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PREPARATION OF Ni POWDERS FABRICATED BY VARIOUS REDUCTIVE GASES

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Abstract. Ni powders were prepared in various gases from commercial NiO ores at different reaction conditions. Nickel oxide (NiO) pellet mixed with carbon powders was reduced in N_2 gases at first reductive stage. In the temperature range from 800 to 1000 °C, the reduction rate of NiO increased with increasing temperature, reaction time, and carbon content in the pellet. At second stage NiO was consecutively reduced by hydrogen (H_2) and the rate of reduction was also found to be increased with increasing temperature from 400 to 1100 °C, reaction time, hydrogen content. It was finally found that NiO was found to be almost completely reduced by hydrogen with 1.0 m^3/hr at 1000 °C for 1 hour.

1. INTRODUCTION

The nickel powders have been used over past decades due to their properties such as magnetism, thermal resistance and chemical activity; and have a wide range of applications including batteries, hard alloys, catalysts, *etc.* [1-4]. Some researchers have studied to make metal powder of Ni using various methods, but none of them was economically feasible [5-6]. Therefore, there is a need to develop simpler and more efficient reductive process for NiO powders. In this study, NiO powder from ores was reduced to Ni using Ni pellet mixed with carbon powder in N_2 gas at first reduction, and direct hydrogen reduction was continuously conducted at second reduction stage on the effect of temperature, time, and carbon/hydrogen content.

2. EXPERIMENTAL PROCEDURES

Nickel oxide (NiO) powders were brought from the ores at Conickel Nickel Mining in Cuba, and the powders in this work were used after flotation process. Particle size distribution tested and chemical composition of NiO are given in Fig. 1. It is seen that NiO particles are less than 10 mm and mostly between 1 and 3 μm, which is identified in the image taken in SEM. The oxide particle is mainly made up of Ni, and content of Co, Fe, C, and Mn only count for only 0.01% of the total mass. That is, the oxide samples used in this study are found to be almost pure NiO powders.

The reduction of NiO to Ni was carried using two different reducing gases, N_2 and H_2, at each stage in electric furnace, and schematic diagram of experimental apparatus is shown in Fig. 2. NiO pellet, with size of $\varphi 30\times 10$, was prepared by a pressing machine with adding carbon content shown in the figure and the samples on the boat were placed in alumina tube. They were heated with flowing N_2 gas in a controlled gas environment of N_2 gas using a flowmeter at the first stage, and then continuously reduced by H_2 gas with same condition at the sec-

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ond stage. The specimens were taken out of alumina tube and then measured using an electric scale once they are cooled down at room temperature. Finally, the samples were ball-milled several times to make appropriate size of fine particles of Ni metal. The specimen of Ni powder was characterized by X-ray diffraction (XRD) and scanning electron microscopy (SEM). The chemical composition of powder was also identified by inductively coupled plasma (ICP).

3. RESULTS AND DISCUSSION

Reduction of NiO was carried out with consecutive processes using two different gases. NiO pellet mixed with carbon powders was reduced by N_2 gas at first, and then the sample was reduced by H_2 gas in a row. Through reaction equations stated below, NiO sample can be reduced by C, CO(g), and H_2(g) at the temperature range of 800~1000 °C.

$$NiO + C = Ni + CO(g),$$

$$\Delta G = -15.42 \sim -27.83 \text{ kcal } (800 \sim 1100\ ^{\circ}C)$$

$$NiO + CO = Ni + CO_2(g),$$

$$\Delta G = -11.23 \sim -11.22 \text{ kcal } (800 \sim 1100\ ^{\circ}C)$$

$$NiO + H_2 = Ni + H_2O(g),$$

$$\Delta G = -11.11 \sim -13.25 \text{ kcal } (800 \sim 1100\ ^{\circ}C)$$

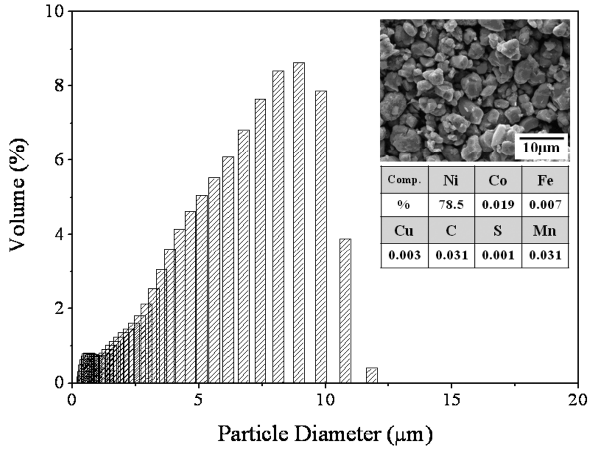

Fig. 1. Particle size distribution and composition of NiO sample.

