

Review

Review on the Use of Alternative Carbon Sources in EAF Steelmaking

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Abstract: Steelmaking in the electric arc furnace (EAF), either scrap-based or based on hydrogen direct reduced iron, will in future contribute substantially to the reduction of CO₂ emissions in the iron and steel industry. However, there still will be the need to introduce carbon into the EAF process either to carburize the steel or to create foaming slag to improve the energy efficiency of the melting process. So, to reach the emission reduction goals set around the world, it will be necessary to substitute fossil charge and injection carbon used in EAF steelmaking with alternative carbon sources. This review presents the recent research on carbon-neutral biomass-based and circular rubber or plastics-based carbon sources and their potential to substitute fossil charge or injection carbon in the EAF process. It also discusses the current state-of-the art and suggests further opportunities and needs for research and development to use alternative carbon sources to produce a really green and carbon neutral and/or fully circular steel.

Keywords: electric arc furnace (EAF); steelmaking; carbon sources; biomass; plastics



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1. Introduction

Apart from the energy production sector, the iron and steel industry is one of the biggest consumers of fossil coal around the world and therefore also one of the biggest industrial emitters of CO₂. In the countries of the European Union (EU-27, as of 2007 to 30 June 2013), between 4% and 7% of anthropogenic CO₂ emissions are estimated to be originating from iron and steel production [1]. According to the International Energy Agency [2], the steel industries coal consumption accounts for 13.7% of the world's total annual production or 1.1 billion t in 2013. About 80% of this coal is coking coal for the use in coke ovens to produce the coke needed especially in blast furnaces for iron production [3]. Up to now, only a very small part of the total energy is supplied by renewables like biomass or waste in the iron and steel industry. In 2015, only 0.1% of the total energy sources of the iron and steel industry in OECD (Organisation for Economic Co-operation and Development) countries were supplied by renewables and waste [4]. For comparison, in the non-ferrous metals industry, the share was equally low whereas in the non-metallic minerals industry the share was as high as 7.2% which certainly can be mainly attributed to cement kilns using all kinds of alternative fuels.

Greenhouse gas (GHG) emissions, especially CO₂ emissions, are an important issue for the steel producers because of national and international GHG emissions reduction plans and/or emissions trading systems. The UN's 2030 Sustainable Development Goals, the Paris Agreement, as well as the European Green Deal all aim to improve the sustainability of industrial production and to reduce CO₂ emissions to combat climate change. Within the EU Emission Trading System (ETS), which the iron and steel industry is subject to, emission targets of 73.2% below 2005 levels have been set [5]. Now, Europe aims to reach a 55% CO₂ emission reduction until 2030 and carbon neutrality and a circular economy by 2050 [6]. This goal cannot be achieved without the iron and steel making industry substituting its fossil carbon consumption by renewable alternatives.

Therefore, the research on the possibilities of a substitution of fossil coals in iron and steelmaking is increasing more and more. The two main routes to produce steel are the integrated blast furnace-basic oxygen furnace (BF-BOF) route and the electric steelmaking route employing the electric arc furnace (EAF). In 2018 the BF-BOF route had a share of the total worldwide crude steel production of 70.7% while EAF steelmaking accounted for 28.9%. However, looking at specific regions the share of EAF steelmaking can be a lot higher. In the EU 41.5% and in North America about 67% of the steel was produced in the EAF route in 2018 [7]. The integrated BF-BOF route is characterized by the two-step process to first produce iron as liquid hot metal in the BF, which is then processed into steel in the BOF. The EAF route, on the other hand, is characterized by the use of mainly solid materials, like scrap, direct reduced iron (DRI), hot briquetted iron (HBI), or pig iron, that are melted in the EAF.

Especially regarding the ironmaking process, which is the main consumer of fossil coals in the iron and steel industry, a number of reviews regarding the use of alternative carbon sources like biomass have already been conducted and published [8–16]. Even so, the amounts are much lower in EAF steelmaking, and fossil carbon sources are used, contributing to the direct GHG emissions of the steelmaking process. However, there are up to now no reviews about the use of alternative carbon sources in EAF steelmaking available. Therefore, in this paper, the use of carbon in EAF steelmaking is first described. Following that, recent research on the use of alternative carbon sources is presented to give an overview on the possibilities to substitute fossil carbon by biomass or waste-based materials.

2. Carbon Use in EAF Steelmaking

In the EAF, iron sources like scrap, DRI/HBI, or pig iron are charged together with materials like alloying elements, slag formers, and carbon sources. The charged material is then melted by a mix of electrical and chemical energy. The electrical energy is introduced into the furnace via electrodes by the electric arcs ignited between electrodes and charged material. The chemical energy is usually introduced by oxyfuel burners but also e.g., by hydrocarbons from scrap contaminants (paint, oil, grease, etc.) and charged carbon. Apart from the steel melt, a slag is also produced based on slag formers like lime and dolomite which will incorporate iron oxides and oxidic impurities from scrap or gangue from DRI/HBI.

