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$LaMn_{1-x}Ni_xO_3$ Perovskites as Oxygen Carriers for Chemical Looping Partial Oxidation of Methane

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Abstract : Chemical looping partial oxidation of methane (CLPOM) is a novel technology to produce high-quality syngas with an auto-thermic process and low equipment investment. The development of oxygen carriers is important for the improvement of the CLPOM performance. In this work, the effect of the nickel-substitution proportion on the performance of $LaMn_{1-y}Ni_yO_{3+\delta}$ perovskites for CLPOM was studied in the aspect of reactivity, syngas selectivity, resistance towards carbon deposition and thermal stability in cyclic redox process. The $LaMn_{1-x}Ni_xO_3$ perovskite oxides with x = 0, 0.1, 0.2 were prepared by the sol-gel method. The performance of $LaMn_{1-y}Ni_yO_{3+\delta}$ perovskites for CLPOM was investigated through the characterization of XRD, H_2-TPR, XPS, and fixed-bed experiments. The characterization and test results suggest that the doping of nickel enhances the generation rate of syngas, leading to high syngas yield, methane conversion, and syngas selectivity. This is attributed to the that the introduction of nickel provides active sites to promote the methane activation on the surface and causes the addition of oxygen vacancies to accelerate the migration of oxygen anion in the bulk of oxygen carrier particles. On the other hand, the introduction of nickel causes carbon deposition to occur earlier. The best substitution proportion of nickel is y=0.1 and $LaMn_{0.9}Ni_{0.1}O_{3+\delta}$ could produce high-quality syngas with a yield of 3.54 $mmol \cdot g^{-1}$, methane conversion of 80.7%, and CO selectivity of 84.8% at 850℃. In addition, the $LaMn_{0.9}Ni_{0.1}O_{3+\delta}$ oxygen carrier exhibits superior and stable performance in the cyclic redox process.

Keywords : chemical looping partial oxidation of methane, $LaMnO_{3+\delta}$, Ni doping, syngas, carbon deposition
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