



Advances in ethanol autothermal reforming



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ABSTRACT

Fuel cell systems powered by bio-fuels such as ethanol are potential solutions to local, small scale electricity needs, especially in remote, off-grid locations. In recent years, many researchers around the world have studied the on-site autothermal reforming of ethanol to hydrogen, which can then be used in the fuel cell. However, a comprehensive review of those efforts has not been carried out. In this paper, a detailed literature review of experimental and numerical research is presented, with a focus on the catalysts, reactor design, and simulation and modelling efforts that sought to understand interactions among fluid flow, heat and mass transfer and chemical kinetics. Our review indicates that although considerable work has been carried out on the development of catalysts, relatively fewer studies report system level experiments and simulations that are necessary before these systems can be commercially deployed. Thereby, we also identify areas for further research in the area of ethanol autothermal reforming.

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1. Introduction

International energy outlook projections indicate that with world GDP rising by 3.6% each year, world energy use will grow by 56% between 2010 and 2040. Since most economic growth is expected to occur in developing countries such as China and India, more than 50% of these increases in energy demand are expected to be attributable to these countries [1]. With these rising energy demands and depleting conventional energy sources,

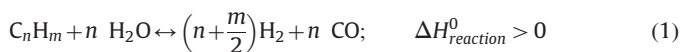
non-conventional sources of energy are being sought [2–6]. Hydrogen as an energy carrier, and fuel cells as energy conversion devices have gained traction recently in the context of the shift toward more efficient and less carbon-intensive energy solutions [7–9]. Broadly, the most common fuel cell systems can be classified according to the primary fuel used: (1) hydrogen based, (2) hydrocarbon based, and (3) solar energy (water-electrolysis) based [10]. For each type of fuel cell system, a great number of research efforts are underway in industry, academia and governments to overcome challenges, reduce cost and commercialize the various technologies involved [11–14].

Of these, fuel cells using pure hydrogen are currently not suitable for auxiliary power or remote power backup, owing to the costs associated with the transport and storage of hydrogen.

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Fuel Cells running on hydrogen produced by water-electrolysis are still not cost effective for remote off-grid applications [15]. And thus, a lot of work has been seen recently on hydrocarbon based fuel cell systems [16–18]. There are three main reforming methods employed to obtain hydrogen from ethanol: steam reforming, partial oxidation and autothermal reforming.

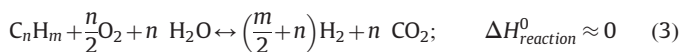
Steam reforming involves reaction of a fuel with steam over a catalyst. This process has the maximum theoretical yield of hydrogen, and has been studied extensively [17,19–22]. However, it is a strongly endothermic reaction requiring external heat, and therefore using steam reforming increases system complexity.



Catalytic partial oxidation, on the other hand involves reaction of an oxygen lean mixture of fuel and air for incomplete combustion. The obvious advantage is that the reaction is exothermic; however, it has the least hydrogen yield of all the reforming reaction. This reaction too has been actively studied in the past decade [23–27].



Autothermal reforming combines the endothermic steam reforming reaction and the exothermic partial oxidation to get a (nearly) thermodynamically neutral reaction. This has a lower H₂-yield than SR, but the thermodynamic neutral nature makes it a better alternative for fuel reforming. This has been studied through various theoretical and experimental studies for comparing these reactions [17,28–35]



Fuel processors can work with conventional fuels like diesel, natural gas and methanol to yield hydrogen rich mixtures that can be directly used to drive fuel cells. However, fuels derived from biomass, because of the additional advantages of being locally available, carbon neutral and renewable, are the most favourable candidates. Much work is being done to promote the use and understanding of these fuels in reforming reactions [36–38]. However among these bio-fuels, ethanol is much favoured over others due to its availability via multiple pathways, many of which are subjects of ongoing research [39–41]. Among other advantages of using ethanol as a fuel source are the ease of reforming, safety in storage, handling and transportation, and its non-toxicity. Ethanol is also preferred as a fuel source as it forms a closed carbon loop, i.e. it adds little or no carbon to the atmosphere via the reforming process [31].

