

# Methane/steam reforming kinetics for solid oxide fuel cells

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## Abstract

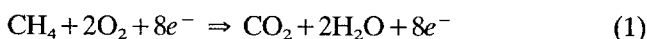
Experiments have been carried out to determine the kinetics of the methane/steam reforming process at anode materials of a solid oxide fuel cell. A nickel cermet was applied consisting of 80 wt.% ZrO<sub>2</sub> and 20 wt.% Ni. The temperature was varied from 700 to 940 °C, the CH<sub>4</sub> partial pressure from 0.11 to 0.33 bar, and the system pressure from 1.1 to 2.8 bar. The influence of the ratio H<sub>2</sub>O/CH<sub>4</sub> was studied, in particular, by increasing this quantity from 2.6 to 8. The tests showed that, within the accuracy of the data, no effect of the H<sub>2</sub>O partial pressure on the catalytic reforming process could be observed. Due to the high conversion rates of CH<sub>4</sub> at high temperatures, however, mass-transfer effects occurred, that must be taken into account when evaluating the steam-reforming data.

*Keywords:* Methane/steam reforming; Kinetics; Solid oxide fuel cells

## 1. Advantage of internal methane/steam reforming

Solid oxide fuel cells (SOFCs) operate at temperatures between 900 and 1000 °C using H<sub>2</sub>-containing gas mixtures, as fuel, and O<sub>2</sub> in the air, as oxidant. In contrast to low-temperature fuel cells the SOFC anode is not poisoned by CO. Experiments even show [1] that CO can be electrochemically converted. The H<sub>2</sub>/CO-rich gas can advantageously be produced by steam reforming from natural gas which consists of 85% methane.

The direct electrochemical conversion of CH<sub>4</sub> in a SOFC is an additional option [2], but is not yet feasible to date. Therefore, the direct oxidation of CH<sub>4</sub>:



must be approximated by a steam-reforming process:



associated with a CO-shift reaction:



The H<sub>2</sub> and CO produced according to reactions (2) can be electrochemically converted. The following reactions occur:

(i) cathode



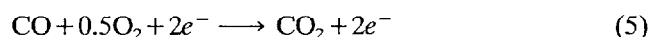
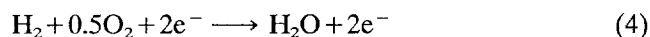
(ii) anode



(iii) anode



The overall reactions are:



The entropy losses of reaction (1) are small resulting in a high thermodynamic efficiency of  $\eta = 99.7\%$ . This is the reason that the direct conversion of CH<sub>4</sub> in SOFCs is a great challenge.

The reforming reaction (2) is strongly endothermic ( $\Delta H_{(2)} = +227.5$  kJ/mol at 1000 °C). The heat necessary for this reaction can be provided by the heat of the irreversible processes produced by the electrochemical reactions (4) and (5). At  $T = 1000$  °C we get an amount of:

$$Q_{\text{irr}} = 3T\Delta S_{(4)}^0 + T\Delta S_{(5)}^0 = -325.54 \text{ kJ/mol}$$

This is a surplus of  $Q = 98.04$  kJ/mol compared with  $\Delta H_{(2)}$  which must be eliminated from the cell by an air stream.

The free energy at 1000 °C available for the electrochemical oxidation of 3H<sub>2</sub> and CO is:

$$\Delta G_{\text{ges}} = 3\Delta G_{(4)}^0 + \Delta G_{(5)}^0 = -704.41 \text{ kJ/mol}$$

Compared with the free energy of the direct oxidation of  $\text{CH}_4$ ,  $\Delta G_{(1)} = -802.55$ , a loss of  $\Delta G$  by 12% occurs during the reforming process which results in a thermodynamic efficiency of  $\eta_{\text{th}} = 87.7\%$ . This value, however, is by 16.6 points higher than that of pure  $\text{H}_2$  oxidation ( $\eta_{\text{th H}_2} = 71.1\%$ ).

