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Reducing the use of Fossil Carbon and Fuels to Defossilize the Electric Steelmaking Route: Evaluation of the Effects of Alternative C-Bearing Materials and Hydrogen in the Electric Arc Furnace

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ABSTRACT

Electric steelmaking is pivotal for the transition toward carbon-lean processes. Replacing fossil carbon and fuels with alternative nonfossil materials can contribute to enhancing the sustainability of this route. The investigations reported in this article explore the use of alternative carbon sources for slag foaming in the Electric Arc Furnace and the utilization of hydrogen in related burners. The effects of using alternative carbon sources are investigated via industrial trials and complementary simulations employing a flowsheet model of the entire electric route. The investigations demonstrate that, although alternative carbon materials can generally lead to fossil CO_2 reduction of up to 15% without negatively affecting the product or most process aspects, high ratios of certain materials, such as 30% tires, can compromise operational safety and result in poor slag foaming. Concerning hydrogen use in burners, simulations show that CO_2 reduction of up to 48% can be achieved in off-gases before post-combustion, accompanied by water vapor increases of up to 31%. Simulations also estimate an increase in hydrogen content in tapped metal of up to twice the reference value; however, this increase can be mitigated by standard vacuum degassing procedures.

1 | Introduction

The European steelmaking sector is undergoing a profound transformation to contribute to achieving the European Green Deal objectives[1] and to promote circular economy concepts according to the new EU Circular Economy Action Plan [2, 3]. Furthermore, recent geopolitical challenges and instabilities have negatively affected fossil material and fuel markets [4–6].

Within this framework, the electric steelmaking route plays a key role in supporting the defossilization of steel production. It is anticipated that the share of scrap-based electric routes will increase in the future [7] and that the electric arc furnace (EAF) will form the basis of novel routes, including direct reduction processes [8–10]

that produce direct reduced iron (DRI) or hot briquetted iron (HBI) to feed the EAF. Scrap-based steelmaking is also well integrated within a circular economy context, as it utilizes secondary raw materials as the main feedstock. However, significant quantities of fossil carbon-bearing materials and fuels are still utilized in electric steelmaking for specific purposes (e.g., slag foaming) or to satisfy part of the process energy demand. Therefore, the sustainability of this route can be further enhanced, and its impacts reduced by replacing fossil carbon and fuels with alternative non-fossil materials. This approach can contribute to reducing fossil CO_2 emissions and counteracting climate change, as well as decreasing the dependency of electric steelworks on unstable fossil carbon/fuel supplying countries.

The growing interest in these topics is clearly evident from the increasing body of literature addressing them.

One of the first overviews of biomass utilization in the ferrous metallurgical industry was provided by Wei et al. [11]; concerning the EAF-scrap-based route, biomass use in a cogeneration system is presented to produce electricity (used in the EAF) and heat. In the review paper by Echterhof [12], among other aspects, interesting results are discussed regarding the use of biomass carbonisates in the EAF: different features of these materials significantly affect the reactions (e.g., in terms of sequence) in which they are involved, influencing foamy slag formation and modifying the timeline of energy supply during the melting process. Mapelli et al. [13] also provide a comprehensive overview of promising results in the use of different renewable carbon sources in steelmaking industries but highlight major issues hampering full-scale industrial exploitation of these materials, such as lack of knowledge, availability, cost, and a still incompletely defined market for alternative carbon-bearing materials. Progress toward biocarbon use in EAF is examined by DiGiovanni and Echterhof [14], who analyze research activities at different scales and identify recent improvements and relevant limitations related to high reactivity and low density of biocarbon used for injection, for which some solutions (e.g., agglomeration) are proposed. Hosni et al. [15] emphasize that the possibility of using biochar in steelmaking is affected by its source and by the process within which it is used, but generally 20%–25% coal/coke substitution is achieved without issues, leading to 5%–50% CO_2 emissions reduction. Alternative carbon sources for steel production are analyzed in depth by Cirilli et al. [16], who consider, from a circular economy perspective, the use of nonbiogenic secondary carbon carriers, e.g., polymers from waste plastics and rubber from tires. Despite this recently renewed interest, the first investigations and initiatives on the use of renewable and alternative carbon-bearing materials in EAF actually date back to the early years of the third millennium [17–19]. For instance, the use of charcoal in EAF-based steelmaking for foaming slag and recarburization first received consistent attention [17], while Gorez et al. [18] describe one of the first investigations on the replacement of fossil carbon with tires in EAF, providing guidelines to preserve performance and safety. Moreover, Joulazadeh [19] investigates the replacement of coke and coal with end-of-life tires in foundry and steelmaking EAF; no detrimental effects were observed on the product, and a decrease in electricity consumption was obtained.

Australian initiatives on the introduction of renewable carbon in integrated and electric routes are described by Mathieson et al. [20] according to the Australian Steel Industry CO_2 Breakthrough Program established in late 2006. Different preliminary investigations were carried out by RWTH Aachen University between 2011 and 2015, focusing on the following aspects related to the use of alternative carbon-bearing materials [21–25] (especially biochar) in EAF: required features, slag foaming, economic analyses, evaluations of CO_2 savings, and reactivity. Zaharia et al. [26] monitored the effects of replacing coke with tires on slag foaming, FeO reduction, and off-gas production by considering different tire/coke ratios in laboratory work and investigating carbon/slag interactions.

Results of novel laboratory, pilot, and industrial trials can be found in more recent literature [27–29]. Bianco and Porisiensi [27] discuss analyses conducted on biomass/biochar features and describe laboratory, pilot, and industrial tests carried out to verify the compatibility of different alternative carbon sources with the EAF process: some issues connected to the features (e.g., density, volatile and fine fractions content) of the alternative carbon-bearing material are reported. In long-term industrial tests on the use of torrefaction biochar in EAF described by Cirilli et al. [28], no significant modifications in steel and slag composition were obtained, but fume temperature increased. Similar results are reported by Echterhof et al. [29] in tests on the use of palm kernel shells to replace fossil carbon in a 140-ton DC EAF. Kieush et al. [30] describe laboratory tests on slag foaming where biocoke is used as an alternative carbon source in EAF and report different behaviors for biocokes containing 5 wt% and 10 wt% of wood pellets: compared with coke, the first one provides better foaming behavior, while the second one shows slightly worse performance. However, all the conducted analyses show that biocoke is a viable alternative to coke in slag foaming.

Since field tests are limited due to intrinsic risks, simulation represents a valuable complementary tool for trials. For instance, Meier et al. [31, 32] use a dynamic model to compare the reaction rate of biomass with hard coals and to propose oxygen use control strategies in EAF. However, as highlighted in the paper by Mapelli et al. [13], existing knowledge gaps (e.g., regarding total achievable CO_2 emissions reduction) and barriers demand further dedicated investigations to pave the way for full-scale industrial use of renewable and alternative carbon-bearing materials.

On the other hand, hydrogen use in EAF burners is a more recent promising option to decarbonize the electric steelmaking route. Some works can be found in the literature demonstrating the suitability of industrial burners originally designed to be fed only with natural gas (NG) to be used with NG/H_2 blends containing 30 vol% H_2 without relevant design/structural modifications [33] and without significant changes in the combustion process or NO_x emissions [34]. Technology providers claim the readiness of EAF burners for use of NG/H_2 blends, even reaching full H_2 inlet [35, 36]. However, in this case as well, complete and consolidated knowledge on the effects of H_2 use in EAF burners on process and product has not yet been achieved. Few literature works address this topic, and they often refer to simulation studies, as industrial data remain quite limited. For instance, Schüttensack et al. [37] analyze variations in EAF off-gas features and liquid steel temperature, while a simulation-based techno-economic and carbon footprint assessment is proposed by Shahabuddin et al. [38].

