

Thermodynamics and Kinetic Modelling of Flash Reduction Ironmaking

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Flash reduction ironmaking is being developed as an alternative pathway for producing DRI from iron ore fines. Producing DRI directly from fines eliminates the pelletising step. Flash reduction in ironmaking was pioneered by Sohn et al., where they used co-current flow of hydrogen at a temperature between 1200 to 1600°C with iron ore particle size between 20 to 53 µm. Calix is developing Zero Emission Steel Technology (ZESTY), a flash hydrogen direct reduction ironmaking process where iron ore powder is fed from the top and 100% H₂ is introduced from the bottom. The working temperature is between 800 to 1100 °C with iron ore particle size < 500 µm. The process employs a vertically oriented reactor with indirect electrical heating, enabling hydrogen to serve solely as a reductant rather than as fuel. The technology has carried out pilot tests with different Australian iron ores and is planning to build a demonstration plant of 30,000-tonne-per-annum in Western Australia. The highest metallisation level of DRI produced from ZESTY pilot test was 98%. Increasing temperature from 900 to 1050°C and H₂/O₂ reduction ratio from 1.2 to 2 has a positive effect on the level of metallisation. Fayalite (Fe₂SiO₄) formation was observed in DRI produced from goethite/hematite ores. Thermodynamic calculations were undertaken to investigate the formation behaviour of fayalite under ZESTY operating conditions and to assess how ore chemistry and selected additives (e.g. CaO, MgO) influence its stability. These studies support a broader investigation into the role of fayalite in flash hydrogen reduction and its relevance to both metallisation and downstream ironmaking processes. Results showed that adding 10 wt% of CaO to the iron ore reduces the amount of H₂ required to reach 100% metallisation level (up to 47%) as CaO can capture SiO₂ and preserve FeO for hydrogen reduction. In a separate strategy, calculations predicted that adding 6.25 vol.% CH₄ to H₂ increased metallisation level from 82% to 86%. Kinetic study of ZESTY at 950 °C showed that porosity of the ore and particle size played an important role in the reduction degree of all types of ores. Reduction by 100% H₂ was 3 times faster compared to 15% H₂. In a separate study of first order kinetic modelling of ZESTY showed that increasing temperature and hydrogen stoichiometric ratio enhanced the level of metallisation. Increasing particle size lowered the metallisation level through decreasing residence time. These findings are consistent with ZESTY pilot plant results.

KEYWORDS: THERMODYNAMICS; KINETIC MODELLING; FLASH REDUCTION; IRONMAKING; ZESTY.

INTRODUCTION

Currently, the ironmaking and steelmaking industries are going through the transition of decarbonisation to achieve its net zero goal by 2050. Hydrogen is widely regarded as one of the most promising clean energy sources because of its high calorific value, good thermal conductivity, and high reaction kinetics [1]. Though full application of H₂ is not currently economically and technologically viable, utilisation of H₂ shows the most promising results for decarbonisation [2]. There is also growing interest in developing technologies that directly use iron ore fines, such as fluidised bed technology [3],

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flash reduction technologies such as flash ironmaking technology (FIT) [4-7] developed by Sohn et al. and Zero Emissions Steel Technology (ZESTY), currently being developed by Calix in Australia [8-13]. These technologies do not require pelletising or sintering steps and can make iron in time scales of seconds to a few minutes. Following its commercialization in the 1950s, flash smelting technology has been widely utilized to produce copper and nickel due to its high efficiency and productivity [14]. The flash reduction concept was not introduced to ironmaking before the development of the FIT process. The FIT process utilises a co-current flow of hydrogen at a temperature between 1200 and 1600°C with iron ore particle size between 20 and 53 μm [6, 7, 15]. This process can use natural gas, hydrogen, or a mixture of both. It has been reported that this process can decrease the consumption of energy by 30 to 60% and reduce the emission of CO_2

by 60 to 96% compared to BF, depending on the usage of hydrogen [16]. On the other hand, ZESTY technology is based on Calix's proprietary Flash Calcination (CFC) technology. The process (as shown in figure 1) uses an indirect heating approach of the CFC technology in which iron ore fines (generally < 500 μm) are gravity-fed from the top of the reactor, with hydrogen introduced from the base in a counter-flow configuration. The iron ore fines are rapidly heated as they fall through the first few meters of the process and are subsequently reduced by the H_2 to a direct reduced iron (DRI) product. Further downstream, this DRI product can be smelted to remove gangue or can be made into briquettes [9, 10]. Since H_2 is only used as a reductant, it is expected to minimise the use of hydrogen in the process. According to Mokhtarani et al. [8], the residence time of ZESTY is between 45 seconds and about 2 minutes, varying with particle size and operating parameters.

