Research Project #26

Oxide Thin-Film Catalysts for Chemical Hydrogen Storage (INW-1 & PGI-7)

Efficient, scalable, and safe hydrogen storage remains one of the key challenges for a future hydrogen economy. Liquid Organic Hydrogen Carriers (LOHCs) such as Benzyltoluene or 1,4-Butanediol (BDO) represent an attractive solution due to their high storage density as well as compatibility with existing fuel infrastructure and safe handling characteristics. While there has been significant progress in catalyst design — including Cu-based systems and Pt/TiO_2 for selective LOHC hydrogenation–dehydrogenation [1][2], catalytic cycles for coupling hydrogen separation with storage [3], and metal-exsolution nanoparticle (NP) catalysts for enhanced thermal stability [4, 5]— we posit an urgent need for well-controlled model systems that allow *operando* insight into catalytic mechanisms toward rational materials and process design.

Atomically-defined epitaxial thin films provide unprecedented structural control, enabling the investigation of structure–function relationships at a level that cannot be achieved on powders. When combined with cutting-edge X-ray and electron scattering, spectroscopy, and imaging techniques at INW-1 and PGI-7, these systems offer a unique opportunity to probe catalytic interfaces, NP evolution, and support interactions across length and time scales and under *operando* conditions.

This PhD project will establish oxide thin films as structurally defined model catalysts for hydrogen-storage-relevant reactions, thereby bridging fundamental surface science with applied catalysis in the field of LOHC hydrogenation–dehydrogenation. The overarching goal is to establish epitaxial oxide thin films as a tunable, cross-scale model platform for thermocatalysis in hydrogen-storage reactions.

The PhD student will pursue the following scientific objectives:

Objective 1: Develop and characterize metal-exsolving perovskite thin films as model catalysts

- Grow epitaxial Ni-doped $SrTiO_3$ and related perovskites with controlled doping levels and surface terminations.
- Induce controlled metal exsolution to generate Ni (or alternative) NPs with well-defined size, density, and anchoring geometry.
- Study NP formation, stability, sintering, and re-exsolution under thermal and reactive conditions (BDO dehydrogenation, LOHC cycling).

Objective 2: Establish γ-Al_2O_3 and TiO_2 (anatase) epitaxial thin films as tunable model supports

- Grow γ-alumina thin films to probe acid sites, defect formation, and structure-dependent adsorption of LOHC reactants, intermediates, and products.
- Grow TiO_2 anatase epitaxial films to investigate strong metal–support interactions (SMSI), including Ti migration ("Ti-creep") onto Pt NPs [5].
- Evaluate how support structure influences catalyst activity, selectivity, and stability.

Objective 3: Determine adsorption geometries, surface structures, reaction pathways under *operando* conditions

- Resolve adsorption geometries of LOHCs and its key intermediates on oxide and metal–oxide surfaces.
- Investigate catalyst surface structure (e.g., NP facets, oxide reconstructions, defect states) during reaction cycles.
- Monitor NP sintering, agglomeration, and dynamic restructuring using *in-situ* and *operando* scattering/spectroscopy/microscopy.

The project combines advanced thin-film growth with cutting-edge X-ray and electron-based techniques to establish epitaxial oxide films as model systems for thermocatalysis. The PhD student will grow high-quality epitaxial γ-Al_2O_3, TiO_2 anatase, and exsolution-active perovskite films using pulsed laser deposition, creating single-crystal-like catalyst surfaces for studying LOHC hydrogenation/dehydrogenation reactions. Structural and morphological evolution will be probed using surface XRD, CTR, and XRR, while GISAXS/GIWAXS will track NP nucleation and sintering under reaction conditions. Chemical and electronic changes will be monitored with near-ambient pressure XPS, complemented by depth-resolved XAS (TEY/TFY) and nanoscale-resolved XPEEM to capture lateral heterogeneity and metal-support interactions.

Catalytic behavior will be assessed through temperature-programmed and controlled hydrogenation/dehydrogenation experiments, enabling direct correlation of structural dynamics with catalytic function. The project will deliver a tunable model platform capable of revealing fundamental mechanisms in hydrogen-storage-relevant catalysts, offering insight into interface restructuring, defect chemistry, nanoparticle evolution, and reactions such as selective BDO dehydrogenation and reversible LOHC cycling. These results will inform rational catalyst design for hydrogen separation, storage, and release, with relevance across thermocatalysis and industrial hydrogen technologies.

The PhD student will work within the combined expertise of INW-1 and PGI-7 at Forschungszentrum Jülich: INW-1 provides leading *operando* X-ray capabilities and catalytic interface science, while PGI-7 offers world-class epitaxial thin-film growth infrastructure, including the Electronic Oxide Cluster for integrated PLD–XPS–XPEEM studies. Additional collaborations with ESRF and PETRA III as well as LOHC and exsolution research groups ensure a highly interdisciplinary environment. The student will receive comprehensive training in thin-film growth, advanced scattering, spectroscopy and imaging, catalysis, and data analysis, forming a strong foundation for a career in energy materials or hydrogen technologies.

Location of the HITEC Fellow	Forschungszentrum Jülich, Institute for a Sustainable Hydrogen Economy - Catalytic Interfaces for Chemical Hydrogen Storage (INW-1), Director: Prof. Dr. Hans-Georg Steinrück https://www.fz-juelich.de/de/inw/unsere-bereiche/inw-1
Partners of the HITEC Project	Forschungszentrum Jülich, Peter Grünberg Institute – Electronic Materials (PGI-7), Director: Prof. Dr. Regina Dittmann https://www.fz-juelich.de/en/pgi/pgi-7
Specific **requirements**	M.Sc. in Physics, Materials Science, or Chemistry. Experience with structural characterization methods — such as scattering, spectroscopy, or imaging using neutrons, X-rays, or electrons — is highly desirable. Proficiency in data analysis, particularly with Python, is also an advantage.
For **project specific** questions please **contact**	Dr. Peter Walter, INW-1, p.walter@fz-juelich.de Dr. Felix Gunkel, PGI-7, f.gunkel@fz-juelich.de

[1] Zhang, S. et al. Selectively Regulated Dehydrogenation of 1,4-Butanediol to γ-Butyrolactone via Tailoring the Structure of Copper Based Catalysts. Catal Lett 155, 240 (2025).
[2] Bong, B. et al. Hydrogen Loading and Release Potential of the LOHC System Benzyltoluene/Perhydro Benzyltoluene over S–Pt/TiO2 Catalyst. ACS Eng. Au 4, 359–367 (2024).
[3] Chen, Y. et al. A catalytic cycle that enables crude hydrogen separation, storage and transportation. Nat Energy https://doi.org/10.1038/s41560-025-01806-9 (2025)
[4] Jeong, H. et al. Emerging Exsolution Materials for Diverse Energy Applications: Design, Mechanism, and Future Prospects. Chem. Mater. 35, 3745–3764 (2023).
[5] Carrillo, A. J., López-García, A., Delgado-Galicia, B. & Serra, J. M. New trends in nanoparticle exsolution. Chem. Commun. 60, 7987–8007 (2024).