

Production of C₂-olefins and Hydrogen from Methane Pyrolysis with Concentrated Solar Energy

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Abstract. A novel concept of CH₄ decomposition into H₂ and carbon materials via a two-step thermochemical and thermo-catalytic conversion process with concentrated solar power was proposed. Solar experiments were conducted for investigating the first high-temperature step of the proposed two-step thermochemical conversion process. This step consists of CH₄ decomposition into C₂ hydrocarbons and hydrogen at high temperatures. A significant effect of the addition of H₂ into CH₄ and a beneficial effect of the use of tungsten strips as catalyst on the increase in the yields of C₂H₂ and C₂H₄ were observed.

INTRODUCTION

For realizing an ideal sustainable energy society envisioned such as in the Paris Agreement on Climate Change, implementations of various possible renewable technologies for reducing carbon dioxide (CO₂) emissions are necessary. One candidate of such technologies is the direct solar conversion of methane (CH₄) to hydrogen fuel (H₂) and solid carbon materials (C) such as carbon nanotubes with concentrated solar energy and thus without co-producing carbon dioxide [1, 2]. However, some technological problems are still remained to be resolved for the practical use of the direct solar conversion; such as rapid and significant pyrolytic carbon deposition on the reactor walls and soot on windows at high temperatures over 1,300 °C.

Aiming to tackle such problem, a new concept for the co-production of hydrogen and valuable carbon materials from methane with concentrated solar power is proposed in this work. In the new concept, a novel two-step conversion process for decomposing CH₄ into H₂ and C is introduced. A series of solar tests was also conducted for investigating the primary process of the proposed conversion process.

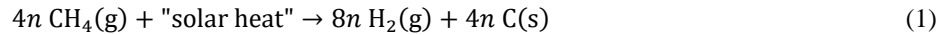
CO-PRODUCTION OF HYDROGEN AND CARBON MATERIALS BY DECOMPOSING METHANE

Several series of solar experiments to decompose CH₄ into H₂ and C have been carried out in the collaboration between PROMES-CNRS and IHI Corporation in the last decade. One is the test with a drop tube type solar reactor in which carbon black particles are continuously entrained into the flow of CH₄ as catalysts. The entrainment of particles in a gas flow improves the rate of heat transfer from the internal surface of the heated tube to the flow of CH₄ via the entrained particles absorbing the heat from the tube surface by radiation and releasing the heat to CH₄ around the particles [3]. The other is the test with an indirectly-heated packed-bed type solar reactor in which some

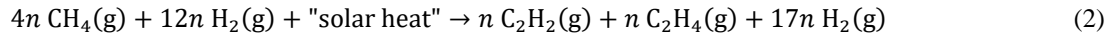
amount of carbon black particles are filled and fixed in the reactor tube as catalysts [4]. The catalytic activity of different carbon catalysts (carbon black, activated charcoal) was also studied between 900 and 1,200°C in an in-house solar thermogravimetric device [5]. Finally, other than the above, the test with an empty tube type reactor without using any catalysts was also carried out at temperatures up to about 1,550°C in the collaboration [6]. In the results of the above series of tests for producing H₂ and C by heating and decomposing CH₄ with concentrated solar power, very small amounts of acetylene (C₂H₂), ethylene (C₂H₄), and ethane (C₂H₆) were produced in addition to H₂ and C as products of the decomposition of CH₄. Such fact became an opportunity to devise and propose a new two-step approach for the co-production of H₂ and C materials from CH₄ with concentrated solar power.

New Approach for the Co-production of Hydrogen and Carbon Materials from Methane

A new methodology for co-producing H₂ and C materials from CH₄ [7,8] is proposed in this study. The main concept of this approach is to divide the conventional process of CH₄ decomposition,



into the following two elementary thermochemical conversion processes;



and



The former (the first step, Eq. (2)) is devoted to the synthesis of C₂-olefins such as C₂H₂ and C₂H₄ (and very small amount of C₂H₆) from CH₄ mixed with H₂ at high temperatures from 1,200 to 1,500 °C; the latter (the second step, Eq. (3)) corresponds to the production of H₂ and C from C₂-olefins at low temperatures from 500 to 1,000 °C.

