Transition metals over C-based materials: unraveling performances and challenges

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Significance and Relevance

In the pursuit of carbon neutrality, the conversion of CO_2 to methane or even to syngas will be a pivotal process. This study aims to examine the catalytic performance of cutting-edge carbon-based catalysts, as an alternative to conventional alumina-based ones, in CO_2 hydrogenation and dry reforming reactions. The findings will offer a comprehensive insight into the influence of the support and its impurities on the selectivity of different products.

Preferred and 2nd choice for the topic: Fundamental advances in understanding catalysis, Circular economy
Preferred presentation: *Oral*

Introduction and Motivations

The capture and utilization of CO_2 are regarded as a promising strategy for mitigating the environmental challenges confronting the globe and for achieving carbon neutrality by 2050[1]. In order to reduce CO_2 emissions, in particular those arising from the power sector, the use of hydrogenation reactions to produce CH_4 or CO through Sabatier's or reverse water gas shift (rWGS) reactions appears to be a promising strategy. Furthermore, the production of syngas through dry reforming represents another un-exploited reaction. In general, formulated metal supported catalysts suffer deactivation due to coke deposition and/or particle sintering[2-5], thus hindering the exploitation at the industrial scale. For these reasons, the development of new catalysts is a challenge. Among the potential options, carbon-based materials are well-suited as catalyst supports due to their low cost, natural abundance, high stability in both acidic and basic conditions, and high catalytic activity in several reactions[6-7]. In this study, carbon-based supports, including commercial graphene nanoplatelet (Gnp), homemade Gnp prepared via the molten salt method, and reduced graphene oxide, were employed as a support for Ni, Co, and/or Ru-based catalysts for CO_2 hydrogenation and dry reforming reactions.

Materials and Methods

The catalysts, with a total metal loading of 2 and 5 wt.% for Ni and Co-based ones and 0.5 wt.% for Ru-based ones, were supported on commercial (C) or homemade (H) graphene nanoplatelets (Gnp) and reduced graphene oxide (rGO). They were prepared by incipient wetness impregnation using ethanol as the solvent. The prepared materials were characterized by a range of techniques, including vibrational spectroscopies, XRD, FE-SEM, SEM-EDXS, and TEM. Prior to testing in Sabatier's, rWGS, and dry reforming reactions, an in-situ drying-reducing process was investigated and performed. A novel method for determining carbon loss during drying-reduction tests was developed and performed using online IR spectroscopy.

Results and Discussion

In Figure 1, CO_2 hydrogenation catalytic performances with the prepared catalysts are shown in terms of yield to methane and CO. At all temperatures investigated, the carbon-based catalysts exhibited a higher catalytic performance in rWGS reaction than Sabatier's one. It is noteworthy that the cobalt catalyst exhibited enhanced catalytic activity at low temperatures (T < 632 K), with a CO yield of 38% at 632 K. The Ni and Ru-based catalyst supported on homemade Gnp demonstrated

superior catalytic activity in terms of CO production compared to those supported on commercial ones. Conversely, catalysts supported on rGO demonstrated limited catalytic activity in both reactions.
The characterization of exhaust catalysts revealed partial graphitization of Gnp and the presence of distributed Ru- and Ni-based particles (with an average dimension of 5-10 and 10-25 nm, respectively), even though sulfur impurities (0.5 wt%) appears upon in-depth investigations.

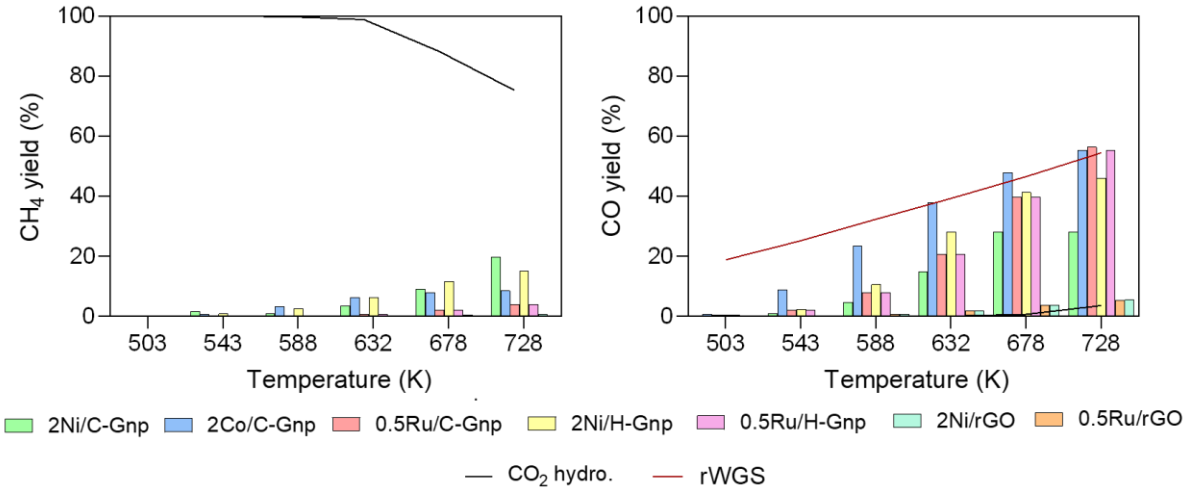

Figure 1. CO_2 hydrogenation catalytic performances with carbon-based catalysts

Co- and Ni-based catalysts showed promising results in dry reforming reaction obtaining a CO yield above 80% in the temperature range 773-973 K and at present time characterization of the exhaust materials is undergoing.

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