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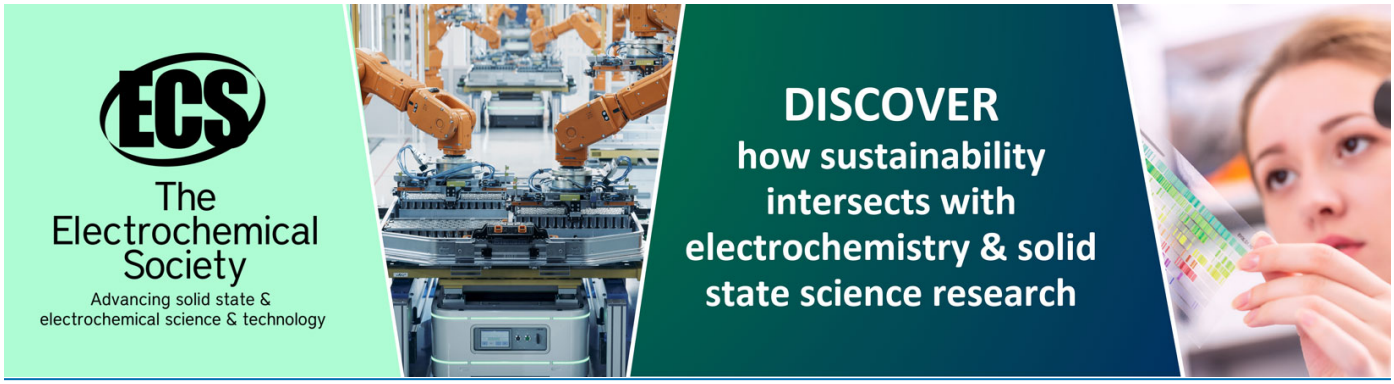

Development of a new laboratory-scale reduction facility for the hydrogen plasma smelting reduction of iron ore based on a multi-electrode arc furnace concept

F Hoffelner[1], M A Zarl[2] and J Schenk[1,2]

[1] Lehrstuhl Eisen- und Stahlmetallurgie, Montanuniversität Leoben, Franz-Josef-Straße 18, Leoben 8700, Austria
[2] K1-MET GmbH, Stahlstraße 14, Linz 4020, Austria

felix.hoffelner@unileoben.ac.at

Abstract. Steel production accounts for a significant share of industrial CO_2 emissions. The HPSR process is a possible alternative to reduce these emissions massively if not completely negate them. In principle, Fe-ore is reduced at high temperatures in the plasma of a DC electric arc. hydrogen reacts with the oxidic melt at the gas-liquid interface. Various concepts for the hydrogen plasma reduction of iron ore have been investigated, but the process technology has not yet surpassed the demonstration scale (TRL5). Experimental setups for charging masses from a few grams to a few hundred kilograms have been realized. Further investigations on the process stability and the reaction kinetics are still necessary. An improved laboratory-scale furnace concept shall provide the basis for the fundamental research. An existing laboratory facility is the starting point for designing and constructing the new plasma furnace. There are several problems with this experimental setup. Mainly, the reactor's dimensions and power supply limitations restrict the arc's length. The first leads to problems with excessive refractory wear, while the latter limits the variation of process parameters. Strong cooling when using Fe crucibles and the unstable nature of the arc complicate the process control. A promising concept to deal with the problem of arc stability is the use of multiple electrodes in a direct current arc furnace. Together with an optimized furnace geometry, new potential for further investigations can open. Using a multi-cathode furnace is also promising to further explore ferroalloy production via hydrogen plasma reduction. An electric arc furnace was designed based on the requirements for the planned plasma reduction facility. The energy requirement was based on assumptions for heat transfer from the arc to the melt, walls, and lid and continuous transfer through the individual furnace parts. Considerations of power supply, hearth dimensions, refractory design, controlled gas atmosphere, and the implementation of auxiliary equipment were central to creating an ideal basis for various experimental setups.