Both advantages – the cooling effect by the endothermic reaction and the high thermodynamic efficiency – are the reasons that internal steam reforming should be applied to the SOFC technology.

## 2. State-of-the-art

There is only a small amount of kinetic reforming data available obtained from nickel-cermet material. Lee et al. [3] investigated the  $\text{CH}_4$  conversion at different  $\text{NiO}/\text{ZrO}_2$  materials, varying the  $\text{H}_2\text{O}/\text{CH}_4$  ratio between 2 and 7.4. They crushed the cermet into 2.4 to 4.8 mm pieces and put them, for the tests, into a rotatable reactor basket. The reactor temperature varied from 800 to 1000 °C. The results showed a strong dependence of the  $\text{CH}_4$  conversion rate on the water partial pressure. The corresponding relationship is approximated by the following equation:

$$\frac{\dot{r}_{\text{CH}_4}}{P_{\text{CH}_4}} = k_0 \exp(-E/RT) p_{\text{H}_2\text{O}}^\gamma \quad (6)$$

where  $k_0$ ,  $E$  and  $\gamma$  vary for the different cermet compositions as follows:

(i) frequency factor:  $490 < k_0 < 4775 \left( \frac{\text{mol CH}_4}{\text{g catal. h}} \right)$

(ii) activation energy:  $7.55 \times 10^4 < E < 9.84 \times 10^4 \text{ (J/mol)}$

(iii)  $\text{H}_2\text{O}$ -reaction order:  $-1.28 < \gamma < -1.25$

It seems rather unlikely that the influence of the  $\text{H}_2\text{O}$  partial pressure is so strong. This becomes evident when Eq. (6) is rewritten by introducing the total pressure,  $P$ , and the molar ratios,  $X$ , of  $\text{CH}_4$  and  $\text{H}_2\text{O}$ , respectively. We get:

$$\dot{r}_{\text{CH}_4} = k_0 \exp(-E/RT) P^{1+\gamma} (X_{\text{CH}_4} X_{\text{H}_2\text{O}}) \quad (7)$$

For a constant feed gas composition and the  $\text{H}_2\text{O}$  reaction order of  $\gamma = -1.25$  we find that the absolute value of the  $\text{CH}_4$  conversion rate would decrease with increasing system pressure  $P$ , i.e., in spite of increasing  $p_{\text{CH}_4}$ . At a system pressure of 10 bar, for instance,  $\dot{r}_{\text{CH}_4}$  would decrease by a factor of 1.78 compared with ambient conditions.

The application of Lee's et al. [3] results to SOFC anode conditions is difficult since the conversion rate is related to the mass of the catalyst. This leads to the question, how deep the reaction penetrates into the

cermet. As the reforming reaction is very fast at the high SOFC temperatures, the reaction may occur only at the surface of the electrode and the thickness of the catalyst layer may have little influence. Therefore, the surface area may be a more practical reference quantity.

Parsons and Randall [4] report on experiments running a SOFC at 960 °C under internal  $\text{CH}_4$  reforming conditions. The active area was 41 mm × 41 mm. In these experiments the  $\text{H}_2\text{O}/\text{CH}_4$  ratio was equal to three. The results were fitted to the following equation:

$$\dot{r} = k(p_{\text{CH}_4})^{1.25} \quad (8)$$

The reaction rate constant,  $k$ , varied between 1.01 and 1.43 mol/(s m<sup>2</sup> bar<sup>1.25</sup>), dependent on whether the cell was operated under open-circuit conditions or load (40 mA/cm<sup>2</sup>). The temperature and the  $\text{CH}_4$  partial pressure have not been varied in the experiments.

Yentekakis et al. [5] studied the methane/steam reforming for Ni-YSZ cermets (yttria stabilized zirconia) in the temperature range from 800 to 930 °C. They mounted the Ni cermet on the bottom of a YSZ crucible. The  $\text{H}_2\text{O}/\text{CH}_4$  ratio investigated was very low and varied from 0.15 to 2. The tests indicated a strong influence of the  $\text{H}_2\text{O}$  partial pressure on the reforming reaction because of the  $\text{H}_2\text{O}$  understoichiometry. For each particular temperature level, however, a maximum of the conversion rate was identified, which is not well understood.