Carbon sources are of great importance in the electric arc furnace. In modern electric arc furnaces, the share of energy input from fossil fuels like natural gas and coal is over 40% of the total energy input [17]. In addition to their energetic use as a substitute for electrical energy, carbon sources are used in particular as slag foaming agents [18]. Solid carbon sources, like coal, petrol, coke, etc., are used in the EAF in two ways. The charge carbon is charged together with the scrap or other iron sources and additives at the beginning of the heat. This carbon serves to carburize the melt, contributes thereby to the slag foaming and by direct oxidation during meltdown realizes a chemical energy input. The injection carbon on the other is injected into the EAF via lances or injectors together with oxygen to generate CO bubbles within the slag and thereby to foam the slag.

According to a study by the International Iron and Steel Institute (IISI), on average, about 12 kg of coal are used per ton of steel produced in the EAF [19]. With a global steel production in 2018 of around 520 million t via the EAF route, this corresponds to a fossil coal consumption of over 6 million tons. This coal use causes specific CO₂ emissions of about 43 kg/t steel and related to the total EAF steel production in 2018 CO₂ emissions of about 22 million tons.

Looking at values of direct CO₂ emissions of EAF steelmaking compiled in studies for the European Commission the benchmark is given with 59 kg CO₂/t steel [20] while the average is given with 102 kg CO₂/t steel [21]. Based on these values, the use of fossil coal in the EAF, besides the use of natural gas and the graphite electrode consumption, is causing

about 40–70% of the direct CO₂ emissions of the EAF steelmaking process. Figure 1 shows a schematic of carbon input and direct emissions of the EAF.

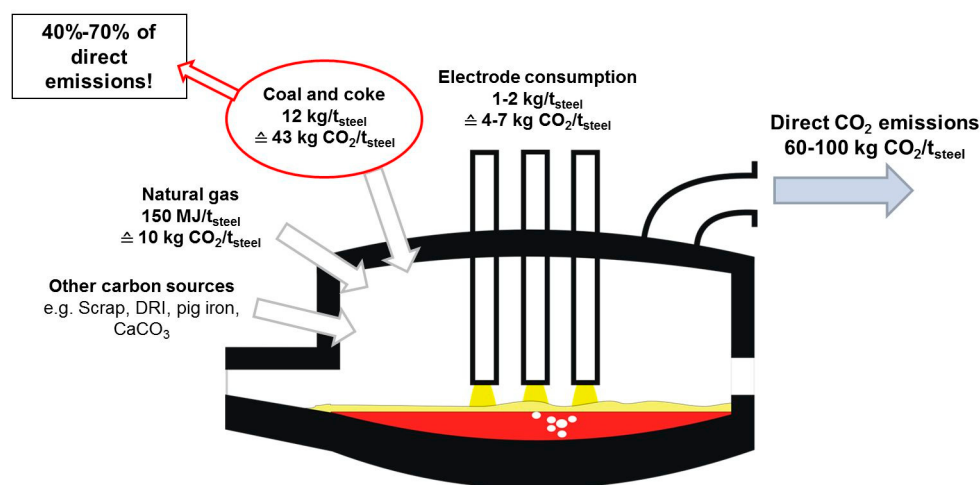


Figure 1. Direct CO₂ emissions of a typical EAF [22].

2.1. Charge Carbon

In comparison to injection carbon, the demands placed on charge carbon are lower. Of particular interest are the calorific value, the reaction behavior as well, as the carburization capacity. If the primary aim of using charge carbon is to carburize the melt, the reactivity must be low enough that the carbon does not burn prematurely but can dissolve in the melt. If the chemical energy input in the melting phase is of primary interest, a high reactivity and therefore a quick energy release may be more important.

Consequently, for charge carbon, the calorific value is of primary importance. With a corresponding cost–benefit ratio, lower carbon contents and higher ash contents are also tolerable as long as no components with the potential to reduce steel quality are introduced into the EAF. Apart from that, the carbon source must have sufficient physical properties to allow for safe handling, storage, and charging into the EAF.

2.2. Injection Carbon

Slag foaming in the EAF is a well-established and widely used method to significantly increase the efficiency of energy transfer in the furnace. The shielding of the electric arcs by the foaming slag reduces the energy losses via the water-cooled furnace walls and roof, thus enabling a significantly improved energy transfer from the arc into the melt. In addition, the slag foaming has a stabilizing effect on the arcs and reduces the noise emissions of the EAF. Approximately 5–10 kg of injection carbon are used in electric arc furnaces [23].

The foaming of the slag by CO/CO₂ gas bubbles occurs in the EAF process via oxidation of carbon dissolved in the molten steel by oxides in the slag (reaction (1)). This foaming process is enhanced and maintained by injecting carbon into the slag. The injected carbon can thereby react directly with the iron oxide according to reaction (2) or reduce the iron oxide indirectly according to reactions (3) and (4) via an intermediate gasification step [24].





