Doctoral Thesis in Material Science and Engineering

Mixing Time and Decarburization Reactions in Side-blown Metallurgical Converters

A Practical Approach using CFD and Thermodynamics

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Stockholm, Sweden 2023
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Abstract

The side-blowing Argon Oxygen converter (AOD), known for its intense gas stirring and turbulent nature, poses complex fluid dynamics and thermodynamic challenges. Modeling has played a substantial role in the development of metallurgical converters, particularly in understanding jet behavior, mixing, flow patterns, and chemical reactions. Flow characteristics and mixing time are recognized as crucial factors that enhance the efficiency and decarburization rate in metallurgical reactors. However, to the best of the author’s knowledge, no prior study has investigated the impact of mixing time on the decarburization reaction. While most studies suggest that reducing mixing time is beneficial, it is reasonable to assume that there might be a point at which further reduction in mixing time does not lead to an increase in reaction rates. Adjustments like tilting the converter or repositioning the nozzles could improve decarburization efficiency by altering pressure conditions and mixing. This study aims to explore how these factors affect the decarburization reaction in side-blowed converters through modeling. The work has been done in a few steps resulting in different supplements.

Side-blowing water model experiments were carried out to investigate how a vessel inclination would affect the mixing time. The results showed a clear increase in mixing time when higher inclination angles (14°) were applied. However, studying the non-reacting water models could only give insight to mixing efficiency and not provide information about decarburization efficiency.

A numerical model capable of integrating mass and heat transfer with high temperature chemical reactions was developed to aid in this investigation. First, the model was applied to an ascending gas bubble in liquid steel. The effect of pressure was investigated by injecting the bubble in different bath depths. It was shown that a mere oxygen bubble injected at the nozzle position under industrial conditions did not decarburize efficiently, rather dissolved into the steel. Only pressure levels at the bath surface could maintain gas as a stable phase and decarburize efficiently.

With high grid resolutions the model consumed a lot of computational time calculating equilibrium locally in each cell with gas and liquid present. Therefore, a more practical approach was taken to study the AOD converter that showed high agreement to the first decarburization step when comparing against two industrial heats. It was shown that with a coarse Computational Fluid Dynamic (CFD) solution the model could be practical, yet fundamental. In the study it was also found that no chromium oxidation was found in one of the heats at the beginning of the process when the initial carbon content was high. The trends were compared against an industrial online process model and showed similar behavior.

With further developments, the model was tested with different treatments of the thermodynamic coupling, including reactions limited by turbulence in an intensely stirred side-blown reactor. The mixing time was shown to have an insignificant effect on the decarburization rate. The system was governed by thermodynamics and gas supply rate.

Overall, this work developed a general model capable of coupling chemical reactions with CFD. The use of this model led to the conclusion that an inclination of the vessel within practical operational angles would not benefit the decarburization rate in the early stages of decarburization. With increased mixing times and small pressure variations from the lowered bath height, the benefits to decarburization might not be worth compared to the engineering challenges posed by such changes. Even relocating the nozzle would require large and impractical height differences to acquire the pressure decrease needed to benefit thermodynamically.
Sammanfattning

AOD konvertern, känd för sin intensiva gasomrörning och turbulenta natur, ställer komplexa utmaningar inom fluidmekanik och termodynamik. Modellering har spelat en betydande roll i utvecklingen av metallurgiska konverter, särskilt för att förstå jetbeteende, omrörning, flödesmönster och kemiska reaktioner. Flödessegenskaper och omrörningstid betraktas som avgörande faktorer som förbättrar effektiviteten och kolfärsningshastigheten i metallurgiska reaktorer. Men enligt författarens kännedom har ingen tidigare studie undersökt effekten av omrörningstid på kolfärsning. Även om de flesta studier föreslår att en minskning av omrörningstiden är fördelaktig, är det rimligt att anta att det kan finnas en punkt där ytterligare minskning av omrörningstiden inte leder till en ökning av reaktionshastigheten. Justeringar som att luta konvertern eller omplacerat dy桑na kan förbättra kolfärsningseffektiviteten genom att ändra tryckförhållandena och omrörningen. Målet med denna studie är att utforska hur dessa faktorer påverkar kolfärsningsreaktionen i sidobåsta konverter genom modellering. Arbetet har utförts i flera steg med olika tillägg.

Experiment med vattenmodeller av sidobåst gas utfördes för att undersöka hur en lutning av behållaren skulle påverka omrörningstiden. Resultaten visade tydligt en ökning av omrörningstiden när högre lutningsinklagar (14°) användes. Dock kunde studier av icke-reactiva vattenmodeller bara ge insikt om omrörningseffektiviteten och inte ge information om hur effektiv kolfärsningen är.


Med ytterligare utveckling testades modellen med olika behandlingar av termodynamisk koppling, inklusive reaktioner begränsade av turbulens i en intensivt omrörd sidobåst reaktor. Omrörningstiden visade sig ha en obetydlig effekt på kolfärsnings hastigheten; istället styrdes systemet av termodynamik och gasleveranshastighet.

Sammanfattningssvis utvecklades en generell modell i detta arbete som kan koppla samman kemiska reaktioner med fluidynamiska beräkningar. Användningen av denna modell ledde till slutsatsen att en lutning av behållaren inom praktiska driftsinklagar inte skulle gynna färsningshastigheten i de tidiga stadierna av kolfärsning. Med ökade omrörningstider och små tryckvariationer från sänkt badnivå skulle födelarna med kolfärsning vara små jämfört med de tekniska utmaningar som sådana förändringar medför. Även omplacerat av dysan skulle kräva stora och opraktiska höjdpunkt i för att uppnå den trycksänkning som behövs för att gynna termodynamiken.
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Stockholm, September 2023

Serg Chanouian
Supplements

The current work is based on the following supplements:

**Supplement I**

**Inclination Effect on Mixing Time in a Gas–Stirred Side–Blown Converter.**

Chanouian, S., Ahlin, B., Tillander, A. and Ersson, M. *steel research int.* 2021, 92: 2100044. [https://doi.org/10.1002/srin.202100044](https://doi.org/10.1002/srin.202100044)

(Parts of the work was presented in the ESTAD21 conference)

**Supplement II**

**Decarburization Reactions in the Argon–Oxygen Decarburization Converter Using Coupled Computational Fluid Dynamics and Thermodynamics Databases.**

Chanouian, S., Ahlin, B., Tillander, A. and Ersson, M. *steel research int.* 2022, 93: 2200156. [https://doi.org/10.1002/srin.202200156](https://doi.org/10.1002/srin.202200156)

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**Supplement III**

**Modelling Decarburization in the AOD Converter: A Practical CFD-Based Approach with Chemical Reactions.**

Chanouian, S., Pitkäla, J., Larsson, H., Ersson, M.

(Under revision in Metallurgical and Materials transaction B)

**Supplement IV**

**The Importance of Mixing Time in Intensely Stirred Metallurgical Reactors: Applied on Decarburization Reactions.**

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The contributions of the author to the supplements are as follows:

Supplement I
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Supplement II
Literature review, Model conceptualization, Model setup, Analysis, Writing the manuscript.

Supplement III
Literature review, Model conceptualization, Model setup, Analysis, Writing the manuscript.

Supplement IV
Literature review, Model conceptualization, Model setup, Analysis, Writing the manuscript.
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Nomenclature

Modified Froude number \( Fr' \) \([\text{-}]\)
Density \( \rho \) \([\text{kg}\cdot\text{m}^{-3}]\)
Volumetric gas flow rate \( Q \) \([\text{Nm}^{-3}\cdot\text{min}^{-1}]\)
Gravitational acceleration \( g \) \([\text{m}\cdot\text{s}^{-2}]\)
Inner nozzle diameter \( d_n \) \([\text{m}]\)
Penetration length \( L \) \([\text{m}]\)
Scale factor \( \lambda \) \([\text{-}]\)
Inclination angle \( \theta \) \([\degree]\)
Velocity vector \( \vec{v} \) \([\text{m}\cdot\text{s}^{-1}]\)
Time \( t \) \([\text{s}]\)
Pressure \( p \) \([\text{Pa}]\)
Dynamic viscosity \( \mu \) \([\text{kg}\cdot\text{m}^{-1}\cdot\text{s}^{-1}]\)
Turbulent viscosity \( \mu_t \) \([\text{kg}\cdot\text{m}^{-1}\cdot\text{s}^{-1}]\)
Total energy \( E \) \([\text{J}]\)
Effective thermal conductivity \( k_{\text{eff}} \) \([\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}]\)
Temperature \( T \) \([\text{K}]\)
Shear energy \( \tilde{\tau}_{\text{eff}} \) \([\text{J}]\)
Specific Enthalpy \( h \) \([\text{J}\cdot\text{kg}^{-1}]\)
Specific heat \( c_p \) \([\text{J}\cdot\text{kg}^{-1}\cdot\text{K}^{-1}]\)
Kinematic viscosity \( v \) \([\text{m}^2\cdot\text{s}^{-1}]\)
Scalar \( \phi \) \([\text{-}]\)
Diffusion coefficient \( \Gamma \) \([\text{kg}\cdot\text{m}^{-3}\cdot\text{s}^{-1}]\)
Molecular mass diffusivity \( D_m \) \([\text{m}^2\cdot\text{s}^{-1}]\)
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Introduction

1. Introduction

1.1 Background

In 2022, around 55 million tons of stainless steel were produced around the world[1]. Stainless steels are used primarily for their corrosion resistant material properties caused by the thin layer of passive, stable chromic oxide. Although there exists a vast amount of different stainless steels with various compositions, the common ground is that they contain at least 10% chromium[2]. The general route to produce stainless steel is a two-stage process where scrap and alloying elements are melted in the Electric Arc furnace (EAF). The melt is then transferred into the Argon Oxygen Decarburization (AOD) process where the carbon levels are reduced (decarburization). Decarburization is an important refinement step in stainless steelmaking to reduce the carbon content in the steel. This process predominantly involves the introduction of oxygen gas and is a significant stage in secondary steelmaking processes. In the AOD, an added complexity arises from the necessity to preserve the stainless properties acquired by elements like chromium. Consequently, a challenge emerges in achieving the oxidation of excess carbon while avoiding the oxidation of chromium. The primary reactions governing this interplay in the AOD process are outlined as follows in equation 1-3[3]:

\[2[\text{C}] + \{\text{O}_2\} \rightarrow 2\{\text{CO}\}\]  \hspace{1cm} (1)

\[4[\text{Cr}] + 3\{\text{O}_2\} \rightarrow 2\{\text{Cr}_2\text{O}_3\}\]  \hspace{1cm} (2)

\[3[\text{C}] + (\text{Cr}_2\text{O}_3) \leftrightarrow 2[\text{Cr}] + 3\{\text{CO}\}\]  \hspace{1cm} (3)

Where [] denotes dissolved in the melt, {} means it is gaseous, and () that it is in the slag.

The primary method of stainless-steel production since the late 1960s has been the AOD process. The process generally consists of three stages: decarburization, reduction, and desulfurization. In the decarburization stage, a combination of inert gas and oxygen is introduced into the converter body through side nozzles (occasionally also a top lance). This mixture facilitates the oxidation of carbon in the melt. The decarburization process consists of several sequential stages, with the initial stage employing the highest oxygen concentration. Subsequent stages proceed with a reduction in oxygen and an increase in inert gas. These steps are designed to maintain a low partial pressure of CO in the gas bubbles, preventing the oxidation of Chromium in equation 3. The partial pressure of CO in the gas bubble can be illustrated as expressed in equation 4.

\[p_{CO} = \frac{n_{CO}}{\sum_{i=1}^{N} n_i} P_h\]  \hspace{1cm} (4)

Where, \(p_{CO}\) is the partial pressure of CO, \(n\) is the moles with \(i\) representing the index for the gas component in the gas mixture with \(N\) components, and \(P_h\) is the static pressure at the height \(h\).

An alternative to the AOD process is the vacuum oxygen decarburization converter (VODC) process[4], wherein vacuum capabilities are incorporated during the later stages to reduce the partial pressure of CO. However, adopting the VODC process involves a relatively substantial investment, and the long-term operational costs can be uncertain. Furthermore, the VODC process complicates the addition of scrap materials to the procedure.
Introduction

It is suggested that when a mixture of inert and oxygen gas is ascending in the melt, chromium and iron are primarily oxidized. As the oxides ascend through the melt, carried by the inert gas, they oxidize carbon[5]. However, with higher dissolved carbon content in the melt it has also been found that oxygen tends to primarily oxidize carbon[6,7] even at higher partial pressures found at the nozzle region. This means that a variation of practical parameters in the side-blowing converter such as tilting the converter or repositioning the nozzles vertically, to reduce the ferro-static pressure imposed by the liquid above could introduce a more efficient decarburization. These practical alterations will not only affect the thermodynamic conditions for the decarburization but also the fluid dynamics hence, impacting the stirring efficiency. The focus of this study will be to fundamentally investigate how pressure conditions and mixing efficiency affect the decarburization reaction in side-blowed converters.

Due to the vigorous gas stirring and the turbulent characteristics within a side-blowing AOD converter, this process presents intricate fluid dynamics and thermodynamic challenges. Nonetheless, the growing interest in this process has ignited enthusiasm for enhancing its efficacy. Consequently, substantial efforts have been dedicated to investigating the system through modeling approaches such as numerical and physical modeling. Models of these kinds are well-suited for situations involving phenomena that are otherwise unobservable or difficult to measure. They offer the potential for cost-effective design solutions and transparency. In addition, it is well-established that the steel industry contributes to greenhouse gas emissions[8], with the stainless-steel sector inherently categorized under CO2 emissions[9]. While the precise impact of the AOD process on these emissions hasn’t been determined by the author, it’s worth noting that advancements in the process of decarburization through modeling could hold promise as a stride toward sustainability. Especially by employing models to eliminate the trial-and-error approach.

Modelling has been an extensive part of the metallurgical converter development with focuses on jet behavior, mixing and flow characteristics, and chemical reactions. A convenient method to study fluid flow has been to scale down metal reactors to physical models by ensuring similar forces between the reactor and the prototype, which is commonly achieved by satisfying the geometric similarity and the modified Froude number[10,11], equation 5. The latter yields the ratio between the inertial forces of the injected gas and the buoyancy forces acting on the gas bubbles. With this, studies on the behavior of horizontally injected gas into liquid have brought forth semi-empirical equations of the gas penetration length of horizontally submerged nozzles[12,13], equation 6[12].

\[
Fr' = \frac{\rho_g Q^2}{\rho_l g d_n^2}
\]

\[
L = 10.7 Fr^{0.46} d_n \left(\frac{\rho_g}{\rho_l}\right)^{0.35}
\]

Where, \(\rho\) is the density with indices \(g\) and \(l\) representing gas and liquid respectively, \(Q\) is the gas flow rate, \(g\) is the gravitational acceleration and \(d_n\) is the nozzle diameter.

Advances in side-blowing physical modelling (mostly water models) have revealed insights into typical flow patterns and mixing time. Mixing time is generally defined as the time it takes for a local change in a domain to homogenize up to 95%. This is an effective parameter to measure the stirring efficiency in converters. Kim and Fruehan[14] studied the effect a slag layer has on the mixing time and found that the mixing time in the water model is increased with the presence of an oil layer due
to interference with the circulatory motion created by the rising gas plume. Different parameter combinations of nozzle number, angular positions, side and top-blowing, and gas blowing rate were studied[15-17]. The studies found a governing role of gas flow rates and angle separation in the side nozzles on the flow characteristics and the mixing effectiveness. An increase of the gas flow rate through the side nozzle initially decreased the mixing time up to a flow rate where it increased again. The top lance generally increased the mixing time in all configurations. In addition, a larger angular separation between six nozzles (27°) improved mixing efficiency. A prolonged mixing time was also observed in another study where a top lance was introduced, with or without oil acting as a slag layer[18]. Variation in the vessel shape[19] and diameter[20] showed an increase in mixing time with an increasing diameter and a negligible change when an oblong shaped vessel was applied. Even the bath height has been shown to affect the flow and mixing characteristics[21,22]. Raising the bath height initially increases the mixing time until a point where it remains constant with an optimal fill level in the range 0.7 < height/diameter < 0.75.

Although down-scaled physical models have shown to be useful for understanding metallurgical converters, they are commonly accompanied by numerical models with Computational Fluid Dynamics (CFD)[23]. The advantage of numerical models is the possibility of scaling up to industrial scale and provides the freedom of varying parameters such as including amounts of liquid and slag phases, or varying flow rates of gas [24]. Furthermore, numerical models can be used to investigate the outcome of adding another nozzle to the AOD converter[25]. However, the accuracy of CFD models comes at the dispense of computing time and incredibly accurate solutions of, for instance, a ladle converter has reported simulation-times of up to 9 months[26]. Overall, these studies have collectively contributed to understanding the mixing and flow dynamics in side-blown converters. The general flow pattern observed is a large circulatory motion caused by the buoyancy force acting on the gas bubbles and the drag force between these bubbles and the melt as the bubbles ascend. The fluid is dragged by the bubbles/bubble plume, up to the bath surface, and pushed to the opposite wall causing wave like motions at the surface, where it is later dragged down alongside the walls and back to the nozzle region forming a large, centered vortex.

Throughout the years, the numerical development in side-blown converters such as the AOD converter has extended beyond fluid dynamics to also consider chemical reactions. The AOD converter’s critical carbon content and oxygen distribution has been analyzed with a kinetic reaction model where oxygen distribution in the steel relies on reaction affinities to oxygen[27-31]. Prediction of dissolved nitrogen and metal/slag compositions using thermodynamic software with reliable databases, effectively tracked nitrogen changes across the AOD stages[32]. To overcome the static pressure limitations and one cell equilibrium, a kinetic model considered conservation equations, evolving to encompass parallel reversible reactions within a rising gas plume, initially studying a stagnant bubble and expanding to a full AOD converter[6,33-35]. An effective equilibrium reaction zone (EERZ) model[36] assumed equilibrium of phases located in zones at reaction interfaces, simplifying mass transfer equations with uniform mass transfer coefficients. This approach demands specific physical descriptions of kinetics in each zone. These types of models can be used as online process models due to their short computational time. However, there also exist models that combine CFD [7,37] with chemical reactions at local volumes which eliminates the interfacial data requirements at the expense of being computationally intensive. This is due to its mass transfer descriptions that resolves multiphase features.
1.2 Main Objectives of this work

The overall objective of this thesis was to develop a numerical model capable of combining CFD with high temperature chemical reactions. The purpose of the model was to aid in the investigation on how the pressure and mixing efficiency affects the decarburization in intensively stirred side-blown converters. The structure of the supplements and their connections is presented in Figure 1. Firstly, the mixing time in a side-blown converter was measured in a physical water model (Supplement I). A tilt angle was applied to reduce the bath height above the nozzles and the difference in mixing time was discussed. The tilt angle was a practical means to alter the pressure conditions in the plume of the converter. Thereafter, a coupled fluid and thermodynamic model was developed based on a local cell equilibrium concept to study the decarburization of an ascending oxygen bubble in liquid steel (Supplement II). This was done to evaluate the model in laminar flow conditions and fundamentally investigate what influence the dynamic variation of pressure had on the decarburization reaction. Further, as the model was capable of coupling fluid dynamics in ANSYS Fluent with chemical reactions in Thermo-Calc, an industrial case was investigated. The parameters from a side-blowing industrial AOD converter with high turbulence and continuous oxygen supply were employed to the model (Supplement III). The purpose was to study the temperature and the bulk concentrations of the industrial case in a coarse but practical CFD solution capable of running full duration process steps, where the kinetics are included. Finally, the importance of mixing time on the decarburization reactions in a hypothetical side-blowing metallurgical reactor was studied (Supplement IV). The aim was to keep the practicality of the model yet extend the model to include reaction-rate limitations imposed by cell-local mixing (turbulence), in order to study the importance of mixing time on the decarburization reactions.
Supplement I
Experimental study on mixing time in side-blown converters.

Supplement II
Numerical study on ascending bubble with chemical reactions.

Supplement III
Practical coupled model applied on an industrial AOD Converter.

Supplement IV
Study on the importance of mixing time in intensely stirred metallurgical reactors.

Figure 1. Thesis structure and supplement connections.
Introduction
2. Methodology

The thesis consists of experimental, numerical flow, and thermodynamic modeling. The methods and theory behind each part will be described in the following sections.

2.1 Physical model

The experimental work was carried out at the water laboratory in KTH-Royal Institute of Technology. A vessel with two mounted side nozzles was employed to study the relative effects of stirring posed by an inclined converter. Scaling between the water tank and a proposed industrial AOD converter was employed to acquire a system with reasonable gas flow rates causing intense stirring. The scale factor was decided between the average of the large and small diameter in the vessel as seen in Figure 2a) and the converter hydraulic diameter. Subsequently, the gas flow rate for one nozzle was rescaled according to equation 7[10,11] to obtain a more realistically related penetration length and gas plume to the bath volume.

\[
Q_m = \frac{Q_f}{\lambda^{(5/2)}}
\]  

(7)

Where \( Q \) is the gas flow rate with indices \( m \) and \( f \) denoting model and full scale respectively, \( \lambda \) is the geometric scale factor.

The gas flow rate was further used in equation 5 to meet the modified Froude similarity criteria and obtaining an inner nozzle diameter, see Table 1.

![Figure 2. a) Schematic illustration of vessel dimensions in the physical water model and b) real image of the water model.](image)
Table 1. Physical model parameters.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Nomenclature</th>
<th>Full scale</th>
<th>Model</th>
</tr>
</thead>
<tbody>
<tr>
<td>Scale</td>
<td>$\lambda$ [-]</td>
<td>1:1</td>
<td>1:5.3</td>
</tr>
<tr>
<td>Tilting angles</td>
<td>$\theta$ [°]</td>
<td>-</td>
<td>0, 5.5, 14</td>
</tr>
<tr>
<td>Number of side wall nozzles</td>
<td>-</td>
<td>6</td>
<td>2</td>
</tr>
<tr>
<td>Inner nozzle diameter</td>
<td>$d_n$ [m]</td>
<td>0.017</td>
<td>0.0046</td>
</tr>
<tr>
<td>Density of liquid (water/steel)</td>
<td>$\rho_l$ [kg m$^{-3}$]</td>
<td>7033</td>
<td>998</td>
</tr>
<tr>
<td>Density of gas (Process gas/Air)</td>
<td>$\rho_g$ [kg m$^{-3}$]</td>
<td>1.41</td>
<td>1.3</td>
</tr>
<tr>
<td>Gas flow rate per nozzle</td>
<td>$Q$ [Nm$^{-2}$.min$^{-1}$]</td>
<td>15.8</td>
<td>0.24</td>
</tr>
<tr>
<td>Modified Froude number</td>
<td>$Fr'$ [-]</td>
<td>1001</td>
<td>1001</td>
</tr>
</tbody>
</table>

Conductivity probes, capable of measuring both conductivity and temperature, were positioned on the lower wall across from the nozzles, and at a 90° angle from the nozzle as illustrated in Figure 3. The temperature was always monitored to ensure room temperature (25°C) and obtaining a similar kinematic viscosity of the water to molten steel. Each nozzle was connected to a separate flow meter to monitor the gas flow rate when continuous air was supplied. Three inclination angles, namely 0°, 5.5°, and 14°, were introduced using a mechanical construction that tilted in a specific direction. This resulted in a slight downward orientation of the nozzles. Both the penetration length and the mixing time were measured in all three tilting angles. However, when measuring the penetration length, only one nozzle was connected. The penetration length was captured using a high-speed camera (1010 fps) positioned perpendicular to the axial flow direction whilst the vessel was placed in a water filled box to compensate for optical distortion. The optical measurement was conducted using the ImageJ software on 60 images in a series of 3000 images (1 per 50 images). The penetration length was measured at the end of the jet along the nozzle centerline. For inclinations of 5.5 and 14 degrees, the penetration length was measured along the centerline and horizontally at the plume, which matches the centerline for an inclination angle of 0 degrees, as shown in Figure 4. Further, 60 seconds after the flow meters stabilized at the active flow rate, a 100 ml NaCl-solution (20%) was poured at the position depicted in Figure 3, and the mixing time was measured with the conductivity probes. This procedure was repeated five times with all angles and 4 gas flow rates: 60, 120, 180, and 240 NL·min$^{-1}$. The mixing time was determined from the probe that measured the longest period for the conductivity in bath to reach within ±5% of complete homogenization.
2.2 Numerical models

In the study of multiphase phenomena, two primary modeling methods are commonly employed: the Volume of Fluid (VOF) approach and the Eulerian (or Euler-Euler) approach. The VOF model solves only one set of conservation equations for all phases and tracks phases by quantifying the volume fraction of each phase within a grid cell. This approach is particularly useful when a sharp interface phenomenon is of interest. On the other hand, the Eulerian approach tracks phases by continuously monitoring their presence and properties throughout the entire computational domain. The Eulerian
Methodology

This approach focuses on solving separate conservation equations for each phase and linking the phases with interaction forces.

The VOF model solves fewer equations but is generally more computationally intensive since it resolves features of the flow that the Eulerian approach models. This is especially apparent when dealing with complex and dynamic interfaces. Since the VOF method provides a more detailed presentation of the phase interface, it was a natural choice for the bubble ascending in liquid steel. The Eulerian method can model a large number of bubbles without explicitly resolving them. It is therefore much more suitable when larger length scales are considered. It is also possible to combine the Eulerian model with a VOF approach, but this was not considered here.

2.2.1 Transport equations VOF

The governing conservation equations of mass momentum and energy for the VOF model used in a transient flow are presented below[38]:

The VOF approach tracks the interface between phases with the solution of the continuity equation (equation 8), for the volume fraction of the $q^{th}$ phase, where $\alpha_q$ is computed in each control volume based on the constraint given in equation 9.

$$\frac{\partial}{\partial t} (\alpha_q \rho_q) + \nabla \cdot (\alpha_q \rho_q \vec{v}_q) = 0$$

(8)

Where, $\rho_q$ and $\vec{v}_q$ are the density and velocity vector for phase $q$ respectively, $t$ is the time.

$$\sum_{q=1}^{n} \alpha_q = 1$$

(9)

Conservation of momentum, equation 10.

$$\frac{\partial}{\partial t} (\rho \vec{v}) + \nabla \cdot (\rho \vec{v} \vec{v}) = -\nabla p + \nabla \cdot \left[ (\mu + \mu_t) (\nabla \vec{v} + \nabla \vec{v}^T) \right] + \rho \vec{g} + \vec{f}$$

(10)

Where $\rho$ and $\mu$ are the density and the viscosity respectively. The material properties are determined by the presence of the phases in the control volume. $\vec{v}$ is the velocity vector, $p$ is the pressure, $\vec{g}$ is the gravitational acceleration and $\mu_t$ is the turbulent viscosity. The force $\vec{f}$ represents the surface tension force employing the Continuum Surface Force (CSF) expressed as equation 11[38,39].

$$\vec{f} = \sigma_{qr} \frac{\rho \kappa_{q} \nabla \alpha_q}{\sqrt{5} (\rho_q + \rho_r)} , \ n = \nabla \alpha_q , \ \kappa = \nabla \cdot \hat{n} , \ \frac{\hat{n}}{\|n\|}$$

(11)

Where, $\sigma_{qr}$ is the surface tension coefficient between two phases $q$ and $r$, $\rho$ is the volume-averaged density, $n$ is the surface normal defined as the gradient of the volume fraction of phase $q$, $\kappa$ is the surface curvature that is computed from local gradients of the surface normal and is defined in terms of the divergence of the unit normal, $\hat{n}$.

