# ABSTRACT

Constantly declining ore grades and the growing steel demand stimulate the broader utilization of magnetite ore, a primary but most underutilized iron ore. Magnetite is less reducible and prone to swelling during reduction. However, magnetite pellets made from magnetite fines are porous and highly reducible. Recently, several alternative routes of ironmaking have emerged for the sustainable development of the iron and steel sectors. One such process is based on a rotary hearth furnace (RHF), where iron-bearing fines or steel plant waste can be reduced in the form of a cold-bonded composite pellet/ briquette with some carbonaceous material and subsequently utilized in a blast furnace as metalized charge or in an electric arc furnace (EAF), depending on its grade. The quality of the burden material in RHF can be compromised because a rotating hearth carries the material in a circular path through different temperature regimes, while the material remains stationary. Naturally cured pellets with comparatively lower strength could be used safely in this reactor. Besides, such pellets also have enough pores to accommodate any volume change due to swelling during magnetite reduction, avoiding catastrophic failure.

RHF is a dual-fuel reactor, where heat and reduction fuels are different; solid coal acts as a reductant, and a low-carbon fuel like natural gas is used for heating, mitigating $CO_2$ emissions. Besides, intimate contact between the carbon and iron ore particles in the pellets improves kinetics and reduces the reductant carbon requirement. Despite having several benefits over other reactors, RHF suffers from low productivity due to heat transfer limitations through the pellet bed that restrict the number of layers over the hearth. Therefore, a significant scope exists to enhance the heat and mass conductance in a multi-layer bed RHF. The present study also investigates the fundamentals of the reduction behaviour of magnetite-coal composite pellet in a three-layer bed laboratory-scale RHF, a study conspicuously absent in the literature.

In the present research, the reduction of magnetite-coal composite pellets has been carried out in a laboratory-scale electrically heated three-layer pellet bed RHF to investigate the layer-wise reduction behaviour of magnetite-coal composite pellets with varying carbon content, a crucial parameter influencing the pellet's heat effects and carbon efficiency. Some experiments have also been carried out in a muffle furnace to investigate the time-dependent data, the effect of particle size, and carbon efficiency.

The muffle furnace study of a single magnetite-coal composite pellet indicated that at comparatively lower temperatures (1150°C), the degree of reduction (DOR) increased monotonically with a decrease in particle size of iron ore and coal fines. While at a higher temperature of 1200°C, it passed through a maximum in the particle size range $106 < \text{size} < 150$ µm at 1200°C. At a further higher temperature (1250°C), the overall DOR was higher without any significant change with a change in particle size. The above results were attributed to competing factors like the specific surface area of particles, the porosity of the pellet, and thermal activation with changes in particle size and temperature.

With an increase in carbon level in the initial pellet (measured in terms of C/O ratio), the DOR passed through a maximum at a C/O atomic ratio of 0.75, attributed to counteracting effects of enhanced carbon activity and higher heat and mass transport resistance at higher coal levels due to its non-conducting ash. The apparent activation energy for the reduction of pellets containing C/O = 0.75 was estimated as 83 kJ/mol, indicating a significant role of heat and mass transfer in kinetics. This finding was ratified by plotting DOR versus DOM, where the experimental best-fit line was found to drift significantly from the stoichiometric line.

For three-layer pellet bed reduction at RHF, it was observed that the DOR passed through a maximum at a C/O ratio of 0.75, irrespective of layers, which was consistent with the results obtained for single pellet reduction in a muffle furnace. A maximum DOR was observed at around 85% for the top layer, followed by the bottom layer, and lowest at the middle layer. Higher reduction at the bottom layer was attributed to the volatile contribution to reduction at a comparatively slower heating rate. The average dry strength of the green and reduced pellets was estimated at 125 N and 2000 N, respectively; such strengths were sufficient for handling green pellets in RHF and reduced pellets for charging in BF.

Finally, the reduction of magnetite-coal composite pellets at laboratory-scale multi-layer bed RHF has been successfully demonstrated with efficient reduction and shrinkage (up to 85% DOR in 10 minutes) and without any swelling, alleviating the magnetite's inherent problem of swelling and sluggish rate of reduction.

As an extension, the present research also studied the effect of various carbonaceous materials on the reduction efficiency of composite pellets in a three-layer bed RHF. However, the iron ore used was hematite in this case. Four different types of carbonaceous material, namely low fixed carbon coal A, high carbon coal B, coal char, and wood char, were tested in a three-layer bed RHF. Coal char produced the maximum reduction. Coal B pellets produced

a higher reduction than coal A pellets; however, they produced a lower reduction than wood char pellets, although coal B contained comparable fixed carbon to that of wood char. All three layers showed a similar trend. The morphology of reduced pellets indicated more surface cracks in coal A, followed by coal B, wood char, and coal char. Fayalite, a low-melting eutectic, only showed its limited presence at the top layer in coal char pellets. In wood char pellets, fayalite was also present in the middle and top layers. Fayalite was observed in all layers for both coal pellets. Fayalite is liquid, and it wets pores, hampering gas passage for reduction. The phase evolution was comprehensively ratified in all the above studies with FactSage analysis, X-ray diffraction (XRD), and scanning electron microscopy (SEM) coupled with energy-dispersive spectroscopy (EDS) analysis.

***Keywords***: Rotary hearth furnace, Multi-layer pellet bed, Reduction behaviour, Microstructure, Magnetite ore, Forms of carbon, Iron ore-carbon composite pellet, Thermodynamic modeling, Characterization.