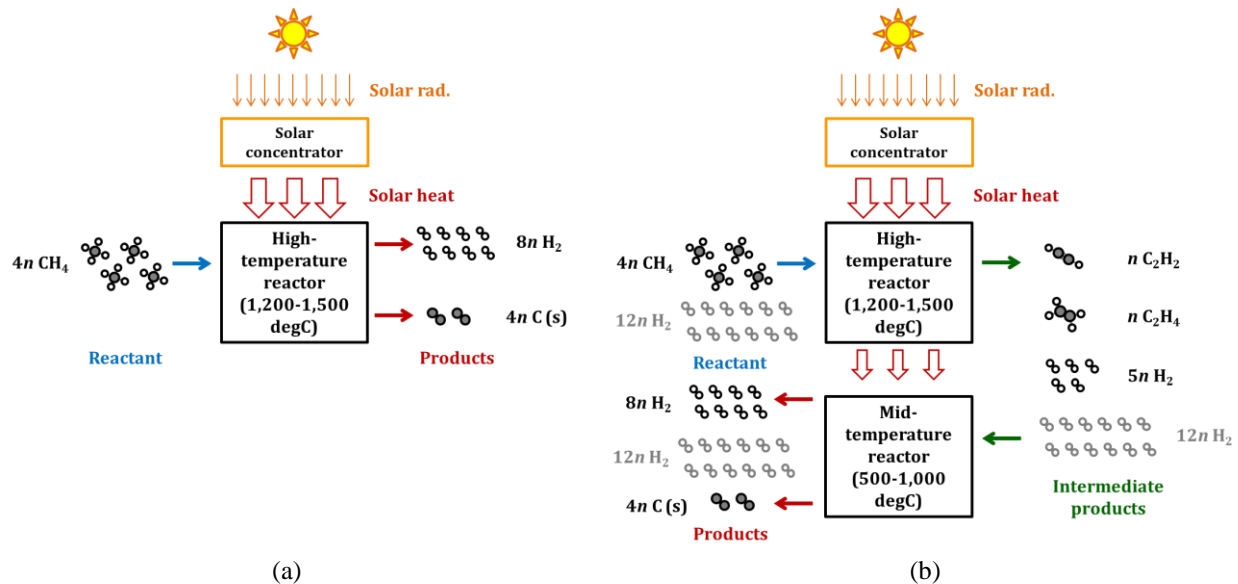


FIGURE 1. Illustrations of the conventional and the newly-proposed thermochemical processes for decomposing CH₄ into H₂ and C materials: (a) The conventional process by which CH₄ is directly decomposed into H₂ and C materials through only one-step thermochemical process at high-temperature conditions; (b) The proposed process by which CH₄ is decomposed into H₂ and C materials through two-step thermochemical processes synthesizing a few types of C₂-olefins as intermediate products.

In the first step of the above two-step conversion process, the main part of C in CH_4 should contribute to produce C_2 -olefins; the other part should contribute to produce solid carbons. The mole fractions of CH_4 and H_2 on the left-hand side of Eq. (2) should be adjusted for optimizing the yields of C_2 -olefins; the H_2/CH_4 ratio of 3 ($= 12n/4n$) is assumed in the above in one instance. A rapid cooling process (quenching operation) is required ideally at the outlet of the reactor for increasing the yields of C_2 -olefins. Rare metals such as molybdenum (Mo) and tungsten (W) may be used as catalysts for improving the yields of C_2 -olefins [9].

In the second step, a part ($12n$) of the total amount of H_2 ($20n$) on the right-hand side of Eq. (3) has to be recycled for the left-hand side of Eq. (2) for the first step. Thus, the net production of H_2 in the above instance with the H_2/CH_4 ratio of 3 is $8n$. Metal catalysts such as iron (Fe) and nickel (Ni) should be used as catalysts for decomposing C_2 -olefins and remained CH_4 into H_2 and C materials. The yields of H_2 and valuable C materials increase significantly when one feeds C_2 -olefins instead of CH_4 in the second step.

Advantages Obtained by Introducing the New Approach

One of the advantages obtained by introducing the above two-step thermochemical process into the decomposition of CH_4 into H_2 and C materials is that the amount of solid carbons (such as carbon black and graphite) produced in the reactor tube at high temperatures decreases because some amount of C from CH_4 forms the part of C_2 -olefins as gases instead of solid materials. Such characteristic can contribute to the inhibition of the obstruction of reactor tube due to the production of solid carbons in the tube at high temperatures.

Another advantage is a diversification of the types of carbon materials produced while decomposing CH_4 into H_2 and C. The proposed thermochemical process produces not only carbon black in the first step at a high-temperature condition but also carbon nanotubes in the second step at a low-temperature condition.

PRELIMINARY INVESTIGATION OF THE CO-PRODUCTION OF C_2 -OLEFINS AND HYDROGEN FROM METHANE

At the first stage of this project, the first step (Eq. (2)) of the two-step conversion process for synthesizing C_2 -olefins from CH_4 at high temperatures was focused and investigated. The test facilities and setup that were used and the considered test method and conditions for the investigation are described below.

Solar Test Facilities and Setups Used for the Investigation



FIGURE 2. The 1.5kW horizontal axis solar furnace at PROMES-CNRS in Odeillo and a tubular-type packed-bed solar reactor co-developed in the collaboration between PROMES-CNRS and IHI Corporation used for investigating the co-production of C_2 -olefins and H_2 from CH_4 .

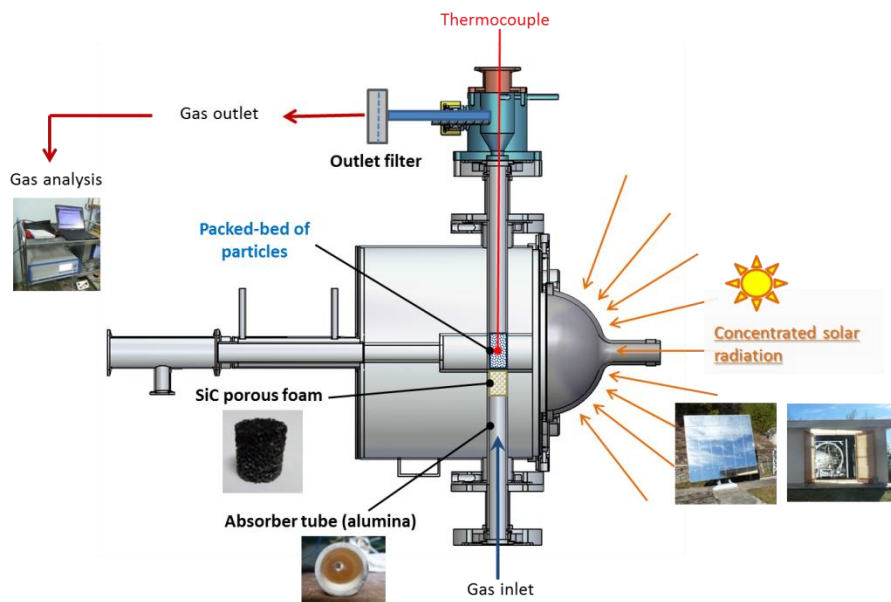


FIGURE 3. A schematic of the tubular solar reactor used in the investigation of the co-production of C_2 -olefins and H_2 from CH_4 .

