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Solar hydrogen production from the thermal splitting of methane in a high temperature solar chemical reactor

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Abstract

This study addresses the single-step thermal decomposition (pyrolysis) of methane without catalysts. The process co-produces hydrogen-rich gas and high-grade carbon black (CB) from concentrated solar energy and methane. It is an unconventional route for potentially cost effective hydrogen production from solar energy without emitting carbon dioxide since solid carbon is sequestered.

A high temperature solar chemical reactor has been designed to study the thermal splitting of methane for hydrogen generation. It features a nozzle-type graphite receiver which absorbs the solar power and transfers the heat to the flow of reactant at a temperature that allows dissociation. Theoretical and experimental investigations have been performed to study the performances of the solar reactor. The experimental set-up and effect of operating conditions are described in this paper. In addition, simulation results are presented to interpret the experimental results and to improve the solar reactor concept. The temperature, geometry of the graphite nozzle, gas flow rates, and CH_4 mole fraction have a strong effect on the final chemical conversion of methane. Numerical simulations have shown that a simple tubular receiver is not enough efficient to heat the bulk gas in the central zone, thus limiting the chemical conversion. In that case, the reaction takes place only within a thin region located near the hot graphite wall. The maximum CH_4 conversion (98%) was obtained with an improved nozzle, which allows a more efficient gas heating due to its higher heat exchange area.
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Keywords: Hydrogen; Production; Methane; Cracking; Solar reactor; Carbon black

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1. Introduction

Fossil fuels will continue to play a major role in global energy supply in the near to medium term future. But increasing consumption of fuel gases (methane and hydrogen) will displace oil, coal, and nuclear as the world's principal source of energy. Sustainable energy carriers based on hydrogen produced by renewable energy resources are considered as an ultimate long term option.

Currently, hydrogen is produced mainly through the steam reforming of natural gas (NG), and carbon black is produced by partial combustion of hydrocarbons. In the short to medium term, NG will continue to be the major source of hydrogen

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Nomenclature

a	reaction order
$C_i(r, z)$	molar concentration of species i in the gas flow (mol/m^3)
C_{pi}	heat capacity of species i (J/mol K)
C_p	heat capacity of the gas mixture (J/mol K) $= \sum C_{pi} \cdot y_i$
d	diameter of the nozzle (0.01 m)
D_{CH_4}	molecular diffusion coefficient of CH_4 in argon (m^2/s)
E_a	activation energy (J/mol)
F_{CH_4}	inlet molar flow rate of CH_4 (mol/s)
F_{Ar}	molar flow rate of argon (mol/s)
h_p	heat transfer coefficient (W/m^2 K)
k	kinetic constant (s^{-1})
k_0	frequency factor (s^{-1})
L	nozzle length (0.061 m)
M_i	molecular weight of species i (kg/mol)
Q	total gas flow rate (m^3/s)
Q_{solar}	solar power input (W)
R	nozzle radius (0.005 m)
r	mean radius of carbon nano-particles (30 nm)
r_v	net rate of CH_4 dissociation reaction (mol m^{-3} s^{-1})
$T(r, z)$	temperature (K)
$T_g(z)$	mean temperature of the gas on a section of the nozzle (K)
T_w	wall temperature (K)
T_0	temperature of the inlet gas (K)
u_m	mean velocity of the inlet gas (m/s)
$u_z(r)$	axial gas velocity (m/s)
u	vector of gas velocity (m/s)
X	conversion of methane
Y	yield of hydrogen
y_i	mole fraction of species i
z	axial position (m)
ΔH_T	reaction enthalpy at T (J/mol)
ν_{CH_4}	stoichiometric coefficient of CH_4 (-1)
ρ_c	density of carbon particles (2000 kg/m^3)
ρ	density of the gas mixture (kg/m^3) $= \sum \rho_i \cdot y_i$
λ_i	thermal conductivity of species i (W/m K)
λ	thermal conductivity of the gas mixture (W/m K) $= [\sum y_i (M_i)^{0.5} \cdot \lambda_i] / [\sum y_i (M_i)^{0.5}]$

(Barreto et al., 2003), because it is the most cost-effective process. Nevertheless, the use of NG as a chemical feedstock and as a fuel in the current hydrogen production processes induces emissions of large quantities of carbon dioxide responsible for global warming.

For addressing climate change and meeting the target of Kyoto Protocol, the ultimate engineering solution to energy and environmental concerns will rely on a complete technology substitution process, which has to take place during a transition period during which CO_2 emission intensity should decrease (decarbonisation of energy production). By favoring the use of NG instead of solid or liquid fossil fuels and by proposing an alternate and clean solution to the conventional fossil fuel steam reforming process, the solar thermal cracking of NG lies on the decarbonisation roadmap and prepares the path to the hydrogen economy (Steinberg, 1999; Muradov, 1993; Fulcheri and Schwob, 1995).

Solar decarbonisation of natural gas is a mid-term route for producing hydrogen in a large scale from renewable energy without greenhouse gas emission, and it thus offers a viable transition path to solar hydrogen (solar energy storage and transportation). The process thermally decomposes methane in a high temperature chemical reactor heated by concentrated solar energy.

The main economical impact is the production of two valuable products, hydrogen (H_2) and a high-value nano-material, Carbon Black (CB). The main advantages are the followings: a clean renewable energy is used to produce the energy carrier H_2; solar energy is stored into a transportable fuel; H_2 is produced with zero CO_2 emission (carbon can be easily sequestered); marketable high-grade CB is synthesized; finally, fossil fuels are saved with respect to conventional methods for H_2 and CB production.

This solar route saves the process energy and the CO_2 emission associated with the production of both products by classical methods. For this solar-thermal process, fossil energy usage and emissions are actually avoided or greatly reduced. Recent work carried out in the USA compared the co-product solar thermal processing to conventional steam reforming and “furnace black” process (Dahl et al., 2004). According to these authors, the overall fossil fuel savings are 277 MJ per kg H_2 produced, and the

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