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# Iron ore wires as consumable electrodes for the hydrogen plasma smelting reduction in future green steel production

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## ABSTRACT

In this work we investigate the feasibility and optimisation pathways for using oxide-filled consumable electrodes as both ore feeding system and reducing/melting arc carrier in the hydrogen plasma smelting reduction process. Similar in nature to Söderberg-type electrodes, but free of C-containing substances, this approach has the potential advantage of eliminating the carbon emissions stemming from conventional electric arc furnace electrodes while drastically simplifying the ore feeding into the process. Using a commercial welding setup with a 1.2 mm thick oxide-cored steel wire, area investigations indicate that approximately 50% of the introduced iron ore could be reduced to metallic iron at 100 A arc current under an Ar-10% H<sub>2</sub> atmosphere independent from deposition time. The reduction efficiency was negatively affected by increasing arc current and it was increased by using the wire as the anode. Based on the performed variation of deposition parameters, microstructural characterisation results, high speed footage and first upscaling trials, the key scientific questions and engineering pathways for technological optimisiation towards future green steel production technology are outlined and discussed.

#### 1. Introduction

The steel industry constitutes one of the biggest sources of global greenhouse gasses, contributing to around 8% of total  $CO_2$  emissions. With the global economic and political tensions around fossil fuels and atmospheric pollution, this has prompted a lot of innovation in this sector with the aim of decarbonisation and a growing green steel industry. Numerous technologies and solutions are currently in development to meet the many challenges posed by this goal at every level, from mining and smelting to recycling and logistics [1]. Hydrogen metallurgy is predicted to play a crucial role in the possible pathways to a sustainable future for steelmaking by acting as a carbon-free reducing agent, with industry developments towards an affordable green hydrogen economy being paramount to achieving this [2,3]. Hydrogen is already used to produce direct reduced iron (HDRI) [4], which is subsequently melted in an electric arc furnace (EAF) [5] or with other electric smelting technologies e.g., submerged arc furnace (SAF), open

bath slag furnace (OSBF). The HDRI approach is currently near state-ofthe-art for crude steel production with capacities higher than 1 Mio. tons a year [6]. Other promising routes are also in development such as the hydrogen plasma smelting reduction process (HPSR), molten oxide electrolysis (MOE) [7] and electrowinning (Siderwin) [8]. The HPSR process is particularly interesting among these technologies due to its simple flow sheet and the possibility of producing steel in a single step. The SuSteel HPSR project, although still in the scale-up process, has already begun its third phase and will show a fully integrated process to reach a TRL of 7 in 2025 [9]. While the basic HPSR principle shows promising results [10,11], many open questions remain concerning the final layout of the technology.

The plasma reduction process is majorly affected by the specifics of ore feeding and the characteristics of the generated plasma arc [12]. The design of the cathodic electrode influences both and is therefore crucial to the function of the reactor. Recent approaches at the lab- and demoplant scale have used a hollow graphite electrode through which fine ore

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and reducing gas is fed into the arc zone. Graphite, although susceptible to electrode erosion through sublimation and oxidation, which contributes as a source of CO<sub>2</sub> emissions, does not suffer from electrode insulation due to oxide splatter [13]. Molten oxide expelled from the melt pool which splatters and deposits onto the electrode gets reduced by the graphite and drops back into the melt. Coating of graphite electrodes to reduce their consumption has also been investigated in the context of EAF, with metal oxides and nitrides reducing high temperature oxidation and improving mechanical wear [14]. During the development of this lance, the possible replacement of the graphite electrode by a thin-walled steel tube containing compacted iron ore was considered. A similar variation in electrode design is already exploited in welding technologies, a clear analogy being the differences between GMAW (gas metal arc welding) and TIG (tungsten inert gas) welding. The cored wire would function as a consumable electrode to act simultaneously as an arc carrier and an ore feeding system, shown schematically in Fig. 1. This setup is similar in nature to a Söderberg type electrode but is free of C-containing substances, and is already utilised in the production of aluminium or silicon [15].

