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## ACCEPTED MANUSCRIPT

### Oxidative Coupling of Methane in Solid Oxide Fuel Cell Tubular Membrane Reactor with High Ethylene Yield

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#### Abstract

A Mn-Ce-Na<sub>2</sub>WO<sub>4</sub>/SiO<sub>2</sub> catalyst integrated solid oxide fuel cell tubular membrane reactor design is developed for oxidative coupling of methane (OCM). The OCM reaction carried out using the tubular membrane reactor establishes both superb hydrocarbon selectivity and outstanding ethylene-to-ethane ratio over the conventional fixed bed OCM or the button cell reactor. A remarkable performance of 60.7% methane conversion with 41.6% C2+ selectivity, 5.8 ethylene-to-ethane ratio and 19.4% ethylene yield is achieved using the tubular membrane reactor and the Mn-Ce-Na<sub>2</sub>WO<sub>4</sub>/SiO<sub>2</sub> catalyst, demonstrating considerable advantages over conventional OCM.

#### Keywords

OCM, SOFC, methane, ethylene, membrane reactor, Mn-Ce-Na<sub>2</sub>WO<sub>4</sub>

#### 1. Introduction

The increasing demands in energy along with the expected long-term decline in petroleum reserves require alternative hydrocarbon feedstocks for the production of important industrial chemical intermediates such as ethylene. Methane (CH<sub>4</sub>), the major component of natural gas, is a promising starting material because it has the desirable carbon-hydrogen ratio as well as abundant reserves.[1] The efficient conversion of methane, which otherwise is primarily used as fuel for power generation, to value-added chemicals is highly demanded. Conventionally, multiple steps are required to convert methane into value-added chemicals, where methane is first reformed to synthesis gas (CO and  $H_2$ ), followed by the Fischer-Tropsch process to produce alkenes or the catalytic conversion to produce methanol and the subsequent methanol-to-olefin (MTO) processes. However, the capital and maintenance costs are relatively high in these rather complicated processes. A direct route for the conversion of methane into value-added chemicals is highly demanded.[2,3]

Oxidative coupling of methane (OCM) has been studied for decades [4–13] and is recently being commercialized by Siluria Technologies. In a typical OCM process, methane and pure oxygen are co-fed

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