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Effects of pressure and flow rate on the efficiency and performance of autothermal reforming systems for hydrogen production

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Abstract

Computational modelling for microchannel reactor design was conducted in the attempt to fully understand autothermal reforming phenomena in continuous flow reactors. The effects of pressure and flow rate on the efficiency and performance were evaluated by performing computational fluid dynamics under different design conditions. The reactor efficiency and performance were assessed by means of the reactant conversion, product yield, reaction rate, hydrogen productivity, and output power. Recommendations for designing an autothermal reforming system were made and strategies for performing efficient operation were set forth. The results indicated that the pressure and flow rate set strict limits on operation, and there is a trade-off between high productivity and low conversion. The pressure and flow rate play competing roles in the reactor efficiency and performance. Lower pressures and flow rates can increase the conversion and yield, but higher pressures and flow rates can significantly improve the hydrogen productivity and output power, which is essential to the success of start-up and acceleration of the downstream equipment. There may exist an optimum pressure and flow rate in terms of both efficiency and performance. The calculated output power is of the order of thousands of kilowatts per cubic meter.
Keywords: Reactor performance; Reactor efficiency; Chemical processes; Hydrogen productivity; Autothermal reforming; Transport phenomena

1. Introduction

Elementary hydrogen finds its principal industrial application in the manufacture of ammonia [1, 2] and in the hydrogenation of carbon monoxide and organic compounds [3, 4]. There are certain important properties of molecular hydrogen. The extremely low melting and boiling points result from weak forces of attraction between the molecules. The existence of these weak intermolecular forces is also revealed by the fact that, when hydrogen gas expands from high to low pressure at room temperature, its temperature rises, whereas the temperature of most other gases falls. According to thermodynamic principles, this implies that repulsive forces exceed attractive forces between hydrogen molecules at room temperature-otherwise, the expansion would cool the hydrogen. The cooling effect becomes so pronounced at temperatures below that of liquid nitrogen that the effect is utilized to achieve the liquefaction temperature of hydrogen gas itself.

The most important industrial method for the production of hydrogen is the catalytic steam-hydrocarbon process, in which gaseous or vaporized hydrocarbons are treated with steam at high pressure over a nickel catalyst to produce carbon oxides and hydrogen [5, 6]. The primary reaction products are processed further in various ways, depending on the desired application of the hydrogen [7, 8]. Another important process for hydrogen production is the noncatalytic partial oxidation of hydrocarbons under elevated pressures [9, 10]. This process requires a feed system for delivering

precise rates of fuel and oxygen, burners of special design to give rapid mixing of the reactants, a refractory-lined reactor, and a cooling system to recover heat from the effluent gases [11, 12]. The latter process is exothermic, in contrast to the endothermic steam-hydrocarbon process.

In a third process, which is referred to as the pressure catalytic partial oxidation method, the two preceding processes are combined to maintain the required reaction temperature without external heating of the catalyst bed [13, 14]. Superheated steam and hydrocarbons are mixed, preheated, and blended with heated oxygen in a diffuser at the top of the catalytic reactor [15, 16]. The oxygen reacts with the hydrocarbons in a space above the catalyst [17, 18]. The reactants then pass through a bed of nickel catalyst in which the steam-hydrocarbon reactions proceed almost to chemical equilibrium [17, 18]. A commercial method has been developed for separating hydrogen from carbon monoxide synthesis gas by diffusion. The gas flows under pressure through bundles of tiny hollow polyester fibres through whose walls the hydrogen passes.

Little research has been conducted to understand and optimise the physical and chemical processes in millisecond microchannel reactors, in which it may be impossible to simultaneously achieve high efficiency and performance. In the present study, computational modelling for microchannel reactor design was conducted in the attempt to fully understand autothermal reforming phenomena in continuous flow reactors. The effects of pressure and flow rate on the efficiency and performance were evaluated by performing computational fluid dynamics under different design conditions. The reactor efficiency and performance were assessed by means of the reactant conversion, product yield, reaction rate, hydrogen productivity, and output power. Recommendations for designing an autothermal reforming system were made and strategies for performing efficient operation were set forth. The present study aims to understand and optimise the autothermal reforming process in a millisecond microchannel reactor. Particular emphasis is placed upon the effects of pressure and flow rate on the efficiency and performance of the reactor system.

2. Model development

2.1. Description of the system

Preferably, chemical reactors are designed to minimize carbon formation and maximize hydrogen production by optimizing operation conditions. In industry, steam reforming reactions are typically the preferred processes for the production of hydrogen [19, 20]. Endothermic steam reforming processes do not require oxygen but external heat sources are necessary [21, 22]. Additionally, steam reforming offers advantages of and higher thermal efficiencies and hydrogen yields [23, 24]. In contrast, autothermal reforming reactions are thermally neutral processes, which allows for reactors to reduce fuel slip, but complex and expensive oxygen separation units are usually necessary [23, 24]. Autothermal reforming processes do not require external heat sources, thereby radically simplifying the design process and substantially reducing the start-up time [23, 24]. Heat is provided internally in the autothermal reforming process. Specifically, the catalytic partial oxidation of the fuel is accompanied by the release of heat, which raises the temperature so that the steam reforming reaction can proceed on the catalyst surface [23, 24]. Consequently, physical processes that transfer energy and mass must be considered essential for autothermal reforming. Autothermal reforming reactors have other attributes, for example, greater potential for economies of scale, lower capital cost, and relative compactness [25, 26]. For such reactors to operate properly, however, feed compositions must be controlled and adjusted precisely to improve the efficiency and performance while preventing coke formation.

A microchannel reactor is utilized to continuously produce hydrogen by auto-thermally reforming a methanol feed. The reactor in which the autothermal reforming reaction of methanol is carried out is illustrated schematically in Figure 1 with the wall and washcoat indicated. Unless otherwise specified,