In the context of future EAF/HPSR operations, FeO-based consumable electrodes could eliminate a significant cost factor, since fine ores can be used without agglomeration (pelleting or sintering) and at various grain sizes. The differences in ore feeding and the heat transfer of the plasma source to the melt also mean the power input and feeding are no longer independent. Investigations into feeding rates, power input, and polarity as well as pressure, gas and ore composition are needed to optimise parameters and assess the viability of such an approach with existing HPSR technologies and the possible scale-up routes. Research in welding technologies has shown that high temperatures and electromagnetic forces effectively liquidize and atomize the iron ore at the electrode tip (spray arc) [16]. In the HPSR process, this would result in a high density of ore particles passing through the hottest and most reactive zone of the plasma arc [17]. The spray arcing effect should then increase in-flight and in-bath reduction underneath the plasma column which would correspond to an increased hydrogen utilisation and reduction degree compared to hollow graphite electrodes. In the specific case of Fe reduction, further research is needed to evaluate the potential of this approach in large-scale industrial production and gain a broader perspective for future reactor design. The objective of this work is therefore to demonstrate the general feasibility of utilising ore filled consumable electrodes as both ore feeding system and reducing/melting arc carriers in the HPSR carriers, in order to gain first insights into the remaining scientific questions and engineering challenges which have to be overcome to apply them in future green steel production.

#### 2. Materials and methods

## 2.1. Reduction experiments

Exploring and validating the proposed concept with welding technology, the production of consumable oxide wires could be realised with established production techniques for cored wires. In welding with metal cored wires, the filling is mainly used to add additional alloying elements, for deoxidation and to stabilise the arc [18]. While solid wires are relatively simply drawn from a larger initial wire diameter to the final size, the manufacture of cored wires requires several production stages [19,20]. A thin metal strip is formed via rolls into a U-shaped profile and filled with a powder in a continuous process. In further steps, the filled strip is closed (either with or without an overlap) forming a powder containing tube, which is then reduced to the required final diameter in several drawing steps. The wires used in this study were of 1.6 mm outer diameter, formed without an overlap from a 0.3 mm thick unalloyed steel strip as a casing. They were filled with iron ore (hematite) powders with the maximum possible filling ratio. As can be seen from the cross section in Fig. 2 (c), this resulted in an oxide sectional area of about 0.54 mm<sup>2</sup> and a metal sectional area of about 1.09 mm<sup>2</sup>, yielding a metallic fraction of 67%. This very high ratio could not be decreased here in this proof-of-concept study due to the small wire diameter using conventional welding technology. For potential upscaling, the ore-filled electrodes should be manufactured with both, the largest outer diameter and the thinner wall thickness as possible to maximise the ore input and minimise the amount of steel to be needlessly remolten. Two types of wires were investigated here, hereafter



Fig. 1. Schematic representation of the proposed consumable electrode concept modified for hydrogen plasma smelting reduction of iron ores purposes. A spool of hematite-containing cored wire is fed at a defined deposition rate.



**Fig. 2.** (a) Image of the commercial GMAW machine EWM picomig puls utilised for the deposition and reduction of the hematite-containing wires. (b) Enlarged view of the reaction chamber showing the gas nozzle encapsulating the wire feed system as well as the water-cooled Cu hearth on which the material was deposited. (c) Image of a deposited sample and cross-section view of the original ore-filled wire.

named as wire "A" and "B". Wire "A" was filled with 100% iron ore, whereas wire "B" in addition to the iron ore contains further small amounts (<1 wt%) of Na- and K-based compounds whose lower ionization energy has the potential to stabilise the arc transfer, a fact that positively contributes for an efficient reduction process [21].

