Development of a High-Fidelity Numerical Framework for Fully Resolving Combustion of Liquid Iron Droplets in Air

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Abstract
Direct combustion of metal powders in air can be utilized as a sustainable carbon dioxide emission free solution for energy storage and production. In particular, iron is considered a promising metallic fuel, due to its abundance and unique combustion properties. Nevertheless, proper design of iron-air combustors remains an outstanding engineering challenge that requires the development of advanced numerical simulation capabilities. In the current work, we present a unique numerical framework for fully resolving the combustion process of single and multiple iron liquid droplets in air. The suggested numerical framework is comprised of two main models: 1) an Immersed Boundary Method for fully resolved droplet-gas interactions, and 2) a zero-dimension numerical model for iron droplet combustion, which takes into account the underlying physical mechanisms, such as diffusion processes and reaction rates. First, we extensively validate each model against results from the literature. Then, we present a one-way coupled approach that integrates the two models together. Finally, we utilize the newly developed numerical framework for conducting detailed high-fidelity simulations of a reacting iron liquid droplet in air. The simulation results demonstrate the complex interactions between liquid iron droplets and the surrounding air during the combustion process.

Keywords
Metal Fuels, Iron Combustion, Computational Fluid Dynamics, Alternative Fuels, Energy Conversion and Storage.

Introduction
Energy and transportation systems are highly dependent on fossil fuels, which can be easily transported, stored, and combusted with air yielding extremely high-energy densities. However, one of the main fossil fuels' combustion products is carbon dioxide (CO_2), which is known today as a greenhouse gas responsible for climate change. Renewable energy sources, such as solar and wind power, can produce energy without any direct carbon footprint. Yet, these energy sources are inherently intermittent and create a mismatch between energy supply and demand. Thus, it is today well-established that energy storage is indispensable for effective utilization of renewable energy sources. Many existing storage methods, such as hydrogen or batteries, suffer from either low energy densities (per unit volume) and significant issues with reliable long-term storage[9]. It is therefore necessary to develop new methods for efficient energy storage and production.
Direct combustion of metal powders in air has been investigated for many decades[1], yet, it is usually overlooked as a potential renewable energy source[3]. Many metals have higher energy densities than gasoline, diesel, and various jet fuels[5]. Moreover, metal powders can be combusted in air and generate high power densities, whereas the main combustion products are metal oxides[3]. Thus, metal-air combustion produces zero emissions of carbon dioxide[4]. Metallic fuels can also be easily transported and stored almost indefinitely for long-durations using properly sealed containers[2] with comparable safety as common liquid hydrocarbon fuels[3]. It is therefore envisioned to establish the metal-fuel cycle[3]. Metallic fuels can be used for energy production and transportation applications via direct combustion. Then, the metal oxide products can be collected and regenerated into metallic fuels using renewable energy sources[23].

Any excess of metallic fuel as well as metallic waste from various industries[16] will be stored for future use. As a result, a closed sustainable cycle can be realized without any carbon footprint. Realization of a metal-air combustor heavily relies on the resulting combustion products. Typically, a metal dispersion system controls the amount of metal powder and air injected into the combustor, and the metal oxide products can be separated via a specially designed cyclone separator. This sort of separation is only possible for micron-sized particles[8] and for a combustion regime where the particle size after combustion remains almost unchanged.
Iron is the only metal that posses these combustion characteristics under stoichiometric and atmospheric air conditions[3]. Moreover, iron is the 4^{th} most abundant element in Earth's crust[22] and iron powder waste is a common industrial by-product[16]. It is also well-known that iron-air laminar flames have comparable flame thicknesses and temperatures as hydrocarbon-air laminar flames[3;13]. Thus, iron is a very promising candidate metallic fuel for direct combustion in air. Several experimental and theoretical studies dealt with iron powder ignition and combustion processes. Sun et al.[19] developed a simplified model to predict the combustion time of micron-sized iron droplets. Mi et al.[11] developed a mathematical model for prediction of solid iron particles' ignition temperature, whereas two main iron-oxide layers were considered. Recently, detailed experiments of solid iron particle ignition via a laser beam were conducted[10;13;14]. These experiments involved thermal imaging sensors for measuring the particle temperature during the combustion process.
It should be noted that any realistic combustion process involves a multiple number of droplets. Thus, even full understanding of the combustion processes for a single iron droplet is not sufficient for designing a realistic combustor. Moreover, it is well-established that collective effects can significantly alter the combustion characteristics in comparison with the case of a single droplet[17]. For instance, important design parameters, such as ignition temperature, combustion regime, maximum temperature and more are highly affected by droplet-gas and droplet-droplet interactions. For a suspension, different phenomena, such as iron droplet clustering and agglomeration, can completely change the resulting velocity field, temperature distribution, and oxygen concentration. Note that the impact of collective effects has been addressed by simplified models, see for instance[11]. However, as far as we know, there is no numerical framework that can fully resolve all the physical phenomena associated with multiple interacting iron droplets combustion in air. This sort of numerical frameworks, which allow high-fidelity simulations, are very common today for inert particles. For instance,[21] presented a methodology for investigating heat transfer processes of agglomerates by using Immersed Boundary Method techniques. In their work, agglomerates with varied particle densities were simulated with the relevant particle interaction physics. Then, based on the simulation results, simple correlations were constructed. For combustion of iron droplets, such high-fidelity simulations, which can provide vital fundamental understanding on the combustion characteristics, still do not exist. Accordingly, in the current work, we present a new high-fidelity one-way coupled approach for fully resolving the combustion process of liquid iron droplets.

