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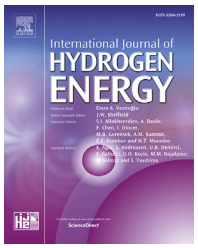

Solid oxide fuel cell operating on liquid organic hydrogen carrier-based hydrogen – making full use of heat integration potentials

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ABSTRACT

Our contribution demonstrates the technological potential of coupling Liquid Organic Hydrogen Carrier (LOHC)-based hydrogen storage and hydrogen-based Solid Oxide Fuel Cell (SOFC) operation. As SOFC operation creates waste heat at a temperature level of more than 600 °C, clever heat transfer from the SOFC operation to the LOHC dehydrogenation process is possible and results in an overall efficiency of 45% (electric output of SOFC vs. lower heating value of LOHC-bound hydrogen). Moreover, we demonstrate that LOHC vapour does not harm the operational stability of a typical 150 W SOFC short stack. By operating the stack with LOHC-saturated hydrogen, operation times of more than 10 years have been simulated without noticeable degradation of SOFC performance.

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Introduction

Liquid Organic Hydrogen Carriers (LOHC) are liquid organic compounds that can be reversibly hydrogenated and dehydrogenated, thereby storing hydrogen [1–3]. Hydrogen storage in form of liquids offers several obvious advantages compared to hydrogen storage in form of gases or solids. Liquids can be stored in high storage densities in tanks and conveyed easily by pumps. Pumping a liquid hydrogen storage

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medium between storage tank and conversion units offers the advantage of decoupling the storage volume (relevant for the MWh of stored energy) from the conversion units (relevant for the MW of power). Moreover, in such a decoupled hydrogen storage system, only the conversion units and the storage material in the conversion unit is heated and under elevated pressure, while the vast majority of the storage volume is kept at ambient conditions in a very cheap tank system.

LOHC systems always consist of pairs of molecules, i.e. at least one hydrogen-lean compound and one hydrogen-rich compound. To store hydrogen, the latter is contacted with a suitable heterogeneous catalyst and the hydrogen-lean compound of the LOHC system has to form the corresponding hydrogen-rich LOHC molecule. At times or places of hydrogen or energy need, the hydrogen-rich form can release hydrogen in presence of a suitable dehydrogenation catalyst at elevated temperature and low pressures. Very importantly, all components of the LOHC system should be liquids under normal conditions to ensure perfect compatibility with the existing infrastructure for fuels [4] Moreover, suitable LOHC systems should be characterized by a wide liquid range (low melting point, high boiling point) of all components as well as by reversible thermodynamics and fast kinetics (in almost perfect selectivity) of the hydrogenation and dehydrogenation reactions. Furthermore, all components of the LOHC systems should be favourable with respect to their toxicity and ecotoxicity [2].

All these requirements are very well fulfilled by the LOHC system dibenzyltoluene (H0-DBT)/perhydro dibenzyltoluene (H18-DBT) that has been proposed by some of us in 2014 [3,5]. Compared to the earlier discussed system toluene/methylcylohexane [3,6,7] this system takes advantage of its higher density (density H18-DBT (20 °C) is 0.92 g/ml compared to 0.77 g/ml for methylcyclohexane) leading to higher volumetric hydrogen density (57 g H_2/litre for H18-DBT vs. 47 g H_2/litre for methylcyclohexane). Its higher boiling point (390 °C for H0-DBT vs. 110 °C for toluene) and its higher intrinsic safety (flashpoint H0-DBT is 200 °C vs. 4 °C for toluene) facilitate the LOHC handling and improve intrinsic safety of this energy storage system. Compared to the also well-studied LOHC pair N-ethylcarbazole/perhydro-N-ethylcarbazole system [8–10], the H0-DBT/H18-DBT system provides a significantly better thermal stability. Isomeric mixtures of dibenzyltoluenes are used for decades on a multi thousand-ton scale as heat transfer oils, for example a well-known trademark is "Marlotherm SH®". Due to the well-established use of dibenzyltoluene in the industry for high temperature applications, H0-DBT is characterized by wide industrial acceptance, full registration, technical availability at low price, well established quality standards and full toxicological and ecotoxicological assessment. The latter identifies dibenzyltoluene as a non-toxic, non-mutagenic and non-carcinogenic compound [11]. Fig. 1 shows the hydrogen storage cycle using the LOHC-system H0-DBT/H18-DBT.

To charge the LOHC with hydrogen, H0-DBT is contacted in the hydrogenation unit with the hydrogenation catalyst (typically Ni- or Ru-based) and hydrogen at the pressure level of the source of hydrogen (e.g. a PEM electrolyzer or a methane reformer), typically 10–50 bar, at temperatures above 80 °C. The exothermic hydrogenation reaction forms H18-DBT that can be stored and transported like diesel. At the time and at the place of hydrogen demand, H18-DBT is contacted in a dehydrogenation reactor with the dehydrogenation catalyst (typically Pt-based) at 1–5 bar hydrogen pressure and 260–320 °C. For this endothermic step that liberates hydrogen and forms back the H0-form of the LOHC system, the same amount of heat has to be provided to the reactor that has been liberated previously in the exothermic hydrogenation step.

Like for all other purely hydrocarbon LOHC systems, the amount of hydrogenation/dehydrogenation heat is relatively high in the case of H18-DBT dehydrogenation: 65 kJ/mol H_2 have to be provided which corresponds to 27% of the lower heating value of the hydrogen stored in the H18-DBT system [12]. The need to provide this heat at the required temperature level during times and at the location of energy demand has been regarded in the literature so far as a major drawback of hydrogen and energy storage using LOHC systems [13].

In this contribution we present a new way to overcome this drawback, namely the combination of the endothermic hydrogen liberation from H18-DBT and the exothermic conversion of this hydrogen in a Solid Oxide Fuel Cell (SOFC). We demonstrate that the combination of these two units in optimized heat integration provides a way to cover the heat of LOHC dehydrogenation to a very large extent by the waste heat of the SOFC unit. Thus the efficiency of the SOFC unit operated on LOHC-bound hydrogen becomes very close to that of the SOFC operated on pure hydrogen. Both options offer – in contrast to SOFC operation on natural gas or LPG – an attractive way for infrastructure independent, CO_2-free electricity production combined to a very convenient way of high-capacity energy storage. The amount of stored energy is only dependent on the size and filling of the LOHC tank attached to the SOFC unit.

SOFCs are characterized by an operation temperature range between 700 and 1000 °C, depending on the cell type used [14]. However, the operation temperature does not have a major impact on the exhaust gas temperature leaving the system, because a big portion of the energy in the gas is necessary to pre-heat the incoming air to the required stack inlet temperature [15–18]. Depending on the system configuration, the exhaust gas temperature can vary between 300 and 900 °C. In general, this fits quite well to the requirements of the LOHC operation, except for the fact that an overheating of the LOHC has to be avoided. The maximum film temperature of H18-DBT should not exceed 350 °C to ensure its long-term stability.

The electrical system net efficiency can achieve high values in case of an optimal SOFC configuration. For natural gas as fuel in combination with internal steam reforming, values in the range of 60% can be achieved [19–23]. In case of using hydrogen as fuel, efficiencies tend to be smaller because a smaller portion of the energy content in hydrogen can be transferred into electrical energy. Operating at the same cell voltage and fuel utilization this results in 17% lower stack efficiency. Based on standard configurations electrical system net efficiencies in the range of 50% are achievable with hydrogen as the SOFC fuel [24].

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