Preparation of Ni/γ-Al_2O_3 Catalyst Using Microwave Plasma Jet Combined with Spouted Bed

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Abstract: Microwave plasma jet combined with a spouted bed was applied for Ni/γ-Al_2O_3 catalyst preparation. Despite short treatment time, Ni/γ-Al_2O_3 catalyst was successfully produced using the plasma spouted bed and its catalytic activity for ethylene hydrogenation showed higher activity than those prepared by conventional method using an electric furnace. Catalyst characterizations were carried out by XRD, BET and SEM. The plasma spouted bed was suggested to prepare active catalyst effectively.

Keywords: Microwave plasma, Spouted bed, Ni/γ-Al_2O_3 catalyst.

1. Introduction

Microwave plasma is an electrodeless discharge and thus there is no contamination by the electrode. The plasma is applicable for particle processing [1]. By combining the microwave plasma with the spouted bed, it is expected that a large amount of particles can be uniformly treated in a short period of time [2]. The conventional method to prepare catalyst using an electric furnace requires high temperature and takes large amount of time for the calcination process. This can be improved by replacing electric furnace with microwave induced plasma due to high temperature and high activity of the plasma. In the present work, microwave plasma jet combined with spouted bed was applied for the preparation of nickel-impregnated alumina catalyst (Ni/γ-Al_2O_3) and its catalytic activity was evaluated.

2. Experimental

2.1 Spouted bed reactor with microwave plasma jet

The spouted bed reactor with microwave plasma is shown in Fig.1. The plasma spouted bed consisted of a quartz glass having an internal diameter of 27mm, a nozzle diameter of 9mm and a length of 370mm. The inlet was inclined upwardly at an angle of 60°. The plasma was generated by a microwave generator under atmospheric pressure and injected to spout the particles from the bottom of the spouted bed. A mixture of Ar and H_2 was used as a plasma gas.

2.2 Preparation of Ni/γ-Al_2O_3 Catalyst

Ni/γ-Al_2O_3 catalyst containing 15wt% of Ni was prepared by the plasma spouted bed as follows:

<Step 1> $Ni(NO_3)_2 6H_2O$ was dissolved in distilled water, and $Al(OH)_3$ powder was added into solution.

<Step 2> The solution was dried at 80°C until slurry-like.

<Step 3> The slurry was dried at 110°C for 12 hours and crushed.

<Step 4> The crushed particles were treated in the plasma spouted bed with the following condition: power of 150W, Ar flow rate of 3L/min, H_2 flow rate of 40ml/min and treatment time with 8 min.

For comparison, Ni/γ-Al_2O_3 catalyst was also prepared by the conventional method with the electric furnace as follows:

<Step 4c> The crushed particles were heated at 700 °C in N_2 atmosphere for 4 h followed by H_2 addition at 700 °C for reduction for 2 h.

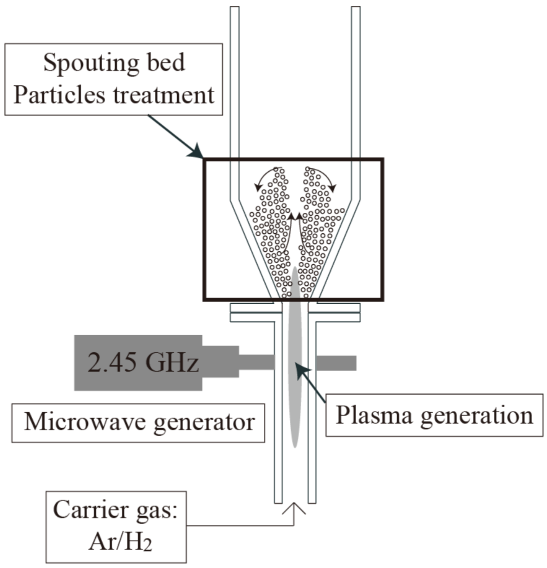

Fig. 1 Schematic diagram of microwave plasma spouted bed.

2.3 Evaluation of Ni/γ-Al_2O_3 Catalyst

Hydrogenation reaction of ethylene was chosen for the evaluation of catalyst activity. This reaction was often used because of its simplicity and less side reaction. The conversion of ethylene was analysed using a gas chromatography.

2.4 Catalyst characterization

The samples prepared here were analyzed by X-ray diffraction (XRD, PANalytical) and scanning electron microscopy (SEM, Keyence VE-7800) to characterize their crystallinity and surface morphology, respectively. The specific surface area of the catalysts was measured by nitrogen adsorption method (BET, Quantachrome instruments).

