

Water Gas-Shift Reaction Catalysis by Ruthenium Based Complexes

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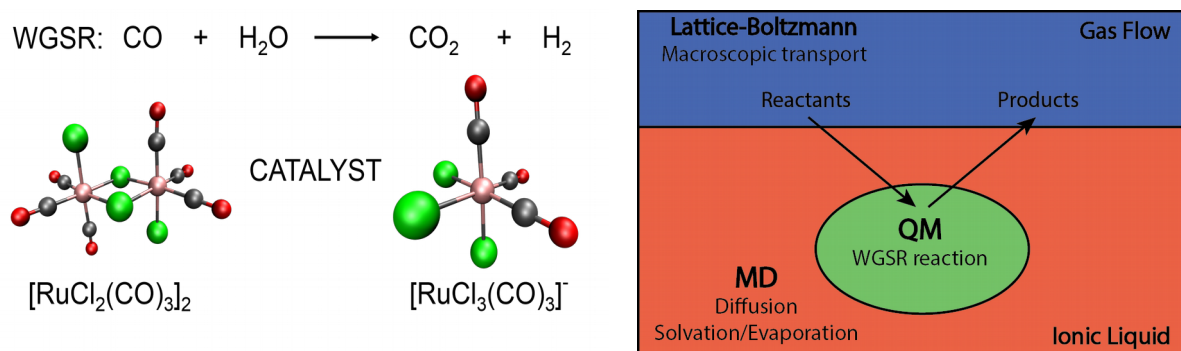
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The Water Gas-Shift reaction (WGSR) is a process which results in the conversion of carbon monoxide and water into hydrogen and carbon dioxide, usually in presence of a catalytic material. The abundance of water, the toxicity of carbon monoxide and the importance of hydrogen as potential fuel of the future are some of the arguments that incite the interest of the industrial and scientific communities in this particular reaction [1].

In this study we take a closer look into the WGSR catalyzed by a Ruthenium based catalyst ($[\text{RuCl}_2(\text{CO})_3]$) known for its high efficiency in the supported ionic liquid phase (SILP) [2, 3]. We tackle the problem of the unresolved mechanism for this specific catalyst and discuss monomer-dimer equilibrium and potential active species. To achieve this we use a plethora of well established quantum-chemical techniques ranging from optimizations and vibrational analysis to one-dimensional “coordinate driving” and multi-dimensional nudged elastic band searches. The theoretical framework for all of the calculations is density functional theory using the gradient corrected BP86 functional, which has proven to be successful for this class of transition metal complexes [4].

The final goal of this study is to identify the most likely mechanism involving this catalyst and to gain access to reaction rates which can then be refined using various approaches to account for the effect of the solvent. These rates, alongside the diffusion coefficients obtained in molecular dynamics simulations, can then be used in modeling macroscopic transport of the reactants and products. This would allow for prediction and optimization of input and output of modern SILP reactors.

[1] C. Ratnasamy, J. P. Wagner, *Cat. Rev.: Sci. Eng.*, **2009**, 51:3, 325-440

[2] S. Werner, N. Szesni, A. Bittermann, M. J. Schneider, P. Härter, M. Haumann, P. Wasserscheid, *Appl. Catal. A*, **2010**, 377, 70-75

[3] S. Werner, N. Szesni, M. Kaiser, R. W. Fischer, M. Haumann, P. Wasserscheid, *ChemCatChem*, **2010**, 2, 1399-1402

[4] M. Sobota, S. Schernich, H. Schulz, W. Hieringer, N. Paape, P. Wasserscheid, A. Görling, M. Laurin, J. Libuda, *Phys. Chem. Chem. Phys.*, **2012**, 14, 10603-10612