Depositions were conducted for both wire types in separate sets of experiments using a commercial GMAW machine "EWM picomig puls", in a setup pictured in Fig. 2. The deposition experiments were conducted using a commercial job program with maximised voltage correction (+5 V) for 5, 10, and 30 s with three current levels (50, 100, 200 A), for which the machine settings resulted in 23.2, 24.6 and 31.0 V of the arc at wire feed velocities of 4.7, 9.7 and 21.6 m/min respectively. For both

types of wires, additional experiments were also conducted in which the polarities were reversed (i.e., the wire then connected to the negative polarity, hereafter referred to as "minus"). For the reversed polarity experiments, the wires were deposited for 10 s at 100 A nominally. Ar with 10 vol% H<sub>2</sub> was fed at a rate of ca. 10 l/min through a conventional GMAW gas nozzle as depicted in Fig. 2b.

The efficacy of upscaling the consumable electrode concept was explored by implementing a protype cathode cartridge into the existing laboratory HPSR facility at Montanuniversität Leoben (MUL). This lab scale device has been used in numerous studies on hydrogen plasma smelting reduction, and complete explainations of its function can be found in in previous works [21,22]. A schematic diagram of the plasma



Fig. 3. Schematic diagram of the constructed cathode ore cartridge, the ore-filled cavity coloured in brown, and the plasma reactor [23]. 1) conductor to cathode, 2) viewport for the camera system, 3) refractory lined lid with copper cooling pipes, 4) steel cathode cartridge, 5) ignition pin, 6) viewport for optical emission spectrometer, 7) steel crucible, 8) bottom electrode, 9) refractory lining.

reactor itself is included in Fig. 3 adapted from [23], with the difference in this work being the exchange of a hollow graphite cathode for a steel ore-cartridge. It was operated at a total gas flow rate of 5 Nl min<sup>-1</sup> at 90–100 A and 40–70 V for an average power input of 5.2 kW over ~13 min. For the first three minutes the plasma was fed only with pure argon, and for the remaining 10 min with an Ar—H<sub>2</sub> mixture (40% H<sub>2</sub>). The cathode cartridge was constructed from steel and acted as both the cathode and the ore feeding system. The outer and inner shells were made from 2 mm thick steel tubing, and the inner tube was tapped with a standard M14 thread to enable easy attachment to HPSR device. The bottom of the cartridge was welded up, and the cavity filled with 56.2 g of iron ore. After generating an argon plasma arc, an Ar—H<sub>2</sub> (40%) mixture melts the cartridge and reduces its contents, depositing the products into the steel crucible anode.

#### 2.2. Characterisation

Cross-sections of the deposited samples were prepared by cutting them with a diamond wire saw (Haas-Drahtsägen, HDS 15) along the largest dimension of the specimen and polishing them down to 1  $\mu$ m diamond paste and OP-S. Overview images were obtained by stitching optical microscopy (OM) images of 2.5× magnification using the IMS software. Longitudinal sections of the wires after the deposition experiments were obtained by embedding them in conductive resin and grinding them down to half of their thickness prior to final polishing. The microstructures of both the deposited samples as well as the corresponding wire tips were analysed by scanning electron microscopy (SEM) coupled with energy dispersive spectroscopy (EDS). The metallic fraction for each sample was derived from the surface area of obtained iron compared to the total surface of the cross-section of the corresponding specimens using the imageJ software package on SEM images. These represent only approximate values allowing relative comparisons between different plasma parameters, as they do not take micrometric iron and slag inclusions as well as porosities into account, and are derived on two-dimensional sections only. The cross-section of the sample deposited using the HPSR facility at MUL was metallographically prepared as described above and visualized via SEM. A point X-ray diffraction (XRD) measurement was performed within the remaining oxide portion to qualitatively identify the formed constituents. For this purpose, a diffractometer D8 Advance A25-X1 was used, equipped with a cobalt  $K_{\alpha}$  X-ray source, operated at 35 kV and 40 mA.