Starting from this background, the present work aims to provide deeper insight into the effects of using alternative carbon-bearing materials and hydrogen on the EAF process and product. A synergistic approach is followed to investigate anthracite and foaming coal replacement by combining simulations and industrial trials that are complementary to each other. A detailed flowsheet model covering the entire EAF-based production route is utilized for simulations considering different types of alternative carbon sources, while experimental investigations are carried out on a full-scale industrial EAF. Different materials and replacement rates are considered, and their impact is evaluated with respect to process, product, fume production, CO_2 emissions, and safety.

The effects of hydrogen use in EAF burners are investigated through simulations using the previously mentioned model after some adaptations. The focus is to explore variations in the composition of EAF off-gases before post-combustion and in tapped

steel, with particular emphasis on hydrogen content due to its relevance and detrimental effect on the final product. As an added value compared to standard simulations, the availability of a flowsheet model covering the entire EAF-based route up to continuous casting enables preliminary assessment of whether the possible increase in hydrogen content in tapped steel can be compensated by standard secondary metallurgy operations, e.g., Vacuum Degassing (VD). Moreover, the integration of a proton exchange membrane (PEM) electrolyzer model allows estimation of the increase in electricity demand by the EAF area to produce the required hydrogen compared to standard EAF operation (i.e., with burners fed by NG).

2 | Experimental Section

2.1 | Reference Industrial Context

The scrap-based electric steelworks where field trials were conducted is also the reference plant considered in the study for model tuning and adaptation. The facility has one EAF with a capacity of ∼150 tons, which is loaded with two baskets. The EAF has three injectors through which primary and secondary oxygen and NG are introduced. The first and second injectors also inject foaming coal, while the third one injects lime. Slag foaming is initiated by adding anthracite through the 5th hole and then continued with the injection of foaming coal. Tapping can be considered the start of secondary metallurgy, since additions of deoxidants, slag formers, fluxes, and Fe-alloys are performed. Afterward, the ladle moves to the secondary metallurgy station, which includes two ladle furnaces (LF) with two wire injectors (one per LF), and one VD unit. Once the secondary metallurgy process is completed, the ladle is transported to continuous casting (CC). Special steels are the main products, and the different produced special steel grades can be clustered into the following steel families (i.e., groups of similar steel grades): alloyed case hardening, alloyed quenched & tempered, bearing, carbon case hardening, free-cutting, microalloyed, and spring.

2.2 | Overview of the Simulation Model and the Preliminary Adaptation Steps

For scenario analyses, a flowsheet model developed in Aspen Plus V11 is used, which represents the EAF production route up to the beginning of CC. The model was initially developed [39] for environmental and energy impact assessment under conventional and unconventional process conditions [40, 41] and has been adapted and upgraded over the years for detailed investigations on more specific aspects of the process, such as slag characterization [42–44]. The model considers all steps that generally characterize electric steelmaking: charge and melting, material additions and injections to the EAF, the EAF process itself, deslagging and tapping, additions at tapping and ladle transportation, LF treatment, VD and final stages of secondary metallurgy, as well as the initial stage of continuous casting with steel receipt in the tundish. Several standard unit blocks and customized calculators and design specification units are combined to consider different phenomena and to estimate the amount, temperature, and composition of molten steel at different process stages, slag amount and composition, required electricity, etc., and in general to compute mass and energy balances. The model

inputs (e.g., metallic and nonmetallic charges and additions, desired temperatures, gas injections, minimum VD pressure) and the data required for model tuning (e.g., steel composition) are variables that are typically measured in standard industrial practice. Therefore, the model can be and has been easily customized, validated, and used for different steelworks and steel families. The modularity of the model also enables adaptations to carry out specific analyses. This is the case for the adaptations required for investigations related to the replacement of fossil carbon used in the foaming process and of NG to feed the EAF burners.

Alternative carbon sources (e.g., biomass, biochar, tires, plastics) are modeled according to supplier data as non-conventional solids, i.e., materials that are not pure species, and are characterized through ultimate analysis, proximate analysis, and sulfur analysis due to lack of equilibrium and physical property data. Since oxygen and hydrogen content to be included in the ultimate analysis were not available, a dedicated model was used that estimates these parameters so that the High Heating Value (HHV) of the carbon-bearing material corresponds to the known value. The suitability of this approach was demonstrated by the good agreement between actual and simulated HHV for almost all materials considered, as shown in Figure 1. Only for plastics is there a higher deviation, which can be explained by inaccuracies in the available data; indeed, the value of 97.2 wt% fixed carbon is extremely high even for materials derived from waste plastics specifically designed for use in steelmaking processes (e.g., as reducing agents).

Alternative carbon-bearing material streams have been added to the model via additional blocks that manage their use and consider the related effects. Literature information [45–47] and industrial data from a first set of field trials (IND1_FT) on the replacement of anthracite added at the fifth hole (used to initiate foaming) were used to tune the model. Subsequently, the adapted model was tested using industrial data that had not been used in the tuning stage. Figure 2 compares the steel composition (iron content is not reported) before tapping and after secondary metallurgy for three exemplary heats belonging to three different steel families, demonstrating the good level of model accuracy. Similar results were obtained for other tested heats.

Regarding other process variables estimated by the model, Table 1 depicts model performance in terms of mean average percentage error (MAPE) of the provided estimates, which is defined as follows:

$$\text{MAPE} = \frac{100}{n} \sum_{i=1}^{n} \left| \frac{V_r - V_s}{V_s} \right| \quad (1)$$

where n is the number of observations (i.e., tests simulations), V_r are the actual industrial values, and V_s are the simulated values.

Excluding LF slag, for which heat-based comparisons were not possible, MAPE values are all below or equal to 13.5%; therefore, model accuracy was considered suitable for the purposes of the investigations to be carried out.

Further minor adaptations were needed to simulate hydrogen use in EAF burners. Primarily, streams and dedicated blocks were added to the model to manage H_2 blends with NG. For instance, design specification blocks managing gas flows were
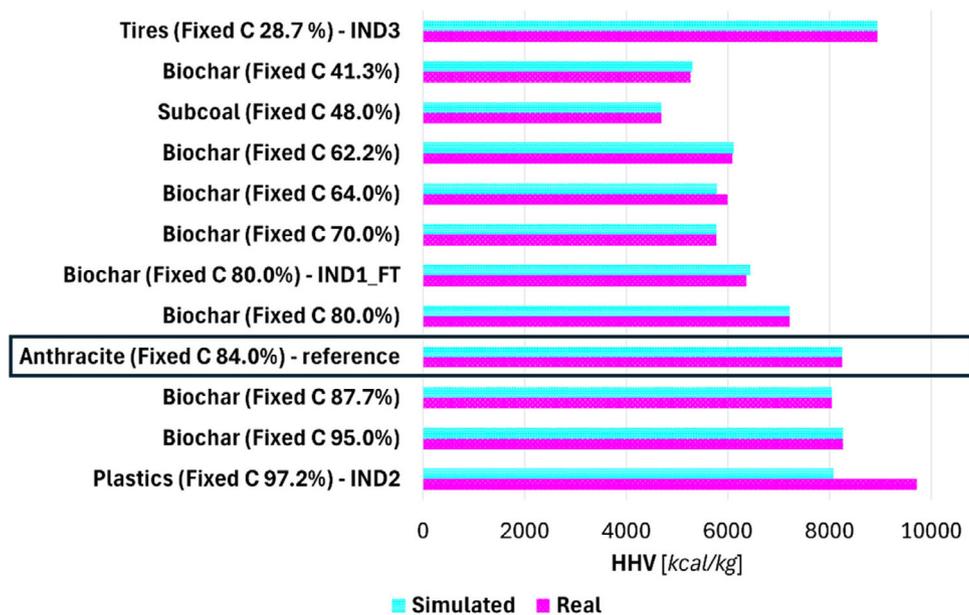
FIGURE 1 | Comparison between actual and simulated HHV of modeled carbon-bearing materials, including reference anthracite. The labels IND1_FT, IND2, and IND3 refer to materials tested in industrial trials (see Section 2.3.1); FT is the acronym for First Trials.

added to ensure that the same energy is provided by burners when fed with different blends. Previously cited literature works were used as references for some adjustments. Additionally, an available PEM electrolyzer flowsheet model [48] was integrated into the EAF model in scenarios related to hydrogen use in burners to estimate global EAF electricity demand, including the contribution required to produce the amount of hydrogen used in the burners.

