

Highly coupled multiphysics simulation of iron combustion: Application to the ASTM G124 test

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Abstract

In the context of pure oxygen storage and transportation, such as production facilities, spacecraft propulsion systems, and medical oxygen supply, severe accidents still occasionally occur. These incidents, including facility damage, storage tank explosions, and even the complete loss of spacecraft, remain rare but often have catastrophic consequences. They continue to occur despite the use of materials specifically selected and qualified through standardized flammability tests, such as ASTM G124-18. Consequently, improving the understanding of metallic combustion in oxygen-rich environments remains an important scientific and industrial challenge.

In collaboration with industrial partners from the oxygen industry, the present work proposes a numerical approach to investigate the combustion of pure iron and to improve the current understanding of the underlying physical mechanisms.

The study focuses on the development of a high-fidelity numerical simulation of the ASTM G124 test, in which a metallic rod is ignited by a laser and its regression rate is experimentally measured.

The main challenge lies in the strong coupling between the multiple physical phenomena required to build a predictive model. The combustion of iron in a pure oxygen atmosphere involves the simultaneous interaction of heat transfer, gas-phase flow, liquid iron and molten oxide hydrodynamics, species diffusion, and the formation and detachment of droplets composed of both liquid iron and oxide.

A two-dimensional axisymmetric model of a laser-ignited, self-sustained burning iron rod is proposed. A conventional multiphase thermo-hydrodynamic Eulerian formulation is coupled with two species transport equations describing the diffusion of oxygen and iron, from which the combustion heat release is evaluated. Attention is paid to the interaction between the different phases. Two coupled phase-field equations are introduced: the first distinguishes the gas phase from condensed materials (iron and oxide), while the second separates the oxide phase from molten iron. This approach enables the appropriate material properties to be assigned to each phase and provides an accurate localization of the combustion front.

Finally, the numerical predictions are compared with experimental observations. Because of the extreme conditions encountered during the ASTM G124 test, direct measurements of local temperature and velocity are not available. The validation therefore relies on comparing the experimentally observed and numerically predicted droplet detachment times.

Good agreement is obtained, demonstrating the ability of the proposed model to reproduce the main features of iron combustion under pure oxygen conditions.

Keywords: Heat transfers, multiphase fluid flows, Eulerian approach, metal combustion, species transport.

Introduction

If the principle is not obvious in the common sense, some metallic materials are able to burn in specific conditions of temperature and pressure. For instance, aluminum base powder is currently used to improve combustion energy of spacecraft propellant. In this case, the combustion occurs in gaseous phase, and this is the most studied case of metal combustion. Our configuration is slightly different and far less represented in the literature. Indeed, regarding combustion of iron base materials, in oxygen rich atmosphere, the “flame” is supposed to be in the liquid phase not in the gas [1], [2]. This particularity

makes the experimental studies harder and more qualitative than quantitative. So, accurate understanding of iron combustion is not really reached today. For these reasons, a numerical approach is necessary to improve physical behavior understanding and address combustion mechanism prediction.

The present study concerns the combustion of pure iron in pure oxygen. The community of metal flammability is mainly centered on experimental investigations and coupled multiphysics models are rare or inexistant, so this first study focuses on simplified configuration.

This study take place in the context of oxygen security mainly driven by space agencies (as NASA)

and oxygen supply industries (as AirLiquid). For transportation and storage of pure oxygen at high pressure, the most common material remains steel. As a first step in these investigations, the present study concerns only iron, and alloying elements will be added in the future.