Zanibelli et al. [6] reported on steam reforming at crushed cermet materials treated by new techniques. The conversion rates displayed in the figures could not be evaluated in the present work with respect to kinetic data.

As a whole, the published experimental results show a considerable discrepancy, in particular with respect to the influence of the  $\text{H}_2\text{O}/\text{CH}_4$  parameter. The reasons may be of different origins. Since the reforming reaction is very fast at high temperatures, mass-transfer problems may have affected the results. Therefore, the main aim of the present work is to look for this transport phenomenon and to elaborate its influence on the reaction kinetics.

## 3. Analysis

The  $\text{CH}_4$  reforming reaction is governed by the  $\text{CH}_4$  partial pressure immediately at the surface of the nickel cermet, here denoted as wall partial pressure  $(p_{\text{CH}_4})_w$ . This must be lower than the bulk partial pressure  $(p_{\text{CH}_4})_b$ , to allow for a mass flux towards the cermet surface. The real situation at the catalyst surface is rather complicated since the reforming reaction is associated with an increase of the mole rate and accompanied by a CO shift reaction. For a rough estimation,

however, let us assume that Newton’s law for the convective mass transfer is applicable:

$$\dot{i} = \beta_c \frac{p_{CH_4,b} - (p_{CH_4})_w}{RT} \tag{9}$$

where  $\dot{i}$  is the mass flux in mol/(m<sup>2</sup> s N/m<sup>2</sup>),  $\beta_c$  the mass transfer coefficient in m/s,  $R$  the gas constant, and  $T$  the absolute temperature. If  $\dot{i}$  is known from the reforming experiment and  $\beta_c$  from empirical equations or tests,  $(p_{CH_4})_w$  can be calculated from Eq. (9) by writing:

$$(p_{CH_4})_w = (p_{CH_4})_b - \frac{\dot{i}RT}{\beta_c} \tag{10}$$

The CH<sub>4</sub> partial wall pressure has to appear in the kinetic expression of the CH<sub>4</sub> conversion assumed to be of the Arrhenius-type and linear in  $(p_{CH_4})_w$ :

$$\dot{i} = k_0 \left( 1 - \frac{p_{CO}p_{H_2}^3}{p_{CH_4}p_{H_2O}K_p} \right) \exp(-E/RT)(p_{CH_4})_w \tag{11}$$

The term in the first brackets:

$$f_K = \left( 1 - \frac{p_{CO}p_{H_2}^3}{p_{CH_4}p_{H_2O}K_p} \right) \tag{12}$$

accounts for the fact that  $\dot{i}$  vanishes when the gas composition has reached the thermodynamic equilibrium expressed by the equilibrium constant  $K_p$ .

Eq. (11) can also be written as a Newton-type equation, similar to Eq. (9):

$$\dot{i} = \beta_{ref} \frac{(p_{CH_4})_w}{RT} \tag{13}$$

where  $\beta_{ref}$  is equivalent to:

$$\beta_{ref} = RTk_0f_K \exp(-E/RT) \tag{14}$$

Eqs. (13) and (9) describe two mass-transfer phenomena combined in series. Only the total effect of both phenomena can be measured in the reforming experiment and merely the bulk partial pressure is known. Thus we define:

$$\dot{i} = \beta_{tot} \frac{(p_{CH_4})_b}{RT} \tag{15}$$

The relationship between  $\beta_c$ ,  $\beta_{ref}$  and  $\beta_{tot}$  is given by the equation of mass transfer in series:

$$\frac{1}{\beta_{tot}} = \frac{1}{\beta_c} + \frac{1}{\beta_{ref}} \tag{16}$$

from which  $\beta_{ref}$  can be derived when  $\beta_c$  and  $\beta_{tot}$  are known. Fig. 1 shows the relationship Eq. (16) and the range of application relevant in this investigation.