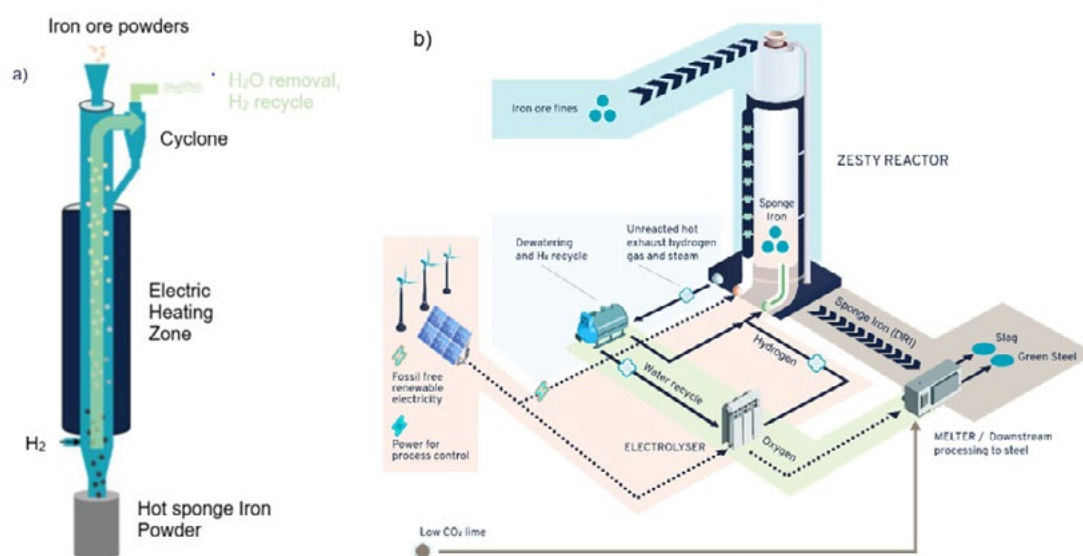


Fig.1 - (a) Schematic of the ZESTY reactor (b) Process flow diagram of the ZESTY process [9].

The present paper describes the current status of the technology, the findings from ZESTY's pilot testing, thermodynamic modelling on the formation of fayalite, kinetics of the ZESTY process, and future development.

PILOT PLANT RESULTS

In 2022, Calix carried out proof-of-concept campaigns with four different iron ores, including a siderite (FeCO_3) sourced from Texas, USA, two hematite goethite ($\text{Fe}_2\text{O}_3/\text{FeOOH}$) ores from Western Australia, and a magne-

tite ore (Fe_3O_4) from Western Australia [9]. The results showed that the siderite and goethite/hematite achieved metallisation levels of 91% (FC43-Siderite), 91% (HG57-goethite/hematite), and 81% (FC43-Siderite) at 950°C, whereas MA68 (magnetite) reached metallisation levels of 54% at 1050 °C. The lower metallisation level in magnetite ore was likely due to its dense structure, whereas the rest of the ores had porous structures (as shown in figure 2).