the coating is 0.10 mm in thickness, the wall is 0.20 mm in thickness, the channels are 0.7 mm in height, and the flowthrough path is 50.0 mm in length. Unless otherwise noted, the feed contains methanol, steam, and oxygen in a molar ratio of methanol to steam of 100:100 and in a molar ratio of methanol to oxygen of 100:23 with oxygen as the limiting reactant, such that the oxygen concentration is minimal on the catalyst surface. The reactor operates with an inlet velocity of 2.0 m/s and with an inlet temperature of 100 °C at one atmosphere pressure, whereupon the reaction proceeds by the contact time in milliseconds.
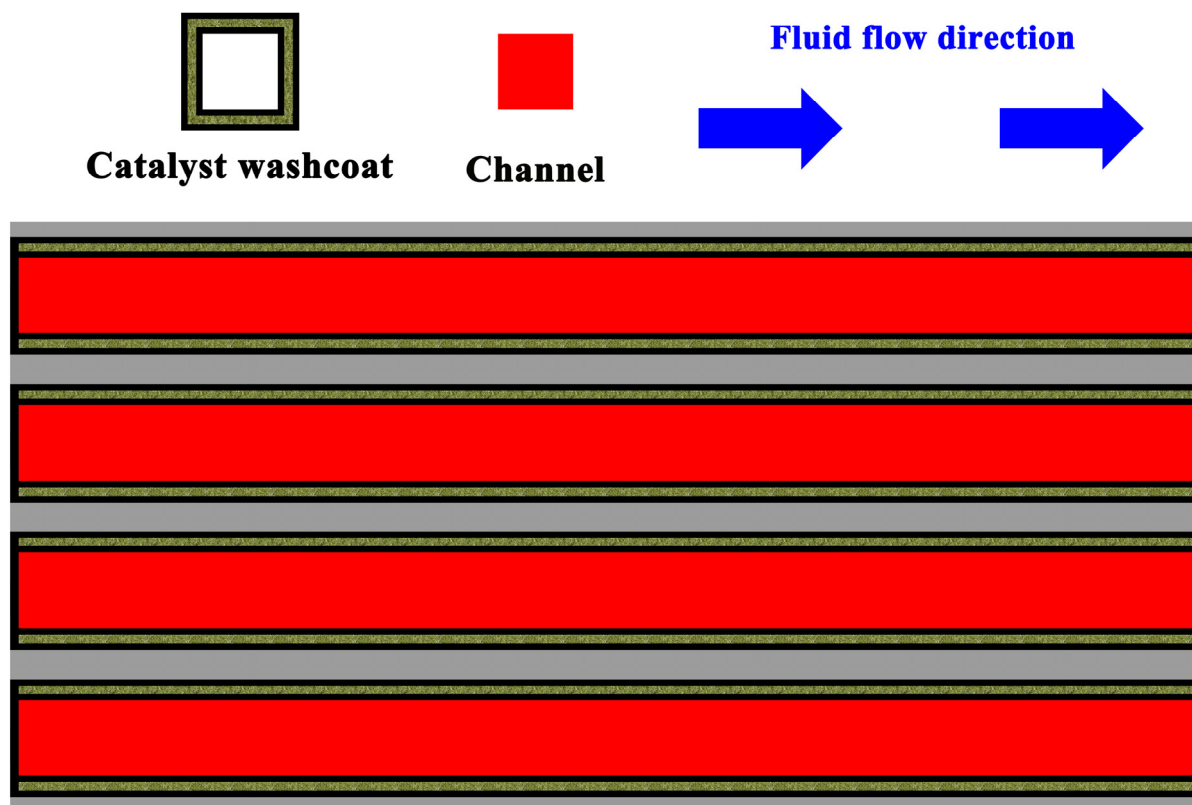

Figure 1. Illustration representation of the autothermal reforming reactor for the production of hydrogen with the fluid flow direction indicated.
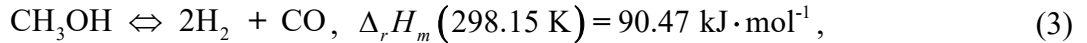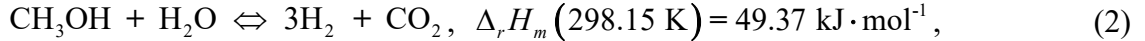
2.2. Chemical kinetics

Chemical kinetics is the branch of physical chemistry that is concerned with understanding the rates of chemical reactions. It is to be contrasted with thermodynamics, which deals with the direction in which a process occurs but in itself tells nothing about its rate. The principles of chemical kinetics apply to purely physical processes as well as to chemical reactions. One reason for the importance of kinetics is that it provides evidence for the mechanisms of chemical processes. Besides being of intrinsic scientific interest, knowledge of reaction mechanisms is of practical use in deciding what is the most effective way of causing a reaction to occur. Many commercial processes can take place by alternative reaction paths, and knowledge of the mechanisms makes it possible to choose reaction conditions that favour one path over others. A chemical reaction is, by definition, one in which chemical substances are transformed into other substances, which means that chemical bonds are broken and formed so that there are changes in the relative positions of atoms in molecules. At the same time, there are shifts in the arrangements of the electrons that form the chemical bonds. A description of a reaction mechanism must therefore deal with the movements and speeds of atoms and electrons. The detailed mechanism by which a chemical process occurs is referred to as the reaction path, or pathway. The rate of a reaction is defined in terms of the rates with which the products are formed and the reactants are consumed. For chemical systems it is usual to deal with the concentrations

of substances, which is defined as the amount of substance per unit volume. The rate can then be defined as the concentration of a substance that is consumed or produced in unit time. Sometimes it is more convenient to express rates as numbers of molecules formed or consumed in unit time.

At the temperatures involved, the oxidation reaction over the catalyst surface is extremely rapid

$$CH_3OH + 1.5O_2 \Leftrightarrow H_2O + CO_2, \quad \Delta_r H_m(298.15\ K) = -726.60\ kJ \cdot mol^{-1}. \tag{1}$$
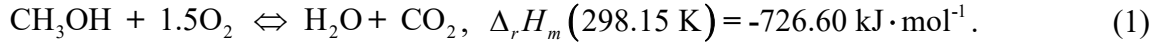
The reforming and decomposition reactions which convert methanol to hydrogen in the reactor are endothermic are represented by

$$CH_3OH + H_2O \Leftrightarrow 3H_2 + CO_2, \quad \Delta_r H_m(298.15\ K) = 49.37\ kJ \cdot mol^{-1}, \tag{2}$$

$$CH_3OH \Leftrightarrow 2H_2 + CO, \quad \Delta_r H_m(298.15\ K) = 90.47\ kJ \cdot mol^{-1}, \tag{3}$$

in which H_m is the molar enthalpy and the subscript r refers to the reaction.

The water-gas shift reaction is represented by

$$CO + H_2O \Leftrightarrow H_2 + CO_2, \quad \Delta_r H_m(298.15\ K) = -41.10\ kJ \cdot mol^{-1}. \tag{4}$$
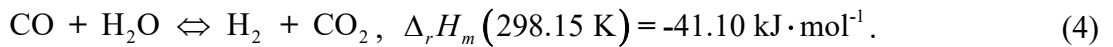
An oxygen-methanol molar ratio of 25:100 is stoichiometrically required. The stoichiometric relationship is determined by

$$CH_3OH + 0.25O_2 + 0.5H_2O \Leftrightarrow 2.5H_2 + CO_2, \quad \Delta_r H_m(298.15\ K) = -12.00\ kJ \cdot mol^{-1}. \tag{5}$$
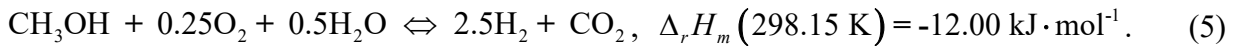
The kinetic model [27, 28] accounts for the reforming, decomposition, and water-gas shift processes. The kinetic model [29, 30] accounts for the oxidation process. The following equations allow the calculation of the rates of the above reactions:

