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Enhanced fluidized bed methanation over a Ni/Al_2O_3 catalyst for production of synthetic natural gas

Jun Li [a,*], Li Zhou [a], Pengcheng Li [a,b], Qingshan Zhu [a,*], Jiajian Gao [a,b], Fangna Gu [a], Fabing Su [a]

[a] State Key Laboratory of Multiphase Complex Systems, Institute of Process Engineering, Chinese Academy of Sciences, Beijing 100190, China
[b] University of Chinese Academy of Sciences, Beijing 100049, China

HIGHLIGHTS

- The CO methanation performance over a Ni/Al_2O_3 catalyst was investigated in a fluidized bed reactor.
- The methanation performance of the catalyst depends on the fluidization quality.
- The fluidization and methanation reaction can be improved by adding Al_2O_3 particles.
- The CO methanation reaction was controlled by two different mechanisms.
- The fluidized bed showed more excellent carbon resistance than the fixed bed.

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ABSTRACT

The fluidization behavior and CO methanation performance over a typical Ni/Al_2O_3 catalyst for synthetic natural gas production was systematically investigated in a fluidized bed reactor. The results indicated that the methanation performance of the catalyst depends, to a large extent, on the fluidization quality in the fluidized-bed methanation reactor. The pure Ni/Al_2O_3 catalyst with the particle sizes of 10–100 μm failed to fluid, but the fluidization can be significantly improved by adding Al_2O_3 particles. The methanation reaction in the fluidized bed reactor was also substantially improved by adding Al_2O_3 particles, and the yield and selectivity of CH_4 in the fluidized bed with Al_2O_3 addition were much higher than those of without Al_2O_3 addition. The CO methanation reaction was controlled by two different mechanisms, i.e. the surface reaction controlling in lower temperature range (250–330 °C) and external diffusion controlling in higher temperature range (>330 °C). Stability test demonstrated that the fluidized bed methanation reactor showed higher CO conversion, CH_4 selectivity and yield, and more excellent carbon resistance than the fixed bed methanation reactor under high temperature, which is beneficial for reclaiming the high temperature steam.

1. Introduction

Conversion of coal and dry biomass to synthetic natural gas (SNG) via gasification and the subsequent methanation of the synthetic gas (syngas) have attracted much attention in recent years [1–3], due to the increasing demand of natural gas and the relative shortage of natural gas resources. Another important driver for investigation in methanation technology is the mitigation of carbon dioxide emissions.

Methanation of syngas ($CO + 3H_2 \rightarrow CH_4 + H_2O$), as the key process of SNG processes, is a highly exothermic reaction accompanied by a large decrease in mole number [4,5]. Removing the reaction heat has been one of the key problems of the methanation process for avoiding deactivation of catalysts due to sintering of metal particles, carbon deposition and sulpur poisoning induced by higher temperature sites [6–8].

Methanation reactor was one of the core technologies in methanation process [9]. Since 1950s, efforts have been made to develop methanation reactor systems, such as fixed bed reactors, fluidized bed reactors, slurry reactors [10–12]. The fluidization of catalyst particles leads to superior transfer and hydrodynamic characteristics which are important for reactions with significant thermal effect and involving catalyst regeneration. Kopyscinski et al. [1] described details of the fluidized methanation processes history, including the multiple-feed fluidized bed reactor by Greyson et al. and Schlesinger et al. [13,14], the Bi-Gas project initiated in 1963 by Bituminous Coal Research Inc. (BCR, United States) aiming at producing SNG from coal [15–17], the Comflux process with the concept of simultaneously water gas shift and methanation

* Corresponding authors. Tel.: +86 10 82544820; fax: +86 10 62536108.
E-mail addresses: junli@home.ipe.ac.cn (J. Li), qszhu@home.ipe.ac.cn (Q. Zhu).

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