Paper ID: A402

Methane Production Over Ni Supported F-Sba-15: Different Amount ff Ni Loadings

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EXTENDED ABSTRACT

Formation of bulky wastes of carbon dioxide (CO_2) from burning of fossil fuels had impacted to the earth's environmental issues. The CO_2 gases trapped in the ozone layer and thus mitigate the climate change through the greenhouse gases (GHGs) effect. Numerous attentions have been drawn towards recycling and transformation of CO_2 gases into more valuable products via CO_2 methanation by using variety of supported metal catalysts [1–2]. For the choice of support, mesoporous material type of support such as SBA-15 is preferred due to its favorable textural properties with higher surface area (600-1,000 m^2/g), larger pore size diameter (5-30 nm), higher thermal and hydrothermal stability as well as highly uniform-arranged mesopores. Nickel (Ni) is selected as type of metal due to its low price and easily available [3]. However, if the carbon deposition and metal sintering occurred over metal-based support, it may rapidly trigger the deactivation of catalyst due to weaker interaction between metal and support [4], and thus resulted to a lower catalytic performance towards methane production. Therefore, this study highlighted on the modification structure of SBA-15 support into fibrous type (F-SBA-15) in order to produce higher accessibility of metal to be dispersed into it due to the formation of higher surface area and wide pore diameter, in agreement with Firmansyah et al. [5]. In addition, different amount of Ni loadings (1, 3, 5, and 10 wt.%) onto F-SBA-15 support for methane production were also controlled. Their physical properties were characterized using XRD, BET, and FTIR. In-situ FTIR adsorbed pyrrole analysis revealed the presence of basic sites originated in the catalysts. The catalytic activities of CO_2 methanation were performed using stainless steel fixed bed reactor. Meanwhile, the presence of coke on the surface of all spent Ni-based F-SBA-15 were investigated using XRD analysis.

As shown in Fig. 1, increment of Ni loadings (1-5 wt.%) resulted to an increase in catalytic performance, and slightly decrease at higher Ni loading (10 wt.%). 5%Ni/F-SBA-15 exhibited the most excellent catalytic performance (CO_2 conversion = 98.9 %, CH_4 selectivity = 99.6 % and CH_4 yield = 98.5 %) due to the favorable structure of catalyst, whereby numerous substitution of silanol groups with large quantity of Ni species resulted to a stronger metal-support interaction. However, a slight decrease in catalytic performance and stability of 10%Ni/F-SBA-15 might be related to the agglomeration of Ni particles, indicating the limitation in substitution of Ni species into the silanol groups of F-SBA-15 support upon the bulk Ni phase.

Fig. 2(A) shows the long-term stability test of different amount of Ni loadings onto F-SBA-15 catalysts at 673 K. It was observed that the most optimum and stable catalyst towards CO_2 methanation was exhibited by 5%Ni/F-SBA-15. This phenomenon might be due to the numerous substitution of silanol groups with large quantity of Ni species, thus resulted towards a better dispersion and stronger interaction between Ni and F-SBA-15 support. Apart from that, it might be inhibited metal sintering during the reaction and thus resulted in an excellent catalytic performance. For 10%Ni/F-SBA-15, a slightly reduction of catalytic stability of catalyst was observed, which closely related with the limitation substitution of silanol groups at higher amount of Ni loading, thus lowering Ni dispersion as well as weakening the metal-support interaction. The analysis of carbon formation and Ni sintering over the spent catalysts after 120 h reaction was investigated by XRD analysis and the results are shown in Fig. 2 (B). As shown in Figure 2(B), 5%Ni/F-SBA-15 possessed only a small amount of undesirable carbon formation, in agreement with the previous assumptions stated in above discussion. Therefore, it is reasonable to conclude that 5wt% is the optimal wt.% for Ni-F-SBA-15 and the influence of Ni loading is worth to be explored.

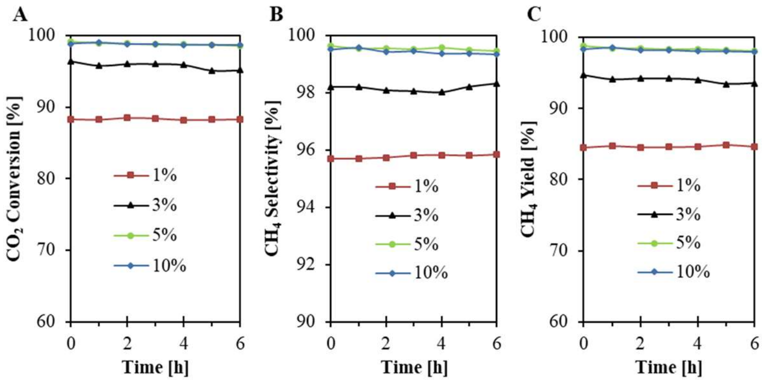

Figure 1: Catalytic performances of all different Ni loadings onto FSBA-15 support (1, 3, 5 and 10 wt.%) towards CO_2 methanation. Reaction conditions: T = 673 K, GHSV = 24,900 mLg-1h-1, H_2/CO_2 = 1/4, time-on-stream = 6 h.

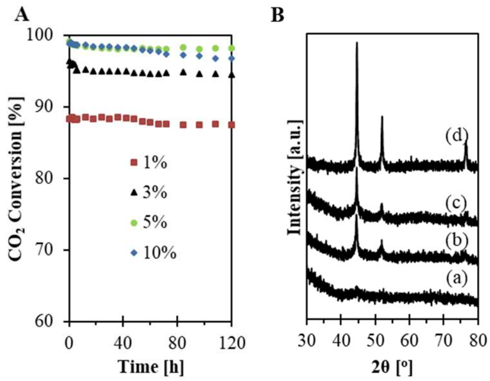

Fig. 2: (A) Long-term stability test of fresh (a) 1%Ni/F-SBA-15, (b) 3%Ni/F-SBA-15, (c) 5%Ni/F-SBA-15, and (d) 10%Ni/F-SBA-15 catalysts and (B) their characterization of each spent catalyst after reacted for 120 h time-on-stream at 673 K towards CO_2 methanation.

Keywords: Ni/F-SBA-15; Methane; Ni loadings; Metal-support interaction; Coke deposition.

Acknowledgment

This study was supported by Ministry of Education Malaysia through Research Acculturation Grant Scheme (RDU151414) and Fundamental Research Grant Scheme (RDU150126).

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