$$r_R = \left[k_R K^{\vartheta}_{CH_3O^{(\alpha)}} \left(p_{CH_3OH} p_{H_2}^{-0.5} \right) \left(1 - k_R^{-1} p_{H_2}^3 p_{CO_2} p_{CH_3OH}^{-1} p_{H_2O}^{-1} \right) c_{S_\alpha}^{\pi} c_{S_{\alpha'}}^{\pi} \right]$$
$$\cdot \left[\left(1 + K^{\vartheta}_{CH_3O^{(\alpha)}} \left(p_{CH_3OH} p_{H_2}^{-0.5} \right) + K^{\vartheta}_{HCOO^{(\alpha)}} p_{CO_2} p_{H_2}^{0.5} + K^{\vartheta}_{OH^{(\alpha)}} \left(p_{H_2O} p_{H_2}^{-0.5} \right) \right) \left(1 + K^{0.5}_{H^{(\alpha')}} p_{H_2}^{0.5} \right) \right]^{-1}, \tag{6}$$

$$r_D = \left[k_D K^{\vartheta}_{CH_3O^{(\beta)}} \left(p_{CH_3OH} p_{H_2}^{-0.5} \right) \left(1 - k_D^{-1} p_{H_2}^2 p_{CO} p_{CH_3OH}^{-1} \right) c_{S_\beta}^{\pi} c_{S_{\beta'}}^{\pi} \right]$$
$$\cdot \left[\left(1 + K^{\vartheta}_{CH_3O^{(\beta)}} \left(p_{CH_3OH} p_{H_2}^{-0.5} \right) + K^{\vartheta}_{OH^{(\beta)}} \left(p_{H_2O} p_{H_2}^{-0.5} \right) \right) \left(1 + K^{0.5}_{H^{(\beta')}} p_{H_2}^{0.5} \right) \right]^{-1}, \tag{7}$$

$$r_W = \left[k_W^{\vartheta} K^{\vartheta}_{OH^{(\alpha)}} \left(p_{CO} p_{H_2O} p_{H_2}^{-0.5} \right) \left(1 - k_W^{-1} p_{H_2} p_{CO_2} p_{CO}^{-1} p_{H_2O}^{-1} \right) \left(c_{S_\alpha}^{\pi} \right)^2 \right]$$
$$\cdot \left[\left(1 + K^{\vartheta}_{CH_3O^{(\alpha)}} \left(p_{CH_3OH} p_{H_2}^{-0.5} \right) + K^{\vartheta}_{HCOO^{(\alpha)}} p_{CO_2} p_{H_2}^{0.5} + K^{\vartheta}_{OH^{(\alpha)}} \left(p_{H_2O} p_{H_2}^{-0.5} \right) \right) \right]^{-2}, \tag{8}$$

$$r_O = 10^7 e^{-115000 R^{-1} T^{-1}} p_{CH_3OH}^{0.18} p_{O_2}^{0.18} p_{H_2O}^{-0.14}, \tag{9}$$

in which ϑ, K, c_S^{π}, and k is the composite parameter, equilibrium constant, total surface concentration, and rate constant, respectively, the subscripts α' and β' and the superscripts (α') and (β') refer to the adsorption site, the subscripts α and β and the superscripts (α) and (β) refer to the active site related to the reaction, and the subscripts R, D, W, and O refer to the reforming, decomposition, water-gas shift, and oxidation reactions, respectively.

2.3. Model validation

The model is validated experimentally by comparisons with measurements [31, 32]. The chemical reaction proceeds in a microchannel reactor. The reaction mixture is delivered to the channels using a manifold that is triangular in shape. Additionally, the flowthrough path is 33.0 mm in length, and the

channels are 0.6 mm in height and 0.5 mm in width. Furthermore, the reactor and vaporizer are identical in structure and shape, and the channel walls are manufactured with stainless steel. Numerical simulations are carried out using the model with the reactor dimensions described above. Comparisons are performed between the measured and calculated methanol conversions and hydrogen production rates at different volumetric flow rates, and the results are presented in Figures 2(a) and 2(b), respectively, at different channel wall temperatures. Sophisticated means are employed to measure the volumetric flow rate of the liquid feed entering the vaporizer chamber at room temperature, while the reaction is proceeding in the reaction chamber. In the channel wall temperature range specified, the calculated results are in reasonably good agreement with the measured data.
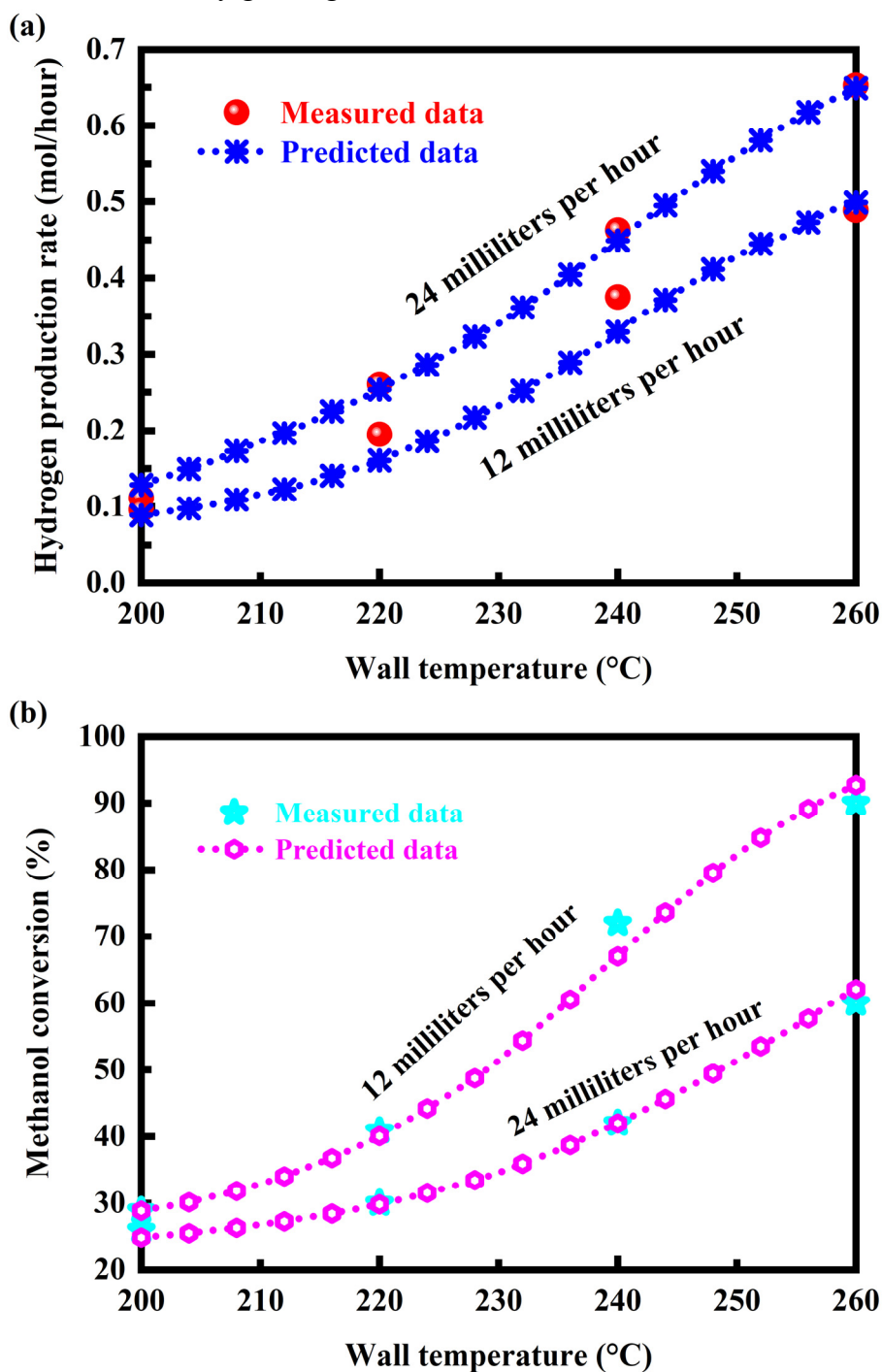

