# Sujet de master M2 / Campagne 2019

## *$N_2O$ - a forgotten greenhouse gas - decomposition by Rh/CeZr catalysts*

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Nitrous oxide ($N_2O$) is considered as one of the most powerful greenhouse effect gases, almost 300 times higher than $CO_2$ molecules. It has been found that it contributed nearly 5% of total US greenhouse emissions (2015) [1]. $N_2O$ emissions, frequently observed in agricultural soil management as well as undesirable by-product of chemical processes e.g. after-treatment emission control catalysts (SCR, LNT, DOC, TWC…), have not been yet regulated by European Community but required attention. One of the frontiers of research highlights the importance of $NO_x$ catalyst for an efficient reduction of $N_2O$ but there exist alternative post-treatment approaches like the catalytic $N_2O$ decomposition. A few studies have proved that $Rh/Ce_xZr_{1-x}O_2$ (CeZr) is a very effective catalyst for $N_2O$ decomposition [2,3]: although both the metal and the support contribute to catalytic activity, the redox properties of ceria seem to be fundamental for this reaction. The correlation between substrate and reaction mechanisms are still not clear even if some studies showed that the improvement could be associated to the oxygen-deficient sites on ceria support [2].

This project aims to understand the role of both Rh and CeZr active sites in order to determine the reaction mechanism of $N_2O$ decomposition. Rh/CeZr catalysts were tested in $N_2O$ decomposition at several temperatures. Previous results confirmed that the Rh/CeZr catalyst is an exceptional catalyst $N_2O$ decomposition at high temperature and, even at low temperature still active. These results also confirmed that the active sites for $N_2O$ decomposition are not only placed on rhodium but also on ceria support. Further experiences, however, need to be performed to understand and verify the metal and support contribution of reaction mechanism. Thus, the *Rh/CeZr catalyst will be characterise by several in-situ* techniques, including *DRIFTS, isotopic exchange and Raman measurements*. Moreover, the *influence of different reaction conditions* (previous catalyst treatment, presence of water, lean conditions, etc.) *will be also studied* in order to optimize the overall process, in view of its further practical development. Finally, the *influence of a second metal*: Bimetallic catalysts (Rh-Cu, Rh-Mn, Rh-Ag…) *will be analyzed* in order to decrease catalyst costs and improve the catalyst properties for $N_2O$ decomposition.


[1] K. Larsen, J. Larsen, W. Herndon, S. Mohan, T. Houser, Taking Stock 2017: Adjusting Expectations for US GHG Emissions, Energy & Natural resources, May 24, 2017.

[2] K. Hashimoto, N. Toukai, R. Hamada, S. Imamura, Catal. Letters 50 (1988) 193-198.

[3] S. Imamura, R. Hamada, Y. Saito, K. Hashimoto, H. Jindai, J. Mol. Catal. A: Chem. 139 (1999) 55-62.





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