1. Introduction

Steel production accounts for 7% of worldwide greenhouse gas emissions [1]. As the predominant production route, we know today, the blast furnace process results from historical development and optimization. The necessity of using coke as a reducing agent limits the potential for further reductions in CO_2 emissions to a minimal degree. Additionally, the inherent nature of the process prevents the complete mitigation of greenhouse gas emissions. hydrogen plasma smelting reduction (HPSR) is a possible alternative to reduce these emissions massively if not completely negate them [2]. The process

reduces Fe-oxides from a liquid phase of molten iron ore in a single reactor. This is one crucial benefit compared to the blast furnace process, which must be coupled with a basic oxygen furnace to produce crude steel. In principle, Fe-ore is reduced at high temperatures in a transferred direct current (DC) electric arc plasma. The current hypothesis is that hydrogen reacts with the oxidic melt at the gas-liquid interface in the direct vicinity of the electric arc via equations (1)–(6) (species are denoted (), {}, [] according to their presence in the slag, gas, and metallic phases) [3,4]. The according reaction enthalpies are displayed at 1600 °C. This temperature was chosen as a requirement for the presence of a continuous liquid metal bath in the presented furnace concept. From the molecular H_2, introduced as the reducing agent, atomic H and ionized H^+ are generated in the plasma of the electric arc. While all species are assumed to contribute to the overall reaction, H^+ is only present in limited amounts.

Extremely high temperatures prevail in the plasma region. Temperatures of up to 10000 K were derived from Optical Emission Spectroscopy (OES) measurements under process conditions [5]. Calculations with FactSage 8.2 predict that a mixture of atomic H with approximately 2.7% H^+ would be present under equilibrium conditions at these temperatures [3]. Collecting detailed information on the contribution of each species to the overall reaction seems challenging, and the authors have not come across such information to date. Although considering ionization processes in the plasma column, a distinct possibility for (5) and (6) cannot be denied.

$$2(Fe^{3+}) + (O^{2-}) + \{H_2\} \rightarrow 2(Fe^{2+}) + \{H_2O\} \qquad \Delta H_r^{1600°C} = 24.3\ kJ \qquad (1)$$

$$2(Fe^{3+}) + (O^{2-}) + \{H_2\} \rightarrow 2(Fe^{2+}) + \{H_2O\} \qquad \Delta H_r^{1600°C} = 24.3\ kJ \qquad (2)$$

$$2(Fe^{3+}) + (O^{2-}) + 2\{H\} \rightarrow 2(Fe^{2+}) + \{H_2O\} \qquad \Delta H_r^{1600°C} = -429\ kJ \qquad (3)$$

$$(Fe^{2+}) + (O^{2-}) + 2\{H\} \rightarrow [Fe] + \{H_2O\} \qquad \Delta H_r^{1600°C} = -462\ kJ \qquad (4)$$

$$2(Fe^{3+}) + (O^{2-}) + 2\{H^+\} + 2e^- \rightarrow 2(Fe^{2+}) + \{H_2O\} \qquad \Delta H_r^{1600°C} = -2.95\ MJ \qquad (5)$$

$$(Fe^{2+}) + (O^{2-}) + 2\{H^+\} + 2e^- \rightarrow [Fe] + \{H_2O\} \qquad \Delta H_r^{1600°C} = -2.98\ MJ \qquad (6)$$

To implement the HPSR process on a larger scale with sufficient productivity, precise knowledge of the overall reaction and its kinetics is necessary. Although a detailed understanding of the mechanistic processes would be preferable, an accurate representation of the overall reduction progress and its dependencies is sufficient for practical purposes. In addition to the kinetics of the HPSR process, it is also essential to investigate the required conditions to maintain a stable arc in atmospheres with a high H_2 content in the gas feed. An adapted furnace concept needs to be developed to address these challenges. Several concepts for the hydrogen plasma reduction of iron ore have been investigated, such as liquid film reactors with non-transferred arc, in-bath reduction facilities with transferred arc, and in-flight reduction with fine ore injected into the arc zone [6]. However, HPSR technology has not yet surpassed the demonstration scale. Experimental setups with charge masses ranging from a few grams to a few hundred kilograms have been realized.

Investigations on thermodynamically more stable oxides such as Cr_2O_3 or TiO_2 have shown that these also can be reduced with hydrogen plasmas. Kitamura et al. [7] reported that Cr_2O_3 could be reduced to the metallic state via in-flight reduction. TiO_2 and Al_2O_3 were shown to be vaporized as their suboxides and then precipitate as stable oxides. Plaul [8] conducted reduction trials on Fe-ore by adding Cr_2O_3 with an HPSR setup. The results indicated that reducing a Fe-Cr alloy was possible, but higher energy input would be necessary for high Cr yields. The new HPSR laboratory facility could be used to investigate the potential for ferroalloy production via this process.

