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Thermal plasmas for industrial high-temperature processes:

Modelling studies of aluminium smelting and steam cracking

Master's thesis in Innovative Sustainable Energy Engineering

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Master's Thesis 2022

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Department of Space, Earth and Environment <u>Division of Energy Technology</u> Chalmers University of Technology Gothenburg, Sweden 2022 Thermal plasmas for industrial high-temperature processes Modelling studies of aluminium smelting and steam cracking ABHISHEK DAHIYA

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Cover: A view of an aluminium smelting furnace similar to the one used by STENA METAL AB, © GHI Smart Furnaces

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Abstract

Industrial emissions constitute approximately 30% of total greenhouse gas emissions in 2019. Several industrial sectors have taken steps to replace their energy sources with renewables such are solar and wind power. However, many industrial processes require high temperatures which have traditionally been achieved by combustion of fossil fuels. It has proved challenging to develop alternative processes to reduce carbon dioxide emissions from these processes. One such process is secondary aluminium smelting, and as much as one-third of all aluminium produced globally comes from scrap products. An important part of the recycling process is melting and alloying with current state of the art furnaces being equipped with oxy-fuel burners. Partial or complete electrification of this process could cut emissions and reduce dependence on fossil supply and prices. Though, the high temperature and melt rates, fundamental to the process, are not achievable through Direct Electric Heating.

A potential alternative for high temperature processes is to switch fossil fuel burners with electrically generated thermal plasma using plasma torches. This study presents a comparison of oxy-propane and CO₂-based plasma burners in the aluminium smelting process. Real process data as delivered by an industrial partner is used to establish a reference case. With process parameters kept constant, the radiative heat load from a plasma torch is modelled in a first step. Process conditions, energy costs and emissions from using a plasma torch in the process is evaluated and compared to the reference case with oxy-fired technology. It was found that the energy costs are 39.1% higher and an increase in melting time by 14%. Although, along with reduced dependence on gas, process modification leads to 94.7% cut in carbon emissions from primary energy. Additionally, this master thesis also includes a brief study of the expected effects from exchanging the burners for plasma torches in a steam cracker.

Keywords: Thermal plasma technology; Aluminium Recycling; Radiative Heat transfer

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List of Abbreviations

P2X	Power to X
CCUS	Carbon Capture, Utilisation and Storage
URTF	Universal Rotary Tilting Furnaces
NOx	Nitrogen Oxides
SOx	Sulphur Oxides
Cp	Specific heat at constant pressure
RTE	Radiation heat Transfer Equation
SNBM	Statistical Narrow Band Model
DTM	Discrete Transfer Method

1. Introduction

The ferocity of recent wildfires, rate of melting glaciers and diminution in rainfall in arid regions confirm that the world is changing. The last seven years have been the warmest seven years in the recorded history, highlighting the drastic trend of the global warming [1]. Fortunately, changing policies and nation states' outlook towards climate issues reflects in the fact that 77 countries have committed to net-zero carbon emissions by 2050 [2]. This rapid transition requires contribution from all sectors, including industries. Industries accounted for about 38% of global final energy use in 2020 and coal is still the dominating source [3]. Due to the growth of global population and economy, demand for goods and materials is certain to rise. Moreover, demand for chemicals, steel and cement is about three times higher in Sustainable Development Scenario than in baseline projection by 2070 [4].

Industrial sector's emissions constitute a significant share, that is, 40% of the total energy related carbon emissions in 2019, after assigning heat and electricity emissions to final sectors [5]. Heavy emitting industries must therefore rapidly reduce their emissions to meet nationally set targets. However, the industrial sector is one of the hardest sectors to decarbonise. This owes to lack of commercially available technologies and long lifespan of industrial facilities. Several industries have taken steps to replace their energy sources with renewables such are solar and wind power. However, many industrial processes require high temperature which have traditionally been achieved by combustion of fossil fuels, and it has proved challenging to develop alternate process to reduce carbon dioxide emissions from these processes. These alternate technologies include Power-to-X, carbon capture and storage and direct electric heating.



Figure 1.1 – Example figure of concept for a green hydrogen network

The concept of Power-to-X (P2X) is to transform surplus renewable energy into liquid or gaseous fuels via electrolysis and other methods. Decreasing cost of renewable electricity from wind and solar technology have drawn research interest towards P2X. Electrolyser systems as presented in Figure 1.1 create green hydrogen as a fuel for transport and industry sectors, are one of the major candidates in this area. Green hydrogen could further be used for synthesis of carbon neutral fuels like methane or methanol by chemically hydrogenating carbon dioxide. While the technology has been proven to be technically feasible, its economic viability remains a challenge. The current cost of green hydrogen is in the range of 2.5 - 6 USD per kilogram [6], but needs to reduce to one-third to be economically competitive in current carbon tax scenario [7]. Storage and transport of hydrogen pose another challenge.

Carbon capture, utilization and storage (CCUS), i.e., to capture the carbon dioxide after combustion, is another alternative to reduce emissions from the industrial sector. Figure 1.2 describes various aspects of CCUS. Carbon dioxide is either captured from flue gas streams or directly from air and compressed to be transported. Compressed CO₂ is then either stored permanently underground or utilized as feedstock to create fuels [8] and polymer products [9]. Currently there are thirty commercial CCUS facilities in operation globally having combined storage capacity of forty million tons of CO₂. However, just 3% of this capacity is in use for heavy industry [10]. This owes to the heavy energy penalty from the capture process which is far too huge for industries functioning in competitive markets [11]. If not used on site, captured carbon dioxide must be compressed and transported. This poses further infrastructure challenges.



Figure 1.2 – CCUS network

Another option is to exchange the fuel combustion for renewable electricity. Direct Electric Heating or ohmic resistive heating, which utilizes Joule Heating effect by passing current through a heating element, is being explored for some industrial processes. However, its challenging to reach the high temperature required for some processes. Moreover, high radiative heat transfer rates which are normal for combustion are not achievable through Direct Electric Heating. This makes remodelling of existing furnaces into electrical one particularly challenging and requires huge investment for replacing existing infrastructure. All these shortcomings of the three discussed proven technologies indicate the need for a technology suitable for high temperature industrial processes.