## 3. Results

Different pre-programmed characteristic curves (i.e., internal coupling of arc current, voltage, wire feed, short circuit behaviour etc.) of the GMAW machine used in this study did not show a significant effect on the deposition process. Being obviously optimised for welding, where a stable arc ensuring reliable metal transfer and heat input is paramount, the usage of ore-filled cored wires resulted in the formation of - here unwanted – short plasma columns (despite the maximised voltage) with frequent short circuits and large droplets, as can be seen in the stills from highspeed footage displayed in Fig. 4. All these phenomena minimise the ore/plasma interaction surface and thus potentially lower the reduction efficiency. Best results with regards to process stability (i.e., minimal short circuits and longest arcs) were obtained with the "standard" setting of the welding machine used here, albeit with a significant auto-regulation of the arc parameters (current, voltage and wire feed) by the welding machine during the deposition. From an operator's point of view, no significant differences between the two wire types could be observed, nor for changes in polarity. All samples exhibited a pronounced degassing during solidification once the deposition was



**Fig. 4.** High speed footage of the performed trials using filler wires and standard welding equipment, illustrating the suboptimal material transfer conditions for the desired melting reduction of this off-the-shelf equipment. (a) Standard characteristic curve at 41 V and 78 A average; comparatively large arc burning on the oxide core of the wire, but formation of large droplets at the wire side. (b) Characteristic curve for stainless steel welding at 187 A and 20.4 V; short and more stable arc with small droplets, but numerous short circuits. For the purpose of reduction, a stable but long arc with material transfer in the form of small droplets should offer the maximum reduction potential.

stopped, indicating the release of dissolved oxygen, water vapour and/ or shielding/reducing gas.

The effects of the various parameters on the reduction efficiency are shown in Fig. 5.

As can be seen in Fig. 5 (a), the initial metallic fraction of the wire of approximately 67% was increased to about 87% at a nominal arc power of 100 A, meaning that more than half of the ore that was fed in has been reduced to iron during the plasma deposition (acknowledging the challenges in reliable quantification of the metallic fraction as mentioned in the experimental details and referred to in the discussion section). This remains almost constant with changes of the deposition time, indicating that the reduction potential of the arc is roughly matching the ore feed rate in the currently non-optimised setup. The metallic fraction decreased though with rising arc power (current) at constant deposition time of 10 s (Fig. 5b), from almost 90% at 50 A down to 82% at 150 A. Both trends are similar in nature and extent for both wire types. A similarly strong effect can be observed for the arc polarity (Fig. 5 c), where the metallic fraction decreases about 7% when the wire electrode was used as the cathode ("minus"). This might be attributed to changes in thermal effects and/or the formation of differing plasma specimens and kinetic phenomena of ionised specimens being accelerated within the electric field.

Figure 6 shows the microstructural characterisation of the wire A deposited at 100 A for 10 s, under a positive polarity. Fig. 6 (a) shows the cross-section of the sample, which has a reduction level of about 86% (i. e., 86% of it is already composed of metal). A large porosity is present inside the reduced iron due to the fast solidification rates imposed on the samples after the wire deposition is completed, permitting the entrapment of large amounts of gas. In some specific pores, unreduced oxide domains can decorate their interface with the reduced iron, as exemplified by the arrow in Fig. 6 (a) for the big star-shape pore. Such oxides have the same morphology as that found in the original hematite utilised to fill the wires, suggesting thus that they could not get completely melted during the process. Smaller pores are also found within the unreduced oxides sitting atop the major metallic Fe portion of the sample (see, for example, the regions surrounding the frame "c" in Fig. 6