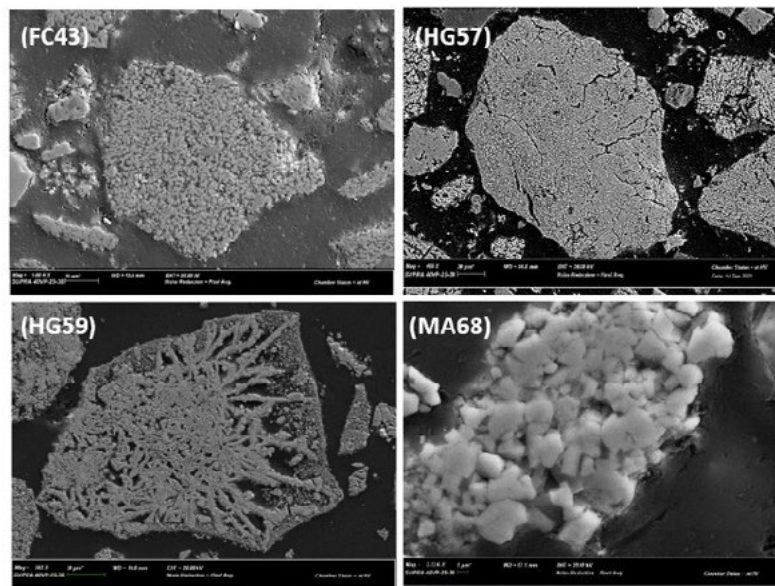


Fig.2 - SEM images of porous structure of FC43, HG57, HG59, while denser MA68 iron ore [9].

In 2024, Calix conducted a pilot plant testing campaign (over 100 runs) with six Australian-sourced iron ores. The pilot plant consists of a vertical reactor tube (ID = 0.2m, l = 18m) heated by 18 independently controlled electrical furnace zones. In the pilot study, trials were conducted in the range of 950 to 1050°C with iron ore at a feed rate between 60 and 200 kg/h in semi-continuous mode. H₂ was injected at the base of the reactor at stoichiometric ratios between 1.2 and 2 [13]. The details of the testing campaign are described in separate papers [12, 13].

Increasing temperature increased the metallisation level of all ores. Figure 3 shows a comparison among all the goethite hematite ores, including the proof-of-concept

ores. Metallisation level increased from 49% to 92% and 61% to 98% for ores HG57 and SC43, respectively, when the temperature increased from 750°C to 950°C. There is evidence indicating that higher temperatures increase metallisation; however, some variability is present in the data, making the trend less distinct, particularly when comparing results between 950 °C and 1050 °C. It is important to note that some degree of sintering occurred in the reactor at temperatures exceeding 1000 °C, potentially hindering reduction kinetics and resulting in a lower metallisation level for this scenario. Increasing the hydrogen stoichiometric ratio also increased the metallisation level [12].

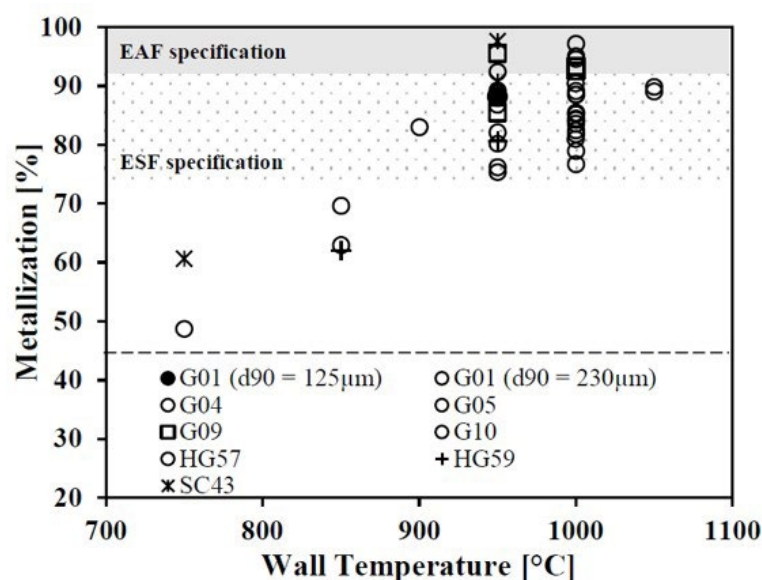


Fig.3 - Metallisation level of the DRI fines with changing temperature for goethite hematite ores [12].

Particle size has a significant effect on the level of metallisation. With increasing particle size, the level of metallisation generally decreases (as shown in figure 4). When the particle size decreased from 700 to 100 μm , the level of

metallisation increased. This result is consistent with the findings from the previous proof-of-concept campaign [10].