A potential alternative for high temperature processes is to switch fossil fuel burners with electrically generated thermal plasma using plasma torches. A plasma torch, or plasmatron, is an apparatus which conventionally uses an electric arc to heat a small volume of gas to extreme temperature, which subsequently heat the remaining working gas to state of thermal plasma [12]. This master thesis work aims to examine a thermal plasma torch, with the focus on the study of radiative heat flux and radiative intensity. It further explores if changing fossil fuel burners for a thermal plasma is a suitable alternative to lower emissions of carbon dioxide in an aluminium smelting furnace. This is analysed through calculations of the heat transfer as well as the overall heat balance. Changes in process parameters and emissions are discussed for a plasma operated aluminium smelting furnace. Additionally, the retrofit of a steam cracker is studied in this thesis, exchanging the current burners for plasma torches. However, this case should be considered as a calculation example based on estimated dimensions and temperatures, evaluating the expected changes in radiative heat transfer.

The remainder of the work is structured as follows. Chapter 2 provides a background for both the discussed industrial process and summarizes current state of plasmatron technology. In chapter 3, a framework and models used for the evaluation are described. Chapter 4 and 5 further presents the result and derived conclusion.

2. Background

This chapter presents a background on the topics of interest. The first part describes the working, types and recent advancements in thermal plasma torch technology. The second part discusses the scope of aluminium recycling and method used for smelting process.

2.1 Plasma Technology

Plasma is a state of matter like solid, liquid and gas, consisting of a mixture of ions, electrons and uncharged particles. It is the most abundant sate of matter in universe and constituting all the stars, nebulas, etc [13]. The degree of ionization is the fraction of atoms that are ionized, which depends on the temperature in the case of thermal plasma. Plasma state differs from gas in terms of having extremely high electrical conductivity and susceptibility to electromagnetic field due to presence of charged particles – electrons and ions. Artificial plasma is conventionally created by passing gas through an electrical arc. The process of dielectric breakdown produces enormous heat which in turn ionizes the gas stream by stripping away electrons from particles. The device used is called a plasma torch or plasmatron.

2.1.1 Plasma Torch



Figure 2.1 – DC non-transfer arc plasma torch [14]

The figure above represents a typical plasma torch. It consists of two electrodes (1,2) with produces an electric arc (4), whereas the working gas enters through a porous media (3) which electrically insulates both electrodes. This arc heats a small volume of gas (G) to extreme temperature which in turn heats up remaining working gas (G₂). Three major design categories for the torches are – DC plasma troches (transferred arc and non-transferred arc) and inductively coupled plasma torches. In a non-transfer

arc device, both the electrodes are a part of the torch and plasma projects out through gas pressure. Whereas, in transfer arc torch, the workpiece act as the second electrode. Electrodes are typically made of either a refractory material like tungsten or graphite, etc., which are expended gradually by sublimation or water-cooled metal with high thermal conductivity like copper. Transferred arc plasma devices are fundamentally more efficient than their counterpart as heat loss to the water-cooled torch body is minimized. This is because plasma is created outside the torch body and only one electrode needs to be cooled. Figure 2.2 and 2.3 presents schematics of a DC transfer arc plasma torch and an inductive plasmatron respectively [12].



Figure 2.2 - DC transfer arc plasma torch

Figure 2.3 – Inductive plasma torch

Inductively coupled plasma torches are characterised by absence of electrodes. These use electromagnetic induction to create high temperature plasma discharge. Plasma gas is insulated from the coil which eliminates the possibility of contamination by metallic vapor, and cooling losses are lower than in DC arc torches. However, DC plasma produces higher local energy density, stable operation and better control. These advantages make DC non-transfer arc plasma torch better suited to industrial processes discussed in this study.

2.1.2 Plasma Gases

Currently, most common gases used in plasma torch are argon, helium, nitrogen, hydrogen and air [15]. The choice of gas is based on compatibility with the process, reactivity, gas enthalpy and cost. The flow rate of plasma gas must be correctly balanced with the electrical power to get a stable arc. Diatomic gases such as nitrogen and hydrogen undergo dissociation reaction before being ionized, thus have higher energy content then inert, monatomic, gases such as argon and helium. For example, commonly, argon is used when an inert gas environment is needed in metallurgical

applications hydrogen is used as reducing agent and provides higher energy density and heat transfer rates [15]. For applications to substitute fossil burners with plasma burners, air is the cheapest alternative. However, high temperatures in plasma core would mean considerable NOx emissions. Carbon dioxide has good potential for use in plasma burners, being an asymmetric molecule that participates in radiation process by absorption and emission [16]. This would fundamentally mean higher radiation heat rates as compared to other transparent working gas and better suited to match combustion heat rates. Thus, carbon dioxide is the preferred working gas for the current study.

2.1.3 Industrial Application and Advantages

Thermal plasmas provide numerous advantages for industrial processes including high temperature, high energy density and relatively simple design. Plasma torches have been used for years in low power application such as welding, plasma cutting and surface treatment. Figure 2.4 shows common application mapped on Volt-Ampere diagram with power rating [15]. Based on the technical maturity and quantity of installations, surface modification and coating seem to be the most important application of thermal plasma [17]. High concentration of radical species in plasma makes it attractive for chemical synthesis and metallurgical application. DC-arc type thermal plasmas are also used for synthesis of nanoparticles like carbon nano tubes [18]. Rao et al. states that retrofitting plasma torches for fuel oil burners lead to lower operating costs, capital costs and greenhouse gas emissions [19]. Other advantages of plasma torch over other energy sources includes lower installation size, rapid start-up and shut-down capability.



Figure 2.4 – Plasma torch applications with power rating [15]

Moreover, plasma technology has been increasingly used for treatment of hazardous waste. High temperature in plasma is capable of decompose highly noble organic compounds such as halomethanes and transforms inorganic fraction into inert slag which could be safely disposed. Demonstrations confirm that the steam plasma torches can be used to destroy R-12 refrigerants with a removal efficiency of 99.9999% [19]. Other advantages of using thermal plasma for waste treatment is fast heating rates which decompose waste into stable vitrified slag and low off-gas flow rates [18]. In any waste processing installation, off-gas cleaning is a major economic factor, and the costs increase with increasing gas flows. Studies and technology providers claim electrode life of over 1000 operational hours [19].