Besides carbon, coal usually also contains varying amounts of volatiles, namely hydrogen which is oxidized by FeO to H₂O (reaction (5)). Furthermore, the hydrogen takes part in side reactions (6) (water-gas reaction) and (7) (water-gas shift reaction). Especially the water-gas shift reaction supports the gasification of solid carbon and thereby accelerates the reaction of slag and coal. Hydrogen in the gas phase also increases the carbon gasification rate since H₂ and H₂O react more rapidly with carbon and slag compared to CO and CO₂. Moreover, FeO also reacts faster with hydrogen compared to CO.

Hayes [25] found that the reaction constant of the FeO reduction by H₂ is one order of magnitude greater than that for FeO reduction by CO at 1300 °C and even 5 times greater at 1600 °C. In agreement, Xie and Belton [26] found that reduction rates of ferric iron in slag by H₂/H₂O are a factor of 2–3 times higher than those by CO/CO₂. King [27] demonstrated that the rate of carbon gasification increases linearly with H₂ concentration in carrier gas for hydrogen concentration of up to 4%.

The requirements on the injection carbon for a good foaming effect primarily concern a high reactivity and thus the highest possible carbon content and a low ash content of the coal as well as a defined particle size distribution for pneumatic conveying. With regard to the plant technology used for injection, it should also be noted that no explosion protection is required for the current use of petroleum coke and anthracite coal and is therefore not usually implemented in terms of plant technology. If suitable alternative carbon sources fall into explosion protection classes, substitution may already fail at this point for economic reasons.

3. Alternative Carbon Sources

When using alternative carbon sources, it must generally be considered that the properties of these materials can differ greatly from those of common fossil coals. These differences can have an impact on the steelmaking process in the electric arc furnace. Research has been carried out so far on the use of biomass-based as well as rubber and plastics-based alternatives in EAF steelmaking.

3.1. Biomass Based Alternatives

The research on biomass-based alternative carbon sources includes technical and life cycle assessment studies, fundamental research on slag foaming by biomass and biochars as well as pilot and industrial scale investigations of the use of biomass and charcoal in the EAF.

Mathieson et al. [28] investigated the potential of biomass use in the steel industry of Australia. The focus here was on reducing fossil CO₂ emissions from the metallurgical processes of the blast furnace and electric arc furnace route, with an emphasis on the blast furnace route. In conclusion, the fundamental suitability of biomass carbonisates as a feedstock in iron and steel production is highlighted. They report the CO₂ mitigation potential through biochar utilization for Australian EAF steelmakers with about 6–12%. However, it has to be noted that this is based on the Australian electricity production mix. This results in a high share of 78% of the total emissions of 0.5 t CO₂/t crude steel in EAF steel production, which is caused by electrical energy consumption.

Norgate and Langberg [29] and subsequently Norgate et al. [30] used the life cycle assessment (LCA) methodology to assess the substitution of fossil carbon sources with charcoal as a fuel and reductant in the iron and steelmaking industry of Australia. In addition, they considered economic aspects of the use of carbonisates from biomass and estimated the land requirements for substituting fossil coal with biomass carbonisates. For a complete replacement of fossil carbon sources by biomass carbonisates, they calculate a saving of CO₂ emissions in relation to total (i.e., direct and indirect) emissions of 5.5–11% for the production route in the electric arc furnace. Taking charcoal production by-product

credits into account, they calculated reduction rates of GHG emissions for the EAF route of 7.3–14.7%.

However, these relatively low reduction values for the EAF route stated by Norgate et al. are due to the indirect emissions from electrical energy consumption of the EAF. The calculations are based on the specific emissions of the Australian electricity mix, which are comparatively high at 0.987 kg CO₂/kWh due to a very high proportion of coal-fired power plants. Demus et al. [31] showed that, for the EU-27, with a different electricity mix and specific GHG emissions of 0.43 kg CO₂/kWh, in a very similar scenario, the relative GHG emission reduction potential is almost 29% for the substitution of fossil carbon sources with charcoal in the EAF route.

Sampaio et al. [32] describe an indirect way for the production of green steel in the EAF. They propose the use of cold pig iron (CPI) produced in charcoal operated blast furnaces in combination with scrap. In this scenario the carbon introduced into the EAF by the CPI is considered as carbon-neutral. In [33] Sampaio et al. discuss the use of large amounts of CPI in EAF steelmaking. Based on operational data of a Brazilian steel plant, they compare steel production based on scrap and with an addition of 35% CPI. The use of CPI results in lower residual levels in the produced steel and introduces high amounts of carbon-neutral chemical energy into the system. The high concentrations of carbon (4.3–4.5%), silicon (0.5–1%), and manganese (0.3–0.6%) in the CPI can deliver energy at a rate of about 3.6 kWh/Nm³ oxygen injected into the EAF. The intense CO formation and boiling action during decarburization of the steel melt carburized by the CPI supports the removal of dissolved gases and leads to reduced nitrogen concentrations in the tapped steel of less than 50 ppm compared 90 ppm in scrap-only heats. In [34] Sampaio et al. again discuss the possibility of using in the EAF hot metal from a mini blast furnace utilizing biomass/charcoal as reducing agent. They also present a biomass carbonization process coupled with the mini blast furnace increasing the overall energy efficiency of the coupled processes.