The investigation was conducted by using a tubular packed-bed solar reactor developed in previous collaboration between PROMES-CNRS and IHI Corporation [4,5,6,10]. The reactor was installed at the focus of a 2 m-diameter parabolic dish concentrator at PROMES-CNRS in Odeillo, France (Fig. 2).

The solar reactor mainly consists of two distinct parts: i.e. the cavity-type solar receiver and the tubular chemical reaction zone separated from the surrounding atmosphere and to each other with appropriate sealing parts, as shown in Fig. 3. The reactor tube is inserted vertically into the cavity solar receiver (45 mm height, 40 mm width, and 55 mm depth) that is surrounded by outer graphite walls, carbon felt, and ceramic fiber insulators and closed at the front by the dome-shaped glass window. The insulation materials are used to minimize the heat losses from the cavity receiver. As a result, the cylindrical reactor shell made of stainless shell (254 mm outer diameter, 218 mm length) is maintained at low temperature without the need to implement an active water-cooling system. The only water-cooled components containing sealing rings are the front face (made of aluminum alloy) used to maintain the glass window and the two flanges used to fix the vertical alumina tube at the two extremities. A 10 mm-thick insulating carbon felt buffer is wrapped over the graphite cavity walls to avoid the direct contact between ceramic wool and hot cavity walls. A 15 mm-diameter aperture at the front graphite wall of the cavity is used to absorb the concentrated solar irradiation through the hemispherical glass window. An insulating layer of carbon felt with a corresponding hole is placed over this front wall to reduce thermal losses. Then, the vertical alumina tube (25 mm outer diameter, 20 mm inner diameter) placed inside the cavity is heated mainly by radiation from the surroundings cavity walls and serves as a heat transfer wall to transmit the solar power from the receiver to the reactor tube and the gases and catalysts inside the tube. A pyrometer placed at the front of the reactor (Heitronics KT15, 4.9-5.5 μm) serves as optical device for direct measurement of the outer surface temperature of the reactor tube (using CaF_2 optical window). B-type thermocouple shielded with alumina is also used to measure the temperature of the catalyst bed (TC1).

Procedures for the Investigation with the Solar Reactor

Argon (Ar) as the inert carrier gas, CH_4 , and H_2 are supplied to the reactor tube through the gas inlet in the upward direction and the mixture of Ar, CH_4 and H_2 is heated in the reactor tube as indirect solar heating. The heat energy absorbed into the gas mixture mainly contributes to the increase in the temperature of the gas mixture and the chemical decomposition of CH_4 . The flow rates of Ar, CH_4 and H_2 are supplied and controlled by mass-flow

controller (MFC, Brooks Instruments model 5850S), individually. The CH_4 mole fraction was controlled by adjusting the flow rate of Ar as the inert dilute gas and the H_2/CH_4 molar ratio was controlled while generally maintaining a total volumetric flow rate of gas at 0.5 NL/min. Pure Ar was used during the heating of the solar reactor in the solar test and the cooling of the reactor after the test. The cavity located at the center of the solar reactor was constantly swept by the flow of nitrogen (N_2) at the rate of 0.5 NL/min to provide inert atmosphere around the cavity made of graphite which is rapidly oxidized if oxygen (O_2) exists in its surroundings. The cavity and the reactor tube were preliminary purged to remove residual air by the means of successive vacuum pumping and inert gases were filled before starting the heating with concentrated solar power. The solar experiments were then performed at slightly over atmospheric pressure (88 kPa whereas the atmospheric pressure of the surrounding air is at 85 kPa at the site at over 1,500 meters altitude above sea level) because of the pressure drop caused by the particle bed and the other components such as the filter and the lines for gas analysis.

In solar tests for this packed-bed type reactor, the mixture of Ar, CH_4 , and H_2 to be heated in the reactor tube was injected from the bottom side of the tube. The reactive CH_4 was injected once the steady state at a targeted temperature (i.e. isothermal) condition was achieved. Although most part of the carbon formed by the decomposition of CH_4 was deposited on the catalyst surface and was thus trapped in the particle bed, a carbon filter was implemented at the outlet for gas cleaning in order to trap the remaining fine particulates and to protect the analytical systems. The gaseous products were finally quantified by online gas analysis using a gas chromatograph (Varian CP4900 micro-GC) equipped with two columns using Ar carrier gas and thermal conductivity detectors (MolSieve 5A PLOT for H_2 and CH_4 , and PoraPLOT U for H_2 , CH_4 , and light C_2 -olefins C_2H_2 , C_2H_4 , C_2H_6), with gas sampling every about 90 s. In addition, the outlet gas was analyzed online by a specific analyzer (NGA 2000 MLT3) for continuous H_2 and CH_4 analysis (by thermal conductivity and nondispersive IR detection, respectively).