Conservation of energy, equation 12.
\[
\frac{\partial}{\partial t} (\rho E) + \nabla \cdot (\rho \mathbf{u} (\rho E + p)) = \nabla \cdot \left( (k_{\text{eff}} \nabla T) + (\overline{\tau}_{\text{eff}} \cdot \mathbf{v}) \right)
\]  \hspace{1cm} (12)

Where, \(E\) is the total energy expressed in equation 13 treated as a mass averaged variable, \(k_{\text{eff}}\) is the effective thermal conductivity, \(T\) is the temperature, and \(\overline{\tau}_{\text{eff}}\) is the shear energy.

\[
E = \frac{\sum_{q=1}^{n} \alpha_q \rho_q E_q}{\sum_{q=1}^{n} \alpha_q \rho_q}, \quad E_q = h_q - \frac{p}{\rho_q} + \frac{\nu_x^2}{2}
\]  \hspace{1cm} (13)

Where \(h_q\) is the specific enthalpy for phase \(q\).

Conservation of scalar transport in multiphase flows of scalar \(i\) in phase \(q\) is denoted by \(\Phi_{q,i}^l\), equation 14.

\[
\frac{\partial}{\partial t} (\alpha_q \rho_q \bar{\phi}_q) + \nabla \cdot (\alpha_q \rho_q \bar{\mathbf{v}}_q \Phi_{q,i}^l - \alpha_q I_{q,i}^l \nabla \Phi_{q,i}^l) = 0, \quad i = 1, \ldots, N
\]  \hspace{1cm} (14)

Where, \(I_{q,i}^l\) is the diffusion coefficient in turbulent flows and is expressed in equation 15.

\[
I_{q,i}^l = \rho_q D_{m,q} + \frac{\mu_{k,q}}{S C_t}
\]  \hspace{1cm} (15)

Where \(D_{m,q}\) is the molecular mass diffusivity and \(S C_t\) is the turbulent Schmidt number (0.7) which is formulated as \(\frac{\mu_{k}}{\rho D_t}\) where \(D_t\) is the turbulent diffusivity.

2.2.2 Transport equations Eulerian

The governing conservation equations of mass momentum and energy for the Eulerian models used in a transient flow are presented below[38]:

Conservation of mass, equation 16.

\[
\frac{\partial}{\partial t} (\alpha_q \rho_q) + \nabla \cdot (\alpha_q \rho_q \bar{\mathbf{v}}_q) = 0
\]  \hspace{1cm} (16)

Where the volume of phase \(q\), \(V_q\) is presented in equation 17.

\[
V_q = \int_V \alpha_q dV, \quad \sum_{q=1}^{n} \alpha_q = 1
\]  \hspace{1cm} (17)

In the Eulerian approach the momentum equation can be seen in equation 18.

\[
\frac{\partial}{\partial t} (\alpha_q \rho_q \bar{\mathbf{v}}_q) + \nabla \cdot (\alpha_q \rho_q \bar{\mathbf{v}}_q \bar{\mathbf{v}}_q) = -\alpha_q \bar{\mathbf{v}} p + \nabla \cdot \tau_q + \alpha_q \rho_q \bar{\mathbf{g}} + \sum_{r=1}^{n} (K_{r,q} (\bar{\mathbf{v}}_r - \bar{\mathbf{v}}_q)) + \bar{F}_{\text{vm},q}
\]  \hspace{1cm} (18)

Where, \(\tau_q\) is the \(q^{th}\) phase stress-strain tensor, \(p\) is the pressure shared by the phases, \(\bar{F}_{\text{vm},q}\) is the virtual mass force. \(K_{r,q}\), defined in equation 19 for fluid-fluid exchange, is the momentum exchange coefficient.
Methodology

\[ K_{rq} = \frac{\rho rf}{6\tau_p} d_{b,r} A_i \]  
(19)

Where, \( f \) is the drag function expressed in equation 20 according to Schiller-Naumann[40], \( \tau_p \) is the particulate relaxation time, \( d_{b,r} \) is the bubble diameter of phase \( r \), and \( A_i \) is the interfacial area.

\[ f = \frac{C_D Re}{24} \]  
(20)

Where \( C_D \) (equation 21) and \( Re \) is the drag coefficient and Reynolds number respectively.

\[ C_D = \begin{cases} 24(1 + 0.15 Re^{0.687}) & \text{if } Re \leq 1000 \\ \frac{Re}{0.44} & \text{if } Re \geq 1000 \end{cases} \]  
(21)

The energy is described by a separate enthalpy for the \( q^{th} \) phase as seen in equation 22.

\[ \frac{\partial}{\partial t}(\alpha_q \rho_q h_q) + \nabla \cdot (\alpha_q \rho_q \vec{v}_q h_q) = \alpha_q \frac{\partial p_q}{\partial t} + \nabla \cdot \left[ (k_{eff,q} \nabla T_q) - (\overline{\tau}_{eff,q} \cdot \vec{v}_q) \right] \]  
(22)

Where, \( h_q \) is the specific enthalpy of the \( q^{th} \) phase expressed in equation 23, \( k_{eff,q} \) is the effective thermal conductivity for phase \( q \), and \( \overline{\tau}_{eff,q} \) is the shear energy.

\[ h_q = e_q + \frac{p}{\rho_q} \]  
(23)

where \( e_q \) is the specific internal energy for phase \( q \).

2.2.3 Turbulence

The ascending bubble in supplement II was solved with laminar conditions while the realizable \( k - \varepsilon \)[41] was employed for the converter simulations in supplement III and IV. It has been demonstrated in the literature[26] that more sophisticated turbulence models such as the Large eddy simulation (LES) can possibly attain higher accuracy. However, due to their significant computational demands and the expected increase in computational time when dealing with chemical reactions, they are impractical for the current project within the constraints of today's computational hardware.

Equation 24 express the turbulent viscosity in relation to the kinetic energy \( (k) \) and the turbulent dissipation rate \( (\varepsilon) \). The transport equations for \( k \) and \( \varepsilon \) are presented in equation 25 and 26 respectively.

\[ \mu_{t,q} = \rho_q C_m \frac{k_q^2}{\varepsilon_q} \]  
(24)

\[ \frac{\partial (\alpha_q \rho_q k_q)}{\partial t} + \nabla \cdot (\alpha_q \rho_q \vec{v}_q k_q) = \nabla \cdot \left( \alpha_q \left( \mu_q + \frac{\mu_{t,q}}{\sigma_k} \right) \nabla k_q \right) + \alpha_q (G_{k,q} + G_b) - \alpha_q \rho_q \varepsilon_q \]  
(25)

\[ \frac{\partial (\alpha_q \rho_q \varepsilon_q)}{\partial t} + \nabla \cdot (\alpha_q \rho_q \vec{v}_q \varepsilon_q) = \nabla \cdot \left( \alpha_q \left( \mu_q + \frac{\mu_{t,q}}{\sigma_\varepsilon} \right) \nabla \varepsilon_q \right) + \alpha_q \rho_q C_\varepsilon S \varepsilon - \rho_q C_2 \frac{\varepsilon^2}{k + \sqrt{\varepsilon}} \]  
(26)
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Where, $G_k$ and $G_b$ are the generation of turbulence kinetic energy due to mean velocity gradients and buoyancy respectively, $\sigma_k$ and $\sigma_b$ are the turbulent Prandtl numbers for $k$ and $b$ respectively, and $\nu$ is the kinematic viscosity. $C_1$ and $C_2$ are constants while $C_3$ is expressed in equation 27, $C_{\varepsilon}$ is a coefficient which determines the influence of buoyancy on $\varepsilon$, and $C_\mu$ is a function of local flow parameters as seen equation 28 and 29. The default values for all constants were used.

$$C_1 = \max \left[ 0.43, \frac{\sqrt{2S_{ij}S_{ij}k/\varepsilon}}{\sqrt{2S_{ij}S_{ij}k}/\varepsilon + 5} \right]$$  \hspace{1cm} (27)

Where $S_{ij}$ is the strain rate expressed in equation 29.

$$C_\mu = \frac{1}{A_0 + A_s \frac{k^2}{\varepsilon}}, \quad U^* \equiv \sqrt{S_{ij}S_{ij} + \tilde{\Omega}_{ij}\tilde{\Omega}_{ij}}, \quad \tilde{\Omega}_{ij} = \Omega_{ij} - 2\varepsilon_{ij}\omega_k, \quad \Omega_{ij} = \tilde{\Omega}_{ij} - \varepsilon_{ij}\omega_k$$  \hspace{1cm} (28)

Where $\tilde{\Omega}_{ij}$ is the mean rate of rotation tensor in a rotating reference frame, $\omega_k$ is the angular velocity, and the model constants are $A_0 = 4.04$ and $A_s$ is given in equation 29.

$$A_s = \sqrt{6} \cos \varphi, \quad \varphi = \frac{1}{3} \arccos \sqrt{6} W, \quad W = \frac{S_{ij}S_{jk}S_{ki}}{S^3}, \quad S = \sqrt{S_{ij}S_{ij}}, \quad S_{ij} = \frac{1}{2} \left( \frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right)$$  \hspace{1cm} (29)

2.2.4 Introducing chemical reactions to CFD

The fluid mass and heat transfer in the domain was modelled with ANSYS Fluent v.2019R3-2022R2 (“Fluent” or “CFD” from now on) and the chemical reactions were calculated in Thermo-Calc v.2020b-2021b[42] (“TC” from now on). The coupling between the softwares was achieved by using the C programming language and Application Programming Interfaces (API), present in Fluent as User Defined Functions (UDF) and in TC as “TQ”. The thermodynamic database TCS Metal Oxide solutions (TCOX10)[43] in TC was employed for all models. This database is designed for slags and oxides which provides a description of solid and liquid slag, liquid steel, and gas phase. The liquid slag and liquid steel can be treated as one phase with different compositions separated by a miscibility gap. This is referred to as ionic liquids in TC. This facilitates the composition of the liquid phase to continuously change between metallic and oxidic in equilibrium. In an AOD process, the temperature of the steel typically varies between 1600-1800°C[3]. At these elevated temperatures, it is reasonable to expect rapid chemical reactions that are primarily limited by mass transfer. Given this context, the model treats the reactions by assuming local cell equilibrium. Therefore, the minimization of Gibbs free energy method was employed to compute the multi-component phase equilibrium in TC. In this work, the largest system was constrained to include only the elements C, Cr, Fe, N, Ni, and O. This restriction was implemented to minimize computational time in TC by incorporating relevant chemical reactions to the decarburization process while minimizing the number of reactions within each computational cell.

Heat and mass transfer is combined with high-temperature reactions as illustrated in Figure 5. The process assumes a one-way coupling, meaning that the reactions do not affect the flow pattern. This enables an incompressible solver and a frozen flow field approximation where User Defined Scalars (UDS) are used to transport each element. The phase composition, temperature, and pressure within
Methodology

each cell (that participates in a reaction) is passed from Fluent to TC at the end of each timestep. This is a non-equilibrium state for all phase-fractions. The enthalpy is calculated separately for each phase in TC before a final calculation is performed with all phases and the total enthalpy as the equilibrium conditions. The new phase composition and temperature are then updated in the CFD solver as the new equilibrium state. This allows local cell changes in the phase compositions and temperature at the end of each timestep which are transported in and out of the reaction zone at the following timestep.

In this way, the cell values are updated directly at the end of each timestep with no reaction rate in the cell. The reaction rate is introduced (Supplement IV) through turbulence. This reaction rate sets a limit on how much of the cell mass can reach equilibrium during a timestep by applying the Eddie Dissipation Concept (EDC)[38,44]. The concept is rooted in the transfer of energy from larger eddies to the smallest eddies, often referred to as fine structures, within turbulent flow. The primary generation of turbulent kinetic energy arises from the interactions between the larger eddies and the mean flow. In this process, mechanical energy is transferred from the eddy structures to the fine structures. The majority of the kinetic energy dissipation into heat occurs within these fine structures due to the work done by molecular forces on the turbulent eddies. The EDC model assumes that the reaction of species takes place within these fine structures that occupies a volume fraction represented by $\xi^{**}$ in equation 30. It further assumes that the species undergo reactions in the fine structures over a timescale ($\tau^*$) as described in equation 31.

$$\xi^{**} = \left( C_e \frac{v_e}{k_2} \left( \frac{1}{\hat{q}} \right)^{\frac{1}{2}} \right)^3$$  \hspace{1cm} (30)

$$\tau^* = C_r \left( \frac{v}{\hat{q}} \right)^{\frac{1}{2}}$$  \hspace{1cm} (31)

Where, $C_e$ and $C_r$ are the volume fraction constant (2.1337) and the time scale constant (0.4082) respectively.

In this work, these variables are applied in each cell. Thereby, the volume fraction of the fine structures in each cell and the reaction time scale of species that react in the fine structures, would determine the time it takes for the fine structures in the cell to react. However, since the timescale of the fine structures may not be equal to the global timestep, a scaling is performed according to equation 32. Thus, introducing the equilibrium fraction ($\eta^{eq}$), which represents the volume fraction of a cell that reaches equilibrium during a timestep. This means that although the current work uses an equilibrium approach, it is also dependent on the timestep taken in the CFD solver, effectively introducing a reaction rate. An equilibrium fraction lower than one means that the cell did not fully reach equilibrium during a timestep, while an equilibrium fraction greater than one means that the entire cell has reached equilibrium within the timestep. An illustration of the equilibrium fraction is presented in Figure 6.

$$\eta^{eq} = \min\left( \left( \xi^{**} \cdot \frac{\tau}{\tau^*} \right), 1 \right)$$  \hspace{1cm} (32)
In the reactive multiphase flow addressed in this work there are two systems, one is the control volume in CFD and the other is the control mass in TC. The control volume approach enables the transfer of mass, heat, and work across the system boundaries, while the control mass only allows heat and work to be exchanged across the system boundaries, meaning that the total mass and atoms in the system remains constant. Consequently, a distinction exists between the two systems, as
the control volume approach maintains constant volume during both equilibrium and non-equilibrium states. This could be managed by implementing a two-way coupling, where fluid properties and mass transfer between phases become integrated into the iterative process of the CFD solver. However, it will excessively increase the computational time. Thus, with emphasis on a practical solution, this work employs a constant density approach where the density of each control volume (CFD) is constant. On the other hand, the control mass (TC) allows the change of density and phase fractions between states. Therefore, to preserve the mass of the elements between the systems, the phase composition for each phase is scaled before updating the element mass in the CFD solver. In this way, the mass of a phase can increase or decrease while density remains constant. When the scaled phase composition sum is less than one, it suggests that the density for the given phase is likely to be lower, whereas a value greater than one suggests a higher density in equilibrium state as seen in Figure 7. This reactive multiphase approach is time efficient and allows a fully incompressible solver that conserves the mass of each element.

**Figure 7.** Depicts the difference between a control volume and a control mass and illustrates the constant density concept where the total mass is conserved.

### 2.3 Numerical procedure

The numerical modeling consisted of three different parts. First a bubble ascended in liquid steel while reacting in different pressure conditions. Then a continuous gas supply was added to a coarse mixing model of an 80-ton AOD converter which considered chemical reactions. The results were then compared to measurements from industrial heats. Finally, the effect of mixing time on the decarburization was investigated with a hypothetical metallurgical reactor based on water model experiments. The numerical setup for each model is presented in Table 2. In addition, some representable starting compositions and temperatures of each model are presented in Table 4. Note that different compositions have been simulated in some of the models. The following conditions and assumptions are applied to the models:
Methodology

- Gas and liquid were treated as incompressible Newtonian fluids (all supplements).
- The physical properties were constant, Table 3 (all supplements).
- VOF model (Supplement II), Eulerian model (supplement III, IV).
- 2D axisymmetric domain (supplement II) and Full 3D domain (supplement III, IV).
- Non isothermal system (all supplements).
- Adiabatic system (supplement II, IV).
- Constant heat loss through convection, conduction, and radiation (supplement III).
- No material additions (all supplements).
- Frozen flow field in quasi steady state (supplement III, IV).
- Reactions only take place in gas-liquid multiphase cells (all supplements).
- Gas injected at 300K (supplement III) or instantaneously adopts the steel temperature (supplement II, IV).

**Table 2. Numerical models set-up.**

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Bubble</th>
<th>AOD</th>
<th>Metallurgical reactor</th>
</tr>
</thead>
<tbody>
<tr>
<td>Model</td>
<td>Volume of fluid</td>
<td>Eulerian Multiphase</td>
<td>Eulerian Multiphase</td>
</tr>
<tr>
<td>Solver type</td>
<td>Pressure based, transient</td>
<td>Pressure based, transient</td>
<td>Pressure based, transient</td>
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<tr>
<td>Formulation</td>
<td>Explicit</td>
<td>Implicit</td>
<td>Implicit</td>
</tr>
<tr>
<td>Pressure-velocity coupling scheme</td>
<td>PISO</td>
<td>Phase coupled SIMPLE</td>
<td>Phase coupled SIMPLE</td>
</tr>
<tr>
<td>Inlet</td>
<td>-</td>
<td>Mass flow rate</td>
<td>Mass source</td>
</tr>
<tr>
<td>Wall</td>
<td>Non-slip</td>
<td>Non-slip</td>
<td>Non-slip</td>
</tr>
<tr>
<td>Outlet</td>
<td>Pressure outlet</td>
<td>Degassing</td>
<td>Degassing</td>
</tr>
</tbody>
</table>

*SIMPLE = Semi-implicit method for pressure linked equations. PISO = pressure-implicit with splitting of operators.*
### Methodology

#### Table 3. Modeling constants.

<table>
<thead>
<tr>
<th>Properties</th>
<th>Symbol</th>
<th>Bubble</th>
<th>AOD</th>
<th>Metallurgical reactor</th>
</tr>
</thead>
<tbody>
<tr>
<td>Density</td>
<td>$\rho_g$ [kg·m⁻³]</td>
<td>1.3</td>
<td>1.3</td>
<td>1.3</td>
</tr>
<tr>
<td></td>
<td>$\rho_l$</td>
<td>7000</td>
<td>7000</td>
<td>7000</td>
</tr>
<tr>
<td>Dynamic viscosity</td>
<td>$\mu_g$ [kg·m⁻¹·s⁻¹]</td>
<td>$4.9\cdot10^4$[45]</td>
<td>$1.8\cdot10^5$</td>
<td>$1.8\cdot10^5$</td>
</tr>
<tr>
<td></td>
<td>$\mu_l$</td>
<td>$6.7\cdot10^3$[46]</td>
<td>$6.3\cdot10^3$[47]</td>
<td>$6.3\cdot10^3$</td>
</tr>
<tr>
<td>Mass flow rate per inlet</td>
<td>$m_g$ [kg·s⁻¹]</td>
<td>-</td>
<td>3.67</td>
<td>5.2$\cdot10^3$</td>
</tr>
<tr>
<td>Bubble diameter</td>
<td>$d_{b,a}$ [m]</td>
<td>$6\cdot10^{-2}$</td>
<td>$1\cdot10^{-2}$</td>
<td>$1\cdot10^{-2}$</td>
</tr>
<tr>
<td></td>
<td>$d_{b,l}$</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Outlet pressure</td>
<td>$p_{\text{outlet}}$ [bar]</td>
<td>1.72, 1.65, 1.01</td>
<td>1.01</td>
<td>1.01</td>
</tr>
<tr>
<td>Specific heat</td>
<td>$c_{p,g}$ [J·kg⁻¹·K⁻¹]</td>
<td>1006</td>
<td>1006</td>
<td>1006</td>
</tr>
<tr>
<td></td>
<td>$c_{p,l}$</td>
<td>800[48]</td>
<td>800</td>
<td>800</td>
</tr>
<tr>
<td>Thermal conductivity</td>
<td>$k_g$ [W·m⁻¹·K⁻¹]</td>
<td>$2.4\cdot10^{-2}$</td>
<td>$2.4\cdot10^{-2}$</td>
<td>$2.4\cdot10^{-2}$</td>
</tr>
<tr>
<td></td>
<td>$k_l$</td>
<td>64[46]</td>
<td>64</td>
<td>64</td>
</tr>
</tbody>
</table>

*Indices g and l denotes gas and liquid steel.*

#### Table 4. Initial composition and temperature.

<table>
<thead>
<tr>
<th>Model</th>
<th>Mass% C</th>
<th>Mass% Cr</th>
<th>Mass% Fe</th>
<th>Mass% N</th>
<th>Mass% Ni</th>
<th>Mass% O</th>
<th>Temperature[K]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bubble</td>
<td>Melt</td>
<td>1.5</td>
<td>26.0</td>
<td>68.0</td>
<td>0.09</td>
<td>4.50</td>
<td>1873</td>
</tr>
<tr>
<td></td>
<td>Gas</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>100</td>
</tr>
<tr>
<td>AOD</td>
<td>Melt</td>
<td>0.988</td>
<td>22.77</td>
<td>74.44</td>
<td>0.231</td>
<td>1.51</td>
<td>1713</td>
</tr>
<tr>
<td></td>
<td>Gas</td>
<td>-</td>
<td>-</td>
<td>24</td>
<td>-</td>
<td>-</td>
<td>76</td>
</tr>
<tr>
<td>Metallurgical reactor</td>
<td>Melt</td>
<td>0.1</td>
<td>-</td>
<td>99.87</td>
<td>-</td>
<td>-</td>
<td>1793</td>
</tr>
<tr>
<td></td>
<td>Gas</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>100</td>
</tr>
</tbody>
</table>
Methodology

Further assumptions regarding the process gas injection method were employed to the coupled reaction model. Throughout this work, 4 different coupled model techniques were developed with different levels of resolution to the reaction mechanism. These are referred to as “coupled methods” and differentiate in the process gas injection method, reaction zone, and the transportation of reacted gas products. It should be noted that these methods were only configurations concerning the UDS/elements that were being transported.

The common denominator between the methods were that they allowed gas, liquid slag, liquid steel, and solid slag to participate in equilibrium calculations. However, the transport of the solid slag phase was excluded from the CFD solution. Hence, it was assumed that all solid oxides formed in reactions were inert and escaped the domain (i.e., to a top slag), Figure 6. This simplification was based on the added complexity to the model which instead is considered an opportunity for future developments of the model. Figure 8 illustrates the differences between reaction methods and are described as follows:

- **Coupled method 1**: The equilibrium frequency (similar to a reaction rate) was set as the inverse of the gas hold-up of the plume (0.6 s), while the transport of mass, and energy took place at the regular CFD timestep. The process gas was distributed across all cells in the reaction zone. This assumed that process gas was spread through the plume without triggering reactions until fully populating the plume. After each equilibrium reaction, the gaseous products were assumed to escape within the gas-holdup time. This method was computationally cheap due to the reduced number of equilibrium reactions.

- **Coupled method 2**: The equilibrium frequency was set to the inverse of the CFD – timestep. However, this method investigated an extreme case where all injected process gas reacted at the injection zone. After each equilibrium reaction, the gaseous products were assumed to escape.

- **Coupled method 3**: The equilibrium frequency was set to the inverse of the CFD–timestep and the reactions were limited to the injection zone. However, the transportation of gaseous products was included in the domain and were allowed to react freely with the melt at different heights in the plume. This added complexity significantly increased the number of cells participating in equilibrium reactions at each timestep which increased the simulation time accordingly. Full equilibrium was assumed to be established in all cells where reactions took place (essentially \( \eta^{eq} = 1 \) was imposed in all cells).

- **Coupled method 4**: This method was similar to method 3. However, the reaction rate was limited by turbulence, i.e., the equilibrium fraction was calculated according to equation 32, and was therefore within a range of values in the plume depending on the local turbulence values \( 0 \leq \eta^{eq} \leq 1 \).
Figure 8. Illustrates the coupled methods and their application to the models/supplements in this work. The white dashed box represents the gas injection region, and the black dashed box represents the transportation of gaseous products. Reactions are allowed to occur in both the white and black box.

2.3.1 Mixing time

Prior to adding chemical reactions, the mixing time was measured in a frozen flow field adopted after the simulation progressed over a certain time, using a transient formulation. This period was considered to achieve a quasi-steady state where the flow was fully developed.

The mixing times in the numerical models were monitored by adding a UDS acting as tracer with the same fluid properties as the bath. A volumetric method was implemented to monitor the mixing time accordingly. Assuming that the tracer concentration within the domain follows a normal distribution, it is possible to use the standard deviation (equation 33) to determine how well-mixed the system is. Approximately 67% of the cells exhibit a homogenization degree within ± 5% when considering one standard deviation, and 95% of the cells exhibit the same homogenization degree when considering two standard deviations. Consequently, the mixing time is defined as the point at which the relative standard deviation of the tracer in the domain falls below 2.5% (second standard deviation).

\[
\sigma_{std} = \sqrt{\frac{1}{N} \sum_{i=1}^{N} \left( \frac{x_i - x_{\infty}}{x_{\infty}} \cdot 100 \right)^2}
\]  

(33)

Where, \(N\) denotes the number of cells, \(x_i\) and \(x_{\infty}\) are the tracer concentration in the cell and the mean concentration respectively.

2.3.2 Ascending Bubble (supplement II)

A gas bubble ascending in liquid steel was calculated with chemical reactions in a 2D axisymmetric domain. This simplification was made to minimize the computational time while capturing the decarburization under different pressure. The gas bubble was patched into a domain with a height of 0.3m. The boundary conditions on the domain are illustrated in Figure 9. Outlet gauge pressures were employed to capture the ferro-static pressure from different bath heights. Essentially, this represented a bubble injection at the depths of 1.3, 1.2, and 0.3m, corresponding to a ferro-static pressure of 0.89, 0.82, and 0.2 bar respectively.
Figure 9. Ascending bubble domain setup. Different outlet pressures represent certain depths in the liquid steel bath.

All CFD simulations were performed in ANSYS Fluent R3 2019 on a quadrilateral grid consisting of 12,000 cells, with an adaptive timestep of $10^{-4}$ s and a criterion to meet global courant number 0.1. The equilibrium reactions took place every $10^{th}$ timestep and the real time calculation period for the coupled model was set to 0.3 s.

2.3.3 AOD Converter (supplement III)

In intensely stirred metallurgical reactors such as the AOD converter, modelling the entrainment of a gas jet from submerged side nozzles into liquid puts a severe limitation on the overall timestep. The combination of high velocities, gas-liquid interfaces, and turbulence demands small cells in the jet region as well as small global timesteps and thus requires substantial computational power. The anticipated increase in simulation time when combining the fluid transport with chemical reactions led to the decision of utilizing coarser models for the transportation of incompressible fluid flow. This work addressed the computational limitations of side nozzle injection through a numerical trick, where the gas injection was introduced further into the vessel. The injection point represented the jet penetration length at which the gas plume is mainly controlled by the buoyancy acting on the bubbles. By adopting this approach, it became possible to capture the flow patterns and mixing phenomena within the bath resulting from the jet, all without the need to simulate the high velocity and small length scales at the nozzle outlet. With a focus on a practical approach, this was assumed adequate for the investigation of decarburization in systems of industrial significance.

The model geometry and boundary conditions of the domain are illustrated in Figure 10 including the method of injection. The continuous gas supply was injected with a mass flow rate through the 90° arc plateau at a penetration length calculated with equation 6. Figure 10 a) shows the penetration length at the midpoint of the inlet plateau. This estimation was determined from the spread of the plume when controlled by the buoyancy as depicted in Figure 10 b).
Figure 10. a) schematic illustration on model geometry and boundary conditions of the AOD simulation, and b) illustrates the estimation on inlet based on behavior of plume in water model.

