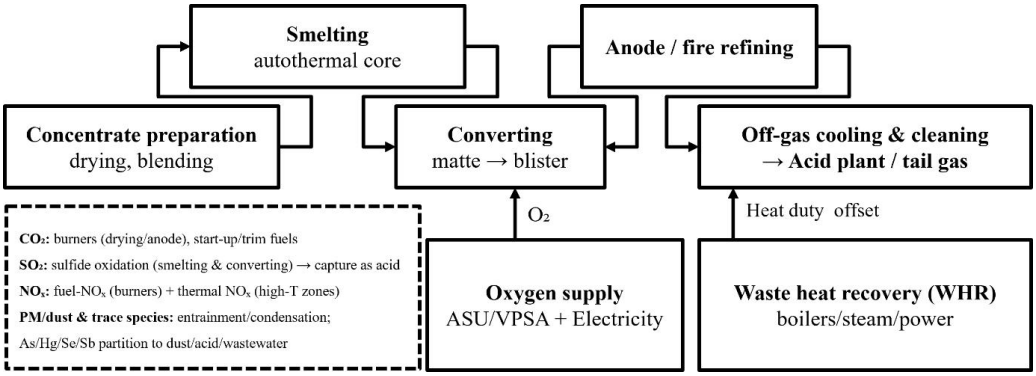

Figure 1. System boundary and emission-source map for carbon-free self-heating copper smelting, highlighting CO_2 sources and key flue-gas pollutants (SO_2, NO_x, PM, and trace species) and their primary capture/transfer pathways.

The types of coupling mechanisms between decarbonization and control of the pollutants in the smelting of copper could be divided into four. The first involves the gas dilution-capture coupling; by increasing the oxygen concentration, the overall off-gas flow is decreased, the concentration of SO_2 rises, and the thermodynamic driving forces and kinetic driving forces of the downstream sulfur capture in acid plants or other sulfur recovery units may be enhanced. Second is the combustion pathway coupling: by removing fossil fuel combustion, CO_2 emitted as well as fuel-NO_x emitted are reduced, but the thermal profiles and residence times undergo changes that could alter thermal NO_x emissions. Third is the temperature-partitioning coupling: Moving between smelting and gas cooling areas, the distributions of semi-volatile species between the matte/slag, dust, and gas phases are determined by the temperature and oxygen potential available. This influences PM formation, dust loading, and composition of captured solids, which subsequently influences recycle ability as well as residue stabilization. Fourth is the energy integration coupling: the better waste heat recovery, the less auxiliary fuel is required, and can be used to offset other electrical demand; however, it also affects cooling rates and condensation, which is important in dust chemistry, acid mist formation, and corrosion control. Any sound framework of technology assessment should take these couplings into consideration and clearly deal with possible tradeoffs such as cross-media transfers and operational reliability[1,12].

Traditionally, copper smelting technology has improved by a series of intensification stages: starting with reverberatory furnaces with huge fuel needs and producing SO_2 gases, to more intensified technologies, including flash smelting and bath smelting versions, which more fully utilize the exothermic heat of sulfide oxidation[13,14]. Oxygen-enriched and oxygen-blown processes are now able to run at high smelting intensity and to generate off-gas streams much more conducive to the production of sulfuric acid when compared with the previously used air-blown process. However, it is difficult to achieve carbon-free performance at a significant scale, at least because of three reasons. The former is the heat balance sensitivity: the autothermal margin is very sensitive to measuring the moisture of the concentrate, gangue material, fluxation needs, and blending grade, all of which can fluctuate based on ore origin and the combined mix approach. The second is constraints in materials and stability: acceleration of refractory corrosion, accretion effects, and magnetite management challenges in slags due to high oxygen potentiality and Bo-Gs gradients, is threatening availability and campaign life. The third is impurity complexity: the greater the extent to which concentrate supply

consists of feeds of higher impurity, the more difficult it is to control trace emissions whilst keeping the smelting process efficient. Dust recycling and impurity build-up pose a threat to environmental performance, unless an appropriate grade of feed is produced and smelting is made rapid because of mobility issues. These facts justify the need to have combined solutions to deal with process thermodynamics and emissions chemistry.

It is against this background that the current review will be able to systematize and critically review research developments on enhancing carbon-free self-heating smelting technology in copper smelting, with a certain focus placed on synergistic carbon emission reduction and flue-gas pollutant minimization. Three guiding questions are used to structure the review: (1) What are the key technical directions so that stable autothermal operation does not require the constant input of fossil carbons during the processes of smelting, converting, and the anode furnace? (2) What are the effects of these pathways on the formation, partitioning, and capture of SO_2, NO_x, PM, and trace pollutants, and what combined designs can achieve the greatest co-benefits and the least tradeoffs? (3) What are the prevailing obstacles to industrial implementation- technical, economical, and operational and what lines of research are most likely to hasten scale-up with quantifiable environmental results?

In answering these questions, the review brings literature in the basic operating principles of the process (heat balance, reaction kinetics, control of oxygen potential), technology routes (energization through intensification of flash and bath smelting, strategies of supplying oxygen, electrification of auxiliaries, waste heat recovery) in addition to integration of sulfuric acid and tail-gaseous cleaning, integrating metal sulfuric acid with other steps, treatment of emissions, and control of impurities[10,15]. One of the similarities is the requirement of similar metrics and clear delimitations. The processes of carbon-free operation can be misrepresented without direct consideration of the inclusion of oxygen plants, electricity and heat recovery credits, and offset of start-up and upset conditions. On the same note, the reduction in pollution should be measured not only by the stack concentrations but also by the capture efficiencies, the production of residues, and the distribution of the hazardous species in the dust, acid, and wastewater streams. Thus, the normalized indicators (e.g., per ton of blister copper or cathode) are placed in focus of this review, and the points where the quality of data and comparability are still inadequate are outlined.

Finally, better carbon-free self-heating smelting cannot be considered a system-level change but rather a free upgrade of a furnace. The comprehensive innovation needed to achieve profound emissions will include (i) the development of autothermal, concentration-variable design of heat balance, (ii) oxygen and heat integration with low-carbon electricity, (iii) the development of robust and corrosion-resistant, off-gas cooling and cleaning architecture, and (iv) the development of impurity storage measures that ensure that the burden is not pushed onto other parties and allow responsible stabilization of residues. This review aims to inform both scientists working on next-generation copper smelting technology and those interested in conducting a retrofit process toward a net-zero and near-zero pollutant emission by clarifying the mechanisms of synergy and defining the conditions in which a co-reduction is a realistic process.

2. Carbon-Free Self-Heating Smelting: Fundamentals and Technical Pathways

2.1. Conceptual Framework of Autothermal Copper Smelting under Carbon-Free Constraints

Self-heating and autothermal copper smelting. Process regimes where the heat needed to drive the melting, reaction progression, and phase separation is provided internally by exothermic oxidation reactions of sulfide minerals and lower-valency species of iron are called self-heating or autothermal copper smelting[10,16]. During the smelting of sulfide concentrates, FeS and Cu -Fe sulfides oxidation provides a large reaction heat and how this heat can be used to maintain steady running relies on how well this heat can counteract endothermic reactions, like moisture evaporation of the concentrate, the degradation of minor phases, reagents in slag formation, sensible heating of solids and gases, and inevitable heat loss through the refractories and cooling systems. Autothermal state should be regarded as a heat-balance state instead of a feature of a particular type of furnace. In the case of industrial operation, self-heating occurs when the net enthalpy produced in the reaction zone and transferred into the molten

bath is high enough to hold the desired matte grade, the slag fluidity, and the settling characteristics without constant auxiliary combustion.

A carbon-free constraint is introduced, and this alters the character of this autothermal state. The traditional smelters can use fossil-fuel burners to start up, dry the concentrate, make intermittent temperature adjustments, or stabilize in response to variations in the feed. Natural gas is usually used as a converters or an anode furnace to provide heat or regulation of oxidation-reduction cycles. In the absence of fossil inputs, it then needs to be the a process based on the controlled intensity of oxidation, oxygen enrichment, sensible-heat recovery, and, in certain instances, electrified addition of heat. Here, carbon-free self-heating smelting does mean no external energy, but a constant production can be maintained without periodic burning of fossil carbon, and any extra energy is made available by low-carbon electricity or recovered process heat. This re-framing increases the significance of setting boundaries of the system and the understanding that the seeming decreases in on-site fuel consumption could be partially compensated by the increases in the emissions to the atmosphere, which are caused by the production of oxygen and the supply of electricity. Carbon-free autothermal design is, therefore, essentially a coupled reaction-engineering, heat-integration, and energy-system-alignment problem[17].

