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## ACCEPTED MANUSCRIPT

# Chemical looping dry reforming of methane: towards shale-gas and biogas valorization

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#### Abstract

Chemical looping dry reforming of methane (CLDRM) is performed by exposing a Ni/CeO<sub>2</sub> to CH<sub>4</sub> and CO<sub>2</sub> in a cyclic way. The solid acts as oxygen carrier producing syngas (CO+H<sub>2</sub>) during exposure to CH<sub>4</sub>, and is re-oxidized during exposure to CO<sub>2</sub>. Absence of CO<sub>2</sub> during syngas production avoids reverse water gas shift reaction and allows high selectivity. Oxygen capacity is restored and residual carbon formed at the surface is removed during exposure to CO<sub>2</sub>. In view of biogas reforming without prior separation of compounds, CLDRM is tested in presence of CO<sub>2</sub> in the CH<sub>4</sub> feed. CLDRM performance remain high as long as methane is in excess with respect to CO<sub>2</sub>. At 800°C, CO<sub>2</sub>/CH<sub>4</sub> ratio up to 0.8 may be used, whereas at 600°C, CO<sub>2</sub>/CH<sub>4</sub> should be kept below 0.4. These results allow considering direct biogas reforming in looping conditions. For shale gas valorization, the reactivity of ethane in CLDR conditions is explored. In all the explored temperature range, ethylene formation is observed. Optimization of conversion and selectivity to syngas from ethane reforming should nevertheless be possible by adapting the amount of solid and reaction conditions to the higher oxygen need for ethane reforming in comparison to that of methane.

#### Keywords:

chemical looping; dry reforming; methane; ethane; biogas; shale-gas

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