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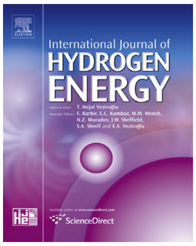

Hydrogen production by steam reforming of DME in a large scale CFB reactor. Part I: Computational model and predictions

Francis A. Elewuwa [a], *Yassir T. Makkawi* [b,*]

[a] *European Bioenergy Research Institute (EBRI), School of Engineering and Applied Science, Aston University, Birmingham B4 7ET, UK*

[b] *Chemical Engineering Department, American University of Sharjah, P.O. Box 26666, Sharjah, United Arab Emirates*

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ABSTRACT

This study presents a computational fluid dynamic (CFD) study of Dimethyl Ether steam reforming (DME-SR) in a large scale Circulating Fluidized Bed (CFB) reactor. The CFD model is based on Eulerian–Eulerian dispersed flow and solved using commercial software (ANSYS FLUENT). The DME-SR reactions scheme and kinetics in the presence of a bifunctional catalyst of $CuO/ZnO/Al_2O_3+ZSM\text{-}5$ were incorporated in the model using in-house developed user-defined function. The model was validated by comparing the predictions with experimental data from the literature. The results revealed for the first time detailed CFB reactor hydrodynamics, gas residence time, temperature distribution and product gas composition at a selected operating condition of 300 °C and steam to DME mass ratio of 3 (molar ratio of 7.62). The spatial variation in the gas species concentrations suggests the existence of three distinct reaction zones but limited temperature variations. The DME conversion and hydrogen yield were found to be 87% and 59% respectively, resulting in a product gas consisting of 72 mol% hydrogen. In part II of this study, the model presented here will be used to optimize the reactor design and study the effect of operating conditions on the reactor performance and products.

Introduction

Hydrogen is currently receiving increasing interest as an alternative source of clean energy and has high potential applications, including the transportation sector and stationary power generation. Traditionally, hydrogen is produced from fossil fuels by steam reforming of natural gas or heavy hydrocarbons; however recently, there is growing research and development activities on hydrogen produced from other sources, such as biomass, methanol (MeOH) and Dimethyl Ether (DME). DME is particularly attractive for hydrogen production because it contains higher mass fraction of hydrogen (13 wt%) and the reforming process can be carried out at a lower temperature (200–350 °C) compared to other options,

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* *Corresponding author*. Tel.: +971 6 515 2167; fax: +971 6 515 2979.
E-mail address: ymakkawi@aus.edu (Y.T. Makkawi).

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such as natural gas reforming for instance [1,2], hence, less energy intensive.

There is appreciable number of published research papers on methane and methanol steam reforming processes, however there is less focus on DME Steam Reforming (DME-SR), particularly in fluidized bed reactors. As far as the author's knowledge, there are no publications on experimental or computational modelling of the DME-SR in a circulating fluidized bed (CFB) system. Owing to their superior heat transfer, intensive solid–gas mixing and the potential integration with catalyst regeneration within a closed loop dual fluidized bed (DFB), it could be easily argued that the circulating fluidized bed is one of the most important technologies for industrial scale hydrogen production.

Numerical investigations have shown the potential of the DFB in carrying out simultaneous reforming reaction, catalyst regeneration and carbon dioxide capture [3–5]. This has been demonstrated for hydrogen production by methane steam reforming were the reforming reactions and carbon dioxide capture are assumed to take place in one reactor while the sorbent regeneration takes place in a second joint reactor, thus forming a continuous closed loop flow. The DFB system has also been frequently reported as a promising technology for other large scale processes involving solid–gas flow [6,7]. Experimental studies on hydrogen production via catalytic steam reforming of methanol have shown that a fluidized bed reactor has a 20% higher conversion efficiency compared to fixed bed reactors [8]. This is mainly due to the advantages of having larger surface contact area, uniform temperature distribution and better control of the gas residence time.