2.3 | Description of the Experiments

2.3.1 | Alternative C-Bearing Materials

The replacement of fossil carbon materials used in the foaming process was investigated by coupling simulation and industrial trials. The aim is twofold: first, to find substitutes for fossil carbon with similar features (the features of anthracite are taken as reference) that do not adversely affect process and product, and second, to assess the effects of using alternative carbon-bearing materials with features inferior to the reference one.

Simulations supported "broader spectrum analyses" of the effects on various process and product variables (e.g., electric energy, fossil CO_2 emissions, steel composition, slag amount) of a larger number of alternative carbon sources, while industrial trials focused primarily on foaming performance and safety issues that represent aspects not covered by our simulations. Specifically, the purposes of the conducted simulations are listed in Table 2.

In industrial trials, several tests were conducted with alternative carbon-bearing materials exemplified in Figure 3, covering the production of steel grades belonging to all steel families mentioned in Section 2.1. Three of the four alternative carbon-bearing materials were considered in simulations (see Figure 1): IND1_FT (Figure 3a), IND2 (Figure 3c), and IND3 (Figure 3d). The biochar in Figure 3b was not simulated, as some parameters fundamental

for appropriate modeling and validation (e.g., HHV) were unavailable. It is a mixture of two similar biochars having the following proximate analysis range: moisture 3.9%–4.2 wt%, fixed carbon 78.5%–81.8 wt%, volatile matter 15.1%–16.6 wt%, and ash 3.0%–4.9 wt%, but the mixing ratio is not available.

The two types of biochar (IND1 trials) were used to replace anthracite due to their higher granulometry; therefore, they were introduced through the 5th hole. Tests with a third type of biochar (not shown in Figure 3) were cancelled because it underwent self-ignition in the hopper. Plastics (IND2 trials) and tires (rubber only—IND3 trials), which have lower granulometry, were tested to substitute foaming coal and were therefore injected directly by the available standard cojets into the liquid steel bath; different percentage mixtures of these materials and fossil coal were tested. Notably, all tested materials have densities lower than fossil coal (i.e., ~900 kg/m³). In particular, plastics have a density of ~400 kg/m³.

Both EAF and production parameters were monitored during trials, e.g., off-gas temperature, acoustic and electrical noise levels, oxygen content in liquid steel, and slag foaming through foaming index, viscosity, and FeO content.

2.3.2 | Hydrogen Use in EAF Burners

Simulations concern gradual replacement of NG used in standard operation (i.e., in the considered cases, 1.1 Nm³NG/t_{tapped_steel} on average) with hydrogen by ensuring that the same amount of energy is provided to the EAF. In the cases considered, complete NG replacement corresponds to ~3.6 Nm³H₂/t_{tapped_steel} on average.

EAF off-gases, steel composition, and other process parameters (e.g., required EAF electric energy, slag amount and composition) are monitored. The simulations include the entire secondary metallurgy station to assess the suitability of current VD operation to mitigate negative impacts on tapped steel (i.e., related to hydrogen
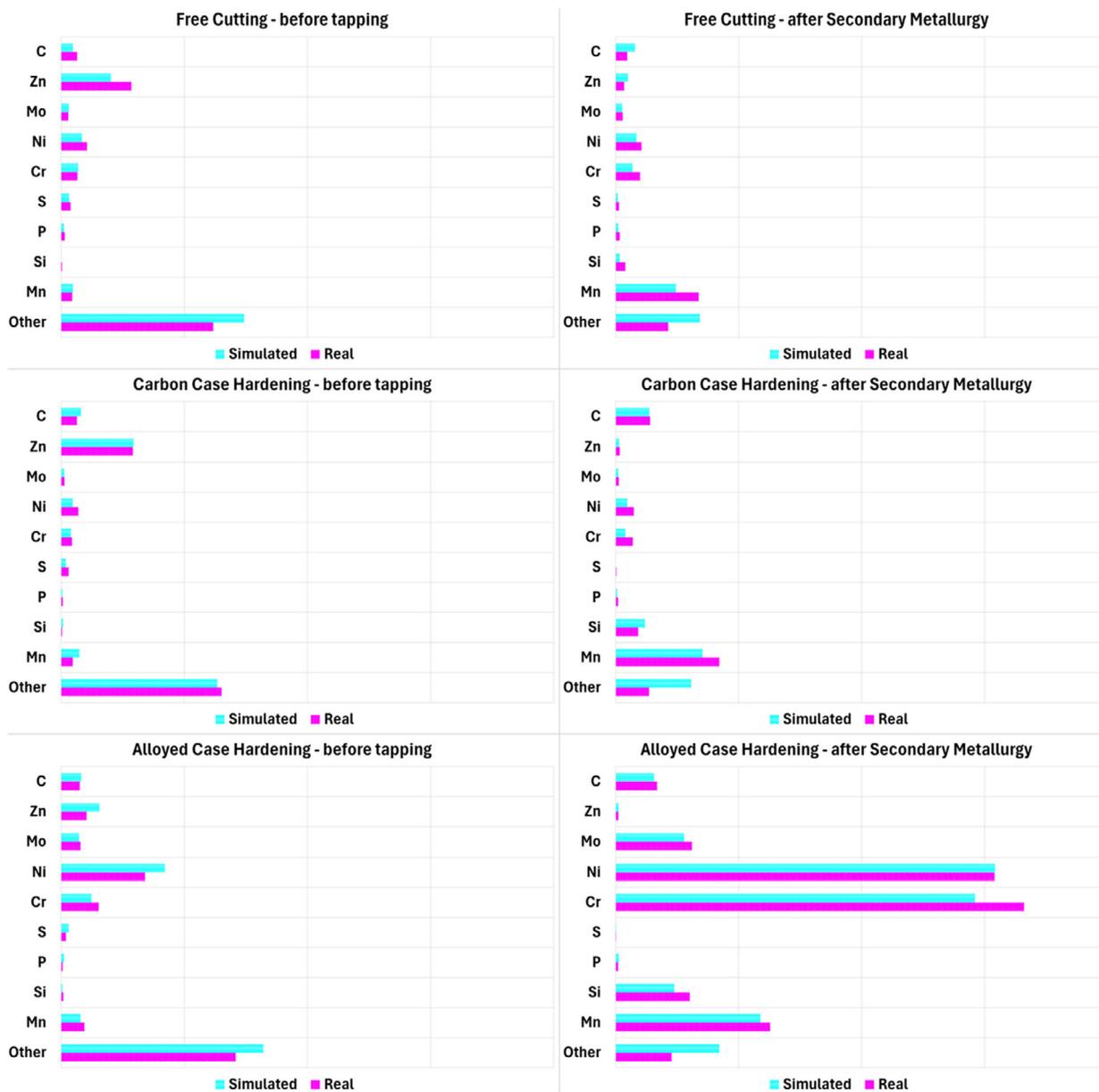
FIGURE 2 | Comparison between actual and simulated steel composition before tapping and after secondary metallurgy for three tested heats belonging to three different steel families (axes scales and values are not reported due to industrial confidentiality constraints).