Coming to the more fundamental research on slag foaming by biomass and biochars, Sahajwalla et al. [35] tested differences in wettability of different carbon carriers in contact with EAF slag. The tested carbon carriers were injection coke, petrol coke, natural graphite, synthetic graphite as well as charcoal. The tests included contact angle and volume measurements of a slag sample on a carbon carrier substrate at 1550 °C. Charcoal in comparison exhibited the lowest wettability and therefore minimal foaming of the slag. Natural graphite showed the most favorable behavior with regard to wettability and volume increase of the slag by foaming.

Yunos et al. [36] investigated the combustion behavior of metallurgical coke as well as palm shell/coke and coconut shell/coke blends in thermal gravimetric analysis (TGA) and a drop tube furnace. The combustion efficiency was increased by increasing amounts of palm shells or coconut shells in the blend due to the added volatile matter in the blend. Yunos et al. [37] also conducted a fundamental study on the formation of foamy slag in the EAF with charcoal. For this purpose, they melted down slag samples from the EAF process together with charcoal from palm kernel shells as well as with metallurgical coke as reference material in lab scale and investigated the interaction of the carbon carrier with the slag. They also conducted TGA tests coupled with a mass spectrometer (MS) of both carbon carriers. Differences in reaction behavior were apparent from the gasses formed in the TGA-MS as well as in the melting trials, the different increase in slag volume as well as the number and size of gas bubbles found in the slag. The tests conducted showed that charcoal from palm kernel shells can be an alternative to fossil metallurgical coke for the process of slag foaming.

Fidalgo et al. [38] investigated the thermal behavior of grape seed and pumpkin seed char at high heating rates (1000 K/s) typical for injection into an EAF atmosphere within a wire mesh reactor. As reference materials four different coals used in EAF steel plants were also tested. It was found that the biochars showed different thermal behaviors with the grape seed char exhibiting higher combustion and gasification reactivities. Therefore,

grape seed char was proposed as potential candidate to substitute injection carbon for slag foaming.

Kalde et al. [39,40] tested various biomass-based materials with regard to their reaction behavior at high temperatures within a high-temperature reactor. Based on off-gas analysis data and overall weight loss of the samples they investigated the time dependent gasification in N₂ and combustion behavior in air. They determined specific amounts of produced gas and gas production rates for a number of materials. Considered here were palm kernel shells, wood chip pellets, walnut shells, olive kernels, pyrolyzed wood char, torrefied biomass pellets, as well as pellets made of hydrothermally carbonized green waste and anthracite as a fossil reference coal. Based on the presented investigation technique, it is possible to at least qualitatively compare gasification and combustion behavior of biomasses and biomass carbonisates to choose the best option with regard to gas production rate and the total specific amount of gases produced.

Huang et al. [41] investigated the reaction behavior of different carbon sources with a synthetic EAF slag by sessile drop tests at 1600 °C. The carbon sources investigated included a slow and fast pyrolysis biochar from woody biomass, graphite, metallurgical coke and char from end-of-life tire pyrolysis. Based on observed reaction behavior and measured contact angles between slag and carbon substrate, the authors conclude that the biochars were the least reactive materials in comparison to the other carbon sources. According to their analysis neither ash content nor carbon crystalline structure are significant factors influencing reactivity of the carbon sources with synthetic slag. They conclude that the wettability of the surfaces and therefore the surface roughness of the carbon particles has the biggest influence on the carbon/slag reaction behavior.

Mansuri et al. [42] investigated the high temperature pyrolysis of waste macadamia shells to prepare a carbon source for iron carburization. After pyrolysis the biochar was used to test the carburization of pure electrolytic iron. The biochar created from the macadamia shells via high temperature pyrolysis showed a carbon content of 98 wt.%. In the carburization test, a fast carburization up to 5.2 wt.% of carbon the iron alloy was reached. The carbon dissolution rate was compared with literature data was found to be higher than other carbon sources like metallurgical coke or coal.

Kongkarat [43] tested rubber tree bark in blends with coal and with coal as a reference as a carburizer for liquid steel. In comparison to coal, the carbon content of the steel increased in contact with rubber tree bark and the bark/coal blends. After 30 min of contact, the final carbon content was about 2.8 wt.% for coal and up to 4.9 wt.% for the rubber tree bark.

Bianco et al. [44,45] report about a research project funded by the European Research Fund for Coal & Steel (RFCS). As part of this GreenEAF project the foaming behavior of various biomass carbonisates with EAF slags was analyzed in laboratory scale. For this purpose, mixtures consisting of EAF slag and reference coal or carbonisate samples were placed in a crucible and melted in a furnace. The change in volume of the resulting foaming slag was measured and a qualitatively greater increase in volume of the slags was found when biomass carbonisates were used compared to fossil coal. The fundamental suitability of biomass carbonisates for slag foaming in the EAF could thus be demonstrated.