Figure 2. (a) Comparisons of the measured and calculated hydrogen production rates at different channel wall temperatures and volumetric flow rates. (b) Comparisons of the measured and calculated methanol conversions at different channel wall temperatures and volumetric flow rates. Reasonably good agreement is obtained between the measured and calculated data.

3. Results and discussion

3.1. Effect of pressure

The two-dimensional contour maps of methanol mole fraction in the reactor under different pressure conditions are illustrated in Figure 3 with the fluid flow direction indicated. Pressure significantly affects the species uniformity in the reactor. At higher pressures, there are no significant species gradients in the fluid flow direction. However, significant species gradients do exist in the transverse direction. High pressures will lead to much steeper species gradients in a direction perpendicular to the fluid flow direction. Accordingly, the rate of diffusion of reactant molecules to the surface of the catalyst is sufficiently low such that the reactor may operate in a mass-transfer controlled regime. Higher pressures tend to facilitate the reaction due to increased reactant concentrations [33, 34]. However, there occurs significant fuel breakthrough or slip, since the methanol mole fraction is high near the exit of the reactor. In contrast, the reaction takes place rapidly in the reactor at lower pressures.
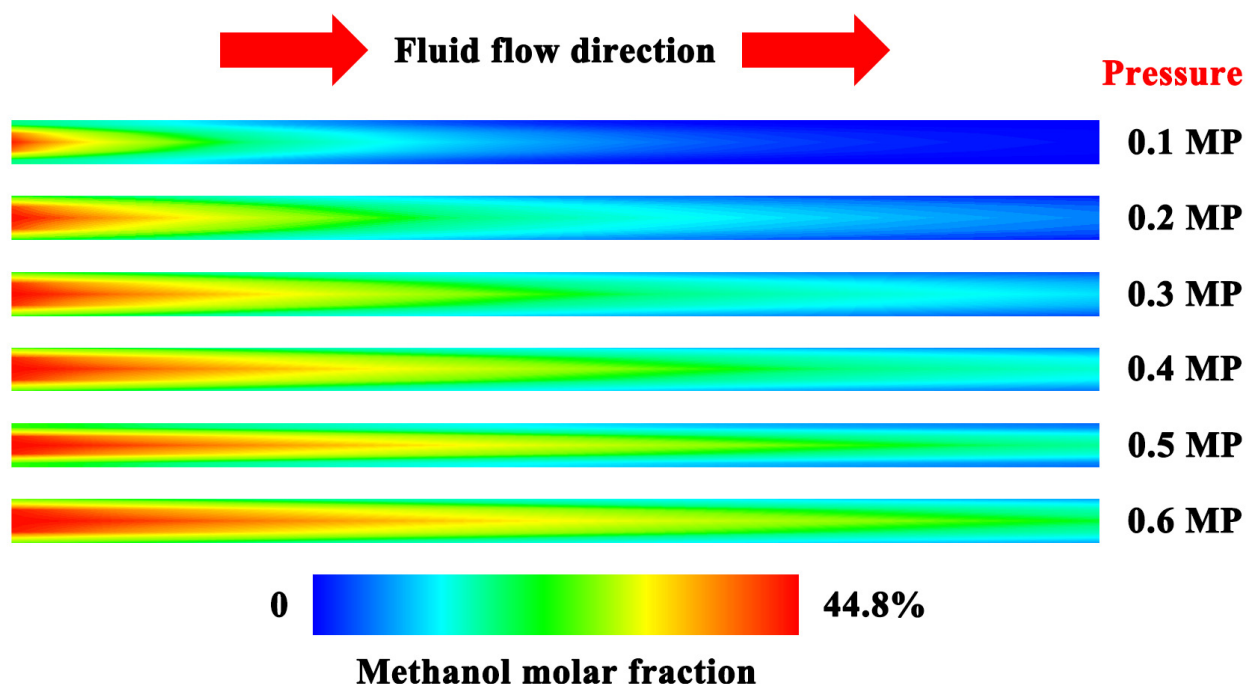

Figure 3. Two-dimensional contour maps of methanol mole fraction in the reactor under different pressure conditions with the fluid flow direction indicated.

The methanol and hydrogen mole fraction profiles along the channel centreline are presented in Figure 4 under different pressure conditions. Lower pressures allow higher conversion of methanol to hydrogen. For example, at one atmosphere pressure, the maximum conversion of methanol to hydrogen is attained, and the reaction is almost completed in the reactor. As the pressure is increased, the methanol mole fraction does not drop rapidly and the hydrogen mole fraction increases slightly along the flowthrough path. At the reactor outlet, the methanol mole fraction increases rapidly and the hydrogen mole fraction decreases significantly with increasing the pressure. Operation of the reactor at high pressures may be disadvantageous, at which there occurs significant fuel breakthrough or slip and thus considerably lower velocities may be required to achieve high conversion and yields. Some processes are so fast that special techniques have to be used to study them. There are two difficulties with fast reactions. One is that the time that it takes to mix reactants or to change the temperature of the system may be significant in comparison with the half-life, so that the initial time cannot be measured accurately. The other difficulty is that the time it takes to measure the amounts of substances may be comparable with the half-life of the reaction. The methods used to overcome these difficulties fall into two classes: flow methods and pulse and probe methods. The kinetic behaviour of an ordinary chemical