Electrodes in plasma torches disintegrate with operation due to thermal decomposition and sublimation. These need to be changed regularly and the associated cost and downtime is an important factor for feasibility of the technology. This has been an area of advancement with better materials and cooling techniques.

The focus of this master thesis project will be to evaluate the possibility of using thermal plasma torches to replace fossil fuel burners in a petrochemical steam cracker and an aluminium smelting furnace. In terms of these applications, use of plasma torch reduces emissions and dependency on fossil fuels. Moreover, simpler design of plasma torches presents a way of transforming current furnaces into electrical furnace without having to invest substantially in new components. Lower gas flow rates in comparison to fossil combustion also reduces need for off-gas treatment.

2.2 Aluminium Recycling

Aluminium is produced by Hall–Héroult process from aluminium oxide coming from the ore, bauxite. The process of electrolysis produces carbon dioxide emissions and consumes a large amount of electricity. Furthermore, the process generates bauxite residue, commonly known as red mud, as a by-product which poses substantial environmental hazard. On the other hand, recycling of aluminium consumes only 5% of energy needed to produce primary aluminium [20]. Moreover, aluminium can be recycled any number of times without any degradation in quality. All this offers sufficient commercial and environmental reasons to recycle aluminium and this has been the case for decades. Figure 2.5 shows the fraction of globally produced aluminium coming from secondary source, i.e., recycling.



Figure 2.5 - Total aluminium production and percent from primary and secondary sources [21]

The process of aluminium recycling starts form collection and sorting of aluminium scrap followed by shredding to right size for further processing. Aluminium scrap contains oil, moisture and various organic compounds such as paints and lacquers which reduces the metal recovery. Therefore, these components are removed sometimes in pre-treatment process such as drying and de-coating. Then the shredded pieces are melted, alloyed and casted to produced desired product. Scarp is melted under a cover of sodium chloride and potassium chloride, called flux, which removes undesirable oxides and shields the metal from oxidation. Commonly used furnaces for the process are Reverberatory and rotary furnaces which use fossils as energy source. Oxygen-enriched burners are employed in modern plants for high melting rates, which is desirable to reduce material losses due to oxidation [21]. Off-gases containing organic compounds are produced during the processing of greasy and oily scrap, as well as scrap coated with organic lacquers or paints. These compounds from off-gases must be eliminated. Moreover, the by-product from smelting process called salt cake has to be treated before being disposed.

2.2.1 The process at STENA's facility

Stena Aluminium, a subsidiary of STENA Metal Group, is Nordic's leading producer of customized aluminium alloys from recycled raw material. Figure 2.6 provides a plan of Stena Aluminium's smelting process located in Älmhult, Sweden. Melting takes place in a universal rotary tilting furnaces (URTF) which is fed with propane and pure oxygen for combustion. As discussed earlier, oxy-fuel combustion provides higher melt rates

which increases aluminium throughput. Furthermore, use of pure oxygen instead of air reduces the amount of off-gases and NO_X emissions.

A flue gas cleaning system is placed after the melting process in the URTF to remove particulate, chlorine, and SO_X, as well as an afterburner to reduce incomplete combustion products. Firstly, the flue gas stream is diluted with additional air to cool it down to 350°C and small residual aluminium particles are caught with a cyclone. This is done to protect the equipment downstream. Then follows an activated carbon silo and a bicarbonate silo for the removal of sulphur, chlorine and organic compounds. Afterwards, the off-gas stream is passed through a bag house filter which separates ash and remaining dust particles. Further downstream is an afterburner unit which heats up the gas to 850°C to eliminate any organic compound that may remain from combustion in URTF. Heat from this step is recovered and transferred to Älmhult's district heating system. After gas cleaning and heat recovery, the exhaust gas exits the smelting plant through a 75-meter-high stack, meeting all required environmental emission standards.



Figure 2.6 – STENA Aluminium recycling plant: Process diagram

Aluminium is highly reactive at temperatures about 600°C. In contact with oxygen, it gets oxidized to form Al₂O₃ which corresponds to a process loss. Aluminium oxide has a lower density as compared to molten aluminium and it thus floats on the surface of molten pool. It also entraps some free aluminium, and this mixture is commonly called dross. The fraction on metallic aluminium in dross can vary between 15 to 80% [21]. The amount of dross generated in the smelting process depends upon category of scrap and estimates range from 1% to 5% of the feed [22]. To minimize dross formation, uses a cover of flux which constitutes of 70 wt.% NaCl and 30 wt.% KCl. Another point to note is that the charging gate of URTF at Stena Aluminium is not airtight sealed which leads to leakage air into the furnace. This leakage air takes part is process chemistry and produces NOx emissions as well.

Replacing the fossil burners in this process with plasma burners could potentially not only cut carbon emissions from the process but may also improve process efficiency by reducing costs and metal loss. Plasma torches provides high energy density and faster melting rates. A suitable working gas in the plasma burners could provide the blanket cover and eliminate the need of chemical fluxes or inert gases. Reddy et. al. states that use of plasma torch reduces dross producing to less than 0.5% [23].

2.3 Steam Cracking

Steam crackers convert the basic petrochemical feedstock such as naphtha, butane, propane and ethane into ethylene, propylene, and other by-products, which may then, e.g., be used for producing plastics. The steam cracking process involves heating of the feedstock with steam at a temperature of about 850°C, conventionally using natural gas combustion in a cracking furnace.

Radiant tubes are placed in the center of burners to provide uniform and effective heating rates. The residence time for the feed in the furnace is in the order of a second. The emissions of greenhouse gases from the process are vast, where the use of natural gas in the furnace is the major source of carbon dioxide emissions. However, carbon emissions of the entire process could be reduced by up to 90% using green electricity instead of combusting natural gas [24]. This has pushed a lot of research interest in developing next generation electrical steam crackers. However, there have been challenges to meet the required heat transfer rates with traditional electrical heating. This causes a longer resident time and lower product throughput. Moreover, these furnaces are huge in terms of both size and investment and have a lifespan of around 40 years. This calls for a modular solution which could be used with existing infrastructure and introducing plasma torches to the furnace could be a potential solution. Furthermore, plasma heating could get much closer to the heat transfer rates achieved with combustion in comparison to traditional heating.