TABLE 1 | MAPE of adapted model in estimating selected EAF-based route variables.

Variable	MAPE
Tapped liquid metal	8.0%
Liquid steel after secondary metallurgy	11.3%
EAF slag	6.1%
LF slag[a]	22.6%
EAF electricity demand	5.2%
LF electricity demand	13.5%

[a]Based only on average industrial data due to lack of heat-based data.

content increase). In addition, hydrogen production with PEM is simulated, and the increase in EAF area electricity demand is estimated.

3 | Results and Discussion

3.1 | Alternative C-Bearing Materials

Various interesting results were obtained in simulations and industrial trials, demonstrating the complementary roles of the investigations conducted.

3.1.1 | Field Trials Results

The biochar used in IND1 trials for replacing fifth-hole anthracite did not lead to production issues or operational problems. Production parameters were not negatively affected, and both steel quality and slag conditioning met the required performance standards.

In contrast, different behaviors were obtained when replacing fossil foaming carbon with plastics (IND2) or with tires (IND3). The main effects are shown in Figures 4 and 5, which

TABLE 2 | Schematic description of the carried out simulations.

| Simulation ID	Main objective	Subcategory	Note
SIM1	Assess the effects of replacing anthracite charged in EAF 5ᵗʰ hole for slag foaming (representing less than 15% of total fossil carbon input)	a	Fixed carbon input
		b	Fixed energy supply
SIM2	Conduct sensitivity analyses on the contents of different compounds in biochar	n.a.	n.a.
SIM3	Investigate the effect of full replacement of fossil carbon for foaming (anthracite + foaming coal) with biochar used in first industrial trials (IND1_FT, see Figure 1), having inferior features compared to reference anthracite	n.a.	Considered biochar main features: • Fixed C content = 80% wt • S content = 0.8% wt • HHV = 6360 kcal/kg
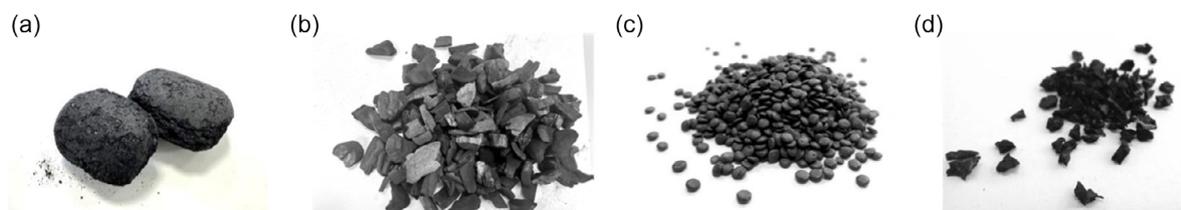
FIGURE 3 | Alternative carbon materials used in industrial trials: (a,b) biochars used to replace anthracite ((a) was tested in first trials (IND1_FT, see Figure 1), whose data were used in model tuning and testing). (c,d) plastics and tires (IND2 and IND3, respectively, see Figure 1), used to replace foaming coal.
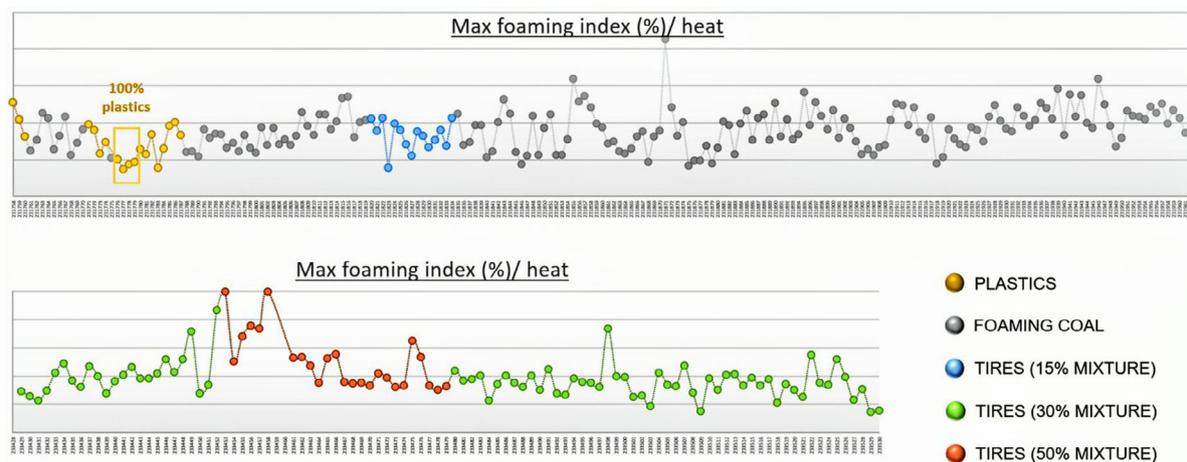
FIGURE 4 | Maximum foaming index trend for different types of injected foaming carbon material; on the *x*-axis, the heat number is reported, while *y*-axis values are not reported due to industrial confidentiality constraints.

represent the maximum foaming index and fume temperature obtained for different heats with standard and alternative carbon sources.

During IND2, although no negative effects were observed on O_2 content at tapping or electric energy consumption, foaming performance was poor. The plastics burned on the surface even before reaching the liquid steel and created flames, resulting in increased fume production and elevated temperature (exceeding the safety trigger). This was more evident when a high ratio of fossil carbon was replaced; unsafe operating conditions were obtained using only plastics, as shown in Figure 6.

During IND3, no negative effects were observed on O_2 content at tapping, but tires showed lower foaming performance compared with coal: high noise levels and worse foaming index were obtained with increasing tire percentage. In this case as well, fume temperature increased, although to a lesser extent compared with IND2. However, when the percentage of injected tires in the carbon-bearing foaming materials mixture reached 30 wt% and 50 wt%, safety alarms were activated due to strong
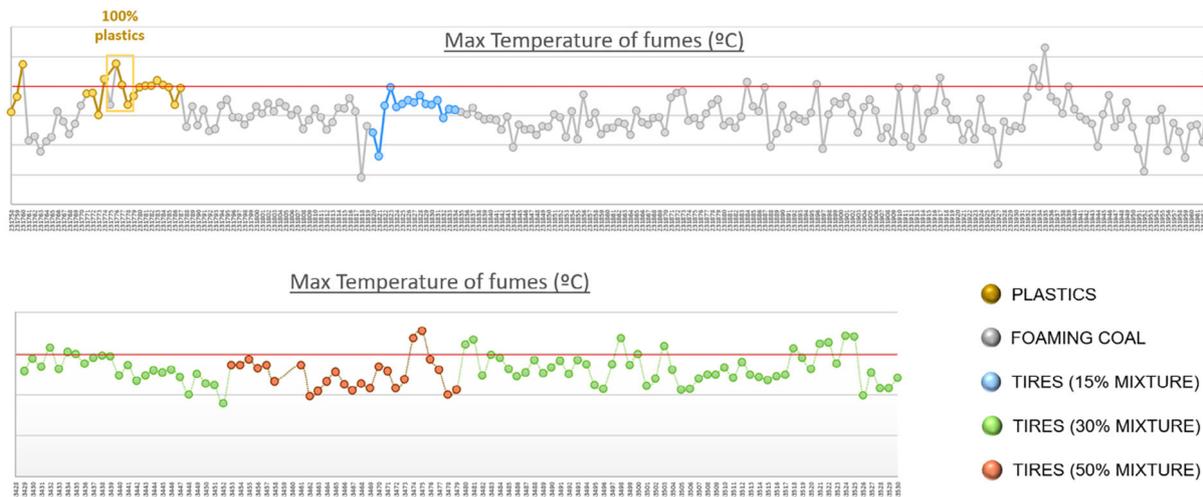
FIGURE 5 | Off-gas temperature trend for different types of injected foaming carbon material; the red line indicates the desired maximum limit; on the x-axis, the heat number is reported, while y-axis values are not reported due to industrial confidentiality constraints.
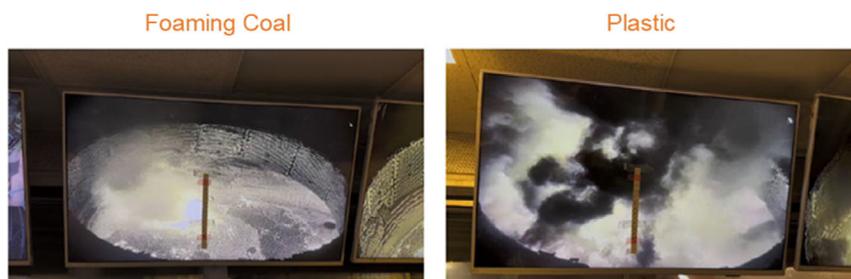
FIGURE 6 | Effects inside the EAF during slag foaming using standard fossil foaming coal (left) and plastics (right—IND2).