Fig. 3 demonstrates reduction ratio of NiO pellet on the effect of temperature and time. The reduction ratio was calculated by weight reduction of oxygen in NiO divided by initial oxygen content in NiO. Reduction of NiO was conducted at 1 m^3/hr of N_2 gas for 1 hr, and it was found that reduction ratio was increased with increasing temperature, but the ratio might reach the maximum value of about 75% at 1100 °C. Reduction ratio of NiO measured at 1000 °C was also increased with increasing time, and the rate was sharply increased for 1 hr, but the

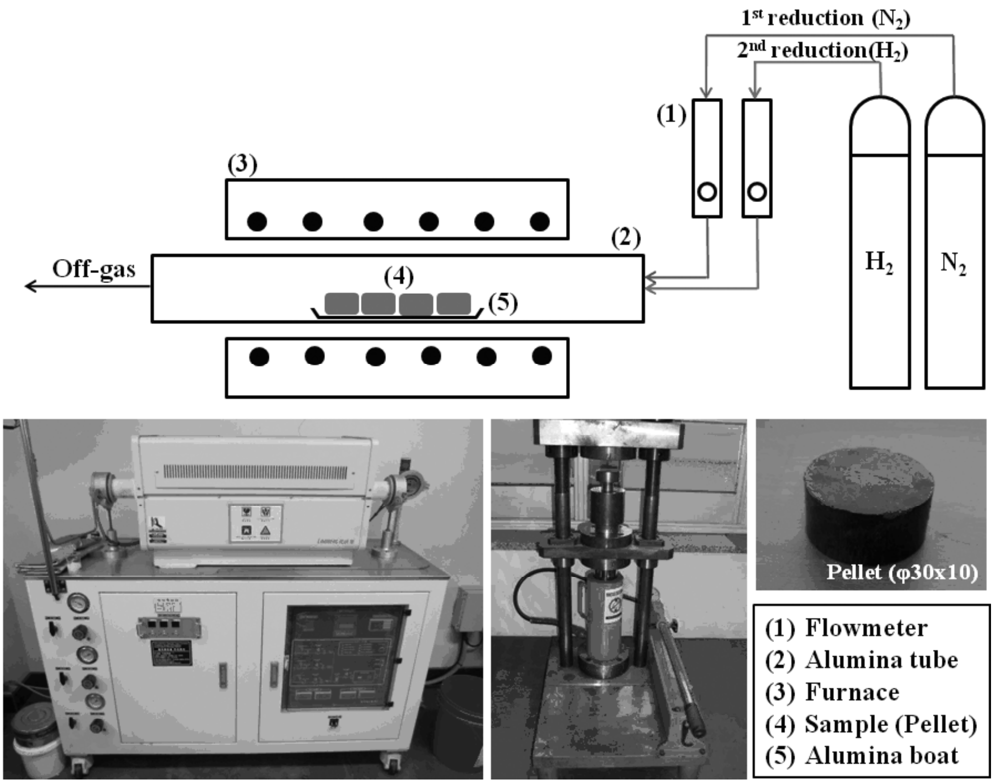

Fig. 2. Experimental apparatus for reduction of NiO pellet.

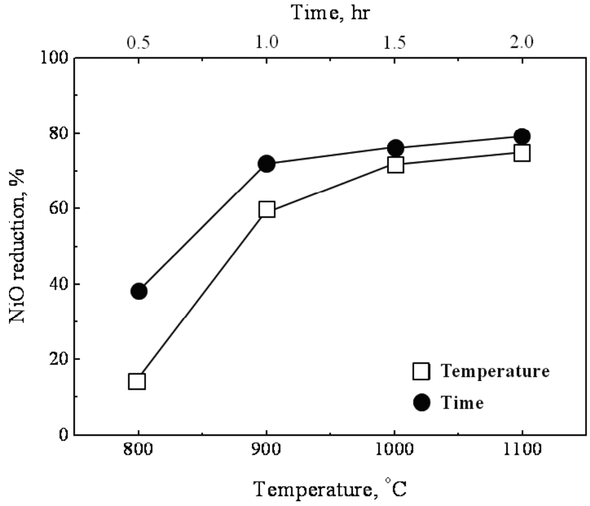

Fig. 3. Variations of NiO reduction ratio in N_2 gas as a function of temperature and time.