Literature review

The literature of the combustion field is quite large, but the combustion of metallic materials is reduced and mainly based on experiment. The main topic is the classification of material thanks to ASTM G124-18 test case [3], or equivalent ISO and NASA, where a metal rod is ignited by pyrotechnic charge and regression rate is measured. This test is poorly reproducible but it was improved few years ago by laser ignition [4], [5]. Physically, the combustion is a sequence of three steps, solid oxidation, liquid ignition and propagation [6]. Neglecting one of these steps or adding experimental inputs can lead to large errors regarding prediction [7], [8]. In the previous works done in the community, some point appears to be sensitive, and mandatory to be included in a predictive simulation:

- Iron oxide as a distinct phase with specific material properties [1], [9], [10].
- Oxide nucleation has to be introduced in a simulation, diffusion only is not able to produce oxide [11], [12].
- Interface transfers are driving combustion.

Therefore, the model must be able to simulate:

- The three steps: solid oxidation, ignition and propagation.
- The Multiphysics coupling.
- Realistic material properties, different for each phase.
- Nucleation.
- Pressure as input parameter (not detailed here).

The present paper aims to detail how different physics are coupled in this complex case and how predictive it can be. So, the first step is a short description of the experimental context, the G124-18 test case. Then, the conservation equations will be described and discussed regarding the physical phenomena needed to be considered. After that, several numerical results will be discussed. Finally, the validity of the model and its limits will be summarized.

Experimental Set Up

As explained previously, the experimental context of this study is the standard test ASTM G124-18. This test built for material classification (burn/no burn tests) was improved by laser ignition to measure input energy and increase repeatability.

The model is built to simulate the laser test.

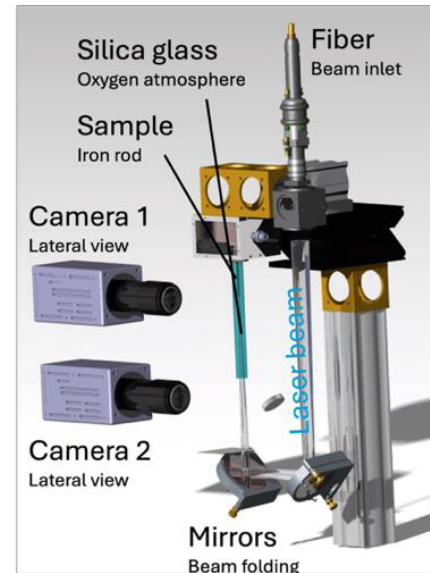


Figure 1. Conception view of the experimental setup.

In Figure 1, the sample is in silica glass where pure oxygen is flowing from the top to the bottom. The beam, coming from the top of the setup, is folded thanks to two mirrors to irradiate the rods from the bottom. Two high speed cameras, calibrated in temperature, are looking at the burning zone laterally and from the bottom.

For the oxygen pressure test, this setup is moved into a pressure chamber.

Numerical Model

Geometrical assumptions

As the sample and its environment (silica glass) are cylinders, authors assume axial symmetry in order to reduce the size of the problem. Moreover, the sample is small regarding the setup size, thus, only a part of the rod and the surrounding oxygen are simulated.

Figure 2 illustrates the selected geometry and the mesh. The model is composed by only one domain (the ABCD rectangle) and the metallic rod is considered thanks to the first Phase Field equation (color bar: 1 is the dense material, -1 is the gas). The mesh is set from a regular coarse quadrilateral mesh (200 μm on the right side) and refined one time (100 μm around the rod). Moreover, a remeshing process adapted to the Phase Field variable is done, reducing the element size to 50 μm close to the interface.

Governing equations

The specificity of this model is the number of physical phenomena occurring during the combustion.

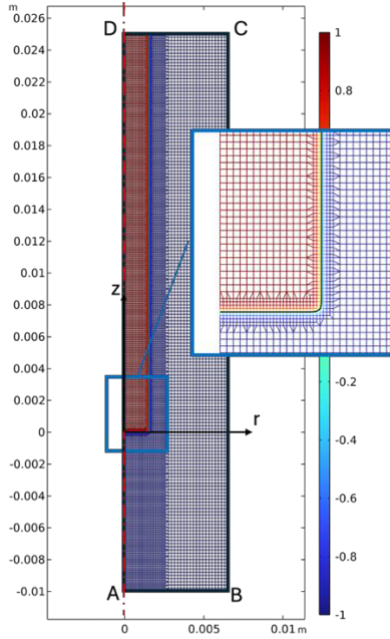


Figure 2: Geometry and mesh.