fume production and temperatures exceeding 600°C (see exemplary Figure 7). Only with 15 wt% of tires in the foaming materials mixture were no problems observed with off-gas temperature.

The operational issues observed are generally related to the lower density of alternative materials compared with standard fossil coal. This leads to lower penetration of these materials into the molten steel and reduced capacity to react with slag formers.

Considering all industrial trials, no differences were observed depending on the produced steel grade.

3.1.2 | Simulation Results

In SIM1, the effect of the considered carbon-bearing materials was not evident on many monitored parameters, but some useful outcomes are depicted in Figures 8 and 9. The figures show exemplary outcomes of SIM1a and SIM1b obtained by simulating production of heats belonging to two considered steel families. Similar results were obtained for other steel families as well. In SIM1a, electric energy consumption decreases when using tires, since a high tire amount is required to reach the desired fixed carbon fed, thus providing higher chemical energy. However, since tires have the highest sulfur content among the considered carbon-bearing
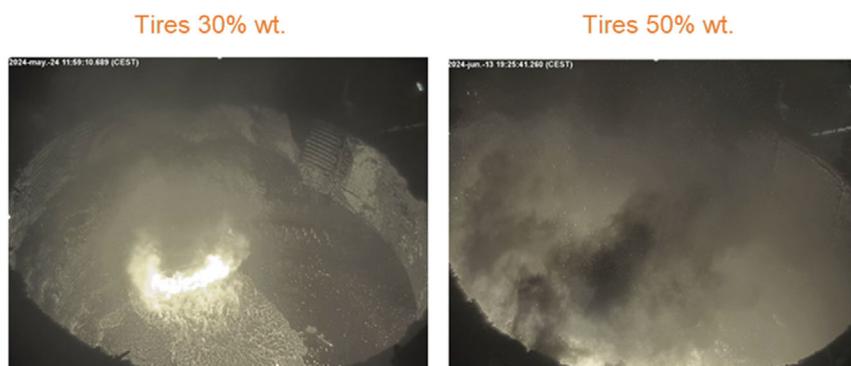
FIGURE 7 | Effects inside the EAF during slag foaming using different tire contents (IND3) in the foaming materials mixture.
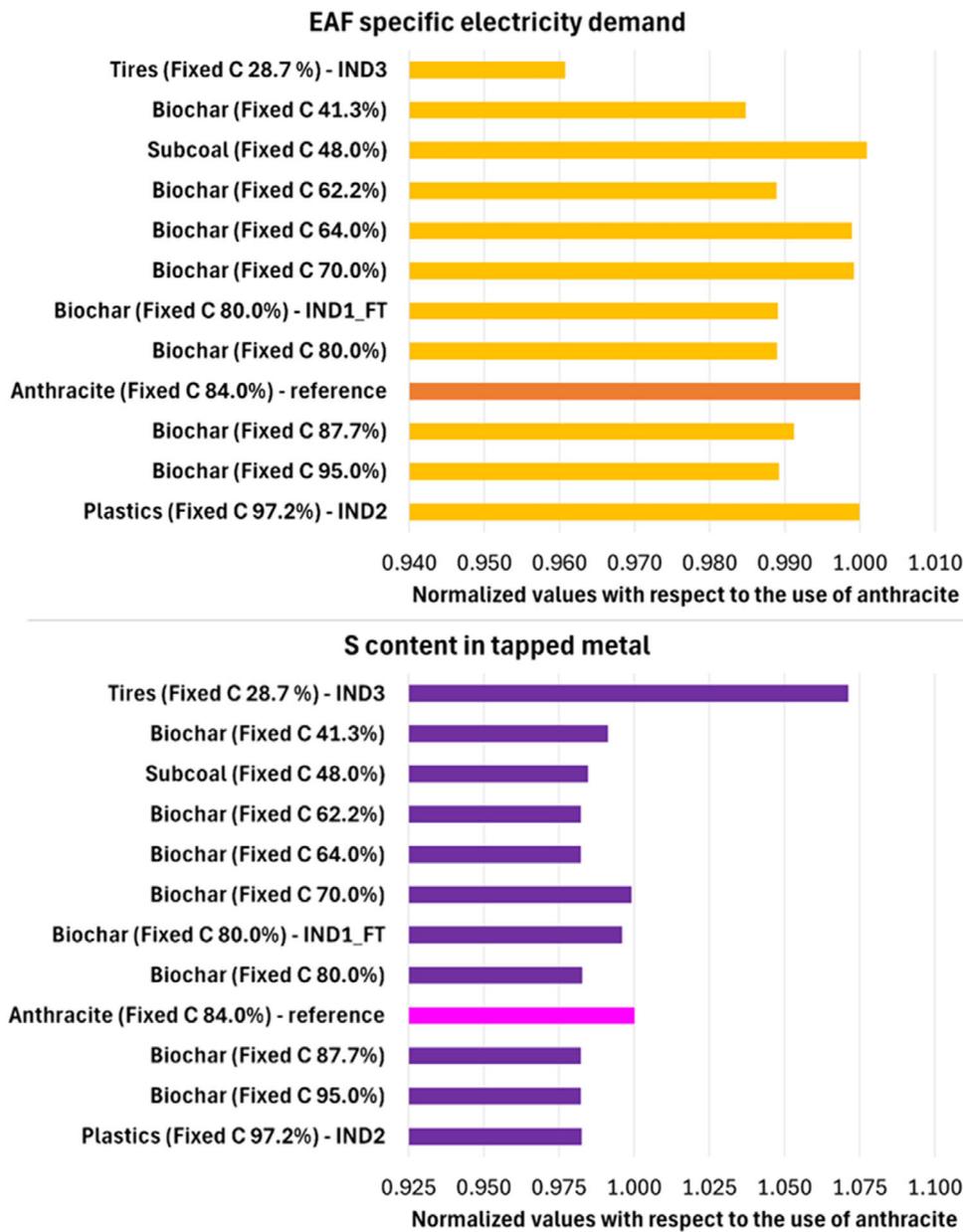
FIGURE 8 | Example of SIM1a outcomes for simulation of heat production belonging to free-cutting steel family.

materials, they lead to the highest sulfur content in tapped metal. Interestingly, the lowest carbon content in tapped metal is also obtained with tires in SIM1b; this can be explained by the highest volatile content in tires compared with other carbon-bearing materials. Nevertheless, a notable decrease in fossil CO_2 emissions in the EAF can be obtained by substituting anthracite with alternative carbon sources.

