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化学链制氢中Fe_2O_3/$LaFeO_3$载氧体的性能研究

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Performance of Fe_2O_3/$LaFeO_3$ as oxygen carrier in chemical-looping hydrogen generation

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摘要 采用柠檬酸络合法制备Fe_2O_3/$LaFeO_3$复合氧化物，将该氧化物作为化学链制氢过程的载氧体，在反应温度为900 ℃、常压下，对Fe_2O_3/CH_4（剂烷比）、进水量、金属负载量进行了考察。结果表明，剂烷比为2：1、进水量为0.1 mL、质量分数15%Fe时载氧体性能最好，甲烷转化率达到60%，单次循环氢气产量为45 mL。将该评价结果与XRD和H_2-TPR表征结果进行关联发现，反应过程的活性位不是金属氧化物，而是吸附氧，而且吸附氧越容易还原，甲烷转化率和氢气产量越高。通过连续60次还原-氧化循环发现，该载氧体上甲烷转化率和氢气产量比较稳定，循环后仍然保持钙钛矿结构。

关键词： 化学链制氢　载氧体　柠檬酸络合法　钙钛矿　甲烷

Abstract: Fe_2O_3/$LaFeO_3$ was prepared by citric acid complexation method and used as oxygen carrier in chemical-looping hydrogen generation; the effect of Fe_2O_3/CH_4 ratio, stream amount and metal loading on the performance of Fe_2O_3/$LaFeO_3$ as oxygen carrier at 900 ℃ and atmospheric pressure were investigated. The results showed that the oxygen carrier performs best with CH_4 conversion of 60% and single-loop H_2 output of 45 mL, when the ratio of Fe_2O_3/CH_4, stream input and Fe loading are 2, 0.1 mL, 15%, respectively. The XRD and H_2-TPR results suggest that the active site is the adsorbed oxygen rather than metal oxide; the easier for the reduction of the adsorbed oxygen, the higher are the conversion of CH_4 and the output of H_2. Moreover, the oxygen carrier is still in perovskite structure and CH_4 conversion and H_2 output remain stable even after 60 cycles.

Key words: chemical-looping hydrogen generation　oxygen carrier　citric acid complexation method　perovskite　methane

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