学 位 論 文 内 容 の 要 旨

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学 位 論 文 題 名

Ironmaking Process using Carbon Deposition by Chemical Vapor Infiltration (CVI) Method
(化学気相浸透 (CVI) 法による炭素析出を用いる製鉄プロセス)

The consumption of iron and steel would increase in the near future to meet the rapid economic growth of several countries such as China, Indonesia, India, and Brazil. However, the iron and steel industry is facing serious problems related to limited resources, energy, and environment simultaneously due to high dependency on high-grade ore and coking coal. A proper strategy should be devised to explore innovative methods for substituting the conventional raw materials and energy resources.

The chemical vapor infiltration (CVI) ironmaking process consists of integrated pyrolysis-tar decomposition over a porous low-grade ore. This process was proposed to solve the aforementioned problems by effective utilization of low-grade coal and biomass. Tar that may cause operational problems such as pipe plugging, condensation, and tar aerosol formation is also resolved in this process. In this thesis, the proposed CVI system is comprehensively examined by discussing several important parameters such as various porous materials, different carbon sources, optimum temperature, kinetic analysis, and product microstructure. In addition, the overall evaluation of the proposed system is discussed on the basis of an exergy analysis to confirm the benefits of the system. The thesis consists of six chapters in which first and last chapters describe general introduction and conclusions, respectively.

Chapter 2 describes a detailed evaluation of the CVI ironmaking process using a low-grade ore as a carbon storage medium and catalyst for tar decomposition. The low-grade ore showed a significant effect on the tar decomposition process by enhancing carbon deposition and total gas product formation. The tar amount obtained from pyrolysis was strongly affected on carbon deposition. The bituminous, a high-grade coal produced the highest carbon deposition due to large tar product composition. A larger amount of combined water within the low-grade ore resulted in pore size less than 4 nm, which was suitable for carbon deposition. The highest amount of carbon deposition was obtained when pyrolysis and tar decomposition were conducted at 800^oC and 600^oC, respectively. A higher temperature of tar decomposition resulted in lower carbon deposition owing to the gasification process. However, sintering started at 800^oC and it significantly diminished the BET surface area of iron ore. Indirect reduction simultaneaously occurred with the decomposition process thereby producing Fe_3O_4 at 600^oC and FeO at 800^oC. The reduction of the CVI ore began at 750^oC, while that of the reference, mixture of Fe_3O_4 and coke started at 1100^oC. The CVI ore exhibited higher reactivity owing to nanoscale contact between the iron ore and carbon.

In chapter 3, the kinetic analysis and carbon deposition phenomenon of tar decomposition over the low-grade ore are examined to understand the CVI process. Both the kinetic constant and deactivation factor are evaluated successfully using a simple proposed model within the ranges 0.13–0.55 s^{-1} and 1.72–2.53 s^{-1}, respectively, at 500^oC-700^oC with the activation energy of 44.86 kJ/mol. The deactivation factor exhibited a similar tendency as the amount of carbon deposition within iron ore pores.

Chapter 4 includes a study of the characteristics of the CVI product related with the microstructure of the porous ore and distribution of carbon deposition. On the basis of TEM images, a layered structure containing around 3-nm-diameter pores was observed after dehydration owing to the removal of the hydroxide (OH) group from FeOOH. This pore size was deemed appropriate for tar decomposition and resultant carbon deposition. EDS results confirmed that carbon distribution was highest near the outer surface and decreased gradually with increasing depth in the inner section of ore. In addition to the ore structure, the type of carbon deposition was also successfully evaluated using Raman spectroscopy. The carbon deposition by the CVI process within ore pores was categorized as amorphous carbon (a:C), with an sp3 content of 19–21%.

Chapter 5 describes the exergy analysis and presents a feasibility study on the applications of the CVI process in the ironmaking industry. The CVI process significantly increases the exergy content of iron ore. On the basis of 3.86 %mass carbon deposition (experimental value) and production of 1000 kg metallic Fe, the exergy loss of the proposed system was found to decrease by about 16.7% compared to that of conventional systems through the recovery of both chemical and thermal tar exergy. When the CVI ore was sent to a sinter plant, the amount of deposited carbon was sufficient to completely replace coke breeze in which the ratio of the CVI ore is above 70% of the total input ore. The total enthalpy of the CVI ore originated from the reoxidation of Fe_3O_4 and deposited carbon. The application of the CVI ore in a sinter plant would result in an extensive decrease of the coke breeze and CO_2 emissions.

On the basis of these results, it could be concluded that the proposed CVI ironmaking process offers promising benefits by alleviating the problems related to resource, energy, and environment simultaneously