

Natural gas reforming method using an inverter plasma

インバータープラズマを用いた天然ガスの改質

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The characteristics of a natural gas reforming method for hydrogen production were investigated. The reforming method combines a plasma process operated by an inverter power supply with a catalytic reaction to improve the energetic efficiency for reforming. A simple process model is presented and examined using a methane gas reforming experiment. The validity of the process model and the temperature dependency of the catalyst activity are discussed with respect to the results.

1. Introduction

To reduce CO₂ gas emission to the atmosphere, it is hoped that natural gas, including a majority of CH₄, will be used in a reforming method that decomposes CH₄ into H₂ gas and C-containing components [1]. Reforming methods that employ plasma discharge have advantages in that no CO₂ is produced through the reforming process itself and the process plant can be manufactured on a compact scale for application in locations such as gas stations and farms. [2,3]. Improvement of the reforming energy efficiency, i.e., the ratio of the combustive energy of produced H₂ divided by the electric power for plasma discharge, is required to realize practical application.

We propose a CH₄ gas reforming method that employs a plasma discharge generated using an inverter power supply with high electric power efficiency. This method features a reforming process that involves the combination of the plasma chemical reaction and a catalytic reaction to improve the reforming energy efficiency.

In this article, we describe the process model for the proposed method and the experimental apparatus. We then present experimental results and discuss the characteristics of the reforming process.

2. Process Model

The simple process model for CH₄ gas reforming is shown in Fig. 1. The modeled process is divided into stages I and II. In stage I, raw CH₄ gas is supplied to the stage I area and is excited by electrical plasma discharge. A portion of CH₄ is

dissociated as CH₄ → CH₃ + H. The excited gas, which includes CH₃ radicals, flows into the stage II area and is exposed to a catalytic material coated on the substrate. Pure carbon or hydrocarbon is deposited on this substrate from the CH₃ radicals. The substrate is heated to T_{II} to maintain activation of the catalyst.

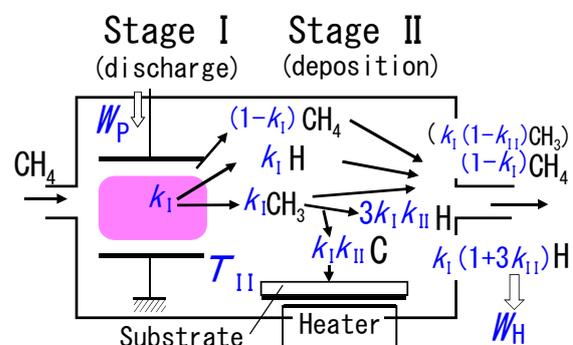


Fig.1. Model for the proposed reforming process. CH₄ gas is introduced and excited at stage I. Carbon from the excited gas is deposited on the heated substrate at stage II. The output gas includes produced H₂ gas and unprocessed CH₄ gas.

Here, the dissociation rate of CH₄ at stage I and that of CH₃ radicals at stage II are defined as k_1 and k_{II} , respectively. The H₂ production rate is thus expressed as $k_1(3k_{II} + 1)$. In addition, the electrical power consumed at stage I and the combustive energy of liberated H₂ gas are defined as W_p and W_H , respectively. The reforming energy efficiency is calculated as W_H/W_p .

3. Experiment

The configuration of the experimental apparatus used to examine the process model is shown in Fig. 2. A plasma is generated between upper and lower parallel-plate electrodes. A substrate is placed on the lower electrode with a heater. Stainless steel 304 alloy plate, which contains the catalytic component (Fe), was used as the substrate. When the substrate temperature reached T_{II} under vacuum, CH_4 gas was introduced into the reaction chamber up to 600 Pa and the plasma discharge was maintained for a further 40 min.

Gas partial pressures in the reaction chamber were analyzed using a gas analyzer with a quadrupole mass-spectrometer and a differential evacuation system. The mass of carbon deposited on the substrate was measured using an electronic balance after processing. The morphology of the deposited carbon was also observed using scanning electric microscopy (SEM).

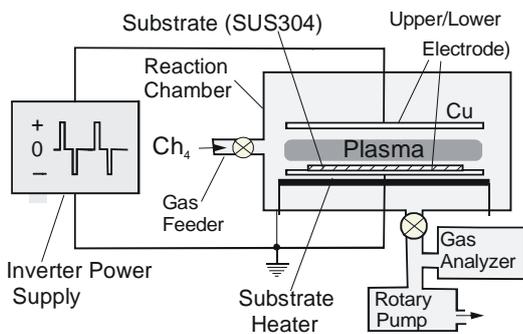


Fig.2. Experimental apparatus configuration for the CH_4 reforming process.

4. Results and Discussions

Typical time evolutions of the H_2 and CH_4 partial pressures in the reaction chamber are presented in Fig. 3. The production of H_2 gas using the proposed plasma process was confirmed. Here, we focus on the relation between carbon deposition and the substrate temperature, T_{II} . Figure 4 shows SEM

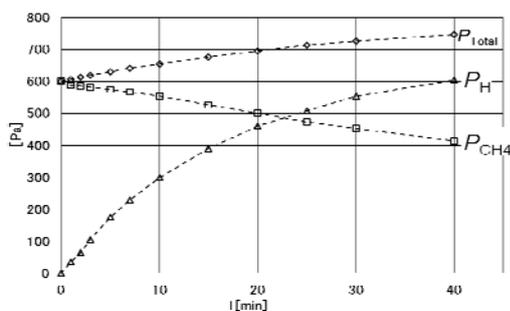


Fig.3. Experimental apparatus configuration for the CH_4 reforming process.

images of the substrates after processing. The catalyst was activated in the temperature region greater than ca. 450°C and a larger amount of carbon was deposited.

In the lecture, we will describe the validity of the process model and the dependency of the reforming energy efficiency on T_{II} .

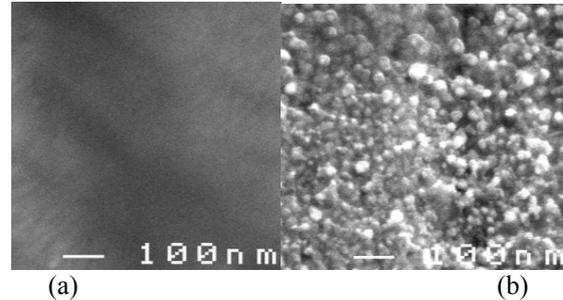


Fig.4. SEM images of substrate surfaces after processing at substrate temperatures of (a) $T_{II} \approx 350$ and (b) $T_{II} \approx 450^\circ\text{C}$.

Acknowledgments

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