The whole experimental set-up was settled at the focus of a solar concentrating system consisting of a horizontal-axis medium size solar furnace (comprising a sun-tracking heliostat and a 2 m-diameter parabolic dish solar concentrator). The focal point of the concentrator (0.85 m ahead of the parabolic mirror) was located at the cavity aperture for maximum solar radiation absorption inside the cavity receiver. The absorbed solar power that determines the temperature can be adjusted by the means of an intermediate shutter placed between the heliostat and the solar concentrator.

Operating Conditions for the Solar Tests

Three types of bed materials put in the reactor tube were considered in this series of the solar test as illustrated in Fig. 4: (α) non-catalytic silicon carbide (SiC) particles, (β) either Mo or W powder/particles mixed with non-catalytic Al_2O_3 particles, (γ) either Mo or W strips.

The mean diameter of SiC particles is 1.4 mm. The diameter of Mo powder is less than 150 μm ; that of W powder is 12 μm ; that of W particles is 500 - 2,000 μm . The diameter of Al_2O_3 particles is 1.14 mm. The widths, lengths, and thickness of the Mo and W strips are 20 mm, 40 mm, and 0.2 mm, respectively. Figure 5 shows the appearance of these particles and the strips.

In the case of the former two packed particle bed reactor, two cylindrical porous ceramic foams (reticulated porous ceramic RPC made of β - SiC , 20 mm diameter, 20 mm height, 30 ppi) were used to support the particles at the center of the heating section of the reactor tube. On the other hand, in the case of the use of Mo/W strips as the catalyst material, one ceramic foam was enough to support the strips around the center of heating section of the reactor tube. An inner supporting tube was placed below the foams inside the reactor tube for the purpose of supporting the foams from the bottom side.

The mass of bed material, the H_2/CH_4 ratios, the total volumetric flow rates of the gas mixture of Ar, CH_4 , and H_2 , and the temperatures of the bed materials attained in the test for each case of the above (α), (β), and (γ) are tabulated in Table 1. The H_2/CH_4 ratio at the inlet of the reactor was varied as a main parameter at each temperature conditions. In most cases, the total volumetric flow rate was kept constant at 0.5 or 1.0 NL/min.

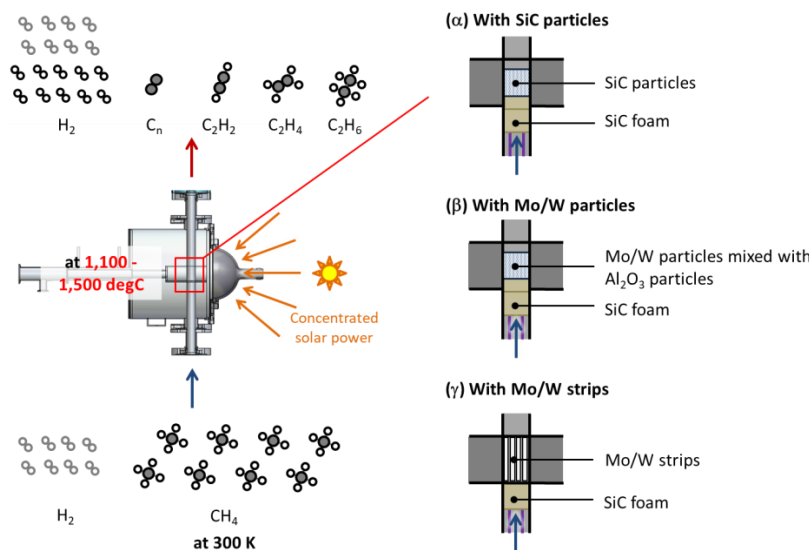


FIGURE 4. Schematic of the solar experiments for investigating the co-production of C_2 -olefins and H_2 by decomposing CH_4 at high temperatures using 1.5 kW solar furnace at PROMES-CNRS in Odeillo: (α) in the case that SiC particles are used as bed materials; (β) in the case that either Mo or W particles mixed with Al_2O_3 particles are used as bed materials; (γ) in the case that Mo or W strips are used as bed materials.

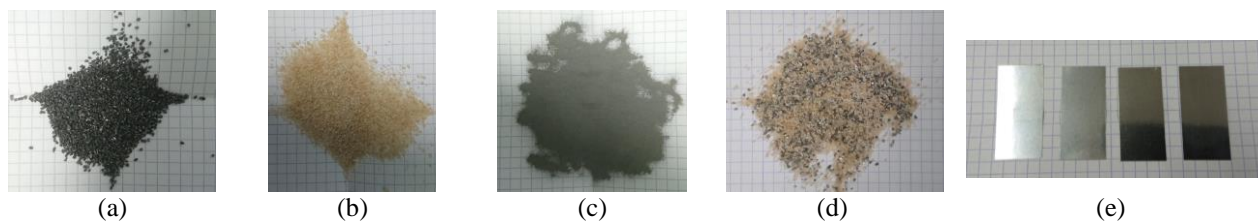


FIGURE 5. Pictures of the bed materials used in the solar experiments for investigating the co-production of C_2 -olefins and H_2 by decomposing CH_4 at high temperatures: (a) SiC particles (1.4 mm diameter); (b) Al_2O_3 particles (1.14 mm diameter); (c) W particles (12 μm diameter); (d) W particles (12 μm diameter) mixed with the Al_2O_3 particles; (e) W strips; the width of the grid of the paper in the picture is 5 mm.)