All CFD simulations were performed in ANSYS Fluent R2 2022 on a butterfly grid consisting of 835 cells, with a fixed timestep of 0.1 s. The equilibrium reactions took place every 6th timestep in a frozen flow field after a 20 s flow development in transient formulation. The real time calculation period for the coupled model was set to approximately 10 minutes representing the first step of the decarburization process.

2.3.4 Side-blown Metallurgical reactor (supplement IV)

This model was based on the physical water model and adopted a numerical trick to model the effect of penetration length instead of solving the gas entrainment directly. Here, the gas was introduced through two cell zones as mass sources according to equation 34,[38], acting as the source of gas from the two nozzles in the physical model, see Figure 11. A metal reactor model was developed by utilizing the configuration of the water model and adjusting the model to replicate the physical characteristics of liquid steel. This was made to provide a solution for decarburization within a theoretical metal reactor, addressing both the energy equation and chemical reactions. Two variations of the mixing time were tested. In the first, the effective diffusion coefficient was applied to all scalar transport as specified in equation 15. In the second measurement, only a laminar diffusion coefficient was assigned to the scalar transport with a constant value of $1 \times 10^{-9}$ kg m$^{-1}$ s$^{-1}$. The latter assumed a minimal diffusion influence on the mixing process, where only laminar diffusion is in effect compared to the much more dominant turbulent diffusion. This approach allowed the investigation of how the mixing time impacts the chemical reactions while at the same time keeping the mass flow rate conditions consistent. Obviously, the mixing time could be altered by changing the gas flow rate, but since the gas is reactive it is not completely trivial to do this without influencing the decarburization process. The current approach artificially modified the mixing time in the reactor without changing other important variables.
Figure 11. a) schematic illustration on model geometry and boundary conditions of the metallurgical reactor simulation, and b) illustrates the estimation on cell zone mass source.

\[ S_m = \frac{\dot{m}}{V_c} \]  

(34)

Mass source added to injection zone in supplement IV, where \( \dot{m} \) is the mass flow rate and \( V_c \) is the cell zone volume.

All CFD simulations were performed in ANSYS Fluent R2 2022 on a butterfly grid consisting of 43956 cells, with a fixed timestep of 0.01 s during flow development and 0.1 during reactions. The equilibrium reactions took place in a frozen flow field after a 70 s flow development in transient formulation. The real time calculation period for the coupled model was set to approximately 10 minutes.
3. Results

3.1 Inclination effect on mixing time and penetration length

The average penetration length and the standard deviation measured with all three inclination angles are presented in Table 5 and Figure 12. The overall results show an insignificant increase or decrease in penetration length when tilting the vessel from 0° to 14°. However, applying a 14° inclination shows an increase in mixing time ($T_m$) from 0° at all flow rates, as seen in Table 6 with the highest mixing time increase of 39% at 60 NL·m⁻¹ and the lowest increase of 13.4% at 180 NL·m⁻¹. The average mixing time and the standard deviation for all inclination angles and flow rates are presented in Table 7 and Figure 13. As can be seen from the figures, apart from 120 NL·m⁻¹, an inclination of 5.5° does not affect the mixing time.

Table 5. Penetration length in water model with inclination angles 0°, 5.5°, and 14°.

<table>
<thead>
<tr>
<th>Penetration length [cm]</th>
<th>0°</th>
<th>5.5°</th>
<th>14°</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nozzle centerline</td>
<td>13.2 ± 2.0</td>
<td>9.2 ± 1.4</td>
<td>10.9 ± 1.6</td>
</tr>
<tr>
<td>Plume</td>
<td>13.2 ± 2.0</td>
<td>11.1 ± 1.5</td>
<td>15.1 ± 1.8</td>
</tr>
</tbody>
</table>

Figure 12. The graph illustrates the penetration length in the nozzle centerline and the horizontal plume for all angles in the water model. Error bars represent the standard deviation based on the pulsation of the jet.
Results

Table 6. Increase of mixing time in percentage between 0° and 14° in the water model.

<table>
<thead>
<tr>
<th>Flow rate [NI·m⁻¹]</th>
<th>Increase in T_mb between 0° and 14° [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>60</td>
<td>39.0</td>
</tr>
<tr>
<td>120</td>
<td>27.2</td>
</tr>
<tr>
<td>180</td>
<td>13.4</td>
</tr>
<tr>
<td>240</td>
<td>14.6</td>
</tr>
</tbody>
</table>

Table 7. The average mixing time and standard deviation for 5 set of trials at all angles.

<table>
<thead>
<tr>
<th>Angle [°]</th>
<th>T_mb 60 NI·m⁻¹</th>
<th>T_mb 120 NI·m⁻¹</th>
<th>T_mb 180 NI·m⁻¹</th>
<th>T_mb 240 NI·m⁻¹</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>23.0 ± 2.0</td>
<td>16.9 ± 1.7</td>
<td>15.7 ± 1.5</td>
<td>15.8 ± 1.0</td>
</tr>
<tr>
<td>5.5</td>
<td>24.9 ± 2.0</td>
<td>20.2 ± 0.4</td>
<td>15.9 ± 0.8</td>
<td>15.2 ± 0.7</td>
</tr>
<tr>
<td>14</td>
<td>31.9 ± 2.0</td>
<td>21.5 ± 2.3</td>
<td>17.8 ± 0.5</td>
<td>18.1 ± 1.1</td>
</tr>
</tbody>
</table>

Figure 13. Mixing time over inclination with error bars representing the standard deviation over 5 sets of experiments.

3.2 Bubble ascending in liquid steel

Figure 14 presents the bubble shape evolution and temperature elevation during ascension. The bubble evolves from a sphere to a spherical cap and breaks up to a toroid while reactions at the interface elevates the temperature from 1873 to 1890.
The carbon evolution in the oxygen bubble ascending from different bath levels over 0.3 seconds is presented in Figure 15. At 1.3 and 1.2 m depth (high pressure), the graph shows that the initial reactions between the bubble and the steel produce off gases (mostly CO) while dissolving into the steel at later stages. However, at 0.3 m depth (low pressure), the bubble remains intact with gas as a stable phase in the system. In this case, the carbon content increases drastically until 0.2 seconds where the bubble is close to fully saturated with carbon, as depicted from Figure 16. Nitrogen showed similar behavior and no oxides were formed during any of the simulations with the composition of steel presented in Table 4. However, it is illustrated in Figure 17 that when the carbon content in the steel was reduced to 0.5 mass%, chromium oxide was formed. The chromium oxide seen in the figure was inert and did not participate in the next reactions. In addition, it was transported as a liquid phase.
Figure 15. Normalized carbon mass against the initial carbon mass over 0.3 s under different pressure, in the oxygen bubble.

Figure 16. Carbon gradients in oxygen bubble during ascendance from a 0.3 m depth in steel.
Results

Figure 17. Inert chromium oxide formation during bubble ascendence.

When the gas bubble is a mixture of oxygen, nitrogen, and argon the model predicts a stable gas phase at all depths. While the nitrogen partly dissolves back into the steel, the carbon is seen to almost reach saturation in the bubble at higher depths (high pressure). However, at lower depths (low pressure), the carbon and nitrogen in the steel reacts with the gas to produce off-gases, Figure 18. The carbon does not reach a saturation level at the lower depths during the simulated times due to the increased nitrogen solubility. It is seen that decarburization is most effective at the depths closer to the bath surface where the ferro-static pressure is low.
3.3 Decarburization in the AOD

The mixing time is around 10 seconds in the model and was calculated with finer grids where an increase of approximately 80% was found between the finest mesh of 5280 cells and the coarsest mesh of 835 cells. This was shown in supplement III to have minor effects on the reactions hence, the coarse mesh was employed for the simulations. The flow behavior of the liquid steel in the AOD converter is presented in Figure 19 where a large circular motion governed by the gas plume is depicted by the arrows, as well as the corresponding local temperature distribution.
**Results**

**Figure 19.** The flow pattern of the liquid steel (direction of arrows) is displayed to the left with the reaction zone/gas plume as the transparent orange volume. The temperature contours on the right are illustrated after a blowing period of 580s (835 cells).

**Figure 20-21** show the mass percentage of elements in the liquid steel throughout the decarburization step of two industrial AOD heats including model predictions of this work, and the data acquired from the steel plants online process model (OPM). The two heats vary in initial composition, temperature, and total mass. **Table 8** presents the mass distribution of oxygen to each element through their respective reactions in both the industrial heat and the developed model. Similarity is found between the model and the two industrial heats. Temperature, chromium- and nickel content all show a high agreement to the measured heat data, while carbon and nitrogen show a difference. From the difference in mass of oxygen distributed to CO, the results indicate an excess decarburization due to the absence of Si and Mn in the thermodynamic treatment.

**Table 8.** Distribution of oxygen mass for oxidation of different elements [kg].

<table>
<thead>
<tr>
<th>Oxygen to:</th>
<th>[CO]</th>
<th>(Cr₂O₃)</th>
<th>(MnO)</th>
<th>(SiO₂)</th>
<th>[O]</th>
<th>Injected oxygen</th>
<th>Used oxygen</th>
<th>O→[CO]</th>
<th>O→(MnO)⁺</th>
<th>O→(SiO₂)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Heat 1: I</td>
<td>723</td>
<td>650</td>
<td>104</td>
<td>38</td>
<td>-</td>
<td>1578</td>
<td>1515</td>
<td>-202</td>
<td>142</td>
<td></td>
</tr>
<tr>
<td>Heat 1: S</td>
<td>925</td>
<td>617</td>
<td>-</td>
<td>-</td>
<td>31</td>
<td>1578</td>
<td>1573</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Heat 2: I</td>
<td>1239</td>
<td>500</td>
<td>82</td>
<td>230</td>
<td>-</td>
<td>2056</td>
<td>2051</td>
<td>-282</td>
<td>312</td>
<td></td>
</tr>
<tr>
<td>Heat 2: S</td>
<td>1520</td>
<td>500</td>
<td>-</td>
<td>-</td>
<td>31</td>
<td>2056</td>
<td>2051</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

S = Simulation, I = Industrial heat. (g) = gaseous, (s) = solid oxide, [] = dissolved in liquid steel.

The temperature in **Figure 20 a)** is elevated rather linearly with a slightly higher rate at the beginning (0-200 s) and the end (400-600 s) with a lower rate of increase in between. The decarburization is
overpredicted in the model as the rate of decarburization in Figure 20 b) is higher at the beginning of the blowing period compared to the OPM. The Chromium oxidation in Figure 20 c) shows a high rate of oxidation (0-200 s) and is followed by a lower rate in the middle (200-400 s), and a resurgence (400-600) to match the measured industrial value. In Figure 20 d) the nitrogen content is seen to shortly increase (0-20 s) followed by a decrease until it increases again at approximately 300 s to an overpredicted value at the end. The nickel content increases similar to the industrial data due to the decrease in the other elements, Figure 20 e). The OPM also show good predictions compared to the industrial measurements.
Figure 20. Show the evolution of a) temperature, b) mass% carbon, c) mass% chromium, d) mass% Nitrogen, and e) mass% nickel over the blowing period of 10 minutes for heat 1.
Results

Similar to the previous heat, the model predicts temperature, chromium, and nickel very well while overpredicting the decarburization rate and the nitrogen content in the melt. The temperature evolution in heat 2 can be seen in Figure 21 a), showing a clear correlation to the carbon and chromium oxidation. The initial carbon content in this heat (1.33 mass%) is higher than in heat 1 (0.988 mass %). Initially a decrease of carbon content is seen in Figure 21 b) while chromium oxidation is very low, Figure 21 c). The carbon content is reduced at a linear rate until approximately 400 seconds where the decarburization rate starts to level out. At this point, the chromium oxidation rate is excessively increasing, reducing the chromium content very close to measured data. The OPM predicts a similar chromium evolution through the blowing time. However, even though the decarburization rate in the model follows the trend of the OPM well, it is still over predicted, resulting in a lower carbon content than measured data and OPM. The nitrogen is rapidly decreased in the first 100 seconds until it starts to dissolve back into the melt (200-800 s) and ending in an over predicted value compared to the measured data.
Figure 21. Show the evolution of a) temperature, b) mass% carbon, c) mass% chromium, d) mass% nitrogen, and e) mass% nickel over the blowing period of 10 minutes for heat 2.
3.4 Side-blown Metallurgical reactor

3.4.1 Mixing time and flow

The mixing time was measured for tracers injected in 9 different positions as seen in Figure 22. The average mixing time and the interval of these tracers are compared to the water model in Figure 23. The results indicate that injection points located in the region with high convective force shows a faster mixing. The injection points 1, 4, 5, 8, and 9 are in the trajectory of the strong circular current posed by the gas plume as depicted in Figure 22. The mixing time was calculated with finer grids where the maximum grid convergence value of 13% determined the mesh reasonable for the coupled model (supplement IV).

![Diagram](image)

**Figure 22.** Positions of tracer injections in the domain. Note that point 1 is the tracer injection point in the water model presented in supplement I. The circular flow field and velocity magnitude of the water caused by the gas plume is illustrated in iso-planes. The flow direction is illustrated by the vectors and streamlines.
The influence of the diffusion coefficient on mixing time in the metal reactor is presented in Table 9. The position of tracer injection did not only result in a variation of mixing time but also on the diffusive and convective contribution to the mixing. This can be seen with points 5 and 9, where the mixing time increased approximately 20 s (270%) compared to the maximum increase of 170 s (1000%), when only using laminar diffusion coefficient.

Table 9. Mixing time in the metal reactor with effective diffusion coefficient and (only) laminar diffusion coefficient.

<table>
<thead>
<tr>
<th>Tracer</th>
<th>Effective diffusion [s]</th>
<th>Laminar diffusion [s]</th>
<th>$\Delta_{mix}$ [s]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>9</td>
<td>123</td>
<td>114</td>
</tr>
<tr>
<td>2</td>
<td>14.4</td>
<td>175</td>
<td>160</td>
</tr>
<tr>
<td>3</td>
<td>14.4</td>
<td>180</td>
<td>165</td>
</tr>
<tr>
<td>4</td>
<td>8.3</td>
<td>104</td>
<td>96</td>
</tr>
<tr>
<td>5</td>
<td>10.4</td>
<td>29</td>
<td>19</td>
</tr>
<tr>
<td>6</td>
<td>15.9</td>
<td>186</td>
<td>170</td>
</tr>
<tr>
<td>7</td>
<td>15.9</td>
<td>185</td>
<td>169</td>
</tr>
<tr>
<td>8</td>
<td>10.2</td>
<td>70</td>
<td>60</td>
</tr>
<tr>
<td>9</td>
<td>9.6</td>
<td>32</td>
<td>22</td>
</tr>
<tr>
<td>Average</td>
<td>12 ± 3</td>
<td>120 ± 61</td>
<td></td>
</tr>
</tbody>
</table>
3.4.2 Coupled methods

A comparison between the different coupled methods 1-4 used in the metal reactor is presented in Figure 24, with the bulk mass of all elements and the temperature in the domain. The coupled methods will further be referred to as “CM1-4” with “a, b, and c” for depicting the different cases where parameters such as diffusion coefficient and initial composition vary, Table 10. As an example, coupled method 1, case a will be named “CM1a”.

Table 10. Parameters used for cases a, b, and c in conducted simulations.

<table>
<thead>
<tr>
<th>Case</th>
<th>W(C)</th>
<th>W(Fe)</th>
<th>W(O)</th>
<th>Temperature [K]</th>
<th>Diffusion coefficient [kg·m⁻¹·s⁻¹]</th>
</tr>
</thead>
<tbody>
<tr>
<td>a</td>
<td>0.001</td>
<td>0.9987</td>
<td>0.0003</td>
<td>1793</td>
<td>Effective</td>
</tr>
<tr>
<td>b</td>
<td>0.001</td>
<td>0.9987</td>
<td>0.0003</td>
<td>1793</td>
<td>Laminar</td>
</tr>
<tr>
<td>c</td>
<td>0.001</td>
<td>0.9989</td>
<td>0.0001</td>
<td>1793</td>
<td>Effective</td>
</tr>
</tbody>
</table>

Figure 24 a) show that the initial decarburization rate is highest in CM1a (oxygen distributed in the entire gas plume) and lowest in CM2a (reactions restricted to jet zone). A slight difference in the decarburization rate is seen between CM1a and CM3,4a (120-450 s) where the decarburization rate is initially similar and starts to decrease in CM3,4a approximately 120 seconds into the blowing process. CM3a and CM4a behave similarly with the difference being the turbulence limitation of reactions. This indicates a small difference between perfect and limited mixing in the cell for this system and composition range. The iron and oxygen content in the melt are presented in Figure 24 b) and c) where the decrease in iron means forming solid oxides, while an increase in oxygen represents either liquid oxides or dissolved oxygen. Thus, CM2a shows an earlier solid and liquid oxide formation compared to the other cases. However, CM1a initially oxidizes iron at a higher rate and reaches similar iron content at the end of the simulation. CM4a exhibits the lowest iron oxide formation and oxygen content in the melt, which is reflected in the temperature graph, Figure 24 d).
Figure 24. The graphs compare the element and temperature evolution in the domain for all coupled models CM1-4a.

3.4.3 Mixing time effect on coupled methods

The mixing time increase with the absent turbulent diffusion does not affect the decarburization as seen in Figure 25 a). The bulk carbon content in the domain shows no significant difference between CM1,2,4a (effective diffusion applied) and CM1,2,4b (only laminar diffusion applied), indicating a process limited mainly by thermodynamics and gas injection rate. Local variations between the two cases can be seen in Figure 25 b) and Figure 26. Even though transport of the reacted melt is faster in CM4a than in CM4b and the maximum/minimum values of carbon in CM4b is higher/lower, the bulk carbon content is not affected as much over the entire blowing time.
Results

**Figure 25.** a) The carbon mass in the domain over time employing effective diffusion in CM-1a,2a,4a and laminar diffusion in CM-1a,2a,4a. b) Illustrates the maximum, average, and minimum carbon content in the melt over 20s for CM4a and CM4b.

**Figure 26.** Illustrates the local variation of carbon mass fraction between CM4a and CM4b during the initial 7 seconds of the simulation.
3.4.4 Effect of Initial oxygen content

The initial oxygen content in the metal reactor melt is seen to affect the decarburization rate in CM1, 3, and 4, where Figure 27 reveals an increased decarburization rate for the first seconds with higher initial oxygen level in the melt. However, this trend was not found in CM2, Fig 27 b) and a slightly higher difference between the cases is found in CM3 and CM4 than in CM1. The cases a and c (lower initial oxygen content) tend towards the same decarburization rate at the end as the initial effects wear off.

Figure 27. The graphs compare the mass of carbon in the domain over time in CM1-4 between case a and c (lower initial oxygen content in the melt).
Results
#### 4. Discussion

A physical and numerical model was developed to investigate the effect of mixing time and pressure on the decarburization reactions in intensively stirred side-blowed reactors such as the AOD converter. The numerical model coupled fluid dynamics with high temperature chemical reactions and has been tested in different phenomena related to the AOD process throughout this work. With the aid of these models, this section will discuss the results presented in the previous section.

##### 4.1 Simulation time

Table 11 displays the overall simulation time for each case when incorporating reactions. The selection of computing core configurations for each case was based on the necessity to balance multiple simulations while optimizing computational resources. Given the computational intensity of these simulations, the allocation of available cores across various cases allowed for conducting more tests within a reasonable timeframe. Among the metal reactor cases considered, CM1, CM3, and CM4 calculates equilibrium for an equal number of cells. However, there are distinct variations in their reaction mechanisms. Notably, CM1 initiates reactions only every 6 timesteps, whereas both CM3 and CM4 trigger reactions at each timestep. As a result, one can expect an increase in simulation time for CM3 and 4 when restricted to 8 or 1 computing cores. While CM2, similar to CM3 and 4, initiates reactions at every timestep, its simulation time remains relatively shorter. This difference can be attributed to the reduced number of cells participating in reactions during each individual timestep. It is evident that as the core count increases, the simulation time decreases, with the primary constraint being the number of cells involved in chemical reactions.

**Table 11. Comparison between simulation time and computing cores for the different cases.**

<table>
<thead>
<tr>
<th>Case</th>
<th>Blowing time [s]</th>
<th>Simulation time [h]</th>
<th>Nr of cells in reactions zone</th>
<th>Reaction frequency [s]</th>
<th>Cores</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bubble CM3</td>
<td>0.3</td>
<td>7</td>
<td>~100</td>
<td>0.001</td>
<td>1</td>
</tr>
<tr>
<td>AOD CM1</td>
<td>600</td>
<td>10</td>
<td>~100</td>
<td>0.6</td>
<td>1</td>
</tr>
<tr>
<td>Metal reactor CM1</td>
<td>600</td>
<td>22</td>
<td>~1600</td>
<td>0.6</td>
<td>8</td>
</tr>
<tr>
<td>Metal reactor CM2</td>
<td>600</td>
<td>6</td>
<td>~60</td>
<td>0.1</td>
<td>8</td>
</tr>
<tr>
<td>Metal reactor CM3</td>
<td>600</td>
<td>20</td>
<td>~1600</td>
<td>0.1</td>
<td>16</td>
</tr>
<tr>
<td>Metal reactor CM4</td>
<td>600</td>
<td>20</td>
<td>~1600</td>
<td>0.1</td>
<td>16</td>
</tr>
</tbody>
</table>

##### 4.2 Physical water model and penetration length

In this study the hydraulic diameter was calculated to solve the complexity of the geometric similarity due to the presence of the refractory blocks in the AOD converter. With this diameter, a scale factor between the physical model and the converter was used to find a reasonable gas flow rate and a nozzle diameter that satisfies the modified Froude number. This resulted in nozzle dimensions that did not satisfy geometric similarity. The mixing time is believed to be dependent on the characteristics of the forming gas stream hence, it was important to satisfy the dynamic similarity at
Discussion

the cost of the geometric similarity for the nozzle. Nonetheless, the physical model only utilizes two nozzles and can therefore never fully represent the mixing time of the real AOD converter with, say, six nozzles. However, it does provide the possibility to investigate the effects an inclination would have on mixing time in a side-blowing metallurgical reactor, and validation points to numerical models.

The inclination did not show any clear effect on the penetration length according to the results. This supports earlier work[49] stating that a variation in bath height does not affect the jet penetration length. Conversely, more recent work[13] has found contradictory results with a quantitative investigation. The work measured the penetration length of 3000 images leading to a better representation of the mean and standard deviation between the angles applied. When the inclination angle is large, the bubbles move in the direction of that angle due to kinetic energy, and when this energy is exhausted, they start to rise due to buoyancy. Larger angle values lead to bubbles rising higher and being more spread out. Even in current study the horizontal component of the penetration depth for 14° increased about 5.6% further than an inclination of 5.5°. This was when comparing the minimum penetration depth of 14° to the maximum penetration depth of 5.5°.

4.3 Mixing time and its effect on decarburization.

According to the results of the water model, large inclination angles (14°) increased the mixing time while an inclination angle of 5.5° did not appear to have any effect. The flow behavior observed in the model was similar to that of other studies[20-22,24,25] where the ascending bubbles drag the water, due to buoyancy, along the nozzle wall to the surface creating a hump at the interface. Subsequently the water moves to the opposite wall with a wave-like motion at the surface and follows the wall down to the bottom and then back to the nozzles. This forms a large circular motion in the bath which also was observed in the numerical model. As the inclination angle increased, the wave like motion at the surface was observed to intensify by the larger vessel oscillations. Inclination angles to submerged side-blowing have previously been demonstrated to result in more pronounced oscillations due to the phase boundary movement[21]. The turbulent surface waves and the oscillatory motion of the upper portion of the bath may lead to dissipative interactions with the bubble plume and the fluid flow. Although these oscillations are believed to affect the stirring and the mixing time, the effect cannot be estimated from this or previous studies. However, the bath height to diameter ratio has been shown to affect the mixing time[20,22] where an optimal ratio was found at 0.7-0.75. The change of bath height and/or geometry does affect the flow structure in the vessel[21] which is eminent when tilting the vessel. For instance, the bath height above the nozzle is decreased and the rising plume is pushed toward the nozzle wall, Figure 28. Without a comprehensive and quantitative study of the transient flow-field, it becomes challenging to attribute any main specific phenomenon to the mixing time.
Figure 28. Qualitative images illustrating the movement of the rising plume towards the nozzle wall from a) 0° inclination angle to b) 5.5°, and c) 14°.

In the numerical models, several mixing times were measured at different times where the flow field was frozen. The flow field reached a quasi-steady state and did not change after a certain time at the current mesh resolutions. In the industrial process, the mixing phenomena is very complex and difficult to measure, hence, unknown. However, for a down-scaled water model of an AOD converter with similar configuration as the numerical AOD model used in this study, a variation of parameters resulted in a mixing time interval of 20-60 s with impulse tracer injection, and 11-24 s with continuous tracer injection[22]. Also, Visuri et al.[50] found mixing times ranging from 7-26 s with pulse tracer injection when varying parameters such as tracer injection position and gas flow rates. The work estimated the mixing time to range between 9-12 s in the decarburization stage of the AOD converter. Further, Haas et al.[18] found mixing times between 11-17 s and Wei et al.[17] found a range of 33-47 s. Other simulations of the full scale-system[25] reported a mixing time range from 100-116 s. With the coarse CFD models used in this work, the mixing time was slightly over-predicted. In addition, the degassing surface did not consider the wave-like motion of the free surface, which impacts the flow in the upper section of the bath. The absence of this free surface movement, along with the coarse mesh, likely contributes to the emergence of a quasi-steady state flow field in the simulation. As a result, the physical model does not replicate the same quasi-steady flow field observed in the numerical model. To achieve a closer resemblance to the physical model, it would be necessary to incorporate a free surface, employ a finer mesh, and possibly utilize a more advanced turbulence model like Large Eddy Simulation (LES). Implementing these changes would significantly extend the simulation time, making it unfeasible to complete this work, given that the primary aim of this study was to develop a practically usable model.

It was found that the location of the tracer injection point was important e.g., injecting the tracer close to the inlet (point 9 in Figure 22) would result in a faster mixing compared to injections closer to the walls such as point 2 or 3. In intense side-blown converters, where a significant portion of the bath is influenced by circular motion and strong currents, it is generally anticipated that the convective contribution to mixing will be substantial. In addition, the diffusion contribution to the mixing would have a smaller effect in these regions. Although this was not studied in the physical water model within this work, the position of tracer injection has been shown to affect the mixing time in previous work[50]. Thus, it is suggested that the injection of tracer when measuring mixing
Discussion

time is located appropriately to the application of the process being modeled. As an example, in the decarburization step of an AOD converter, the reaction zone (nozzle inlet or plume) would give a representative mixing time to correlate with the decarburization. However, the mixing time for material additions of any kind or the reduction step would be better represented by injecting at the bath surface. This means that there may be several mixing times in a metallurgical reactor, the two main ones being the reaction zone mixing time and the other being the materials addition mixing time. It is clear that this behavior is mainly due to the convective flow pattern where an injected volume can shear in various directions and follow different currents depending on the injection location.

