



Experiment title:

Detection and evolution of active site during catalytic production of H₂ (methane dry reforming and water gas shift reactions) over Mo_xC/HSAGs and RuMo_xC/HSAGs catalysts

Experiment number:

CH-4081

Beamline:

BM23

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18

Local contact(s):

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Report:

Mo and Ru-Mo catalysts supported on high surface area graphites (with values of surface areas in the 100-400 m²/g range), were studied by XAS during treatments consisting in the reduction of these metal components and their simultaneous carburization. Statistical factor analysis (PCA) applied to set of spectra has allowed following the chemical states both of Mo and Ru metal components during generation of carbide phases and provides crucial information concerning both the carbon-Mo/Ru interface and the resulting effects on the carbides phases. Thus, Mo initially presents as Mo⁶⁺ is reduced when increasing the temperature under a H₂/Ar mixture. Over 400°C gasification of C support takes place and the genesis of Mo_xC species starts. When using CH₄/H₂/He mixture the presence of methane promotes both the reduction process and the carburization of Mo.

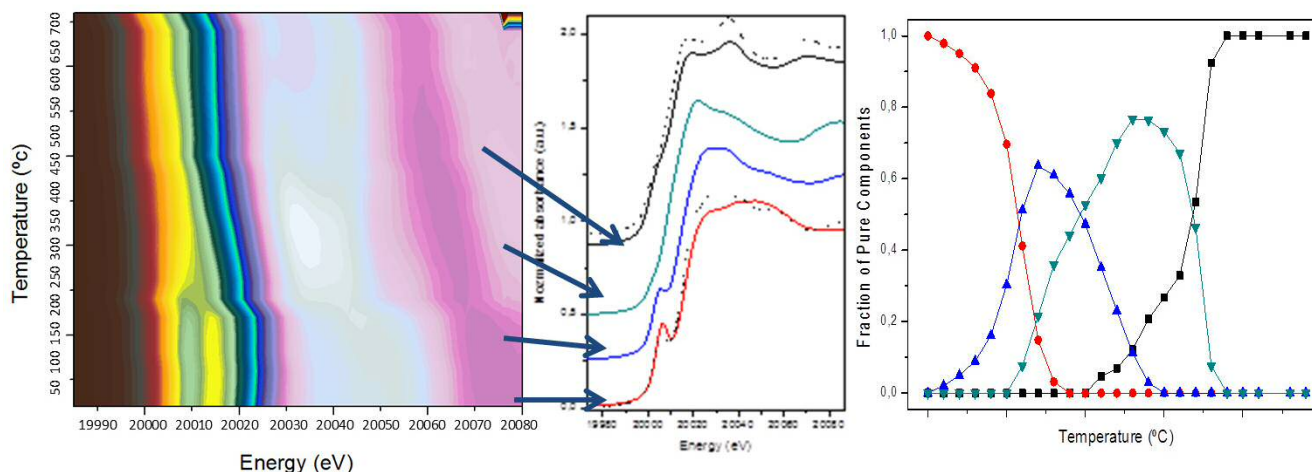


Fig.1. Temporal evolution of Mo species in Mo/C catalysts during carburization treatment (CH₄/H₂/He). Dash lines correspond to MoO₆ and Mo₂C references for initial and final species respectively.

In samples where Ru was added as promoter, the chemical states of Ru was also followed (Ru K-edge) in order to define the structural or electronic effects of this promoter and its role in the behaviour of Mo_xC species. Ru promotes Mo carbide phase and induces some structural modifications. On another hand, a Ru_xC is also detected. This last specie is not observed in Mo absence (Fig 2C).