The determination of  $\beta_c$  can be made by applying empirical equations in the case of standard geometries. They are available in the form:

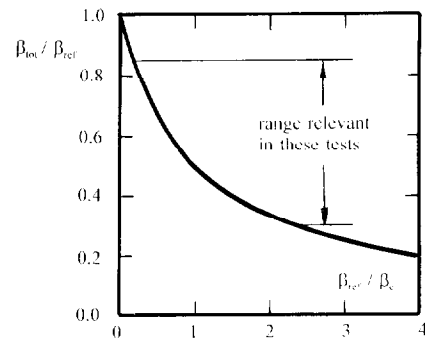


Fig. 1. Relationship between the total mass-transfer coefficient and that due to convection,  $\beta_c$ , and reforming,  $\beta_{ref}$ .

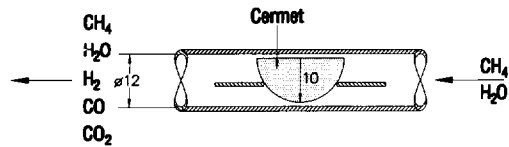


Fig. 2. Setup of the reforming experiments.

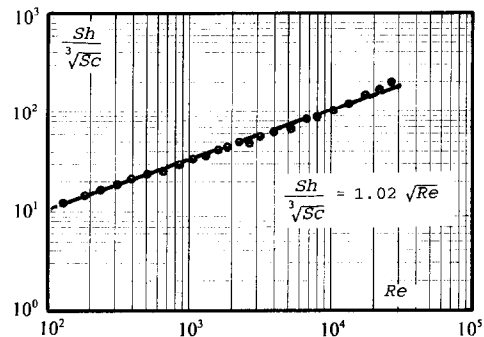


Fig. 3. Mass transfer from the cermet, experimentally determined by means of the naphthalene-to-air sublimation method.

$$Sh = \frac{\beta_c L}{D} = Af_{st} Re^{n1} Sc^{n2} \tag{17}$$

The Sherwood number,  $Sh$ , which represents the dimensionless convective mass-transfer coefficient depends on the flow conditions expressed by the Reynolds number,  $Re$ , and the Schmidt number,  $Sc$ . The constant  $A$  is a shape parameter. The Stefan function,  $f_{st} = P / (P - (p_{CH_4})_b)$  accounts for mass transfer at high partial pressure differences across the boundary layer.

To ensure that, for the evaluation of the present tests, the mass-transfer coefficient,  $\beta_c$ , is a reliable quantity, separate mass-transfer experiments using the method of naphthalene sublimation in air were carried out. In these experiments, the same geometrical conditions were applied as used for the reforming tests (see Fig. 2). The method is very well established and leads to results of high accuracy.

Fig. 3 represents the corresponding experimental data and a fit to a curve of the form of Eq. (17). Under naphthalene sublimation conditions  $f_{st} = 1$ . Thus the

actual mass-transfer coefficient can be determined by means of Eq. (18).

$$\beta_c = \frac{D}{L} \frac{P}{P - (p_{\text{CH}_4})_b} 1.02 \sqrt{Re} \sqrt[3]{Sc} \quad (18)$$

where  $D$  is the diffusion coefficient of  $\text{CH}_4$  in the entrance gas mixture consisting of  $\text{CH}_4$ ,  $\text{H}_2\text{O}$  and in some cases additionally of  $\text{N}_2$ . It is determined according to Ref. [7];  $L$  is the characteristic length of the test specimen, i.e. in the actual case the disk diameter. As the flow data and properties are known in each of the reforming tests the convective mass-transfer coefficient can be determined according to the above equation.

The evaluation procedure of the present reforming data was as follows:  $\beta_c$  and the  $\text{CH}_4$  partial wall pressure,  $(p_{\text{CH}_4})_w$ , were determined from Eqs. (18) and (10), respectively. The experimental conversion rate can now be referred to the wall partial pressure and be plotted against the quantity  $1/T$ .