In the coupled models, the mixing time was artificially increased by removing the turbulent diffusion to observe its effect on the reactions without changing the gas flow rate in the system. This did not affect the reactions, as observed in the result section. The mixing time increases by more than 270% at the reaction zone which corresponds to about 20 seconds. Even though local variations were observed over 20 seconds, these time frames only correspond up to 4% of the total blowing time and may be the reason for its low impact on the end results. Thus, the current system is governed by thermodynamics and the gas supply rather than the mixing in the process. It has been suggested in earlier studies[27-30], that at the early stages of decarburization, the system is governed by the supply of oxygen. Although, it is possible that this increase of mixing time might affect the later stages of the AOD process or a have a larger impact on less intensely stirred system such as the ladle. While an increase in mixing time may initially seem detrimental to the process, it's important to note that this doesn't necessarily guarantee a negative impact on the overall process. Therefore, solely examining mixing time in investigations might overlook the bigger picture, as it cannot be definitively stated that a shorter mixing time always benefits the process.

4.4 Pressure and its effect on decarburization

Regardless of pressure, it appears that initially high carbon content in the melt will drive the system to primarily decarburize the steel rather than oxidizing chromium. This was found in both the bubble simulations (supplement II) and in the AOD simulations (supplement III) which covers the whole pressure range of the plume. This behavior has also been observed in previous models[6,7] and it is consistent with thermodynamics. At ferro-static pressures imposed by the melt close to the nozzles, a small pressure decrease by approximately 0.067 bar, did not affect the decarburization rate by much, as seen from the bubble simulations (Figure 18). This decrease corresponds to a 10 cm elevation change which is also approximately one layer of refractory bricks. However, it was found that a pressure decrease of 0.6 bar (1 m elevation change) had a large effect on the decarburization with a considerable higher carbon removal to the bubble closer to the bath surface, as seen in Figure 18. When comparing these pressures to the metal reactor model (supplement IV), the injection region is at a ferro-static pressure of approximately 0.3 bar. In the CM2a case in the metal reactor, the reactions only occur in the injection region. However, the pressure in the injection region is low enough to admit a reasonable decarburization rate. Solid oxides are formed at a higher rate compared to the other metal reactor cases where reactions occur across the entire plume. If the ferro-static pressure would be even higher (e.g., a head of 1.3 m), then the decarburization would be negatively affected. CM1a showed the highest decarburization rate due to the injected oxygen being evenly distributed across the reaction zone, which permits a larger amount of oxygen to react at lower partial pressure. CM3a and CM4a both allow the reacting products at the jet region to be transported to the top and react at lower partial pressure. However, at this stage gas also consists of CO hence does not decarburize as effectively as CM1a. The decarburization rate decreases at the end

46
of the blowing process which is commonly described as a decrease in carbon removal efficiency[7,28]. In industrial practice, this is usually a sign to change the ratio of the gas mixture to reduce the partial pressure and prevent oxidation of chromium. As this step is not applied in this study, oxygen is utilized in the formation of Iron oxide. However, there is still carbon oxidation near the bath surface as visualized in Figure 29 which distinct CM1, 3, and 4 against CM2. This also explains the difference in lowest carbon content between CM2 and the other cases.

![Figure 29](image)

**Figure 29.** iso planes visualize a snapshot of the carbon content at the end of the simulation in CM1-4 and distinctively show the lack of decarburization close to the bath in CM2. Note, the gradient in the upper iso-planes in CM2 is the effect of the flow spreading the reacted melt.

An initial offset in time of the carbon curve with lower initial oxygen content in the metal reactor was seen in Figure 27 for CM1c, 3c, and 4c. Initially, more of the injected oxygen was dissolved in the c cases before decarburization reached the same rate between the a and c cases. In CM2, all injected oxygen only reacts in the injection zone which is enough oxygen to dissolve into the melt and to decarburize the steel at the similar rate, unaffected by the initial oxygen content. In the other cases the gas reacts with the steel over the entire plume where the equilibrium is pressure dependent. The amount of oxygen that dissolves into the steel varies depending on the pressure. The smaller offset difference between CM1 and CM3,4 is due to the oxygen being distributed over the entire plume in CM1 whereas it needs to be transported in CM3 and 4.
Overall, CM1 did present a good agreement with industrial heats (industrial AOD case, supplement III). As shown in the results, the discrepancy in carbon between the model and the industrial heat is likely attributed to the lack of Silicon and Manganese treatment in the model. These elements were shown to also oxidize in the industrial AOD and were not accounted for in the simulations (not present in the thermodynamic system). The overprediction of nitrogen could possibly be an effect of distributing the gas evenly across the reaction zone instead of transporting the gas with reaction limitations. The effect of escaping the domain after reaction prevents the gas bubbles from reacting during ascendance. In the bubble simulation it was seen that nitrogen almost completely dissolved into the steel under high pressure while producing nitrogen gas at lower pressures Figure 18 b). It is therefore natural to test CM3 and CM4 in the AOD system. The decarburization rate in CM4 was slightly lower than in CM3, while the formation of iron oxide occurred earlier and at a higher rate in CM3. This was anticipated as the only difference between the two methods was the turbulence limitation in CM4. Naturally, CM4 is not as effective as CM3. In addition, it can be seen in Figure 30 that the equilibrium fraction at the lower part of the plume has a region lower than one. This means that the reactions taking place at highest ferro-static pressure do not have time to fully reach equilibrium and explains the less effective iron oxidation in CM4 compared to CM3, where the equilibrium fraction is set to unity. In the metal reactor, the difference between CM1 and CM3, 4 was not large, indicating that the CM1 simplification could save computational time. However, the pressure in the reaction zone has clearly shown to affect the reactions, especially the decarburization. Thus, when introducing CM3 or CM4 to the AOD system, the pressure at the reaction zone will be closer to 2 bars rather than 1.3 bars as in the metal reactor. This might result in a larger difference between CM1 and CM3, 4. It is therefore important to include solid slag as part of the CFD solution. In the event of chromium oxidation at the jet region, the transportation of these oxides will allow reduction to CO closer to the bath surface.

Figure 30. Iso planes visualize the region where the reactions are limited by turbulence with the equilibrium fraction in CM4. The transparent black volume represents the plume/reaction zone. Note, all equilibrium fraction values above 1 are displayed as 1.
In the AOD process, tilting the converter or relocating nozzles vertically is possible to accomplish although to a limited extent as it imposes practical challenges. As seen in Figure 28, tilting the converter this way would increase the wear of the refractory by the shear forces of the plume being pushed towards the wall. Especially for an inclination of 14° or above. It would create an engineering challenge to redistribute refractory blocks and the pressure difference at the nozzle region might not be large enough. Hence, the efficiency in decarburization rate would be neglectable. Further, even though the water model does not fully represent an AOD converter, it showed that the mixing time in a side-blown converter increases with higher inclination angles. Therefore, should the mixing time affect the decarburization rate at later stages of the process, it would be for the worse. It is therefore not suggested to tilt the converter during the decarburization stages, unless this is done for other purposes. In addition, elevating the nozzles 10 cm would have an insignificant effect on the decarburization rate and is therefore not suggested. Substantially larger decreases in ferro-static pressure are needed in order to improve the thermodynamic conditions during decarburization. More flexible systems might be able to incorporate this, such as utilizing a height adjusted submerged top lance during the decarburization stages.
Discussion
5. Conclusion

Throughout this thesis, the focus has been to develop a general numerical model that integrates CFD with high-temperature chemical reactions. The primary objective was to understand the interplay between pressure, mixing efficiency, and decarburization processes within intensively stirred side-blown converters. Through a series of designed experiments and simulations, the study investigated various parts of this complex system. It began by examining mixing time alterations with tilt angles in a physical water model. Subsequently, a fluid and thermodynamic model based on local cell equilibrium explored the decarburization dynamics within laminar flow conditions. Expanding its scope to practical applications, the model was tested against an industrial case involving an AOD converter, where turbulence and continuous oxygen supply played pivotal roles. Finally, the research explored the implications of mixing time on decarburization reactions within a hypothetical metallurgical reactor. This approach not only contributes valuable insights to the field but also highlights the adaptability and robustness of the developed numerical model in addressing the complexities of real-world industrial processes. This model holds great promise for advancing our knowledge and optimizing operations in metallurgical engineering.

Overall, it can be concluded that intensely stirred metallurgical reactors are more affected by the thermodynamics rather than the mixing time. Further, the pressure only impacts the decarburization when the variation in pressure is large. The specific conclusions from this work can be summarized as follows:

- When applying an inclination, the shape of the bath volume changes, the bath height to diameter ratio decreases as the bath height above the nozzle is decreased, and the wave like motion at the surface intensifies. This interferes with the flow pattern and results in an increase in mixing time at higher inclination angles. Tilting the vessel by 14° increased the mixing time by approximately 14% at high gas flow rates and 39% at low gas flow rates. An increase of 5.5° did, however, not show any effect on the mixing time.

- The placement of the tracer injection holds significance in the calculation of mixing time. Injection points situated within the ascending plume or areas influenced by vigorous currents consistently demonstrate accelerated tracer mixing. Given that reactions predominantly transpire at the gas plume interface, it is advisable to employ tracer injection within this region for all systems where reactions originate from this interaction. This approach ensures a representative measurement of mixing time.

- Mixing time modifications by eliminating the turbulent mass diffusion component had an insignificant impact on the carbon and iron compositions throughout the entire process. Despite a substantial increase in mixing time, approximately 270%, it became evident that the primary determinants in this system are thermodynamics and gas injection rates. Investigations solely centered on mixing time may overlook the mark, as it cannot be definitively concluded that a shorter mixing time consistently benefits the reactive process.

- Rapid decarburization reactions saturate a gas bubble with carbon. Saturation time depends on both pressure and composition of the gas. In the case of a 60 mm oxygen bubble injected at the surface under laminar conditions, carbon saturation is achieved in 0.2–0.25 seconds, whereas a bubble containing oxygen, nitrogen, and argon necessitates more than 0.3 seconds. Although it is not established what bubble sizes dominate in metallurgical reactors,
Conclusion

some authors report the range 5-70 mm, which means that 60 mm bubbles in laminar flow likely estimates a longer than average saturation time.

- The influence of ferro-static pressure on the reactions became increasingly pronounced with greater differences in injection depth. When a bubble is injected closer to the bath surface, it saturates at elevated carbon levels due to the reduced partial pressure of gas elements. Conversely, under higher pressure conditions representing deeper injection depths, without any inert gas, the bubble dissolves entirely into the steel.

- Industrial data has been employed to validate the AOD simulations. The results from the simulations and the industrial data exhibit strong agreement in temperature, chromium, and nickel. However, a discrepancy is observed in carbon content, which can be attributed to the absence of Si and Mn in the thermodynamic description. In the early stages of the decarburization process, element oxidation is primarily limited by oxygen supply rather than mass transfer. Regardless of pressure, steel grades with lower carbon content initially exhibit high rates of oxidation for both carbon and chromium, while higher carbon content leads to initial decarburization before chromium oxidation occurs, much in accordance with thermodynamics.

- Analysis of different coupled methods with various assumptions, showed that the highest initial decarburization rate was found in a case where oxygen was distributed across the gas plume, which fostered a lower equilibrium pressure for a major part of the reactions. The least favorable decarburization rate and the highest iron losses were exhibited in the case where all reactions were assumed to occur solely in the injection zone. A turbulence limiter for the reaction rate was also introduced which influenced decarburization, solid oxide formation and temperature evolution.
6. **Sustainability and recommendations for future work**

6.1 **Future work**

This thesis has presented a model that demonstrates the feasibility of both detailed and practical yet fundamental approaches to modeling complex metallurgical reactors. The model achieves this by coupling Thermodynamic software with CFD. However, to model the full AOD process or other metallurgical processes, further developments need to be addressed. The model’s future holds promising opportunities in the following development areas:

- Including the transport of the solid slag phase and the reactions at the metal-slag interface to attain a deeper complexity to the model and provide the possibility for more isolated and detailed simulations. This will also require description of systems with more elements leading to higher computational demands. Since the current model can utilize parallel decomposition of the domain, this increase in resolution is possible.
- Including material additions (e.g., alloying and/or scrap addition) is also vital to model the full AOD process for obvious reasons.
- Including the free surface and applying a finer grid would better represent the dynamics and add higher accuracy to the stirring. This could possibly give more insight at the cost of increased simulation time. Considering the success of one-cell approaches (process models) with kinetic coefficients trimmed over many industrial heats, the two previously mentioned development areas are likely more important. The uncertainty in real process conditions puts a limit on the usefulness of developing high accuracy models even if simulation time is not an issue.

6.2 **Sustainability**

This work has successfully developed a general model capable of predicting complex metallurgical reactors. The model is useful in investigating fundamentals as well as practical questions related to actual plant operation. Metallurgical processes are expensive and time-consuming. Therefore, pilot experiments and experiments in large scale productions will be very costly and seen as too risky for smaller companies looking to develop their processes. By using fluid dynamic/thermodynamic modeling, more information can be obtained, providing better security while eliminating risks, which can give smaller companies a chance to compete. Moreover, the utilization of computer simulations diminishes the need for industrial trial and error experiments, leading to decreased economic and environmental consequences. As such the use of this model could contribute in an integrated way to the following UN Sustainable Development Goals:

- UN Sustainable Development Goal 9: Industry, Innovation, and Infrastructure by promoting sustainable industrial innovation and infrastructure.
- UN Sustainable Development Goal 12: Responsible Consumption and Production by reducing material usage.
- UN Sustainable Development Goal 13: Climate Action by reducing greenhouse gas emissions through modeling that contributes to fewer industrial trials.
7. References


References

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Inclination Effect on Mixing Time in a Gas–Stirred Side–Blown Converter

Serg Chanouian,* Björn Ahlin, Anders Tilliander, and Mikael Ersson

Small-scale physical models are commonly used to investigate gas-stirred processes in steelmaking practice. The argon oxygen decarburization (AOD) converter is among various processes widely used in the metallurgy field and utilizes side blowing of oxygen and inert gas for mixing in the bath. Herein, the effect of the converter inclination on mixing time and jet-penetration length with a side-blown physical model is investigated. Scaling with the modified Froude number is applied on data from a real industrial AOD converter to achieve a system with reasonable gas flow rates. During the experiments, water is used to simulate liquid steel and air is blown through side-mounted nozzles for stirring. A NaCl tracer is added and subsequent conductivity measurements are used to measure mixing time. Overall, the penetration length is shown to be independent of inclination angle. The mixing time is found to be influenced by the change of the height of diameter ratio, change of geometry in the bath volume, gas flow rate, and the intensified wave motion at the interface caused by the inclination of the vessel. The mixing time increase with 14% when 14° angle is applied.

1. Introduction

In steelmaking practice, gas stirring is a common way to homogenize liquid steel processes ranging from various types of converters to ladle stirring. Due to the extreme environment in the process, involving high temperature and turbulent flows, small-scale physical models are commonly used to investigate the processes, see Table 1. Indeed, these models provide the tools for understanding underlying phenomena in steel productions. More specifically, water models are widely used to model liquid steel, which provides safer, more cost efficient, and simpler experiments. However, due to the underlying complexity of the real systems scaling can be difficult and acquires attention to the similarity criterions such as geometric and dynamic similarities.[1]

One of the various metallurgical gas stirring processes is the argon oxygen decarburization (AOD) converter. The process is widely used and accounts for the majority of the world’s stainless-steel production.[2] During the AOD process, oxygen and inert gases are injected through side-mounted nozzles to decarburize the molten steel. However, other precious metals, such as chromium, are oxidized while decarburizing. Therefore, varying ratios of gases are injected in different steps to minimize the chromium oxidation while maximizing the decarburization. In Equation (1), the relationship between carbon and chromium oxidation is described.

\[ 3\{C\} + (Cr_2O_3) \leftrightarrow 2\{Cr\} + 3\{CO\} \]  

where \( \{ \) is dissolved in liquid steel, (\) is gas phase, and (\) is slag phase.

Throughout the process, several chemical reactions occur. It is well established that the kinetics of these reactions are influenced by the mixing of the bath. Consequently, fluid flow transport in gas stirring processes such as the AOD converter are of utmost importance in the steel industry and has been studied by physical models, see Table 1.

The behavior of horizontal gas injection into a liquid using a submerged nozzle was investigated by Hoefele and Brimacombe.[3] The study consisted of laboratory work using different combinations of gas/liquid to characterize flow and jet behavior. The gas penetration length at the nozzle centerline was measured and found to depend on the modified Froude number and the gas–liquid density ratio according to Equation (2).

\[ L = 10.7F_r^{0.44}d_n\left(\frac{\rho_g}{\rho_l}\right)^{0.35} \]  

where \( d_n \) is the nozzle diameter, \( \rho_g \) is the density of gas, \( \rho_l \) is the density of liquid, and \( F_r \) is the modified Froude number given by Equation (3).

\[ F_r' = \frac{F_r}{\rho_l\rho_g} \]  

where \( Q \) is the volumetric gas flow rate and \( g \) is the gravitational acceleration.
Flow patterns and jet submersion of a side-blowing converter was studied by Fabritius et al.\textsuperscript{[10]} with the help of a down-scaled physical model using the modified Froude’s number and the specific mixing energy. The specific mixing energy is described by the specific kinetic energy and the specific buoyancy energy. Among the conclusions, they found that the horizontal jet penetrated further into the vessel when injecting air compared with only using oxygen gas. As a result, the rising gas plume was transferred further into the vessel creating a different flow pattern which was more suitable for the mixing giving a 27% shorter mixing time in the physical model. Further, a physical model was used by Bjurström et al.\textsuperscript{[11]} to study the penetration length and the fluid flow structure in a side-blowing water model. The focus was to study the effect of gas flow rate and bath heights in different combinations. Accordingly, the main findings were that the penetration length was dependent mostly on the gas flow rate. Moreover, the fluid flow structure was dependent on both the flow rate and the bath height.

With help of a physical model, the authors Fruehan and Kim\textsuperscript{[12]} studied the effect of operating variables such as position and number of nozzles on mass transfer between metal and slag using water and oil. The work measures mixing time on different nozzle positions with and without the presence of oil. It was found that the presence of oil increased the mixing time significantly due to resistance against the circularly velocity of the fluid created by the plume near the surface. Further, the most effective mixing time was achieved with a side-blowing nozzle compared with bottom blowing.

Wei et al.\textsuperscript{[6-8]} thoroughly investigated the fluid mixing characteristics of an AOD converter, with several combination of different parameters used in practice with the help of a down-scaled physical model. The work considered the effect on fluid flow characteristics caused by different combinations of nozzle number, angular positions, gas flow rates, side blowing, and side blowing combined with top blowing. The authors claim to have reached very high similarities between the model and the industrial converter, especially regarding the kinematic similarity which in previous work have been sufficient, although not necessarily accurate according to the authors. In addition, the authors conclude that all the parameters investigated influences the mixing time and each combination has one setup that is most efficient for mixing. However, applying top blowing does have a detrimental effect on the mixing. On most cases, top blowing showed an increase in mixing time. Also, Haas et al.\textsuperscrap{[9]} studied the effect of top blowing on the mixing in the AOD converter with a physical water model including oil on the surface acting as slag. The results agreed with the work of Wei where top blowing increased the mixing time both with and without a slag layer.

Further, Ternstedt et al.\textsuperscrap{[10]} studied the effect of vessel diameter, bath height and gas flow rate on mixing time in a side-blowing converter using water models. The results showed that the mixing time was almost independent of bath height. However, it was stated that the mixing time decreased with an increased gas flow rate and that an increase in vessel diameter increased the mixing time. Other papers\textsuperscrap{[11-13]} found that an increase in the bath height affected the mixing time up until a certain point. For fill levels from 75 to 100% of the original bath height an increase in the mixing time was found. For bath heights from and above 100%, the mixing time remains constant. Furthermore, Jiang et al.\textsuperscrap{[14]} studied the effect of the bath height on the mixing time in a horizontal cylindrical water model with side- and bottom-blown air. In contrary to previous studies\textsuperscrap{[11-13]} they found that the mixing time is decreased with an increase in the bath height, regardless to what gas flow rate is used. The authors explain the results mathematically by calculating stirring energy from rising gas bubbles.

Samuelsson et al.\textsuperscrap{[15]} studied the feasibility to decrease the decarburization rate in an AOD converter by increasing the fill
volume with the help of geometry change of the vessel. The authors carried out mixing time experiment on two water models with circular and oblong cross section, respectively, to increase volume without tampering with bath heights. The results concluded that the difference in mixing time between the two geometries were insignificant, meaning that an increase in volume without changing bath height by applying a geometry change could increase productivity in the industrial AOD. This is because more steel could be produced per charge under the same amount of time.

A side-blowing water model scaled from a newly developed pilot plant reactor was developed by Bölke et al.\textsuperscript{14} to conduct experiments that investigate both the mixing time and the penetration length. The work carried out conductivity measurement to determine the mixing time behavior for different gas flow rates as well as adding more nozzles. In addition, the gas penetration length was measured for each case to investigate its dependence on gas flow rate. The work concluded that the mixing time decreased with an increased gas flow rate similar to other studies. However, when using three nozzles compared with a single nozzle the mixing time significantly increased. This was explained by an apparent calmer and steadier bath surface due to the use of three nozzles. Also, two of the nozzles were faced toward each other working as counterparts that disturb the circular flow movement. The gas penetration length was seen to increase as the flow rate increased. It is important to note that the effect of pulsations was considered when measuring the penetration length.

In sum, there are various studies on mixing in gas stirred physical models and the majority are down-scaled from an industrial vessel. Additional studies regarding the effect of gas stirring have been carried out.\textsuperscript{17,20} Worth noticing, the studies by Odenthal and coworkers\textsuperscript{11,17} concluded that a mixture of gas with a larger portion of inert gas increase the oscillations in the system, a slight inclination on the vessel toward the nozzles increases converter vibrations and that a high fill-level results in a slight drop in forces and torques.

Even though various factors affecting stirring conditions in gas-blowed converters have been studied there are still aspects that need to be considered. One aspect is the effect on mixing time due to practical changes such as an inclination of the converter or vertical relocation of the side nozzles. Such changes affect the ferrostatic pressure in the rising gas plume. By tilting the converter, it is possible to affect the distance from the nozzles to the free surface. Similarly, a vertical relocation of the side nozzles will also affect the distance to the free surface. To the authors’ knowledge, no studies have been reported on the mixing time when applying an inclination (i.e., tilting) to a side-blowed converter. The hypothesis is that there is no total thermodynamic equilibrium in an AOD converter. Instead, there are local equilibriums where reactions take place. Therefore, an inclination of the converter, which lowers the bath height above the nozzles, would in turn lower the ferrostatic pressure around the nozzles. From a thermodynamic perspective this could influence the reaction presented in Equation (1) by reducing the partial pressure of CO. Thus, favoring carbon oxidation over chromium oxidation during decarburization in an AOD converter. Moreover, as stirring in the vessel is affected by an inclination, it is important to investigate how an inclination might affect the stirring. More specifically, it is of importance to investigate if there are any drawbacks to the mixing of the bath and to what extent these drawbacks outweigh the possible benefit on the chemical reactions.

Therefore, in this work, the aim is to investigate gas penetration length and mixing time as well as study the circulatory flow characteristics when tilting a down-scaled water model of an AOD converter. This work is also carried out with the purpose of obtaining validation points to develop a numerical model, which later can be used to investigate the effect on chemical reactions.

2. Experimental Section

2.1. Physical Model and Scaling

The scaling method and construction of the water model was used to acquire a system with reasonable gas flow rates similar to the industrial AOD converter. Thus, it was only possible to investigate relative effects such as the effect on stirring posed by an inclined converter. Therefore, the results could only indicate the behavior of the flow and should not be perceived as a representation of the industrial converter.

A previously downscaled AOD-converter\textsuperscript{15} was used for the experimental procedure. However, some modification to the dimensions of the vessel was necessary to match a 70-ton industrial AOD-converter with six submerged side nozzles. Linear scaling was carried out between the industrial converter and the water tank with a scale factor of 1:5.33 with reference to the vessel diameter. The tank was manufactured in Plexiglas with the capacity for up to six nozzle inlets, whereas the nozzles were 3D printed in plastic to match the specified dimensions shown in Figure 1. For this work, only two nozzles were used.

Due to the presence of refractory blocks in the industrial converter and the decrease in diameter at the lower part of the vessel, achieving geometrical similarity was more complex considering asymmetry in the real system. The approach used to acquire a scale factor was to calculate a hydraulic diameter for the different

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{figure1.png}
\caption{Schematic figure of physical model downscaled from a 70-ton AOD-converter.}
\end{figure}
courses of refractory blocks in the industrial converter and calculate an average diameter, see Equation (4) and (5) and Figure 2.

\[ D_H = \frac{A}{P} \]  

(4)

where \( A \) is the area and \( P \) is the perimeter.

\[ \frac{D_{H1} + D_{H2} + \ldots + D_{Hn}}{n} = D_{Hc} \]  

(5)

where \( D_{Hc} \) is the average hydraulic diameter of the converter and \( n \) is the number of courses.

The ratio between \( D_{Hc} \) and the model diameter decides the scale factor, \( \lambda \).

Despite the number of nozzles used for this case the gas flow rate was rescaled and calculated for 1 out of six nozzles using Equation (6) to obtain a more realistic penetration length and gas plume in relation to the bath volume^{[19]}

\[ Q_m = \frac{Q_f}{\lambda^2} \]  

(6)

where \( Q_m \), with \( m \) and \( f \), as indexes for “model” and “full-scale,” respectively, is the gas flow rate and \( \lambda \) is the scale factor.

Further, the calculated gas flow rate was used in Froude’s modified number, Equation (3) to reach the Froude similarity criteria as well as acquiring a reasonable nozzle diameter. The parameters and units of the physical model and the converter are shown in Table 2.

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Symbol</th>
<th>Unit</th>
<th>Converter</th>
<th>Model</th>
</tr>
</thead>
<tbody>
<tr>
<td>Scale</td>
<td>( \lambda )</td>
<td>[-]</td>
<td>1:1</td>
<td>1:5.33</td>
</tr>
<tr>
<td>Tilting angle</td>
<td>( \alpha )</td>
<td>[°]</td>
<td>-</td>
<td>0, 5.5, 14</td>
</tr>
<tr>
<td>Number of side wall nozzles</td>
<td>( n )</td>
<td>[-]</td>
<td>6</td>
<td>2</td>
</tr>
<tr>
<td>Inner nozzle diameter</td>
<td>( d_n )</td>
<td>[m]</td>
<td>0.017</td>
<td>0.0046</td>
</tr>
<tr>
<td>Density of melt/water</td>
<td>( \rho_1 )</td>
<td>[kg m(^{-3})]</td>
<td>7033</td>
<td>998</td>
</tr>
<tr>
<td>Density of process gas/Air</td>
<td>( \rho_g )</td>
<td>[kg m(^{-3})]</td>
<td>1.41</td>
<td>1.293</td>
</tr>
<tr>
<td>Gas flow rate (total)</td>
<td>( Q_{\text{tot}} )</td>
<td>[Nm(^{-3}) min(^{-1})]</td>
<td>95</td>
<td>1.44</td>
</tr>
<tr>
<td>Gas flow rate per nozzle</td>
<td>( Q )</td>
<td>[Nm(^{-3}) min(^{-1})]</td>
<td>13.83</td>
<td>0.24</td>
</tr>
<tr>
<td>Modified Froude number</td>
<td>( Fr' )</td>
<td>[-]</td>
<td>1001</td>
<td>1001</td>
</tr>
<tr>
<td>Kinematic viscosity</td>
<td>( \nu )</td>
<td>[m(^2) s(^{-1})]</td>
<td>9.3 \times 10(^{-7})</td>
<td>1.1 \times 10(^{-6})</td>
</tr>
</tbody>
</table>

Figure 2. (Left) Illustration of refractory blocks at lower part of a converter. (Right) The top view of one course.