Several groups studied the HPSR process using different setups over the past 40 years. Nakamura et al. [9] used a single W cathode on iron ore in a water-cooled crucible. 500 g of ore were reduced with arc powers of 15 kW using Ar-H_2-mixtures with 10–50% H_2. Voltages ranged from 40–

60 V with currents of 250–350 A. Kamiya et al. [10] also performed reduction experiments with transferred DC arcs on FeO-containing oxide melts. They used cooled Cu crucibles and MgO refractory crucibles in different experimental setups. Experiments with 25–75 g pure Fe_2O_3 were conducted in the cooled crucible. Experiments in MgO crucibles were performed on pre-reduced fine ore dissolved in synthetic slag above liquid Fe. Behera et al. [11] performed experiments in a larger setup with charge masses of up to 7 kg. A water-cooled Cu crucible was used in conjunction with a non-transferred plasma torch using a W cathode and a Cu anode. For charge masses of 1000 g, a power of 12–15 kW was specified. They assessed a variety of bath depths with crucible diameters of 100 and 200 mm. Their findings suggested that the depth-diameter ratio of the oxide bath limits the possible reduction degree. Too shallow baths do not allow full Fe recovery, according to their results. Souza Filho et al. [12] experimented with a transferred arc in a cooled Cu crucible. Their procedure consisted of interrupted reduction cycles of 1 min each. During this time, they used an arc with a relatively high power of 35 kW on only a few grams of charged Fe_2O_3 material. Between the reduction periods, the gas atmosphere was renewed to an Ar-10% H_2 mixture while the reactor was sealed entirely during the reduction time. Bäck, Plaul, Zarl, et al. [2,13–15] extensively researched the HPSR process with a transferred arc system. The setup consisted of a single graphite cathode with Fe-crucibles with a power input from 7–15 kW. Samples of approximately 100 g ore were used to investigate various aspects of the process.

Crucibles of laboratory-scale arc furnaces for the HPSR process are mostly heavily cooled. While this approach makes sense for very small facilities and certain experiments, it hinders or, to some extent, prevents other experimental setups. To further investigate the HPSR process, a furnace design with a constant pool of molten metal and oxide is of interest. This enables experimental conditions closer to those of the envisioned industrial HPSR process. Typically, laboratory-scale DC EAFs are designed as insulated systems with limited cooling. The only parts to be extensively cooled are design aspects that require increased heat fluxes, such as a bottom anode, measuring equipment or refractory concepts. Table 1 presents data on the power requirements of several laboratory-scale furnaces, which will be compared against the results obtained in this study.

Table 1. Data on operational ranges for power supplies of laboratory-scale EAFs.

Type	Capacity (kg)	Power (kW)	Voltage (V)	Current (A)	Reference
DC	100	350	30–250	2000	HPSR Donawitz
DC	45	20	80	400	Knight et al. [16]
DC	12	50	35–50	1250	Xue et al. [17]
AC with a bottom electrode	3	25	85	300	Mandal et al. [18,19]

A demonstrational reduction plant for the HPSR process with a charge weight of around 100 kg is operated by K1-MET at voestalpine Stahl Donawitz in Leoben. This facility represents a scaled-up version of the existing laboratory facility at the Montanuniversität Leoben. The new furnace shall bridge the gap between existing 100 g and 100 kg facilities for in-bath reduction. An additional motivation for constructing a multi-cathode furnace (MCF) is to expand the possibilities for investigations on the HPSR process compared to the existing facility. Primarily investigations on the kinetics are interesting to better understand the reactions involved. Also, the possibility of introducing the reactants directly into the hotspot of the plasma gives rise to new possibilities regarding process control and stability.

2. Design and calculations

The starting point for designing and constructing the new plasma furnace is the laboratory facility with about 100 g of feed material. This design is intended to melt pre-charged ore and a continuous feed of ore fines during the reduction. H_2 is injected via a hollow electrode, reducing the molten iron ore. An iron crucible or a pin-type electrode inside a refractory crucible is the second electrode to close the

electric circuit. While the existing facility allows refractory crucibles, it was initially designed to use steel crucibles. Experiences with the demonstrational plant in Leoben also influenced the design decisions. The proposed reactor design also consider relevant aspects based on studies of the HPSR process and DC EAFs, which are discussed in the introduction section.

Several problems exist in the experimental setup of the laboratory facility. The dimensions of the system and limitations regarding the supplied voltage restrict the length of the arc. This also leads to problems with premature wear of crucibles made from refractory material. During operation in an iron crucible, strong cooling complicates the process control. The unstable nature of the arc is also problematic, which makes process control even more difficult. The mentioned limitations restrict investigations on larger parameter areas in the existing furnace.