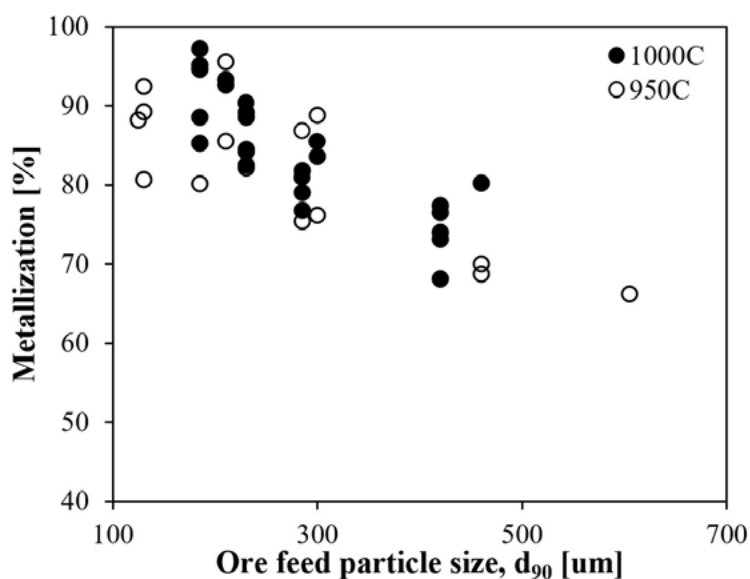


Fig.4 - Metallisation level with varying ore particle size [12].

In the pilot campaign, the two magnetite ores (M07, M12) and one pre-oxidised magnetite ore (OM07-pre-oxide M07) were tested. The maximum level of metallisation

achieved for magnetite ore was about 70% for the pre-oxidised ore. Further work is underway to optimise ZESTY for magnetite ores.

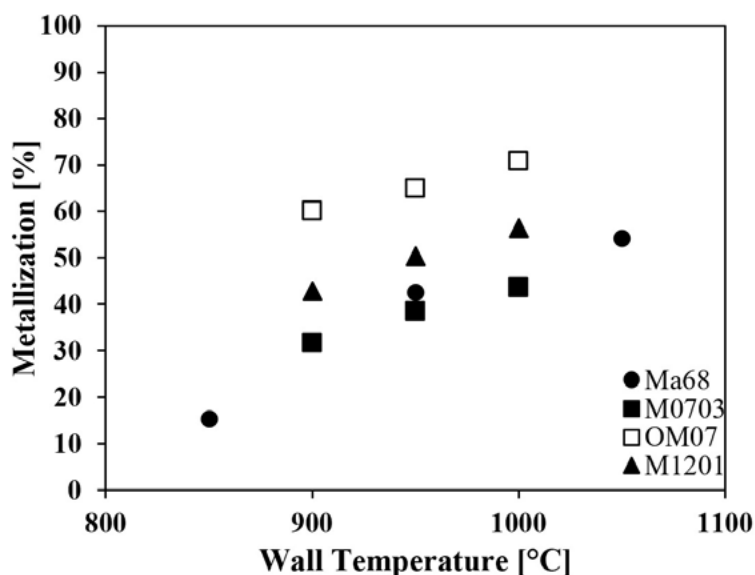


Fig.5 - Metallisation level of the DRI fines with changing temperature for magnetite ores [12].

Characterisation of the DRI fines derived from all goethite hematite ores showed formation of fayalite. Fayalite (Fe_2SiO_4) is the iron-rich member of the olivine group. Semi-quantitative analysis of the XRD results indicated that the

amount of fayalite formation ranged from 5% to 20%. The high fayalite results were associated with silica-rich ores, and further work is underway to limit its formation.

The accumulation of water vapour in recycled reducing gas is a common challenge in all hydrogen-based direct reduction processes. Increasing H_2O concentration decreases the $\text{H}_2/\text{H}_2\text{O}$ ratio and therefore lowers the reducing potential of the gas. However, due to the short residence times in flash reduction, the impact of water vapour accumulation in the recycle loop on reaction kinetics must be carefully assessed and managed. Currently, ZESTY operates with excess H_2 (stoichiometric ratio of 2) to keep the $\text{H}_2/\text{H}_2\text{O}$ ratio high, but further work is required to understand the trade-off.