2.2. Experimental Procedure

The gas penetration length was measured using one nozzle while the mixing time was measured using two. For mixing, the symmetry of an even number of nozzles was deemed important. However, for penetration length measurements, it was believed to have little effect.

Water, with a temperature ranging from 23 to 25 °C was continuously measured by a combined temperature and conductivity probe. Water used to simulate molten steel due to their similar kinematic viscosities and air was injected through the two nozzles to simulate process gas. The conductivity probes were placed 1 [cm] above the bottom. The first probe (P9) was placed at the wall opposite to the nozzles and the second probe (P10) at the wall between the nozzle and the first probe, as shown in Figure 3. Each nozzle was connected to a separate flowmeter (Bronkhorst EL–FLOW SELECT F–203AV) which controlled

Figure 3. Schematic illustration of equipment set up and experimental procedure.
the gas flow rate to 240 Nl min\(^{-1}\) through the software FlowDDE provided by Bronkhorst. The air was supplied to the flowmeters through a pressure regulator, see schematic in Figure 3.

Furthermore, a mechanical construction was built with the possibility to tilt the converter in three different angles, 0, 5.5, and 14°, as shown in Figure 4.

The penetration length was filmed with a high-speed camera (Motion Blitz Cube 4, 1010 fps) placed orthogonally to the axial flow direction and measured optically with the software ImageJ. The vessel was placed in a Plexiglass box which was filled with water to compensate for optical distortion when measuring the penetration length. In addition, for higher accuracy, only one nozzle was connected to inject air. Oscillating pressures due to the large density ratio between the gas and liquid transmits and affects the upstream jet causing pulsations.\(^{[20]}\) Therefore, a series of 3000 pictures was captured where only 60 were measured, meaning, one per 50 pictures. The penetration length was determined to be at the end of the jet on the nozzle centerline which corresponds to 0 volume fraction of air.\(^{[21,22]}\) It is worth noticing that for the 5.5 and 14° inclinations, the penetration length was measured along the nozzle centerline and horizontally at the plume corresponding to the centerline for the 0° inclination, see Figure 5. Further, the penetration length was also calculated with Equation (2). As the penetration length is measured with 2D images of a 3D phenomenon, it is possible that the jet dispersion away from the nozzle centerline is not captured.

The mixing time was studied by adding a 100 [ml] tracer in form of a 20% NaCl-solution (75 g NaCl in 300 mL water) and measuring the electrical conductivity of the bath with the two conductivity probes. The solution was poured manually at the tracer point shown in Figure 3. The conductivity probes feed the data to a conductivity meter in one second span for a total interval of 120 s in each trial. In addition, the flow was fully developed before adding the tracer solution. This was considered to be achieved 60 s after the flowmeters had stabilized.

The mixing time was measured five times for three different tilt angles, 0, 5.5, and 14°, respectively. It was determined as the time for the tracer concentration to reach within ±5% deviation of its final homogenized concentration in the bath. Figure 6 shows an example of one mixing time graph. The mixing time was determined as the largest time given by any probe for each trial. Further, the same method for mixing time measurement was applied for a gas flow rate of 180, 120, and 60 Nl min\(^{-1}\).
3. Results

Due to the pulsation at the nozzle, the gas penetration length fluctuates and therefore, a series of pictures were measured, as previously stated. An irregular periodic pulsation is discovered, where both high- and low-frequency pulsations are found over the course of 3 s. The average penetration length and standard deviation are shown in Table 3 and Figure 7, including the theoretical value from Equation (2). Even though the penetration length along the nozzle centerline indicates a slight decrease for 5.5° compared with 0° inclination, the overall results show no significant increase or decrease in penetration length when tilting the vessel.

The mixing time for all three angles and four flow rates are shown in Table 4 and 5 and Figure 8–11 with the corresponding standard deviation. As shown in the figures, apart from the 120 Ni min⁻¹ case, the results indicate that an inclination of 5.5° does not affect the mixing time in the bath, whereas an inclination of 14° shows an increase in mixing time for all flow rates investigated.

4. Discussion

4.1. Scaling

In an industrial converter, there is an unavoidable presence of refractory blocks that complicates the geometric similarities. Therefore, in this study, a hydraulic diameter was calculated to partially solve the complexity of geometric similarity and obtain a scale factor between the two systems. With this scale factor, a custom bath height that corresponds to the industrial bath height was chosen. Further, the nozzle diameter for the water model was too large according to the scale factor. However, as the mixing time is more dependent on the characteristics of the forming gas stream, it is more important to obtain kinematic similarity rather than geometric similarity for the

<table>
<thead>
<tr>
<th>Angle [°]</th>
<th>Tₘ [s] 60 [Ni min⁻¹]</th>
<th>Tₘ [s] 120 [Ni min⁻¹]</th>
<th>Tₘ [s] 180 [Ni min⁻¹]</th>
<th>Tₘ [s] 240 [Ni min⁻¹]</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>23.0 ± 2.0</td>
<td>16.9 ± 1.7</td>
<td>15.7 ± 1.5</td>
<td>15.8 ± 1.0</td>
</tr>
<tr>
<td>5.5</td>
<td>24.9 ± 2.0</td>
<td>20.2 ± 0.4</td>
<td>15.9 ± 0.8</td>
<td>15.2 ± 0.7</td>
</tr>
<tr>
<td>14</td>
<td>31.9 ± 2.0</td>
<td>21.5 ± 2.3</td>
<td>17.8 ± 0.5</td>
<td>18.1 ± 1.1</td>
</tr>
</tbody>
</table>

Table 4. Mixing time (Tₘ) for five set of trials in each angle and gas flow rate including their averaged value and standard deviation.

<table>
<thead>
<tr>
<th>Flow rate [Ni min⁻¹]</th>
<th>Increase in Tₘ [%] between 0° and 14°</th>
</tr>
</thead>
<tbody>
<tr>
<td>60</td>
<td>39.0</td>
</tr>
<tr>
<td>120</td>
<td>27.2</td>
</tr>
<tr>
<td>180</td>
<td>13.4</td>
</tr>
<tr>
<td>240</td>
<td>14.6</td>
</tr>
</tbody>
</table>

Table 5. The increase in Tₘ between 0° and 14° inclination angle presented in percentage.

Figure 7. Penetration length for all angles including centerline and plume measurements.

Figure 8. Mixing time versus inclination angle 60 Ni min⁻¹.
Figure 9. Mixing time versus inclination angle 120 Nl min$^{-1}$.

Figure 10. Mixing time versus inclination angle 180 Nl min$^{-1}$.

Figure 11. Mixing time versus inclination angle 240 Nl min$^{-1}$.

similarity such as thermal and chemical are not possible to acquire for reasons such as no chemical reactions occur in the physical model and therefore no temperature differences due to chemical reactions. In this sense, phenomena related to temperature such as gas expansion are not present in the physical model. With this in mind, it is also important to notice that the physical model is only using two nozzles and can, therefore, never fully represent the mixing time of the real converter. However, this is not a problem as the study focuses on the behavior of mixing as a result of applying an inclination in the AOD component. Furthermore, another purpose is to have validation points for numerical models. This will later be used to model a full-scale AOD converter with high-temperature chemical reactions.

4.2. Penetration Length and Mixing Time

The obtained penetration length is in accordance with the theoretical penetration length proposed by Hoenle and Brimacombe.[3] In addition, regardless of the inclinations applied to the vessel, the results show no clear indication of increase or decrease in penetration length. This supports the finding of earlier studies[6] where the bath height does not affect the penetration length. It is arguable that the difference in hydrostatic pressure could change the physical properties such as density for the fluids which in turn can change the penetration length.[3] However, the difference in height at the end of the jet for 0° and 14° pose a negligible change in hydrostatic pressure for this physical model and can therefore be ignored. The only apparent difference is if the penetration length is measured in the horizontal direction. For a 14° inclination, the horizontal component of the penetration reaches $\approx 5.6\%$ further in compared with the penetration length at a 5.5° inclination. This is only when comparing the shortest penetration for the 14° case with the longest penetration for the 5.5° case.

As for mixing in the bath, the results point to an increase in mixing time when a high enough inclination such as 14° is applied to the vessel. In contrast, an inclination of 5.5° does not appear to affect the mixing time. This behavior is not isolated by one causal factor but rather dependent on several factors which are discussed in the following paragraphs.

Similar flow behavior, as discovered in earlier studies,[11,17] were also detected in this study. The gas bubbles are forced up to the surface by buoyancy, which in turn cause the water motion. This flow pattern spreads across the bath to the opposite wall and then follows the wall down, and bottom horizontally back to the nozzle hence creating a circular motion. The rising plume creates a bulge at the interface that take part in the violent and intense wave motion at the surface called sloshing. The same pattern is detected for all three angles. However, as the inclination increased the sloshing motion appears more intense and violent and the vessel oscillates to the degree of being visible to the naked eye. This pattern agree well with the work of Odenthal et al.[11] and Wuppermann et al.[3] where it was determined that the oscillations are mainly caused by the phase-boundary surface movement. However, in their work, it was believed that the increased turbulence caused by the vibrations might increase the stirring leading to a lower mixing time. No vibrations were measured in this work, but as earlier mentioned, it was clearly a
more violent motion as the inclination increased. It appears that the mixing time cannot be estimated from how violent the stirring seems.

From earlier studies, it is clear that the bath height-to-diameter ratio is important to mixing in the bath where an increase or decrease in bath height can either be to an advantage or disadvantage. In addition, a change in bath height and/or geometry gives different flow structures. In Figure 12, static flow patterns for different geometries and bath heights are shown schematically. However, these figures are only an indication of the flow structure since the flow structures are transient for these kinds of models. For the same reason, this also concerns the volumetric shape of the water in the vessel. When tilting the vessel, the water inside the tank changes its volumetric shape and therefore affects the circular motion inside which affects the mixing. For instance, when tilting the vessel the hump height created by the rising plume is moved toward the nozzle wall. Consequently, the smaller swirl between the plume and wall found in the 0° case is completely eliminated by a 14° inclination. In addition, when the vessel is tilted the bath height over the nozzles is decreased. This results in a decreased input of buoyancy energy into the system, which in turn affects the mixing energy as buoyancy is the main driving force of the accelerated ascending bubbles. Thus, having a lower effect on the mixing of liquid compared with a non-/small vessel inclination. It should be clarified that an increase in bath height does not always result in better mixing due to buoyancy energy input increase. As seen in Wuppermann et al., the optimal bath height-to-diameter ratio was found to be 0.7–0.75. Therefore, it is important to include the bath height-to-diameter ratio as well as the drag and the buoyancy effects.

In Figure 13, qualitative images of sloshing behavior are displayed. The violent waves at the surface appear to affect the water and bubble entrainment. The fluctuating motion in the upper half of the bath may cause dissipative interaction with the bubble plume. This would be another factor explaining the increase in mixing time when applying an inclination as the sloshing motion appeared more intense with 14° inclination. Without a 3D image of the transient flow-field, it is difficult to attribute the mixing time to any particular phenomenon. Numerical models would be suited for this investigation.

The effect of buoyance on mixing time when tilting the vessel should also be studied for inclination toward the nozzles. Furthermore, adding a symbolic slag layer with higher viscosity such as oil to dampen the intense surface motion is recommended for further studies to investigate the sloshing effect on mixing time.

5. Conclusions

A physical model study has been conducted to investigate the effect of an inclination on the gas penetration length and mixing behavior in a side-blown converter. From the results, it is clear that an inclination affects mixing time. The study show an increase in mixing time when applying higher inclination angles and indicates that several factors need to be considered when dealing with the complexity of mixing time based on the following conclusions. 1) The penetration length is not affected by an inclination of the vessel. 2) An active circularly motion is found in both the inclined and noninclined vessel during the gas blowing process. The shape of the bath volume change when an inclination is applied which interferes with the flow pattern hence affecting the stirring of the bath. 3) When applying an inclination, the bath height-to-diameter ratio over the nozzles changes where the bath height is decreased, thereby resulting in a lower

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**Figure 12.** Schematic drawings on fundamental static flow structures in different height-to-diameter ratios as well as different geometries of water in vessels.
buoyancy input energy and an increase in mixing time. 4) The mixing time increases by ≈14% with an increased inclination angle from 0 to 14° for high gas flow rates. For lower gas flow rates, a 14° inclination has a larger effect on the mixing time where an increase up to 39% is shown. However, from 0 to 5.5°, there is no significant change in mixing time.

If an inclination were to be implemented in an industrial setting on an AOD converter, it is recommended to tilt ≈5.5°. This is due to the fact that there is no significant change in mixing time and the stress from vibrations appeared lower than for a 14° inclination. However, while a 5.5° inclination might be too small to change the thermodynamic conditions; a 14° inclination, in contrast, might be too stressful for the AOD construction. The next step is to apply computational fluid dynamics coupled with high temperature reactions to investigate the possible tradeoff between mixing and thermodynamics for different systems. This would then provide an opportunity to investigate how decarburization is affected by a decrease in bath height due to an inclination or vertical relocation of the side nozzles.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

Research data are not shared.

Keywords

argon oxygen decarburization converter, inclinations, mixing time, penetration lengths, physical modeling

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A Fundamental Investigation of Decarburization Reactions in the Argon–Oxygen Decarburization Converter Using Coupled Computational Fluid Dynamics and Thermodynamics Databases

Serg Chanouian,* Björn Ahlin, Anders Tillander, and Mikael Ersson

Metallurgical converters such as the argon–oxygen decarburization (AOD) converter generally utilize gas blowing for the mixing and refinement of liquid steel. Due to the harsh environment of the complex and opaque system, it is common practice to study the stirring of the process through physical and numerical models. Effective mixing in the bath has an important role in refinement such as decarburization and has been vividly studied before. However, high-temperature chemical reactions that also play a major role are sparsely investigated. With the help of modeling, a computational fluid dynamics model coupled with chemical reactions is developed, allowing the study of both dynamic fluid transport and chemical reactions. Herein, the chemical reactions for a single gas bubble in the AOD are investigated. The study shows that a 60 mm oxygen gas bubble rapidly reacts with the melt and is saturated with carbon in 0.2–0.25 s at low-pressure levels. The saturation time is affected by the pressure and the composition of the injected gas bubble. The impact of ferrostatic pressure on the reactions is more significant at larger depth differences.

1. Introduction

Decarburization is a common step for carbon reduction in the steel-making practice. Decarburization involves oxygen injection to reduce the carbon content in the steel melt and is crucial for secondary steelmaking processes such as the argon–oxygen decarburization (AOD) process. In general, decarburization is similar to other secondary steelmaking processes such as the basic oxygen furnace (BOF). However, in the AOD process, it is also a requirement to maintain the stainless properties acquired by elements such as chromium. Hence, the difficulty is to oxidize the excess carbon without oxidation of chromium described by the main reactions in the AOD:[1]

\[ 2[C] + \{O_2\} = 2(CO) \]  

(1)

\[ 4(Cr^3+) + 3\{O_2\} = 2(\text{Cr}_2\text{O}_3) \]  

(2)

Combined to

\[ 3[C] + \{\text{Cr}_2\text{O}_3\} = 2(Cr) + 3(CO) \]  

(3)

where \( \{\) is the gaseous form, \( [\) is dissolved in liquid, and \( \) is in the slag.

A lot of work has been dedicated to understanding the complexity of the AOD and BOF systems and as gas stirring is involved, numerical modeling has been a popular choice.[2] With numerical modeling, it is possible to lower the design cost and add transparency to the highly complex metallurgical systems, which otherwise cannot be observed or easily measured. Although there exists an extensive amount of work, this work mostly covers the fluid dynamics of the process ranging from sonic jet top lance to more abstract areas such as mixing and homogenization. Therefore, further understanding and development of the complex processes require investigations into different aspects of the thermodynamic parts such as the high-temperature decarburization reactions. One of which is the decarburization zone.

Fruehan[3] studied the reaction sequence and mechanisms for oxidation of chromium and carbon and found that chromium oxidation is considerably faster than carbon oxidation. The study states the possibility that the main reaction at the nozzle region is the oxidation of chromium at the bubble interface. Further, as the bubble rises in the bath, \( \text{Cr}_2\text{O}_3 \) is reduced by carbon. In this way, there are local reactions taking place at one part of the process which further up in the bath reacts with other products due to species transport and implies that total equilibrium exists in the AOD converter. The combined reaction above states that a lower partial pressure of CO benefits decarburization. The partial pressure of CO is partly governed by the ferrostatic pressure,[4] which implies that decarburization is more effective at the surface due to the lower ferrostatic pressure. On the contrary, another hypothesis is that local equilibrium exists in the AOD instead of total equilibrium. For this hypothesis, most of the
Decarburization occurs at the nozzle region which is affected by the ferrostatic pressure imposed by the bath height above the nozzle. Thus, decreasing the height between the nozzle and the bath surface could introduce more efficient decarburization.

Asai and Szekely presented an early mathematical model to describe temperature and composition trajectories for decarburization. The model is based on component balances using driving-force expressions, equilibrium relationships, and heat balance. A new mathematical model for the AOD process was developed by Wei and Zhu et al. However, this model treats all possible oxidation and reduction reactions simultaneously and reaches a combined equilibrium in competition at the liquid/bubble interfaces. With the help of Gibbs free energies of oxidation reactions, the model characterizes the competitive oxidation among the elements and determines the corresponding oxygen distribution. The model also considers the difference in the existing regimes of rate control for decarburization at high and low carbon levels by the assumption that the oxidation rate of elements at high carbon levels is mostly related to the supply of oxygen, while the mass transfer of the liquid bulk to the reaction interface determines the decarburization rate for lower carbon levels. In addition, the model assumes that the nonreacting oxygen blown into the bath escapes at the liquid interface rather than accumulating as a dissolved phase in the steel bath.

Järvinen et al. developed a reaction submodel for a computational fluid dynamics (CFD) solver describing chemical reactions as well as heat and mass transfer taking place on the surface of a gas bubble. The method assumed a fast/infinite kinetic rate, with mass transfer as the dominating mechanism for solving the rates in a controlled component reaction system. The rate expression was based on the modified law of mass action, thus making it possible to simplify and couple it with the conservation equations. The results suggested that at high carbon concentrations, most of the oxygen gas reacts with carbon to form CO and that equilibrium is reached after 0.06 s at the surface interface. The model was further developed to predict local variations in conditions such as total pressure, void fraction, bubble size, etc. This model was also used in the unified interaction parameter (UIP) model to acquire accurate information on the thermodynamic behavior.

A further extension to the reaction model in other studies was developed by Visuri et al. to include the reduction of top slag during the AOD reduction stage. The dominating reactions were assumed at the surface of the emulsifying slag droplets using thermodynamic submodels considering the activities of metal and slag. By including mass transfer reversible reactions and various additions to mass and heat balance, the model was able to predict the reduction stage in the AOD reasonably well.

Another method for existing kinetic process model predictions is the effective equilibrium reaction zone (EERZ) coupled with thermodynamic software such as FactSage or Thermo-Calc. With the help of thermodynamic databases, the local equilibrium calculation approach is applicable and the change of the chemical composition of bulk is controlled by interface reactions and the mass transfer coefficient. EERZ treats the reaction volume between two species as several finite number sections, first, calculating equilibrium at the interface which is followed by the homogenization reaction in the other finite sections.

Despite the earlier achievements in the cost-efficient satisfactory modeling of the AOD process, they rely on stagnant mathematical descriptions of the reaction surface interface. For this study, a more dynamical approach is required; hence, the coupling method between CFD and thermodynamic modeling developed is more suited. Ersson et al. developed a fundamental dynamic model of decarburization in a top-blown converter. The model integrated fluid dynamics with thermodynamic databases allowing thermodynamic reactions and continuous decarburization of molten steel surface over time. This was accomplished by achieving local cell equilibrium with the present chemical species. Andersson et al. developed a fluid flow and reaction model for the decarburization stage of the AOD including liquid, gas, and slag phases. The coupled model is also based on a single-cell local equilibrium between species elements. Furthermore, with a similar approach, Liu et al. developed a new multizon reaction model that couples thermodynamics with CFD. The model predicted maximum hot spot temperature and decarburization rate in a bottom-blown electric arc furnace (EAF).

As seen earlier, numerical modeling of steel processes has grown over the years and is becoming more complex. Developed kinetic process models and coupled models can be used as a tool to understand more about the complex steelmaking process. The purpose of the present study was to develop a coupled fluid and thermodynamic model based on the local cell equilibrium concept. The focus was on the decarburization reactions taking place when an oxygen bubble rises in liquid steel. This was done to investigate fundamental phenomena such as where reactions take place, the effect of ferrostatic pressure, and the kinetics. This study is part of a larger research project to develop a dynamically coupled model of an AOD process and is important to further understand the complexity of the industrial decarburization process. The authors would like to stress that the objective of this study is not to present an overall AOD process simulation, but rather a model that provides the potential for transparency in decarburization reactions. Specifically, the effect of ferrostatic pressure on bubble decarburization is investigated.

2. Experimental Section

2.1. CFD Model

The model tracked the interface between the phases using the volume of fluid (VOF) model by solving the continuity equation of each phase volume fraction. The gas phase is solved with Equation (4)

$$\frac{\partial}{\partial t}(\alpha \rho) + \nabla \cdot (\alpha \rho \mathbf{v}) = S_\alpha + \sum_{p=1}^{n} (m_{\rho_p} - m_{\rho_p})$$

where $\alpha$ is the volume fraction, $\rho$ is the density, $\mathbf{v}$ is the velocity vector with $g$ and $l$ as indices for gas and liquid phases, respectively, $m_{\rho_p}$ is the mass transfer from gas to liquid phase, and $m_{\rho_p}$ is the mass transfer from liquid to gas phase. $S_\alpha$ is an additional source term.

In each control volume, the volume fraction of the liquid phase was computed based on the following constraint, as shown in Equation (5).
\[ \alpha_0 + \alpha_1 = 1 \] (5)

All phases were treated as incompressible Newtonian fluids with constant physical properties, where the model solves a set of mass and momentum conservation equations for the mixture phase in the following form.

\[ \frac{\partial}{\partial t} (\rho \mathbf{v}) + \nabla \cdot (\rho \mathbf{v} \mathbf{v}) = -\nabla p + \nabla \cdot [\mu(\nabla \mathbf{v} + (\nabla \mathbf{v})^T)] + \rho g + \mathbf{F} \] (6)

where \( \mu \) is the dynamic viscosity and \( \rho \) and \( \mathbf{g} \) are the pressure and gravitational acceleration respectively. Furthermore, \( \mathbf{F} \) represents the surface tension force, which was considered using the continuum surface force (CSF) model expressed as Equation (7) for a two-phase system:

\[ \mathbf{F} = \sigma \frac{\rho_1 \mathbf{V}_1 - \rho_2 \mathbf{V}_2}{0.5(\rho_1 + \rho_2)} \] (7)

\( \sigma \) is the surface tension coefficient and \( \mathbf{V} = \nabla \cdot \mathbf{n}, \mathbf{n} = \frac{\mathbf{n}}{n} \).

In all bubble simulations, the flow was considered to be laminar, whereas the first system with water/air was isothermal and for the second system with steel/oxygen, the temperature was treated by sharing the energy equation among the phases according to Equation (8). The water/air system was investigated to establish a decent bubble evolution before applying the steel/oxygen system and reactions. This is explained more clearly in the next sections.

\[ \frac{\partial}{\partial t} (\rho E) + \nabla \cdot (\rho \mathbf{v} E + p) = -\nabla \cdot (k \nabla T) + S_h \] (8)

The specific energy \( \mathbf{E} \) and temperature \( T \) are treated as mass-averaged variables, \( k \) is the effective thermal conductivity, and \( S_h \) is the source term that contains contributions from volumetric heat sources such as radiation.

A 2D axisymmetric computational domain was used under the simplification of a rectilinear bubble ascent to minimize the computational time. Previous work [27] has reported that for the current Eottös (Eq) and Galilei (Ga) numbers, the bubble ascent will be stable, thus further justifying the usage of a 2D axisymmetric model. The wall and outflow boundaries were set to no-slip and pressure outlet conditions, respectively. The geometric reconstruction scheme was used to track the interface and the pressure-implicit with splitting of operators (PISO) scheme was used for the pressure-velocity coupling. [28] The parameters used for both systems are presented in Table 1.

Before coupling with the chemical reactions, the CFD model was tested for grid independence using the recommendations of previous work [29–31] as a starting point. To eliminate the effect of boundaries on the bubble behavior, the solution domain size should be a minimum of 8 times the bubble diameter (\( d_b \)) and 16–25 quadrilateral computational cells were required across the bubble to correctly predict the bubble characteristics. Therefore, an initial case in the water/air system was calculated with 12, 16, 20, and 25 cells across a 0.01 m bubble diameter. The bubble ascent and shape evolution were studied through the terminal velocity and size aspect ratio, as presented in Figure 1. The calculations with 20–25 cells across the bubble showed small deviations to terminal velocity and a maximum deviation of 8.2% for the size aspect ratio. Hence, 25 quadrilateral cells across the bubble were suited for the simulations.

### 2.2. CFD and High-Temperature Chemistry

The steel temperature in an AOD typically varies between 1600 and 1800 °C. Under such high temperatures, it is reasonable to assume that the chemical reactions are rapid and limited by the mass transfer in the phases. Based on this, the model treats the reaction with the assumption of local cell equilibrium. The thermodynamic reactions between phases were calculated with Thermo-Calc [29] v.2010b software with the corresponding database TCS Metal Oxide Solutions (TCOX10). [30] TCOX is a thermodynamic database for slags and oxides. The thermodynamic model treats the liquid steel and the slag phase in a physically correct manner. It is described as one and the same phase (IONIC-LIQUID) with the possibility of having different compositions that are separated by a miscibility gap, for example, IONIC-LIQUID #1, #2, etc. The composition of the liquid phase is allowed to continuously change from metallic to oxidic when in equilibrium, all elements are divided to each side of the miscibility gap. This means that under the condition of equilibrium, the concentration and activities of elements in the slag and the liquid steel are calculated as a function of bulk composition, temperature, oxygen activity, etc. Furthermore, the software provides the calculation of multicomponent phase equilibria with the minimization of the Gibbs free energy method and is used to predict local equilibrium distributions within a defined system. Thermo-Calc allows solving for gas, liquid, and slag phases. In this study, the system consisted of the elements C, Cr, Fe, N, Ni, and O with the corresponding initial composition presented in Table 2.

The two software codes were coupled through two packages, the Thermo-Calc TQ interface which implements functions in the native language of Thermo-Calc and the user-defined function (UDF) which allows for dynamical coupling of functions in ANSYS Fluent. The coupling scheme was built to perform steps within each timestep for each finite volume cell. Initially, conditions such as temperature, pressure, weight fraction, and masses for each element in each phase were used as input from the
transient CFD solution. Thereafter, Thermo-Calc calculated equilibrium separately for each phase to account for the enthalpy and heat contents which served as inputs to calculate the total equilibrium. Finally, new temperatures, compositions, and masses were sent back to the CFD solution for the next timestep, as shown in the flowchart in Figure 2. The thermodynamic element quantities were treated with scalar transport locked to its fluid phase. The slag phase was included in the CFD solution. However, it was treated as an inert scalar in the liquid phase when formed. The reaction only takes place if the gas volume fraction is between 0.3 and 0.9 in the computational cell. This limit was set to decrease the number of equilibrium calculations that also would have only a minor impact on the results.