(a). A closer inspection of the microstructure of the metallic Fe reveals that several droplets of oxides remained pinned inside of it, as shown in Fig. 6 (b). Complementary EDS measurements revealed that they generally consist of a Si oxide core (estimated around 73 at.% O and 27 at.% Si by EDS) plus a Fe-enriched oxide shell (measured as 58.5 at.% O, 39.1 at.% Fe, 2.2 at.% Mn, 0.2 at.% Si). The entrapment of oxides in reduced iron was already reported in preceding works for hydrogen plasma reduction of iron ores [11,27], and their presence can also be explained by the fast solidification rates imposed by the water-cooled Cu hearth on which the samples were deposited, impeding gravimetric separation of the micro domains of liquid oxide atop the denser metallic Fe-enriched liquid before the solidification of the entire specimen. Fig. 6 (c) shows details of the microstructure of the unreduced oxides that solidifies with a dendritic structure and contains droplets of reduced Fe entrapped in it. The inset frame in this figure reveals that the primary interdendritic region (darker grey domains in Fig. 6c) also solidified with a dendritic morphology, thus presenting secondary dendrites and a secondary interdendritic region. The local chemistry of the unreduced oxide was probed via EDS and the elemental distribution maps are shown in Fig. 6 (d). These maps reveal that the primary interdendritic region is mostly enriched in Al and Si. Semi-quantitative analysis reveals an average composition of 65.6  $\pm$  0.6 at.% O; 19.7  $\pm$  0.8 at.% Fe; 9.4  $\pm$ 0.3 at.% Si; 3.0  $\pm$  0.1 at.% Al; 1.2  $\pm$  0.1 at.% Mn; 0.8  $\pm$  0.2 at.% Ca for the primary interdendritic domains, measured in several locations inside the sample. The constituents pertaining to the secondary dendrites and corresponding interdendritic zones are too small to be probed via EDS. Nevertheless, these results suggest that the gangue-related elements contained in the original hematite preferably partition to these zones. The composition of the primary dendrites was measured as 61.3 at.% O; 37.3 at.% Fe; 0.2 at.% Si; 0.5 at.% Al; 0.7 at.% Mn. No substantial contents of other elements, other than Fe, were observed in the reduced iron portions.

Figure 7 (a) shows an overall view of the remaining wire tip A (i.e., non-consumed) after 10 s deposition at 100 A (positive polarity). A large bubble was formed at the tip of this wire, giving its spherical shape. Fig. 7 (b) shows an enlarged view of the transition area between the



**Fig. 5.** (a and b) Approximate metallic fraction based on area evaluations of the deposited specimens (initially 67% within the filled wire) after experiments at arc current levels ranging from 50 to 150 A, and for deposition times of up to 30 s. The experiments were conducted for wires of type A with a positive polarity (plus). (c) Approximate reduction degree of both wire types deposited for 10 s at 100 An under positive and negative polarities based on area evaluations. Wire type A was filled with iron ore only, type B contained small amonts of Na and K-compounds for arc stabilisation.



**Fig. 6.** (a) Cross-section of the sample obtained after the deposition of wire A for 10 s at 100 A, with a positive polarity. The arrows point out the unmelted oxide domains attached to the surface between metal and porosities. (b) Microstructure of the metallic portion of the sample, highlighting the entrapped oxide particles in it. The probed region is highlighted by the red frame "b" depicted in (a). (c) Microstructure of the unreduced oxide domains atop of the reduced iron, acquired for the region denominated as frame "c" in (a). The inset of this figure shows microstructural details of the interdendritic zones. (d) Corresponding elemental distribution maps for the unreduced oxide in "c", showing O (green), Fe (brown), Al (blue) and Si (purple).

original iron oxide filling (top) and the melted zone (bottom), as highlighted by the gold frame "b" in Fig. 7 (a). Fig. 7 (b) shows an intense fragmentation, evidenced by pores and cracks in the ore filling confined within the metallic shell (Fig. 4b). Immediately below it is the molten zone, denser than the original oxide and seen with a light grey contrast in Fig. 7 (b), and whose microstructure is shown in detail in Fig. 7 (c). In the melted zone of the wire, two constituents appear with different grey shades after solidification (the interface between these constituents are given by the dashed line in Fig. 7 c). Round particles are also found in this area (black contrast). The local chemistry of the molten zone was inspected by EDS and the obtained elemental distribution maps are reported in Fig. 7 (d).