reaction is conventionally studied in the first instance by determining how the reaction rate is influenced by certain external factors such as the concentrations of the reacting substances, the temperature, and sometimes the pressure. Sometimes reaction rates depend on reactant concentrations in a more complicated way.
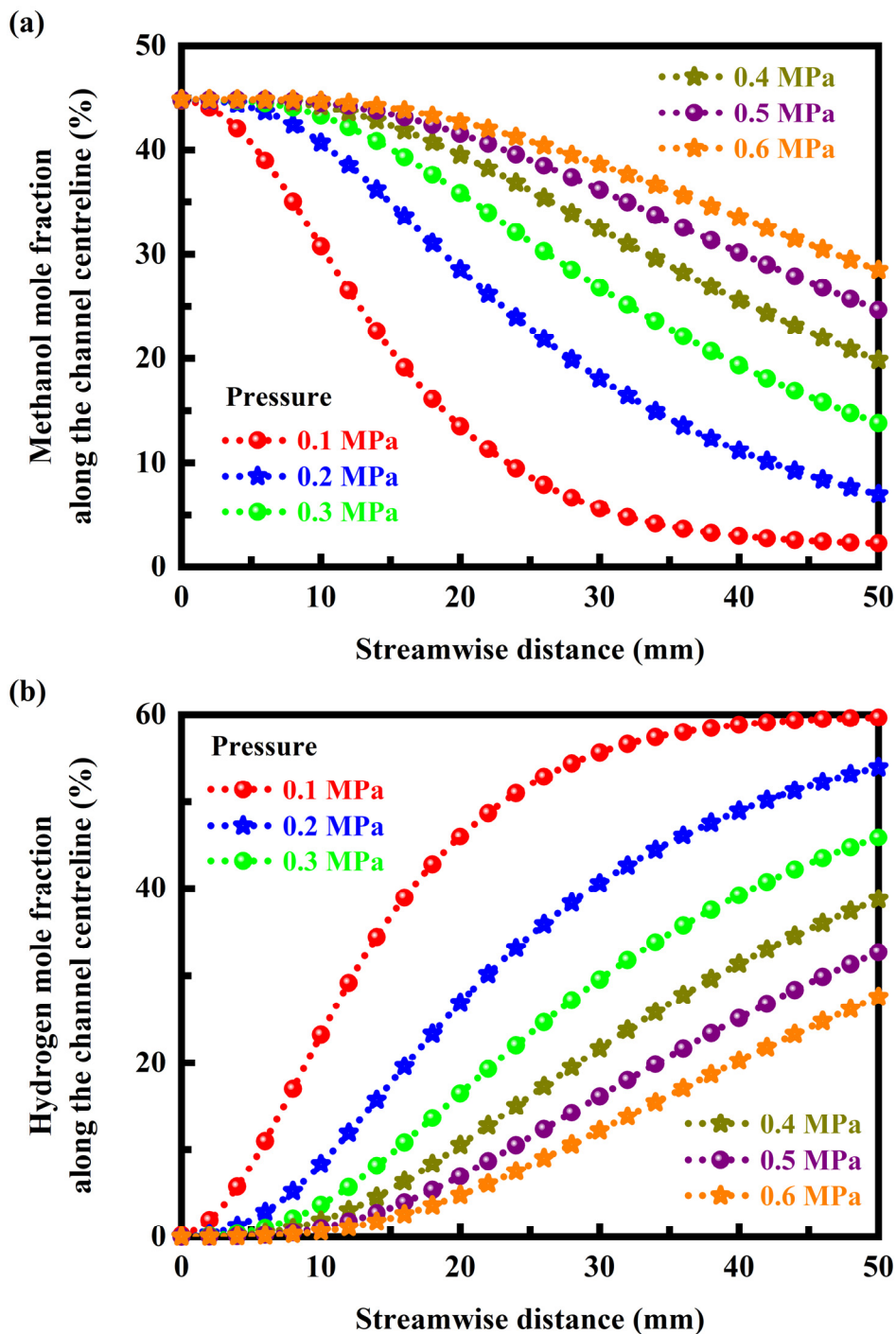
(a)

(b)

Figure 4. (a) Effect of pressure on the methanol mole fraction along the channel centreline. (b) Effect of pressure on the hydrogen mole fraction along the channel centreline.

The effects of pressure on the methanol conversion, hydrogen yield, and hydrogen productivity of the reactor are illustrated in Figure 5 wherein the results obtained for output power are also presented. The conversion and yield are governed largely by the pressure. Lower pressures are advantageous, as they tend to maximize the conversion and yield. As the pressure is increased, both reactant conversion and product yield decrease significantly. Higher pressures cause the conversion and yield to decrease. At higher pressures, the conversion of methanol to hydrogen is limited by the diffusion resistances of reactant molecules from the bulk fluid phase to the catalyst surface as discussed above, which will

inevitably lead to lower conversion and yields. For example, the conversion is about 60 percent and the yield is about 28 percent, if the reactor operates at a pressure of six atmospheres. However, higher pressures may be preferred, as they tend to significantly improve the hydrogen productivity and output power of the reactor, which is essential to the success of start-up and acceleration of the downstream equipment. For example, the maximum hydrogen productivity and output power are obtained in an incomplete conversion regime. Consequently, it is impossible to simultaneously achieve high conversion and productivity in the reactor. At higher pressures, there is a trade-off between low methanol conversion and high hydrogen productivity. At the pressures involved, the calculated output power of the reactor is of the order of thousands of kilowatts or megawatts per cubic meter. As the pressure is increased further, however, the reactor operation may be limited by strong diffusion resistances, which will inevitably lead to lower hydrogen productivity and output power.
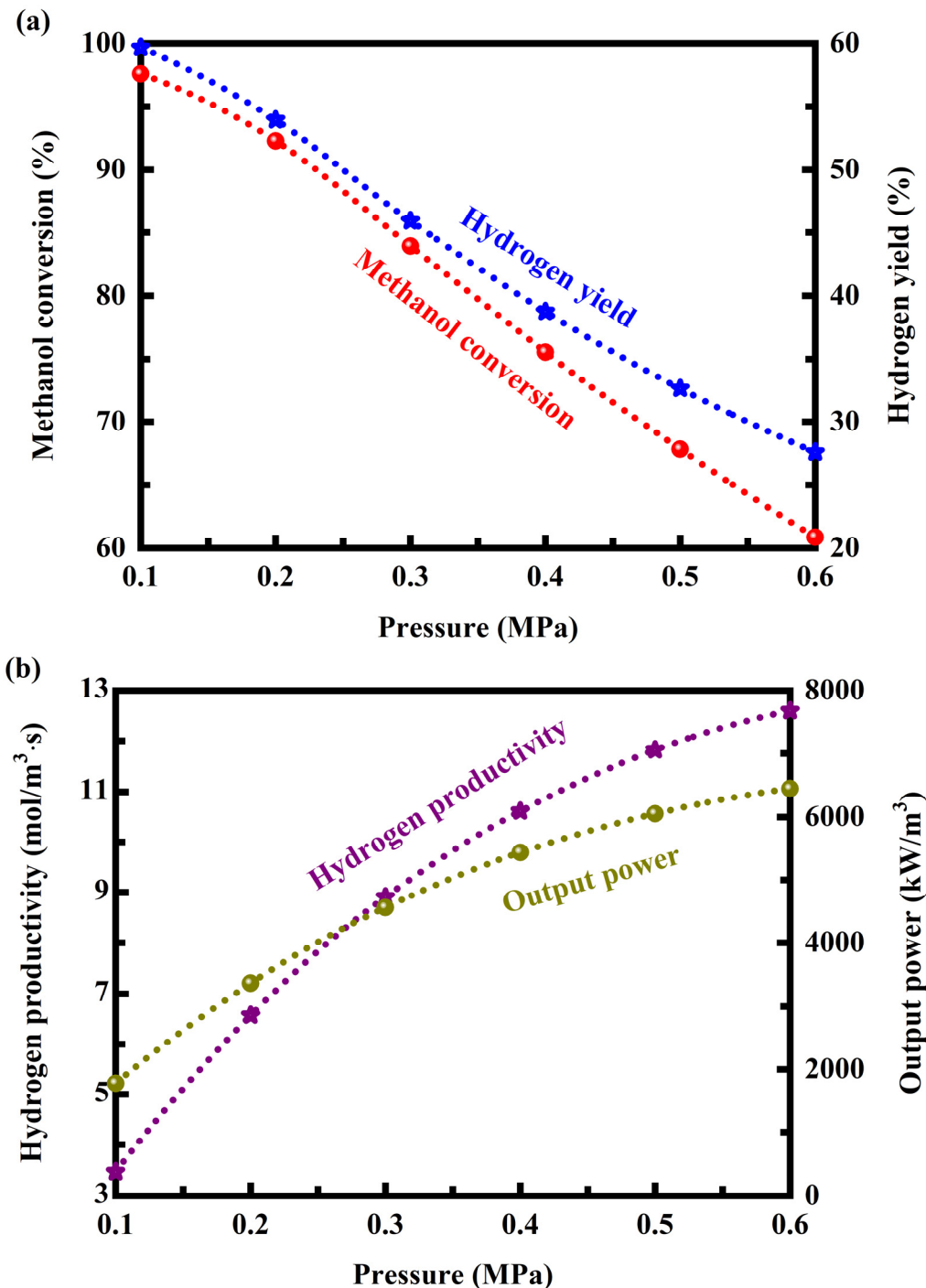

