Low-temperature kinetics of the oxidation of iron powders

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Oral Presentation

Metal fuels, especially iron, show significant potential as a sustainable source for reducing carbon emissions in heat and electricity production. Despite this fact, a detailed understanding of the combustion of iron dust flames is currently missing. In this study, we conduct a thorough kinetic investigation of the oxidation of iron particles in the micrometer size-range. The kinetic study is based on thermogravimetric analysis (TGA), particle characterization, and various modeling approaches. An accurate understanding of the oxidation kinetics is of utmost importance, as it strongly influences dictates the combustion behavior, for example evidenced by the flame speed, prior to thermal runaway (Mich et al. 2023).

To offer a complete picture, a comprehensive TGA test campaign has been conducted, consisting of approximately 100 individual experiments. A few results of these tests are shown in Figure 1. In detail, the data set comprises:

- Isothermal tests with temperatures from 300 to 700 °C, with a particular focus on the 400 to 570 °C range, i.e. below the stability region of wüstite
- TGA tests with a linear increase in temperature ranging from 1 to 15 °C/min
- Stepwise increase of the temperature (300; 400; 500 °C)
- Variation of the gas flow rate and initial solid weight
- Variation of the oxygen concentration of the inlet gas flow
- Two types of iron powders

In addition, most of the tests have been conducted multiple times and two different TGA instruments were used to ensure reproducibility of the results. The first conclusion from the TGA analysis is that oxidation rates increase greatly with temperature, moderately with oxygen content, while particle diameter has a much smaller effect than current models predict at low temperatures. Complete oxidation can be achieved around 600 °C when the temperature is increased gradually. The reaction extent increases very slowly or plateaus after a short rapid initial stage in isothermal tests. It is suggested that the experiments initially deviate from isothermal conditions due to the self-generated heat of the samples and undergo partial sintering.

This series of TGA experiments lays the basis for the development of mathematical models of the oxidation process of single iron particles. The models assume that the oxidation rate is governed by the lattice diffusion of iron through a uniform oxide layer. The developed models use Wagner's theory of ionic diffusion and Tammann, Pilling and Bedworth's (TPB) theory of atomic diffusion to calculate the oxidation rate of the iron particle. The diffusion rate of species through the oxide layer depends on the geometric evolution of the particle during the oxidation process. Three possible geometric configurations have been considered: a spherically symmetric model with a constant contact surface between the iron and the oxide, a spherically symmetric shrinking core model, and an asymmetric shrinking core model. All these models are combined with particle size distributions corresponding to the two powders used in the experiments. The modeling results show improvement over an existing single particle model in two respects: the oxidation process is spanned over a wider temperature range and the gradual smoothening of the conversion curves as the oxidation is completed.

To gain further understanding of the oxidation process, structure characterization techniques (SEM imaging and XRD analysis) have been applied to the raw materials and to the oxidation products under following conditions: at 50% and 100% oxidation at 5 °C/min; at 2 hours after oxidation at 400 °C. The results show differences in the structure evolution under linear increase of temperature and after isothermal test, and also show the gradual growth of oxide layers – mostly magnetite, rest hematite – around iron metal cores.

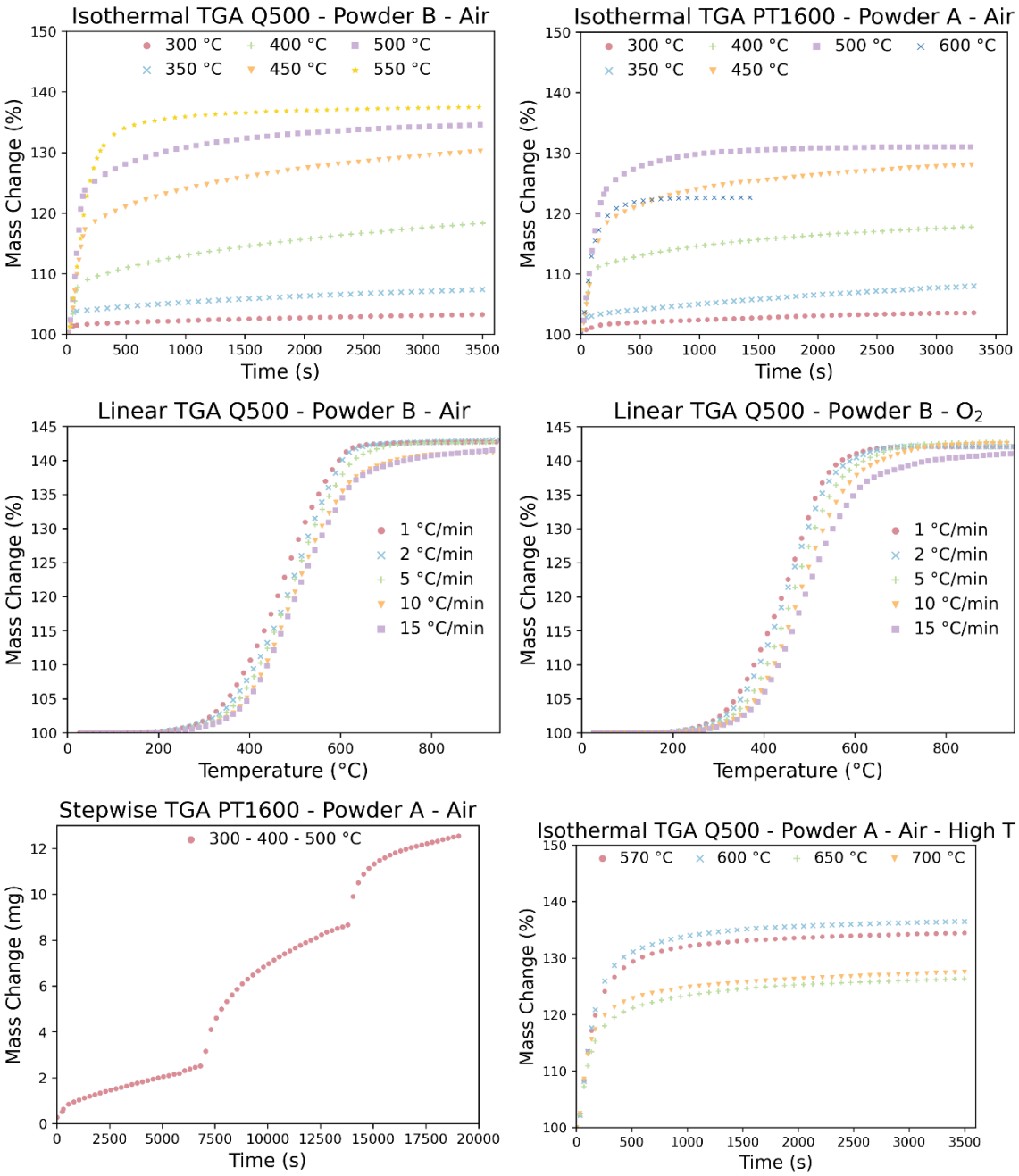

Figure 1: Thermogravimetric analysis of two types of iron using two different apparatuses. The tests were performed under various conditions as detailed in the text