Figure 2.8 shows a typical design configuration of a steam cracker with two rows of radiant tubes and four rows of burners. This study compares radiative heat transfer rates in a cracking furnace with plasma torches to a case of current natural gas burners.



3. Method

This chapter defines the model and methods used in the project. The aim of the project was to assess if the fossil fuel burner could be replaced by plasma burners in specific high temperature industrial processes. The process of aluminium smelting in UTRF is studied in detail with evaluation and analysis of changes in process parameters. Whereas the case for steam cracking is limited to estimation of radiation heat loads for a typical cracker with estimated dimensions and number of burners.

It was fundamental to have a proper understanding of the aluminium smelting process in the current configuration to establish a reference case for comparison and further modifications. Hence, available process data was gathered from Stena Aluminium for one batch of smelting operation. However, several quantities which are not recorded during normal operation. Therefore, models were developed, or literature was used to estimate some of these unknowns. Remaining unknows were calculated by closing the heat and mass balances. Next part of the analysis was to evaluate heat loads through different modes of heat transfer for plasma torch case and reference case. Figure 3.1 shows a graph of burner power with respect to time through the duration of the batch studied. Heat transfer loads were studied at steady-state condition where the burner is working at its full power, that is, 4 MW. The same is represented by yellow bands in figure 3.1.



Figure 3.1 – URTF burner output and steady-operation zone (yellow band)

These heat loads are compared to evaluate the increase in process time and energy input. Further, flow rate and power output for plasma torch are evaluated. In the end, total emissions and energy cost are compared for both the configuration.

3.1 Heat and Mass balance for aluminium smelting

A combined mass and energy balance was carried out for a smelting batch in the rotary furnace at Stena Aluminium's refiner plant in Älmhult. Many of the parameters required for such balances are neither measured nor documented on a regular basis. Figure 3.2 shows all the material streams involved in the process. The scarp fed to the process along with aluminium contains a significant portion of organic matter which is combusted during the process and acts as a heat source. Moreover, some portion of aluminium is oxidized during the process to Al₂O₃. This metal oxidation, also called metal burn-off, provides heat for the melting process. Although, this heat reduces the required amount of fuel for the process, the aluminium spent in the process is far more valuable. Hence, there are measures like addition of salt flux to reduce these oxidation reactions.



Figure 3.2 – Material Streams in smelting process

Table 3.1 contains data on measured quantities as provided by Stena Aluminium for one batch. Off-gas coming out of the furnace is a mixture of flue gases from combustion of propane and organic matter, and partly unreacted leakage air. Dust carried over by off-gas is taken as 0.35% of total scarp feed [27].

Stream	Quantity	Units
Scrap feed	32412	[Kg]
Salt	2530	[Kg]
Oxygen	1961.8	[Nm ³]
Propane	345.1	[Nm ³]
Tapped metal	27496	[Kg]

Table 3.1 – Process data from Stena Aluminium

Slag is the mixture of salt, unburned organics matter and oxidized aluminium left after the metal is tapped. It also contains some quantities of free metal trapped within. The mass balance is carried over separately on both solid and gas phase as the interactions between both the phases are known. These interactions correspond to metal oxidation reaction where oxygen from the gas phase ends up in the slag, represented by Reaction 3.1. Furthermore, organic matter in the feed is combusted which is responsible for mass from solid phase ending up in gas phase. The organic matter is considered to be in the form of long aliphatic chains for which the chemical structure is taken as CH₂ for simplicity in stoichiometric calculations. Reaction 3.2 represents the combustion reaction, and the heat of reaction is 50.3 MJ/kg [28]. Previously, Stena Aluminium in collaboration with Chalmers conducted an internal study on the composition of off-gas stream from the smelting process from the same furnace. It was found that there is no remaining oxygen in the off-gas stream. Hence, all the oxygen remaining after complete combustion of propane and metal burn-off reactions is consumed. The mass of organic substance combusted during the process is calculated from the amount of oxygen available for the combustion. Apart from the pure oxygen fed into the furnace, leakage air is another source.

$$4 A l + 3 O_2 = 2 A l_2 O_3 \tag{3.1}$$

$$2 CH_2 + 3 O_2 = 2 CO_2 + 2 H_2 O \tag{3.2}$$

$$C_3 H_8 + 5 O_2 = 3 C O_2 + 4 H_2 0 \tag{3.3}$$

The primary source of energy for the process is, however, the oxy-propane burner. Reaction 3.3 represents the combustion of propane, and the gross calorific value is 49.35 MJ/kg as per data from Stena Aluminium. This corresponds to a total energy input of 32889 MJ throughout the smelting operation, as observed in figure 3.1. A significant share of the heat required to melt comes from the metal burn-off reaction mentioned earlier, though, the smelting process of contaminated scrap is a complex and unpredictable system. As the objective of an aluminium refiner is to extract a maximum amount of metal from a ton of scrap, metal burn-off rate is one of the most critical factors. Burn-off rate is defined as the fraction of scrap feed which is converted into oxide. Reuter et. al. studied the aluminium scrap melting process in a 20-ton rotary furnace over 26 furnace cycles [27]. Consequently, burn-off rates for different scrap qualities were calculated. Considering the category of scarp based on yield data provided by Stena Aluminium, a burn-off rate of 2.14% is considered for this batch and corresponding heat of reaction is 13,38 MJ of Aluminium oxidized. At elevated temperatures, organic matter also reacts with molten aluminium to form aluminium carbide. This reaction releases hydrogen as a by-product which is then combusted. Reactions 3.4 and 3.5 shows the corresponding reactions, also called carbidisation. Data from Stena Aluminium on composition of slag suggests the presence of carbon content to be around 0.9 wt.% but no traceable amount of aluminium carbide. This suggests that the effect of carbidisation reactions is minimal and is thus ignored for this analysis.

$$4 A l + 3 C H_2 = A L_4 C_3 + 3 H_2$$
 (3.4)

$$6 H_2 + 3 O_2 = 6 H_2 O \tag{3.5}$$

For the energy balance, it is important to account for all energy losses. The major one is the heat that is lost with off-gases. A prior study on Stena Aluminium's smelting process states that the off gases are at 1000°C at the time of leaving the furnace [29]. This is due to short residence time for flue gases inside the furnace and poor convective heat transfer rate. Other studies on full-scale CFD simulations of aluminium scrap melting in rotary furnaces also claim the temperature of the off gas to be around 1000 °C [30] [31]. The energy content in the off gas is given by the equation:

$$Q_{gas} = \sum_{i} m_{i} C p_{i} (T_{i} - T_{ambient})$$
(3.5)

Where summation index '*i*' stands for the major fractions in the off-gas, which are carbon-dioxide, water vapor and nitrogen from leakage air. The off gas is a mixture of flue gases from combustion of propane and organic matter, and partly unreacted leakage air. Thus, energy loss through off gas, Q_{gas} , is a function of just one unknow, that is the mass of leakage air.