2.3. Numerical Procedure

All CFD simulations were performed using ANSYS Fluent R3 201926 with a convergence criterion of $1 \times 10^{-3}$ for all dependent variables except for energy, which was set to converge at $1 \times 10^{-6}$. The effect of the global Courant number ($C_{\text{global}}$) was tested prior to any calculations. Three different calculation methods were tested: 1) $C_{\text{global}} = 1$ with a fixed timestep of $1 \times 10^{-3}$ s, where equilibrium was calculated at each timestep, 2) $C_{\text{global}} = 0.1$ with a fixed timestep of $1 \times 10^{-4}$ s, where equilibrium was calculated every 10 timesteps, and 3) $C_{\text{global}} = 0.1$, with an adaptive timestep of $1 \times 10^{-4}$ s, where equilibrium was calculated every 10 timesteps. Figure 7 in the result section shows a negligible difference between at $C_{\text{global}} = 1$ with a fixed timestep and c) $C_{\text{global}} = 0.1$ with an adaptive timestep; hence, an adaptive timestep up to $1 \times 10^{-4}$ s with the criterion to meet a global Courant number below 0.1 was used for the simulations. All simulations ran for 0.3 s.

The original approach referred to as Case I intended to investigate the effect of ferrostatic pressure on decarburization by patching a spherical oxygen bubble with a 0.06 m$^3$4,35 diameter into a volume of liquid steel. The pressure imposed on the bubble from a 1.3 m bath height was simulated by calculating the bubble ascent in a domain height of 0.3 m with an outlet gauge pressure. The outlet pressure represents the depth of the bath where three levels were calculated, referred to as P1, P2, and P3 corresponding to the bubble being released at the depths of 1.3, 1.2, and 0.3 m, respectively. Figure 3. The weight fraction of elements in each phase was monitored with user-defined scalars (UDS), which applied the convection and diffusion of their corresponding phases.

Previous studies27,28–31,36–39 of the shape evolution of a rising bubble in water/air systems have been experimentally characterized and mapped with dimensionless numbers. For instance, the $E_o$ number was a ratio of gravitational force to the surface tension force $E_o = \frac{\mu \rho \delta k}{\gamma}$, with $\rho$ as liquid density, $g$ as gravitational...
acceleration, $R$ as initial radius, and $\sigma$ as surface tension coefficient. The $Ga$ number was the ratio of the gravitational force to the viscous force $Ga = \frac{\mu_1 \sqrt{V_R R}}{\rho}$. where $\mu_1$ is the dynamic viscosity of the liquid and Morton number $Mo = Eo^2/Ga^4$. Hence, if the values from Table 1 are applied to these dimensionless numbers, the bubble shape behaviors for both water/air and steel/oxygen systems are predicted to be described in Table 3 according to the regimes, as shown in Figure 4.

The second approach referred to as Case II released the bubbles at the same depth corresponding to the pressure levels P1, P2, and P3. However, nitrogen and argon were added to the bubble according to Table 4 to resemble process gas at the first step of decarburization.

### 2.4. Mesh Sensitivity

When patching a circular bubble into a domain consisting of quadrilateral cells, the volume fraction will cover cells within the boundary of the bubble’s interface. However, some cells close to the boundary were excluded, as shown in Figure 5a. Therefore, when refining the grid, the size of the excluded cells close to the boundary differed, which affected the initial mass of the elements inside the bubble, as shown in Figure 5b. Hence, a traditional mesh sensitivity study is not applicable to study the grid dependence on the mass of the elements. Instead, a square bubble was patched to partly resolve this problem, as shown in Figure 5c,d. Three grids, 12,000 (23 cells across bubble), 16,000, and 22,000 cells, were used to investigate the grid independence for the coupled model. An unstructured mesh could be used, that conforms to the initial bubble shape. However, this would create a mesh that is not quadrilateral which is more ideal for this study since quadrilateral meshes are preferred for the VOF. Ideally, an adaptive quadtree mesh would be used, but it is out of scope for the current study.

### 3. Results and Discussions

This study focuses on the model coupling to predict the decarburization reaction of a rising bubble under different pressures. However, prior to coupling with the thermodynamic reactions, a bubble ascent pattern is required.

<table>
<thead>
<tr>
<th>Calculation</th>
<th>$Eo$</th>
<th>$Ga$</th>
<th>$Mo$</th>
<th>Shape evolution</th>
</tr>
</thead>
<tbody>
<tr>
<td>Water-air</td>
<td>3.4</td>
<td>1104</td>
<td>$2.5 \times 10^{-11}$</td>
<td>sphere, ellipsoid cap, wobbly ellipsoid cap</td>
</tr>
<tr>
<td>Steel-oxygen</td>
<td>34</td>
<td>17 076</td>
<td>$4.6 \times 10^{-13}$</td>
<td>sphere, spherical cap, breakup, toroid</td>
</tr>
</tbody>
</table>

### Table 4. Gas composition for case II where the released bubble also contains nitrogen and argon.

<table>
<thead>
<tr>
<th>Mass % C</th>
<th>Mass % Cr</th>
<th>Mass % Fe</th>
<th>Mass % N</th>
<th>Mass % Ni</th>
<th>Mass % O</th>
<th>Mass % Ar</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0.15</td>
<td>0</td>
<td>0.79</td>
<td>0.06</td>
</tr>
</tbody>
</table>
Figure 5. Illustration of gas mass difference between grids when a,b) patching a spherical bubble versus c,d) patching a square bubble.

The bubble shape evolution in water/air systems has been previously studied and characterized with the dimensionless numbers mentioned in the Experimental Section. The topology of the bubble during ascent agrees well with the predicted bubble rise in the study by Tripathi et al.[27] It is described as the asymmetric oscillatory region with a final state of a wobbly ellipsoidal shape, as shown in Figure 6a. The high Bo and Ga numbers in the steel/oxygen system (Case 1) are not within the measured boundaries in the characterization map of Tripathi et al.[27] However, it follows the pattern and behaves as the map suggests with a central breakup. In addition, with the statement that high Bo and Ga numbers result in a steady central breakup to a toroidal shape during the ascent, as shown in Figure 6b.

The axisymmetric simulation might not be suitable for this case due to the suppression of an unstable bubble ascent. However, due to the lack of experimental studies on the correlation between the bubble ascent and dimensionless numbers for a steel/oxygen system, the study relies on the statement of Xu et al.[30] Gas bubbles with a diameter of 10–20 mm in steel rise in a stable linear path and hence, the CFD part of the coupled model is considered reasonable for the study.

Figure 7 shows a small effect of the different calculation setups on oxygen in the gas bubble. It can be seen that the global Courant number, that is, the timestep, has a very small impact on the results. The oxygen content in the bubble shows a similar trend as the other elements after the initial reactions; notice that the normalized mass is plotted in the figure. All further graphs presented in the paper are normalized against the elements’ initial mass denoted as $m_{\text{element}}/m_{\text{element}}^0$ on the Y-axis.

Figure 8 shows the normalized mass of oxygen in the square gas bubbles reacting while ascending in the steel for different grids. The mesh sensitivity study for reactive flow can be seen to differ at most by 4.5% between 12 and 22 k cells. Therefore, with these small differences, it was deemed reasonable to continue with the 12 k mesh, found in the previous mesh sensitivity study, to save computational time.

3.1. Case 1: 60 mm Oxygen Bubble Ascending 0.3 m

The temperature at the bubble–liquid interface and its distribution, controlled by convection during the ascent, is illustrated in Figure 9 for the 60 mm bubble. The temperature is initially set to 1873 K and rises to 1890 K due to the exothermic reactions at the interface. The instantaneous rise in temperature is also found by...
Jarvinen el al.[12] and the level agrees with the operating temperature span in the AOD process during decarburization.[11]

The normalized mass of carbon in gas and steel is presented in Figure 10 for the pressure levels P1 and P2. It appears that the oxygen bubble reacts dramatically with carbon and nitrogen in steel to produce off-gases. However, as the mass of the steel surrounding the bubble is remarkably larger than the bubble mass, the initial local equilibrium in the bubble dynamically adjusts toward a global equilibrium in the system. This is visually represented in Figure 11, where the mass fraction of gas is seen to disappear over time.

To further investigate the possible final state, single-point equilibrium calculations were performed for the cases P1, P2, and P3. P1 and P2 could not sustain any gas at the final thermodynamic equilibrium. On the other hand, at lower pressure levels such as P3, gas becomes more stable, which results in both liquid and gas as stable phases in the system. This is seen in both single-equilibrium and coupled model calculations. The difference in the elements during the bubble ascent is presented in Figure 12 and 13 and a visual representation of the carbon gradient in the bubble over time is illustrated for the P3 level in Figure 14. It can be seen from Figure 12 and 13 that the pressure has a large effect on decarburization and nitrogen removal. It is also possible that the kinetics are fast enough to admit decarburization calculations at pressures close to surface pressure. However, with larger gas flow rates, there will be a larger concentration gradient in the plume region that may admit decarburization at higher pressures.

It is not possible to provide a quantitative validation due to the lack of experimental data. In this study, a qualitative comparison has been done. For this case, the results show that the most active elements are carbon and nitrogen, as illustrated in Figure 15, with the normalized mass of elements in steel for P3. The carbon is drastically increased in the gas bubble before 0.2 s and seems to level out due to saturation of the bubble. This behavior is visually illustrated in Figure 14, where the carbon gradient appears to diminish between the frames 0.2 and 0.25 s. This trend is common in the AOD process and is also predicted in the previous work.[12-14,16,24,25] The period for a fully saturated CO bubble may be affected by the ascending shape evolution (breakup or no breakup), supply of oxygen (size of bubble), and/or mass transfer of carbon to the interface (bubble rising velocity).

Furthermore, as presented in previous works,[7-11,13,14,16,24,25] oxidation of elements such as iron and chromium is inevitable in the AOD process. Despite this, no oxidation of Cr and Fe appears in Figure 14. This can be explained by the scale difference of the simulations between this case and previous work. The previous studies calculate the decarburization of a full AOD converter ranging from 5 min to full process while this case is calculated for a single bubble. Another explanation is connected to the initial conditions such as the amount of dissolved oxygen or carbon in the melt. In the bubble simulation by Jarvinen et al.[12] they found that the major fraction of oxygen oxidizes the carbon in the melt due to the high initial carbon content of 1 mass%-%. This was also found by Anderson et al. with 0.6 mass%-%. The initial dissolved carbon and oxygen content for this case in 1.5 and 0.03 mass%-%, respectively, which implies that the oxygen from the gas bubble is dissolved into the steel instead of oxidizing chromium or iron. Basically, this case
Figure 10. Normalized mass of carbon in a) gas and b) steel in case I. Results from the P3 level are excluded for better visualization of differences between P1 and P2.

Figure 11. Illustration of the gas mass fraction during ascent. At 0.20 s, the bubble has almost completely dissolved into the steel.

represents the first injected oxygen bubble into a freshly tamped melt that has progressed from the earlier step in the production line, for example, the EAF. This context is important to note as it could indicate that the melt is yet to reach the critical conditions required for chromium oxidation. The reactions for different initial conditions will be presented and discussed in Case III.

3.2. Case II: 60 mm Bubble with Oxygen, Nitrogen, and Argon Bubble Ascending 0.3 m

The results for the bubble consisting of oxygen, argon, and nitrogen are presented in Figure 16 and 17. Due to the inert argon gas in the bubble, a gas phase will always be stable in the present system. Looking at Figure 16a, it is understood that the carbon that has entered the bubble does not fully dissolve back into the steel at higher pressure levels such as for P1 and P2 in Case I. However, with lower pressure, more carbon in the steel reacts with oxygen to form off-gases, as shown in Figure 16b. In addition, the time for carbon saturation in the bubble is extended. This is explained in Figure 17. As the bubble partly consists of nitrogen, it will dissolve into the steel at higher pressure levels (P1 and P2). On the other hand, at lower pressure levels (P3), the lowered partial pressure of N₂ increases nitrogen solubility in the bubble. The time to saturate the bubble with carbon is never reached for the low-pressure case (P3). However, after 0.2 s, there is a clear change in the rate at which carbon enters the bubble.

The difference in pressure for P1 and P2 does not appear to have a large effect on decarburization. However, it appears that

Figure 12. Normalized mass of carbon in a) gas and b) steel in Case I. Results from the P3 level are included.
Figure 13. Normalized mass of a) nitrogen in gas and b) oxygen in gas in Case I. Results from the P3 level are included.

Figure 14. Carbon in the 60 mm gas bubble over 0.25 s (Case I, P3).

Figure 15. Normalized mass of elements in steel during bubble ascent (Case I, P3).

the saturation level is increased when the bubble is released at higher bath levels and that a bubble released close to the bath surface has the most efficient decarburization. This is due to the increased solubility of the elements in the gas at higher bath levels, which decreases the partial pressures in the bubble. In addition, the single-cell equilibrium calculations in the Thermo-Calc software agreed with the end results of the coupled model. This means that for a semiclosed system such as these cases with a single bubble release, the bubble in the coupled model will inevitably reach the state that is calculated with a single-cell equilibrium calculation. In practice, the plume in an AOD consists of interactions between multiple bubbles. With this in mind, it is difficult to describe how pressure affects the decarburization reactions in a continuous oxygen supply by modeling a single bubble. However, with the coupled model, it is possible to eliminate the closed system effect by modeling a continuous jet with the inlet positioned at different heights to include the pressure differences. This is important to better represent the practical operation and is planned for future work.

3.3. Case III: 60 mm Bubble Ascending 0.3 m with Different Initial Melt Conditions

In the cases discussed before, the initial composition of the steel represented a newly tapped melt with high carbon and low oxygen content. In these cases, there was no slag formation during the calculations. In Case III, the initial steel melt composition was adjusted to fit a charge further into the decarburization step, as shown in Table 5, and the model was tested for the pressure case P3. The results show that a small amount of slag was formed with the elements: chromium, iron, and oxygen. The slag formation is illustrated as the mass of chromium during the bubble ascent, as shown in Figure 18. It is seen that the slag is being dragged along with the bubble. It is important to note that the slag which forms in these calculations is treated as inert and has the same physical properties as the liquid steel. This topic will also be included in further studies. In Case I and II, the supply of oxygen is restricted to the single bubble introduced. The model showed that at first contact with the melt it was more favorable to decarburize and oxidize the steel than to form slag. As the bubble occupies a small fraction of the domain compared with the steel, it did not affect the composition of the steel to the point of slag formation before
Figure 16. Normalized mass of carbon in gas (case II). The left graph a) is adjusted to fit the data acquired from levels P1 and P2 whereas the right graph b) presents the whole range to include all pressure levels.

Figure 17. Normalized mass of nitrogen in gas (case II).

Table 5. Initial melt composition (case II).  

<table>
<thead>
<tr>
<th>Mass % C</th>
<th>Mass % Cr</th>
<th>Mass % Fe</th>
<th>Mass % N</th>
<th>Mass % Ni</th>
<th>Mass % O</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.5</td>
<td>23.2</td>
<td>69.84</td>
<td>0.16</td>
<td>4.2</td>
<td>0.1</td>
</tr>
</tbody>
</table>

the bubble was saturated with carbon. In the industrial process, the supply of oxygen is much higher than a single bubble and is not comparable to this study. A jet oxygen supply is therefore well suited as the next step of this project. The current method is based on the assumption of very high reaction rates, limited only by local mass transfer. This is reasonable considering the very high temperatures of the process. However, to develop the method further, the next suggested step is to incorporate a reaction rate that is limited by the local cell turbulence. Furthermore, it is not fully understood how solid oxides behave in regard to formation as well as dissolution. The current approach to let the oxides form and afterward be treated as inert (nonreacting) should be seen as a first step to investigate the effect of pressure on the dynamic formation of oxides in the AOD process.

4. Conclusion

A coupled thermodynamic and fluid transport model on a dynamic oxygen bubble in a steel melt was developed to fundamentally investigate pressure differences in decarburization reactions. The successful software coupling has led to some promising development areas. 1) The reactions take place rapidly

Figure 18. Chromium mass in inert slag formation over 0.28 s during bubble rise (case III).
and saturate the gas bubble with carbon as well as increase the temperature. The saturation time is affected by the pressure and the elements of the injected gas bubble. For a 60 mm oxygen bubble injected at the surface, the time to saturate the bubble with carbon is \( \approx 0.2-0.25 \) s, while a bubble consisting of oxygen, nitrogen, and argon requires more than 0.3 s. 2) The impact of ferrostatic pressure on the reactions was more significant at larger depth differences of the injected bubble. A bubble injected closer to the surface is saturated at higher carbon levels due to the lowered partial pressure of the elements in the gas. However, at higher pressure which represents lower injection depths, the bubble completely dissolves into the steel without an inert gas present. Even with an inert gas present, the ending oxygen supply of a single bubble poses difficulties in analyzing the effect of pressure on decarburization. This may be eliminated by modeling a continuous oxygen jet and will be investigated in further studies. The slag formation is dependent on the initial steel melt composition where melts representing early decarburization stages did not form any slag by the small amount of oxygen supplied from a bubble. 4) Further studies will involve a coupled model of a jet oxygen supply to investigate the effect of pressure on the decarburization reactions and study the critical point of chromium oxidation.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available on request from the corresponding author. The data are not publicly available due to privacy or ethical restrictions.

Keywords

argon-oxygen decarburization process, bubbles, computational fluid dynamics, coupled models, decarburization reactions

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Modelling Decarburization in the AOD Converter: A Practical CFD-Based Approach with Chemical Reactions

Serg Chanouian*, Jyrki Pitkala, Henrik Larsson, Mikael Ersson

Abstract

Gas blowing technology is widely used in converter steelmaking to homogenize liquid steel and accelerate chemical reactions, with Argon oxygen decarburization (AOD) being the dominant process for stainless steelmaking. Due to the harsh environment, it is advisable to study the phenomenon using small-scale physical models and numerical simulations before conducting industrial-scale trials. This paper presents a practical computational fluid dynamics (CFD) approach for simulating the AOD process, with chemical reactions considered. This approach can simulate the entire process in a reasonable time using a standard workstation. The simulation employs a Finite Volume Method CFD approach to handle mass, momentum and energy transfer, and a local equilibrium assumption is utilized. The study shows that a practical approach can be used to model the initial stage of decarburization in the AOD process with a reduced accuracy in mass transport calculations. The accuracy of the simulation is validated using industrial data, and good agreement is found.

Keywords: AOD process, decarburization reactions, coupled model, CFD
The Importance of Mixing Time in Intensely Stirred Metallurgical Reactors: Applied on Decarburization Reactions

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Abstract: In metallurgical converter processes, numerical modeling is a useful tool for understanding the complexity of the systems. In this paper, we present a practical model that couples fluid dynamics and chemical reactions to explore the impact of mixing time on decarburization. Using computational fluid dynamics (CFD), in this study, we investigate an arbitrary metallurgical reactor with continuous oxygen supply, focusing on the Fe–C–O system. The model employs local equilibrium, a turbulence limiter, and a finite volume method for mass, momentum, and energy transfer. Tracer injection points in the gas plume’s rising region exhibit faster mixing, and a comparison of reaction cases reveals distinct decarburization rates based on oxygen injection distribution and the influence of turbulence on reactions. Overall, while mixing time matters, the results show that this system is primarily governed by thermodynamics and oxygen supply, and a 270% increase in mixing time increase had a small impact on the end carbon content.

Keywords: mixing time; decarburization; metallurgical processes; computational fluid dynamics

1. Introduction

In intensely stirred metallurgical converter processes such as the argon oxygen decarburization process (AOD), it is common to use physical and numerical modeling as a tool for adding transparency and understanding the otherwise opaque and complex systems. A proper model could be rewarding in terms of knowledge, safety, and cost reduction for converter development. In general, metallurgical converters utilize gas blowing for decarburization. For instance, in the AOD converter, a mixture of oxygen and inert gas is typically introduced through side nozzles to decarbure the steel. The oxygen reacts with carbon in the steel and produces mainly CO. This reaction is driven by the low partial pressure of CO; hence, a mixture of oxygen with inert gas. The reaction also benefits through kinetics which is the transportation of carbon in the steel to the reaction zone. This is accomplished by the stirring that is introduced by blowing gas into the vessel. Thus, the main numerical modeling challenges are fluid transport and high temperature chemical reactions. CFD is a popular choice for modeling fluid mass transport [1–9], see Table 1. Zhu et al. [2] presented an early side-blowing model by implementing a plume model to predict mixing and turbulence behavior in 3D. The mixing time was evaluated using a tracer added at five positions, revealing that the mixing time remained largely unaffected. Notably, these positions were situated at the surface of the bath within an area corresponding to half the bath diameter, effectively placing all tracer positions near the center of the bath. Oscillation studies of a 120-ton AOD converter were carried out in refs. [3–5] with a down-scaled physical model and industrial scale numerical model. By altering the blowing rate and fill level, the researchers observed variations in the mixing times. They found that higher blowing rates led to shorter mixing times, and the mixing time initially increased and then remained constant with increasing fill levels. An optimal fill level range of 0.7 < height/diameter < 0.75 was identified. A comparison between the traditional impulse injection method
and the continuous injection method of a food coloring tracer demonstrated a remarkable reduction in mixing time variations with a continuous injection [4]. This study reported a range of approximately 20–60 s for impulse injection, whereas continuous injection yielded a narrower range of 11–24 s. Also, Visuri et al. [7] reported mixing times in the range of 7–26 s in a physical water model of an AOD with pulse tracer injection. In addition, they found an increase in mixing time when the tracer was injected at lower depths compared to near the bath surface. However, a separate numerical model study [9] that investigated an AOD converter of the same size reported a mixing time range of 100–116 s with six nozzles and 60–120 s with seven nozzles. It is important to note that these results were not validated against any industry or water model data. Tilliander et al. [6] employed a model of an AOD converter encompassing liquid, slag, and gas phases to investigate flow characteristics. The findings revealed that lower gas flow rates induced a clockwise circulation of the steel bath, with the vortex located in the upper region of the converter. Remarkably, this flow pattern aligned with the observations reported by other side-blowing models [2–5,8,9]. However, higher flow rates exhibited a more localized counterclockwise circulation behind and below the gas plume. The penetration length of the increased gas flow rate surpassed the midpoint of the domain, reaching close to the wall opposite to the nozzles. Interestingly, these findings contradict the observations in Refs. [3,9], where the horizontal gas jet penetration did not exceed past the vessel’s midpoint.

Table 1. Previous fluid dynamic models on intensive metallurgical converters that utilize gas blowing.

<table>
<thead>
<tr>
<th>Ref.</th>
<th>Year</th>
<th>Pro</th>
<th>Sys</th>
<th>Inj</th>
<th>M</th>
<th>TM</th>
<th>MM</th>
<th>t_mix</th>
<th>React</th>
<th>Note</th>
<th>First Author</th>
</tr>
</thead>
<tbody>
<tr>
<td>[2]</td>
<td>1998</td>
<td>AOD</td>
<td>WM</td>
<td>b, s = t</td>
<td>N</td>
<td>k</td>
<td>n/a</td>
<td>x</td>
<td>n/a</td>
<td>Plume</td>
<td>Zhu</td>
</tr>
<tr>
<td>[3]</td>
<td>2010</td>
<td>AOD</td>
<td>WM/120 ton</td>
<td>s</td>
<td>P + N</td>
<td>Re</td>
<td>SST-</td>
<td>VOF</td>
<td>x</td>
<td>n/a</td>
<td>Oscillation</td>
</tr>
<tr>
<td>[4]</td>
<td>2012</td>
<td>AOD</td>
<td>WM/120 ton</td>
<td>s</td>
<td>P</td>
<td>n/a</td>
<td>SST-</td>
<td>VOF</td>
<td>n/a</td>
<td>Mixing</td>
<td>Wuppermann</td>
</tr>
<tr>
<td>[5]</td>
<td>2013</td>
<td>AOD</td>
<td>WM/120 ton</td>
<td>s</td>
<td>P + N</td>
<td>SST-SAS</td>
<td>VOF</td>
<td>N/a</td>
<td>n/a</td>
<td>Oscillation</td>
<td>Wuppermann</td>
</tr>
<tr>
<td>[6]</td>
<td>2014</td>
<td>AOD</td>
<td>95 ton</td>
<td>s</td>
<td>N</td>
<td>k</td>
<td>n/a</td>
<td>x</td>
<td>n/a</td>
<td>Flow</td>
<td>Tilliander</td>
</tr>
<tr>
<td>[7]</td>
<td>2016</td>
<td>AOD</td>
<td>WM/150 ton</td>
<td>s</td>
<td>P</td>
<td>n/a</td>
<td>SST-SAS</td>
<td>VOF</td>
<td>n/a</td>
<td>Mixing</td>
<td>Visuri</td>
</tr>
<tr>
<td>[8]</td>
<td>2020</td>
<td>AOD</td>
<td>120 ton</td>
<td>s</td>
<td>N</td>
<td>n/a</td>
<td>SST-SAS</td>
<td>VOF</td>
<td>n/a</td>
<td>Mass transfer</td>
<td>Wimmer</td>
</tr>
<tr>
<td>[9]</td>
<td>2023</td>
<td>AOD</td>
<td>120 ton</td>
<td>s</td>
<td>N</td>
<td>k</td>
<td>n/a</td>
<td>x</td>
<td>n/a</td>
<td>Nozzle configuration</td>
<td>Cheng</td>
</tr>
<tr>
<td>Current</td>
<td>2023</td>
<td>AOD</td>
<td>WM/1.1 ton</td>
<td>s</td>
<td>N</td>
<td>Re</td>
<td>SST-SAS</td>
<td>VOF</td>
<td>n/a</td>
<td>High temperature reactions</td>
<td>Charanigan</td>
</tr>
</tbody>
</table>

Pro, process; Sys, system; WM, water model; Inj, injection method; b, bottom; s, side; t, top; M, model; N, numerical; P, physical; t_m, mixing time; MM, multiphase model; TM, turbulence model; Re, realizable; SST-SAS, shear stress transport-scale adaptive simulation; React, chemical reactions.

In general, the above-mentioned studies collectively contribute to our understanding of mixing and flow dynamics in gas-stirring converters such as the AOD converter, highlighting the importance of various parameters such as fill level, injection methods, nozzle configuration, and gas flow rates. By comprehending these factors, researchers and engineers can strive to optimize converter designs and operating conditions for improved efficiency and performance.

Although it is emphasized that mixing and flow characteristics play an important role in the metallurgical converters, the optimization of metallurgical processes, such as the AOD converter, extends beyond fluid dynamics alone. The numerical development has also considered chemical reactions, where a lot of work has been dedicated to developing reaction models [10–25], involving decarburization of the steel, see Table 2.
Table 2: Previous kinetic and chemical reaction models.