The DME-SR reaction occurs in two-steps: DME hydrolysis and steam reforming of methanol, with the latter being produced from the first step [9–11]. It is this two-steps reaction that necessities the use of a bifunctional catalyst to facilitate both reactions. However, depending on the catalyst used and the reaction parameters, some side reactions may occur which include water-gas shift reaction (WGSR) and DME decomposition reaction [12]. The catalysts that have been frequently reported in DME-SR are alumina or zeolite acid catalyst in combination with metallic copper oxides [10,13–15]. The zeolite component is the preferred choice as acid site for the hydrolysis of the DME because it promotes reaction at a lower temperature compared to the alumina [16]. It is understood that the alumina catalyst promotes the reverse WGSR while the zeolites acid site promotes the forward reaction. Hence, the zeolite help in increasing the hydrogen yield by converting the carbon monoxide formed during the process to hydrogen and carbon dioxide [17]. The metal based catalyst $CuO/ZnO/Al_2O_3$ has been used in methanol steam reforming studies [18–20]. The same metal catalyst, but with an added ZSM-5 zeolite catalyst, has been recently reported in experimental studies of DME-SR [21]. Such a bifunctional catalyst is suitable for achieving very high hydrogen yield with minimum carbon monoxide produced due to presence of WGSR.

Review of the available literature show that numerical studies on DME-SR in fluidized bed reactors are limited. Feng et al. [22] developed a one-dimensional isotherm plug flow model to simulate DME-SR in a fixed bed reactor with bifunctional catalyst $CuO/ZnO/Al_2O_3+ZSM\text{-}5$. The DME hydrolysis reaction was implemented in the model using a simple multi-response objective function with the kinetics derived from the elementary reaction steps of methanol dehydration to DME. Yan et al. [21] conducted a numerical study on DME-SR in a micro-reactor with the same bifunctional catalyst and predicted up to 70–90% DME conversion when operating the reactor within the temperature range of 240–280 °C. Other catalysts, such as mechanically mixed HPA/Al_2O_3 acidic catalyst and Cu/SiO_2 metallic catalyst, have been tested experimentally in a fixed bed reactor and reported to achieve near 100% DME conversion at 290 °C [23]. A Numerical study using STAR-CD software has been reported to investigate the fluid flow, heat transfer and chemical reactions during DME-SR in a fixed bed [12]. The DME-SR kinetics was based on $CuO–NiO/Al_2O_3/ZrO_2+ZSM\text{-}5$ catalyst. The results have shown fast decrease of the temperature at the entrance region of the reactor because of the endothermic nature of the DME-SR reaction. Beyond the entrance region, the temperature was found to increase along the axial length of the bed to the exit. Recently, a mathematical model coupling mass, energy and momentum equations has been reported to investigate the DME SR in a novel fluidized bed reactor using Comsol commercial software [24]. The reactor was assumed to be thermally driven by exhaust gas recycling. The result provided useful data on the effect of varying the exhaust gas velocity and other operating parameters on the DME conversion and hydrogen yield.

In this study, which is the first in a series of two, Eulerian–Eulerian model is used to simulate the DME-SR in a CFB reactor, as part of a proposed concept of closed loop DFB system. The model was solved using commercial software (ANSYS FLUENT). The chemical reactions and kinetic in the presence of bifunctional catalyst $CuO/ZnO/Al_2O_3+ZSM\text{-}5$ were implemented in the model using in-house developed user-defined function (UDF). The model was first validated by comparison with experimental data [17], then used to show the detailed hydrodynamics and thermochemical behaviour of the CFB reactor at a selected operating conditions. In part II of this study, this model will be used for optimizing the reactor design and studying the effect of operating conditions on the reactor performance and products.

Proposed concept and computational domain

It is proposed that the overall DME-SR process is carried out in an industrial scale DFB system. This system mainly consist of two coupled reactors; one for catalytic steam reforming of the DME and the other the thermal regeneration of the catalyst. The solid and gas mixture leaving both reactors are separated using two cyclones as part of a closed loop system. The arrangement of the reactors and the particulate flow throughout the system is described schematically in Fig. 1. In this study, focus is only made on the DME-SR reactor; hence the cyclones and regeneration reactor are outside the scope of the study. The steam reforming reactor is assumed to operate in a riser mode and has the dimensions of 3 m diameter and 15 m height. The choice of this size is made to replicate commercially proven scale of a CFB reactor (e.g. PYROFLOW CFB system of Goodrich Co in Illinois U.S.A as reported in Ref.

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