To analyze in depth the impact on sulfur and carbon contents in tapped steel of feeding tires instead of anthracite via the fifth hole, additional SIM1a and SIM1b scenarios were tested to simulate gradual replacement of anthracite with tires. Figure 10 depicts results obtained for the same heats considered in Figures 8 and 9: carbon and sulfur contents in tapped steel show nearly linear dependence on the percentage of anthracite replacement.

When fossil carbon is used in the foaming process, sulfur content in tapped steel for the Free-Cutting family (i.e., the exemplary family of results reported in Figure 10a) generally lies in the range 0.027 ± 0.004 wt%, while for 25% and 100% anthracite replacement, it exceeds this range for 17% and 33% of simulated heats, respectively. Moreover, for 5% of simulated heats, sulfur content is higher than the maximum value normally measured for this family (i.e., 0.038 wt%) for replacement rates higher than 25%. However, industrial tests demonstrated that such replacement rates are not feasible for operational and safety reasons.

SIM2 simulations exhibit nearly linear correlations of the main monitored parameters with the contents of fixed carbon, sulfur, and moisture in biochar. Notably, carbon content in tapped metal increases linearly with increasing fixed carbon content in biochar: the slope of the straight line ranges between $\sim 3 \cdot 10^{-5}$ and $9 \cdot 10^{-5}$ depending on the simulated steel family. Furthermore, electric energy consumption at the EAF also increases linearly with increasing moisture content in biochar.
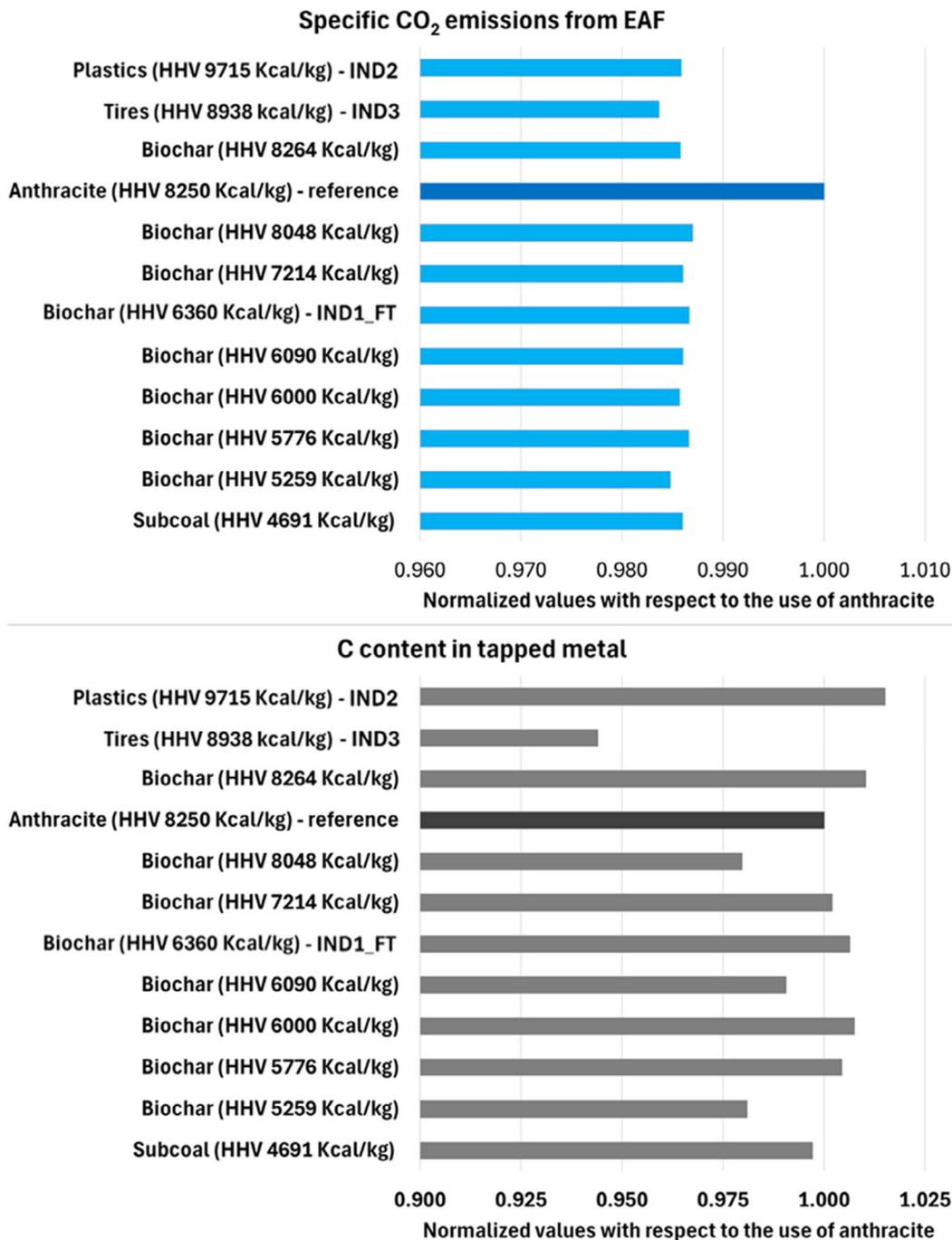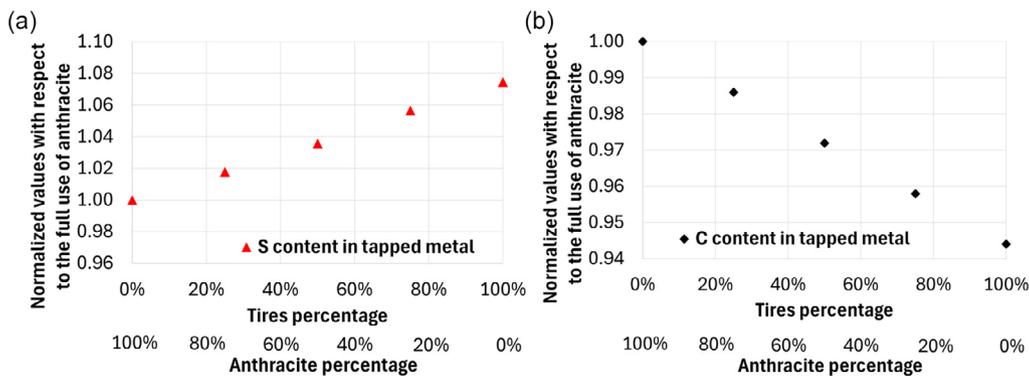
FIGURE 9 | Example of SIM1b outcomes for simulation of heat production belonging to carbon case hardening steel family.

Finally, SIM3 scenarios show that even using biochar with inferior features compared to reference anthracite, ~15% fossil CO_2 emissions reduction can still be obtained by complete replacement of fossil carbon used for the foaming process. Other interesting results include an 8%–11% decrease in sulfur content in tapped metal and a 3%–7% decrease in EAF slag mass quantity. In some cases, an increase in required electric energy of ~2%–8% was observed, which depends on the lower HHV of this biochar compared to reference anthracite; however, the extent of such increase depends on the produced steel family and related operating practices. Carbon content in steel and EAF metallic efficiency remain essentially stable.