High-Fidelity Model for Iron Combustion

In this section, we present the new high-fidelity fully resolved numerical framework for iron droplet combustion. Two independent models are described and validated in detail below: 1) A new compressible fluid dynamics solver with an Immersed Boundary Method approach, for modeling fully resolved gas-solid interactions, and 2) A zero dimension iron droplet combustion model. Then, we demonstrate a one-way coupling approach between the two models.

Immersed Boundary Method

The suggested numerical framework is implemented in our massively parallel compressible flow solver Athena-RFX++, which is an extension of the open-source code Athena++[18]. Athena-RFX++ is based on a finite-volume structured-grid fully compressible formulation with high-order time integration and spatial reconstruction methods. This code exhibits excellent perfor-

mance and scalability, and includes Adaptive Mesh Refinement (AMR), hybrid parallelization strategy, and almost full code vectorization. Thus, Athena-RFX++ can provide many numerical capabilities required for the proposed numerical framework. However, the main disadvantage of this code is a lack to handle complex geometries, such as fully resolved droplets. Nevertheless, it is known that unstructured-grid flow solvers are more computationally expensive in comparison with structured grid flow solvers[6]. In order to resolve this issue, and yet to preserve our current code advantages, we suggest to implement an Immersed Boundary Method (IBM) in the code. IBM allows the body surface to be treated as source terms or approximate boundary conditions. Thus, the body does not have to coincide with the grid. There are many variants of the IBM, see for instance[12;15]. One of the most prominent variants of the IBM is the "Ghost Cell Method"[20]. In this method, the body is treated as approximate boundary conditions forced by neighbouring grid cells. These cells are referred as "Ghost Cells". Since no meshing is required in IBM, it is relatively simple to fully resolve a multiple number of droplets simultaneously, including droplets' equations of motions and interactions with the surrounding gas[21].
Accordingly, we implement an IBM in Athena-RFX++ by enforcing boundary conditions at the cells that are within the solid zone (see the shaded zone in Fig. 1). The values at the fluid cells (denoted as "x" in Fig. 1) are interpolated to the position of the image point (red dot in Fig. 1). Then, the proper boundary condition at the the immersed boundary (yellow dot in Fig. 1) can be enforced at the solid ghost cells (denoted as "+" in Fig. 1).

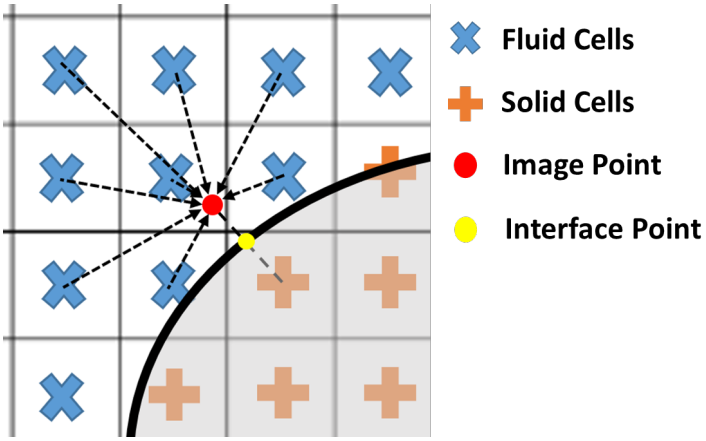

Figure 1. Schematic description of the IBM implementation.

For validating our new IBM implementation, we perform a numerical test that involves a low-Mach number flow. In this test, we solve the Naiver-Stokes equations for the problem of a freestream flow on a cylinder with a Reynolds number of 40 and a Mach number of 0.05. The domain is divided into 1200X600 grid cells with refinement near the cylinder and a domain size of 80DX40D, where D is the cylinder's diameter. Figure 2 presents a validation of our simulation results against experimental data from the literature, see[7].

