Study of the partial substitution of Ni by Nb in perovskite based catalysts applied on partial oxidation of methane using XPD line

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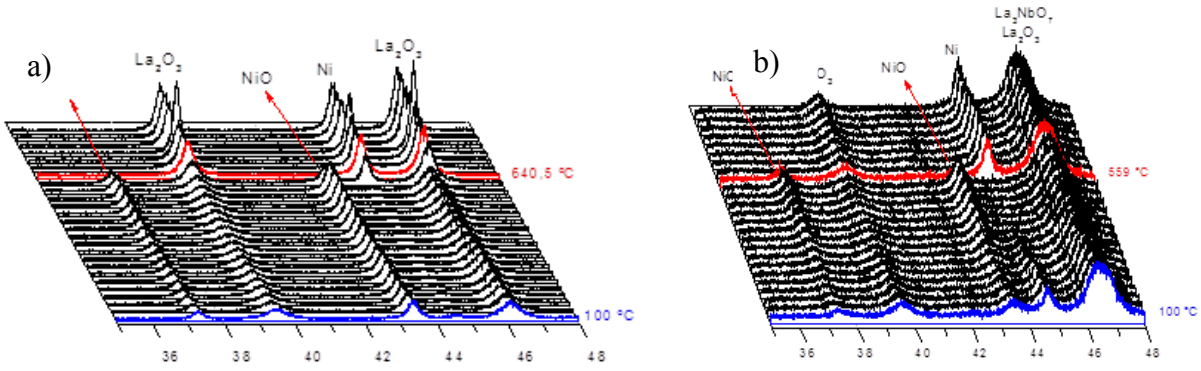
In this work perovskite $LaNiO_3$ and $LaNi_{0.5}Nb_{0.5}O_3$ were investigated as catalyst precursors in the partial oxidation of methane (POM). The objective was investigating the influence of the partial substitution of Ni by Nb in the catalytic performance. The catalysts were obtained by reduction of the perovskite precursors under H_2 flow (30mL/min) at 800 ºC. Analysis of the post reduction x-ray diffraction (XRD in situ) indicated the presence of Ni and La_2O_3 for the catalyst obtained from $LaNiO_3$ and Ni, La_2O_3 and La_3NbO_7 for that derived from $LaNi_{0.5}Nb_{0.5}O_3$. The catalysts were characterized by experiments XRD in situ performed at XPD line (LNLS) at high temperatures and under CH_4 and O_2 flows (2:1 CH_4/O_2 ratio). The diffractograms (Figure 1) indicated that is more difficult to oxidize Ni. Moreover nickel is regenerated at lower temperature in the catalyst containing Nb which was attributed to the presence of La_3NbO_7. Metal oxides such as NiO promote total oxidation of methane instead of partial oxidation producing CO_2 and H_2O [1]. Thus the lower susceptibility to Ni oxidation in the niobium-containing catalyst is quite relevant in the partial oxidation of methane. Tests performed with catalysts obtained from $LaNi_{0.5}Nb_{0.5}O_3$ during 20h at 750 ºC and CH_4/O_2 2:1, even containing approximately half amount of Ni than the catalyst derived from $LaNiO_3$, exhibited CH_4 conversion just 15% lower and ratio H_2/CO close to the



stoichiometric to POM. Based on these results it can be proposed that the partial substitution of Ni by Nb lead to a better resistance to oxidation of the Ni sites and which makes possible a H_2/CO ratio close to the stoichiometric in POM, which can be attributed to the presence of La_3NbO_7 phase.

[1] R. JIN et al. Applied Catalysis A: General 201, 71–80 (2000).

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