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On the assessment of renewable industrial processes: Case study for solar co-production of methanol and power

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HIGHLIGHTS

- Novel evaluation procedure for renewable-hybrid processes is presented.
- Evaluation procedure can be applied to a wide range of processes.
- It is presented how conventional evaluation criteria may yield misleading results.
- Process for production of methanol via solar reforming of natural gas is proposed.
- The solar methanol process simulations indicate promising efficiency.

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ABSTRACT

The transition to renewable energies has mainly happened in the field of electric energy. In industrial processes only a negligible share of the energy consumption is supplied by renewable energies. To increase this share, several processes were proposed in the past. However, for a meaningful evaluation and optimization of such processes, no appropriate criterion exists. In particular, renewable energy is still the limited resource. To identify the best use of renewable energy, a new evaluation procedure for renewable industrial processes is proposed. The novel procedure is applied to the co-production of electricity and methanol from solar energy and natural gas. The SOLME process for solar methanol production is proposed, optimized and evaluated with the proposed criterion. The results indicate that the proposed SOLME process can make more efficient use of the solar energy than conventional solar power plants. Hence, the solar methanol production is promising to initiate a further penetration of solar energy into the chemical industry.

1. Introduction

The need to reduce anthropogenic carbon dioxide emissions in order to inhibit climate change is a widely accepted fact today. Furthermore, a consensus exists that a major factor in achieving this is the extensive replacement of fossil fuels by renewable energies. Many countries are investing a substantial effort in the implementation of renewable energies. However, this implementation has mainly been successful in electricity production. For instance in Germany, which is one of the leading countries with respect to renewable energy implementation, 32.6% of the electricity was produced by renewable energies in 2015 [1]. In contrast to that, only 3.3% of the energy consumption of German industry was supplied by renewable energies in 2014 [2]. The International Energy Agency [3] determined that approximately 32% of the fossil fuel consumption in 2008 can be attributed to electricity production, leaving 68% to other sectors. These numbers show that in order to achieve an extensive replacement of fossil fuels by renewable energies, their utilization beyond electricity production is necessary. Centi et al. [4] discuss the role the chemical industry should play in the transition towards a more sustainable economy. They state that in chemical industry, not only the energy demand has to be supplied by renewable energies, but also the consumption of fossil fuels as feedstock has to be reduced. They report that the utilization of carbon dioxide as feed stock could potentially be a viable option to achieve this aim. However, since carbon dioxide represents a low energy containing feedstock, additional energy is

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Nomenclature

Abbreviations

AHR	air heated reformer
CCGT	combined cycle gas turbine power plant
El	electricity
ESI	Electronic Supplementary Information
NG	natural gas
RC	Rankine cycle
RMP	reference methanol plant
SIP	solar industrial process
SOLME	process for methanol production via solar reforming of natural gas
SPT	solar power tower (Reference)

Symbols

A_{AP}	area of receiver aperture [m^2]
C	concentration ratio of CSP technology
DNI	eirect normal irradiance [kW/m^2]
E	energy input or output [GW h]
ΔE_{Fuel}	reduction in fuel consumption [GW h]
h	efficiency of energy substitution [–]
I	flux density [kW/m^2]
S_{Energy}	energy savings ratio acc. to Bai et al. [30] [–]
T	temperature [K/°C]
η	efficiency
σ	Stefan-Boltzmann constant $\left(5.67 \cdot 10^{-8}\ \frac{W}{m^2\ K^4}\right)$

necessary for the activation of the carbon and to induce reactivity by endothermal transformation into another species [5]. Environmental benefits of carbon dioxide utilization can therefore not be taken for granted [6].

The reforming of natural gas is a widely practiced industrial process, which causes significant carbon dioxide emissions. But it also has the capability of using carbon dioxide as a feedstock. According to Bartholomew and Farrauto [7] 160 mio. tons of ammonia and more than 20 mio. tons of methanol were produced via reforming of natural gas in 2004. According to Bertau et al. [8] the production of methanol has increased to 53 mio. tons in 2011 with 90% being produced via the reforming of natural gas. A further increase in production is projected.