Iron Liquid Droplet Combustion Model

For modeling the liquid iron droplet combustion process, we consider the droplet structure as comprised of three layers: An inner Fe core and two outer iron oxide layers (FeO and $\mathrm{Fe_3O_4}$). The main model assumptions:

- Thermal gradients within the droplet are considered negligible (low Biot number). Hence, the droplet's temperature distribution is assumed uniform.
- Constant densities of the oxides (no thermal expansion).
- Evaporation of the iron and the iron oxides is negligible due to their low vapor pressure.

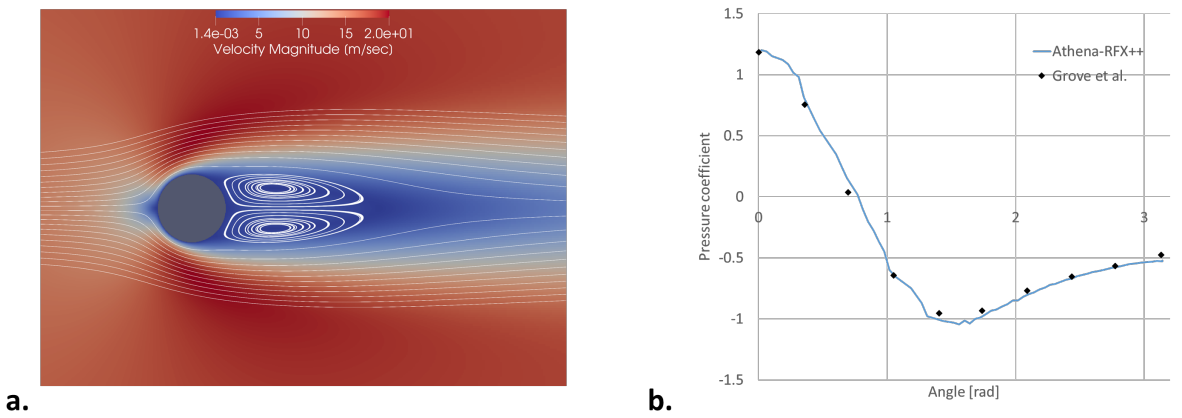

Figure 2. Validation of the IBM implementation - Flow on a cylinder with Re=40 and Ma=0.05. a. Velocity field of the flow. b. Comparison of the our simulation results against experimental data from [7].

- The droplet remains perfectly spherical.

In order to calculate the droplet's temperature as a function of time, an energy balance equation should be solved. Two main terms are considered, the first one is the heat release by chemical reactions, i.e. the transformation of Fe and oxygen into metal oxides. The second is the heat loss to the environment. The overall combustion process highly depends on both terms. For instance, the droplet's combustion characteristics, such as the maximum temperature and the total burn time are determined by this energy balance.

The heat release due to iron oxides' formation is controlled by three rate dependent mechanisms:

- Diffusion of oxygen from the ambient to the droplet surface through an external boundary layer.
- Diffusion of the Fe atoms and oxygen ions into the oxide layers.
- Chemical reaction of the oxygen ions and the Fe, which produces oxides (such as FeO and $\mathrm{Fe_3O_4}$) and heat.

In order to obtain the different species rate of formation, the three aforementioned mechanisms are combined into a single formula. After finding the total rates of the process, the mass rates of the oxides' formation can be calculated yielding the heat release, which is utilized in the energy balance. Then, the energy balance equation is solved numerically for each time step for obtaining the droplet temperature.
Figure 3 presents the thermal history of a 54 micrometer droplet. It can be seen that around 5 ms melting occurs ($\sim$1800K melting point iron) and after $\sim$15 ms, the Fe core is fully consumed by the chemical reactions. Validation is presented in Fig. 4 by comparing the droplet's maximum temperature predicted by our model against experimental results from [14] for atmospheric air. An excellent agreement is achieved between our model predictions and the literature.

Coupled Model

In this section, we present a high fidelity fully-resolved model for combustion of liquid iron droplets by integrating together the zero dimensional iron combustion model and the IBM framework in Athena-RFX++. As a first step, we suggest a one-way coupling approach between the flow field and the iron droplet combustion model, i.e. the droplet affects the gas temperature but the droplet's combustion process is not affected by the fully resolved gas. Also, for simplicity, we assume 2-D planar conditions.