2.2. Heat Balance, Oxygen Potential, and the Determinants of Autothermal Margin

The possible viability of carbon-free self-heating operation can be viewed in terms of an autothermal margin, which is the extent to which the internal heat release is greater than the cumulative heat sinks at a particular operating point[18]. The governing variables are generally transferable, whereas the detailed balance is plant-specific. High levels of sulfur and iron have a strong effect on the amount of heat produced due to the oxidation of sulfides and Fe (II) species as the main source of heat. In comparison, the concentrated moisture, gangue material, and fluxing needs augment the endothermic load by augmenting the evaporation loads and slag masses. Matte grade has an influence on both pathways of reaction and energy partitioning, whereby the greater the matte grade, the deeper the oxidation of iron and sulfur to the required copper enrichment, and this can increase heat release, but the slag chemistry and magnetite formation tendencies move to increase viscosity, settling, and refractory interaction. These thermochemical interactions serve to understand why there are specific forms of concentrates and operating goals with which autothermal behavior is intrinsically stronger and why blending mechanisms and flux choices can prove to be as significant as furnace design in supporting autothermal operation devoid of carbon. The autothermal condition is best understood as a net heat-balance state; therefore, **Figure 2** summarizes the dominant heat sources and sinks and illustrates the concept of 'autothermal margin' under carbon-free constraints.

The rate of oxidation, as well as the proportion of heat of reaction allocated to both the gas and molten phases, is determined by oxygen potential, which is dominated by oxygen enrichment and the ratio of oxygen to feed[19]. Greater concentrations of oxygen in intensified systems cause less dilution of inert nitrogen to occur, and as a result, enhance high temperature in the flame or reaction zone and enhance SO_2 in the off-gas. From a heat-balance point of view, oxygen enrichment enhances the capacity of sulfide oxidation heat to sustain bath temperature since it decreases the aggregate gas sensible-heat loss that comes along with nitrogen and may decrease the aggregate gas flow through the furnace. Meanwhile, an increase in oxygen potential encourages the inclination of magnetite formation within slag and might raise refractory corrosion unless compensated through appropriate slag chemistry and temperature regulation. The practical meaning of this is that a carbon-free self-heating operation is not just attained by increasing oxygen, but that an operating regime is realized over which the strength of the oxidation is sufficient to support the temperature and to retain the slag properties within manageable limits and the wear of the refractory within acceptable limits.

Since a totally carbon-free design cannot depend on burners of fossil fuels to supplement temporary shortages, the performance metric of heat-balance resilience is characteristic of it. Fluctuations in feed moisture, sulfur, and impurity concentrations can change the autothermal margin, and only minor alterations can disrupt temperature regulation or fluidity of slag in high-intensity operation. To this end, industrially plausible self-heating processes without carbon need not only a preferable average heat balance, but sufficient dynamical controllability, such as rapid oxygen regulation, thorough feed preparation, and heat buffering with molten inventory or integrated heat recovery[20,21].

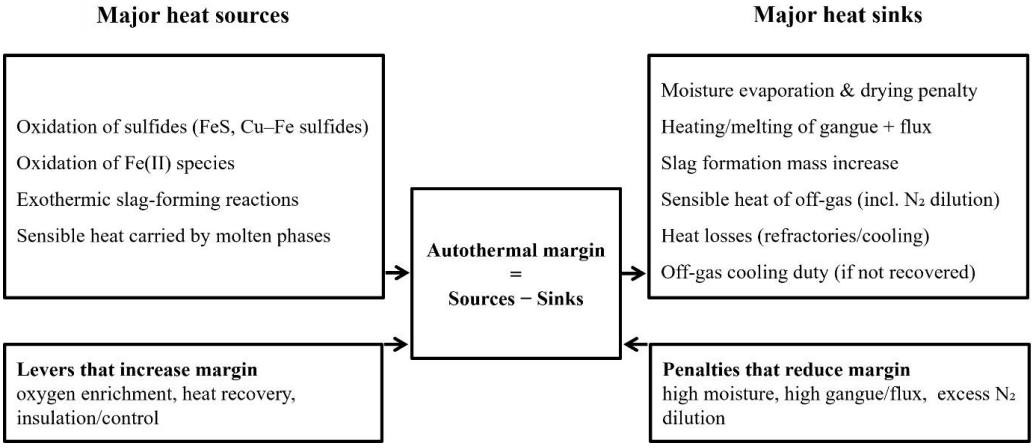

Figure 2. Conceptual heat-balance structure of autothermal copper smelting under carbon-free constraints, illustrating major heat sources/sinks and the definition of autothermal margin.

2.3. Process Intensification through Oxygen Enrichment and Oxygen-Blown Operation

One of the most commonly proven levers to intensify smelting and allow it to be operated in an autothermal mode is oxygen enrichment[22]. Enrichment reduces the amount of nitrogen that must be diluted and raises the partial pressure of the reactive species, thus enhancing the oxidation kinetics and permitting smaller furnace volumes or higher throughput in existing furnace volumes by a factor of two. These effects are especially useful in self-heating smelting, where they enhance the accuracy of the match between the heat generation of reactions and the thermal requirements of melting and slag extraction, and tend to minimize or remove the supplemental fuel requirement of steady-state operation.

The switch to an air-blown to either an oxygen-enriched or oxygen-blown environment, however, creates a grouping of interrelated effects that define the carbon-free envelope of feasibility. An increase in oxygen can cause local temperatures to rise and force heat release into smaller reaction zones, altering heat-transfer mechanisms and possibly raising peak thermal loads on refractories. The resulting increase in SO_2 concentration is beneficial in terms of enhancing downstream capturing of sulfur and acid plants; however, need close monitoring of gas cooling patterns to avoid corrosion and mist development of acid. Besides, the elevation of oxygen potentials has the potential to modify the partitioning of elements and stability of oxides in slag, and the control of magnetite turns out to be a common problem in highly intensified regimes. Thus, oxygen-based intensification can be viewed as a system intervention that simultaneously involves furnace thermodynamics, gas analogies, and residue chemistry, which should all be addressed to provide the actual carbon-free self-heating operation throughout the integrated flowsheet[23].

Carbon accounting: Oxygen enrichment sends part of the decarbonization load upstream to the oxygen supply system[1,24]. The net climate benefit is also based on the mix of the electricity and on the actual energy consumption of the oxygen plant, and how the oxygen use will be maximized in comparison to throughput and metallurgical targets. This brings an imperative in design: carbon-free self-heating smelting is most plausible when the oxygen generation is to be accompanied by low-carbon electricity, and the design of the process is such that it reduces the amount of unnecessary oxygen consumption by better control over the oxidation effectiveness and minimizing the leaking of air.

2.4. Technology Classes Enabling Carbon-Free Self-Heating Smelting in Copper Production

Even though the general principles of autothermal can be generalized, the routes to carbon-free self-heating vary among the families of smelting by suspensions and by baths due to the difference in the reaction environment and heat-transfer mechanisms[25]. In suspension or flash-type smelting, oxidation of the dried particles of the concentrate proceeds very quickly, and a portion of the oxidation is carried out in the air as the particles fly. This design maintains high specific throughput and is capable of strongly autothermal operation when the concentrate drying is also efficient,

and oxygen enrichment is also tuned. The carbon-free requirements in flash-style systems commonly focus on the elimination of fossil fuel in the drying process and start-up, temperature stabilization of the reaction by using oxygen and feed controls, and waste heat recovery in the uptake and boiler systems, which do not lead to excessive dust deposition and corrosive environments.

Bath smelting apparatus, such as top-submerged lance and bottom-blown, depend on intense mixing and injection of gases into a melt in which oxidation and smelting reactions take place in the liquid [26,27]. They might be highly operationally flexible in the sense of feed size distribution and tolerate greater feed moisture levels than strict flash systems, which can be valuable when carbon-free operation is sought in case drying is energetically or operationally limited. But bath smelting can also offer various difficulties of carbon-free self-heating, especially with refractory consumption in your intense turbulence, localized high-temperature regions around lances, and control of slag chemistry at high oxygen potentials. The route to becoming a carbon-free operation in such systems usually consists of the use of optimal oxygen injection strategies to maintain the temperature without external fuel, constant freeze lining or refractory cover, and efficient off-gas cooling and dust management, which is in line with high dust loading.

Hybrid designs and integrated smelting-converting designs are intended to unite high kinetics of intensified oxidation with better thermal efficiency and simplified flowsheets [3]. Their features are desirable for carbon-free self-heating since they can minimize the final thermal steps that need to be made with external heat and generate more consistent high-strength SO_2 gas that can be used in downstream capture. However, integration enhances the interconnection among unit activities: occurrences in one area may spread quickly, and the division of impurities can become complicated. Due to that, hybrid pathways present specifically severe challenges to process control, monitoring, and metallurgical strength, and their environmental performance cannot be assessed only on a steady-state basis. Although autothermal principles are shared, the feasibility window and dominant constraints differ by furnace family and integration level; **Table 2** compares the main carbon-free self-heating pathway classes in terms of enabling mechanisms, off-gas trends, constraints, and retrofit suitability. To provide a unified taxonomy for the remainder of the review, **Figure 3** organizes these pathways into a classification tree, highlighting cross-cutting enablers such as oxygen supply, waste-heat recovery, electrification, and advanced control.