<table>
<thead>
<tr>
<th>Ref.</th>
<th>Year</th>
<th>Proc</th>
<th>SE</th>
<th>DB</th>
<th>Ph</th>
<th>Met</th>
<th>Kinetics</th>
<th>SP</th>
<th>CFD</th>
<th>t_melt</th>
<th>Note</th>
<th>First Author</th>
</tr>
</thead>
<tbody>
<tr>
<td>[10,11]</td>
<td>2002</td>
<td>AOD</td>
<td>Std, Mn, Si</td>
<td>Lit</td>
<td>g. l_{melt}, s</td>
<td>OABS</td>
<td>Mass transfer of carbon in steel</td>
<td>SH</td>
<td>n/a</td>
<td>n/a</td>
<td>Melt analysis, OD, CRE</td>
<td>Wei &amp; Zhu</td>
</tr>
<tr>
<td>[12-14]</td>
<td>2007-2011</td>
<td>AOD</td>
<td>Std, Mn, Si</td>
<td>Lit</td>
<td>g. l_{melt}, s</td>
<td>OABS</td>
<td>Mass transfer of carbon in steel</td>
<td>SH</td>
<td>n/a</td>
<td>n/a</td>
<td>Combined top and side blowing</td>
<td>Zhu, Shi, Wei</td>
</tr>
<tr>
<td>[15]</td>
<td>2008</td>
<td>BOF</td>
<td>Fe, C, O, Si</td>
<td>TC</td>
<td>g. l_{melt}, s</td>
<td>GFM</td>
<td>CFD</td>
<td>HV</td>
<td>x</td>
<td>n/a</td>
<td>Top blowing</td>
<td>Ersson</td>
</tr>
<tr>
<td>[16]</td>
<td>2010</td>
<td>AOD</td>
<td>Std, Mn, Si, N</td>
<td>HSC, Lit</td>
<td>g. l_{melt}, s</td>
<td>LAMB</td>
<td>Conservation eq</td>
<td>HV</td>
<td>n/a</td>
<td>n/a</td>
<td>Decarburization in bubble</td>
<td>Järvinen</td>
</tr>
<tr>
<td>[17-19]</td>
<td>2011-2016</td>
<td>AOD</td>
<td>Std, Mn, Si, N, Ni</td>
<td>HSC, Lit</td>
<td>g. l_{melt}, s</td>
<td>LAMB</td>
<td>Conservation eq</td>
<td>HV</td>
<td>n/a</td>
<td>n/a</td>
<td>Melt analysis in AOD</td>
<td>Järvinen &amp; Sauli</td>
</tr>
<tr>
<td>[20]</td>
<td>2011</td>
<td>RH</td>
<td>Fe, C, O</td>
<td>FS</td>
<td>g. l_{melt}, s</td>
<td>GFM</td>
<td>EERZ</td>
<td>ZV</td>
<td>n/a</td>
<td>n/a</td>
<td>Decarburization in RH</td>
<td>Van Ende</td>
</tr>
<tr>
<td>[21,22]</td>
<td>2013</td>
<td>AOD</td>
<td>Std, Ni</td>
<td>TC + TA2</td>
<td>g. l_{melt}, s</td>
<td>CV + GFM</td>
<td>CFD</td>
<td>HV</td>
<td>n/a</td>
<td>n/a</td>
<td>LMV, CRE</td>
<td>Andersson</td>
</tr>
<tr>
<td>[23]</td>
<td>2016</td>
<td>LF</td>
<td>Std + 16</td>
<td>FS</td>
<td>g. l_{melt}, s</td>
<td>GFM</td>
<td>EERZ</td>
<td>ZV</td>
<td>n/a</td>
<td>n/a</td>
<td>Kinetic LF model</td>
<td>Van Ende</td>
</tr>
<tr>
<td>[24]</td>
<td>2020</td>
<td>LF</td>
<td>Fe, Mn, O+3</td>
<td>TC</td>
<td>g. l_{melt}, s</td>
<td>GFM</td>
<td>EERZ</td>
<td>ZV</td>
<td>n/a</td>
<td>n/a</td>
<td>Kinetic LF model</td>
<td>Mason</td>
</tr>
<tr>
<td>[25]</td>
<td>2022</td>
<td>AOD</td>
<td>Std + 16</td>
<td>TC</td>
<td>g. l_{melt}, s</td>
<td>GFM</td>
<td>n/a</td>
<td>SH</td>
<td>n/a</td>
<td>n/a</td>
<td>Nitrogen in AOD</td>
<td>Wei, WJ</td>
</tr>
<tr>
<td>Current</td>
<td>2023</td>
<td>AOD</td>
<td>Fe, C, O</td>
<td>TC</td>
<td>g. l_{melt}, s</td>
<td>CV + GFM</td>
<td>CFD</td>
<td>HV</td>
<td>x</td>
<td>x</td>
<td>Mixing effect on Decarburization</td>
<td>Chanouzian</td>
</tr>
</tbody>
</table>

Pro, process; RH, Rustahl Heraeus; LF, ladle furnace; SE, system elements; Std (standard), Fe-Cr-Co; DB, database; Lit, Literature; HSC, HSC chemistry software; FS, FactSage; TC, Thermo-Calc; TA2, TimeAOD2; Ph, phases; g, gas; l_{melt}, liquid metal; l_{s}, liquid slag; s, slag; i, inclusion; Met, method; OABS, oxygen affinity based selectivity; LAMB, law of mass action based; GFM, Gibbs free energy minimization; CV, control volume; SP, system pressure; SH, static homogeneous; HV, hydrostatic variation; ZV, zone variation; t_melt, mixing time; OD, oxygen distribution; CRE, carbon removal efficiency; LMV, local melt variations.

The critical carbon content and oxygen distribution in an AOD converter were investigated using a numerical kinetic reaction model [10,11]. Oxygen is distributed to different components in the steel based on the relative affinity of their reactions. The work suggests that in the early stages of decarburization, when the carbon levels are high, the incoming oxygen is rapidly consumed. As a result, it is controlled by the supply of oxygen. At low carbon levels, the decarburization rate is predominantly influenced by the mass transfer of carbon from the molten steel bulk to the reaction interface. Additionally, a refined version of the model has been developed to study decarburization in a combined top- and side-blowing converter, which was benchmarked against industrial heats under oxidizing and reducing conditions [12-14].

Another notable study, by Wei et al., focused on the development and testing of an AOD model capable of predicting the dissolved nitrogen content and other metal/slag compositions [25]. This model utilized thermodynamic equilibrium calculations using commercial software with reliable databases and was successfully applied to process data from 12 production-scale heats. It effectively predicted changes in dissolved nitrogen content throughout the entire AOD process, considering multiple phases such as liquid metal, liquid slag, solid slag, and gas. However, certain areas require further attention such as the implementation of different total gas pressures for nitrogen and carbon reactions, and incorporating kinetic mechanisms into the model. One drawback of the above-mentioned models is their treatment of pressure as static and homogeneous, which is particularly limiting in one-cell calculations. However, these calculations have proven to be fast and efficient for process control purposes. To overcome the limitations associated with a static pressure, Järvinen et al. addressed conservation equations in their kinetic and chemical...
reaction model of the AOD process [16–19]. They developed a rate expression based on the modified law of mass action, initially studying the model on a stagnant bubble and later expanding it to a full AOD converter with locally varying conditions of a rising gas plume. This method was further enhanced to include multiple parallel reversible reactions controlled by mass transfer.

An alternative approach that differentiates itself from the reaction interface models mentioned above is the effective equilibrium reaction zone model (EERZ) [20,23,24]. Unlike the reaction interface models, the EERZ method assumes equilibrium of all phases located in an effective reaction zone at the interface, simplifying mass transfer equations by assuming equal mass transfer coefficients for a given solution. By considering the reaction volume at the interface, the mass transfer equations can be transformed into simple thermodynamic calculations, facilitating linkage to thermodynamic databases. However, physical descriptions of reaction mechanisms and kinetics for each zone are still required.

Ersson et al. [15] introduced a CFD model coupled with chemical reactions in a top blown converter which was based on local control volume equilibrium after each timestep. Andersson et al. [21,22] later applied this coupling method in the AOD converter. This approach offers a significant advantage as it eliminates the need for information regarding the interfacial surface area. In this method, if an interphase exists, the entire cell is assumed to be in equilibrium. However, this approach demands significantly greater computational effort, especially when an accurate description of mass transfer to the interface is required. This is due to the necessity of resolving multiphase features in the computational cells which further amplifies the computational requirements.

Overall, it has been highlighted that flow characteristics and mixing time are important contributing factors to a more effective process and decarburization rate in metallurgical reactors. Specifically, at low carbon levels, the rate is governed by the mass transfer of carbon in the bulk to the reaction interface. However, to the best of our knowledge, no previous study has examined the influence of mixing time on the decarburization reaction. In most studies, it has been concluded that a decrease in mixing time is good. However, it is also reasonable to assume that there is a point where a decrease in mixing time would not yield any increase in the reaction rates. Therefore, the objective of this study is to develop a practical model that combines fluid dynamics with chemical reactions, building upon our previous work [26,27]. An arbitrary metallurgical side-blowing reactor with continuous oxygen supply is utilized in this study to investigate the aforementioned effect focusing on the decarburization reaction in a Fe-C-O system.

2. Materials and Methods

The numerical model was based on a side-blowing physical water model conducted in ref. [26]. The physical water model was used as validation for the mixing time and flow structure of the system. Subsequently, a thermodynamic database and chemical reactions were integrated into the numerical model to investigate the impact that mixing time had on decarburization. The immersion of a fast-moving jet into the liquid puts a severe limitation on the overall timestep. This is due to the very high courant numbers owing to a combination of high velocities, sharp gas–liquid interfaces, and small cells needed in the jet region. Thus, simulating high-velocity gas–liquid injections require substantial computational power. This is commonly addressed through a numerical technique [3,5,28,29], where the characteristics of the jet flow are separately simulated and then imported into the vessel in another simulation. To account for the expected significant increase in simulation time when employing a combined model that incorporates heat transfer and reactions, a decision was made to utilize a coarse model for incompressible fluid flow transportation. The injection of gas into the system was introduced further away from the wall, representing the point where the gas plume is mainly controlled by the buoyancy, i.e., after the jet penetration depth is reached. By employing this approach, it becomes feasible to capture the flow structure and mixing within the bath caused by the jet, without the necessity of simulating the high velocity and small length scales at the nozzle.
outlet. With an emphasis on the practical approach, it was assumed that this would be sufficient to study decarburization in systems of industrial significance.

2.1. General Equations

The Eulerian multiphase [30] model was employed to solve the conservation of mass and momentum for each phase under a shared pressure according to Equations (1) and (2), respectively. The momentum transfer between the phases is modeled with a drag term where a diameter is specified for the secondary phase:

\[ \frac{\partial}{\partial t} \left( \alpha_q \rho_q \vec{v}_q \right) + \nabla \cdot \left( \alpha_q \rho_q \vec{v}_q \vec{v}_q \right) = \sum_{p=1}^{n} (m_{rq} - m_{qr}) q p + S_q \]  

where the phases are denoted as \( q \) and \( r \), \( \alpha \) is the volume fraction of the phase, \( \rho \) is the density, \( \vec{v} \) is the phase velocity, and \( m_{rq} / m_{qr} \) are the mass transfer from one phase to the other. The last term \( S_q \) is a source term which by default is zero.

\[ \frac{\partial}{\partial t} \left( \alpha_q \rho_q \vec{v}_q \vec{v}_q \right) + \nabla \cdot \left( \alpha_q \rho_q \vec{v}_q \vec{v}_q \vec{v}_q \right) = -\alpha_q \nabla \rho + \nabla \cdot \tau_q + \alpha_q \rho_q \vec{g} + \sum_{p=1}^{n} \left( K_q \left( \vec{v}_q - \vec{v}_q \right) + m_{rq} \vec{v}_q - m_{qr} \vec{v}_q \right) + \left( \vec{F}_q + \vec{F}_{lq} + \vec{F}_{w,q} + \vec{F}_{td,q} \right) \]  

Here, \( p \) is the pressure shared by the phases, \( \tau \) is the stress strain tensor as seen in Equation (3), and the gravitational acceleration is denoted as \( \vec{g} \). \( K_q \) is the interphase momentum exchange coefficient defined in Equation (4), and \( \alpha_q / \alpha_q \) are interphase velocity. The last terms are \( \vec{F}_q \) external body force, \( \vec{F}_{lq} \) lift force, \( \vec{F}_{w,q} \) wall lubrication force, \( \vec{F}_{vm,q} \) virtual mass force, and \( \vec{F}_{td,q} \) turbulent dispersion force acting between the different phases.

\[ \tau_q = \alpha_q \mu_{eff,q} \left( \nabla \vec{v}_q + \nabla \vec{v}_q^T \right) \]  

Here, \( \mu_{eff,q} \) is the effective viscosity for phase \( q \).

\[ K_q = \frac{\rho f}{6 \alpha_q} d_{b,r} A_i \]  

Here, \( f \) is the drag function defined in Equation (5) according to the Schiller–Naumann model [31], \( \tau_i \) is the particulate relaxation time, \( d_{b,r} \) the diameter of bubbles of phase \( r \), and \( A_i \) is the interfacial area.

\[ f = \frac{C_D \Re}{24} \]  

Here, \( C_D \) is the drag coefficient defined in Equation (6) and \( \Re \) is the Reynolds number:

\[ C_D = \begin{cases} \frac{24(1+0.158\Re^{0.66})}{\Re}, & \Re \leq 1000 \\ 0.44, & \Re \geq 1000 \end{cases} \]  

Here, the relative Reynolds number is described in Equation (7):

\[ \Re = \frac{\rho q \vec{v}_q \cdot \vec{v}_q |\vec{v}_q|}{\mu_q} \]  

where, \( \mu_q \) is the viscosity for phase \( q \).
Many types of engineering problems targeted at the metallurgical industry describe turbulence with two equations for effective viscosity approximation such as the k-ε or k-ω models (Table 1), as they have become the industry standard. While it has been demonstrated in the literature [28, 29] that higher accuracy can be attained through the utilization of more sophisticated turbulence models, it is to be acknowledged that this advantage is accompanied by an increase in computational time. With an emphasis to developing a practical model, it was decided to treat the turbulence in the domain with the k-ε realizable model [32], where Equation (8) expresses the turbulent viscosity in relation to the kinetic energy (k) and turbulent dissipation rate (ε) of phase q. Here, the viscosity coefficient \( C_{\mu} \) is a function of local flow parameters. Furthermore, Equations (9) and (10) present the modeled transport equations for k and ε, respectively:

\[
\mu_t = \rho_q C_{\mu} \frac{k^2}{\varepsilon}
\]

\[
\frac{\partial \left( \rho_q \varepsilon \right)}{\partial t} + \nabla \cdot \left( \rho_q \varepsilon \mathbf{v} \right) = \nabla \cdot \left( \rho_q \left( \mu_t + \frac{\mu_{\varepsilon}}{\varepsilon} \right) \nabla \varepsilon \right) + \rho_q \left( \varepsilon \mu_t C_{\varepsilon} + C_{\mu} \right) - \rho_q \varepsilon \frac{\partial k}{\partial t}
\]

\[
\frac{\partial \left( \rho_q k \right)}{\partial t} + \nabla \cdot \left( \rho_q \mathbf{v} \rho_q k \mathbf{v} \right) = \nabla \cdot \left( \rho_q \left( \mu_t + \frac{\mu_{\varepsilon}}{\varepsilon} \right) \nabla k \right) + \rho_q \left( \varepsilon k C_{k} - k \frac{\partial \varepsilon}{\partial t} \right)
\]

In this context, \( C_{\mu} \) represents the production of turbulent kinetic energy resulting from velocity gradient, while \( C_{\varepsilon} \) signifies the production of turbulent kinetic energy due to buoyancy. The turbulent Prandtl numbers for k and ε are denoted as \( C_{tk} \) and \( C_{\varepsilon} \), respectively, and \( \gamma \) represents the kinematic viscosity. \( C_{k} \) and \( C_{\varepsilon} \) are constants while \( C_{\mu} \) is a coefficient which determines the influence of buoyancy on ε, and \( C_{1} \) is expressed as Equation (11):

\[
C_1 = \max \left[ 0.43, \frac{\eta}{\eta + 5} \right], \quad \eta = \frac{k}{\varepsilon}, \quad S = \sqrt{2S_{ij}S_{ij}}
\]

Based on the first law of thermodynamics, the energy equation is expressed in Equation (12):

\[
\frac{\partial}{\partial t} \left( \rho_q \varepsilon \right) + \nabla \cdot \left( \rho_q \varepsilon \mathbf{v} \right) = \rho_q \frac{\partial T}{\partial t} + \nabla \cdot \left( \mathbf{k}_{eff} \nabla T - \left( \mathbf{\tau}_{eff} \cdot \mathbf{v} \right) \right) + S_h
\]

The specific enthalpy \( h \) is presented in Equation (13), \( \mathbf{k}_{eff} \) is the effective thermal conductivity, \( \mathbf{\tau}_{eff} \) is the shear energy, and \( S_h \) is a source term.

\[
h = e + \frac{p}{\rho}
\]

where, \( e \) is the specific internal energy.

2.2. Boundary Conditions and Setup

The geometry was designed based on the water model in ref. [26], as shown in Figure 1, where gas injection occurs through two cell zones as a mass source, according to Equation (14) [30]. This represented the two side injections in the physical model, where the axial momentum of the jet is almost completely dissipated, and buoyancy drives the jet in the vertical direction. The volumes were positioned at some distance from the jet inlet, having their horizontal endpoint aligned with the average penetration depth measured in the physical model. This assumption was based on observations of the physical model,
Figure 2. The buoyant plume region starts at some distance from the wall and ends at, approximately, the jet penetration depth.

\[ \frac{m}{V} \]  \hspace{1cm} (14)

where, \( m \) is the mass flow rate and \( V \) is the volume of the cell zone.

Figure 1. Schematic illustration of numerical geometry, side and top views.

Figure 2. Image capturing the penetration depth of the physical water model. The red-dashed box represents the injection zone utilized in the numerical model.

A flat surface approach was used with degassing as boundary conditions and the walls were set to non-slip. The phase coupled SIMPLE algorithm (semi-implicit method for pressure-linked equations) was employed to solve the pressure velocity coupling, and the
spatial discretization utilized the second-order upwind for momentum, pressure, and first order turbulence, and user-defined scalars (UDS), with first-order implicit time integration. The key parameters for the system are presented in Table 3.

2.3. Fluent and Thermo-Calc Coupling Scheme

The model coupled ANSYS Fluent v2022 R2 [36] with Thermo-Calc v2021b [33] ("TC" from now on). This is achieved using the C programming language, with the aid of the application programming interfaces (API) present in ANSYS Fluent and TC. The API in Fluent is the user-defined function (UDF) and the API in TC is called TQ. The model uses Fluent for the heat and mass transfer in the domain and TC for the high temperature reactions. The TCS metal oxide database (TCOX10) [34] database was used for the current setup. TCOX is a thermodynamic database designed for slags and oxides, enabling the description of solid slag, liquid steel, and slag as phases with varying compositions separated by a miscibility gap. These phases are referred to as liquid slags, where, e.g., liquid #1 represents liquid slag and liquid #2 represents liquid slag, facilitating a continuous transition in composition from metallic to oxidic states in equilibrium. Thermo-Calc typically employs the minimization of Gibbs free energy method to compute multi-component phase equilibria, accommodating different phases such as gas, liquid, and slag. For the purpose of this study, the system was limited to the elements C, Fe, and O, thereby reducing computational time in TC by minimizing the number of chemical reactions and including the ones relevant to the decarburization process.

The process of solving for heat and mass transfer combined with high temperature reactions is described as follows: First, an assumption is made that there is a one-way coupling between the flow and the reactions, meaning that the reactions do not affect the flow pattern (velocity field). This means that an incompressible solver with a frozen flow field approximation can be used. At the end of each timestep, before calling TC, the pressure, temperature, and composition of elements are collected in cells that undergone reactions. These data are passed from Fluent to TC. The enthalpy of each phase is calculated separately using TC. Since it is likely that each phase is in a non-equilibrium state, a final equilibrium calculation is performed with all phases and the total enthalpy as equilibrium conditions to find the temperature and new phase compositions of the cell.

In this way, the composition per phase and temperature changes locally in the cell at the end of the timestep, and the mass transfer handles the macro kinetics, i.e., the transport of mass in and out of the reaction zone at the next timestep, Figure 3. Even though the values are updated directly at the end of each timestep, a reaction rate is needed for each cell. In the current work, this is introduced through a turbulence dependent reaction rate that sets a limit on how much of the cell mass can reach equilibrium during a timestep, which is discussed in the case section below. When addressing reactive multiphase flows, the preservation of mass is crucial. In the present work, the approach taken ensures that in TC, while heat and work may be exchanged across the system boundaries, the total mass and atoms of the system remains constant (control mass). In contrast, Fluent adopts an open system (control volume) approach, enabling the transfer of heat, work, and mass across the system boundaries, as depicted in Figure 4a. Note that each computational cell is treated as a separate system. The difference between these two approaches is that the control volume approach does not allow a change in system volume between equilibrium and
non-equilibrium states. This restriction can be handled by using two-way coupling where fluid properties and mass transfer between phases are made part of the CFD solver iterative process. However, this would increase the computational time dramatically, which is why the current model proposes a different approach. Here, constant density is used in each control volume, and the frozen flow field does not admit any changes in phase fractions in the CFD solution field. Conversely, in the control mass, the density and the phase fractions are allowed to vary when a new equilibrium state is calculated. Thus, a mass conservation of the elements in phases can be obtained by scaling the element composition for each phase when the element mass is updated in the CFD solver. This admits a state where the mass of a certain phase can increase or decrease, while the density is kept constant. A scale factor that is lower than one indicates that the density (or pressure) likely would be lower for the phase in question, while a value higher than one indicates a higher density (or pressure). The density and phase fraction are kept constant so the CFD solver will not experience a change in the flow field due to the reactions (i.e., a “one-way coupling”). This is a practical approach that conserves the mass of each element while allowing the use of a fully incompressible solver. Figure 4b illustrates the concept of keeping the density constant during the reactive process.

Figure 3. Flowchart of the fluid transport and chemical reactions coupling scheme.

Figure 4. (a) Illustration of the difference between the control volume and control mass approach; (b) illustration of the mass conservation with constant density where total mass is conserved.
2.4. Numerical Procedure

2.4.1. Water Model

Prior to adding chemical reactions, the water model simulation was configured to progress using a transient formulation over a duration of 50 s, employing a timestep size of 0.01 s. Subsequently, a frozen flow field was adopted, and the concentration of an inert scalar was tracked to monitor the mixing time. To examine if the frozen flow field was at a quasi-steady state, this process was repeated three times, with each iteration continuing with a 10 s increment in the transient flow field simulation. The mixing time was determined using a frozen flow field at specific time intervals of 50, 60, and 70 s during the transient flow.

2.4.2. Mixing Time

The mixing time represents a characteristic period dictating the duration required for a change to propagate throughout the majority of a given process. Specifically, the mixing time is defined based on a predetermined criterion that indicates the achievement of a highly homogeneous state. In this study, the criterion for determining the mixing time is set at 95% total homogenization, denoting the time necessary to reduce concentration differences to within ±5% of the mean concentration. The simulation employed a user-defined scalar (UDS) model, which solved Equation (15) [36], in order to calculate the mixing time:

\[
\frac{\partial}{\partial t} (a_q \rho_q \phi_q^i) + \nabla \cdot \left( a_q \rho_q \phi_q^i \nabla \phi_q^i - a_q \Gamma_q^{i} \nabla \phi_q^i \right) = 0, \quad i = 1, \ldots, N
\]  

(15)

Here, the arbitrary scalar \( i \) in phase \( q \) is represented by \( \phi_q^i \), while \( \Gamma_q^{i} \) is the diffusion coefficient in turbulent flows determined by Equation (16):

\[
\Gamma = \rho D_m + \frac{\mu_t}{Sc_t}
\]  

(16)

where, the molecular mass diffusivity is \( D_m \) and \( SC_t \) is the turbulent Schmidt number (0.7) which is expressed as \( \frac{\mu_t}{\rho D_m} \) with \( D_t \) being turbulent diffusivity.

Nine different tracers with the same fluid properties as the water were injected into different positions in the domain, see Figure 5, in order to observe the effect of injection position on the mixing time. The mixing time of each tracer was monitored with a volumetric method which was implemented by tracking the standard deviation of the mass-averaged tracer concentration using Equation (17):

\[
\sigma_{std} = \sqrt{\frac{1}{N} \sum_{i=1}^{N} \left( \frac{x_i - x_{\infty}}{x_{\infty}} \times 100 \right)^2}
\]  

(17)

Here, the number of cells is denoted as \( N \); \( x_i \) and \( x_{\infty} \) are the concentration of tracer in the cell and the mean concentration, respectively.

Under the assumption of a normal distribution of tracer concentration within the domain, on the one hand, one standard deviation captures the homogenization degree (±5%) within approximately 67% of the cells. On the other hand, two standard deviations capture 95% of the cells, serving as an appropriate representation of the mixing time in this case. Thus, the mixing time was determined as the point when the relative standard deviation of the tracer in the domain dropped below 2.5%.

This mixing time was measured in 3 different butterfly-structured meshes, of sizes: 16,562 (coarse), 43,956 (medium), and 76,545 (fine) The mesh sensitivity was studied using the mixing time of all tracer injections in the fine, medium, and coarse meshes.
2.4.3. Theoretical Model Cases

A metal reactor model was developed using the medium mesh of the water model. In this setup, the model was modified to simulate the physical properties of liquid steel. The objective of this model was to obtain a solution for decarburization in a hypothetical metal reactor, where the energy equation was solved as well as accounting for chemical reactions. The key system parameters can be found in Table 4.

Table 4. Key parameters of metal reactor simulation.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Nomenclature</th>
<th>Liquid Steel (l)</th>
<th>Gas (g)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Density</td>
<td>$\rho$ (kg m$^{-3}$)</td>
<td>7000</td>
<td>1.3</td>
</tr>
<tr>
<td>Dynamic viscosity</td>
<td>$\mu$ (kg m$^{-1}$ s$^{-1}$)</td>
<td>$6.3 \times 10^{-3}$</td>
<td>$1.8 \times 10^{-3}$</td>
</tr>
<tr>
<td>Mass flow rate per</td>
<td>$m$ (kg s$^{-1}$)</td>
<td>-</td>
<td>0.0052</td>
</tr>
<tr>
<td>inlet</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Bubble diameter</td>
<td>$d_b$ (m)</td>
<td>-</td>
<td>0.01</td>
</tr>
<tr>
<td>Degassing pressure</td>
<td>$p$ (Pa)</td>
<td>101,325</td>
<td>101,325</td>
</tr>
<tr>
<td>Specific heat</td>
<td>$c_p$ (J kg$^{-1}$ K$^{-1}$)</td>
<td>800</td>
<td>1006</td>
</tr>
<tr>
<td>Thermal conductivity</td>
<td>$k_c$ (W m$^{-1}$ K$^{-1}$)</td>
<td>64</td>
<td>0.0242</td>
</tr>
</tbody>
</table>

The measurement of mixing time was conducted following the above-mentioned procedure, wherein a fixed flow field was considered after a transient flow of 70 s, accompanied by the injection of tracers at nine distinct points. However, to account for the combined influence of advection and diffusion on the mixing process [35], two measurements were performed. In the first, an effective diffusion coefficient was applied to all tracers in accordance with Equation (16), while in the second, only a laminar diffusion coefficient was assumed by assigning a constant of $1 \times 10^{-9}$ kg m$^{-1}$ s$^{-1}$. The latter assumes a negligible diffusion contribution to the mixing. This enabled an examination of the mixing impact on the chemical reactions, while maintaining consistent conditions for mass flow rate, which otherwise certainly would impact the decarburization.

