Non-isothermal reaction of iron ore-coal mixtures

by

Theresa Coetsee

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Supervisor: Professor P.C. Pistorius

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Theresa Coetsee
Supervisor: Professor P.C. Pistorius
Department of Material Science and Metallurgical Engineering
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ABSTRACT

Extensive work is reported in literature on the reduction of iron oxides with carbonaceous reductants. Most of this work considered isothermal reaction of the material mixture, although as shown in some studies, isothermal reaction conditions are not often the norm because of sample size and heating arrangement in the experiment. In industrial processes, such as the rotary hearth type processes and the IFCON® process for iron ore reduction, the norm is non-isothermal reaction. Simulation of industrial processes should take non-isothermal reaction into account if the heat transfer effects within the process are to be investigated. To avoid the complications of coal volatiles in the experimental set-up, few studies were done with coal as reductant. The primary aim of the work presented here is to quantify radiation heat transfer to the surface of an iron ore and coal mixture heated uni-directionally from the sample surface to show the importance of heat transfer in the IFCON® process. Secondary aim of this work are to show the effects of layer thickness, coal volatiles, phase chemistry and particle size in this reaction system. The experimental set-up consists of a tube furnace modified to transport the sample into and out of the experimental tube furnace heating zone under a protected atmosphere, whilst the product gas is analysed throughout the experiment by quadropole mass spectrometer. The sample surface temperature, heating zone temperatures and material bed temperatures were measured throughout the experiment. A sample cutter-splitter was developed to divide the reacted sample into three horizontal segments for chemical analyses. The sample surface temperature and the heating zone temperatures were used as inputs to a radiation network calculation to quantify radiation heat transferred to the sample surface. The radiation network calculation was calibrated against heat-mass balance calculations for pre-reduced ore and graphite samples reacted at furnace temperatures of 1300, 1400 and 1500°C. The results show that radiative and conduction heat transfer control prevails for 16 mm to 40 mm material layers heated uni-directionally from the material layer surface. It is shown that coal volatiles contribute to reduction in the stagnant material layer. Also, smaller particle sizes result in increased reaction rates because of a decrease in the diffusion limited effects which were seen in reaction of the base size of coal and ore particles.

Keywords: heat transfer, uni-directionally, radiation network, radiation, conduction, coal, iron ore, temperature, furnace, material layer

Nie-isotermiese reaksie van ystererts-steenkool mengsels

deur
Theresa Coetsee
Promotor: Professor P.C. Pistorius
Departement Materiaalkunde en Metallurgiese Ingenieurswese
Philosophiae Doctor (Metallurgiese Ingenieurswese)

OPSOMMING

‘n Groot aantal studies in die literatuur handel oor die reduksie van ysteroksied met koolstof. In die meeste studies word verhitting van die mengsel beskou as isotermies. Sommige studies toon egter dat isotermiese verhitting selde plaasvind as gevolg van monstergrootte en verhittingsmetode soos aangewend in die eksperimentele opstelling. In industriële prosesse waarin ystererts gereduseer word, soos die roterende herd tipe prosesse en die IFCON® proses is nie-isotermiese reaksie die norm. In die simulasie van industriële prosesse behoort nie-isotermiese reaksie in oorweging geneem te word om die effek van hitteoordrag te ondersoek. Min studies is gedoen waarin steenkool as reduktant aangewend is omdat die teenwoordigheid van steenkoolvlugstowwe ’n komplekse opstelling vereis. Die primêre doelwit van hierdie studie is om stralingshitteoordrag na die monsteroppervlak van ‘n ystererts-steenkool mengsel, eendimensioneel verhit vanaf die monsteroppervlak, te kwantifiseer om daardeur aan te toon dat hitteoordrag belangrik is in die IFCON® proses. Die sekondêre doelwit van die studie is om die invloed van laagdikte, steenkoolvlugstowwe, fasechemie en partikelgrootte in hierdie reaksiesisteem aan te toon. Die eksperimentele opstelling bestaan uit ‘n buisoond wat aangepas is om die monster onder ‘n beskermende atmosfeer in en uit die warm sone van die oond te verplaas met deurlopende produkgasanalises deur middel van ‘n massaspektrometer. Die monsteroppervlak-temperatuur, verhittings sone temperature en temperture in die materiaallaag is deurlopend in die eksperiment gemeet. ‘n Monster snyer-verdeler is ontwikkel om die gereageerde monster in drie horisontale segmente te verdeel vir chemiese analises. Die monsteroppervlak-temperatuur en die verhittings sone temperature dien as insetparameters tot ‘n stralingsnetwerk berekening waarmee hitteoordrag na die monsteroppervlak bereken word. Die stralingsnetwerk berekening is gekalibreer teenoor die massa-hitte balans berekening vir voorgereduseerde ystererts-grafiet monsters gereageer teen 1300, 1400 en 1500°C. Die resultate toon dat stralings- en geleidings hitteoordragbeheer plaasvind vir materiaal laagdiktes van 16 mm tot 40 mm. Die resultate toon dat steenkoolvlugstowwe bydra tot reduksie in ‘n stagnante ystererts-steenkool materiaal laag. Reaksie van kleiner partikels toon verhoogte reaksietempo as gevolg van ‘n afname in diffusie beperkende effekte, soos waargeneem in reaksie van die basis partikelgrootte vir steenkool en ystererts.