Figure 5. (a) Effects of pressure on the methanol conversion and hydrogen yield of the reactor. (b) Effects of pressure on the hydrogen productivity and output power of the reactor.

The rate profiles of the autothermal reforming reaction along the length of the reactor are presented in Figure 6 under different pressure conditions. As the pressure is increased, the reaction rate is also increased due to increased reactant concentrations. Higher pressures cause the reaction zone to be shifted significantly downstream. The reaction zone shifts about past half the length of the reactor, if the reactor operates at a pressure of six atmospheres. The reaction front may eventually exit the reactor, if the pressure is increased further. Higher pressures offer the potential to improve the rate of the reaction. However, the reactant molecules cannot be transferred to the surface of the catalyst fast enough to keep up with the rapid rate of the reaction, whereupon the reactor operates in a mass-transfer controlled regime, as discussed above. In such a situation, a lowly active catalyst is indistinguishable from a highly active catalyst, as the reaction rate is not determined by the intrinsic activity of the catalyst [35, 36]. High conversion and yields cannot be achieved under the same conditions, if the intrinsic activity is increased above that required for mass transfer control. In such a situation, the reactor may be adjusted in length to meet the needs of the conversion. Two different theoretical approaches to chemical kinetics have led to an understanding of the details of how elementary chemical reactions occur. Both of these are based on the idea of potential-energy surfaces, which are models showing how the potential energy of a reaction system varies with certain critical interatomic distances. The course of an elementary reaction is represented by the movement of the system over the potential-energy surface. One theoretical approach to the problem involves studying the region of the potential-energy surface that corresponds to the highest point on the energy barrier that separates the reactants from the products. This approach is relatively simple and leads to explicit general expressions for the reaction rate. The second approach involves considering the dynamics of the motion of the system over the potential-energy surface. The second theoretical approach to chemical kinetics is referred to as molecular dynamics, or reaction dynamics. It is a more detailed treatment of reactions and is designed to investigate the atomic motions that occur during a chemical reaction and the quantum states of the reactant and product molecules. Such studies are important in testing the validity of transition-state theory and similar treatments.
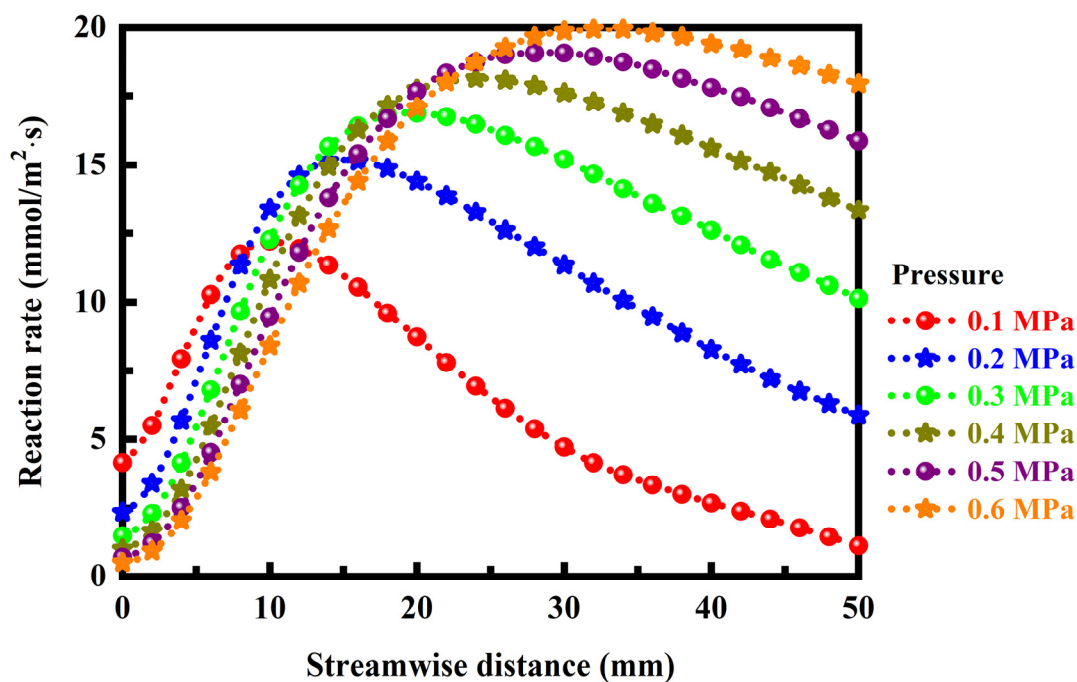

Figure 6. Rate profiles of the autothermal reforming reaction along the length of the reactor under different pressure conditions.

The pressure plays a competing role in the reactor efficiency and performance. Higher pressures tend to decrease the conversion and yield, but at the same time higher pressures may be advantageous,

as they tend to significantly improve the hydrogen productivity and output power of the reactor by accelerating the rate of the reaction. Therefore, there may exist an optimum pressure in terms of both efficiency and performance. It is impossible to achieve the object of maximum efficiency, while avoiding the problem of low performance.

3.2. Effect of flow rate

The chemical reaction proceeds at a level that may depend heavily upon the flow rate of the gaseous reactants in the reactor [37, 38]. The reactor within which the autothermal reforming process is carried out for productive purposes is devised so that continuous operation is possible under variable flow rate conditions. The effect of inlet velocity on the chemical process in the reactor is investigated in order to achieve variable hydrogen production capacity.

