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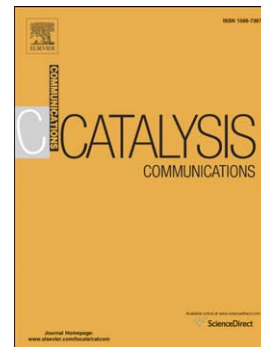
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# Hydrothermal synthesis of sulfur-resistant MoS<sub>2</sub> catalyst for methanation reaction

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## Abstract

The flower-like MoS<sub>2</sub> with high activity for sulfur-resistant methanation reaction was prepared by hydrothermal method. S/Mo mole ratios in feed and sulfidation condition during preparation were studied. The results show that higher S/Mo mole ratio increases the activity. Sulfidation in 3% H<sub>2</sub>S/H<sub>2</sub> results in structure with longer and more ordered multi-layered MoS<sub>2</sub> which increases the activity and stability.

*Keywords:* Hydrothermal method; MoS<sub>2</sub>; Sulfur-resistant catalyst; Methanation reaction; Sulfidation

## 1. Introduction

Since the crisis of environmental contamination and energy shortage, it has become increasingly important to use fossil resources efficiently and cleanly<sup>[1]</sup>. The development of synthetic natural gas (SNG) from syngas is a relatively reasonable and clean way to utilize coal<sup>[2]</sup>. Methanation is the key process for SNG production. Traditionally, Ni-based catalysts are adopted in the methanation reaction, in which they exhibit high CO conversions and good CH<sub>4</sub> selectivity from syngas<sup>[3]</sup>. However, sintering of the Ni particles and coke formation result in deactivation of Ni/Al<sub>2</sub>O<sub>3</sub>. These issues were addressed by the studies of Su. et al<sup>[4-6]</sup>, leading to the significant improvement for Ni-based catalysts. In addition, Ni-based catalysts are sensitive to poisons such as sulfur and its compounds. Therefore, pre-treatment for syngas, i.e. desulfurization is necessary and essential before methanation, which will also increase the cost of methanation from SNG production on Ni-based catalysts<sup>[7]</sup>.

In recent years, MoS<sub>2</sub>-based catalysts have been widely explored in hydrotreatment processes, such as hydrodesulfurization (HDS)<sup>[8-10]</sup>, water-gas-shift reaction<sup>[11]</sup> and sulfur-resistant methanation<sup>[12]</sup>, because of their high catalytic activity and sulfur resistance property.

Due to the sulfur resistance property, MoS<sub>2</sub> catalysts for methanation received widespread concern. Mo-based catalyst can be applied not only to lower H<sub>2</sub>/CO ratio, but also sulfur-contained atmosphere, which can eliminate both the water-gas-shift process and sulfur removal process, reducing the cost of methanation from syngas significantly.

Recently, preparation of Mo-based catalyst for sulfur-resistant methanation has been investigated such as sulfidation conditions<sup>[13]</sup>, stepwise sulfidation<sup>[14]</sup> et al for supported Mo-based catalyst. In addition to the sulfidation process, it was found that Al<sub>2</sub>O<sub>3</sub><sup>[15]</sup>, ZrO<sub>2</sub><sup>[16]</sup> and composite supports, such as CeO<sub>2</sub>-Al<sub>2</sub>O<sub>3</sub><sup>[17]</sup>, MgO-Al<sub>2</sub>O<sub>3</sub>, TiO<sub>2</sub>-Al<sub>2</sub>O<sub>3</sub> or ZrO<sub>2</sub>-Al<sub>2</sub>O<sub>3</sub><sup>[18]</sup> can function as supports. Also, promoters such as Co, Ni or La<sup>[19-20]</sup> and suitable precursor improve the performance<sup>[7]</sup> for methanation reaction. Jia Zhongbao et al studied Mo-Ni/ $\gamma$ -Al<sub>2</sub>O<sub>3</sub> and the effect of promoter Co and W<sup>[21]</sup>.

Few researches were focused on the unsupported MoS<sub>2</sub> catalysts, except the thermal decomposition with different Mo precursors<sup>[22-23]</sup>. In this work, we synthesized flower-like MoS<sub>2</sub> by hydrothermal synthesis method. Meanwhile, the performance of the catalysts was evaluated and the effects of sulfidation atmosphere have also been studied.

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