Remark: The rod is not related to a geometrical domain but analytically formulated as an input for the first phase field equation.

In few words, the laser melts the bottom side of the rod. At the melting point, the diffusion property of oxygen inside iron increases, a lot. Oxygen reacts with iron and produces oxide and heat. Once the laser beam is off, the heat generated by reaction compared to heat losses sustain, or not, the combustion.

So, the equations needed to be solved are:

- A first Phase Field to separate the dense material (the rod) from the gas and transport the interface during the melting and drop fall. **Variable Φ_1** .
 - The heat conservation to calculate the temperature and flux balance, leading to melting. **Variable T** .
 - The mass and momentum conservations, to consider flows in the melt zone and gas, drop formation and drop fall. **Variable $\vec{u}$ and p** .
 - A first species conservation of iron, to calculate the concentration of iron in the domain and particularly in the melt. **Variable c_{fe}** .
 - A second species conservation of oxygen, to calculate the concentration of oxygen. **Variable c_o** .
- The two later are used to simulate mixing and reaction between the two species in the liquid.
- A second Phase Field equation to simulate oxide phase and consider its non-miscible behavior with liquid iron. **Variable Φ_2** .

Thus, the numerical problem is the resolution of six coupled partial differential equations.

Phase Field 1

$$\frac{\partial \Phi_1}{\partial t} + \vec{u} \cdot \vec{\nabla} \Phi_1 = \vec{\nabla} \cdot (M \vec{\nabla} \mu)$$

Where $\vec{u}$ is coming from fluid flow problem.

With boundary conditions:

- [AB] outlet
- [BC] outlet
- [CD] wall
- [DA] symmetry

And initial condition:

$$\Phi_{ini} = -\tanh\left(\frac{r - r_{rod}}{\varepsilon_{int}\sqrt{2}}\right) \frac{\left(\tanh\left(\frac{z - z_{rod}}{\varepsilon_{int}\sqrt{2}}\right) + 1\right)}{2} - \frac{\left(-\tanh\left(\frac{z - z_{rod}}{\varepsilon_{int}\sqrt{2}}\right)\right)}{2}$$

This condition is the one illustrated on Figure 2 (+1 in rod and -1 in gas). It is an analytical formulation of the rod solid shape allowing full disconnection between domain, mesh and rod shape. It should be noted that the phase field approach is used here a numerical tool and is not yet connected to chemical energy of phases.

Heat transfers

$$\rho_\Phi c_{p,eq,\Phi} \frac{\partial T}{\partial t} + \rho_\Phi c_{p,eq,\Phi} (\vec{u} \cdot \vec{\nabla} T) - \vec{\nabla} \cdot (k_\Phi \vec{\nabla} T) = (\varphi_{las} + \varphi_{evap} + \varphi_{ray}) \delta \Phi + S_\Phi$$

Where $\vec{u}$ is coming from fluid flow problem.

The Φ index, indicates the dependency of thermal properties to the phase (for instance, gas properties when $\Phi_1 = -1$).

The source terms, usually applied as boundary conditions, are here applied in the bulk but only at the interface and distributed through the $\delta \Phi$ (phase field variable normal derivative).

These sources terms are respectively, the laser beam with $\varphi_{las} = -\frac{\alpha P_L}{\pi R_L^2} n_z (r < R_L)$ and losses, as radiation with $\varphi_{rad} = \varepsilon \sigma (T^4 - T_\infty^4)$ and latent heat of vaporization with $\varphi_{vap} = \dot{m} L_{vap}$ ($\dot{m}$ is the evaporation rate and L_{vap} the phase change energy). $S_\Phi = \Delta h_f R_{FeO}$ is the reaction energy, coming from the standard enthalpy of formation of FeO Δh_f and reaction rate R_{FeO} .

