

ID: 1090

Effect of Preperation Method on Activity of Ni/Mg/Al Catalysts Prepared From Hydrotalcite Precursors in Dry Reforming of Methane

Baya Djebbari^{*1,2}, Nadia Aider^{1,3}, Fouzia Touahra⁴, V. M. Gonzalez-Delacruz⁵, Juan P. Holgado⁵, Alfonso Caballero, K. Bacharri³ and Djamila Halliche⁴

¹Laboratory of Natural Gas Chemistry, Faculty of Chemistry (USTHB), BP 32 16111 Algiers, Algeria

²Laboratory of Applied Chemistry and Materials (LabCAM), University of M'hamed Bougara of Boumerdes, Avenue de l'Indépendance Boumerdes, 35000. Algeria

³Centre de Recherche Scientifique et Technique en Analyses Physico-chimiques (CRAPC), BP 384-Bou-Ismaïl-RP42004, Tipaza, Algeria

⁴Laboratoire de Chimie Appliquée et de Génie Chimique (LCAGC), Université Mouloud Mammeri, 15000, Tizi-Ouzou, Algeria

⁵Instituto de Ciencia de Materials de Sevilla and Departamento de Química Inorganica, CSIC-University of Sevilla, Avda. Americo Vespucio, 49, 41092 Seville, Spain

*Corresponding author's email: correspondant.email.b.djebbari@univ-boumerdes.dz

ABSTRACT

The reaction of CO₂ methane reforming was tested over hydrotalcite-based catalysts containing nickel. In order to study the effect of Ni/Mg/Al synthesis methods. Ni/Mg/Al solids were prepared via three ways: (i) coprecipitation at basic pH of Ni²⁺, Mg²⁺ and Al³⁺ to obtain NiMgAl sample. (ii) MgAl-NiY sample prepared by Chelation-based techniques. (iii) The final way is common impregnation of Ni salts on MgAl hydrotalcite calcined at 450°C and was noted Ni/MgAl. X-ray diffraction (XRD), Brunauer-Emmett-Teller Method (BET), Temperature Programmed Reduction (TPR), Fourier Transform Infrared Spectroscopy (FTIR), Scanning Electron Microscopy (SEM) were used to describe completely the structural, morphological and surface characteristics of the solids. After reduction at 750°C for one hour, the catalysts were evaluated at the same temperature (750°C), in the dry reforming of methane reaction under continuous flow with CH₄/CO₂ ratio equal to 1, at atmospheric.

Keywords: energy, methane, hydrotalcite, nickel.

1. Introduction

Global warming is one of the major environmental challenges of our time. A major cause of the current global warming trend is the increase in the amount of greenhouse gas released to our environment (CH₄ and CO₂). Therefore, finding new ways to help tackle and/or reduce the level of greenhouse gas is of paramount importance, this could be achieved by either increasing the use of greenhouse gas (CO₂ and CH₄) and/or by reducing energy consumption. Catalytic dry reforming of methane (DRM) (also known as CO₂ reforming of methane) converts both CO₂ and CH₄ to syngas ($CH_4 + CO_2 \leftrightarrow 2CO + 2H_2$); the reaction has been proposed by many researchers as one of the possible solutions targeted at reducing the amount of CO₂ and CH₄ in our environment. The resulting intermediate is widely applied in the Fischer-Tropsch synthesis, [1- 3] to make other value-added hydrocarbons.

2. Experimental

Hydrotalcite-derived catalysts were prepared *via* co-precipitation method following procedure previously described. [4] NiMgAl sample catalyst was synthesized with molar ratio $(nNi^{2+} + nMg^{2+} / nAl^{3+}) = 2$. In order to study the effect of Ni position in within the NiMgAl hydrotalcite-derived catalysts structure (i.e., in the brucite layer or in the interlayer space of hydrotalcite), we embarked on the synthesis of MgAl-NiY sample using the co-precipitation method reported above with molar ratio $(nNi^{2+} / nAl^{3+}) = 2.4$. The synthesis of MgAl-NiY hydrotalcite-derived catalysts layered double hydroxide (LDH) included two steps. Following a procedure previously described [5].



3. Results and Discussion

The diffractograms of the NiMgAl and MgAl-NiY solids both show reflection peaks at $2\theta = 60^\circ$, with the same value of d_{110} (1.51 \AA). This suggests that no change in the cation-cation distance in the brucite-like layers has occurred. The lattice parameter calculation gave the value of 3.02 \AA which is in good agreement with the literature [6]. The value of parameter c of the MgAl-NiY solid is 44.97 \AA whereas it is approximately 22.14 \AA in the case of the solid NiMgAl. Indeed, the intercalation of $[\text{Ni}(\text{Y})_2]$ species between the sheets of brucite led to the extension of the spacing d_{003} and consequently an increase in the lattice parameter c .