The reforming tests were conducted, such as the gas composition was far from the thermodynamic equilibrium, i.e.  $f_k$  from Eq. (12) was not lower than 0.99.

#### 4. Experimental

A sketch of the experimental setup is shown in Fig. 4. The cermet is mounted in a steel tube of 12 mm internal diameter which can be heated up to 1000 °C in an electrical furnace. Five thermocouples are installed along the test section to register the actual temperature distribution. The feed gas ( $\text{CH}_4$ ) is mixed with steam immediately before entering the test tube. An entrance length of 300 mm was found to be sufficient to achieve

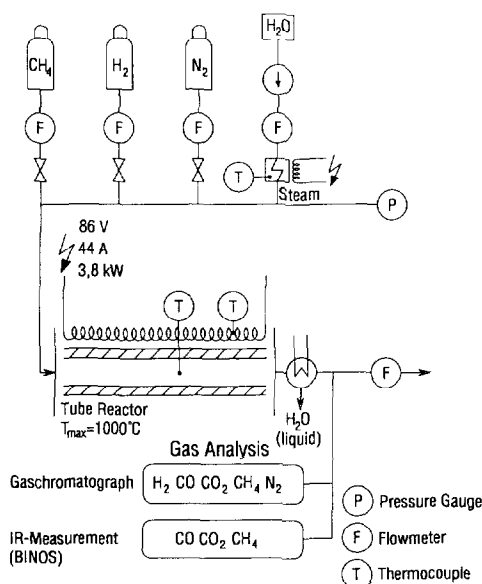


Fig. 4. Flow-sheet of the reforming experiments.

the required test temperature. The composition of the gas after the reforming process is analysed by means of gas chromatography and checked by a IR Binos Analyzer system. Both equipments yielded consistent results. Before the product gas reached the analyser, the steam was removed by cooling down to 2 °C.

Details of the test section are depicted in Fig. 2. The cermet is a semi-disk and is 1.4 mm thick. Its total surface area is  $3.86 \times 10^{-4} \text{ m}^2$ . The cermet material consists of 20 wt.% Ni and 80 wt.%  $\text{ZrO}_2$ . This specification, probably, does not meet the electrochemical requirements of a SOFC cermet. The electrochemical suitability, however, was of minor interest for the present reforming tests.

#### 5. Results

The conditions of the SOFC internal reforming process considerably vary along the channel length of the cell. For modelling purposes, therefore, a differential treatment of the problem is necessary. The experimental consequence is that the  $\text{CH}_4$  conversion rates must be kept low. In the present work values between 4 and 15%, in a few cases up to 25% are applied.

The effect of the temperature on the reaction was studied by increasing the temperature from 700 to 940 °C. The  $\text{CH}_4$  partial pressure was varied from 0.105 to 0.325 bar and the steam/methane ratio from 2.6 to 8. System pressures,  $P$ , were applied in the range from 1.1 to 2.8 bar. In addition, particular runs were provided with increased mass-flow rates to vary the ratio of  $\beta_c/\beta_{\text{ref}}$ .

An important requirement for kinetic experiments is the catalytic stability of the test specimen. Since the present experiments had to be carried out over a period of more than four months, the reference point conditions –  $T=900 \text{ °C}$ ;  $P=2.8 \text{ bar}$ ;  $\text{H}_2\text{O}/\text{CH}_4=8$ ;  $r_{\text{CH}_4}=2.317 \cdot 10^{-4} \text{ mol/s}$  – were checked after each run. No degradation effect was observed throughout the whole period.