Tests were also carried out in a pilot-scale EAF, in which fossil charge coal was replaced by biomass carbonisates. Compared to the operation with fossil charge coal, clear differences in the reaction sequences could be determined with the help of gas analysis when biomass carbonisates were used. This was, e.g., reflected in a different timeline of the energy supply in the melting process. The differences were attributed to different reactivities, physical properties, compositions, and carbon contents of the biomass carbonisates in comparison to the fossil charge coal. Notwithstanding this, the general suitability of the biomass carbonisates as batch carbon could be established as no difference in steel quality could be detected [46].

Initial trials on an industrial electric arc furnace with as-is biomass carbonisate from pyrolysis processes were also carried out [45]. The trials showed clear problems in handling

and charging of the fine-grained material into the EAF. However, the industrial trials were able to confirm that no negative influence on product quality is to be expected from the use of biomass carbonisates.

Demus et al. [22,47] subsequently conducted trials on the briquetting of biomass carbonisates and tested the use of the briquettes made from biomass carbonisate in a pilot EAF. The results of the melting tests showed again that the biomass carbonisates have no negative influence on the process. The briquettes made from biomass carbonisate showed similar combustion behavior to conventional charge coal (anthracite coal) and can thus basically be regarded as an alternative feedstock to fossil coal.

Funke et al. [48] investigated the use of a biomass carbonisate from wheat straw fast pyrolysis as charge carbon substitute in the EAF. The carbonisate is a by-product of a biofuel production process and was pretreated by agglomeration prior to the melting trials in a pilot-scale EAF. Molasses in combination with water proved to be a good binder, resulting in sufficiently strong agglomerates. The reaction behavior and release of chemical energy exhibited by the agglomerate was comparable to biochar from slow pyrolysis and also to anthracite coal typically used in EAFs.

Baracchini et al. [49] report on the GreenEAF2 project funded by the RFCS, which is a follow-up to the GreenEAF project. This project was a pilot project to demonstrate the use of biomass and biomass carbonisates in industrial scale. Within the project, various biomass carbonisates but also virgin biomasses available on the market have been sampled and characterized. In subsequent trials the substitution of injection and charge carbon was tested. The industrial injection trials delivered mixed results regarding the achieved slag foaming. Possible reasons identified are the lower biochar density resulting in a reduced penetration of the slag layer by the injected biochar as well as a reduced reactivity of the biochar with iron oxides. The substitution of charge carbon by biochar and biomass, also reported in Cirilli et al. [50] and Echterhof et al. [51], was evaluated positively. The long-term trials of more than 1500 heats resulted in no detrimental effects on steel or slag quality or furnace operation. Moreover, one campaign of about 300 heats using a mix of biomass (palm kernel shells) and fossil coal even resulted in a reduction of the specific energy consumption of the EAF of about 6%.

Meier et al. [52] conducted simulations of the use of biomass in the EAF based on the case of one of the steel plants in the GreenEAF2 project. They used a dynamic EAF process model to simulate complete heats implementing biomass (palm kernel shells) as a charge carbon substitute. The differences between palm kernel shells and anthracite coal like the increased amounts of volatiles have been included in the model. The model was able to deliver results, e.g., regarding the off-gas composition and evolution which were in sufficient agreement with measured off-gas compositions.

Robinson et al. [53] report on lab-scale and industrial carburizing trials with two types of biochar from woody biomass and synthetic graphite and anthracite as reference materials. The woody biomasses tested are commercial wood chips from logging residues and commercial wood pellets from sawdust. All samples were briquetted and added into a molten iron-carbon alloy. The laboratory tests showed that the biochar from sawdust behaved similar to high quality anthracite and showed similar dissolution kinetics. For the industrial trials, a 50 t EAF was used where about 600 kg of carbon sources like anthracite are usually added to the charge material. In trials, one third of the anthracite charge carbon was substituted by the biochar from sawdust. The test heats did not show any deviations from standard operating conditions.

3.2. Rubber and Plastics Based Alternatives

Another possibility for the substitution of coal or anthracite in the EAF is the use of used tires or waste plastics as a carbon source. Used tires contain carbon in the synthetic and/or natural rubber, in textiles and as carbon black. They also contain a significant amount of steel wire, which can be recycled in the EAF. Natural rubber included in the tire can even be considered as carbon-neutral.