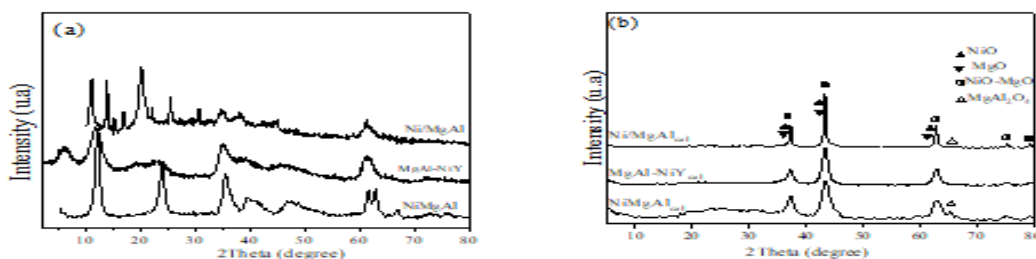


Figure 1: X-ray diffraction diagrams (XRD) (a) for synthesized precursors (b) Respective mixed oxides

The catalytic performance of the different catalysts for CO_2 reforming of methane were evaluated at a reaction temperature of 750°C using a 1/1 of CH_4/CO_2 feed ratio, after an “in-situ” H_2 reduction at 750°C for 1 hour. The main products from this reaction were H_2 and CO . The performances levels of the catalysts remain constant after 8 hours of reaction time, indicating that no or little deactivation took place within this period, at least under the conditions used. If we focus on the CH_4 conversion, we could say that catalysts NiMgAl (ca. 94%) and MgAl-NiY (ca. 78%), followed by Ni/MgAl (ca. 61%). The conversion for CO_2 shows a similar tendency: NiMgAl ca. 95.0%, MgAl-NiY (86.0%), Ni/MgAl ca. 76%. for the NiMgAl, presenting almost double conversion for CO_2 (90%) than for CH_4 .

H_2/CO ratios obtained at 750°C , in presence of Ni based-catalysts. This figure is a direct consequence of the differences in the conversion levels of CH_4 and CO_2 as can be seen, during the test (after 8 h on stream), the value of the H_2/CO ratio remains approximately constant for all the catalysts. For NiMgAl sample, the H_2/CO ratio is almost that corresponding to the thermodynamic equilibrium (ca. 1.0). On the other hand, MgAl-NiY and Ni/MgAl shows a H_2/CO ratio smaller than unity. The deviation from the stoichiometric ratio, the occurrence of the reverse water-gas shift reaction.

4. Conclusions

Different types of catalysts of the type containing Ni/Mg/Al have been prepared by thermal decomposition. The synthesized catalysts were successfully characterized and applied for syngas production from CO_2 reforming of methane at 750°C . The hydrotalcite structures were confirmed using XRD analysis, while the intercalation of Ni in the interlayer space was verified using both XRD and FTIR. The results also showed that the activity and stability of Ni-based catalysts were related to the Ni positioning; the best catalytic activity was observed with NiMgAl.

References

- [1] Centi, G., Perathoner, S. (2009). Opportunities and prospects in the chemical recycling of carbon dioxide to fuels. *Catalysis Today*, 148, 191-205.
- [2] Al-Fatesh, A. S., Naeem, M.A., Fakeeha, A. H., Abasaed, A. E. (2013). CO_2 Reforming of Methane to Produce Syngas over $\gamma\text{-Al}_2\text{O}_3$ -Supported Ni-Sr Catalysts. *Bulletin of the Chemical Society of Japan*, 86, 742-748.
- [3] Usman, M., Daud, W. W., Abbas, H. F. (2015). Dry reforming of methane: influence of process parameters- A Review Renew. *Renewable and Sustainable Energy Reviews*, 45, 710-744.
- [5] Touahra, F., Chebout, R., Lerari, D., Halliche, D., Bachari, K. (2019) Role of the nanoparticles of Cu-Co alloy derived from perovskite in dry reforming of methane. *Energy*, 171, 465-474.
- [6] Aider, N., Touahra, F., Bali, F., Djebarri, B., Lerari, D., Bachari, K., Halliche, D. (2018) Improvement of catalytic stability and carbon resistance in the process of CO_2 reforming of methane by CoAl and CoFe hydrotalcite-derived catalysts. *International Journal of Hydrogen Energy*, 43, 8256-8266.