3. Results and discussion

Fig. 2 shows XRD patterns of the catalysts prepared by the plasma spouted bed and the conventional electric furnace. According to the XRD analysis, the peaks of NiO were not detected in both methods which means the treatment time of 8min in the plasma spouted bed was enough to reduce NiO into Ni due to high reducing power of hydrogen radical in the plasma. In both samples, the peaks of γ-Al_2O_3 phase were detected. However, in the plasma treated Ni/γ-Al_2O_3 catalyst, the peaks of boehmite[AlOOH] were still found. It means that the treatment time of 8min was not enough to change the whole aluminum hydroxide into γ-Al_2O_3. A major difference between diffraction patterns of the two samples is the intensity of peaks of metallic Ni at $2\theta = 44.0$ and 51.25. Ni/γ-Al_2O_3 catalyst prepared by the electric furnace shows sharp and intense peaks than those prepared by the plasma spouted bed. It is known that half width at half maximum(HWHM) of the peak is inversely proportional to the crystallite size. The HWHM of Ni/γ-Al_2O_3 catalyst prepared by the plasma spouted bed and the electric furnace was 0.42[deg] and 0.27[deg], respectively. Thus, Ni/γ-Al_2O_3 catalyst prepared by the plasma spouted bed indicates smaller crystallite size than that prepared by electric furnace. Fig.3 shows the conversion of ethylene to ethane as a function of reaction time. In the both samples, the conversion was relatively steady. Despite the same amount of loaded nickel, the catalyst synthesized by the plasma spouted bed showed higher catalytic activity than that prepared by the electric furnace. The catalytic activity mainly depends on the crystallite size of metal which affects the specific surface area of metal on the surface. Therefore, it was suggested that the treatment by the plasma spouted bed made the crystallite size of metallic nickel smaller which caused more reactive site for ethylene hydrogenation.

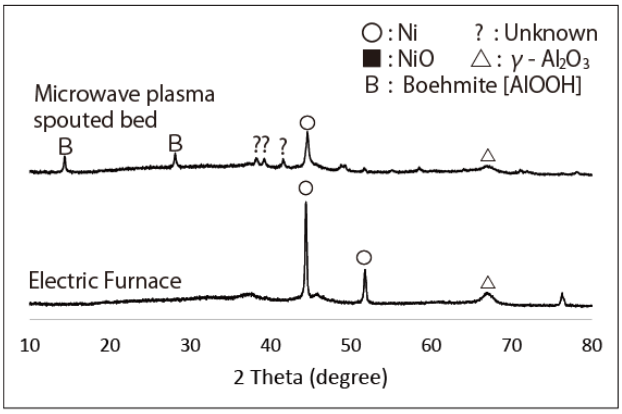

Fig. 2 XRD spectra of Ni/γ-Al_2O_3 catalyst prepared by microwave plasma spouted bed and electric furnace.

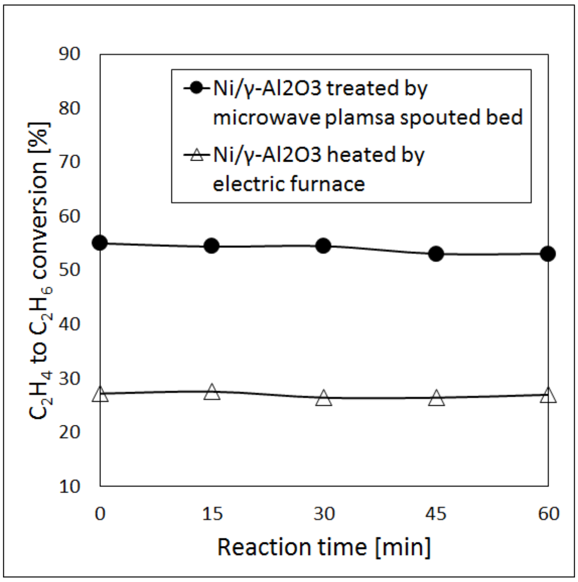

Fig. 3 C_2H_4 to C_2H_6 conversion as a function of reaction time at catalytic temperature of 130°C with the gas flow rate of C_2H_4:60mL/min, H_2:60mL/min, and Ar:100mL/min in the presence of Ni/γ-Al_2O_3 catalyst.

4. Conclusion

The microwave plasma jet combined with spouted bed was applied for Ni/γ-Al_2O_3 catalyst preparation. For comparison, Ni/γ-Al_2O_3 catalyst was also prepared by the conventional method using the electric furnace. From XRD analysis, Ni/γ-Al_2O_3 catalyst was successfully produced in both ways, however small crystallite size of metallic nickel was indicated in the plasma spouted bed treatment. The catalytic activity of the catalyst prepared by the plasma spouted bed showed higher catalytic activity than that prepared by the electric furnace. It was suggested that the plasma spouted bed had a potential to prepare active catalyst effectively.

References

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