THERMODYNAMIC MODELING

Thermodynamic modelling was used to explore techniques to increase metallisation levels and suppress fayalite formation. Calculations were carried out in FactSage

8.2®. The details of the modelling work are described elsewhere [17].

Figure 6 shows how the addition of CaO to a simulated ore changes metallisation levels at different reduction temperatures compared to a simulated ore without any CaO. The solid line indicates metallisation levels of a simulated ore by hydrogen with a composition of 90 wt.% FeO and 10 wt.% of SiO_2 at different reduction temperatures, whereas the dashed line shows metallisation levels of the same with 10 wt.% of CaO. Adding CaO improved the reduction of FeO-10wt.% SiO_2 mixtures, as it moves the lines to the left, which means a lower H_2/FeO ratio is sufficient to achieve higher metallisation. This can be attributed to the presence of CaO, which captures SiO_2 , and thereby preserves FeO for reduction by hydrogen [17].

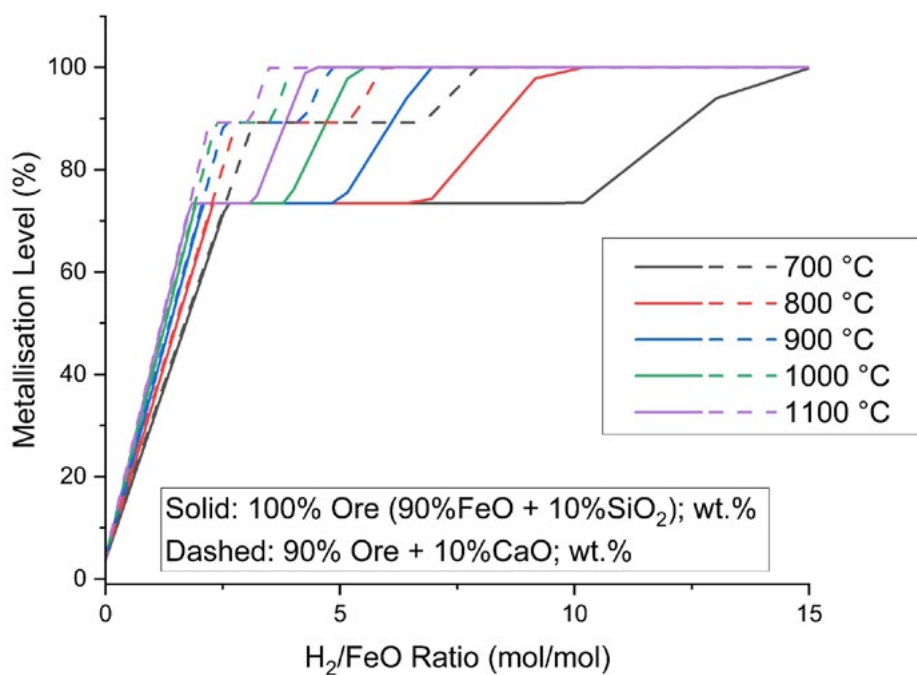


Fig.6 - Metallisation level of DRI vs H_2/FeO ratio at different temperatures [17].

In a different investigation on fayalite suppression, a portion of H_2 is substituted by CH_4 , and CO separately. The effect of this gas addition on the metallisation level and carbon content of the DRI is studied and shown in figure 7. The metallisation of the DRI increased from 82% to 87% when 6.25% of CH_4 was added to H_2 . The metallisation

level improves because the addition of CH_4 reduces Fe in the other oxide phases, including fayalite. Adding CH_4 also increased the carbon content of the product, as observed in figure 7. On the other hand, partially substituting H_2 with CO reduces the metallisation level, though the carbon content increased quite rapidly.