The boundary conditions are:

- [AB] out flowing ($-\lambda \frac{\partial T}{\partial n} = 0$)
- [BC] out flowing ($-\lambda \frac{\partial T}{\partial n} = 0$)

- [CD] thermal insulation ($-\lambda \frac{\partial T}{\partial n} = 0$)
- [DA] symmetry ($-\lambda \frac{\partial T}{\partial n} = 0$)

And initial condition is a fixed temperature $T(r, z, t = 0) = 293^\circ$

Fluid flows

Mass conservation:

$$\vec{\nabla} \cdot \vec{u} = 0$$

Momentum conservation:

$$\rho_\Phi \frac{\partial \vec{u}}{\partial t} + \rho_\Phi (\vec{u} \cdot \vec{\nabla}) \vec{u} = \vec{\nabla} \cdot [-p\vec{I} + \vec{\tau}] + \rho_\Phi \vec{g} (1 + \alpha_p (T - T_{fusion})) + \gamma(T) \kappa \delta \Phi$$

Here again, material properties are phase dependent (Φ index). The buoyancy volume force is applied in the bulk and interfacial effect as Laplace pressure is set in $\delta \Phi$.

As for the thermal problem, boundary conditions are related to gas mainly.

- [AB] outlet ($p = p_0$)
- [BC] outlet ($p = p_0$)
- [CD] wall ($\vec{u} = 0$)
- [DA] symmetry $\vec{u} \cdot \vec{n} = 0$

And initial condition: $\vec{u} = \vec{0}$ and $p = p_0$

Species transport

For oxygen

$$\frac{\partial c_O}{\partial t} + \vec{\nabla} \cdot D_O(T, \Phi) \vec{\nabla} c_O + \vec{u} \cdot \vec{\nabla} c_O + S_{interf} = \begin{cases} R_{FeO} & \text{if } \Phi > 0 \text{ (metal)} \\ 0 & \text{if } \Phi < 0 \text{ (gas)} \end{cases}$$

With S_{interf} to drive interface oxygen flux from [13].

For iron

$$\frac{\partial c_{Fe}}{\partial t} + \vec{\nabla} \cdot D_{Fe}(T, \Phi) \vec{\nabla} c_{Fe} + \vec{u} \cdot \vec{\nabla} c_{Fe} = \begin{cases} R_{FeO} & \text{if } \Phi > 0 \text{ (metal)} \\ 0 & \text{if } \Phi < 0 \text{ (gas)} \end{cases}$$

Where D_O and D_{Fe} are respectively diffusion coefficient of oxygen and iron in the different phases and materials (gas, solid iron, liquid iron and FeO oxide).

They are formulated as:

$$D_{O \rightarrow Fe}(T) (\Phi_1 > 0) (\Phi_2 < 0) + D_O(T, \Phi) = D_{O \rightarrow FeO}(T) (\Phi_1 > 0) (\Phi_2 > 0) + D_{O \rightarrow O_2}(T) (\Phi_1 < 0) (\Phi_2 < 0)$$

R_{FeO} is the reaction rate $= c_0 c_{Fe} A \exp\left(-\frac{E}{RT}\right)$ calculated from concentrations c_i for each species, activation energy E and temperature T .

Phase Field 2

$$\frac{\partial \Phi_2}{\partial t} + \vec{u} \cdot \vec{\nabla} \Phi_2 = \vec{\nabla} \cdot (M \vec{\nabla} \mu) + R_t$$

The second phase field equation aims to identify the oxide phase in order to set its own physical properties. So, in the oxide $\Phi_2 = +1$ and $\Phi_2 = -1$ everywhere else.

Moreover, the oxide (FeO) is not yet formed at the initial time step.