However, in open literature it is observed that compared to autothermal reforming, ethanol steam reforming has received more attention due to its relatively higher conversion efficiency [22,42–44]. We have not found a comprehensive literature survey on autothermal reforming in recent years. This paper aims to review the technological advances in autothermal reforming of ethanol for hydrogen production in the last five years. In this context, we review catalyst selection, reactor design and simulation studies.

Section 2 describes in detail the catalysts developed for ethanol autothermal reforming. It also touches on the aspect of catalyst stability. Section 3 briefly describes previous works in reactor development, in particular for ethanol autothermal reforming. Section 4 summarizes the simulation and modelling studies that have been carried out to understand the process.

2. Catalysts for ethanol ATR

In the ethanol ATR process, catalysts play a very important part in ensuring conversion and preventing coke formation. However, each catalyst may promote the reforming reaction through distinct pathways, and therefore catalyst selection plays an important role. Each catalyst has different selectivity towards hydrogen and other side products, and for fuel cell system purposes, the aim is to maximize H₂ selectivity while minimizing catalyst coking.

Very broadly, the catalysts can be divided as (1) noble metal-based, (2) non-noble metal-based and (3) bi-metallic catalysts. During the last decade, noble metal catalysts had been the subject of extensive research due to their high conversion efficiencies and high H₂ selectivities. However, a survey of recent literature suggests that the focus of research has now shifted to development of non-noble metal catalysts that can replace the conventional noble metal catalysts. This is primarily because of the cost involved in noble metal catalysts which may adversely impact market acceptability. Few research works also focus on developing novel catalysts which involve nanostructures and composite phases [45,46]. For instance, perovskite phase also has been studied extensively for ethanol ATR [47–51].

2.1. Noble metal catalysts

Most catalysts in this category are prepared by incipient wetness impregnation method. Incipient wetness impregnation (IW or IWI), also called capillary impregnation or dry impregnation, is a commonly used technique for the synthesis of heterogeneous catalysts. Typically, the active metal precursor is dissolved in an aqueous or organic solution. The metal-containing solution is then added to a catalyst support containing the same pore volume as the volume of the solution that was added. Capillary action draws the solution into the pores. The catalyst can then be dried and calcined to drive off the volatile components within the solution, depositing the metal on the catalyst surface. Rh, Pt, Pd, Ru and Ir have been extensively studied for ethanol autothermal reforming [52–54].

Chen et al. used Ir/La₂O₃, and studied the effect of in situ dispersion of Ir [55]. They proposed that the excellent performance of Ir/La₂O₃ was due to the formation of La₂O₂CO₃ and an in situ dispersion effect of Ir on La₂O₂CO₃. Due to the in situ dispersion effect, the resulting catalyst possessed enhanced ATR performance in terms of activity and stability. Based on the insight into the in situ dispersion mechanism of Ir over La₂O₂CO₃-II, a supersonic-assisted impregnation method was used to improve the surface iridium concentration and reduce the initial Ir domain size. This significantly enhanced the activity and stability of Ir/La₂O₃ catalysts for the ATR reaction. A systematic study for identifying the key relationships between the catalyst morphology/structure and catalytic performance in autothermal reforming of ethanol was carried out over a series of Ir/CeO₂ catalysts by Cai et al. [56]. They showed that the dispersion of both the ceria support and the noble metal phase control key parameters such as the diffusion of oxygenated adspecies along the ceria surface, surface basicity, and the concentration of coupled sites formed at the metal/support interface between peripheral ceria defects and coordinately unsaturated sites on Ir particles. All of these factors are found to be critical for ethanol conversion, selectivity toward hydrogen production, and resistance to aging.

Rh on various supports have been independently by various researchers [57–59]. It has been reported to be a very efficient catalyst for conversion of ethanol, generally with 100% conversion at low temperatures with high selectivity towards hydrogen. Pt, Pd and Ru have been similarly studied [57,58,60–63]. The results for both simple catalysts and those employed in a membrane reactor

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