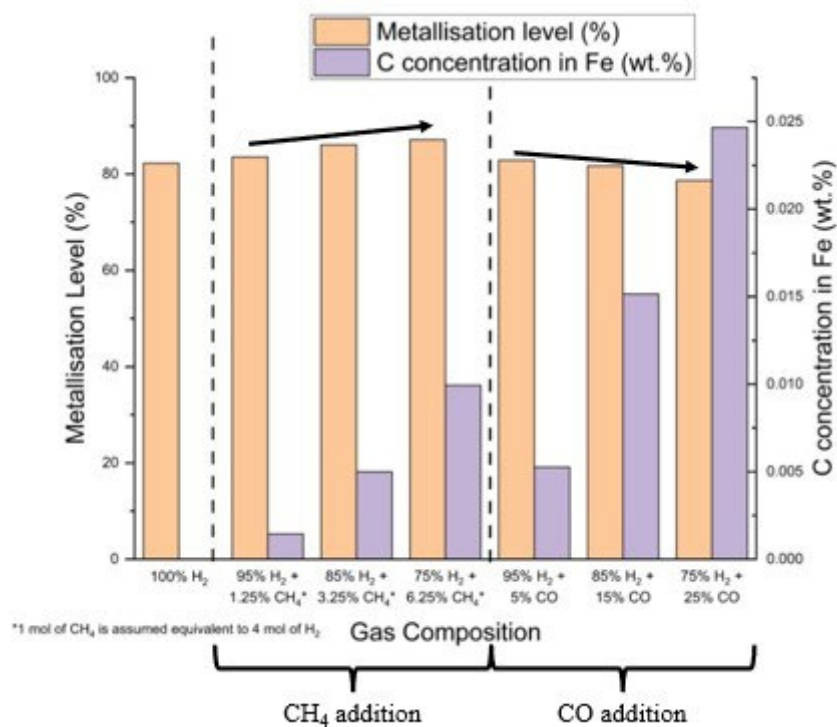


Fig.7 - The effect of the addition of CH₄ and CO separately to H₂ in the reduction process [17].

KINETIC STUDY

Nuraeni et al. [10] carried out a kinetic study of the ZESTY process in a TGA (Thermogravimetric Analysis) furnace

and studied the mass loss from a 0.100g sample. The study showed that the reduction by 100% H₂ was three times faster compared to 15% H₂ (as shown in figure 8).

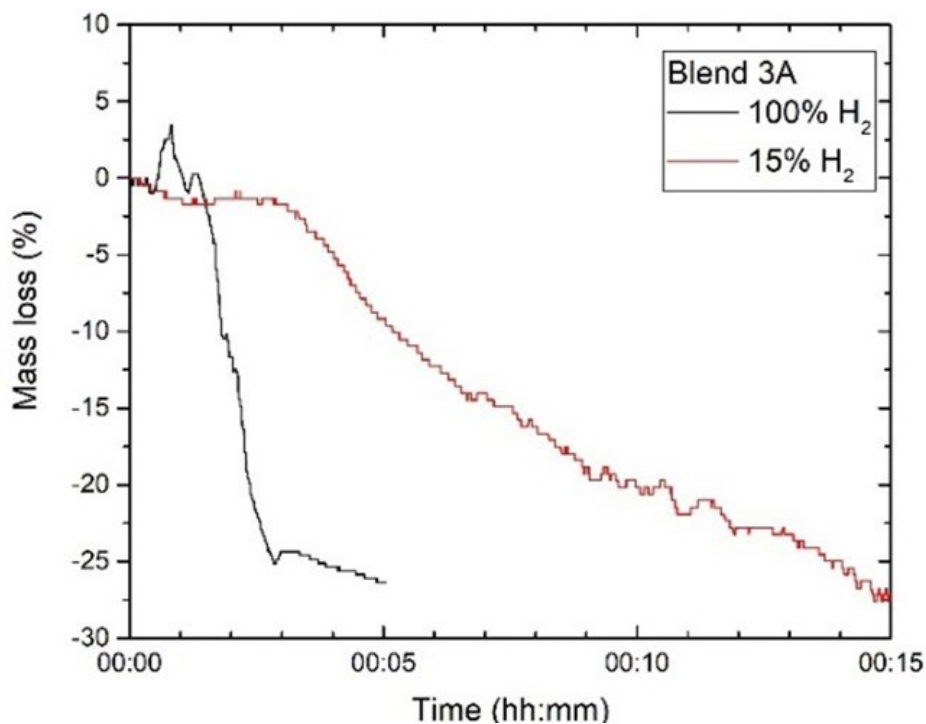


Fig.8 - Mass loss of Blend 3A/magnetite (125-300 μm) reduction by 100% H₂, 950 °C, 5 minutes [10].