TABLE 1. List of operating conditions of the solar tests for investigating the co-production of C_2 -olefins and H_2 by decomposing CH_4 at high temperatures using 1.5 kW solar furnace at PROMES-CNRS in Odeillo.

	Type of bed material	Size of bed materials	Mass of bed materials, g	H_2/CH_4 ratio, -	Total vol. flow rate, L/min	Bed temperature, °C
(α)	SiC particles	$\phi = 1.4$ mm	9.71	0.0, 2.0, 4.0	0.5 - 0.9	1,100 - 1,500
(β)-1	Mo powder / Al_2O_3 particles	$\phi < 150$ μm / $\phi = 1.14$ mm	7.92 / 13.13	0.0, 1.0, 2.0	0.5	1,300
(β)-2	W powder / Al_2O_3 particles	$\phi < 12$ μm / $\phi = 1.14$ mm	20.5 / 13.13	0.0, 1.0, 2.0	0.5	1,100 - 1,300
(β)-3	W particles / Al_2O_3 particles	$\phi = 500-2,000$ μm / $\phi = 1.14$ mm	29.7 / 13.13	0.0, 1.0, 2.0, 3.0, 6.0	0.5 - 1.0	1,300 - 1,400
(γ)-1	Empty tube	-	-	0.0, 1.0, 2.0, 3.0	0.5 - 1.0	1,100 - 1,400
(γ)-2	Mo strips	4 pcs. \times 40 mm \times 20 mm \times 0.2 mm	6.0 = 4 pcs. \times 1.5	0.0, 1.0, 2.0, 3.0, 6.0	0.5 - 1.0	1,400
(γ)-3	W strips	4 pcs. \times 40 mm \times 20 mm \times 0.2 mm	13.2 = 4 pcs. \times 3.3	0.0, 1.0, 2.0, 3.0, 6.0	0.5 - 1.0	1,300 - 1,400

Results of the Solar Tests

The main results of the solar experiments are described below. In the following discussion, the cases of (α), (β)-2, (γ)-1, and (γ)-3 in Table 1 are focused for highlighting the effect of the addition of H_2 into CH_4 and the use of rare metal particles and strips on the increase in the yields of C_2H_2 and C_2H_4 .

Case (α), solar test results with non-catalytic SiC particles

Figure 6 shows a series of the results obtained by the solar tests for the decomposition of CH_4 with non-catalytic SiC particles. The CH_4 conversion and H_2 yield increase as increasing the temperature in any case of the H_2/CH_4 ratio. However, they decrease as increasing the H_2/CH_4 ratio. The CH_4 conversion decreased from 0.83 to 0.52 and from 0.94 to 0.81 at 1,300 °C and 1,400 °C, respectively, when the ratio changed from 0 to 4. The H_2 yield decreased from 0.74 to 0.36 and from 0.86 to 0.63 at 1,300 °C and 1,400 °C, respectively, when the ratio changed as well.

On the other hand, the C_2H_2 yield (carbon-based) and the C_2H_4 yield (carbon-based) increase slightly as increasing the H_2/CH_4 ratio, excepting the condition at 1,200 °C in both cases. The C_2H_2 yield increased from 0.16 to 0.24 and from 0.15 to 0.25 at 1,300 °C and 1,400 °C, respectively, when the ratio changed from 0 to 4. The C_2H_4 yield increased from 0.05 to 0.10 and from 0.05 to 0.11 at 1,300 °C and 1,400 °C, respectively, when the ratio changed as well. The C_2H_6 yield keeps 0.00 at any conditions of the ratio and the temperature.

Case (β)-2, solar test results with W powder mixed with Al_2O_3 particles

Figure 7 shows a series of the results obtained by the solar tests for the decomposition of CH_4 with W particles mixed with Al_2O_3 particles. The CH_4 conversion and H_2 yield increase as increasing the temperature in any case of the H_2/CH_4 ratio. And, they decrease as increasing the H_2/CH_4 ratio, as well as in the case (α) with SiC particles, but very rapidly comparing with that case with SiC particles. The CH_4 conversion decreased from 0.89 to 0.70 at 1,300 °C when the H_2/CH_4 ratio changed from 0 to 2. The H_2 yield decreased from 0.86 to 0.63 at the same temperature when the ratio changed as well.

In this case, the C_2H_2 and C_2H_4 yields show no apparent influence of the change in H_2/CH_4 ratio. The C_2H_2 yield decreased from 0.03 to 0.00 when the ratio changed from 0 to 2, while the C_2H_4 yield increased from 0.02 to 0.06 when the ratio changed as well.

Case (γ)-1, solar test results with empty tube

Figure 8 shows a series of the results obtained by the solar tests for the decomposition of CH_4 with empty tube. The CH_4 conversion and the H_2 yield increase as increasing the temperature and they decrease as increasing the H_2/CH_4 ratio, as well as in the above cases. The CH_4 conversion decreased significantly from 0.95 to 0.69 at 1,400 °C when the H_2/CH_4 ratio changed from 0 to 3. The H_2 yield decreased more significantly from 0.86 to 0.39 at the same temperature when the ratio changed as well.