3.1.3 | Summary of Achievements and Comparison with Literature

In the literature, several types of biochar have been separately investigated on different EAFs and at various experimental scales. The procedures for feeding them to the EAF are also variable and can differ from those considered here. However, to enable viable and continuous use of carbon alternatives in standard operating practice, any actual steelworks needs assurance that many feedstocks are suitable so that they have a sufficiently wide portfolio of solutions to adopt for at least partial replacement of fossil carbon sources, performing selection based on costs, availability, and main features. Therefore, in this work, a wide range of non-fossil

FIGURE 10 | Impact of using tires on steel quality: (a) trend of sulfur content in SIM1a scenarios and (b) trend of carbon content in SIM1b scenarios versus percentage of anthracite replacement with tires.
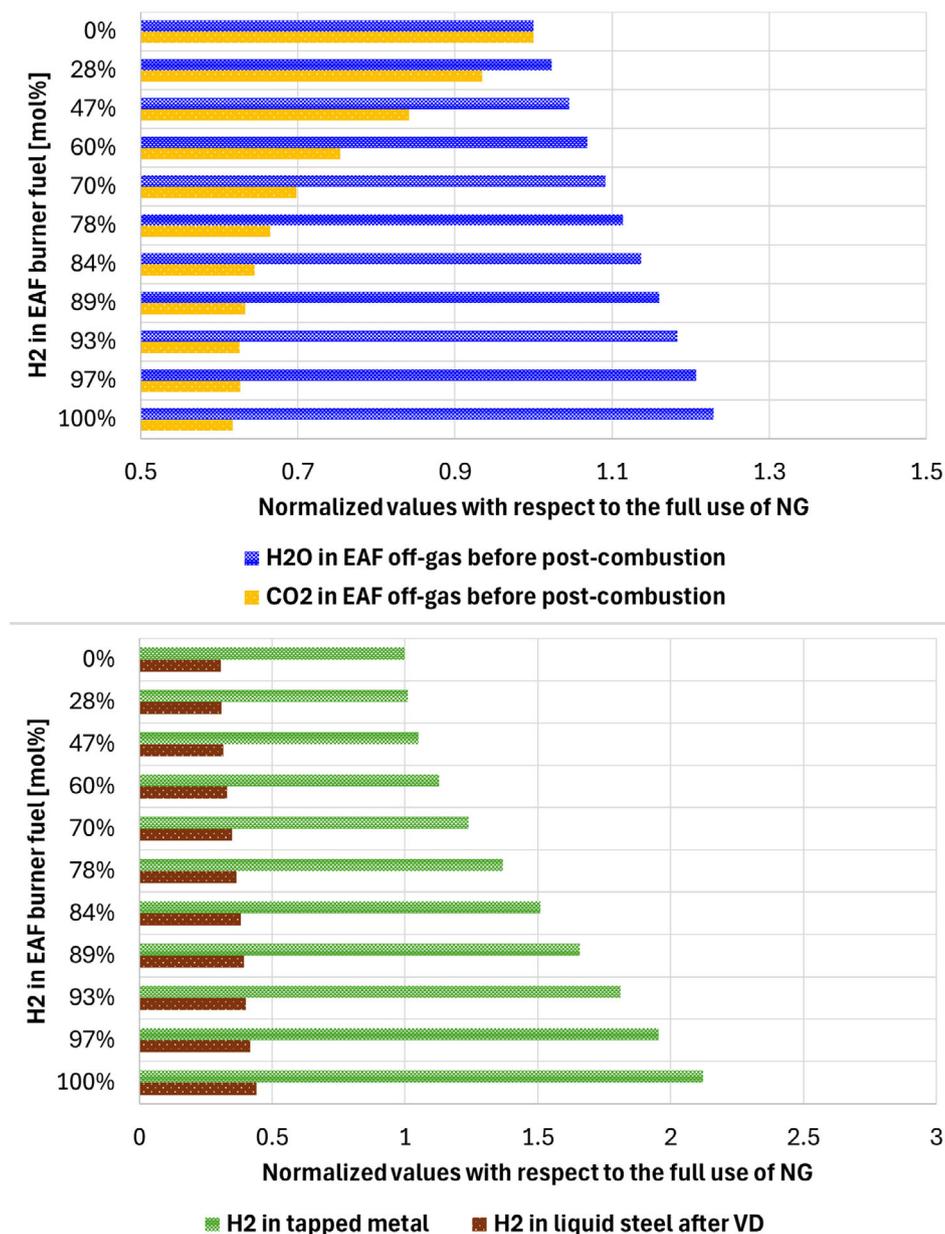
FIGURE 11 | Example of outcomes for simulation of hydrogen use in EAF burners for production of a heat belonging to the alloyed case hardening steel family: H_2O and CO_2 contents in EAF off-gas before post-combustion (top); H_2 in tapped metal and in liquid steel after VD (bottom).

carbon-bearing materials, including low-quality ones, were tested on the same EAF, where two feeding modes are typically adopted. The results obtained in all biochar-based industrial and simulation trials are aligned with the literature: no detrimental impacts are observed on steel and furnace operations, as stated by Echterhof [12] and as found in laboratory, pilot, and industrial trials carried out by Demus et al. [22], Reichel et al. [24], Echterhof et al. [29], Bianco et al. [27], and Cirilli et al. [28]. Only carbon and sulfur contents in steel are slightly affected, showing a linear correlation with the contents of these elements in the carbon-bearing material. This was also observed by Demus et al. [22] and Echterhof et al. [29], although they analyzed far fewer materials compared to the present work. Moreover, electricity demand increases with the moisture content of all alternative carbon-bearing materials, while tires generally lead to decreased electricity demand, as reported by Joulazadeh [19].

The obtained reduction in fossil CO_2 emissions, although dependent on EAF operating conditions and produced steel, ranges from ~1.5% (for lower replacement rates) up to ~15% (for complete replacement of fossil carbon used in the foaming process). These results match the values provided by Mathieson et al. [20] (i.e., between 1.6% and 3.1%) and Echterhof et al. [21] (i.e., between 12% and 15%).

Plastics, tires, and, in general, materials with lower density and granulometry and higher volatile matter contents lead to issues related to handling, lower/unstable slag foaming, higher reactivity, and consequent increases in fume temperature and flame intensity, in line with literature results for rubber [26], tires [19], and fine and low-density biochars [14, 23, 27–29, 31, 32]. Simulations also show that tire addition increases sulfur content in tapped steel. However, industrial trials demonstrated that replacement is viable only below a 30% rate, where the increase in sulfur content is tolerable in most cases.

To summarize, our combined experimental and simulation analyses of different types of alternative carbon-bearing materials confirm their suitability for slag foaming and expand the range

of potential options to replace fossil carbon to reduce the carbon footprint of electric steelworks without significant impacts on process and product. Moreover, several barriers and constraints for the use of alternative carbon-bearing materials in EAF are identified: fixed carbon content and density must be as high as possible, volatile matter must be lower than 20 wt%, moisture must be as low as possible, and sulfur content must be limited.
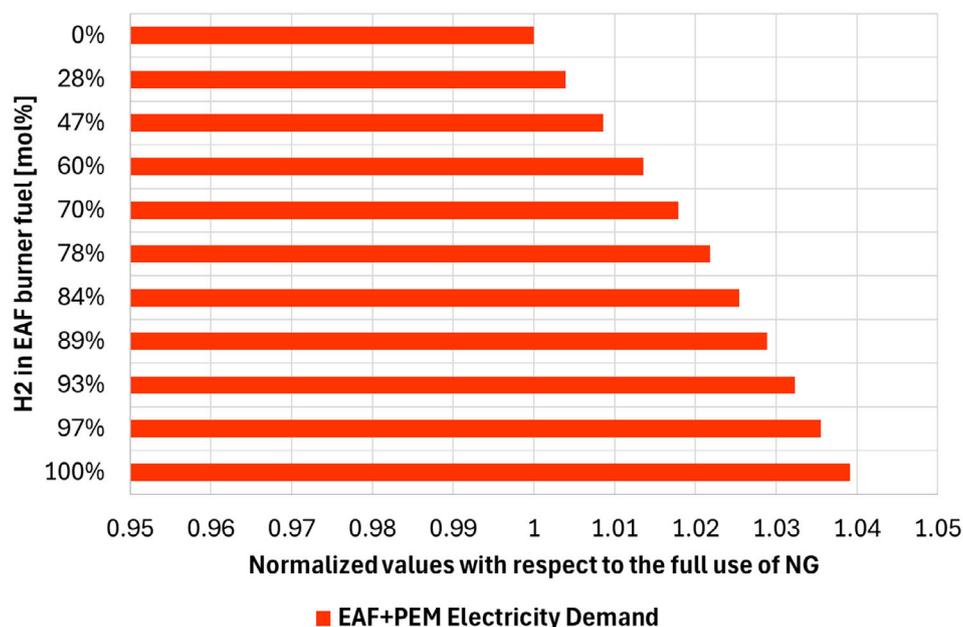
3.2 | Use of Hydrogen in EAF Burners

The most interesting simulation results concerning the effects on process and product of replacing NG with H_2 in EAF burners are summarized in Figure 11, which shows the outcomes for an exemplary heat producing steel belonging to the Alloyed Case Hardening steel family. Similar behaviors were obtained for other steel families as well.