Heat is also lost to the surroundings from the outer walls of the furnace. This loss is calculated using the equation 3.6. Ambient temperature is taken as 15 °C whereas the temperature of outside wall of the furnace during operation is 75 °C. A CAD model was constructed for the rotary furnace present at Stena Aluminium's plant. Outside wall surface area, A_{wall} , was found to be 54.9 m². Average heat transfer coefficient was taken to be 15 W/m² K, as suggested for a similar rotary furnace by B. Zhou et al. [31]. Radiative heat losses is not considered as temperature of outside wall is rather low.

$$Q_{wall} = h * A_{wall} * (T_{wall} - T_{ambient})$$
(3.6)

Water produced through combustion reactions 3.2 and 3.3 exists the furnace along with flue gas. Equation 3.5 covers the sensible heat loss associated with these water vapours. However, gross calorific value was considered for combustion energy of propane and organic matter. Hence, heat loss associated with latent heat of produced vapours needs to be considered. This corresponds to 2256.4 kJ/kg of water vapor produced. Table 3.2 gives the energy content of remaining material streams [27].

Stream	Energy	Units
Heat in tapped metal	1265	MJ/ton tapped metal
Heat in Slag	1026	MJ/ton slag
Heat in off-gas dust	910	MJ/ton dust

Table 3.2 – Energy content of various streams

A model was developed for coupled mass and energy balance using the data and information discussed above. As mentioned, the mass balance is carried over separately for both solid and gas phase considering their interaction in the model. Two main unknow quantities calculated from the model were the mass of leakage air into the furnace and amount of slag left after tapping out the molten metal.

3.2 Radiation Model

Radiative heat transfer is the most dominant mode of heat transfer in a rotary furnace or steam crackers [32]. Thus, its fundamental to accurately predict the radiative conditions inside the furnace. Furnaces have an atmosphere rich of gases which participate in radiation process by absorption and emission at elevated temperatures, such as in combustion. However, symmetric diatomic molecules like nitrogen and oxygen molecules are fundamentally transparent to radiation. Carbon dioxide and water vapor are the two main gases that must be considered when it comes to radiative heat transfer. Other gases such as carbon monoxide also participate in radiative heat transfer. However, the concentration of gases other than carbon dioxide and water vapor are so low that contribution from them could be neglected. The temperature, pressure, and composition of a gas mixture determine the emission and absorption characteristics of its constituents.

For a given wave number, v, the radiative heat transfer equation (RTE) given below describes change in radiative intensity along a set direction \hat{s} .

$$\frac{dI_{\nu}}{ds} = \kappa_{\nu} * I_{b\nu} - (\kappa_{\nu} + \sigma_{s\nu}) * I_{\nu} + \frac{\sigma_{s\nu}}{4\pi} \int_{0}^{4\pi} I_{\nu}(\widehat{s}_{\iota}) * \Phi_{\nu}(\widehat{s}_{\iota}, \hat{s}) * d\Omega_{\iota}$$
(3.7)

The three terms on the right side represent the contribution from emission, absorption and scattering away from direction \hat{s} , and scattering into the direction \hat{s} from surrounding directions \hat{s}_i , where $d\Omega_i$ is the solid angle of the ray in the \hat{s}_i direction. κ and σ_s are absorption and scattering coefficients. Whereas, the scattering phase function, Φ_v represents the probability that a ray from the \hat{s}_i direction will be scattered into the \hat{s} direction. Scattering effects only occurs when there are particles present. Therefore, for the case of gas radiation, scattering may be neglected [16]. This is also justified as soot particle are absent in the case for thermal plasma and minimal for the case of complete combustion of propane in pure oxygen. Therefor the RTE simplifies to:

$$\frac{dI_{\nu}}{ds} = \kappa_{\nu} * I_{b\nu} - \kappa_{\nu} * I_{\nu} = \kappa_{\nu} (I_{b\nu} - I_{\nu})$$
(3.8)

Ideally, the RTE should be solved for each spectral line and for all possible directions, but this is not computationally possible. Regarding the gas radiative properties of the gases, for a large system, a fixed average absorption coefficient could be used over all wavelengths. However, there exists models which are midway these two extremes in terms of accuracy and complexity. One such approach is the statistical narrow-band model (SNBM). The RTE is solved for a set number of directions using a discrete transfer method (DTM).

In SNBM, the spectrum is divided into narrow bands and an average value of the absorption coefficient is considered for each band. The spectrum must be divided into bands that are both wide enough to contain many lines to allow statistical treatment and narrow enough to enable for constant black-body radiation assumption in each band. An in-house model based on DTM is used for calculating radiation intensities in the rotary furnace. It considers representative rays in volume between centre and furnace walls in predetermined directions. The rays are set to be directed to a point and distributed uniformly in the angular direction. The furnace is modelled as an indefinitely long axisymmetric cylinder. Gunnarsson [33] provides a detailed overview of the model used in the study. Radiation characteristics of a gas mixture depends upon temperature, pressure, and composition of a gas mixture. The model uses temperature and concentration profile of carbon dioxide and water vapor in the furnace to calculate radiation intensity as a function of radial distance in a rotary furnace.

3.2.1 Gas and temperature profiles for aluminium smelting

Concentration of carbon dioxide and water vapor for the case of oxy-propane combustion is known from mass balance analysis. For the plasma torch case, carbon dioxide was considered as the working gas and only source of water vapours is from combustion of organic matter. The concentration profile was considered uniform throughout the furnace. Yang *et al.* [34] conducted a full scale CFD analysis of aluminium scrap melting process in a rotary furnace. As result, it provides temperature distribution inside the furnace at process times, as shown in figure 3.3.