To cover the planned investigations, the scope of the furnace must meet some essential requirements. Multiple electrodes in parallel will be used to investigate their influence on the stability of the arc column. The objective is to create one central hotspot between the single independently powered electrodes, as the single arc columns are expected to attract each other [20]. This opens opportunities for experiments with varying arc power and hotspot size and for using different numbers of electrodes. It also presents the possibility of introducing charge material of various constitutions directly into the hot spot. If a more stable arc can be achieved in the new setup, OES measurement in the hot spot would allow further investigations on the plasma conditions. The new concept will represent the basis for further optimizations of the HPRS process. As the furnace is to be used in a laboratory environment with various parameters, the focus is on achieving the highest possible flexibility rather than efficient operation. Therefore, the furnace design aspects and associated calculations are liberal estimates and not fully optimized results.

The thermal losses for the proposed concept were estimated to determine the necessary power for the furnace. These calculations are based on a rudimentary replacement system, which can be seen in Figure 1. The overall transfer efficiency from an electric arc to an open bath was determined by Ameling et al. [21] to be 36%. Due to the presence of an H_2 atmosphere, it was assumed that a shorter arc was present, leading to a value of 40% being used during calculations. In their research, Alexis et al. [22] studied heat transfer from the arc to the molten bath through different mechanisms. Although using higher power in the MW range for their calculations, they found that shorter arcs with lower currents are more effective for energy transfer to the bath. They determined that radiative transfer, convective transfer, and heat generated by anodic effects contributed 42%, 36%, and 22%, respectively. This work utilized these findings to make estimations and calculate the radiative heat transfer from the arc to the bath. The calculation of heat transfer between the solid surfaces within the furnace and the bath in this study only accounted for radiative exchange and assumed that the bath, wall, lid, and cathodes had isothermal surfaces.

The view factors between the surfaces were calculated with Python. A rudimentary model of the inner furnace geometry was created with the PyVista module, and the view factors were calculated based on the PyViewFactor module [23,24]. The view factors were calculated with three cathodes of 16 mm diameter in place, including their shadowing effect. The calculated view factors exhibit good agreement with those calculated by Hay et al. [25] for an open bath configuration. Conductive heat transfer through the refractory materials and the steel shell was calculated from the inner surfaces. Although we are dealing with a small geometry with comparatively thick refractory layers, the transfer was assumed to be one-dimensional trough planes (8) and cylinder (9) areas, and additional terms for the edges were added in parallel [26]. Conduction through the anode and the cathode were also included in the calculation. The thermal resistances of different furnace layers and contact resistances were added in series and parallel to respective replacement values for the hearth, the wall, and the lid. The heat flux through these three parts was then calculated according to (7) [26]. Heat transfer from the outer steel shell was assumed as radiative and convective transport against surroundings at 30 °C. Wall and hearth thicknesses for the insulation layer were adjusted so that the outer surfaces would not surpass 200 °C.

The outer surface of the lid is additionally considered to be actively cooled to a temperature of 70 °C. This is especially important as it accommodates the cathodes and auxiliary equipment. Furthermore, it

must maintain a secure seal and prevent water condensation. The necessary power input from the arc was calculated on the premise that temperatures of 1850 K for the bath could be assured. Figure 1 shows the cylindrical replacement system for the performed calculations, and the data used for the calculations can be found in Table 2. Following are the equations used for the calculation of the thermal fluxes:

$$q = \frac{\Delta T}{\sum R_{\mathrm{th}}} \tag{7}$$

$$R_{\mathrm{th,plane}} = \frac{\Delta x}{k \cdot A} \tag{8}$$

$$R_{th,cylinder} = \frac{ln\left(\frac{r_{\mathrm{o}}}{r_{\mathrm{i}}}\right)}{2\pi kl} \tag{9}$$

where q is the thermal flux (W), R_{th} the thermal resistance (W·K^{-1}), ΔT the temperature difference (K), Δx the wall thickness (m), k the thermal conductivity (W·m^{-1}·K^{-1}), A the area of the plane (m^2), r_o and r_i the outer and inner radius (m), and l cylinder length (m).