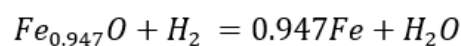
In another study by Mokhtarani et al. [8], a process model for flash reduction was developed for hematite ores. The modelling was carried out using Aspen Plus with two kinetic models: the modified Sohn and the mixed control models with a set of assumptions. The reactor was divided into 9 well-mixed zones, each 2 m long. Top and bottom zones were taken as pre-heating zones where no reactions occurred. The heat transfer in the seven reactor zones was assumed to be infinitely fast, so that the gases and particles were the same temperature as each other and as the wall. The particles were all at the mean diameter of the size distribution of the given sample. No fine particles were elutriated from the reactor. Hematite reduc-

tion was simplified as a single-step reaction from Fe_2O_3 to Fe, neglecting intermediate oxide phases. These simplifications were adopted to evaluate reactor-scale trends in metallization and residence time rather than provide a fully predictive description of the underlying multiphase and multistep reduction mechanisms. The Sohn kinetic model [18] is based on nucleation and growth kinetics. It incorporates the effect of temperature and partial pressure of H_2 and water vapor. The model has also been validated with experimental results. The reduction of FeO by hydrogen during the final stage of hematite reduction formed the basis for the development of the following empirical equation for hematite conversion [8]:

$$-\ln(1 - X) = 4.41 * 10^7 * e^{-\frac{214000}{RT}} * (p\text{H}_2 - \frac{p\text{H}_2\text{O}}{K})$$

where, X= fractional reduction degree, R=gas constant, T=temperature, p=partial pressure (atmospheric), t=res-

idence time (s), and K = equilibrium constant. The constants were calculated for the reaction below:



On the other hand, a mixed-control model incorporates both chemical reaction kinetics and diffusion resistance. A three-dimensional diffusion model (D3) together with the first-order reaction model (ROM1) is used for calculating the reduction of hematite. The details of the models are described in a separate paper [8]. The modelling results indicated that increasing the temperature and the hydrogen stoichiometric ratio improves the level of metallisation. Also, increasing particle size reduced the level of metallisation through decreasing the residence time. Ta-

ble 1 shows the comparison between simulated and experimental results. The findings from the model are consistent with the findings from the ZESTY pilot plant results [8]. It is important to note that the experimental results for FeO-110 and FeO-108 show variation from the general trend. The observed deviation may be attributed to differences in metallization calculations based on ICP-AES analytical results obtained from pilot-scale experiments.

Tab.1 - Comparison between results from the modified mixed kinetic model and experimental work at 950 °C and Hydrogen Stoichiometric Ratio of 2 [8].

Sample	$d_{av}(\mu\text{m})$	Residence time (s)	Metallisation level (%)	
			Model	Experiment
FeO-109	60	86	84	91
FeO-114	90	80	81.9	86
FeO-110	220	54	68.6	70
FeO-108	280	52	67.6	74

CONCLUSIONS

ZESTY's pilot plant results showed that temperature and hydrogen stoichiometric ratio positively affect metallisation level, whereas increasing particle size decreases metallisation level. The kinetic modelling by Mokhtarani et al. [8] showed similar findings to the pilot tests. Thermodynamic calculations showed that adding CH_4 to H_2 or adding 10% CaO to the high silica ore can suppress the formation of fayalite. These estimations need to be verified by further experimental work.

As part of its development, ZESTY has completed the Front-End Engineering Design (FEED) study for the 30,000-tonne-per-annum demonstration project. The main objective of the project was to examine and showcase the feasibility of a standalone, full-scale processing module [13, 19]. Calix is currently developing a demonstration facility capable of producing 30,000 tonnes per annum of hydrogen direct reduced iron (H_2 -DRI) or hot

briquetted iron (HBI), funded by an AUD 44.9 million grant from the Australian Renewable Energy Agency, with additional investment of more than AUD 35 million from Rio Tinto under a joint development agreement [20]. The key challenges and successes of this technology depend on its ability to adapt to the current high cost of H_2 , improved understanding of fayalite formation, optimisation for magnetite ores, innovative engineering solutions, and addressing briquetting requirements to meet existing shipping standards to enhance market flexibility.

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