Figure 3.3 – Temperature distribution in rotary furnace (used with courtesy of [34])

This temperature distribution was used as an input for the radiation analysis. As the radiation model has a radial symmetry, the furnace was discretized into three longitudinal zones and model used to calculate radiation intensities separately for each zone. Three zones were considered enough to simplify temperature contours in figure 3.3 to fit the radial symmetry. Figure 3.4 shows the three zones and corresponding temperature profile.



Radiation intensity function obtained from the model is numerically integrated over the molten pool surface to obtain radiative heat transfer load. Equation below gives the heat transfer rate:

$$H = \int I(r) * \pi * dA * \cos(\theta)$$
(3.9)



Figure 3.5 – Furnace cross-section

The surface of molten metal pool was discretized into strips of area dA for performing the integration as per equation 3.9. θ is the angle between radial direction and normal to pool surface. Whereas r represents the radial position of strip dA. As the function of radiation intensity, I(r), is different for each zone, thus the integration is performed separately for each zone. The sum of heat transfer rates in each three zones gives the total radiative heat transfer load.

3.2.2 Gas and temperature profiles for steam cracker

Figure 3.6 shows top view and dimensions of the steam cracker considered for the analysis in this work. The furnace was set to be 4 meters times 4 meters with two rows of tubes and four rows of burners with a total of 16 burners. Burners are placed such that the rows are equally spaced with each other and the walls. The gas composition was taken as uniform throughout the furnace. Different flame temperatures and gas compositions were analysed to study the effect of both parameters on the radiative heat transfer rates.



Figure 3.6 – Top view of cracking furnace

The different combinations of set gas composition and flame temperature studied are shown in Table 3.3. For these cases, the working gas for the plasma torch was considered to be mainly CO_2 with a smaller amount of H_2O for increased gas radiationand the composition was varied for the studied average temperature of 2000°C.

Case	Maximum flame temp (°C)	Gas composition (CO ₂ % - H ₂ O %)
1	1500	80-20
2	1750	80-20
3	2000	80-20
4	2250	80-20
5	2500	80-20
6	2000	90-10
7	2000	85-15
8	2000	75-25
9	2000	70-30

Table 3.3 – Parameters for various cases in steam cracking furnace

Figure 3.7 shows the temperature profile for the cases with 2000°C as the flame temperature. For all studied cases, the wall temperature is maintained constant at 700°C whereas furnace gas temperature outside of the flame is taken as 800°C. As can be observed, the flames are considered to be quite narrow in comparison to the full size of the steam cracker.



Figure 3.7 – Temperature profile used for a maximum flame temperature of $2000^\circ C$

3.3 Convection and Conduction

Convection heat transfer coefficient in a rotary furnace is considered to mostly depends upon gas flux, G, which is a fraction of flow rate and cross-sectional area. Due to the tilt of the furnace, the freeboard cross/sectional area varies a lot with position along the axis. Hence, the heat transfer coefficients and consequently the convection rate was found separately for each of 3 zones described in prior section. The cross-sectional freeboard area was measured from the CAD model of rotary furnace, shown in Figure 3.6 below.



Figure 3.8 – Sectional view of UTRF with molten pool plane

Equation 3.9 was used to estimate the heat transfer coefficient from gas to molten pool surface [32]. It has been used in multiple studies for rotary kilns [35].

$$h = 0.4 * G^{0.62} \tag{3.10}$$

$$Q_{conv} = h * A * (T_{gas} - T_{pool})$$
(3.11)

A denotes the area of molten pool surface which was calculated to be 13.02 m² from the CAD model.

Conduction mode plays a small role, around 5% to 7%, in overall heat transfer in a rotary furnace [32]. Moreover, it depends on rotational speed, fill ratio and properties of refractory material inside a furnace. These parameters stay the same for both configurations, plasma burner and oxy-fuel burner. Thus, the contribution of conduction mode of heat transfer was taken to be same for both the configurations.

For the steam cracker calculation, convective and conductive heat transfer is no studied since the focus was to study the changes in radiative heat transfer.

3.4 Plasma Torch

In the case for steam cracking furnace to calculate radiative heat loads, only the temperature of the plasma was a factor of interest as far as the torch is concerned which was varied between 1500 and 2500°C as shown is table 3.3.

The plasma torch for aluminium smelting was considered to be of the same power, 4 MW, as the maximum power of installed oxy-propane burner. This allows us to have

the assumption of similar temperature distribution which was fundamental for radiation analysis. Required flow rate of working gas, that is, carbon dioxide needed for the torch was calculated from plasma enthalpy data, which was calculated using FactSage [™] software, and torch efficiency provided by ScanArc Plasma Technologies AB.

Finally, the change in process time was calculated from the difference [36] in overall heat transfer rates for both the configurations and total energy that is needed for the process. Further, carbon emissions and energy costs were compared for both the configurations.

4. Results

This chapter presents the results from the study. Firstly, the result from energy and mass balances for aluminium smelting process are presented and discussed. Then the flow rate and parameters of the plasma torch used for the smelting process are presented. Next section offers a comparison on heat transfer rates and changes in process parameters for propane burner and plasma burner in smelting furnace. Finally, radiative heat load and results for parametric analysis in steam cracker are presented.

4.1 Heat and Mass balance in aluminium smelting

A combined heat and mass balance analysis was carried out to establish an understanding of the current process and to calculate the unknown parameters which are not measured during standard operation at Älmhult plant. The main objective was to calculate the amount of leakage air entering the furnace and the heat input provided by metal oxidation reactions and combustion of organic content. Figure 4.1 shows the share of energy or heat coming from three different sources. The greatest share, 58% comes from the external energy source which is combustion of propane. Therefore, 42% of heat needed for the smelting process is generated internally in the pool of scrap. From the metal burn-off, being to 2.14% of the aluminium load, heat corresponding to about 9281 MJ was released. Whereas the heat from combustion of organic matter is significant at about 14400 MJ, found from the oxygen mass balance since there was no oxygen in the flue gas. Total mass of organic matter undergoing combustion was found to be about 278,35 kg which is around 0.86% of total scrap feed.