The University of New South Wales in cooperation with OneSteel investigated the utilization of waste plastics and rubber and blends of these materials with coke in the EAF. They especially tested the interactions between slag and carbonaceous material with the sessile drop technique. Zaharia et al. [54] investigated metallurgical coke as a reference and two coke/rubber tire blends and their interaction with EAF slag. Based on off-gas data and volume ratios based on visual observations from the sessile drop tests, they concluded that blends of rubber and coke could be used to substitute the coke used in EAF steelmaking. Subsequently, Zaharia et al. [55] again published an investigation of four rubber/coke blends compared to coke with similar results. Zaharia et al. [56] also investigated the combustion behavior of coke, rubber tires and blends of the two in TGA and a drop tube furnace. They found that the combustion performance or burnout increased with increasing rubber content in the blends and therefore correlated directly with the volatile matter content of the blend. In a further publication, Dankwah et al. [57] tested the reduction of FeO-containing slag by blends of end-of-life tires and coke in a horizontal tube furnace in laboratory. They could show that rubber/coke blends exhibited a significantly increased reduction and carburization of the metal than coke alone. It is presumed that the hydrogen introduced by the rubber and the side reactions under participation of this hydrogen led to a faster gasification of the solid carbon and therefore to a faster reduction of the iron oxide in the slag.

Sahajwalla et al. [58] published on the recycling of waste plastics for slag foaming in the EAF. They tested the combustion behavior in a drop tube furnace and subsequently the slag foaming by sessile drop tests. The materials and blends tested were metallurgical coke and a PP (polypropylene)/PE (polyethylene)/coke blend, synthetic graphite and a HDPE (high-density PE)/graphite blend, and petrol coke, HDPE/petrol coke and PP/petrol coke blends. In all cases the volume ratios during slag foaming increased with plastics additions and therefore improved slag foaming. Sahajwalla et al. [59] published a similar investigation on the combustion behavior in a drop tube furnace, this time with HDPE/coke, LLDPE (linear low-density PE)/coke and ABS (acrylonitrile butadiene styrene)/coke blends in comparison to coke. While the combustion efficiency was increased to a similar degree by HDPE and LLDPE, ABS led to an even more significant increase of the combustion efficiency.

Dankwah et al. [60] investigated the kinetics of the reduction of FeO from the EAF slag with HDPE/coke blends in comparison to coke. Due to the high process temperatures in the EAF the polymers decompose into basic hydrocarbons (reaction (8)), especially CH₄ and into carbon and hydrogen respectively (reaction (9)). The hydrogen formed directly reduces iron oxide according to reaction (5) and in the process reacts significantly faster than a reduction with C and CO respectively.



In their investigations, Dankwah et al. could demonstrate that a HDPE/coke blend possesses a significantly increased reaction rate in comparison to pure coke. This is attributed to the hydrogen introduced by the polyethylene and the described reaction mechanisms. Accordingly, it is reasoned that plastics can substitute a part of the coke input into the EAF.

A similar investigation on PP, PET (polyethylene terephthalate), and PU (polyurethane) plastics in a blend with coke was published by Sahajwalla et al. [61]. In all three cases, the blends of polymers with coke exhibited an improved slag foaming behavior compared to pure coke. So, all three blends could be suitable to substitute coke in EAF slag foaming. Sahajwalla et al. [62] again presented results of sessile drop tests with coke, HDPE/coke, rubber/coke, PET/coke and Bakelite/coke blends. While PET/coke and rubber/coke blends showed an increased slag volume, the HDPE/coke blend exhibited a significantly higher slag volume compared to coke. The carbon pickup of the metal was also strongly increased for the HDPE/coke and PET/coke blends in comparison to coke. Sahajwalla

et al. [63] also tested the combustion behavior of HDPE and PP in comparison to metallurgical coke and in blends with coke in a drop tube furnace. Again, they found an increased combustion efficiency with increasing plastics content in the blend. However, the HDPE and PP in the blend was not as effective as rubber investigated previously.

The reduction of FeO-containing EAF slag with PP/coke blends was investigated by Dankwah and Koshy [64]. The extent of reduction was significantly improved by additions of PP to the coke. Also, the carburization of the reduced metal was significantly increased up to 4.95 wt.% in comparison to a reduction by coke alone (0.65 wt.%). Subsequently, Dankwah et al. [65] tested also the reduction of EAF slag by PET/coke blends. Again, they could demonstrate in laboratory that the presence of polymers in the blends increases the extent of reduction and also increases the carburization of the reduced metal up to 5.29 wt.%. Kongkarat et al. [66] investigated the reduction behavior of EAF slag with PET/coke and PU/coke blends. While the PU/coke blends showed a fluctuating slag foaming, the PET/coke blends showed a stable slag foaming. In both cases, the polymer/coke blends showed higher volume ratios than the slag foaming with coke alone. Kongkarat et al. [67] also tested the carburization of pure iron with coke and HDPE/coke blends in a horizontal tube furnace. The addition of HDPE to the coke increased the carbon dissolution into the liquid steel depending on the amount of HDPE in the blend.

Mansuri et al. [68] investigated the high temperature pyrolysis of waste CDs (compact discs), CFRPs (carbon fiber reinforced polymers) and bakelite to prepare a carbon source for iron carburization. After pyrolysis, the three waste polymers show different characteristics with regard to carbon content ranging from 65% to 98%, surface area, and structure.