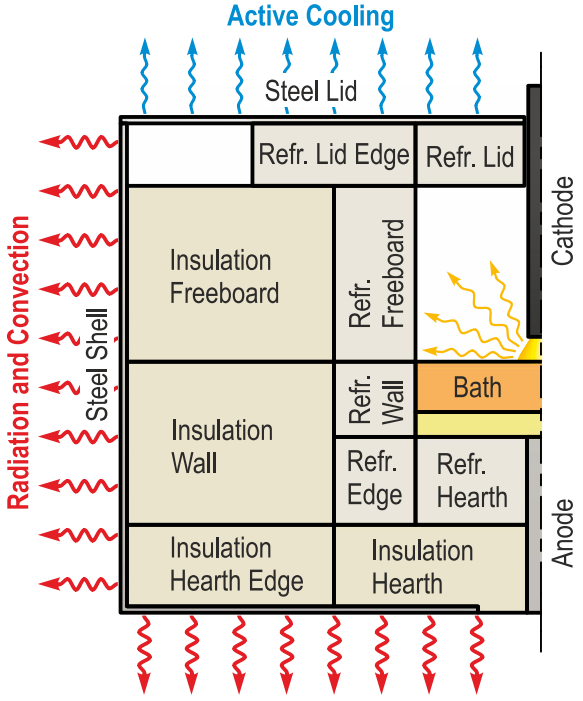

Figure 1. Geometry of the model assumptions for calculating thermal losses.

H_2 and a stabilizing gas will be used during the process. While a process with pure H_2 would be desirable, a certain amount of stabilizing gas may be necessary even at higher voltages. The enthalpies to heat the process gases are included in the thermal balance to calculate the power requirements. The necessary electrical power is calculated for various operating conditions in the steady state. The goal is to guarantee a stable operation throughout all phases of reduction trials. This includes furnace heat-up, batch-wise operation, operation with a continuous charge feed, and tapping the liquid metal.

Additional to the thermal balance, various design aspects for the MCF were considered. These include placement and control of the electrodes, placement for gas injection, and design of the furnace vessel and lid to house all auxiliary equipment.

Table 2. Parameters used for the calculations of thermal losses.

Parameter	Value	Unit	Ref.
Thermal conductivity refractory crucible and lid	5	$(W \cdot m^{-1} \cdot K^{-1})$	[27]
Thermal conductivity insulating bricks	0.6	$(W \cdot m^{-1} \cdot K^{-1})$	[27]
Thermal conductivity graphite	160	$(W \cdot m^{-1} \cdot K^{-1})$	[27]
Thermal contact resistance steel-refractory	0.0036	$(m^2 \cdot K \cdot W^{-1})$	[28]
Convective transfer coefficient hearth	2.4	$(W \cdot m^{-2} \cdot K^{-1})$	[29]
Convective transfer coefficient wall	5	$(W \cdot m^{-2} \cdot K^{-1})$	[29]
Convective transfer coefficient lid	6	$(W \cdot m^{-2} \cdot K^{-1})$	[29]
Emissivity refractory	0.7		[30]
Emissivity oxide melt	0.8		[31]
Emissivity coated furnace shell	0.91		[32]
Specific heat refractory magnesia aluminate T_{avg}= 1300 °C	1400	$(J \cdot kg^{-1} \cdot K^{-1})$	[3]
Specific heat isolation calcium silicate T_{avg}= 640 °C	1050	$(J \cdot kg^{-1} \cdot K^{-1})$	[3]
Specific heat steel shell T_{avg}= 150 °C	500	$(J \cdot kg^{-1} \cdot K^{-1})$	[3]

3. Resulting design aspects and discussion

To perform extensive experiments on the HPSR process on a laboratory scale, the most important goal is to design the laboratory facility as close to the envisioned industrial process as possible. This includes the constant presence of a molten Fe-bath during the in-bath reduction process. Each electrode in the reactor requires a minimum distance to the side walls. Multiple electrodes must also be separated far enough to obtain a usable area at the hotspot. The resulting large bath area leads to a relatively shallow pool. Limiting is mainly the charge weight to restrict the size, complexity, and expenses of the laboratory trials.

Nevertheless, it is necessary to maintain a minimum bath depth to ensure a constant pool across the entire furnace area and prevent the arc from directly burning onto the anode and its surrounding refractories. The consequence of these considerations leads to the necessity of a constant Fe bath of 1 kg below the oxidic melt. The MCF is intended to be used with 2–7 kg Fe-ore charge masses.