The methanol and hydrogen mole fraction profiles along the channel centreline are presented in Figure 7 under different inlet velocity conditions. The flow is laminar in nature and the distance required for reactant molecules to diffuse through the bulk fluid phase and contact the surface of the catalyst is essentially unchanged. The residence time of the gas stream in the reactor is extremely short. The reactor operates at millisecond contact times and therefore the inlet velocity of the entering gas stream may have a strong effect on the reactor efficiency and performance. At the lowest inlet velocity, methanol can be converted efficiently to hydrogen. More specifically, the chemical reaction is almost completed, and the maximum conversion of methanol to hydrogen is attained. As the inlet velocity is increased, the methanol mole fraction decreases slightly and the hydrogen mole fraction does not increase rapidly along the flowthrough path. At the outlet of the reactor, the methanol mole fraction increases steadily and the hydrogen mole fraction decreases greatly with increasing the inlet velocity. Operation of the reactor at higher inlet velocities may be disadvantageous, at which there occurs significant fuel breakthrough or slip due to reduced residence time. In such a situation, the chemical reaction is controlled by the velocity of the reactants passing through the reactor. At higher flow rates, there occurs fuel breakthrough or slip, which leads disadvantageously to incomplete conversion and lower hydrogen yields. Lower flow rates allow for the reactor to reduce fuel slip. Lower flow rates permit the conversion of methanol to hydrogen in an efficient manner. The rate of diffusion to the surface of the catalyst is determined by the concentration gradient and boundary layer thickness [39, 40]. Hence, the flow rate can be adjusted so that the rate of delivery of the reactants to the surface of the catalyst is kinetically controlled or mass-transfer limited to a desired level.
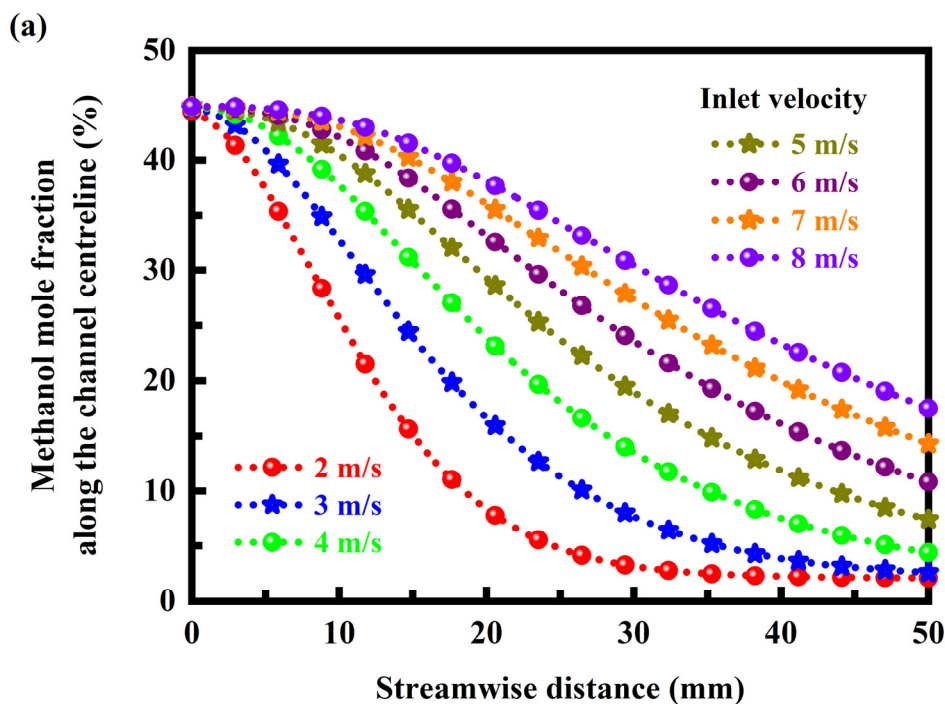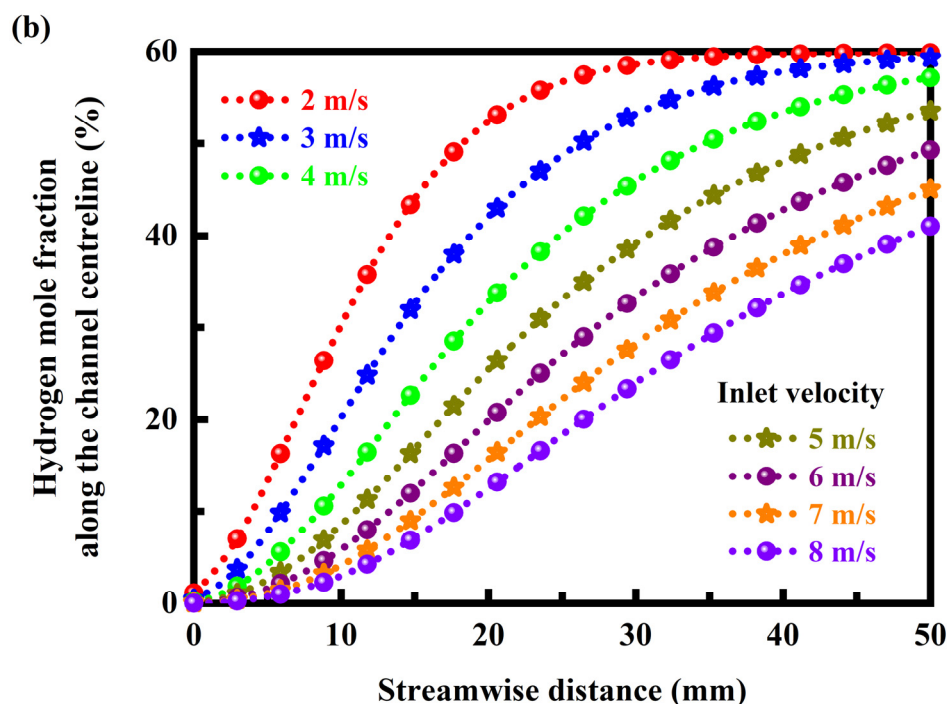
Figure 7. (a) Effect of inlet velocity on the methanol mole fraction along the channel centreline. (b) Effect of inlet velocity on the hydrogen mole fraction along the channel centreline.