In addition to laboratory tests, industrial trials were conducted at different EAF steel mills around the world. Gorez et al. [69] described the use of end-of-life tires as a substitute for charge coal or anthracite in two industrial EAF steel works in France. The tires were added as whole tires, shredded tires and even injected as tire powder. The material was added in bulk, in big bags, via injection and via the fifth hole. They could determine a substitution rate of 1.7 kg tire per kg of carbon. They also could determine that the use of up to 8–12 kg/t steel is possible and has no detrimental effect on product quality, emissions or process behavior. However, the addition of tires instead of coal needs more care to ensure that the tires are not only leading to an increase of temperature in the off-gas dedusting system. Ayed et al. [70] subsequently report that the addition of end-of-life tires was also implemented at another French EAF steel works and at a Belgian EAF steel works. They again state that the placement of the tires within the scrap basket is important to optimize the use of the tires. The tires should be put in the middle of the basket to avoid direct contact of the tires with the hot heel on the one hand and to reduce the burn-off through direct contact with the furnace atmosphere on the other hand.

Sahajwalla et al. [71] report on an industrial trial campaign conducted in 2006 at the OneSteel Sydney steel works. In the trials the injection coke for slag foaming was replaced by an HDPE/coke blend. About 22 heats could be evaluated in comparison to standard operation. The use of the HDPE/coke blend led to a better slag foaming according to visual observations, a reduced specific energy consumption (−3%), a reduced power-on time and a similarly reduced tap-to-tap time leading to an increase in productivity. Sahajwalla et al. [72,73] report on trials conducted at OneSteel Sydney and Laverton steel mills. In both EAFs, the injection of a rubber/coke blend is a standard practice. In addition, the injection of an HDPE/coke blend was tested. As a result, the specific energy consumption is in both cases lower than with coke injection, being the lowest with the HDPE/coke blend. Also, the carbon additions could be reduced by about 12% for the rubber/coke blend and by about 15% for the HDPE/coke blend, while FeO content in the slag was reduced slightly.

Joulazadeh [74] reports on similar trials conducted in Iranian EAF steel plants, where coke and coal were replaced by whole end-of-use tires. The trials were conducted in a 6 t foundry EAF as well as in 25 t and 40 t EAF steel plant. Again, there were no negative effects on product quality or pollutant emissions. Based on the trials Joulazadeh reports a decrease

in electrical energy consumption and a decrease in steel production costs, when end-of-use tires are used in the EAF.

Clauzade et al. [75] studied different use cases for end-of-life tires by means of life cycle assessment. They looked at use cases in civil engineering, in energy recovery, e.g., in cement works, as well as material recycling in steel works. Based on tire composition, they also considered the partial biomass origin of the tires. For the use of tires in EAF steelworks the environmental assessment led to intermediate results compared the use of end-of-life tires e.g., in cement works, while there is a significant environmental benefit.

O’Kane et al. [76] describe the polymer injection technology developed and commercialized by OneSteel using a blend of end-of-life rubber tires and coke. The technology is used as standard operating practice in the OneSteel EAF steel works and was also implemented at a number of other EAF steel works around the world. All installations and trials led to a reduction in specific electrical energy consumption as well as a decrease in injected carbon. O’Kane et al. also present a polymer composite briquette consisting of coke fines, waste LDPE flakes and millscale as well as an LDPE-coke briquette. The briquettes have been investigated in laboratory and industrial scale as alternative iron and carbon units to be charged with the basket. While the millscale briquette achieved good reduction in the laboratory tests, the industrial tests of both briquettes showed problems with increased heat generation and overheating of the dedusting system. However, for trials where nutcoke was replaced by LDPE-coke briquettes an overall reduction of electrical energy consumption by 10 kWh/t and a reduction of the power-on time could be observed.

Fontana et al. [77] report on the implementation of OneSteels polymer injection technology at the European EAF steel works of CELSA Group in Cardiff, UK and Mo I Rana, Norway. In both cases, by the injection of a rubber/coke blend instead of coke, the electrical energy consumption could be reduced, the amount of injected carbon could be decreased, the amount of injected oxygen could be decreased, and the productivity could be increased. Emission measurements could prove that there were no increased emissions from the use of rubber/coke blend.

Cirilli et al. [78] studied the utilization of ASR (auto shredder residue) as a carbon substitute in the EAF. The light fraction of ASR, which mainly consists of plastics, rubber, textile and fiber material, was used to produce 150 t of briquettes by pressure extrusion. These briquettes were then used within industrial trials in an EAF to substitute charge coal. To substitute 100 kg coal, 450 kg of ASR briquettes have been charged into the EAF. In a campaign of 29 heats, it was found that the substitution led to a reduction in electrical energy consumption of about 8 kWh/t, while natural gas consumption, oxygen consumption, and tap-to-tap time were not changed. However, it has to be ensured that the ASR briquettes are not charged in the top of the scrap basket, because this leads to an early and rapid combustion of the ASR increasing the temperature in the off-gas duct. An increased substitution of 200 kg of coal with 900 kg of ASR was also tested but led to a temperature overload of the dedusting system, which could not be tolerated. Emission measurements during the trials showed, that there was no negative effect on emission levels including dioxins and furans.