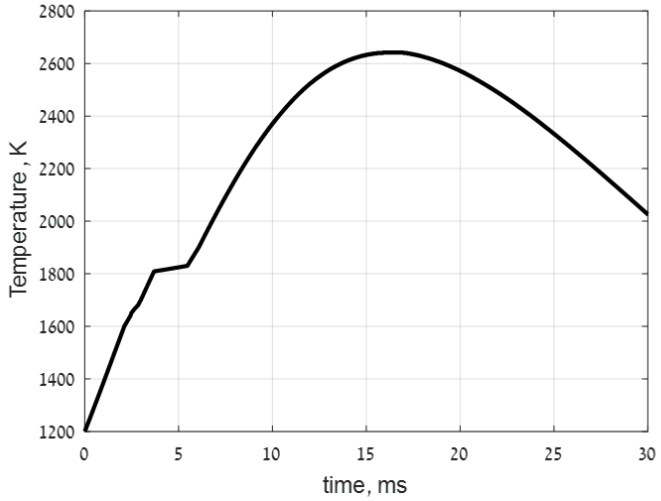

Figure 3. Thermal history according to our iron droplet combustion model.

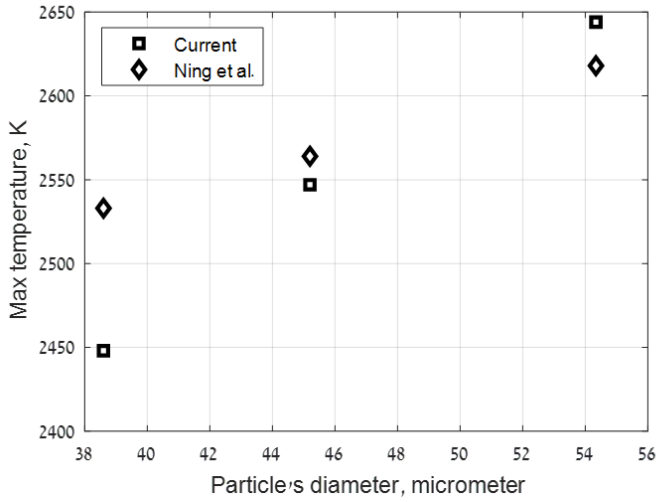

Figure 4. Maximum temperature during iron droplet combustion: Comparison between our model's predictions and experimental data from [14] for different initial droplet diameters under atmospheric air conditions.

Simulation results for the the coupled model are presented in Fig. 5. It can be seen that we can capture the spatial distribution of the surrounding gas temperature, which is heated due to the increase in the droplet's temperature during the combustion process.

Conclusions

In the current work, we presented a high-fidelity numerical framework for simulations of single and multiple liquid iron droplets combustion process. The framework is comprised of an Immersed Boundary Method coupled with a zero dimensional iron combustion model. Each model has been extensively validated against results from the literature. Finally, we demonstrated the one-way coupled numerical framework capability to fully resolve the combustion process of a single liquid iron droplet. Future work will involve inclusion of full droplet-gas two-way coupling, extension of the framework to three-dimensions, and addition of force balance equations for the droplets. The new numerical framework will be utilized for understanding the complex physical phenomena associated with combustion of multiple liquid iron droplets.

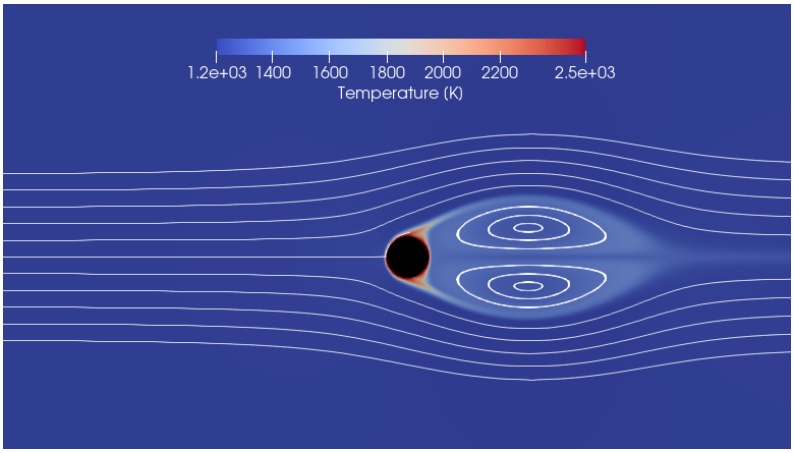

Figure 5. Demonstration of the coupled model approach: Temperature field of the gas surrounding the iron liquid droplet at 16 ms after the start of the combustion process. The droplet's diameter is 54 micrometer and the Re number is 300.

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