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A review on solar thermal syngas production via redox pair-based water/carbon dioxide splitting thermochemical cycles



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ABSTRACT

The high power density, ease of transportation and storage and many years of development of internal combustion engine technologies have put liquid hydrocarbon fuels at a privileged position in our energy mix. Therefore processes that use renewable energy sources to produce liquid hydrocarbon fuels from H_2O and CO_2 are of crucial importance. Concentrated Solar Power (CSP) can be employed as the only energy source for the renewable production of hydrogen from water either indirectly, e.g. by supplying the electricity for electrolysis, or directly by supplying the necessary heat for thermochemically producing hydrogen. Among the various thermochemical cycles tested so far for CSP-driven hydrogen production via water splitting (WS), those based on redox-pair oxide systems, are directly adaptable to carbon dioxide splitting (CDS) and/or combined CO_2/H_2O splitting for the production of CO or syngas, respectively. The acknowledgement of this fact has recently revived the interest of the scientific community on such technologies. The current article presents the development, evolution and current status of CSP-aided syngas production via such redox-pair-based thermochemical cycles. At first the various redox oxide material compositions tested for water/carbon dioxide splitting are presented and their redox chemistries are discussed. Then the selection of suitable solar reactors is addressed in conjunction with the boundary conditions imposed by the redox systems as well as the heat demands, technical peculiarities and requirements of the cycle steps. The various solar reactor concepts proposed and employed for such reactions and their current status of development are presented. Finally, topics where further work is needed for commercialization of the technology are identified and discussed.

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1. Introduction

Conventional, fossil, liquid hydrocarbon fuels will continue to cover a major portion of the ever-increasing world energy requirements for the foreseeable future, basically due to their demand in transportation and the existing relevant extensive infrastructure. In this perspective, the direct use of non-fossil, Synthetic Liquid Fuels (SLFs) is an intensively pursued alternative. Currently, the term “Synthetic Fuel (synfuel)” refers to a liquid fuel produced at commercial scale from low energy content carbonaceous sources, such as coal, natural gas, oil shale and other biomass, that are upgraded at the expense of additional energy, also obtained from the combustion of fossil fuels [1]. The terms Gas-To-Liquid (GTL) and Coal-To-Liquid (CTL) refer to processes converting natural gas (or other gaseous hydrocarbons) and coal, respectively, into longer-chain liquid hydrocarbons such as gasoline or diesel fuel. In the GTL route, methane-rich gases are converted into SLFs either via direct conversion of methane to methanol in one step, or via syngas as an intermediate.

Syngas is a gas mixture containing varying amounts of CO and H₂ and whose exothermic conversion to fuels has been commercially practiced since a long time ago e.g. via the Fischer–Tropsch (FT) technology [2]. Syngas can be also used as a source of pure hydrogen and carbon monoxide [3,4]. Thus, in fact hydrogen and syngas are the basic raw materials to produce SLFs and chemicals via industrially available processes. These procedures can be rendered more attractive and environmentally friendlier when combined with a renewable energy source, such as solar energy [5]. Indeed, it is generally accepted that only solar-driven technologies offer a permanent solution to both oil independence and climate change due to the unmatched magnitude and availability of solar resource [6]. When solar energy is employed for the production of the raw materials for the synthesis of such fuels, the latter are characterized with the term “solar fuels”. In the broad sense this term can contain in addition to “solar hydrogen”, synthetic liquid hydrocarbons and alcohols produced from reactions between H₂ and CO that have originated from solar-aided dissociation processes as well as metal powders obtained by solar thermal reduction of metal oxides [7].

Fig. 1a (adapted from [6,8]) depicts a partial listing of the various feedstocks and solar energy variances that can be employed to produce such fuels. In this perspective, there are basically three pathways for producing syngas with the aid of solar energy: photochemical/photobiological, thermochemical and electrochemical [9–11]. The first, low-temperature route makes direct use of solar photon energy for photochemical and photobiological processes [11].

The thermochemical route uses solar heat at high temperatures supplied by Concentrated Solar Power (CSP) systems – i.e. special mirror assemblies that track the sun – for performing various high-temperature reactions that produce syngas from transformation of various fossil and non-fossil fuels (Fig. 1b). Such chemical reactions are natural gas steam reforming [12–14], gasification of solid carbonaceous materials like coal or biomass [15–17], or Water Splitting (WS) to hydrogen and oxygen. This last reaction can then be followed either by reaction of H₂ with CO₂ via the Reverse Water–Gas Shift (RWGS) reaction or by reaction of H₂ with CO coming from Carbon Dioxide Splitting (CDS) to CO and O₂, to produce syngas [18,19]. In addition, CSP systems can be employed alternatively to photovoltaics for syngas synthesis indirectly, e.g. by supplying the (solar thermal) electricity for high-temperature electrolysis of steam or of steam/CO₂ mixtures [20,21] according to the third, electrochemical route in Fig. 1a. In the next step, the most promising liquid fuels to be generated from solar syngas are methanol, dimethyl ether (DME) and Fischer–Tropsch diesel [22–24].

Among the thermochemical routes to solar syngas shown in Fig. 1b, solar reforming requires lower temperatures compared to WS/CDS thermochemical processes. However, the latter route employs CO₂ as a reactant. In this perspective lies within the broad, increasing R&D effort on developing effective solutions for reusing and “valorizing” atmospheric CO₂ as a carbon-containing raw material for the production of fuels and chemicals rather than currently treating it as a waste [21]. A review on CSP-aided reforming (dashed line) has been recently published by the present authors [25] whereas CSP-aided coal/biomass steam gasification (dotted line) has been also covered in a series of publications [16,17]. The present work is a review on the activities on CSP-aided syngas production via WS/CDS thermochemical processes (solid line). First, the evolution of thermochemical cycles from WS to CDS is presented. Then the key thermochemistries of the various redox pair materials tested for such applications are delineated, followed by their coupling to relevant solar reactors. In closing, particular issues for future work on the topic are identified and discussed.

2. Redox-pair thermochemical cycles for syngas synthesis: Chemistry issues

The so-called WS thermochemical cycles were proposed initially for the production of hydrogen from the dissociation of water to hydrogen and oxygen. The single-step thermal dissociation of

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