Boundary conditions are set as for Φ_1 even if the oxide will only interact with [DA].

- [AB] outlet
- [BC] outlet
- [CD] wall
- [DA] symmetry

The initial condition is a fixed value of $\Phi_2 = -1$, everywhere in the domain.

The most important term in this equation is the source R_t which leads to the creation or not of oxide, depending on the reaction rate.

$$R_t = \underbrace{c_0 c_{Fe} A \exp\left(-\frac{E}{RT}\right)}_{R_{FeO}} * (c_O > c_0^{max})$$

The condition $c_O > c_0^{max}$ is used to introduce the nucleation aspect. In the case of liquid pure iron, oxide can nucleate only if the concentration is higher than the solubility, here 2% in weight.

Remark: Material properties are summarized **Table 1** in appendix. Missing parameters are supposed to be known.

Numerical consideration

These six-conservation equations are solved on the previously described mesh which leads to almost 160 000 DDL.

A sequential resolution is built from the segregated solver:

1. Φ_1
2. T
3. $\vec{u}, p$
4. c_O, c_{Fe}
5. Φ_2

Different Direct solvers are used for each variable (PARDISO or MUMPS) and α -gen time scheme is used for time resolution (time step fixed at 1.10^{-4} s).

Simulation Results / Discussion

Results presented here are concentrations and temperature in the rod.

Thermal cycle

Looking firstly at the temperature, particularly at the maximum temperature in the rod with the time

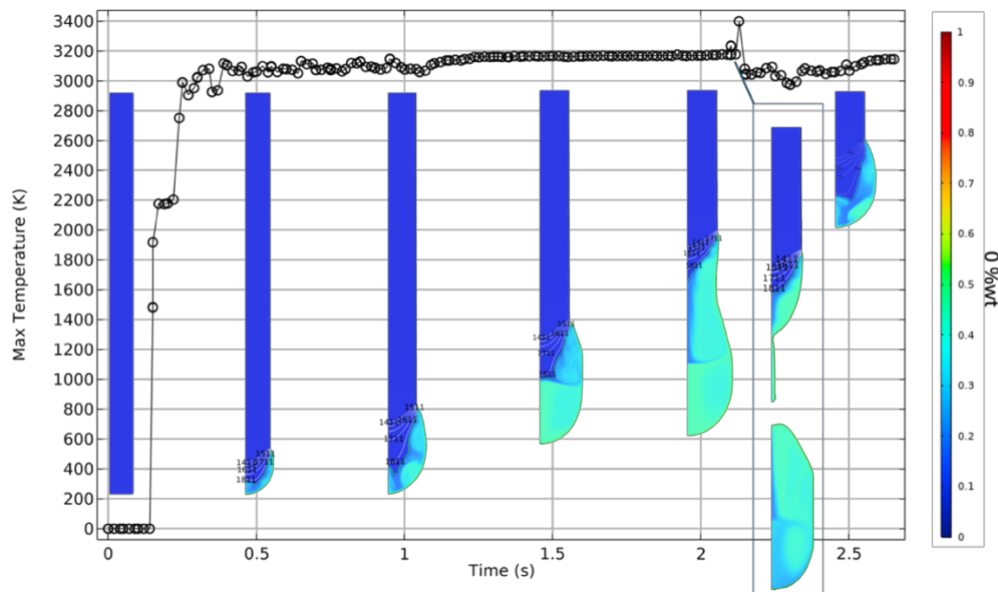


Figure 3 : Maximum temperature and Oxygen concentration field – 700W laser 1s

(Figure 3), one can observe the thermal cycle of a sustainable combustion. The case presented here corresponds to 700W laser heating during 1s. After the laser is switched off, the temperature continues growing indicating that the heat produced by the reaction is still melting the material and continues the oxidation process. At $t = 2.1$ s, in this case, the first drop falls, removing a certain quantity of energy leading to a temperature decrease. Nevertheless, the reaction occurring in the remaining rod is still energetic enough to increase temperature and continue the rod consumption.