Both the C_2H_2 and C_2H_4 yields increase significantly as increasing H_2/CH_4 ratio in this case without any bed materials in the tube. The C_2H_2 yield increased from 0.14 to 0.22 at 1,400 °C when the ratio changed from 0 to 3. The C_2H_4 yield also increased from 0.05 to 0.23 at the same temperature when the ratio changed as well.

Case (γ)-3, solar test results with W strips

Figure 9 shows a series of the results obtained by the solar test for the decomposition of CH_4 with W strips. The CH_4 conversion and the H_2 yield increase as increasing the temperature and they decrease as increasing the H_2/CH_4 ratio, similarly to the above case (γ)-1 without any bed materials. The CH_4 conversion decreased significantly from 0.95 to 0.69 at 1,400 °C when the H_2/CH_4 ratio changed from 0 to 3; this trend is mostly same with the case (γ)-1. On the other hand, the H_2 yield decreased from 0.86 to 0.33 at 1,400 °C when the ratio changed as well, more significantly than the case (γ)-1.

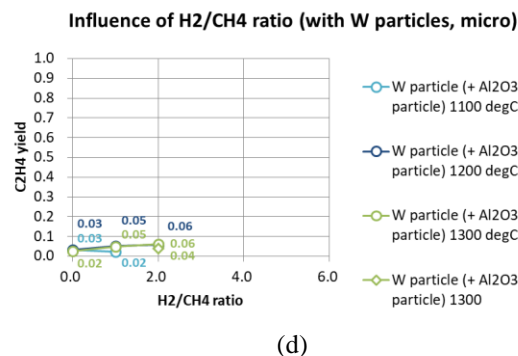
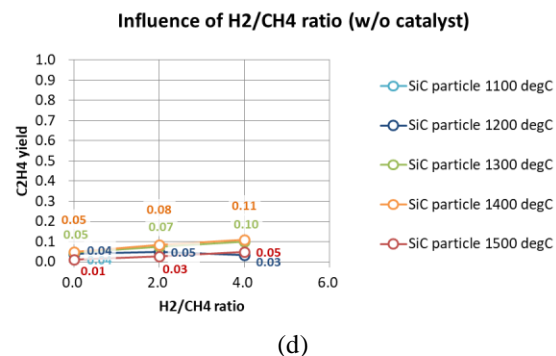
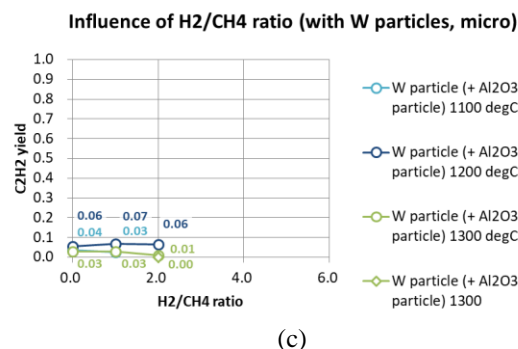
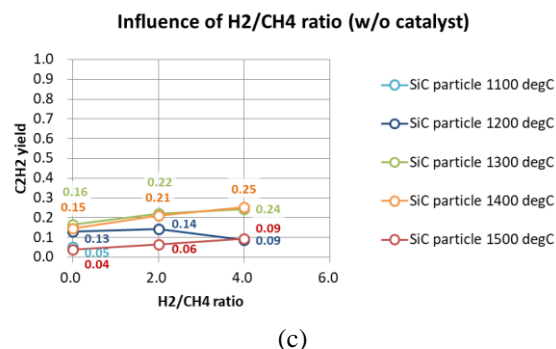
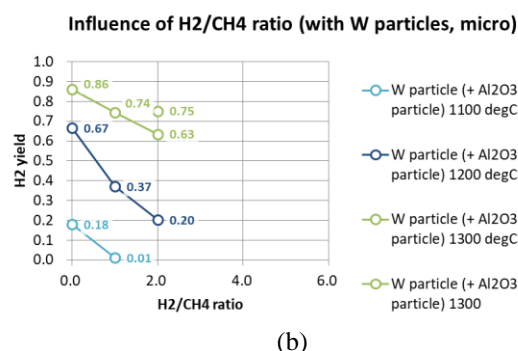
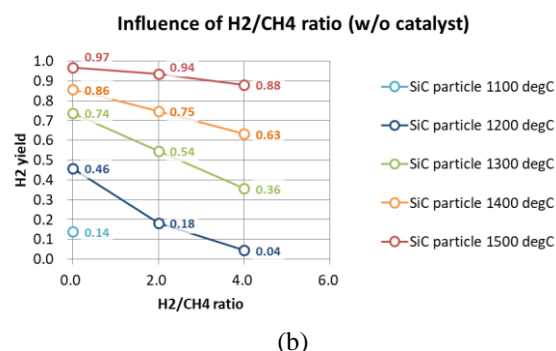
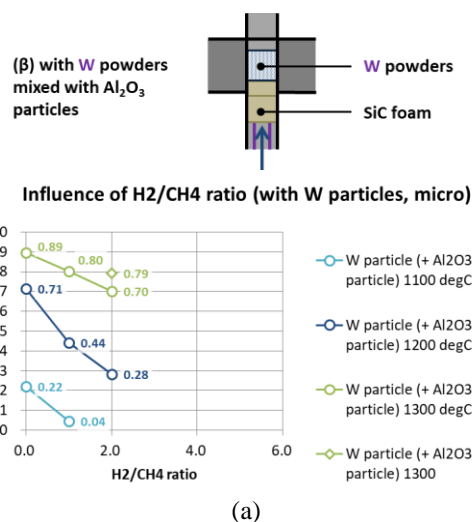
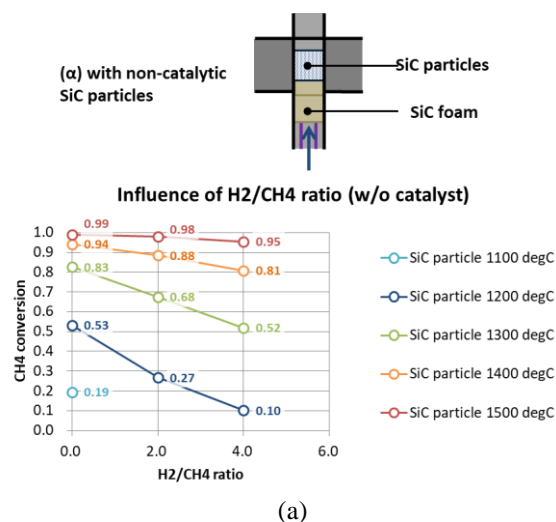