The goal to maintain a constant molten Fe bath beneath the molten oxide layer also opens the possibility of tapping the contents of the reactor. Therefore, a tilting vessel with a spout is planned. This opening also functions as the off-gas channel with the benefit of preheating it for easier handling during tapping. The anode at the bottom of the vessel will consist of several sheet metal fins made from pure iron strips surrounded by a ramming mass. The refractory concept consists of an insulating layer of lightweight refractory bricks and a cast wear lining in contact with the melt. As the furnace is designed for laboratory-scale experimentation, adaptations can easily be made if required. Gas will be supplied by an array of mass flow controllers (MFC). Three pairs of MFCs will distribute H_2 and a stabilizing gas to the single electrodes. Alternatively, they will be used together if the gas is supplied directly to the hot spot.

Assuming a heat-up time for the charge of 30 min and a reduction time of 120 min, the power requirements for the various operational states were calculated. Calculations for the thermal balance result in a required furnace power of 11 kW to maintain a molten bath in the steady state, which constitutes mainly losses. During reduction operation, the primary energy sinks are the reduction of Fe^{3+} to Fe^{2+} and the required enthalpy to heat the process gases and the charged material. For the different operational states, the required power results in 21, 15, and 18 kW for batch-wise heating, batch-wise reduction, and reduction under continuous feed. These calculations do not consider the necessary energy to heat the furnace to steady-state temperatures. As this would take at least 260 min at a furnace power of 21 kW, it seems necessary to pre-heat the refractory material to a certain degree. The most sensible

solution is to use a resistive heater, as the power supply for the arc is already present. The results for the furnace power are in good agreement with the literature data on laboratory-scale EAFs, and a power source with a maximum power of 50 kW seems reasonable for the task at hand. Voltages up to 150 V and current sums up to 350 A are planned for the upper limits.

Three galvanically isolated DC power sources are required because three independent arcs are operated simultaneously on the bath as the anode. Compared to other plasma gases, the voltage drop over an arc length is high in an H_2 atmosphere. This restricts the lower limit voltage, as too low values can lead to arc detachment due to the fluctuating nature of a transferred plasma. At the same time, the power supply must provide a wide range of voltages, currents, and power levels. As one of the main objectives of furnace design is to study kinetic effects and arc stability, the broadest possible variation of these parameters is required.

Keeping a gas-tight seal between all furnace parts is vital for safety reasons. While sealing the off-gas duct and the lid to the furnace vessel is relatively simple. The main design challenge is the lid assembly with the retractable cathodes and additional openings for gas supply, sampling, and observation. The relatively tight space must fit all the equipment while still thermally insulating the furnace. Additional ports in the lid and vessel are planned for sampling and as a provision for future measuring device implementation. Figure 2 shows the preliminary schematics for the furnace design. The three electrodes are positioned circularly around an additional lance for gas supply and injection of fines into the center.

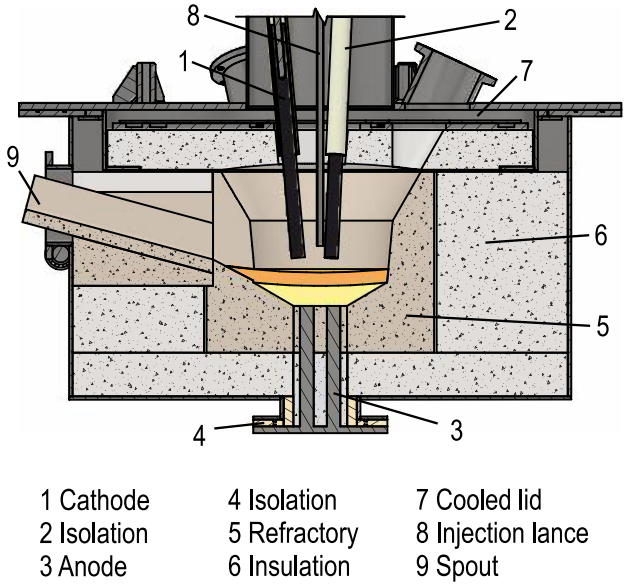

Figure 2. Preliminary schematics for the MCF vessel and refractory design.

4. Conclusion

An MCF DC EAF presents a promising concept for further investigations on the HPSR process. The key requirement is a design as close as possible to the envisioned industrial process. As a result, the following aspects are held to be crucial for the intended task:

- Multiple separately supplied electrodes
- Refractory lined and insulated furnace vessel to create near-process conditions and allow tapping of the contents
- Cooled lid construction to house different auxiliary devices and easy adaption

The engineering work for the laboratory-scale facility is expected to be completed by the third quarter of 2023, with construction targeted for completion by the third quarter of 2024. The data generated from

this facility will be crucial in advancing the HPSR process toward technological readiness, particularly in understanding its kinetics.

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