Computational study of rare-earth oxide catalysts

Understanding the catalytic performance of rare-earth oxides: Toward a knowledge-driven design of catalysts from first-principles calculations

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In Short

- Rare earth oxides (REOs) are promising catalysts
- Quantum-chemical treatment of REOs requires advanced electronic-structure methods and significant computing power
- Theoretical elucidation of reaction mechanisms on REO surfaces
- Design of reactivity descriptors

The full potential of rare earth oxides as versatile catalysts for various chemical transformations with great economic promise is still quite underexplored mainly because a detailed fundamental understanding of the catalytic properties of the REO series is lacking. The goal of the project is two-fold: (i) rationalize different catalytic function of the whole REO series through their first systematic quantum-chemical modelling at an atomic scale and (ii) use this knowledge to help design new catalysts for the oxidative coupling of methane (OCM), an economically highly attractive but not yet implemented in practice process for conversion of natural gas into value-added commodity chemicals [1].

The project will pursue an innovative route for knowledge-driven design of tunable catalysts on the basis of mixed oxides. Our research team aims at finding a highly efficient catalyst for OCM, which is a prerequisite for a major advancement in this field of research. REOs with their exceptionally high oxygen mobility and varying oxidizing capability within the REO series seem to be ideally suitable for this purpose.

Computational studies of REOs are scarce because these systems require the use of advanced electronic-structure methods and significant computing power [2]. Nowadays, with increased progress in computational chemistry, the availability of new computational tools, and with the growing expertise of the computational community in theoretical modelling of heterogeneous catalytic reactions, this complex matter is finally ready to be embarked on.

The detailed mechanism of OCM remains a matter of debate. Some of the main points of discussion regarding the mechanistic picture of OCM are summarized in Fig. 1.

Our theoretical study will be carried out in parallel with the activities of the collaborating experimental group at the same university (Prof. M. Bäumer). We will help catalyst developers by suggesting descriptors of the catalytic activity and by evaluating these descriptors in judiciously selected model systems before actual catalysts are prepared.

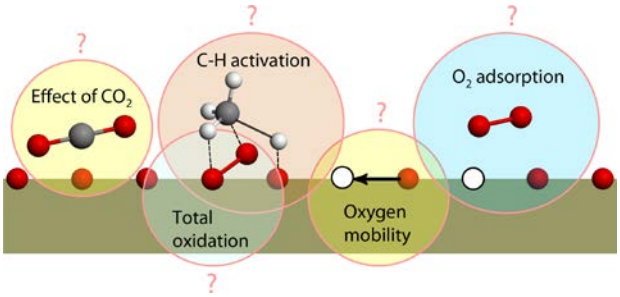

***Figure 1**. Critical questions associated with the hydrocarbon oxidation on REOs.*

Weitere Informationen

[1] Y. Amenomiya, V. I. Birss, M. Goledzinowski, J. Galuszka, A. R. Sanger, *Catal. Rev. – Sci. Eng.* **32**, 163 (1990).

[2] J. Paier, C. Penschke, J. Sauer, *Chem. Rev.* **113**, 3949 (2013).

Förderung

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