Concentration field

Figure 4 illustrates the results at a time step corresponding to 2.1 s, when the first drop starts to fall. On this figure, one can see the activity of oxygen (% in weight), the contours of the FeO oxide ($\Phi_2 = 0$) in red and nucleation or oxidation zones (in purple).

Several interesting points must be noticed:

- After detachment, the drop seems to be fully oxidized.
 - The oxidation front (nucleation zone) is located at the interface liquid iron/oxide. This was supposed in the literature and appears to be validated here [14].
 - Oxygen content is quite homogenous in the liquid oxide, which means that the transport is more sensitive than species diffusion. This was also supposed in the literature and observed here [15].
- In terms of quantitative results, the concentration of oxygen in the liquid FeO is not constant, indeed, as an ionic mixture, the concentration is not

stoichiometric, and oxygen can be in supersaturation. The stoichiometric FeO corresponds to 22 %wt, and the supersaturation observed in literature [14] can reach 20 %wt more. Looking more closely at the oxygen content in the oxide (Figure 4 colormap), one can observe a maximum content close 40%wt (orange color) close to the interface O/FeO and a minimal content between 20 and 25%wt (medium blue) close to the interface Fe_l/FeO. Dark blue corresponds to the liquid iron unoxidized, so with a low oxygen concentration.

General consideration

An important result of the model is the geometry of the liquid phases, both iron and oxide. In Figure 5, two iso-values of temperature are plotted, the melting point " T_{fus} " and 100 K below the melting " -100 K". As the area between oxide boundary and T_{fus} is very thin, authors are interrogative about the theoretical value of the iron phase change temperature, perhaps not being realistic for high thermal dynamics. Showing the value 100 K below melting point illustrates the sensitivity of this assumption.

In Figure 5, a 3D extrapolation view is showing the whole domain, including iron (solid and liquid), oxide and gas. Regarding the gas part, the temperature (right side) increases a lot during the combustion, close to the liquid part, and seems to be affected by the external boundary (the glass tube). One can observe eddies on the thermal field (transport of the gas) and nothing regarding concentration (left side).

Remark: from a numerical point of view, using the Eulerian approach for species diffusion is not

optimal. It can produce some iron diffusion in the gas artificially. A penalization technic is used in this model to avoid losses of iron in gas.

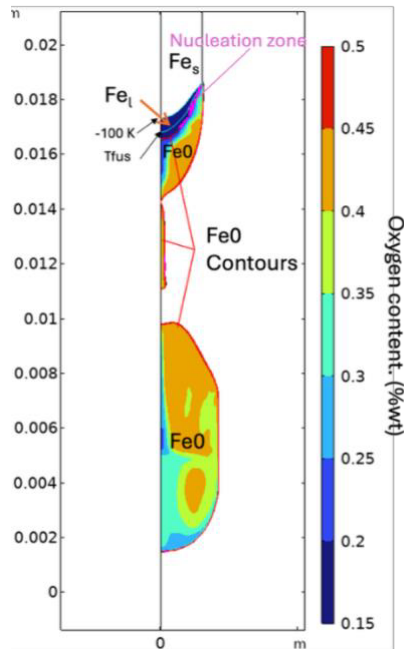


Figure 4: Concentration field of oxygen in the whole rod. Phase identification ($t=2.1s$).