In the reforming of natural gas, the heat of reaction is commonly provided by combustion of additional natural gas. There are two reforming reactions, which can be employed: Steam reforming (Eq. (1)) and dry reforming (Eq. (2)). In the latter, carbon dioxide is used as oxidizing agent, hence a significant consumption of carbon dioxide occurs on the feedstock side [9]. Currently, in industrial reforming plants dry reforming of natural gas is rarely applied. It is probable that future plants will rely on mixed reforming, where both reactions take place. In mixed reforming, the product composition can also be adjusted by variation of the steam to carbon dioxide ratio in the feed. Both types of reforming are based on endothermic reactions, therefore more than 25% of the natural gas consumption are used for provision of heat to the reaction. This can be replaced by utilization of concentrated solar radiation, allowing for an effective reduction of carbon dioxide compared to the conventional reforming process. Furthermore, as recently investigated by Lu et al. [10], dry reforming of methane can be used for chemical energy storage.

$$CH_4 + H_2O \rightleftharpoons 3H_2 + CO \quad \Delta H^0_{R,298K} = 206\ \frac{kJ}{mol} \tag{1}$$

$$CH_4 + CO_2 \rightleftharpoons 2H_2 + 2CO \quad \Delta H^0_{R,298K} = 247\ \frac{kJ}{mol} \tag{2}$$

The reforming product syngas is commonly directly processed further into the final product, e.g. ammonia, methanol or hydrogen. Several authors, such as Bertau et al. [8] and Olah et al. [11] propagated an economy based on methanol, as it combines the advantages of a flexible product that can be used in chemical industry as well as in the energy sector with the advantage of a liquid product that is easily storable and transportable. Because of these advantages, Pérez-Fortes et al. [12] also investigated the production of methanol with captured CO_2 as a measure to reduce CO_2 emissions. Yang and Jackson [13] reported that a rapid increase can be observed in the methanol demand in China, where it is widely used in the chemical industry and as blend-in gasoline. If this growing methanol demand is supplied by conventional reforming of natural gas or gasification of coal it is associated with significant carbon dioxide emissions. In contrast, when the heat of reaction is supplied by solar energy, the associated carbon dioxide emissions can potentially be significantly reduced. The resulting product will be a fuel that is to a part produced from solar energy. Referring to the term solar fuels for fuels produced entirely from solar energy, this methanol may be called a partly solar fuel.

Solar reforming of natural gas is a process, that upgrades a fossil fuel with solar energy. As Steinfeld and Palumbo [14] state, the process can serve as a transition from the current fossil processes towards even more advanced and challenging processes that use solar energy as the only energy input and correspondingly achieve carbon dioxide emissions close to zero. Solar reforming has been investigated since the late 1980s and different concepts for introduction of the solar energy into the reaction volume were proposed [15]. One of those concepts is the indirectly heated solar reforming reactor, as recently investigated by McNaughton [16] and previously in the ASTERIX project [17]. In this concept, a solar receiver converts the radiation into heat and transfers it to a fluid, which then heats the reforming reactor. The advantage of this concept is the high number of degrees of freedom with respect to process design and the possibility to implement a heat storage or co-firing of fuel for off-sun operation. As shown in a previous work, this concept can achieve high efficiencies regarding the use of solar energy to increase the heating value of the reactant gas [18,19]

In this work, a process is developed and simulated that produces methanol via indirectly heated solar reforming of natural gas (**SOL**ar **ME**thanol process - **SOLME**). As plants that use concentrated solar power are usually remote from other facilities, waste heat streams of the process are minimized by heat integration. For the simulation, the whole process is modelled from the solar concentrator (in this case the heliostat field) to the final product methanol. In order to make use of all waste heat, some off-heat is converted into electric power in the process.

The developed process consumes two forms of energy streams: Natural gas and solar energy. It also produces two products: methanol and electric power. Methanol can also be considered an energy stream, but at the time being it is considered for its physical and chemical properties, not mainly for energetic considerations. Due to the use of two energy inputs and two products, optimization of such a process is not unambiguous. Maximization of the energetic or exergetic efficiency may not lead to a result that would actually represent the most beneficial process configuration. More specifically, it has to be defined first, what "beneficial" means for such a process. The need for such a definition will occur in any industrial process that consumes more than one feedstock or produces more than one product. This problem is discussed in further detail in this