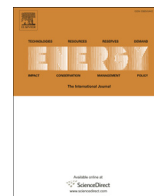




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Energy

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# Addressing fuel recycling in solid oxide fuel cell systems fed by alternative fuels

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## ARTICLE INFO

### Article history:

Received 31 October 2016

Received in revised form

6 March 2017

Accepted 14 March 2017

Available online xxx

### Keywords:

SOFC

Fuel cell

Alternative fuels

Anode recirculation

Methanol

Ethanol

Ammonia

Biogas

## ABSTRACT

An innovative study on anode recirculation in solid oxide fuel cell systems with alternative fuels is carried out and investigated. Alternative fuels under study are ammonia, pure hydrogen, methanol, ethanol, DME and biogas from biomass gasification. It is shown that the amount of anode off-fuel recirculation depends strongly on type of the fuel used in the system. Anode recycling combined with fuel cell utilization factors have an important impact on plant efficiency, which will be analysed here. The current study may provide an in-depth understanding of reasons for using anode off-fuel recycling and its effect on plant efficiency. For example, it is founded that anode recirculation is not needed when the plant is fed by ammonia. Further, it is founded that when the system is fed by pure hydrogen then anode recirculation should be about 20% of the off-fuel if fuel cell utilization factor is 80%. Furthermore, it is founded that for the case with methanol, ethanol and DME then at high utilization factors, low anode recirculation is recommended while at low utilization factors, high anode recirculation is recommended. If the plant is fed by biogas from biomass gasification then for each utilization factor, there exist an optimum anode recirculation at which plant efficiency maximizes.

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## 1. Introduction

With an ever increasing demand for more efficient power production and distribution, some main research and development for the electricity production is identified as efficiency enchantments and pollutant reduction, especially carbon dioxide among others. Alternative fuels have also been recognized as potential element in decreasing emissions locally such final at end users.

Solid Oxide Fuel Cells (SOFCs) are recognized as one of the most promising types of fuel cells, particularly in terms of energy production. Besides pure hydrogen they can be fed variety of fuels such as Natural Gas (NG), ethanol, Di Methyl Ether (DME), methanol and syngas from gasification of biomass or municipal waste. They are expected to produce clean electrical energy at high conversion rates with low noise and low pollutant emissions [1]. They can tolerate sulfur compounds at concentrations higher than those tolerated by other types of fuel cells. Additionally, unlike in most fuel cells, carbon monoxide can be used as a fuel in SOFCs. Due to the above-mentioned advantages, SOFCs are considered to be a strong candidate for either hybrid systems or integration into currently

deployed technologies. Therefore, SOFC plants have been the subject of many studies since the beginning of 90s. For example [2] showed that electrical efficacy of a hybrid plant consisting SOFC, gas turbine and steam turbine may reach about 70% which is encouraging to further investigate on such plants.

Numerous studies on SOFC based systems have been considered in the literature among them SOFC–gas turbines hybrid systems have extensively studied, for example the study of [3] shows that plant efficiency reaches about 60% at full load while its part-load (until 50%) efficiencies are also above 50%. In the study of [4], the net efficiency of a SOFC plant was calculated to be about 28–29% when it is fed by biogas from biomass gasification. A study on biogas (assumed to be available in the gas grid without providing the source) fuelled SOFC micro-CPH in Ref. [5] showed that an overall CHP efficiency of about 80% is achievable for single-family detached dwellings. In another study carried out in Ref. [6], it was concluded that a SOFC plant fed by biogas from organic wastes may reaches electrical efficiencies of about 34% at approximately 55% utilization factor. Biogas from wastewater treatment facilities was used in the study of [7] to estimate electrical efficiency of a SOFC plant. The study showed that plant efficiency would be about 41% if the utilization factor was selected to be 65%. A study on syngas from municipal waste gasification carried out in Ref. [8]

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<http://dx.doi.org/10.1016/j.energy.2017.03.082>

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showed that plant efficiency of such integrated gasification-SOFC plant approaches about 43% with utilization factor of about 80%. These are some examples of many that have been explored by researchers for utilization of waste to energy in sustainable modern societies.

SOFC fed by different fuels have also studied by many researchers. In the study of [9], the net efficiency of a 2 kW<sub>el</sub> SOFC plant was calculated to be about 55% when the fuel was methanol. If DME was used as fuel, then the study of [10] showed that the plant efficiency will be about 50%. The study of [11] showed that plant net efficiency of about 53% is achievable when the fuel of SOFC was bioethanol. In Ref. [12] an ammonia fed SOFC integrating with gas turbine was studied and the results shown efficiencies close to 56%. Comparison performance of SOFC plants fed by alternative fuels have also been studied in Ref. [13] in which a single general modelling approach was used for the investigation. This single modelling approach with the same simulating code was also evaluated to ensure accuracy of the modelling and methodology used in the present study as documented in Ref. [13].