Table 2. Comparative matrix of carbon-free self-heating technology pathways in copper smelting, summarizing enabling mechanisms, off-gas trends, dominant constraints, and retrofit suitability.

Pathway/Furnace Family	Autothermal enabler	Carbon-Free Lever(s) across Line	Off-gas Characteristics (Trend)	Key Technical Constraints	Retrofit Suitability
Flash/Suspension smelting	Rapid sulfide oxidation in suspension; high intensity	Electrified/WHR-based drying; oxygen enrichment; minimized trimming burners	Lower gas volume; higher SO_2 strength; potentially fine dust	Drying dependence; dust/boiler fouling; tight temperature window	Medium–High (site dependent)
Bath smelting (top submerged lance (TSL)/bottom-Blown)	Oxidation within turbulent bath; high mixing	Reduced drying requirement; oxygen-blown; heat retention; electrified auxiliaries	Lower gas volume; higher SO_2 strength; aerosol/droplet carryover possible	Refractory wear; lance zone hot spots; slag/magnetite control	Medium
Hybrid/Integrated smelt–convert concepts	Reduced unit operations; continuous heat utilization	Reduced fuel steps; stable high-SO_2 gas to capture	Potentially stable high-strength SO_2; lower dilution	Control complexity; impurity handling; technology maturity	Low–Medium (often greenfield-first)
Carbon-free converting (high-O_2, continuous)	Exothermic Fe/S oxidation	Burnerless converting; improved hooding and capture	Stronger SO_2; reduced fugitives with good sealing	Refractory/hood constraints; transient peaks	Medium
Carbon-free anode/Fire refining interface	Not inherently autothermal	Electrified heating; insulation; heat integration from WHR	Reduced combustion products; altered NO_x profile	Power reliability; equipment durability	Medium–High (stepwise upgrades)
Enabling systems (oxygen + WHR + controls)	System-level	Low-carbon oxygen, WHR steam/power, model predictive control (MPC), monitoring, reporting and verification (MRV)	Stabilizes gas strength; reduces fugitives	Corrosion/deposition; sensor reliability	High

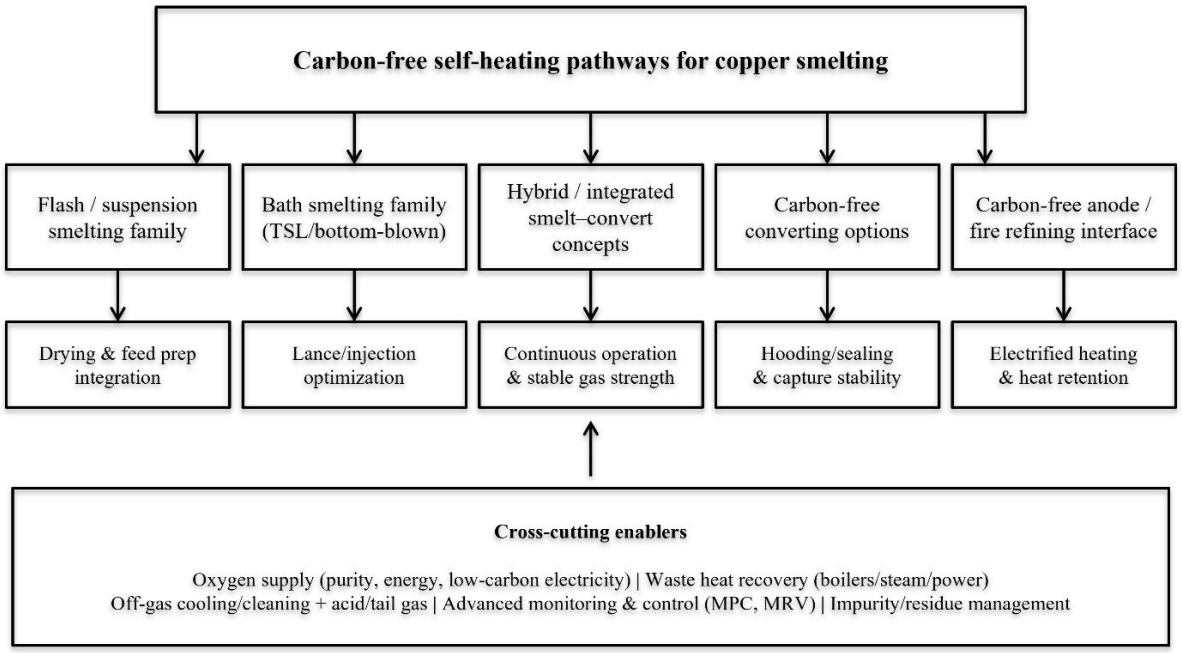

Figure 3. Classification tree of carbon-free self-heating smelting pathways for copper production, including cross-cutting enablers (oxygen supply, waste heat recovery, electrification, and advanced control).

2.5. Extending "Carbon-Free" across Converting and Anode/Fire-Refining Operations

Although smelting itself may be executed in a complete autothermal form, the copper pyrometallurgical line still has converting and anode/fire-refining steps that may continue to be an important source of fossil combustion emissions [28]. In the transformation of matte to blister copper, oxidation of iron and sulfur converts the alloy to copper, and the process is very exothermic, and with suitable control of oxygen supply and heat losses, can be made to operate self-heating. Practically, converting has long involved modes of operation which could involve the use of fuel to allow control of temperature or to control the thermal profile at various blow stages. When subjected to carbon-free limitation, strategies of conversion are directed at enhancing oxygen consumption, enhancing hooding and gas recovery to dilute less and prevent fugitive emissions, and maintaining thermal equilibrium by means of better heat management instead of by auxiliary combustion. Continuous converting concepts are sometimes referred to, since these may help minimize transient extremes and enhance integration with stable downstream gas treatment, although the practicability of the converting concept relies on the properties of the concentrate and impurities and the strength of refractory systems.

The situation with anode furnace and fire refining is a bit different since they usually use burners to provide the heat necessary to maintain the temperature and to facilitate oxidation-reduction processes that can adjust the amount of dissolved oxygen and eliminate impurities [29]. The carbon-free performance of this step is generally accomplished by electrified heating, better insulation, and heat retention, and close coupling with the upstream waste heat recovery, such that thermal losses are minimal. Its operational goal is still metallurgical: to make the right amount and level of impurity and oxygen, and to create an anode good enough to be refined. Carbon-free redesign, therefore, has to maintain product quality and throughput and modify the heat supply mechanism and, possibly, the profile of emissions caused by combustion. Also, NO emissions and local thermal gradients, including those caused by burner removal, can impact the behavior of secondary oxidation and the behavior of furnace lining.

2.6. Start-Up, Transient Operation, and the Practical Meaning of "Carbon-Free" in Industry

Consistent with the operational definition above, a practical challenge in implementing carbon-free self-heating is managing start-up, shutdown, and transient upsets without violating the steady-state carbon-free claim; such non-rou-

tine events are accounted for separately and do not invalidate the carbon-free steady-state paradigm [30]. The controlled warm-up of most smelting furnaces is needed to prevent thermal shock, develop stable freeze lining or refractory temperature gradients, and emergency burners can be required to ensure safety or to avoid solidification during unintended breakages. In a purely theoretical definition, the use of any fossil fuel would be contrary to the notion of carbon-free, but in practical and regulatory terms, it is better to ask whether the continued operation depends on the structural necessity of fossil combustion or just rare contingency use. Thus, carbon-free self-heating is to be understood as a steady-state operating paradigm that is backed by low-carbon utilities, and non-routine fuel consumption and its annual contribution to the total emissions are reported transparently.

The reason why transient behavior is also important is that intensified oxygen-blown operation might be fast-dynamics [31,32]. Variations in feed composition, leakage of air into the furnace, and alterations of the mineralogy of the concentrate can alter the oxygen demand and heat release, which may cause temperature swings or alter slag properties. In a world with carbon-free, this makes oxygen regulation, feed control, and thermal buffering critical in the control of these transients. This is the need which connects fundamentals with enabling technologies, which include, but are not limited to, fast oxygen flow control, matte grade and slag chemistry predictive models, and off-gas composition and temperature powerful instrumentation. Put differently, the concept of carbon-free self-heating smelting is not only a matter of control and integration but of thermodynamic challenge, and its feasibility is limited to the attainment of stable operation within realistic ranges of variability and not merely within idealized steady states.