Figure 4.1 – Share of heat from different sources

Figure 4.2 shows the energy content of various output streams and losses in the process. The energy content of each stream could be calculated from respective mass flow and temperature, except the unknown leakage stream, which is instead found as the residual from an energy balance. The total amount of leakage air entering the furnace during the complete batch was found to be 722 kg. This leakage air provides additional oxygen for combustion of organic matter, which is included in the results shown in Figure 4.1. However, the unreacted fraction of leakage air, majorly nitrogen, exists furnace at high temperature and adds to the off-gas losses. Losses through walls of furnace and heat carried by dust together accounts for less than 0.2% of total energy.



Figure 4.2 – Losses and energy content of outlet streams

4.2 Plasma torch for aluminium smelting

The working gas considered for the plasma torch is pure carbon dioxide. Water vapours would still be present in the furnace due to combustion of organic matter. However, it was found that the concentration of H_2O is considerably reduced as compared to the case with oxy-propane burner. Gas composition used for radiation analysis was therefore estimated to be 90% CO₂ and 10% H_2O for the case with plasma torch. The concentration profile was considered uniform throughout the furnace.

The power output of plasma torch was set to 4 MW. Considering electricity to heat conversion efficiency of 90%, the heat output of the torch is 3.6 MW. Also, the flame temperature of plasma torch was set to 2000°C to maintain the same temperature distribution as for oxy-propane case. This corresponds to 2.81 MJ/kg-CO₂ of plasma

enthalpy and a flow rate of 1.28 kg/sec. For a comparison, the flow rate of off-gas from the furnace in oxy-propane case is 0.89 kg/sec.

4.3 Heat transfer rates in aluminium smelting

Figure 4.3 shows the graph of radiation intensity with respect to radial position in zone A of the furnace for both cases. It could be seen that there is considerable decrease in radiation intensity from oxy-propane case to plasma burner case. This reduction was also observed for zones B and C of the furnace. The drop is due to change in furnace atmosphere, that is the composition of gases inside the furnace.



Figure 4.3 – Radiation intensity vs Radial position

Table 4.1 shows comparison of radiation modes of heat transfer respectively for all three zones. Plasma burner provides about 20% lower total radiative heat load than oxy-propane burner.

Zone	Oxy-Propane Burner	Plasma Burner	
Zone A	525.95 kW	381.23 kW	
Zone B	376.67 kW	300.76 kW	
Zone C	492.86 kW	426.20 kW	
Total	1395.48 kW	1108.20 kW	

Table 4.1 – Radiation Heat transfer rates

A noticeable increase in convection heat transfer rate in plasma case was noted. This could be attributed to higher gas flow rate and consequently better heat transfer coefficient. As reasoned in prior section, conduction was considered to contribute 7% to total heat transfer in the process with oxy-propane burner. This corresponds to 125 kW. The rate of conduction depends upon rotational speed and furnace characteristics which remains same for both configurations, Hence, considered same for both the cases. Table 4.2 provides summery of heat transfer analysis.

Table 4.2 – Total Heat transfer rates	
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Modes	Oxy-Propane Burner	Plasma Burner	
Radiation	1395.48 kW	1108.20 kW	
Convection	243.9 kW	305.4 kW	
Conduction	125 kW	125 kW	
Total	1764.38 kW	1538.6 kW	

Considering these heat loads and total external energy needed to melt, melting time for the case with plasma burner is 14% higher than with oxy-propane burner. Consequently, the heat needed for the process also increases by 14% as power outputs of both burners is the same. Subsequently, the electricity needed for the process would be 11572 kWh, taking into consideration the 90% power-to-heat efficiency of plasma torch.

4.4 Carbon Emissions in aluminium smelting

Total carbon dioxide emission for the original process was calculated to be 2869 kg. Major share of this, about 70%, comes from the combustion of propane and remaining 30% from combustion of organic matter in scrap. Ideally the process with plasma torch with carbon dioxide should be run in closed cycle where the working gas must be recycled. Average carbon intensity of Swedish electricity grid is 10 gm CO₂ per kWh [37]. Considering total electricity consumption for the batch, emissions from electricity use would be 104.1 kg. Therefore, modifying the process with plasma torch cuts CO₂ emissions from energy source by 94.7%. Overall process emissions are reduced by 67%. By further pre-treatment steps for the scrap like de-coating [38], the process could result in net negative emissions. This could be possible as the emissions from combustion of organic matter could end up being captured and recycled in the process. This is further discussed in the next chapter.

4.5 Energy cost in aluminium smelting

Cost for energy was calculated for the analysed batch, based on cost of commodities as per Stena Aluminium. Due to the relatively large electricity demand for the plasma torch case, it was found that there is a 39.1% increase in energy costs for the modified process with plasma torch. Table 4.3 presents detailed results

Commodity	Commodity Consumption		st	Total (Euro)
Oxygen	2611.2 kg	0.13	Euro/kg	339
Propane	666.4 kg	0.38	Euro/kg	253
Total for Oxy-propane case				592
Oxygen	1402 kg	0.13	Euro/kg	188
Electricity	11572 kWh	0.055	Euro/kWh	636
Total for Plasma torch case				824

Table 4.3 – Energy costs for one batch

4.6 Radiation Intensity in cracking furnace

Radiation intensity at tube surface in cracking furnace was evaluated using the model and parameters discussed in section 3.2. Figure 4.4 shows the line-of-sight radiation intensity for the case with 2000°C flame temperature and 80% - 20% concentration of CO_2 and H_2O respectively. The plot shows location with respect to furnace side walls on the x-axis in meters whereas the two rows of tubes are located at a distance of 1.2 and 2.8 meters from each of the side wall. Line-of-sight radiation intensity gives the intensity from one side of the furnace. Hence, contribution from both sides are added to get the total radiative intensity incident on the tubes. That is, the intensities calculated at positions 1.2 and 2.8 m from the left wall are the considered contributions to each radiant tube, with assumed equal view factors.



Figure 4.4 – Radiative Intensity (case 3) for the steam cracker

Peaks in the plot corresponds to location of burner/flame or plasma torch. Following a ray of light leaving the flame, that is following the intensity with increasing distance from a plasma, the intensity decreases as heat is absorbed by the cooler gases surrounding each plasma/flame. Table 4.3 gives the radiative intensity at the wall and at location of tubes for all the studied cases.