These maps show that the darker grey constituent existing near the original iron oxide filling contains a slightly higher oxygen concentration compared to the lighter grey one existing at the top of the melted zone (see white arrow in Fig. 7c and d). These results suggest that the original iron oxide filling (hematite) is therefore not directly reduced into iron in this particular location of the wire and therefore that a transition phase is formed during the reduction process. The interface of this transition zone between unmelted and melted oxide domains is enriched in Al, as revealed in Fig. 7 (d). This figure also shows that Si oxides exist in both unmelted and melted zones in the forms either as precipitates or located in the intergranular zones. The very tip of the same wire, indicated by a red frame "e" in Fig. 7 (a) was also microstructurally and chemically investigated as documented in Fig. 7 (e) and (f), respectively. Droplets of metallic iron is obtained at the tip of the wire and immersed in unreduced iron oxides that solidify with a dendritic structure. Al and Si-enrichment is found in the interdendritic regions of the oxide portion, similar to that observed in the deposited sample reported in Fig. 6.

Results from the upscaling trials at MUL are compiled in Fig. 8. The loaded ore (Fig. 3) was reduced over  $\sim$ 700 s and deposited into a steel crucible anode. This was cross sectioned (Fig. 8 d) and the region

labelled as "1" in Fig. 8 (d) was further probed via X-ray diffraction (XRD) and SEM. The obtained results are documented in Fig. 8 (e). The diffraction data reveals that the oxide domain of the sample is comprised of a complex mixture of magnetite (Fe<sub>3</sub>O<sub>4</sub>), wüstite (Fe<sub>0.92</sub>O), fayalite (Fe<sub>2</sub>SiO<sub>4</sub>) and small metallic Fe domains, the latter appearing as bright round particles in the SEM image shown in the inset of Fig. 8 (e). Quantitative analysis via XRD was not possible in this case due to the strong crystallographic texture displayed by the solidified columnar grains of the sample. However, the obtained results are sufficient to characterize the deposited sample, revealing thus that hematite is fully reduced within the deposition/reduction experiment. Mass spectrometry measurements were performed over the entire course of the experiment, allowing for analysing in real-time the amount of H<sub>2</sub>, Ar, CO and CO<sub>2</sub> during the process. From this, the absolute mass of removed oxygen from the sample could be assessed (Fig. 8 f) as well as the corresponding reduction degree (Fig. 8 g), in terms of the gaseous byproducts (H<sub>2</sub>O, CO and CO<sub>2</sub>). The carbon content in the steel cathode cartridge and ignition pin contributes a small amount of CO and CO<sub>2</sub> as it melts, while the slight uptake in these species after 600 s is due to electrode consumption of the then exposed graphite socket where the cathode was attached. Fig. 8 (g) shows that a maximum of 53% reduction degree was achieved with the plasma-assisted deposition experiments, a reduction level that is still not sufficient to provide complete transformation of the ore into metallic Fe. This observation thus helps confirm that the metallic regions visible in Fig. 8 (d) mainly result from the melting of the steel cartridge (cathode). This results also agrees with the qualitative XRD phase characterisation (Fig. 8 e) which revealed that the reduction achieved was sufficient to fully convert the original hematite (with Fe in an oxidation state of (III) into intermediate Fe(II) oxide constituents.



**Fig. 7.** SEM and EDS analysis of the wire tip originally filled with Oxide A. (a) Overview of the tip after deposition, indicating minimum pororsity of the oxide filling in the as-drawn wire. (b) Magnified SEM images of the transition region from the initial oxide filling to the melted zone, highlighted in (a) by the gold frame. (c) Enlarged view of the regions highlighted by the blue frame "c" shown in (b). The arrow points out a gradient in terms of O. (d) Corresponding EDS maps for the region visualized in (c). The interface between the sub-portions slightly enriched and depleted in O is highlighted by a dashed line. (e) Microstructural details of the very tip of the remaining wire, probed in the region highlighted by the red frame "e" in (a). (f) Corresponding EDS maps of the oxide formed at the extremity of the tip as shown in (e).