3. Synergistic Co-Reduction of CO_2 and Flue-Gas Pollutants

3.1. Coupling Logic between Decarbonization and Air-Pollutant Control in Copper Smelting

Interventions based on decarbonization of copper pyrometallurgy can hardly target only one stream of emissions since the furnace and its off-gas system are a thermochemical network [7]. Modifications to achieve a lower CO_2 level, e.g., removal of auxiliary burners, more aggressive enrichment, intensifying autothermal reactions, and enhancing heat recovery, also alter the off-gas volume and composition, temperature history, dust loading, and semi-volatile elements speciation. The potential of so-called synergy occurs when a single change has similar benefits on climate and air-quality goals, such as lowering both fossil-burning CO_2 and fuel-generated NO_x, and allowing more SO_2 to be captured because of a more powerful SO_2 stream. Tradeoffs, on the other hand, can occur when intensification causes a rise in thermal NO_x or when the condensation chemistry is changed to cause an increase in capture of toxic elements into acid or fine dust that is more difficult to dispose of. Any tight-fisted argument on co-reduction should then not be confined to stack concentrations but rather be a system that takes into consideration the upstream reaction pathways, downstream cooling and cleaning structure, and the disposition of captured species in solids, liquids, and products. Because each decarbonization intervention shifts multiple thermochemical levers simultaneously, **Table 3** summarizes the expected directionality and control points for CO_2 reduction and multi-pollutant outcomes (SO_2, NO_x, PM, and trace species), emphasizing where tradeoffs and cross-media transfers may emerge [33].

Table 3. Intervention-to-outcome co-reduction map showing how carbon-free pathway elements affect CO_2 emissions and flue-gas pollutant behaviour (SO_2, NO_x, PM) and trace-element fate, including potential cross-media transfers.

Intervention (Carbon-Free Pathway Element)	CO_2 Impact Mechanism	SO_2 Impact	NO_x Impact	PM/Dust Impact	Trace Species (Hg/As/Se/Sb) & Cross-Media Considerations
Eliminating fossil burners (drying/anode/etc.)	Removes direct combustion CO_2	Indirect (less dilution if air burners removed)	Reduces fuel-NO_x; may lower baseline NO_x	May reduce soot/combustion aerosols	Shifts temperature history; may alter condensation points
Oxygen enrichment/oxygen-blown operation	Supports autothermal heat balance; reduces auxiliary fuel	Higher SO_2 strength; lower gas volume; better acid-plant suitability	Risk of higher thermal NO_x if peak T increases	Mixed: less dilution but possibly finer dust/aerosols	Can increase volatilization; may increase capture in dust/acid

Table 3. *Cont.*

Intervention (Carbon-Free Pathway Element)	CO_2 Impact Mechanism	SO_2 Impact	NO_x Impact	PM/Dust Impact	Trace Species (Hg/As/Se/Sb) & Cross-Media Considerations
Waste heat recovery (WHR) intensification	Offsets external energy; reduces auxiliary heat demand	Enables stable gas delivery to acid plant if well designed	Indirect (temperature profile changes may affect thermal NO_x)	Deposition/fouling risk if dust is sticky	Cooling profile controls whether toxics condense to dust or enter acid
Improved sealing/hooding and negative pressure	Prevents fugitive losses	Reduces fugitive SO_2	Can reduce uncontrolled NO_x release	Reduces dust leakage	Moves burden to captured streams; requires residue management
Optimized cooling/condensation staging	Indirect via efficiency	Stabilizes acid plant inlet; reduces mist issues	Can reduce thermal NO_x residence time via quench	Improves capture by conditioning particle size	Determines partitioning to dust vs. acid vs. wastewater
Advanced particulate capture (Electrostatic precipitator (ESP)/Baghouse)	Indirect via reduced rework/maintenance	Protects acid plant; supports high sulfur capture	Minor direct effect	Major reduction if well sized/maintained	Captures toxics into dust; drives stabilization/recycle strategy
Tail-gas treatment/polishing	Indirect (electricity use)	Final SO_2 reduction	Typically minimal	Minimal	Must avoid creating secondary wastes without treatment routes

3.2. CO_2 Reduction Mechanisms and Boundary-Sensitive Interpretation

In the smelting and conversion of copper, direct CO_2 emissions are mainly related to the combustion of fossil fuels to dry the concentrate, start-up and stabilization of the furnace, heating of the anode furnace, and other thermal loads where the heat of sulfide oxidation is not used effectively [7,34]. Self-heating methods that are carbon-free mitigate such emissions through the replacement of combustion by regulated oxidation intensity, enhanced heat retention, and, where required, electrified heating. In theory, converting can be autothermal by nature since oxidation of iron and sulfur generates a lot of heat, or when using favorable operating conditions and the sulfide content of the concentrate, intensified smelting can maintain the bath temperature without having to add fuel. The short-term CO_2 advantage is thus attained by eliminating on-site fuels and by enhancing energy efficiency.

Nonetheless, the net CO_2 effect is a boundary effect since enrichment of the oxygen and electrification will cause a portion of the energy requirement to be shifted to electricity [35]. Indirect sources of emissions can be major when powered by grids that are rich in carbon, e.g., oxygen generation, gas handling, and auxiliary electric heating. Therefore, what matters is not just the decrease in on-site fuel use, but the decrease in the emissions of CO_2 -equivalent at the defined boundary, which comprises electricity to operate oxygen plants and large fans and blowers. The issue of heat recovery also makes interpretation difficult due to the fact that recovered steam or electricity may displace external energy, giving rise to credits whose value varies depending on the local energy systems and accounting processes. To a standard treatment, under SCI, CO_2 reductions should then be debated as either a combination of direct displacement of combustion at the expense of the process or enhanced integration of process heat and decarbonization of upstream electricity, realizing that the same process intensification which enhances autothermal performance can also elevate oxygen demand and hence the value of low-carbon electricity.

3.3. SO_2 Strengthening, Sulfur Capture, and the Co-Benefit Pathway

The greatest synergies linked to the carbon-free self-heating operation are the strengthening of the SO_2 in the off-gassing as the level of dilution of nitrogen decreases. In air-blown systems, high off-gas volumes (possibly characterized by lower concentration of SO_2) can interfere with efficient sulfur capture and increase the risk of fugitive emissions since gas handling structures are required to transport large volumes and the capture infrastructure is more challenging to seal and hold at negative pressure. The enrichment with oxygen and the use of oxygen-blown operation can de-

crease the total gas volume and elevate the concentration of SO_2, which tends to enhance the feasibility and efficiency of downstream production of sulfuric acid. Greater SO_2 supplies can be used to enhance conversion efficiencies and minimize the proportion of sulfur that is emitted in uncontrolled form, though gas cooling, dust removal, and acid plant combination must be fabricated to accommodate the changed thermal and particulate loads[36].

Under a carbon-free structure, the gain of the sulfur capture is not a peripheral process; it is part of the logic of the process. A more concentrated process gas, a result of higher-intensity smelting autothermal, is inherently more capturing and valorization capable, and is converting sulfur management from an emissions-controlled responsibility into a steadier unit operation. However, even high-strength SO_2 gas contributes to the significance of strong cooling and corrosion management, as gas composition that promotes the occurrence of acid could enhance the appearance of corrosive condensates if the temperature is not controlled correctly. The synergy is then pegged on the correspondence of furnace intensification and a suitably designed gas handling train that is capable of maintaining capture efficiency without compromising dust chemistry and avoiding acid mist and downtime caused by corrosion[37].

3.4. NO_x Formation Pathways under Carbon-Free Intensified Conditions

The mechanism of NO_x emissions in copper smelting is sensitive to both combustion and high-temperature oxidation conditions. NO_x produced by fuels may have a significant contribution under the circumstances of the auxiliary fossil burners, and the direct co-benefit of eliminating the burners is a reduction in both CO_2 and this component of NO_x. Carbon-free self-heating routes thus generally lower a single significant source of NO_x to the beginning (in drying and anode furnace processes that tend to use hydrocarbon combustion)[38].

Meanwhile, it is possible to enrich oxygen and enhance reaction regions to elevate peak temperatures and alter residence times to facilitate thermal NO_x formation that even occurs in the absence of fuel nitrogen[39]. This hazard is especially applicable in cases of localized high-temperature regions adjacent to burners when starting on or in bath smelting or in reaction shafts when using highly enriched oxygen. The empirical consequence here is that NO_x behavior in which carbon-free intensification is practiced cannot be non-monotonic: the reduction of the baseline NO_x by elimination of burners can be overshadowed by the increases in the process temperature and the oxygen supply, unless special care is taken over temperature and mixing. This is why co-reduction approaches tend to be based on staged oxygen injection, temperature regulation by means of controlled heat transfer and mixing of the bath, as well as a quick quenching in uptake and boiler sections to reduce high-temperature residence time. Such strategies demonstrate a more general rule: it is possible to achieve a smaller end product of NO_x by operating carbon-free, but to achieve a net positive benefit of a lower NO_x value, a focused control of the thermal budget must be maintained instead of assuming that zero-fuel consumption implies a zero NO_x. The coupling pathways are synthesized in **Figure 4**, which links burner elimination, oxygen enrichment, heat recovery, and cooling/capture design to changes in gas volume and temperature history that ultimately govern SO_2 capture, NO_x formation routes, PM behavior, and trace-element partitioning.