Sleutelwoorde: Hitteoordrag, eendimensioneel, stralingsnetwerk, straling, geleiding, steenkool, ystererts, temperatuur, oond, materiaallaag

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INTRODUCTION

The use of coal instead of coke as reductant in the iron and steel industry has become more important because this industry realised that coking coal supply could soon be less than demand (Nashan *et al.*, 2000). Furthermore, the future trend is expected to tend toward process consolidation by reducing the number of reactors needed to produce steel from raw material (Wiesinger, 2000). Accompanied with the trend in coal usage has emerged several processes that use iron ore fines, which are not compatible with older iron making technologies when not agglomerated or pelletised (Sarma and Fruehan, 1998). Most of these processes use natural gas to reduce fine iron ore to directly reduced iron (DRI). A few processes that use coal as reductant to produce DRI have been developed. These are the rotary kiln type processes: SL/RN (Bornman and Ackerman, 1993), Accar (Rierson, 1993), Davy DRC (Haworth *et al.*, 1995) and rotary hearth based processes: Fastmet (Hoffman and Harada, 1997) and Comet (Borlée *et al.*, 1999) processes. Of these processes, only the rotary kiln type processes have been commercialised on a large scale. The only commercially established coal based process to produce hot metal is the Corex process (Flickenschild *et al.*, 1996). Other hot metal processes have been developed: AISI (Aukrust, 1992), Hismelt® (Cusack *et al.*, 1995), Dios (Saito, 1992), Romelt (Romenets *et al.*, 1999), Ausmelt (Floyd, 2000) and Technored (Contrucci, 2000). The first commercial Hismelt® plant has been successfully hot commissioned in October 2005 in Kwinana, Western Australia and production will be ramped up to full capacity over three years to 800 000 t/year (Rio Tinto News Release, 2006).

Thus, it is evident from the development of the above mentioned processes that the use of coal and iron ore fines is becoming more important as traditionally used feed stocks of iron ore and coal are depleted. Only the Comet (Borlée *et al.*, 1999) process uses coal and iron ore fines in a fixed material bed, although in alternate layers, to produce DRI. The hot metal production processes do use coal and iron ore fines, but these raw materials are reacted through bath smelting.

The IFCON® process is a direct steelmaking process reacting iron ore fines and coal in a single vessel to produce crude liquid steel. Material mixture of ore fines, coal and fluxes of -10 mm is fed onto the liquid metal bath to form heaps floating on the metal bath. The freeboard is heated by combustion of natural gas and an air and oxygen blast blown into the freeboard via burners. In addition to the natural gas that is combusted in the freeboard, the coal volatiles and reduction product gas from the heaps are combusted to generate heat in the freeboard. The upper portion of the heap where solid state reduction takes place is heated by fossil fuel energy generated in the freeboard. The bottom ends of the heaps are heated from the metal bath, which is in turn heated by inductors.

As identified by Pistorius (2005) ore-carbon/coal reaction systems form the third type of heat transfer control in which there is a band of reaction temperatures in which the process can take place, if no bulk melting of reactants and products takes place. Most of the work done on mixtures of carbon/coal and iron ore (as reported in the literature) was done with the intent of isothermal reaction, but the use of relatively large sample sizes and/or heat transfer hampering sample containment arrangements resulted in non-isothermal reaction. Therefore, the non-isothermal treatment of the samples was not taken into account, and reaction data from the experiments was used to calculate apparent activation energies at the furnace temperatures. From the magnitude of the apparent activation energies conclusions were made as to the rate controlling mechanism in the experiment. Seaton *et al.* (1983) were the first to highlight the problem of using chemical kinetics alone to make conclusions on the rate controlling step when the mixed ore-carbon/coal sample is reacted non-isothermally.

As pointed out by Vankateswaran and Brimacombe (1977) a lot of work is required to obtain all the necessary detailed fundamental information to describe the process progress in a mixed bed system so that an empirical approach to reaction rate measurements is more effective. Therefore a realistic simulation experiment is required in which the heat transfer rate is quantified from measurement of temperature and reaction extent as functions of reaction time and position within the sample material. Here the development of such a simulation experiment for the solid state reduction under unidirectional radiative heating is described, and results reported and interpreted. The information gained from such an experiment should provide enough information to use in validation of mathematical models that can then be used for process design and testing process sensitivities.