The complementary nature of CO_2 and water vapor in EAF off-gas before post-combustion can be observed: considering all performed simulations, by increasing the H_2 ratio, CO_2 content decreases by ~38%–48% and H_2O increases by ~23%–31%.

The change in off-gas composition was expected. It was also observed by Schüttensack et al. [37], who, however, highlight that CO_2 decrease is not a fixed value but is highly dependent on EAF operating practices and the burner's role.

The H_2O increase can partly explain the increase in H_2 content in tapped steel (up to ~2.5 times considering all simulations): an H_2O-richer atmosphere promotes H_2O splitting at the electrode and, together with unreacted H_2, can contribute to H_2 content in tapped steel. To the best of the authors' knowledge, no other works in the literature have explored this aspect thus far, which lies beyond the scope of the present work but can be explored in future investigations. However, according to the results of secondary metallurgy simulations, VD procedures currently used in the considered steelworks are suitable for maintaining H_2 content at adequate levels (e.g., for the steel family considered in

FIGURE 12 | Example of EAF area electricity demand to produce a heat belonging to the alloyed case hardening steel family when hydrogen used in EAF burners is produced via PEM electrolysis.

Figure 11, it must be lower than 1.5 ppm) even if its concentration in tapped steel is higher.

If the required hydrogen is produced via PEM electrolysis with a specific energy consumption of $\sim$55 kWh/kg$_{H_2}$ on average [48], EAF area electricity demand increases, as exemplified in Figure 12 for the same heat considered in Figure 11. In this case as well, similar results were obtained for all considered steel families.

Overall electricity demand of the EAF area increases by 0.4%–0.8% for burner fuel containing $\sim$30 vol% H$_2$, and this increase reaches 3.9%–4.4% when pure H$_2$ is used. These values correspond, respectively, to overall specific electricity consumption of $\sim$1.6–1.9 kWh/t$_{tapped_steel}$ and 16.2–19.2 kWh/t$_{tapped\_steel}$. These figures are relatively low due to low NG usage in standard practice of the considered EAF and particularly in the simulated cases (see Section 2.3.2).

4 | Conclusions

Considering the defossilization demand of electric steelworks, simulation and industrial trials can be profitably combined to investigate the effects on EAF process and product of replacing fossil carbon and fuels with alternative carbon-bearing materials and green hydrogen.

The combined analyses carried out in the described research work provide the following results concerning the use of alternative carbon-bearing materials in EAF:

- Replacing fossil carbon in the foaming process with different types of biochar can reduce CO$_2$ emissions by up to 15% without critical effects on process or liquid steel. However:

  - Carbon and sulfur contents in tapped steel increase with the increase of related contents in alternative carbon-bearing materials.

  - Electric energy demand increases with the increase of moisture content in alternative carbon sources and also depends on volatile matter and HHV.

- All considered alternative carbon-bearing materials represent promising options to replace fossil carbon, although with different viable replacement rates, especially if one wants to use them in a standard EAF. Plant adaptations may be required to use some of these materials. In particular:

  - In some cases, biochar can undergo self-ignition; thus, suitable handling procedures must be implemented.

  - Low foaming performance, high acoustic and electrical noise, intense flame and fume formations, and high fume temperature (>600°C) were obtained with high percentages of plastics and tires (>15 wt%) injected with standard installed cojets, as they react on the molten bath surface without suitable penetration into molten steel. Therefore, adaptations are required, e.g., modification of injectors or adjustment of injection speeds.

Concerning the use of green hydrogen to replace NG in EAF burners, the following conclusions can be drawn:

- The use of hydrogen in EAF burners is a promising solution to decrease CO$_2$ by up to 48% in EAF off-gases before the post-combustion step, and it is also flexible, as the replacement rate can be adjusted based on green hydrogen availability.

- High replacement of NG with H$_2$ in EAF burners leads to increased H$_2$ in tapped steel, but standard VD operating practices appear suitable to mitigate such an increase.

- Production of green hydrogen via PEM electrolysis increases EAF area electricity demand by up to 19.2 kWh/t$_{tapped\_steel}$ in the considered cases.

Finally, although the present work explored a wide range of non-fossil carbon-bearing materials and green hydrogen to provide a sufficiently wide portfolio of solutions to adopt, it is worth noting that the viability of different options depends not only on technical factors, which are the focus of the present investigation. Economic and logistic aspects greatly affect the availability and costs of both alternative carbon-bearing materials and green hydrogen and vary over time and across different European countries. These aspects are, however, beyond the scope of the present investigation.

5 | Future Work

The proposed investigation methodology provides outcomes aligned with other literature results and enables deeper understanding of the effects of the considered solutions to decrease the carbon footprint of EAF-based steel production. Therefore, further investigations and simulations are ongoing. Pilot trials on hydrogen-enhanced combustion with standard EAF burners and novel hydrogen burners are being carried out together with simulations of process integration solutions and optimized approaches to manage alternative nonfossil materials and fuels. Moreover, alternative fuels (e.g., biomethane) or further hydrogen production methods (e.g., biomethane pyrolysis) will be investigated as further options (possibly combined with those considered here) to improve the sustainability of EAF-based steelmaking route.

Author Contributions

Ismael Matino: conceptualization (lead), data curation (equal), formal analysis (equal), funding acquisition (lead), investigation (lead), methodology (lead), software (lead), validation (lead), visualization (lead), writing – original draft (lead), writing – review and editing (supporting). **Orlando Toscanelli:** data curation (equal), formal analysis (equal), investigation (equal), methodology (equal), software (equal), writing – review and editing (supporting). **Antonella Zaccara:** data curation (equal), formal analysis (equal), investigation (equal), software (equal), validation (equal). **Aintzane Soto Larzabal**: conceptualization (supporting); data curation (lead), funding acquisition (equal), investigation (lead), methodology (equal), resources (equal), supervision (supporting); visualization (equal). **Asier Zubero Lombardia**: data curation (equal), investigation (equal), visualization (supporting); writing – review and editing (supporting). **Jon Hermosa**: data curation (equal), investigation (equal), methodology (equal), writing– review and editing (supporting). **Tamara Rodríguez Durán**: investigation (equal), methodology (equal), project administration (supporting); resources (equal), supervision (supporting); validation (supporting); visualization (equal), writing – review and editing (lead). **Valentina Colla**: conceptualization (lead), formal analysis (equal), funding acquisition (lead), investigation (supporting); methodology (equal), project administration (lead), resources (equal), supervision (lead), validation (equal), visualization (supporting); writing – original draft (supporting); writing – review and editing (supporting).

Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

Data associated with this article cannot be disclosed due to confidentiality requirements of the involved steelmaking facility.

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