3.5. Particulate Matter, Dust Loading, and the Emissions–Recycle Feedback Loop

The copper smelting manufactured particulate matter is formed by a mix of entraining fine solids, mechanical carryover of droplets, fuming and volatilizing of some species, and eventual condensing during the cooling of gases[40,41]. Each of these pathways can be influenced by intensified operation with oxygen blowing. The entrainment tendencies are reduced with a given throughput using a reduced volume of gas; however, stronger reaction intensities and turbulence can enhance the droplet carryover and dust generation, especially in areas of intense injection or during slag foaming. Suspension smelting, the rate of oxidation and the rate of gas velocity can elevate dust production, and bath smelting can accelerate aerosol creation through active mixing and bubble bursting. The final PM performance is then determined by the furnace design, distribution of reaction intensity and the performance of primary separation zones.

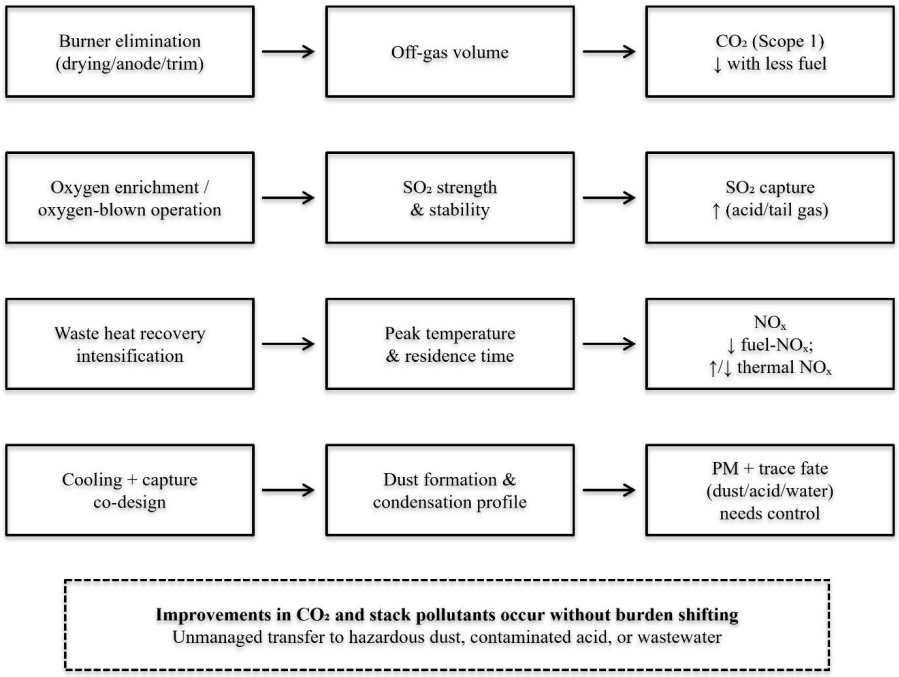

Figure 4. Synergy mechanism network linking decarbonization interventions (burner elimination, oxygen enrichment, waste-heat recovery, cooling/capture design) to gas volume, SO_2 strength, peak temperature, residence time, dust formation, and trace-element partitioning.

With carbon-free self-heating constructions, the dust is located even more centrally since it is both a resource and a hazard. Recycling dust has the benefit of enhancing the recovery of metals and also cutting the amount of waste, but the impurities are also ppb, and this creates a feedback loop in which further impurities are loaded into the system, making control of the emissions more difficult[42]. Increased volatilization by intensification can move a larger portion of these species to the dust and gaseous phases, i.e., hone local air quality and downstream residue management as important factors of both the dust collection strategy and the dust handling strategy. Thus, CO_2 and PM co-reduction cannot be assured: as a smaller quantity of gas can be beneficial to capture better, the chemical and physical properties of dust can be harder in adverse conditions, and thus enhanced filtration, fine cooling profiles, and enhanced residue stabilization activities are necessary.

3.6. Trace Pollutants and Multi-Pollutant Control under Oxygen-Rich Autothermal Regimes

The trace pollutants, which are Hg and the metalloids, i.e., As and Se, show a high level of temperature and chemistry-dependent partitioning behavior[43]. These elements can concentrate in the smelting and conversion into matte, slag, and gas depending on their volatility, attraction to sulfide or oxide phases, and the existing oxygen and sulfur potentials. Spurred oxygen-abundant functioning has the ability to alter these potentials and elevate temperatures, enhancing the proportion of some species that are volatile and subsequently condense as fine particulates or dissolve into acidic plant streams. It poses a serious co-control dilemma: changes in sulfur capture and changes in CO_2 can be accompanied by adjustments in the loading of dangerous species in dust or acid, which can change the quality of acids, catalyst behavior, corrosion, and even hazardous waste.

The conceptualization of multi-pollutant control in this case should then be seen as a moving train, not autonomous devices. The efficacy of the electrostatic precipitators, baghouses, and the wet cleaning system is based on particle size distribution, dust resistivity, moisture, and the gas composition, which shifts under the intensification without carbon[44,45]. The cooling strategy influences the position at which volatile species condense during filtration. Based on this, the synergy of CO_2 reduction and control of trace pollutants is achieved when intensive smelting is combined with controlled condensation, high-efficiency capture of partic-

ulate matter, and management of impurities, which avoids recirculation. In the absence of such integration, the system can merely get the pollutants from one stream to another or generate residues that are more challenging to stabilize.

3.7. Defining and Quantifying Synergy without Burden Shifting

Synergistic co-reduction is a term that suggests a joint enhancement of the climate and pollutant outputs without creating a disproportional new burden in the system[46]. In measuring the synergy, the emissions should be measured in a normalized form that takes into account the production output and whether there is a reduction in the production or an enhancement in the capture. In the case of CO_2, this may generally involve the need to specify emissions in units of blister copper or cathode and to separate the direct fuel-generated emissions and electricity-generated emissions, respectively, of oxygen and the heating of electrified oxygen and electrified heating. In the case of SO_2 and NO_x, it requires stack emissions and capture efficiencies since the low content can co-exist with the great total mass in case of varying gas volumes. In PM and trace species, mass emissions, capture fractions, and what happens to the captured material in recycled dust, stabilized residues, or in polluted acid streams need to be followed to prevent the false finding that capture, that is, capture alone, equals mitigation.

Another condition is a clear consideration of cross-media transfer. The capture can be more aggressive, which makes the amount of dangerous dust or liquid effluent that needs to be treated greater, which could replace the environmental burden placed on air with solid or wastewater management. Therefore, the synergy should be understood as a system effect where the CO_2 and release of atmospheric pollutants will be reduced, and the development of residue will be sufficiently low, and the stabilization pathways will become technically and economically plausible. Understandably, this view tends to correlate with the realities of smelter operation: environmental performance is ultimately assessed by the number of stacks, but also by the results of the whole capture and treatment chain. The greatest synergies in carbon-free self-heating smelting will thus arise when the autothermal intensification, oxygen strategy, heat recovery, and multi-pollutants control are created as an integrated system, but not applied as retrofits[7].

4. Integrated Process Design and Key Enabling Systems

4.1. Oxygen Supply as a Structural Enabler and Its Implications for Net Decarbonization

The self-heating smelting process (carbon-free) depends greatly on oxygen control due to the frequency of oxygen enrichment and the use of oxygen-blown smelting as some of the most effective methods of maintaining the autothermal heat balance and minimizing the off-gassing dilution[47]. Practically, oxygen supply is not a utility as it is structurally a component that determines the thermodynamics of the furnace, the composition of the gases, and the performance of downstream capture. Higher oxygen concentration increases the intensity of reactions and may increase the ratio of heat of sulfide oxidation retained in the molten phases at the expense of higher oxygen concentration causes, a shift of energy demand towards on-site combustion of fossils to electricity production. This change implies that the decarbonization response is very sensitive to both the energy usage of the oxygen plant in question and the electricity carbon intensity. What seems to be a carbon-free process at the locale may provide partial net benefit when oxygen is generated using high-emission electricity, but a low-carbon grid or special supply of renewable energy can turn oxygen intensification into a solid emission reduction plan.

The technical decision of the oxygen supply technology has an influence on the process performance and the emissions[48,49]. Cryogenic separation of air may provide high-purity oxygen that offers stable and high-intensity operation and usually needs significant quantities of electrical power and capital expenditure. In certain designs, pressure swing adsorption and related technologies can provide operational flexibility and reduced capital requirements; however, oxygen purity, as well as capacity, can determine feasible smelting intensity, gas strength, and temperature control. In a self-heating design without carbon, oxygen purity is not a particular requirement: it is combined with nitrogen dilution, volume of off-gases, and heat transfer. Reduced purity amounts of oxygen elevate inert load and can weaken SO_2 strengths, whereas exceptionally high purity amounts of oxygen can elevate peak temperatures and impose refractory and NO_x constraints. This means that the oxygen system has to

be co-optimized with furnace design, off-gas cooling, and the sulfur capture infrastructure as opposed to being chosen based on utility economics.