## 4. Discussion

Despite the use of off-the-shelf welding technology (Fig. 3) not optimised for the metallurgical objectives targeted here, the obtained results successfully demonstrate the general feasibility of using a consumable ore wire as both ore feed system and electrode in the generation of a melting and reducing plasma arc in the HPSR process, with a constant reduction rate of >50% of the injected iron oxide. The observed drop in reduction efficiency (Figs. 5 a and b) with increasing arc power indicates either an increasing amount of metallic evaporation in the hotter arc, the formation of less efficient reducing plasma species [24] and or increasing re-oxidation with more intense deposition conditions, which are difficult to separate in these proof-of-principle trials with comparatively small sample dimensions. Of further interest is the decrease in reduction efficiency with changing the ore wire polarity from positive to negative. With the wire as the cathode, firstly, the temperature of the molten batch (anode) should be higher [25], promoting thermal decomposition of remaining oxide and thus aiding in the overall reduction; secondly, positively charged hydrogen plasma specimens will be accelerated towards the wire and away from the bath in this scenario, which appears to be the dominant effect. Further improvement of the proposed concept of consumable ore-filled electrodes in the HPSR process will therefore focus on obtaining a better insight into plasma characteristics (mainly by optical emission spectroscopy [23], in order to elucidate the interdependent relationships between arc characteristics (current, voltage, length), formation of plasma species  $(H, H^+, H_2^+)$ [26], and resultant reduction kinetics. Parallel highspeed imaging of the arc with increased resolution should allow to gauge-and optimise-the size, surface area and flight time of the molten oxide particles. Such investigations will be performed best on larger scale equipment such as in the MUL HPSR experiments showcased here (Fig. 8). The larger scale reduces the drastic temperature gradients and comparatively high metallic fraction, and enables both more effective diagnostics of the process as well as more representative microstructure characterisation (Figs. 6 and 7). It should be noted though that the obtained metallic fractions of this study represent only approximate values, as they are stemming from SEM-based area evaluations on two-dimensional sections. While offerering the benefit of gaining insight into microstructural build-up of the desposited sample with regards to the dispersion and size distribution of metal within the remaining oxide and vise-versa, possible uncertainties with respect to the precision of the metallic fraction remain from these two-dimensional investigations. Wet chemical determination of the metallic fraction, however, was found to generally offer similar potential uncertainties due to the specific geometric buildup of these lab-scale specimens such as entrapped oxide particles or unevenly distributed constituents outside the measured volume. Largerscale experiments with slower solidifications should allow for a more readily separation between metal and oxidic slag and thus permit more precise evaluations.

Along those lines, our first trials shown here suggest the following improvements for future upscaling and more in-depth investigations, with the target of understanding the possibilities and limitations of consumable ore electrodes for potential industrial application: the oxide fraction within the wire electrode should be drastically improved over the about 33% of this study, which was imposed by the geometrical limitations (minimum steel sheet thickness) of the welding equipment



Fig. 8. Results from the larger scale reactor at MUL. Pictures (a) and (b) are the empty and filled miniature Söderberg plasma electrode, respectively, with image (c) showing the reducing plasma arc generated in the experiment. The polished cross section of the deposited material is shown in (d), with XRD data of region 1 (whose microstructure is given in the inset of this figure) shown in (e). The graphs displayed in (f) and (g) describe the reduction over the course of the measurement by the amount of oxygen in the reaction products and the reduction degree, respectively.