FIGURE 6. Results of the solar tests with non-catalytic SiC particles: (a) CH₄ conversion; (b) H₂ yield; (c) C₂H₂ yield; (d) C₂H₄ yield (carbon-based).

FIGURE 7. Results of the solar tests with W and Al₂O₃ particles: (a) CH₄ conversion; (b) H₂ yield; (c) C₂H₂ yield (carbon-based); (d) C₂H₄ yield (carbon-based).

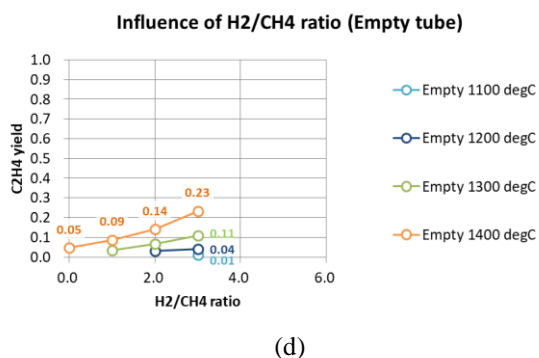
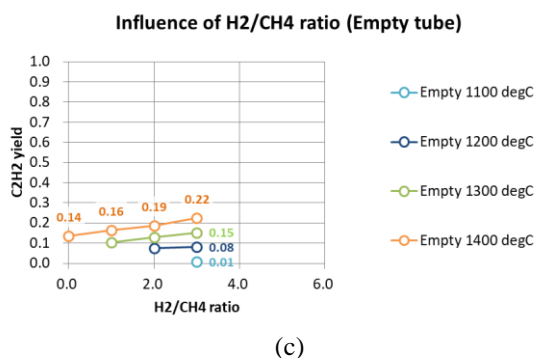
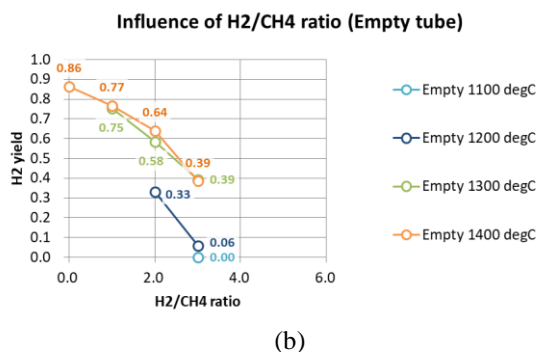
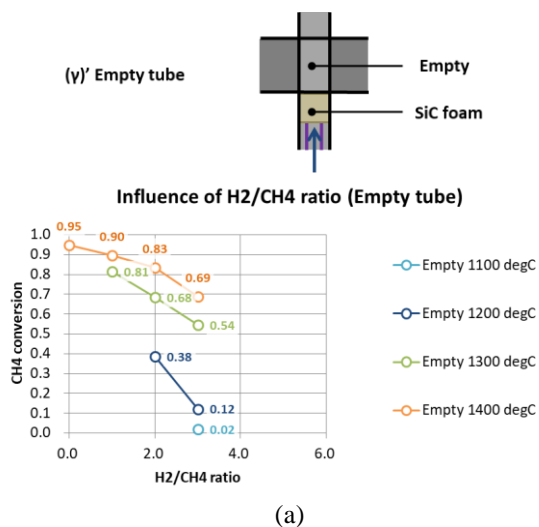


FIGURE 8. Results of the solar tests with empty reactor tube: (a) CH₄ conversion; (b) H₂ yield; (c) C₂H₂ yield (carbon-based); (d) C₂H₄ yield (carbon-based).