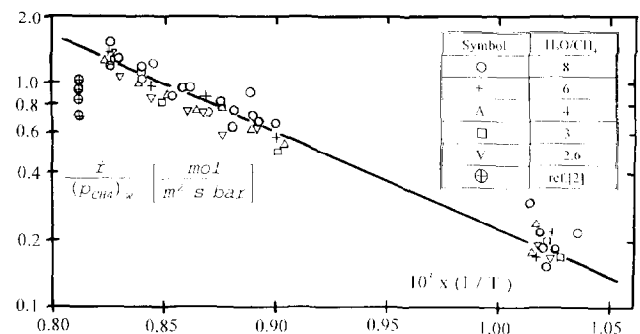


Fig. 5. Arrhenius plot of the reforming results. Data recalculated for mass-transfer effects (Eq. (19)).

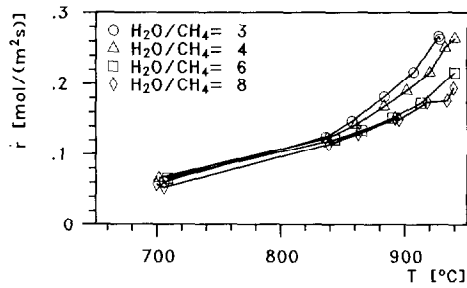


Fig. 6. Methane-conversion rates vs. temperature and  $\text{H}_2\text{O}$  partial pressure.

Fig. 5 represents the experimental data recalculated for the effects of mass transfer. The most important result is that the ratio of  $\text{H}_2\text{O}/\text{CH}_4$  has no dramatic effect on the reforming process. The departures observed for the raw data (Fig. 6) can be explained by mass-transfer processes. The recalculated data show a standard deviation of  $\sigma = \pm 0.147$ . The pre-exponential constant is equal to  $k_0 = 4274 \text{ mol}/(\text{s m}^2 \text{ bar})$  and the activation energy evaluated from the slope of the curve in Fig. 5 is determined to be  $E = 8.2 \times 10^4 \text{ J/mol}$ . Thus, the kinetic of the  $\text{CH}_4$  reforming process can be described by following equation:

$$\frac{\dot{r}}{(p_{\text{CH}_4})_w} = 4274 \left( 1 - \frac{p_{\text{CO}} p_{\text{H}_2}^3}{p_{\text{CH}_4} p_{\text{H}_2\text{O}} K_p} \right) \times \exp\left( - \frac{8.2 \times 10^4}{RT} \right) \frac{\text{mol}}{\text{s m}^2 \text{ bar}} \quad (19)$$

Eq. (19) can be rewritten in terms of a mass-transfer coefficient to make the  $\text{CH}_4$  conversion rate consistent for use in Eq. (16):

$$\beta_{\text{ref}} = 4.274 \times 10^{-2} RT \left( 1 - \frac{p_{\text{CO}} p_{\text{H}_2}^3}{p_{\text{CH}_4} p_{\text{H}_2\text{O}} K_p} \right) \times \exp\left( - \frac{8.2 \times 10^4}{RT} \right) \frac{\text{m}}{\text{s}} \quad (20)$$

## 6. Discussion of the results

The present results demonstrate that for high molar conversion fluxes the conditions of the mass transfer from the bulk to the catalytic surface must be considered carefully. Uncontrolled flow conditions can cause essential errors during the evaluation of the test data.

The activation energy  $E = 8.2 \times 10^4 \text{ J/mol}$  is a reasonable value for the heterogeneous  $\text{CH}_4$  conversion and is in satisfactory agreement with the findings of Lee et al. [3] mentioned above.

The evaluation of the data of Ref. [5] taken at the maximum of the reaction rate for the different temperatures, yields an activation energy in the order of

$2.75 \times 10^5 \text{ J/mol}$  which is an unusually high value for  $\text{CH}_4$  catalytic reactions. Furthermore, the reaction rates are lower by a factor of five at  $900 \text{ }^\circ\text{C}$  and by 20 at  $800 \text{ }^\circ\text{C}$  compared with the present results.