4. Discussion

In view of current developments with regard to GHG-neutral hydrogen-based reduction processes producing H₂-reduced DRI and the subsequent melting in the EAF (e.g., HYBRIT [79], SALCOS [80], H2FUTURE [81]), there will still be the need to introduce carbon into the system either to carburize the steel or to create foaming slag to improve the energy efficiency of the melting process. So, if in the future a substantial part of the steel production shifts to a direct reduction and EAF based route to reach the GHG emission reduction goals set around the world, there will still be a need to use alternative carbon sources to produce a really green and carbon neutral and/or fully circular steel. For the scrap-based EAF route this is true anyway. Here also carbon-neutral slag foaming agents and carburizers will be needed in future.

The literature review could show that there are already solutions available to substitute fossil carbon sources by carbon-neutral biomass-based and circular rubber or plastics-based carbon sources. In some cases, the substitution is already tested in industrial scale or even implemented standard operating practice (SOP) in some EAF steel works. Table 1 summarizes and compares the current state for the different alternative carbon sources discussed. However, there is still room for further research regarding the different use cases of charge and injection carbon, but also regarding the materials used and their treatment or pre-processing.

Table 1. Current state of research and implementation of alternative carbon sources.

Carbon Source	Use	Laboratory	Industrial Tests	SOP ¹	References
<i>Biomass based</i>					
Charcoal from various materials	Injection carbon	3	3	-	[35,37–42,44,45,49]
Virgin biomasses	Injection carbon	3	3	-	[36,39,40,43,49]
Charcoal from various materials	Charge carbon	4	5	-	[44–50,53]
Virgin biomasses	Charge carbon	4	6	-	[49,51,52]
<i>Rubber and plastics based</i>					
Rubber tire/coke blends	Injection carbon	4	9	SOP	[54–57,72,73,76,77]
Polymer/coke blends	Injection carbon	4	7	-	[58–67,71–73]
Pyrolyzed CFRP	Charge carbon	3		-	[68]
Rubber tires	Charge carbon	2	9	SOP	[69,70,74]
Polymer/cokebriquettes	Charge carbon	4	5	-	[76]
ASR briquettes	Charge carbon	2	5	-	[78]

Evaluation of the reported tests and trials according to the technology readiness level [82] by the author; ¹ SOP at least in specific steelworks.

Regarding the use as substitute for injection carbon, currently only blends of polymers and coke have been implemented in industrial scale. While this certainly is a step forward, still a substantial amount of fossil coke is used. Currently, there is no industrially tested solution available for a full substitution of fossil carbon sources for slag foaming. Even so biomass-based materials showed some promising results in laboratory, the industrial tests conducted delivered inconclusive results. Also, there still seems to be further need for research to fully understand all factors influencing the foaming behavior of different alternative carbon sources with EAF slag and the transferability of laboratory results to industrial EAF operation where also aspects like the injection of the material into the slag have to be considered.

Regarding the substitution of charge carbon, the use of biomasses and biomass-based materials seems to be ready for implementation in the EAF. Its implementation mainly seems to be a question of availability and economy at the moment. Also, the use of end-of-use tires has been tested and implemented at industrial EAF steel works for some time now. However, when materials with high volatile matter content like rubber or polymers are used, the charging and also the EAF operation, e.g., post-combustion oxygen use, may need further research and development to ensure, that the energy introduced by the alternative carbon source is used most efficiently within the melting process and does not lead to an overheating and subsequent shutdown of the dedusting system.

Regarding the materials and their treatment or pre-processing, biomasses for example are available from a wide variety of origins and sources all with different compositions and characteristics. Also, there are a number of waste biomasses and wet biomasses, e.g., sewage sludges, that could be made applicable for the EAF by processes like hydrothermal carbonization. In general, the integration of biomass treatment and upgrading processes with the EAF process, e.g., taking advantage of available waste heat potentials, still has further potential for research and development. Moreover, while a lot of waste-plastics, like PP, PET, HDPE, PU, ABS, etc., have already been investigated in the laboratory,

the real material streams, like the ASR mentioned above, can be more complex and thus require additional research regarding possibilities to utilize these circular material streams as an alternative carbon source for EAF steelmaking.

5. Conclusions

This review could show that a lot of research is already available regarding laboratory-scale and also industrial-scale investigation of alternative carbon sources for a substitution of fossil charge and injection carbon in EAF steelmaking. However, it has also been discussed that there is still a lot of potential and need for further research and development in this area.

The EAF steel production route from scrap and/or from hydrogen-reduced DRI will in future play at least an important if not a fundamental role to contribute to the GHG emission reduction in the iron and steel industry. To produce a fully green and carbon-neutral steel, it will be necessary to use alternative carbon sources in the EAF that are either renewable like biomass or at least circular, and maybe in future also produced from renewable sources like plastic or rubber wastes.

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