Despite extensive studies on SOFC based power plants, investigations on anode recycle SOFC systems fed by NG is comparably limited. Anode off-fuel recycling (anode gas recycle) is essential in SOFC systems fed by NG in order to provide steam for the steam reforming reactions in a pre-reformer prior to the SOFC cells. Exclusively all studies on anode recycling are about carbon formation and carbon deposition in the pre-reformer of a natural gas (NG) feed SOFC stack. Most of these studies are on stack level and do not on investigate the effect of anode recycling on system level and plant performances. For example, the experimental studies of [14] showed that the limit for O/C ratio (oxygen-carbon ratio) to avoid carbon formation depends on the purity of gas. Their study showed that the limit of O/C ratio for carbon formation for nickel catalyst was between 0.9 and 1.0 for Russian natural gas and between 1.0 and 1.25 for Danish natural gas. If precious metal catalyst used, then the limit was between 0.5 and 0.75 irrespectively of natural gas composition. The effects of SOFC anode recycle on catalytic diesel reforming and carbon formation was also studied in Ref. [15] experimentally. This study showed that anode recycle is more effective than reformer recycle when it comes to carbon formation in the reformer (off-fuel from SOFC, not reformate gas out of reformer). Steam recycling for internal methane (and/or natural gas) reforming in SOFCs to analyse the carbon deposition using computational fluid dynamic was used in Ref. [16]. This study showed also that anode recycling is need to decreases carbon formation when fuel is methane or natural gas. Electric power generation of 380 W SOFC stack fed by methane with and without anode recycle was demonstrated in Ref. [17]. Their study showed that anode recycle increases stack efficiency by about 10% when anode recycle is used. It was reported in Ref. [18] that cell voltage could be improved by anode off-fuel recycle in solid oxide fuel cell fed by pure methane. The study was on a cell level (not system level) with distinguished conclusion. The study of [19] showed that anode recycling enables the operation of a SOFC stack at low fuel utilizations without sacrificing the electrical efficiency of the stack. The maximum electrical efficiency of 57% was reached at 60% fuel utilization when the fraction of recycled fuel was 66%. If no anode gas recycling was applied then the maximum electrical efficiency was about 53% with about 77.5% fuel utilization.

Despite many studies on anode recycling and carbon formation, studies on anode off-fuel recycling on plant efficiency are very limited and exclusively all are about natural gas (and/or methane) fed fuel. No study on off-fuel recycling with alternative fuels is found in the open literature, which makes the basis of the current study. The effect of anode recycling on plant efficiency using different types of fuels is investigated here which is completely novel and

has not been studies elsewhere. A single study with similar conditions and prerequisites will thus reveal the importance of off-fuel recirculation on plant performance when the fuel is an alternative fuel. The findings in the current study may help SOFC system developer on boosting their plant efficiencies when alternative fuels are used. All foundlings are new and have not been reported elsewhere.

## 2. Plant layout and modelling methodology

Fig. 1 displays a typical SOFC plant with natural gas and/or methane as fuel. A similar layout can also be seen in e.g. Refs. [1,5], and [20]. Air is compressed and preheated in a cathode preheater (CP) before entering the cathode side of the fuel cell. Natural gas (and/or pure methane) is firstly reformed and then preheated in an anode preheater (AP) before entering the anode side of the fuel cell. Depending on the utilization factor, a portion of the feed fuel will leave the anode side without reacting inside the fuel cells. The remaining fuel (off-fuel) and air (off-air) is then sent to a burner for further combustion. The off-gases after the burner is used to pre-heat both incoming air and fuel into the fuel cell. In order to provide steam for the reformer some of the off-fuel is recycled which calls for anode recirculation (or off-fuel recirculation). Even though the main purpose of the off-fuel recirculation is to provide steam for the steam reforming but it will also improve stack efficiency since more fuel is reacted inside the cells and therefore more power will be generated (see e.g. Ref. [19]). On the other hand, since no external steam is provided to the steam reformer (during normal operation) then it will be important that steam-carbon-ratio (S/C-ratio) is approximately above 1.8 to avoid carbon deposition, which has a significant effect on the reformer performance and lifetime, see e.g. Ref. [5]. However, most of the researchers assumes the value of 2 to be on the safe side, such as in Refs. [5,6,21,22]. Note that it is generally believed that carbon deposition can be determined by S/C ratio but the experimental study of e.g. Ref. [23] shows that not only S/C but also the extent of equilibrium in the gas mixtures should be taken into account to control the carbon deposition (O/C ratio). However, as shown in the study of [24] carbon deposition is not an issue in SOFC fed by wood gas from biomass gasification.

However, when changing the fuel into alternative fuels such as ammonia, pure hydrogen, methanol and ethanol then there is no problem on limiting the S/C (or O/C) ratio if a pre-reformer is used (see e.g. the C–H–O ternary diagram in Ref. [5]). If biogas (from biomass gasification) is used, then there will be enough steam in the gas and such problem does neither exist, as discussed in Ref. [24]. For such alternative fuels the question will be if off-fuel recirculation is needed or not and if it is needed then how it will effect on plant performance. This is the basis of the present study, which is entirely new and not been studied elsewhere.

In this study, the thermodynamic results are obtained using the Dynamic Network Analysis (DNA) simulation tool (see, e.g. [25]), established at DTU since 1983. The program has continuously been developed to be generally applicable for different energy systems. It includes a component library, thermodynamic state models for fluids and standard numerical solvers for differential and algebraic equation systems. The component library contents models for heat exchangers, burners, turbo machinery, decanters, energy storages, valves and controllers, among many others. Fig. 2 illustrates the calculation procedure used in the program.

DNA is a component-based simulation tool, meaning that the model is formulated by connecting components together with nodes and adding operating conditions to create a system. The equations include mass and energy conservation for all components and nodes together with the relations for the thermodynamic properties of the fluids in the system. The total mass balance and

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