The data of Parsons and Randall [4] taken at a SOFC cell during operation are by 50 to 25% lower than the authors' recalculated results. Having in mind, that in the present work the corrections for mass-transport effects at  $940 \text{ }^\circ\text{C}$  were in the order of 50 to 100%, a reasonable agreement of both results can be observed.

In order to get an impression of how fast the reforming reaction is, the reforming conversion rate predicted for  $900 \text{ }^\circ\text{C}$  and for a  $\text{CH}_4$  partial pressure of 0.17 bar is referred to the molar flux necessary to sustain a current density of  $j = 3000 \text{ A/m}^2$  by means of an electrochemical reaction. This ratio is a factor of 42 in favour of the reforming process. For  $1000 \text{ }^\circ\text{C}$ , this value is even 81, at  $700 \text{ }^\circ\text{C}$  still 7.4. The consequences for the temperature distribution of a cell with internal reforming are shown in previous calculations [8,9]. The fast endothermic reaction occurs immediately in the entrance section of the fuel cell and leads to local subcooling in this area.

The evaluation of the experimental conversion data requires the knowledge of the  $\text{CH}_4$  diffusion coefficient,  $D$ , in multiple gas mixtures. The value was calculated for the entrance conditions though it is a local quantity. This was done since uncertainties exist in determining the magnitude of the diffusion coefficient and since an appropriate mean value is not easy to define for the present geometry of the cermet. The mass-transfer coefficient is linearly affected by the diffusion coefficient. The more the convective mass transfer becomes a limiting quantity, i.e. for high temperatures, the more important is the accuracy in determining  $D$ .

## 7. Conclusions

Nickel cermet, the anode material of SOFCs, is a suitable catalyst to apply internal methane/steam reforming. The reaction is very fast at high temperatures between 900 and  $1000 \text{ }^\circ\text{C}$  and may be affected by mass-transfer phenomena.

In the present contribution, an apparent effect of  $\text{H}_2\text{O}$  partial pressure on the reforming catalysis could be identified as a mass-transport process from the bulk to the catalytic wall. Though varying the  $\text{H}_2\text{O}/\text{CH}_4$  ratio from 2.6 to 8 no systematic influence of  $\text{H}_2\text{O}$  on the reforming catalysis could be observed, which is in contradiction to the findings in the literature.

An equation can be proposed for the reforming kinetics in the form of the Arrhenius-type independence of the  $\text{H}_2\text{O}$  partial pressure and proportional to the  $\text{CH}_4$  partial pressure. The activation energy, derived

from the slope of the Arrhenius equation is  $E = 8.2 \times 10^4$  J/mol.

### 8. List of symbols

$A$	shape parameter (–)
$D$	diffusion coefficient (m <sup>2</sup> /s)
$E$	activation energy (J/mol)
$f_{st}$	Stefan factor (–)
$f_k$	equilibrium function (–)
$G$	Gibbs free energy (J/mol)
$H$	reaction enthalpy (J/mol)
$j$	current density (A/m <sup>2</sup> )
$k$	pre-exponential factor (case-dependent)
$K_p$	equilibrium constant (case-dependent)
$L$	characteristic length (m)
$n$	exponent (–)
$P$	absolute pressure (N/m <sup>2</sup> )
$p$	partial pressure (N/m <sup>2</sup> )
$Q$	heat rate (J/mol)
$R$	gas constant (8.314 J/(mol K))
$\dot{r}$	conversion rate (mol/(m <sup>2</sup> s))
$S$	entropy (J/(mol K))
$T$	absolute temperature (K)
$u$	velocity (m/s)

### Greek letters

$\beta$	mass-transfer coefficient (m/s)
$\gamma$	exponent (–)
$\eta$	fluid viscosity (kg/(m s))
$\rho$	fluid density (kg/m <sup>3</sup> )

### Characteristic groups

$$\text{Reynolds number } Re = \frac{UL\rho}{\eta}$$

$$\text{Schmidt number } Sc = \frac{\eta}{\rho D}$$

$$\text{Sherwood number } Sh = \frac{\beta L}{D}$$

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