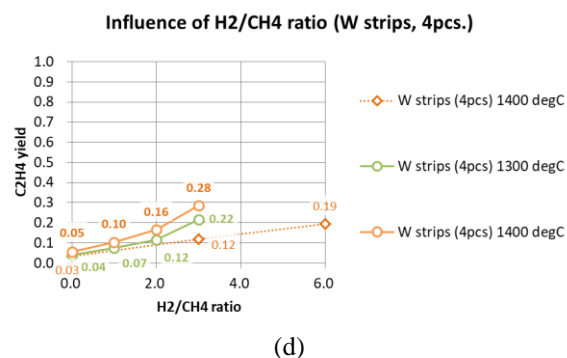
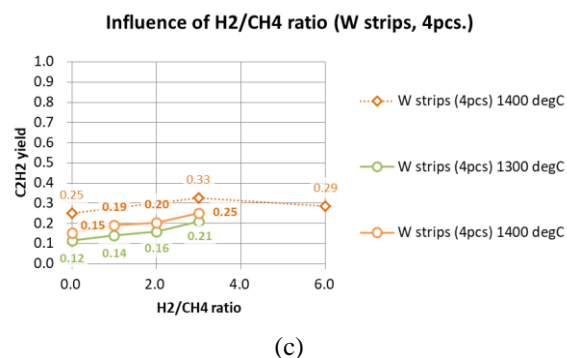
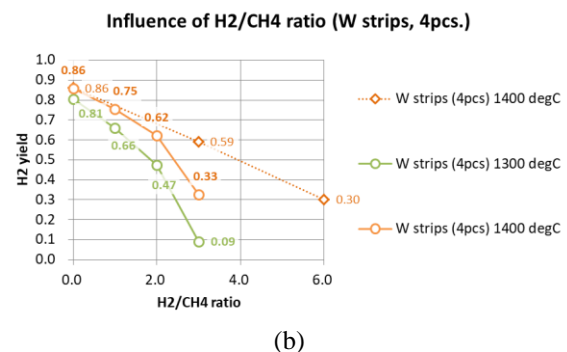
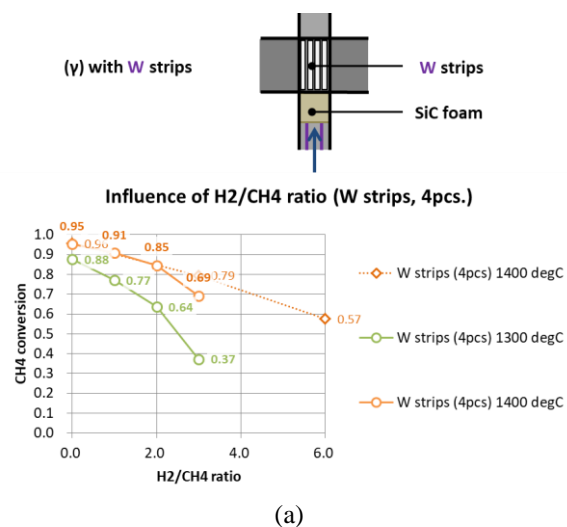


FIGURE 9. Results of the solar tests with W strips: (a) CH₄ conversion; (b) H₂ yield; (c) C₂H₂ yield (carbon-based); (d) C₂H₄ yield (carbon-based).

Both the C_2H_2 and C_2H_4 yields increase significantly as increasing H_2/CH_4 ratio in this case with W strips. The C_2H_2 yield increased from 0.15 to 0.25 at 1,400 °C when the ratio changed from 0 to 3, more significantly than the case (γ)-1. The C_2H_4 yield also increased from 0.05 to 0.28 at the same temperature when the ratio changed as well, more significantly than the case (γ)-1 as with the C_2H_2 yield.

CONCLUSION

- A novel concept for CH_4 decomposition into H_2 and carbon materials via a two-step thermochemical and thermo-catalytic conversion process with concentrated solar power was proposed. Its first step is devoted to the synthesis of C_2 -olefins (C_2H_2 and C_2H_4) from CH_4 mixed with H_2 and rare metal catalysts at high temperatures from 1,200 to 1,500 °C. Its second step is devoted to the production of H_2 and valuable C materials such as carbon nanotubes from the C_2 -olefins at low temperatures from 500 to 1,000 °C.
- Solar experiments were conducted for investigating the first high-temperature step of the proposed two-step thermochemical conversion process. The 1.5 kW horizontal axis solar furnace at PROMES-CNRS in Odeillo and the tubular packed-bed solar reactor co-developed in the collaboration between the PROMES-CNRS and IHI Corporation were used for the investigation.
- Non-catalytic SiC particles, Mo and W powder/particle mixed with Al_2O_3 particles, and Mo and W strips are used as bed materials in the reactor tube for investigating some beneficial effects on the increase in the yields of C_2 -olefins.
- A significant effect of the addition of H_2 into CH_4 on the increase in the yields of C_2 -olefins was observed. The conversion of CH_4 was 81 % and the yields of C_2H_2 and C_2H_4 (carbon-based) were 25 % and 11 %, respectively, when the H_2/CH_4 ratio was 4 and the reactor was heated at 1,400 °C with the non-catalytic SiC particles in the reactor tube. The conversion of CH_4 was 69 % but the yields of C_2H_2 and C_2H_4 (carbon-based) were 22 % and 23 %, respectively, when the H_2/CH_4 ratio was 3 and the reactor was heated at 1,400 °C with no bed materials in the reactor tube.
- No apparent effect (rather a negative effect in some cases) of the use of W powder as the catalyst on the increase in the yields of C_2 -olefins was observed.
- A slight beneficial effect of the use of W strips as the catalyst on the increase in the yields of C_2 -olefins was observed. The yields of C_2H_2 increased from 22 % to 25 % and that of C_2H_4 increased from 23 % to 28 % with the use of W strips.

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