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Hydrogen production from thermo-catalytic decomposition of methane using carbon black catalysts in an indirectly-irradiated tubular packed-bed solar reactor

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ABSTRACT

The solar thermo-catalytic decomposition of methane using carbon black catalysts for CO₂-free hydrogen production is studied in a packed-bed reactor. The indirectly-irradiated reactor is based on a cavity receiver and a tube-type absorber in which a given load of particle catalyst is injected during on-sun operation, while enabling multiple refilling for catalyst replacement. Concentrated solar power is used as an external radiative source for supplying the high temperature process heat and for driving the endothermic reaction. The indirect irradiation via the intermediate opaque tubular absorber results in a more uniform heating of the whole reacting bed volume and thus an easier reaction temperature control and determination. Carbon particles are used for enhancing the rate of the heterogeneous decomposition reaction and the coupling of the reactor with a particle injection system is implemented to operate in semi-continuous mode with possibility of catalyst load renewal after deactivation.

The packed-bed reactor was operated on-sun to investigate the effect of the various operating parameters on the reactor thermochemical performances. Complete methane conversion to hydrogen was achieved (100% selectivity to H₂ with no side-products), but catalyst deactivation occurred progressively because of carbon deposition at the surface, with acetylene as the main evolving by-product. The temperature and the residence time of the flowing gas through the catalyst bed were identified as the main parameters influencing the chemical conversion and hydrogen yield. This type of scalable reactor configuration could be applied for continuous hydrogen production with in-situ catalyst regeneration or substitution without reactor operation break-down at high temperatures. Copyright © 2014, Hydrogen Energy Publications, LLC. Published by Elsevier Ltd. All rights reserved.

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Introduction

Hydrogen is one of the main alternative and sustainable fuel options for decarbonizing transport and power generation systems, with the potential to be truly zero emission, for example, when used in fuel cells. However, this energy carrier has to be produced using renewable energies without CO₂ emission to be really environmentally attractive. Consequently, alternative routes for clean and cost-effective hydrogen production have to be developed [1,2], in substitution for the conventional hydrogen production technologies such as natural gas steam reforming, partial oxidation or coal gasification, that require further CO₂ abatement technologies such as capture and sequestration to reduce their environmental impact. These processes, indeed, are characterized by their high-energy consumption and the concomitant emission of pollutants, derived mainly from the combustion of fossil fuels for heat generation. Hybrid solar-fossil thermochemical processes that make use of an external source of concentrated solar radiation for supplying process heat, offer viable and efficient routes for fossil fuel decarbonization and CO₂ avoidance, and further create a transition path towards solar hydrogen [3–8]. In this context, the solar-driven thermocatalytic decomposition of methane (TCDM) is an attractive option for CO₂-free hydrogen production as the reaction yields hydrogen and solid carbon that can be valorized as a marketable high-value co-product, or also be applied as reducing agent in metallurgical processes. The TCDM process ($\text{CH}_4 \rightarrow \text{C(s)} + 2\text{H}_2$, $\Delta H^\circ = 74.6 \text{ kJ/mol}$) lies on the decarbonization roadmap and prepares the path to the hydrogen economy, as it represents a mid-term transition from fossil fuel to renewable hydrogen energy systems [2]. The main advantages of the solar route include (i) the saving of fossil fuel as no portion of the feedstock is combusted for process heat, (ii) the chemical storage and up-grading of solar energy in the form of solar fuel, (iii) the suppression of greenhouse gas emissions (CO₂, SO₂, NO_x) with respect to conventional processes, and (iv) the absence of products contamination by the combustion gases since solar concentrated energy is used as the external source for supplying the process heat at high temperatures.

The thermal methane dissociation occurs at elevated temperatures (typically above 1300 °C), which suggests the use of catalysts in order to reduce the maximum process temperature and enhance the kinetics. Chiefly, supported metallic-based (transition metals of group VIII, such as Ni, Fe and Co, that lead to the production of carbon nanofibers or nanotubes) and carbonaceous catalysts have been used for TCDM [9–39], and the latter have emerged as attractive candidates because they offer several advantages: (i) higher thermal stability and feedstock flexibility and no sulphur poisoning; (ii) lower price; and (iii) the formed carbon can be used as catalyst precursor, so that the process may be self-sustained by autocatalytic reaction. Among the variety of carbon materials investigated, disordered forms of carbon (e.g., carbon blacks, activated carbons) were generally found more catalytically active than ordered ones (e.g., graphite and diamond powder) [22,23]. Activated carbons with high surface area showed acceptable initial reaction rates at the expense of

being rapidly deactivated, whereas carbon blacks (CBs) showed a lower initial reaction rate, but they became deactivated very slowly. Concerning the regeneration of deactivated carbon [25,39], treatment with activating oxidizing agents (steam, CO₂) at elevated temperature can be considered to remove the deposited carbon and to re-establish the catalytic activity, due to the increase in surface area.

Regarding solar reactor technologies, TCDM was experimentally demonstrated using directly-irradiated particle-flow solar reactors with reactants directly exposed to the concentrated solar irradiation (using CB particles as radiant absorbers) [40], and indirectly-irradiated solar reactors with intermediate tubular or other opaque graphite/ceramic surfaces serving as absorbers and heat conductors [41–45]. Concerning directly heated solar reactors, the direct radiation absorption by particle-laden flow provides efficient means of heat transfer to the reaction site, bypassing the limitations imposed by indirect heat transfer through reactor walls. However, the window for the access of concentrated solar radiation is a critical component because of possible particle deposition, which generally requires the use of inert gas for window protection and imposes associated constraints: (i) the gas stream is mixed with the reactive gas, which dilutes the reaction products and further induces energy penalty, (ii) the residence time of the reactants is reduced, which does not favor the reaction kinetics. These issues justify the need to develop indirectly-irradiated solar reactors applied to thermochemical fuel production for decoupling the solar receiver zone from the reaction zone.

Based on these considerations, the pilot-scale demonstration of methane splitting process in multi-tubular solar reactors (20–50 kW) was recently achieved in the range of 1400–1800 °C, which produced H₂ and CB up to 2.24 Nm³/h (88% H₂ yield) and 330 g/h (49% C yield), respectively [43,44].

Packed-bed type solar reactors have been previously developed for ZnO carbo-thermal reduction or carbonaceous materials gasification [46–48]. The adopted concept consists of two cavities in series, the upper one serves as the solar absorber and contains a windowed aperture to let in concentrated solar radiation. The lower cavity serves as the reaction chamber and contains the packed-bed irradiated by an emitter plate that separates the two cavities. Thus, the emitter plate is directly irradiated and acts as solar absorber and radiant emitter to the lower cavity, for the heating of the bed topmost surface. Its main purpose is also to eliminate contact between the quartz window and the reactants/products, preventing deposition of particles or condensable gases and ensuring a clean window during operation. However, the beam down configuration does not provide uniform heating of the particle bed volume as large temperature gradients exist between the top irradiated surface and the bottom of the bed due to conductive losses and poor volumetric radiation absorption by the bed. Moreover, the reactor can only be operated in batch mode, with the packed-bed shrinking as the reaction progresses. Consequently, such a reactor configuration is suited to solid-consuming reactions but it cannot be applied to the case of catalytic reactions demanding optimal solid–gas contact duration at high temperatures and uniform catalyst heating. Therefore, the efficient utilization of such

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