4.2. Waste Heat Recovery and Energy Cascading under Intensified Autothermal Regimes

Waste heat recovery is one of the fundamental levers to obtain a carbon-free performance since it transforms high-temperatures off-gas enthalpy into useful energy, which can compensate for the auxiliary heat demand, electricity use, or even external steam needs [50]. Uptake boilers and other heat recovery devices are often used in conjunction with smelting furnaces in modern copper smelting; however, the intensified oxygen-rich operation both varies the thermal profile and changes the particulate loading of the gas, thus modifying the design requirements of the reliable operation of a heat recovery device. Increased SO_2 and decreased gas volumetric contents have the opportunity of enhancing thermal efficiency and decreasing fan power; however, higher temperature and altered dust chemistry could spur deposition, erosion, and corrosion hazards on heat-exchange surfaces. The success of aggressive heat recovery thus lies in the control of the rate of gas cooling and in the prevention of the conditions that can lead to sticky dust deposition or corrosive condensate formation.

With carbon-free constraints, energy cascading is especially significant since it will widen the range of ways in which internally produced heat can supplant fossil fuels [51]. Recycled steam may be used to aid in electricity production, heating processes, and in certain applications, concentrate drying or preheating to eliminate the use of burners. Nevertheless, the incorporation of heat recovery alongside drying and feed preparation should be taken at a slow pace. In case drying is relocated to lower-grade heat in order to minimize the consumption of fuel, the heat-balance resilience can be lower in case of a rise in feed moisture or unstable operation of drying. On the other hand, the proper utilization of the recovered heat can stabilize feed preparation and minimize variability that poses a threat to the autothermal margin. The practical design problem is accordingly how to distribute recovered heat to improve the system stability as a whole, and not to maximize recovery individually. With high-rate operation, it frequently means trading off deep heat recovery with the need to have controlled quenching to control the behavior of dust condensation and the integrity of downstream equipment.

4.3. Off-Gas Cooling Architecture, Dust Conditioning, and Corrosion Control as Co-Design Constraints

The off-gas train at copper smelting serves several purposes all at the same time: it cools down the hot-valued process gas, conditions the particles to be effectively collected, protects corrosive species, protects the equipment down the line, and provides gas to sulfuric production or sulfur capture alternative with the right temperature and composition [37]. These demands are enhanced by carbon-free self-heating pathways since oxygen enrichment is likely to raise SO_2 concentration and may raise the peak temperatures of gases, whereas modifications in the reaction environment may raise dust fineness and vary the composition of condensed phases. Within this framework, off-gas cooling is not an afterthought design consideration; it is an upstream design control that determines the destiny of pollutants and the dependability of operations.

Cooling rate and temperature staging control either the formation of solids by semi-volatile species upstream of particulate control devices or the formation of later units. Provided condensation is encouraged further along its course, the traces can be stored in dust, which might make the gas-phase control easier but enhances the risky nature of solids and puts a strain on dust-handling systems. In case of a delay in condensation, these species can be transferred to wet gas cleaning streams or acid plant streams, which can have an impact on acid purity, catalyst performance, and corrosion. This tradeoff is especially applicable to those elements with high-temperature-sensitive capture pathways, including Hg and As. In addition, there are conditions that may enhance the development of acidic condensates with intensified SO_2 streams, and thus, careful choices of materials and temperature are essential to prevent corrosion in boilers, ducts, and cleaning equipment. Thus, cooling architecture design, such as quench sections, waste heat boilers, and gas conditioning stages, should be implemented in conjunction with a multi-pollutant capture strategy and the desired so-called synergy results [52].

4.4. High-Efficiency Particulate Capture and Its Interaction with Intensified Smelting Conditions

Particulate control is a basis standard of regulatory compliance as well as the downstream performance of sulfur capture [53]. The loadings, finer particle properties, and more complicated dust chemistry can be experienced by particulate capture systems under oxygen-rich autothermal conditions. High mass capture efficiencies can be realized using electrostatic precipitators and fabric filters, which are sensitive to the characteristics of particles, the temperature of the gas, moisture and electrical properties subject to change with the intensification of the process. Specifically, the resistivity and stickiness of dust may vary with composition and cooling history and affect collection efficiency and maintenance load.

During carbon-free self-heating smelting, the management of particulate matter has other strategic significance since the recycling of dust is usually performed to reuse the precious metals and to suppress the production of waste [54]. Recycling may enhance the efficiency of resources, but it may also focus impurities and loops of feedback that enhance the instability and distribution of dangerous elements. This renders the capture system as being a component of a larger impurity management plan as opposed to a mere end-of-pipe tool. The design goal goes further than ensuring maximum capture efficiency to the generation of a dust stream that could be safely and efficiently treated, recycled, or stabilized. Consequently, the particulate control needs to be synchronized with upstream decisions that regulate the formation of dust, such as furnace turbulence, injection settings, and slag chemistry, along with downstream treatment paths of impurity-rich residues.

4.5. Sulfuric Acid Plant Integration and Tail-Gas Treatment in a Carbon-Free Framework

One of the most well-known and economically relevant pathways in the control of emissions in copper smelting is the sulfur capture by the production of sulfuric acid [55,56]. Enrichment of oxygen, which reinforces the SO_2 level, may enhance the functioning of the acid plants by supplying them with a better feed gas, which has the potential of improving the conversion efficiency, thus decreasing the amount of tail gas that needs treatment. Simultaneously, the heightened activity may make one more sensitive to the changes in the gas composition, temperature, and dust carryover, and the stable integration of smelting and acid production becomes even more important. The upstream gas conditioning train must be able to provide high-strength SO_2 streams with sustained temperature and effective dust removal to secure catalysts and heat exchangers; the process may become a disadvantage.

Tail-gas treatment gains more significant value as the regulations grow stricter and the smelters become interested in achieving close-to-zero pollutant emissions in addition to deep decarbonization. Tail-gas control performance is affected by the concentration of inlet SO_2, oxygen concentration, moisture, and the presence of catalyst poisons or aerosol-forming species [57]. Some of these can be mitigated by carbon-free self-heating operation, to reduce the gas volume, and stabilize the SO_2 strength, yet it can also introduce new problems, as smelting may become more severe and traces can contaminate catalysts' life or acid mists. Therefore, acid plant and tail-gas design have to be considered as an integrated system optimization, as the oxygen strategy, and off-gas cooling, instead of a fixed downstream module.

4.6. Electrification of Auxiliary Heat Demands and the Role of Grid Carbon Intensity

To remove the use of fossil combustion throughout the smelting line, thermal loads that are not entirely met by autothermal heat are frequently electrified, especially in the process of preparing concentrates and in anode furnace practice [58]. The electrification may be of any type (resistive heating, induction, or other sources of electric heating), and its climate benefit is highly dependent on the carbon intensity of electricity. Even in areas where the grid is low-carbon, the conversion of remnants of fossil fuel consumption into significant net CO_2 reduction by electrification can support the carbon-free self-heating paradigm. Where electricity is generated by high-carbon sources, electrification can cause emissions to be redistributed vertically without providing the intended net improvement, except when combined with clean power acquisition or on-site renewable linking.

Thermal controllability is also changed with electrification. Fast response and close temperature control [59].

Electric heating may be useful in transient control and the maintenance of product consistency. This controllability may reinforce autothermal resilience through the decrease of the dependence on combustion trimming, as well as allow more straightforward integration of oxygen control and feed control. Nevertheless, the electrified systems have to be carefully engineered to be able to survive in the severe furnace conditions, and may place new limitations on the reliability of power supply, peak demand control, and equipment life. Electrification, thus, should be seen as an enabling element that needs to be co-designed with heat recovery, oxygen supply, and process control, and not to be an equivalent one to one replacement of burners.

4.7. Digitalization, Instrumentation, and Advanced Control for Carbon-Free Operational Stability

The fact that carbon-free self-heating smelting charges a premium on stability is due to the fact that, since the standard fuel trimming is thrown off, the operator of the plant must rely on combustion to deal with heat-balance anomalies. To maintain a constant autothermal margin at variable feed conditions, the delivery of oxygen, feed rate, and, in certain cases, flux addition and temperature control of the downstream units must be controlled rapidly and precisely. This requirement increases the importance of digitalization and sophisticated control measures, such as soft sensors that can determine matte grade, slag chemistry, or oxygen potential based on measurable parameters, such as off-gas composition, temperatures, and flow rates[47].

More advanced control strategies can be used to facilitate co-reduction goals by coordinating multiple setpoints at a time, e.g., keeping the SO_2 strength of the acid plant at target levels and controlling temperature to reduce thermal NO_x, and managing dust generation by maintaining constant flow regimes[60]. Models predictive and data-driven control can also be used to control transients related to the variability of feeds and equipment limitations to decrease the number of off-spec conditions that can cause an emergency combustion or high fugitive emissions. Moreover, strong monitoring can be used to make real carbon-free claims as it allows clear measures, reports, and verification of both the CO_2 and pollutant emissions within the system boundary. In that respect, instrumentation and control can be regarded not as auxiliary utilities but as key facilitators bridging the gap between the physics of the autothermal process and its environmental implications and industrial dependability.