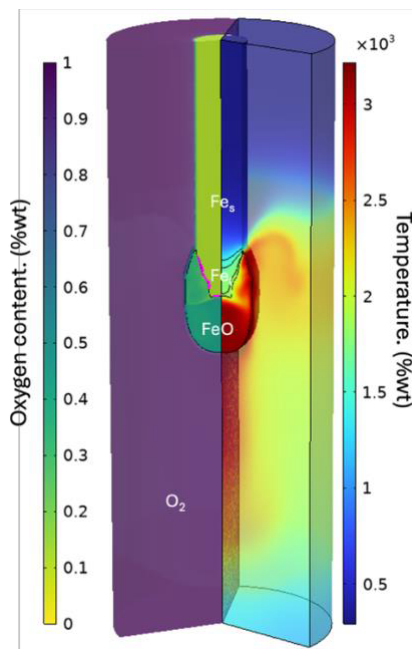


Figure 5: Oxygen content and temperature field in the whole geometry ($t = 1.5s$).

Figure 6 summarizes comparison between numerical results and experiments regarding times to produce the first drop. Despite noisy experimental data, the model seems to be nearly predictive in terms of oxide drops formation.

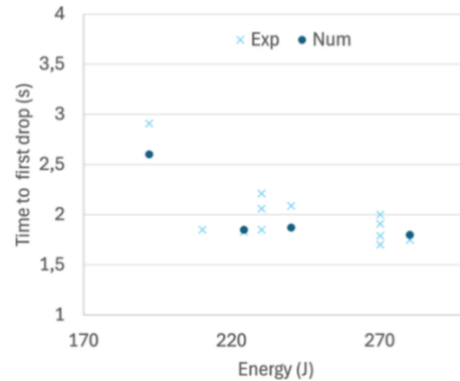


Figure 6: Comparison to experiments.

Conclusions

In this paper, authors have described a full model of iron combustion under oxygen rich atmosphere. The complexity was the coupling between the three physics (heat transfers, fluid flows and species diffusion) for two chemical species and with free boundary problems. A double Eulerian Phase Field approach was proposed to manage dense material/gas interface and FeO/iron interface. This approach gave realistic results, locally (species concentrations in oxide) and generally (time to first drop). The next steps will be an improvement of experimental validation (temperature, velocities, ...) and integration of alloy elements.

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Appendix

Table 1: Material properties

Gas: $\Phi_1 = -1$	Value
Density (ρ)	1.2. kg/m ³
Viscosity (μ)	1.10 ⁻⁵ Pa.s
Thermal conductivity (λ)	0.02 W/m/K
Specific Heat (c_p)	1000 J/kg/K
Iron: $\Phi_1 = +1, \Phi_2 = -1$	Value
Density (ρ)	[16]
Viscosity (μ)	[16]
Thermal conductivity (λ)	[16]
Specific Heat (c_p)	[16]
Heat of fusion (L_m)	250 kJ/kg
Heat of vaporization (L_{vap})	6400 kJ/kg
Melting point	1811 K
Surface tension (γ)	[16]
Thermal expansion (α_p)	[16]
Iron: $\Phi_1 = +1, \Phi_2 = +1$	Value
Density (ρ)	[17]
Viscosity (μ)	[18]
Thermal conductivity (λ)	[19]
Specific Heat (c_p)	[20]
Surface tension (γ)	[17]
Melting point	1650 K
Diffusion coefficients	Value
$D_{O \rightarrow Fe}$ solid phase	2.10 ⁻¹⁶ m ² /s

$D_{Fe \rightarrow Fe}$ solid phase	2.10 ⁻¹⁶ m ² /s
$D_{O \rightarrow Fe}$ liquid phase	3.2.10 ⁻⁷ m ² /s
$D_{Fe \rightarrow Fe}$ liquid phase	2.10 ⁻¹⁶ m ² /s
$D_{O \rightarrow FeO}$ liquid phase	3.2.10 ⁻⁸ m ² /s
$D_{Fe \rightarrow FeO}$ liquid phase	4.2.10 ⁻⁹ m ² /s
$D_{O \rightarrow O}$ gas phase	2.10 ⁻¹⁶ m ² /s
$D_{Fe \rightarrow O}$ gas phase	2.10 ⁻¹⁶ m ² /s
$D_{FeO \rightarrow O}$ gas phase	2.10 ⁻¹⁶ m ² /s