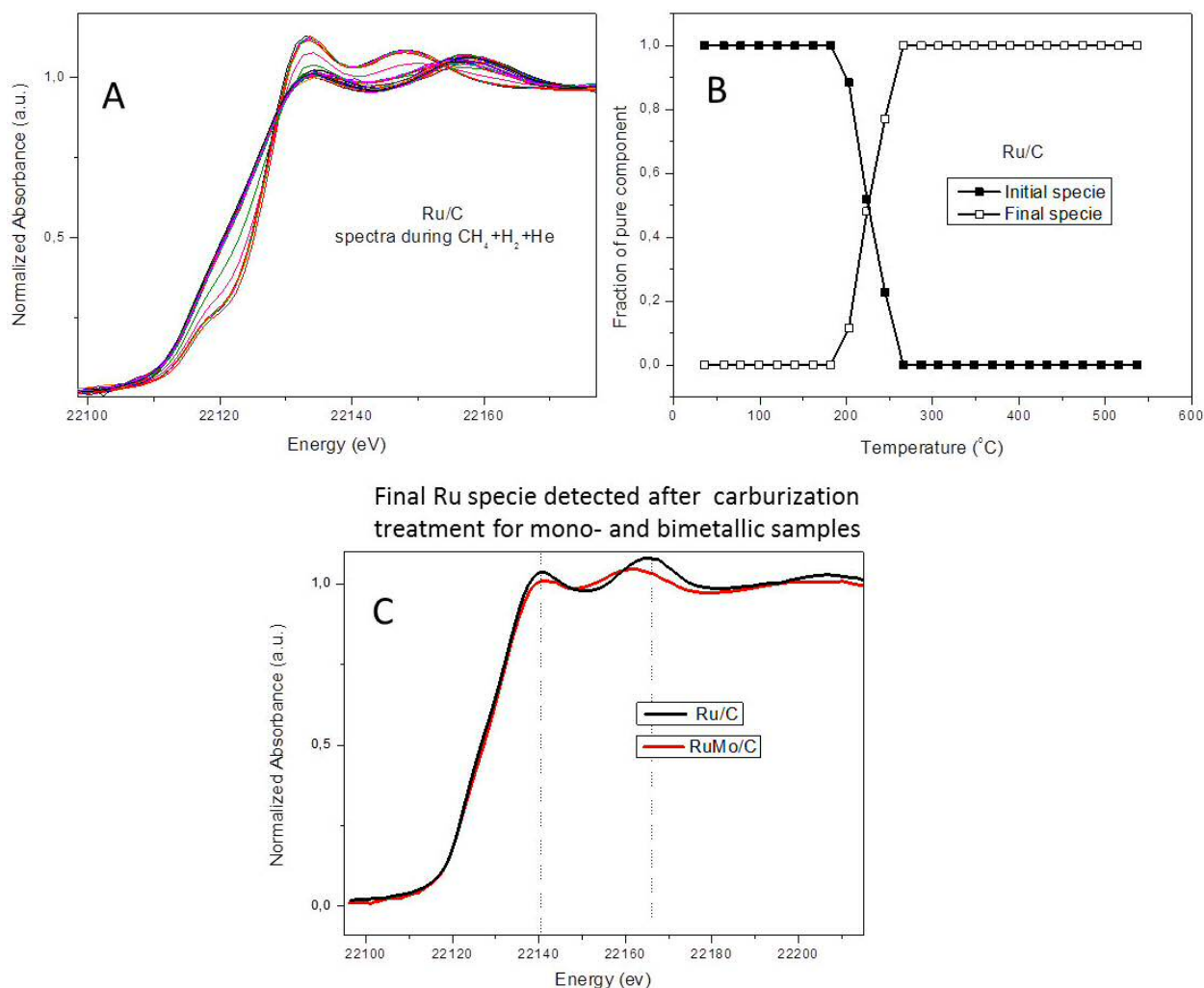


Fig.2. Ru K-edge XANES spectra (A) and concentration profiles (B) corresponding to pure chemical species (obtained by PCA) present during carburization treatment ($\text{CH}_4/\text{H}_2/\text{He}$) for Ru/C. (C) Comparison of final species for Ru and RuMo/C.

Therefore, the results show the synergetic action of the active species (Mo_xC) and the promoter (Ru). These catalysts exhibit an interesting behaviour in the dry reforming of methane (DRM) and water gas shift (WGS) reactions, both reactions of applied/industrial relevance because produce hydrogen with high selectivity.

Thus, we will try to correlate this extense structural and electronic characterization of the formed catalytic structures with the chemistry performance of these systems.

Finally, acknowledgment to Dr. S. Gatla for supporting the experiments carried out at BM23. We would also like to thank Dr G. Agostinni for the extensive help he gave during this measurement.