5. Comparative Assessment, Barriers, and Research Needs

5.1. Rationale for Comparative Evaluation and the Need for Harmonized Metrics

Individual comparisons between carbon-free self-heating smelting pathways in diverse furnace families and plant designs cannot be entirely dependent on qualitative descriptions, as the performance in this context is extremely sensitive to the composition of the concentrates, the oxygen strategy, the design of the heat recovery, and the level of integration of the gas treatment systems[61]. An effective comparative analysis should be based on harmonized measures and open frontiers. Practically, the most informative comparisons put outcomes on a functional basis, e.g., one ton of blister copper or cathode; direct on-site emissions are explicitly separated into indirect emissions associated with the generation of oxygen and the consumption of electricity. The same is used with the idea of the pollutant control, so that reporting of the stack concentrations only may be misconceiving when the volumes of gases vary with the increasing oxygen content, and, therefore, the total mass emissions, the capture efficiencies, and the destinations of captured species in dust, acid, and residues are required to evade false conclusions. The absence of a unified review system can make two systems seem equal in a single set of indicators and radically different in terms of net CO_2 effect, capture of sulfur, or dangerous residue load.

Another issue is that concepts of carbon-free self-heating cut across process and utility realms[51]. The effectiveness of the seizure of SO_2, NO_x, and PM is determined by the strength of hooding, sealing, cooling, and capture systems, which can vary significantly according to location, and the net emission performance of an oxygen-blown autothermal smelter is determined by the intensity of electricity carbon, and the particular energy consumption of the oxygen plant. The comparative evaluation should then be viewed as a system-level evaluation that connects the performance of metallurgical to utility demand and downstream environmental controls, or as an estimation of results and not as an

evaluation of furnace choice being the best determinant of results.

5.2. Benchmarking of Technology Classes: Performance Envelopes and Typical Tradeoffs

The smelting pathways, which are based on suspensions, such as flash-type arrangements, often have large reaction rates and have a high autothermal character in the case where feed drying is successful and oxygen supply is properly regulated[25]. Their principal comparative advantages are the high throughput, comparatively small reactor volumes, and the capacity to generate off-gases that can be effectively counterbalanced by the production of sulfuric acid when operating under enriched oxygen conditions. Nonetheless, the same high-intensity reaction environment may give high dust loading and dust size distributions and provide high uptake design, waste heat boilers and particulate capture requirements. In systems without the use of carbon, the reliance on feed preparation is an even stronger factor of viability: when the drying must be fueled by fossil fuel and cannot be reliably replaced by recovered heat or electrification, the entire description of carbon-free is undermined even where the smelting furnace per se is autothermal.

Top-submerged lance and bottom-blown bath smelting routes are more flexible in the variability of feed and can be used with higher moisture or coarser feed. This can be used to accommodate carbon-free solutions in cases where the drying of the concentrate in question is limited, and can be equally used to relocate problems to refractory wear, bath hydrodynamics, and local thermal loads around the injection locations. The performance in relation to others will be the ability of the system to sustain steady slag chemistry, control the formation of umagite, and freeze lining or integrity of refractory with increased oxygen delivery. Highly turbulent settings can also result in large aerosol and droplet carryover by bath systems, which influence the properties of PM control and dust recycle[62,63].

The concept of hybrid and integrated smelting-converting focuses on enhancing the total energy efficiency and minimizing the number of thermal stages that require external heat, which is desirable to be carbon-free[3,64]. Their comparative advantage is their potential to produce high-strength SO_2 gas and enhanced heat integration with greater stability and continuity. However, the more tightly unit operations are integrated, the more coupled they will be, and therefore disturbances will spread more easily and potentially decrease operational robustness in case control systems and materials performance are not mature enough. Compared to others, such concepts typically have a positive theoretical niche in mass and energy balance studies, but their practice in industry requires an extremely long lifetime, the ability to tolerate impurities, and maintainability.

5.3. Techno-Economic Drivers and Sensitivity to External Conditions

Economic viability of carbon-free self-heating smelting is calculated as an aggregate of the cost of intensified oxygen supply, upgraded heat recovery, enhanced gas cleaning, and electrification of auxiliary heat requirement, offset by savings due to cut back in the cost of fossil fuel, better value of sulfur capture, and the prospective energy credits on the recovered heat[65,66]. Examples of capital expenditures that can dominate are oxygen plant, boiler upgrades, particulate control devices, and acid plant or tail-gas treatment enhancements, and operating expenses are usually dominated by electricity cost, oxygen requirements, refractory replacement rates, and the maintenance burden in dusty high-temperature sections.

The vulnerability to external conditions is especially strong. Carbon intensity and electricity price determine operating cost and net emissions performance, and these change greatly as a function of the region and over time. The composition of it, in particular, its level of sulfur, moisture, gangue, and composition impurities, strongly influences the autothermal margin, mass of slag, and chemical composition of the dust, and thus, alters the energy requirements and the needs of environmental control. Economic performance is also influenced by regulatory regimes that impose the costs of the cut of SO_2 and other air pollutants, tighter tail-gas regulations, or a carbon price. Consequently, no universal order of rankings of pathways can be established, and instead, the pathways can have a viable and economically appealing area of a multi-dimensional space characterized by characteristics of the concentrate, presence or absence of electricity, presence or absence of existing infrastructure, and regulatory restrictions[3,67].

5.4. Technical Barriers in Achieving Sustained Carbon-Free Self-Heating Operation

The major obstacle is the weakness of the heat-balance stability to realistic fluctuation [68,69]. Self-heating designs with no carbon emissions eliminate the use of auxiliary combustion, eliminating a standard way to mitigate deficits in autothermal margin in the short term. Changes in the moisture content of the concentrate, mineralogy, sulfur content, and blending may hence cause temperature instability, alterations in slag viscosity, or poor separation performance unless oxygen control and feed conditioning are sufficiently active. This problem is enhanced in strongly enhanced oxygen-blown regimes, in which the reaction processes can be rapid and local thermal gradients may be strong.

Second is materials performance. The presence of high oxygen potentials and high temperature activity enhances refractory corrosion and wears, particularly where there is a combination of turbulence and chemical attacks [70]. The formation of magnetite in slags may complicate operation further due to flow and heat transfer disruption via accretion formation that leads to higher viscosity. A stable protective freeze lining or control over refractory chemistry operating in enhanced circumstances is thus core to industrial viability, and its challenge is compounded when plants proceed to higher oxygen content levels in an attempt to do away with any remaining fuel need.

The third obstacle is related to impurity management and residue management. Numerous copper concentrates include large quantities of As, Sb, Hg, Pb, as well as halides, and their separation can change in harsh autothermal circumstances [71,72]. Should these species report to dust or acid streams in more of them, the cleaning of gases and the stabilization of residues will be more demanding and may raise levels of hazardous wastes or the amount of contaminants in acid that can be sold. Dust recycling to reuse valuable metals may form loop accumulations, which can raise the levels of impurities and the risk of emissions sprite, or cause a periodic purge stream to be discharged, which must be controlled. In this way, it becomes the impurity chemistry and the strength of stabilization pathways which limit environmental performance and not just sulfur capture.

There exists a fourth barrier of operational reliability and safety constraints [73]. Start-up and upset conditions may demand close thermal control to avoid freezing up or damage, and most plants have back-up burners or emergency operations, which involve the use of fossil fuel. Under the operational definition adopted here, these realities do not nullify the carbon-free steady-state claim, provided that start-up/upset fuel volumes are transparently reported and annualized emissions remain negligible relative to total output. A steady-state pathway that is fuel-free can still have non-trivial annual emissions, either because it frequently suffers an upset or because, due to considerations such as maintenance constraints, it has to start up again and again. To connect observed deployment barriers with actionable engineering responses, **Table 4** consolidates the dominant constraints, mitigation strategies, priority research needs, and the recommended KPIs required for comparable reporting and verification.

Table 4. Summary of key barriers, mitigation strategies, priority research needs, and recommended KPIs for verifying carbon-free self-heating operation and multi-pollutant co-control.