Further, the coupling to TC was introduced to the frozen flow field with adiabatic boundaries and UDS to transport each element. The thermodynamic system consisted of liquid steel with C, Fe, and O in all of the below mentioned cases and with oxygen as process gas supplied into the system. The process gas assumed the steel temperature instantaneously, and the reactions took place if the gas volume fraction was between 0.1–0.9
in the computational cell. Table 5 presents the initial composition and temperature of the following cases and simulations. A workstation, equipped with two AMD EPYC 7301 processors featuring 16 cores each, and a combined RAM capacity of 128 GB, computed all cases with 10 min of real time for each case, employing a timestep of 0.1 s. Three simulations were conducted for all cases with the first (a), setting the reference, the second (b), using only a laminar mass diffusion coefficient (i.e., no turbulent diffusion) in order to artificially increase the mixing time, and the third (c), changing the initial oxygen mass fraction. All the simulations assumed that solid oxides formed during reactions would be inert after formation and would, therefore, not participate in any subsequent reactions. This assumption was made to reduce the number of transport equations to be solved due to the extremely low amounts of solid oxides forming in the current process (Figure 6).

### Table 5. Initial composition and temperature of cases in conducted simulations.

<table>
<thead>
<tr>
<th>Test</th>
<th>W (C)</th>
<th>W (Fe)</th>
<th>W (O)</th>
<th>Temperature (K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>a</td>
<td>0.001</td>
<td>0.9987</td>
<td>0.0003</td>
<td>1793</td>
</tr>
<tr>
<td>b</td>
<td>0.001</td>
<td>0.9987</td>
<td>0.0003</td>
<td>1793</td>
</tr>
<tr>
<td>c</td>
<td>0.001</td>
<td>0.9989</td>
<td>0.0001</td>
<td>1793</td>
</tr>
</tbody>
</table>

![Schematic illustration of turbulent reaction rate treatment in coupled model](image)

**Figure 6.** Schematic illustration of turbulent reaction rate treatment in coupled model, where \( m \) denotes the mass and \( T \) is the temperature in the cell.

**Case 1—Simplified Oxygen Injection, No Transport of Products**

The first case was created to investigate how a simplified oxygen injection model would affect the results. The equilibrium reaction frequency was set as the inverse of the gas hold-up of the plume (0.6 s). The injected oxygen was distributed equally across all
cells participating in the reactions. This assumes that the oxygen could spread across the plume region without triggering any reactions until the plume was filled with the oxygen that entered the domain during the gas holdup time. The gaseous products (mainly CO) were assumed to leave the domain within the gas holdup time. Case 1 reduces the number of equilibrium calculations at the expense of not accounting for the full transport process taking place in the plume. It should be noted that the transport of elements and energy in the liquid melt took place at the regular CFD timestep.

Case 2—Oxygen Injection in the Jet Zone, No Transport of Products

The second case looked at another extreme, namely what happens if all oxygen reacts at the injection zone. The equilibrium reaction frequency was set to the inverse of the timestep. Similar to Case 1, there was no transport of gaseous products. The main difference compared to Case 1 was the higher reaction frequency and the higher average reaction pressure due to all reactions taking place in the injection zone.

Case 3—Oxygen Injection in the Jet Zone

In the third case, the reactions were limited to the injection zone, similar to Case 2, and the equilibrium reaction frequency was set to the inverse of the timestep. However, this time, the gaseous products were transported in the domain and were allowed to react freely with the liquid melt if they were in the domain. The cost of this added complexity substantially increased the number of cells in which reactions took place during a timestep, thereby increasing the simulation time accordingly.

Case 4—Oxygen Injection in the Jet Zone, Reactions Limited by Turbulence

The fourth case is similar to Case 3. However, in the previous cases, the only limitations to the reactions were the transportation of elements in and out of the reaction zone and the oxygen rate supply. The reaction rate in Case 3 was essentially infinite, where the entire amount of gas present in a cell would react with the entire amount of liquid in that cell. In Case 4, a reaction rate limited by turbulence was introduced by applying the Eddie dissipation concept (EDC) [30,36]. The concept is based on the energy transfer from large eddies to the smallest eddies referred to as fine structures in turbulent flow. The primary generation of turbulent kinetic energy occurs through the interactions between larger eddies and the mean flow. In this process, mechanical energy is transferred from the eddy structures to the fine structures. Most of the dissipation of kinetic energy into heat happens in these fine structures due to work done by molecular forces on the turbulent eddies. The EDC model assumes that the reactions occur in the fine scales occupied by the volume fraction which can be expressed by Equation (18), and the reacting species are assumed to react over a timescale expressed as Equation (19):

\[ \xi^{**} = C_{\xi} \left( \frac{\nu \varepsilon}{K^2} \right)^{\frac{1}{4}} \]

\[ \tau^{*} = C_{\tau} \left( \frac{\nu}{K} \right)^{\frac{1}{2}} \]

Here, \( C_{\xi} \) and \( C_{\tau} \) are the volume fraction constant (2.1337) and time scale constant (0.4082), respectively.

With Equations (18) and (19), it was possible to assume an equilibrium fraction within each cell over a timestep (\( t_{s} \)), according to Equation (20):

\[ f^{eq} = \xi^{**} \frac{t_{s}}{\tau^{*}} \]
As an example, if the equilibrium fraction were 0.5 in a cell, only half of the element masses would react in TC, the new phase composition would then blend with the unreacted mass in the computational cell, as illustrated in Figure 6.

3. Results and Discussion

The decarburization reaction was modeled in a side-blowing metal reactor with fluid transport and chemical reactions. The model was rather coarse to focus on a practical approach to coupled modeling with reasonable computational time. The mixing time measurements of three grid resolutions were compared to a physical water model, presented in Figure 7. It is clear from the figure that the average mixing time is converging to a value slightly below the experimental range. All simulations reached a quasi-steady state and showed no variation in mixing time at 50, 60, and 70 s. Instead, the vertical arrows, seen in Figure 7a, represent the interval on mixing times acquired from injecting the tracer in different positions, as depicted in Figure 7b (from Figure 5). These results suggest that the location of the injection point is important when determining the mixing time, as Points 1, 4, 5, 8, and 9 clearly show a faster mixing of the tracer. These injection points are located in the trajectory of the commonly reported [2-5,8,9] large circulation which appears in side-blowing models. In such side-blowing processes, the convective contribution to the mixing is very high, as seen in Figure 8, where a large portion of the bath is affected by strong currents. Similar results were also found in experiments carried out by Visuri et al. [7]. As discussed in ref [35], mixing refers to the process of achieving homogenization in a substance that is initially heterogeneous, accomplished through either bulk motion at a macroscopic level and or molecular diffusion at a microscopic level. However, when considering molecular diffusion alone, it is often inadequate for rapidly attaining a uniform mixture due to the limited speed of the process. Additionally, the small scales involved in mixing make it impractical to generate turbulent flow, which is commonly employed in macro-scale systems to achieve rapid mixing. With this in mind, it is suggested that the position of the tracer injection be located near the reaction zone of the process being modeled when measuring mixing time, due to the convective contribution varying across the domain. Also, note that the water model only used one tracer injection point, which approximately corresponded to Point 1 in Figure 5.

Apart from using a relatively coarse mesh, there are some other limitations in the comparison against the water model. The absence of quantitative velocity measurements in the physical model restricts the possibility of conducting a detailed comparative analysis. Also, the degassing surface fails to account for the wave-like motion of the free surface, thereby influencing the flow in the upper region of the bath. The lack of this free surface movement as well as the coarse mesh likely contribute to the quasi-steady state flow field arising in the simulation. Consequently, the physical model does not achieve the same quasi-steady flow field observed in the numerical model. However, to achieve a higher level of resemblance to the physical model would require the incorporation of a free surface and a finer mesh, and possibly a more advanced turbulence model such as the large eddy simulation (LES). It is worth noting that implementing these improvements would significantly increase the computational time, as observed in the study conducted by Haas et al. [28,29], where a single simulation took several months to complete. Given the focus of this current study, i.e., to develop a practical model, it was determined that the numerical model, which successfully generated a flow field consistent with expectations for side-blowing models as well as exhibiting a reasonable mixing time, was sufficient for the next step where a coupling to chemical reactions was added.
Figure 7. Mixing time comparison between the numerical model and the water model. (a) Shows the average value of mixing time based on all tracer injections which are displayed in (b).

Among the coarse, medium, and fine mesh, the error estimate, according to the maximum grid convergence index (GCI) [37], was approximately 13% for the medium mesh (GCI$_{132}$), which was deemed acceptable for the purposes of this study. The GCI measured at the most important plume injection point (Point 9 in Figure 5) had a value of 7%. Thus, the medium mesh was used for all subsequent simulations. Please refer to Table 6 for the GCI values related to the mixing time at all tracer injection points.
3.1. Theoretical Model Cases

Figure 9 and Table 7 present the mixing time differences between using the effective diffusion coefficient and using only the laminar diffusion coefficient. This generally has a large impact on mixing time. Similar to the water model simulations, there are variations in mixing times depending on the location of the tracer injection. However, when the turbulent diffusion contribution to mixing is removed, the convective contribution still has a large impact at Points 5 and 9. At these points, the mixing time only increases approximately 20 s (270%) compared to the maximum which increases approximately 170 s (1000%). Since Point 9 is in the plume, it will likely be the most representative mixing time, as the plume is the reaction zone, hence, the region of change in the process. This is important to remember when comparing test a to b.
Figure 9. Mixing times for different tracer injections with varying diffusion coefficients.

Table 7. Mixing time differences in the metal reactor with effective diffusion coefficient and (only) the laminar diffusion coefficient.

<table>
<thead>
<tr>
<th>Tracer</th>
<th>Effective Diffusion (s)</th>
<th>Laminar Diffusion (s)</th>
<th>$\Delta_{\text{mix}}$ (s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>9</td>
<td>123</td>
<td>114</td>
</tr>
<tr>
<td>2</td>
<td>14.4</td>
<td>175</td>
<td>160</td>
</tr>
<tr>
<td>3</td>
<td>14.4</td>
<td>180</td>
<td>165</td>
</tr>
<tr>
<td>4</td>
<td>8.3</td>
<td>104</td>
<td>96</td>
</tr>
<tr>
<td>5</td>
<td>10.4</td>
<td>29</td>
<td>19</td>
</tr>
<tr>
<td>6</td>
<td>15.9</td>
<td>186</td>
<td>170</td>
</tr>
<tr>
<td>7</td>
<td>15.9</td>
<td>185</td>
<td>169</td>
</tr>
<tr>
<td>8</td>
<td>10.2</td>
<td>70</td>
<td>60</td>
</tr>
<tr>
<td>9</td>
<td>9.6</td>
<td>32</td>
<td>22</td>
</tr>
<tr>
<td>Average</td>
<td>12 $\pm$ 3</td>
<td>120 $\pm$ 61</td>
<td></td>
</tr>
</tbody>
</table>

3.1.1. Comparison of Cases 1–4a

Table 8 presents the total simulation time for each case setup when the reactions were included. The choice of core configuration for each case was determined by the need to balance multiple simulations while optimizing computational resources. Given the computational intensity of the simulations, distributing the available cores across multiple cases allowed for more tests within a reasonable time frame. Among the cases considered, Cases 1, 3, and 4 involve calculations for an equal number of cells. However, there are distinct variations in their reaction mechanisms, notably, Case 1 triggers reactions only every six timesteps, while both Case 3 and Case 4 triggers reactions at every timestep. Consequently, one can anticipate an increase in simulation time for Cases 3 and 4 when restricted to eight computing cores. While Case 2, like Cases 3 and 4, initiates reactions at each timestep, its simulation time requirement remains comparatively shorter. This discrepancy can be attributed to the reduced number of cells that partake in reactions during each individual timestep. As the core count increases, the simulation time decreases, with the main limitation being the number of cells partaking in chemical reactions.
Table 8. The simulation time for calculations including chemical reactions for 600 s.

<table>
<thead>
<tr>
<th>Case</th>
<th>Time (h)</th>
<th>Cores</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>22</td>
<td>8</td>
</tr>
<tr>
<td>2</td>
<td>6</td>
<td>8</td>
</tr>
<tr>
<td>3</td>
<td>20</td>
<td>16</td>
</tr>
<tr>
<td>4</td>
<td>20</td>
<td>16</td>
</tr>
</tbody>
</table>

A comparative analysis of Case 1a through Case 4a, encompassing their respective reactions, is depicted in Figure 10, wherein the mass of all elements and the mean temperature across the domain are displayed. The results demonstrate noticeable differences among the cases. The initial observation, as depicted in Figure 10a, reveals a diminished decarburization rate in Case 2a as compared with the other cases. This difference can be attributed to the increased pressure at which all reactions occur, since all reactions only occur in the jet region. It is also seen that Case 1a exhibits the highest initial rate of decarburization, which should be due to the distribution of oxygen across the entire gas plume that, in turn, permits a lower pressure equilibrium for a larger portion of the reactions. The difference when comparing Case 1a to Cases 3a and 4a is not large, indicating that the simplification in Case 1a could be used for the current state of the process. Cases 3a and 4a are quite similar, indicating that the turbulence mixing limitation on the reactions does not pose a significant difference compared to perfect mixing in the cells, when it comes to decarburization reactions within the current composition range. Figure 10b,c show the iron and oxygen content in the melt. A decrease in iron content indicates that formation of solid oxides has occurred, while an increase in oxygen indicates that liquid oxides have formed and/or that the dissolved oxygen content in the melt has increased. It is seen that Case 2a leads to an earlier formation of liquid and solid oxides compared to the other cases. However, at the end of the simulation, the differences in iron content are very small between Cases 2a and 1a, and small between Cases 2a and 3a. Only Case 4a exhibits significantly lower iron losses at the end of the simulation. When observing the oxygen content at the end of the simulation, it is also seen that the differences are small among Cases 1–3a, and Case 4a has a lower oxygen content in the melt. The temperature, Figure 10d, follows the decarburization and oxide formation found in Figure 10a–c. It can be concluded that the turbulence-limited reactions of Case 4a lead to less solid and liquid iron oxide formation, while maintaining a similar carbon content as Cases 1a and 3a at the end of the simulation. Forcing all reactions to only occur at the injection zone (Case 2a) leads to the worst rate of decarburization and the highest iron losses. The differences are relatively small, though and the largest impact can be seen in the temperature evolution of the various cases.

In Cases 1a to 3a, where the equilibrium fraction \( \eta^G \) is set to unity for all cells, regardless of turbulence, the supplied oxygen within the cell is completely consumed during the equilibrium reaction. However, except for Case 2a, all cases enable reactions to occur throughout the entire plume, considering a pressure variance of approximately 30,000 Pa. This pressure difference has implications for the decarburization process, primarily because of the lower partial pressure of CO gas in proximity to the surface. In Case 1a, the injected oxygen is evenly distributed throughout the reaction zone. In contrast, Case 3a and Case 4a include the transport of reacted gas from the inlet to the surface of the bath. Consequently, Case 1a will facilitate a greater amount of oxygen to react at a lower partial pressure of CO, which explains the higher decarburization rate. While the carbon content decreases to lower levels, oxygen is utilized in the formation of iron oxide. However, even as the decarburization rate diminishes, there is still ongoing carbon oxidation near the upper region of the bath. This constitutes the primary distinction between Case 2a and the remaining cases, as illustrated in Figure 11, where a carbon concentration snapshot can be seen at the end of the simulation (equilibrium state at the end of a timestep). The pressure in the reaction zone clearly has an effect, which is to be expected. The impact of the turbulence limited reactions is evident in all graphs presented in Figure 10, particularly
when comparing Case 3a and Case 4a. It is observed that the decarburization rate in Case 4a is lower compared to Case 3a, while the formation of iron oxide occurs earlier (at around 250 s) and at a higher rate than in Case 4a (at around 280 s). Consequently, this leads to a reduced temperature evolution in Case 4a. These observations are in line with expectations, considering that the equilibrium fraction $\eta^{eq}$, due to turbulence, is set to unity for all cells in Case 3a, while it varies in Case 4a, as shown in Figure 12. Note that the difference between Cases 3a and 4a is not particularly large, which is easily explained since the equilibrium fraction is close to or higher than one in a majority of the gas plume. The lower part of the plume has a region where the equilibrium fraction is lower than one, which means that the reactions taking place at the highest ferro-static pressure do not have time to fully reach equilibrium within one timestep. This influences the formation of solid oxides and the temperature, as was described previously.

Figure 10. The graphs show the element and temperature evolution in the domain over time: (a) Carbon mass; (b) Iron mass; (c) Oxygen mass; (d) Temperature.
Figure 11. Iso-planes display the carbon content at the end of the simulation (660 s) in Cases 1a–4a. Note, there is no carbon oxidation in the above region of Case 2a, the gradient in the upper iso-planes is the effect of the flow spreading the reacted melt.

Figure 12. Iso-planes display the equilibrium fraction due to turbulence mixing in the cells for Case 4. The transparent black volume (plume shape) illustrates the reaction zone. Note that values higher than one are displayed as 1.
3.1.2. Test 2, Mixing Time Effect on Reactions in Cases 1–4

Figure 13 presents the mass of carbon and iron in Cases 1a, 2a, and 4a (including turbulent diffusion of mass) and Cases 1b, 2b, and 4b (only laminar diffusion of mass). It is seen that the increase in mixing time caused by removing turbulent diffusion does not affect the composition throughout the process, both for carbon (Figure 13a) and iron (Figure 13b). The removal of turbulent diffusion is an artificial way of increasing the mixing time, to observe the effects of such an increase without changing the gas flow rate in the system. As stated above, the reaction zone is the source of change in the process wherein the mixing time only increases approximately 20 s, when turbulent diffusion of mass is neglected (see Table 7). It should be noted that the mixing time increases by more than 270%. However, this increase only corresponds to about 4% of the total process time, which may be the reason for the low impact it has on the end results. Therefore, the current process is mainly limited by thermodynamics and the rate of gas injection. In other process stages (e.g., later decarburizing stages in an AOD), it is possible that a corresponding increase in mixing time might affect the end state. It is also likely that less intensely stirred systems such as ladles might experience a completely different response. In Figure 14, the local changes of carbon content over 7 s are displayed for Cases 4a, and 4b. The figure shows that the transport of the reacted melt across the domain is faster when turbulent diffusion is considered, providing local variations between the cases. However, as seen in Figure 15, the local variations seen in Figure 14 do not affect the carbon content as much. Naturally, the maximum/minimum values of Case 4b (with only laminar mass diffusion) are higher/lower compared to Case 4a where the higher mass diffusion leads to smoother gradients. Overall, the results, once again, point to a system that is governed by thermodynamics and the supply of oxygen rather than the mixing in the process. So, while an increase in mixing time of 270% might sound detrimental to the process, it is not certain that it will negatively affect the process. To put it in other words, investigations that only focus on mixing time may miss the target since it cannot be concluded that a lower mixing time is always beneficial to the process.

3.1.3. Test 3, Initial Oxygen Effect in Cases 1–4

The carbon mass is compared between Cases 1–4a and Cases 1–4c, wherein the initial oxygen content is decreased, as outlined in Table 5. Initial oxygen levels in the melt are seldom part of the sampling done at steel plants. The dissolved oxygen content in the melt may, or may not, be at an equilibrium state at the start of the process. This analysis looks at the effects of a variation in the initial oxygen content in the melt. Figure 16 reveals that the oxygen content in the melt plays a significant role in decarburization. Cases 1a, 3a, and 4a all show an increased rate of decarburization for the first 300 s compared to the lower initial oxygen content in the melt of Cases 1c, 3c, and 4c. Cases 2a and 2c do not show this trend. This distinction can be attributed to the reactions taking place in the plume, as the oxygen in the melt reacts with the ascending gas. Notably, there is a more pronounced difference observed in Case 3 and Case 4 compared to Case 1. This discrepancy arises from the fact that the injected oxygen primarily reacts at the inlet, resulting in the gas plume consisting mostly of CO in Case 3 and Case 4. Consequently, the oxygen in the melt contributes to a higher oxygen content within each cell and exerts a more significant influence on the reactions, since there will be more oxygen available higher up in the plume. Conversely, in Case 1, the injected oxygen is already distributed throughout the plume, thereby diminishing the contribution of the initial dissolved oxygen in the melt to the reactions. The cases all tend towards the same decarburization rate with increasing time as the effect of the initial oxygen content wears off. Regardless, it is important to note that measuring the oxygen content of the melt can be important to accurately predict the initial stage of decarburization. The different behavior in Case 2 can be described by the limited reaction zone that is only close to the nozzles, i.e., no additionally available oxygen at lower ferro-static pressure as in the other cases. Also, when considering Figure 10b,c, it is understood that Case 2a almost immediately forms liquid iron oxides. At a lower initial
oxygen content, there will be less liquid iron oxide formation, while approximately the same amount of oxygen is participating in the decarburization reactions, as can also be seen in Case 4 in Figure 17.

![Graphs showing changes in element mass over time](image)

**Figure 13.** The element mass in the domain over time using effective diffusion (Cases 1a, 2a, and 4a) as well as only laminar diffusion (Cases 1b, 2b, and 4b): (a) Carbon mass; (b) iron mass.
Figure 14. Four time frames between 0 and 7 s, showing the local changes of carbon content in Case 4a (effective diffusion) and 4b (laminar diffusion only).

Figure 15. Compares the maximum, average, and minimum carbon content in the melt over 20 s for Cases 4a (including turbulent diffusion) and 4b (laminar diffusion only).
Figure 16. The graphs show the mass of carbon in the domain over time for Cases 1–4 between Test a (higher initial oxygen content) and Test c: (a) Case 1; (b) Case 2; (c) Case 3; (d) Case 4.
Figure 17. The graphs show the mass of oxygen in the domain over time for Case 4 between Test a (higher initial oxygen content) and Test c.

At the current state, the model provides a practical approach to coupling between CFD and chemical reactions by sacrificing some accuracy in the flow and energy transport calculations. While this provided the possibility to investigate the impact of mixing time on the decarburization and added transparency to the system, further work is needed to better represent the complexity in the dynamics and mixing of such intensely stirred processes, e.g., including a free surface and applying a finer mesh. In addition, the reaction between metal and slag as well as the mass transport of solid slags needs to be investigated to judge if it is necessary to resolve those phases for more accurate description of industrial cases. For instance, chromium oxidation is unwanted in the AOD converter which is partially prevented by lowering the partial pressure of the CO gas to force decarburization instead. However, as seen in this work, at a lower pressure, oxidation of elements in the liquid such as iron is feasible. Thus, in an industrial AOD where the pressure is even higher at the injection zone, chromium will also oxidize. As these oxides are driven upwards to lower pressure regions, they continuously react with the gas and melt. This complex phenomenon can only be thoroughly studied when the model includes the transport of solid slag and the reactions between solid slag and melt. Cases 3 and 4, in particular, are interesting candidates for these model developments as they are expected to provide better insight into what happens if reaction kinetics in the plume has a larger influence on other process steps. Since the current model already utilizes parallel decomposition of the domain, an increase in resolution is possible and a straightforward next step. The other developments remain as opportunities for further studies.

4. Conclusions

A practical model that was capable of simulating fluid dynamics with chemical reactions in a side-feeding metallurgical reactor was developed to specifically study the effect of mixing time on decarburization. The model employed cell-local equilibrium with a turbulence limiter, while mass, momentum, and energy transfer were calculated using a finite volume method CFD approach. The mixing time and flow structure of the bath were validated against a water model, and then different reaction methods were investigated. Overall, the findings show the interplay between mixing, pressure, oxygen distribution, and turbulence in the decarburization process.

Specifically:
The location of the tracer injection is important when determining mixing time. Injection points located in the rising plume or in regions where the bath is affected by strong currents clearly showed a faster mixing of the tracer. Since reactions occur at the interface of the gas plume, it is recommended to use tracer injection in this region in all systems where reactions originate from this interaction, in order to get a representative mixing time.

The highest initial decarburization rate was found in the case where oxygen was distributed across the gas plume, which fostered a lower equilibrium pressure for reactions. The least favorable decarburization rate and the highest iron losses were exhibited in the case where all reactions were assumed to occur solely in the injection zone. A turbulence limiter for the reaction rate was also introduced which influenced decarburization, solid oxide formation, and temperature evolution.

The mixing time was artificially manipulated in one case by removing the turbulent mass diffusion contribution, leading to an increased mixing time. Surprisingly, this had minimal influence on the carbon and iron compositions throughout the process. Despite an increase in mixing time of approximately 270%, it was clear that the dominant drivers of this system are thermodynamics and gas injection rates. The current process’s complexity is shaped more by thermodynamics and oxygen supply than by mixing time. To put it in other words, investigations that only focus on the mixing time may miss the target, since it cannot be concluded that a lower mixing time is always beneficial to the reactive process.

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**Conflicts of Interest:** The authors declare no conflict of interest.

**Nomenclature**

\[
\begin{align*}
\alpha & \quad \text{Volume fraction} \\
\mathbf{v} & \quad \text{Velocity vector} \\
\rho & \quad \text{Density} \\
m_{pt} & \quad \text{Mass transfer between phases} \\
p & \quad \text{Pressure} \\
\mathbf{g} & \quad \text{Gravitational acceleration} \\
K_{pt} & \quad \text{Momentum exchange coefficient} \\
\mu_{eff} & \quad \text{Effective viscosity} \\
\tau_p & \quad \text{Particulate relaxation time} \\
d_b & \quad \text{Bubble diameter} \\
A_i & \quad \text{Interfacial area} \\
C_{D} & \quad \text{Drag coefficient} \\
Re & \quad \text{Reynolds number} \\
\mu & \quad \text{Molecular viscosity} \\
\mu_t & \quad \text{Turbulent viscosity} \\
k & \quad \text{Turbulent kinetic energy}
\end{align*}
\]
<table>
<thead>
<tr>
<th>Symbol</th>
<th>Definition</th>
</tr>
</thead>
<tbody>
<tr>
<td>ε</td>
<td>Turbulent dissipation rate [m²·s⁻³]</td>
</tr>
<tr>
<td>σ_k</td>
<td>Turbulent Prandtl number for k [-]</td>
</tr>
<tr>
<td>σ_ε</td>
<td>Turbulent Prandtl number for ε [-]</td>
</tr>
<tr>
<td>ν</td>
<td>Kinematic viscosity [m²·s⁻¹]</td>
</tr>
<tr>
<td>h</td>
<td>Specific enthalpy [J·kg⁻¹]</td>
</tr>
<tr>
<td>k_eff</td>
<td>Effective thermal conductivity [W·m⁻¹·K⁻¹]</td>
</tr>
<tr>
<td>τ_eff</td>
<td>Shear energy [J]</td>
</tr>
<tr>
<td>e</td>
<td>Specific energy [J·kg⁻¹]</td>
</tr>
<tr>
<td>m</td>
<td>Mass flow rate [kg·s⁻¹]</td>
</tr>
<tr>
<td>V</td>
<td>Volume [m³]</td>
</tr>
<tr>
<td>ϕ_q</td>
<td>Scalar in phase q [-]</td>
</tr>
<tr>
<td>Γ</td>
<td>Diffusion coefficient [kg·m⁻³·s⁻¹]</td>
</tr>
<tr>
<td>D_m</td>
<td>Molecular mass diffusivity [m²·s⁻¹]</td>
</tr>
<tr>
<td>D_1</td>
<td>Turbulent diffusivity [m²·s⁻¹]</td>
</tr>
<tr>
<td>S C_l</td>
<td>Turbulent Schmidt number (0.7) [-]</td>
</tr>
<tr>
<td>χ^l</td>
<td>Volume fraction occupied by fine structures [-]</td>
</tr>
<tr>
<td>C_x</td>
<td>Volume fraction constant [-]</td>
</tr>
<tr>
<td>τ^l</td>
<td>Reaction time scale in fine structures [s]</td>
</tr>
<tr>
<td>C_r</td>
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<td>η^l</td>
<td>Equilibrium fraction [-]</td>
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<td>t_i</td>
<td>Timestep [s]</td>
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References


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