The study of the detailed processes of reaction mechanisms is important for many reasons, including the help it gives in understanding and controlling chemical reactions [41, 42]. Many reactions of great commercial importance can proceed by more than one reaction path [43, 44]. Knowledge of the reaction mechanisms involved may make it possible to choose reaction conditions favouring one path over another, thereby giving maximum amounts of desired products and minimum amounts of undesired products [45, 46]. Furthermore, on the basis of reaction mechanisms, it is sometimes possible to find correlations between systems not otherwise obviously related [47, 48]. The effects of inlet velocity on the methanol conversion, hydrogen yield, and hydrogen productivity of the reactor are illustrated in Figure 8 wherein the results obtained for output power are also presented. The flow rate sets strict limits on operation. Specifically, the reactor operation is substantially affected by the flow rate of the feed mixture or the ratio of residence time to transverse mass transfer time. Incomplete conversion conditions can be clearly identifiable by calculation over a wide range of flow rates. At very lower flow rates, the conversion is almost complete and the yield is high. As the inlet velocity is increased, the conversion and yield drop rapidly, but the hydrogen productivity and output power of the reactor increases significantly in an almost linear fashion, which is essential to the success of start-up and acceleration of the downstream equipment. The flow rate of the gases entering the reaction chamber is desirably above the flow rate at which the conversion is almost complete. While high flow rates will lead disadvantageously to a hydrogen yield loss, the flow rate of the gaseous reactants in the reactor must be sufficiently high to maximize the hydrogen productivity and output power. The maximum hydrogen productivity and output power are obtained in the regime where the conversion is incomplete or methanol is converted inefficiently to hydrogen. At higher flow rates, there is a trade-off between high hydrogen productivity and low methanol conversion. The calculated output power of the reactor is of the order of thousands of kilowatts or megawatts per cubic meter. As the inlet velocity is increased further, however, the reactor operation may be limited by insufficient residence times, which will inevitably lead to lower hydrogen productivity. For certain processes that are particularly adaptable to continuous operation, reactors are devised so that the chemical reaction may continue while starting materials are introduced continuously or intermittently. For a reaction to occur, it is not sufficient that it be energetically product-favoured. The reaction must also occur at an observable rate. Several factors

influence reaction rates, including the concentrations of reactants, the temperature, and the presence of catalysts. The concentration affects the rate at which reacting molecules collide, a prerequisite for any reaction. Temperature is influential because reactions occur only if collisions between reactant molecules are sufficiently energetic. The proportion of molecules with sufficient energy to react is related to the temperature. Catalysts affect rates by providing a lower energy pathway by which a reaction can occur.
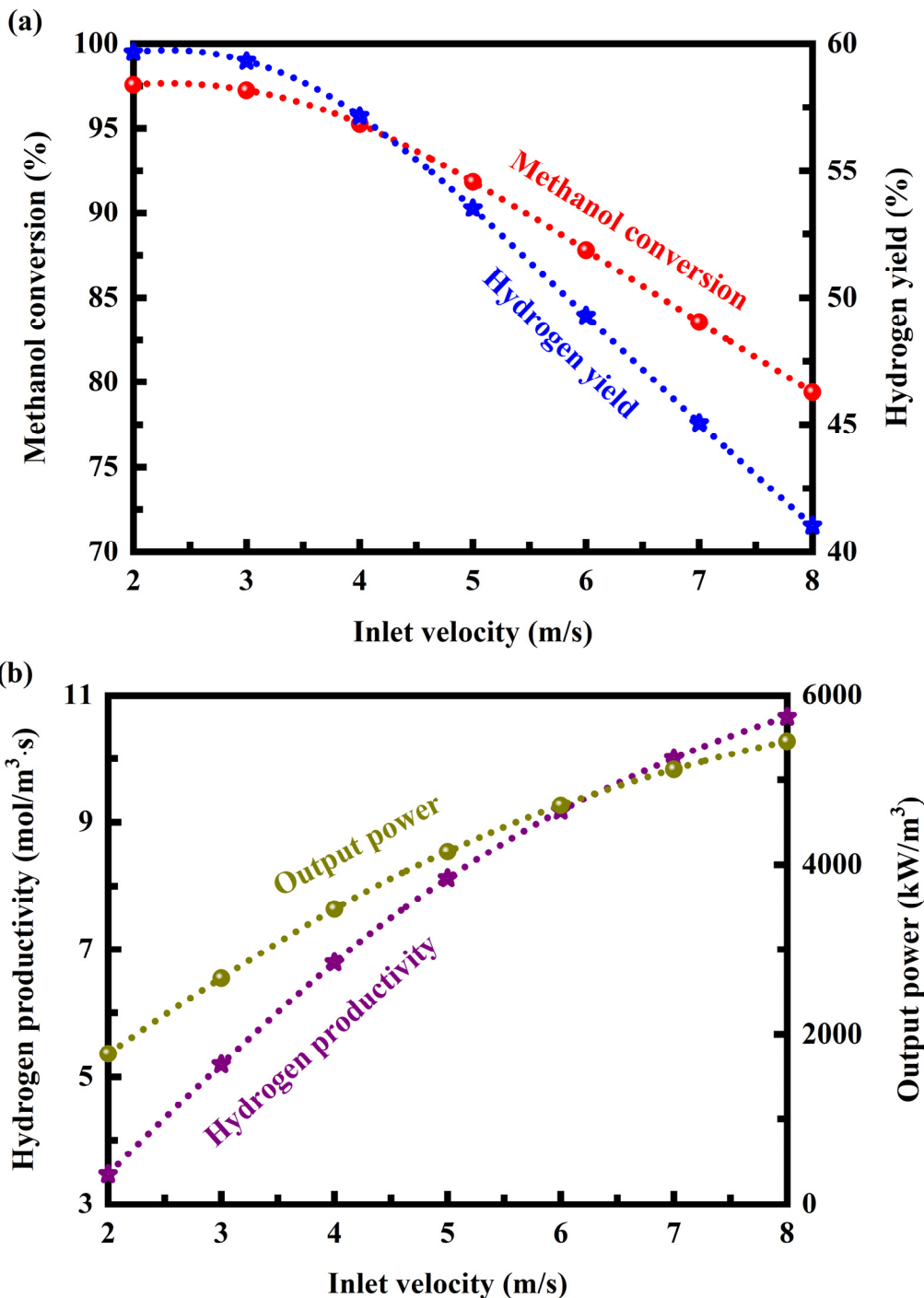

Figure 8. (a) Effects of inlet velocity on the methanol conversion and hydrogen yield of the reactor. (b) Effects of inlet velocity on the hydrogen productivity and output power of the reactor.

The flow rate plays a competing role in the reactor efficiency and performance, as similar as the pressure. Lower flow rates will lead to improved conversion and yields in the reactor. On the other hand, higher flow rates may be advantageous, as they tend to significantly improve the hydrogen productivity, which can increase the output power of the reactor. Therefore, there may exist an optimum flow rate in terms of both efficiency and performance.

4. Conclusions

   Computational modelling for microchannel reactor design was conducted in the attempt to fully understand autothermal reforming phenomena in millisecond reactors. The effects of pressure, and flow rate were investigated by means of various efficiency and performance criteria. The main conclusions are summarized as follows:

- The reactor operation is substantially affected by the flow rate and pressure. At higher pressures or flow rates, there is a trade-off between high hydrogen productivity and low methanol conversion.
- Pressure significantly affects the species uniformity in the reactor. The methanol conversion and hydrogen yield are governed largely by pressure. Operation at elevated pressures may be disadvantageous, at which there occurs significant fuel breakthrough or slip.
- The pressure and flow rate play competing roles in the reactor efficiency and performance. Higher pressures and flow rates tend to decrease the conversion and yield, but at the same time higher pressures and flow rates may be advantageous, as they tend to significantly improve the hydrogen productivity and output power of the reactor, which is essential to the success of start-up and acceleration of the downstream equipment. Therefore, there may exist an optimum pressure and flow rate in terms of both efficiency and performance.
- The calculated output power of the reactor is of the order of thousands of kilowatts per cubic meter.

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