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Description of reaction rate for steam methane reforming throughout a channel in combination with diffusion in a porous catalytic wall: Extension to concentration variation along flow direction

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ABSTRACT

In a reactor with a porous catalyst on a channel wall, the local reaction rate and composition cannot be easily measured. This study built a model for evaluating the overall reaction rate by predicting the distribution of the concentration of chemical species in a porous catalyst as well as in a gas stream. Prior to taking into account the streamwise distribution, the validity of the function of the concentration profile in steam methane reforming, which is one-dimensional to the thickness direction of catalyst, was confirmed by comparison with the results of an experiment. Next, the model was coupled with the streamwise variation in the concentration. Consequently, we derived a model in which the local concentration can be specified at any position with the concentration at the inlet of the channel as the only input variable. Finally, a criterion for the condition in which the maximum yield of products can be obtained with the minimum volume was proposed with nondimensional groups.

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Introduction

Hydrogen has been utilized for many purposes such as a fuel in internal combustion engines [1], gas turbines [2] and fuel cells [3], and as a material to form other products, e.g. the gas to liquid (GTL) process [4]. Steam methane reforming (SMR), expressed by the following equation, is the most popular way to produce hydrogen [5].



A catalyst is generally used to enhance the reaction of SMR. To improve the performance of the catalyst in terms of activity and durability, various catalyst materials and their

supports have been examined [6] [7]. Although many expressions for the reaction rate, which is one of the most important factors in designing a reactor, have been proposed [8–10], universal expression have not been proposed because each material has its own kinetics. Therefore, prior to the application of a newly developed catalyst in a reactor, the reaction rate must be obtained through experiments.

In addition to chemical aspects, the arrangement of the catalyst has a strong influence on the performance of a reactor. Although catalyst pellets are packed into a reactor in most large-scale plants, a packed catalyst has the disadvantages of a high pressure drop and the risk of plugging or channeling, particularly in small systems [11]. Therefore, in some reactors, the catalyst is coated on a channel wall

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[12–14]. In such systems, the transport phenomena at the interface between the gas stream and the catalytic wall are important. Furthermore, we must take into account mass transfer inside the catalyst layer, that is, internal diffusion, because catalysts generally have a porous structure.

In our previous paper [15], we proposed a method for determining the reaction rate of SMR in a porous catalyst on a channel wall. Generally, it is difficult to acquire local information not only within a porous catalyst but also in a gas stream. Thus, in the proposed method, changes in the bulk concentrations of gas species were measured experimentally and introduced into the one-dimensional diffusion equation in the thickness direction in the catalyst layer. As a result, a power-law-type reaction rate with an upper limit was found to be appropriate for expressing the local reaction rate. Moreover, the dimensionless numbers derived from the reaction rate expression and diffusion equation provide a quantitative criterion for the rate-limiting condition.

The model developed in our previous paper [15] was one-dimensional in the thickness direction of the catalyst layer, because catalysts had a short length. To design an actual catalyst with a certain length, numerical simulation using the reaction rate estimated from an experiment with a short catalyst is a possible method. For a wider application range, the aim of this study is to develop a model that can specify the local phenomena at any position in a reactor even if the gas composition varies along the flow direction. To this end, we attempt to describe the concentrations of species in a gas stream and inside a porous catalyst as functions of position.

In this paper, we first derive a function for the concentration in the thickness direction of the catalyst on the basis of the experimental data in our previous paper [15]. Next, an experiment is performed with catalysts of different lengths and thicknesses. Finally, we derive equations for the concentration distribution in the case of variation along the flow direction. In this model, the overall reaction rate throughout the reactor can be evaluated from the gas composition at the inlet.

Concentration profile in one dimension

Before developing a two-dimensional model, we derive the one-dimensional concentration distribution perpendicular to the flow direction and verify its validity by comparison with experimental results.

Basic equations

As described in our previous paper [15], the overall reaction can be considered on the basis of the diffusion and reaction inside the porous catalyst. Although there are some diffusion models, as investigated by Solsvik et al. [16], a simple equation is chosen to express the concentration profile in this study.

If the diffusion is one-dimensional in the y -direction, the conservation equation for species j can be expressed in terms of the local reaction rate r_{st} as follows:

$$\frac{d}{dy} \left(D_j^{\text{eff}} \frac{dC_j}{dy} \right) = n_j r_{st}, \quad (2)$$

where D_j^{eff} , C_j and n_j are the effective diffusion coefficient, molar concentration and stoichiometric coefficient, respectively. The flux at the interface between the gas phase and the catalyst layer, that is, $D_j^{\text{eff}} dC_{\text{CH}_4}/dy$ at $y = 0$, corresponds to the converted methane rate, which is the change in concentration in the bulk gas stream. To fit the experimental data, we assumed that r_{st} could be expressed by a power-law-type reaction rate with an upper limit in our previous paper [15],

$$r_{st} = \min \left\{ k_{st} (p_{\text{CH}_4})^a (p_{\text{H}_2\text{O}})^b, r_c (= \text{const.}) \right\}. \quad (3)$$

This expression for the reaction rate indicates that when the partial pressures of methane and steam, p_{CH_4} and $p_{\text{H}_2\text{O}}$ respectively, are sufficiently large, the reaction rate settles to a constant value of r_c . In many papers that represented the reaction rate of SMR with a power-law-type equation, the exponential index a has a value close to unity, whereas the absolute value of b is smaller than that of a at the wide range of the concentration ratio of steam to methane, i.e., the steam-to-carbon ratio [15,17–19]. Moreover, the change in $p_{\text{H}_2\text{O}}$ is generally not significant because the steam-to-carbon ratio is relatively large in actual conditions of reactors. Therefore, we assume that the reaction rate is proportional to the concentration of methane and independent of the concentration of steam as follows:

$$r_{st} = \min \left\{ kC, r_c (= \text{const.}) \right\}, \quad (4)$$

where, for simple notation, C and k are the concentration of methane and the reaction rate constant, respectively. Fig. 1(a) shows a sketch of the reaction rate as a function of the concentration. If the concentration is larger than the threshold concentration C_c , which is calculated as r_c/k , the reaction rate becomes constant.

By integrating Eq. (2) and using Eq. (4), the local concentration of methane can be expressed as a function of the position y . As shown in Fig. 1(b), we consider three different cases with different values of the concentration at the interface between the gas flow and the catalyst layer. Hereafter, such a concentration is denoted as C_{sur} .

When C_{sur} is smaller than C_c , the local reaction rate depends on the concentration at position y , as in mode [i] in Fig. 1(b). In mode [ii], a region in which the reaction rate is constant appears in the upper layer. Upon further increasing C_{sur} , the reaction rate of the entire field becomes constant. The diffusion equation at each mode is summarized in Table 1. Here, the diffusion coefficient is assumed to be constant because it is almost constant along the depth direction as described in our previous paper [15].

The equations in Table 1 are integrated with boundary conditions. For every case, $C = C_{\text{sur}}$ at $y = 0$ and $dC/dy = 0$ at $y = H$. In mode [ii], at the intermediate point of $y = y_c$, an additional condition that connects the two different reaction rates is necessary. The obtained solutions are also listed in Table 1, accompanied with the flux from the catalyst to the gas stream, which corresponds to the converted methane rate.

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