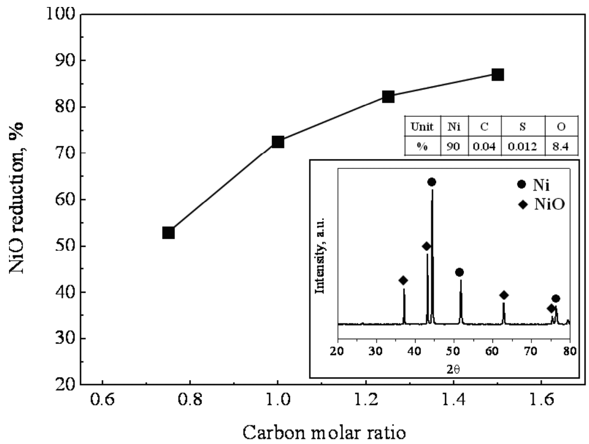

Fig. 4. Variations of NiO reduction ratio with carbon molar ratio.

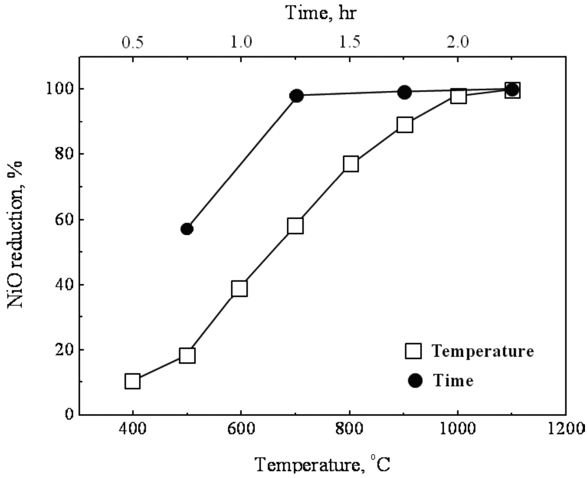

Fig. 5. Variations of NiO reduction ratio in H_2 gas as a function of temperature and time.

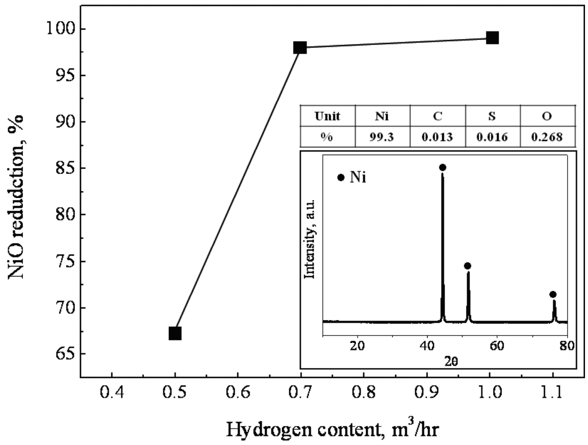

Fig. 6. Variations of NiO reduction ratio with hydrogen content ratio.

slope was shown to be slowing down after the initial stage.

Reduction ratio of NiO was measured with increasing carbon content at 1000 °C with 1 m³/hr of N_2 gas shown in Fig. 4. Increasing carbon molar ratio led to increase reduction ratio of NiO, but Ni and NiO phases were both detected in XRD pattern indicating that adding carbon could not reduce NiO to Ni powder successfully. The sample after ball-milled was characterized by ICP analysis and Ni was found to be made up of major component counting for 90% with some oxygen content in the powder. At the first reduction, reduction ratio of NiO to Ni was shown to be highest at 1000 °C of temperature, 1 hr of reaction time, and 1.5 of molar ratio.

At the second reduction, NiO was continuously reduced by H_2 gas as a function of temperature, time, and H_2 gas content. Fig. 5 illustrates that the ratio of NiO reduction was gradually increased with increasing temperature and maximum reduction ratio for almost 100% was achieved over 1000 °C for 1 hr in flow rate with 0.7 m³/hr of H_2 gas. In addition, the reduction ratio was also found to be increased with reaction time and the ratio was abruptly increased for 1 hr indicating that reduction of NiO to Ni could be accomplished quickly compared to the reduction with carbon and N_2 gas conducted at the first reduction.

Fig. 6 shows the ratio of NiO reduction conducted by increasing flow rate of H_2 gas at 1000 °C for 1hr. It is clearly seen that NiO could be almost completely reduced to Ni using H_2 gas over 0.7 m³/hr. The sample reduced by 1.0 m³/hr of H_2 gas was examined by XRD and ICP analysis and Ni peak was only detected in XRD patterns. As result of ICP analysis, it might be confirmed that NiO powder was completely reduced to Ni with only small amount of impurities.

4. CONCLUSIONS

1. The ratio of NiO reduction mixed with carbon powders was increased with increasing temperature and reaction time under N_2 gas at first stage, and it reached the maximum value of about 75% at 1100 °C for 2 hours.

2. At the second stage of NiO reduction, the ratio was gradually increased with increasing temperature and reaction time using H_2 gas, and maximum reduction ratio about almost 100% was achieved over 1000 °C for 1 hr in H_2 gas flow rate with 0.7 m^3/hr.

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