Case	Maximum flame temp (°C)	Gas composition (CO ₂ % - H ₂ O %)	Radiation Intensity at wall (kW/m ₂ .sr)	Radiation Intensity at tubes (kW/m ₂ .sr)
1	1500	80-20	21.10	42.94
2	1750	80-20	23.56	46.84
3	2000	80-20	26.86	52.32
4	2250	80-20	30.97	58.00
5	2500	80-20	35.84	65.07
6	2000	90-10	25.91	49.93
7	2000	85-15	26.44	51.00
8	2000	75-25	27.20	52.66
9	2000	70-30	27.50	53.32

Table 4.4 – Radiation intensity at wall and tube

5. Discussion and Future work

This chapter builds on the knowledge gained from process modelling and discusses possible process schemes for aluminium smelting using plasma torch as the heat source.

5.1 Open-cycle operation for smelting process

With open-cycle operation, everything downstream the furnace stays the same as in the original configuration and described in section 2.2.1. However, the working gas used for the torch is expanded and must be replenished. The flow rate for torch with carbon dioxide and the working fluid was found to be 1.28 kg/sec. This corresponds to requirement of around 12 ton of carbon dioxide for just one batch. Apart from the problems with storing such large volume of gas on premises, the cost associated with buying the gas makes it unfeasible. Moreover, it will increase the carbon dioxide emissions many folds.

Therefore, carbon dioxide is not suited for use as working gas without a recycling system due to the cost of gas itself. For such a configuration, air would be much more suited as a working gas. It would still provide benefit of substituting propane with

electricity and corresponding emission reduction with minimum capital expenditure. Apart for the torch itself and associated power system, an air compression unit would be the only addition component needed.

However, the current study was focused on using carbon dioxide as a working gas for its radiative properties. It would best imitate the conditions as observed in combustion chambers currently. Usage for air which is mostly transparent to radiation would reduce the heat transfer loads, further increasing the process time and electricity demand. It must also be evaluated how a significant increase in oxygen concentration inside the furnace would affect the process chemistry. Moreover, NOx emissions would be a problematic due to high temperatures inside the furnace.

5.2 Closed-Cycle Operation for smelting process

For the reasons discussed earlier, a system to recycle off-gas would be necessary to use carbon dioxide as the working gas. In plasma case, the off gas coming out from afterburner unit would consist of just carbon dioxide and water vapours. This could be recycled back to plasma torch in either a wet or dry stream after minor compression. With both open and closed cycle processes, oxygen is still needed in the process for burning of organic matter and for metal burn-off reactions. Evaluation of an optimum feed of oxygen for highest yield of AI from scrap would interesting to study as a future prospect.

De-coating is a pre-treatment step for scrap where it is heated at temperature well below melting point in oxygen less environment. This leads to pyrolysis of organic matter and releases about 90% of organic matter as volatile compounds. De-coating increases the yield form scrap [38] and reduces the need for oxygen inside the rotary furnace during melting process. Although, it must be evaluated how the carbon residue left after pyrolysis affects the yield of aluminium in the process.

5.3 Steam Cracking process

The radiative intensity at the tube surface in a cracking furnace was evaluated. Table 4.3 presented the intensities for all the cases considered. It could be observed that the radiative intensity at the tubes was much higher than at the wall of the furnace, as expected due to the tubes being placed between the rows of burners/plasma torches. Figure 5.1 shows the intensity with respect to the maximum flame temperature at a constant gas composition, i.e., cases 1-5.



Figure 5.1 – Intensity dependance on flame temperature

The intensity increases at both wall and tubes with increasing flame temperature, and it is interesting to note that the dependance is rather strong. Therefore, it seems that an increased temperature would be beneficial for the process due to the increased heat load on the tubes. However, this will probably also be related to increased flue gas temperatures and thereby increased heat losses. Further examination of the process would therefore be recommended. Figure 5.2 shows the intensity with respect to gas composition at a constant flame temperature of 2000°C, i.e., cases 3 & 6-9. This shows that increasing proportion of H₂O increases the radiation intensity. This is due to H₂O being a stronger absorber, and therefore emitter, of radiative heat in comparison to CO₂. It would therefore be interesting to increase the gas concentration of H₂O even further in future studies. However, using H₂O as the working gas could eventually cause some operational related problems with the plasma torch and needs evaluation. Also for future studies could be to examine possible ways to increase the radiation from the plasma by introducing particles to the high temperature zones since particles are considered to be broad emitters of radiative heat.



Figure 5.2 – Intensity dependence on gas composition

To better evaluate the findings from the steam cracker simulations, it would be good to compare to present conditions in a real steam cracker. That is, comparing the findings in this work to similar simulations based on actual dimensions, temperature conditions and gas compositions from a steam cracker in operation.

6. Conclusion

Aluminium smelting process in a rotary furnace was modelled with oxy-propane burner and a plasma torch for comparison of the two cases. It was understood that heat load for thermal plasma falls short of that from combustion. This results in an increase in process time of around 14%. This could be mitigated by using a plasma torch of higher output. However, how that would affect the temperature distribution inside the furnace and process chemistry is a matter of future investigation. Furthermore, with the electricity and fuel prices as in year 2021, there is about 39% increase in energy cost for the batch.

It was found that the use of carbon dioxide in open cycle configuration would not be viable. Hence, a flue gas recycling system is fundamental to avoid additional emissions and feasibility of the whole process. Modifying the process with plasma torch cuts CO₂ emissions from energy source by 94.7%, owing to very low carbon intensity of Swedish power sector. Overall process emissions are reduced by 67%, in case no pre-treatment steps are employed. To conclude, the adoption of the process of aluminium scrap melting with plasma torch as an energy source is possible with additional energy costs.

Exchanging the burners for plasma torches in a steam cracker was also examined in this work. Based on estimated dimensions, burner placements, gas temperatures and gas compositions, it was shown that the heat load to the tubes is increased with the plasma temperature as well as with an increased gas concentration of water. There is a potential to install plasma torches to a steam cracker, but a heat load comparison based on dimensions and operational data from a real steam cracker would be beneficial to draw better conclusions.

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