Barrier Category	Typical Manifestation in Carbon-Free Autothermal Regimes	Mitigation/Engineering Strategy	Research Need (What to Prove)	KPI(s) to Report
Heat-balance resilience	Temperature swings with feed moisture/gangue variability	Fast oxygen/feed control; buffering; improved feed prep; heat integration	Dynamic models + validation under variability	Autothermal margin; temperature variance; downtime events
Refractory & freeze-lining durability	Accelerated corrosion/erosion under high O_2 potential and turbulence	Slag chemistry control; materials upgrades; optimized injection	Long-duration campaigns; corrosion mechanisms	Campaign life; refractory wear rate; maintenance frequency
Slag chemistry & magnetite control	High viscosity, accretions, poor settling	Controlled oxygen potential; flux optimization; temperature window control	Coupled thermodynamics–computational fluid dynamics(CFD); slag property data	Slag viscosity proxy; magnetite fraction; Cu-in-slag losses
Dust/boiler deposition & corrosion	Fouling in WHR/ducting under altered dust chemistry	Staged cooling; sootblowing strategy; materials selection	Predictive deposition models; corrosion mapping	Boiler availability; pressure drop; cleaning intervals

Table 4. *Cont.*

Barrier Category	Typical Manifestation in Carbon-Free Autothermal Regimes	Mitigation/Engineering Strategy	Research Need (What to Prove)	KPI(s) to Report
Impurity loops (As/Hg/Sb/Se)	Accumulation via dust recycle; contamination of acid	Partition control; selective dust treatment; stabilized purge	Partitioning models; stabilization routes	Impurity mass balance closure; acid quality; hazardous residue tonnage
NO_x control under high intensity	Thermal NO_x increases with peak T	Staged oxygen; quench control; residence time management	Temperature–NO_x correlations in real furnaces	NO_x mass/t; peak-T proxy; O_2 staging effectiveness
Boundary/MRV credibility	"Carbon-free" claims omit oxygen/electricity or start-up fuel	Standardized accounting; continuous monitoring	Harmonized reporting standards	Scope 1 vs. 2 CO_2e; oxygen kWh/t; annualized start-up fuel

5.5. Barriers Specific to Off-Gas Integration and Multi-Pollutant Control

Carbon-free intensification also tends to generate higher SO_2 streams and smaller volumes of gases, which may enhance sulfur capture but also impose bottlenecks on cooling of gases, dust removal, and corrosion control [74]. The boilers and downstream ducts of waste heat could encounter increased deposition risks or corrosion in case the dust chemistry changes to be more adhesive or corrosive condensates. It is also possible to have acid plant integration become more sensitive to trace contaminants that poison catalysts or lower the quality of acids, particularly where increased operation causes an increase in the transfer of semi-volatile elements into the gas phase. Additional aspects that increase the significance of stable inlet conditions and good control of fine particulate and aerosols include tail-gas limits, since minor variations can transform into compliance risks in high-performance systems [75].

Comparatively, whether deep co-reduction is possible or not becomes frequently constrained by the ability of the capture train to run under high load conditions of dust and impurities at high efficiency, rather than by whether deep co-reduction is possible or not. This reliability is necessary through the co-design of furnace operation and off-gas treatment, with a controlled design of the cooling profiles, dust conditioning, and partitioning of impurities [76]. In the case of these couplings being ignored, systems can encounter burden shifting, e.g., more hazardous residue production or greater wastewater treatment requirements, and this nullifies the premise of synergy.

5.6. Priority Research Needs: Toward Predictive Design and Verified Environmental Performance

The focus of current research on carbon-free self-heating of copper smelting is less on individual laboratory phenomena and more on forecasting the result of such a system. The first requirement is coupled modeling, which combines thermodynamics, reaction kinetics, and fluid dynamics, and emissions formation and partitioning. Autothermal margin, matte grade control, slag chemistry evolution, dust generation, and trace element speciation at the same time, models would allow rational optimization of oxygen strategy, furnace configuration, and cooling architecture under the changeable feed conditions [77]. These models need to be confirmed by plant-scale measurements since small-scale testing cannot usually scale industrial turbulence, residence time distributions, and multistage condensation behavior.

The second priority is pilot and demonstration campaigns with a long duration that produce open and comparable datasets. Performance claims are often based on short tests or steady-state snapshots, which are insensitive to variability, maintenance limits, and the dynamics of loops of impurity recycle. Evidence of long-term demonstrations of normalized KPIs (such as data of start-up/shutdown frequency, refractory life, dust recycle ratios, and residue stabilization performance) would be valuable in enhancing the evidence base on which pathway was selected [3].

The development of integrated impurity management strategies that can support the process of decarbonization as well as the control of pollutants is the third priority. This

involves managing the location of impurity condensation, developing capture trains that isolate dangerous species into manageable streams, and developing stabilization paths that can be used with circular metal recovery objectives. Meanwhile, materials, such as refractories, protective lining, and corrosion-resistant alloys, need to advance so that high oxygen potentials and increased heat recovery can be achieved without an excessive maintenance burden [28,78].

The fourth priority is in the area of measurement, reporting, and verification. Since carbon-free claims are becoming more important in investment and policy, the capability to prove the system boundary emissions reduction becomes a necessity. It will need strong monitoring of the off-gas composition and flow, oxygen use and electricity consumption, and the destination of the captured pollutants in solids and liquids. Both stability in operations and credible accounting can be aided by digitalization and sophisticated control; however, it should be backed by high-quality sensors and be similar across sites and technologies to compare them.

Along with these research requirements, there is a suggestion that the move to carbon-free self-heating smelting is not as limited by the inherent availability of autothermal reaction heat as it is by the capacity to control coupled thermal, chemical, and environmental dynamics in a predictable and testable manner. The most effective developments will then be the ones that consider the furnace design, oxygen and heat systems, and the multi-pollutant control as an aspect that presents a consistent, quantifiable model that is in line with the industrial working reality [79,80].

6. Conclusions

Carbon-free self-heating smelting is a technically feasible and strategically significant route to decarbonize primary copper production and, at the same time, enhance influence over the flue gas contaminating the atmosphere. Its main assumption is that the exothermic oxidation of sulfide minerals and iron-bearing phases can be the major driving force of the thermal process to smelt and convert it, with the additional smelting and conversion so that routine fossil-fuel burning can be removed or significantly cut down when heat balance, oxygen supply, and process integration are integrated into one system. This paradigm, when combined with adequate oxygen enrichment and effective heat recovery, not only reduces the direct CO_2 emissions but also transforms the off-gas properties in a manner that can enhance downstream capture, such as lowering the volume of gases and increasing the strength of SO_2, which is conducive to the production of sulfuric acid and the minimization of fugitive losses.

Another point that has been raised in the review is that CO_2 and air pollutants can also be reduced in a synergistic manner, but with conditions. Measures of the same intensification that allow autothermal operation without the use of fuels can decrease the quantity of NO_x generated by fuel and intensify SO_2 captures, but can also increase the risk of thermal NO_x formation and modify dust generation, particle size distributions, and the fractionation of semi-volatile and toxic species like As, Hg, and Se. Such reconfigurations can transfer the burdens of stack emissions to residue captures, acid streams, or wastewater in the event that cooling, condensation control, particulate capture, and impurity management are not co-designed with the intensified furnace regime. Hence, plausible co-reduction must be achieved by system-wide optimization whereby the furnace, off-gas cooling and cleaning train, sulfuric acid plant, and residue stabilization routes are viewed as an environmental control architecture as opposed to a series of units.

In the considered classes of technologies, there is no standard furnace type. Both suspension-based and bath-based smelting provide their own unique strengths and limitations in terms of autothermal margin, feed flexibility, dust behavior, refractory life and retrofit. Their net decarbonization performance is highly boundary-specific, especially over the carbon intensity of electricity and the energy demand of oxygen production specific. This is why the advertisements of carbon-free functioning should be supported by the open account of oxygen and electricity consumption, the use of start-up and upset fuel, and the destiny of the trapped pollutants. The harmonized metrics and normalized reporting in the form of mass emissions and capture efficiencies per ton of blister copper or cathode is critical to compare alternatives and prevent inappropriate conclusions that can be drawn on the basis of concentration data only.

In the future, the ability to deploy intensified, oxygen-rich, low-fuel systems at realistic feed variability and at impurity loading levels will not be determined by the availability of reaction heat, but will be determined by operational and environmental robustness. Breaking down these obstacles

will entail the development of the next generation of predictive models, linking thermodynamics, kinetics, and fluid dynamics to dust and trace element speciation; demonstration campaigns that record long-term performance with standard KPIs; better refractory and corrosion-resistant materials in high-intensity environments; and combined impurity control and residue stabilization schemes to avoid the hazardous accumulation and shifting of burden. Measurement, reporting, and verification frameworks that have the potential of substantiating net emissions reductions across system boundaries and investment and regulatory decision-making are equally important.

In short, enhancing carbon-free self-heating copper smelting technology is a consistent path to achieving climate and air-quality goals, although the integration is required: oxygen supply must be optimized to be low-carbon electricity, waste heat recovery must be designed to work with dust and corrosion limits and multi-pollutant capture must be engineered to manage not only SO_2 and PM, but also the trace toxic species that are increasingly becoming the defining variables of environmental performance. With the copper industry facing an increased demand and pressure to tighten emissions controls, the shift towards non-incremental efficiency improvement via integrated carbon-free autothermal systems, where co-reduction of CO_2 and flue-gas pollutants is a verifiable system, is becoming an urgent priority and a viable future direction of next-generation smelter design and retrofit.

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