used here. As already demonstrated in the MUL experiments, thicker wires towards Söderberg-type electrodes would be of interest, with accordingly similar challenges. The most critical factor being securing the oxide core – which is liquefying towards the tip – within the thin steel casing. Here mechanical supports might be implemented, as well as exploiting heat both from radiation of arc and batch as well as electrical

conduction through the power carrying wire section (Fig. 1) for selfbaking, possibly assisted by co-implemented sintering agents such as bentonite [16]. Even reducing agents such as carbon-based substances (ideally stemming from renewable sources such as biochar [5] or slag forming constituents might be added to the ore filling, with proper consideration of their effects on arc stability and efficiency in hydrogen

usage. As well as optimising the electrode, the power source for arc generation will have to be adapted to the specific process characteristics, with the goal of achieving a stable and yet long arc (overcoming the limitations of the welding machinery used here, which limits arc length by strictly coupling wire feed with arc power) to maximise flight time of the oxide particles and thus exposure to the reducing hydrogen plasma. Simultaneously, the arc current should be sufficiently intense to achieve thorough atomisation of the liquefied ore at the tip of the electrode to maximise surface area of the droplets, but without excessive burn-off of the electrode material (achieving a stable arc length). Ideally the ore droplets would be thus entirely reduced to metallic iron before they even enter the melt pool (i.e., over its flight inside the plasma volume). All together these optimisations and adaptions would allow investigation of the limits of achievable reduction using a consumable oxide electrode in order to lay the basis for future HPSR reactor design, i.e. identifying the potential need for additional heat sources and reduction mechanisms (such as conventional non-consumable electrodes), or utilising it in the already proposed pathway of hybridisation of HPSR with solid-state direct (pre) reduction for optimised hydrogen usage efficiency [27,28].

## 5. Summary and conclusions

This work demonstrates the general feasibility of utilising oxidefilled consumable electrodes as both ore feeding system and reducing/ melting arc carrier in the HPSR production of green steel. Similar in nature to Söderberg-type electrodes (but free of C-containing substances), this approach has the potential advantage to eliminate the carbon emissions occurring in conventional electric arc furnaces, as well as simplifying the ore feeding, which is currently based on co-injection through the electrode along with the hydrogen-containing reduction gas. Based on the basic trials using a commercial oxide-cored welding wire system, the following conclusions can be drawn:

- Over 50% of the iron ore, using a 1.2 mm thick steel coated wire, was reduced to metallic iron at 100 A arc current under an Ar-10% H<sub>2</sub> atmosphere independent from deposition time. The reduction efficiency was negatively affected by increasing arc current power and was increased by using the wire as the anode.
- (2) Fundamental scientific questions are the relationships between arc parameters, formed plasma species and the resulting reduction efficiency. Here optical emission spectroscopy and highspeed imaging techniques can most effectively deployed, preferably in a larger scale reactor as developed by MUL.
- (3) For further increasing the reduction efficiency, increasing both flight time (via the arc length) and surface area (liquid ore atomisation similar to a spray arc) of the fed-in ore should be targeted by developing an appropriate power source.
- (4) Efforts towards upscaling will be concerned with adding reducing agents such as biochar, sintering additives, as well as slag formers, while increasing the ore fraction through larger diameter wires or pipes.
- (5) Main goal of future research is to identify the potential and limitations of the achievable reduction degree through consumable ore electrodes in order to gauge their applicability in future HPSR reactor design.

## Author statement

The work described has not been published previously, is not under consideration for publication elsewhere, its publication is approved by all authors, and, if accepted, will not be published elsewhere in the same form, in English or in any other language, including electronically without the written consent of the copyright-holder.

## Contributions

HS and AZ conceptualised the work and performed the deposition experiments. AH prepared the wire electrodes and performed the high speed imaging. IS and LC conducted the microstructural analysis. HS, LC, IS, AZ, and CQ wrote the original manuscript. All authors discussed in high detail the obtained findings and commented on the written manuscript.

## **Declaration of Competing Interest**

None.

## Data availability

Data will be made available on request.

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