Priority Programme

“Material Synthesis near Room Temperature”

Project Description – Project Proposal

Reducible oxide materials: knowledge-driven design of novel low-temperature synthesis routes

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Summary of proposal

Reducible oxides are key components in future technologies. The low-temperature synthesis in ionic liquids (ILs) holds the potential to synthesize such materials via simple and efficient routes and with novel tailor-made properties. Towards a knowledge-based design of such synthesis routes, however, we urgently require an in-depth understanding of the interfacial chemistry that controls synthesis and materials properties. The project aims at providing this molecular-level understanding. We follow a model approach that combines surface science and spectroscopy in ultrahigh vacuum (UHV) with studies under realistic conditions and on real materials. To this end, we use a broad spectrum of state-of-the-art experimental methods. In this project, we scrutinize the synthesis of ceria-based materials in imidazolium-based ILs. We cover all steps of materials synthesis, from hydrolysis, via nucleation, growth and aggregation of nanoparticles, to the final and critical step of solvent removal. We study the elementary steps of precursor conversion using ultraclean IL films in UHV, identifying intermediates and exploring their conversion kinetics. Using time-resolved in-situ spectroscopy, we will then investigate the interfacial chemistry of nucleation and particle growth. These studies will provide detailed insight into growth mechanisms and the processes that control composition, impurities, stoichiometry, structure, and particle shape. Making use of a newly available toolbox of well-defined ceria model surfaces, we will follow the chemistry at the IL/ceria interface. Adsorption, desorption, surface reactions, and

decomposition mechanisms will be studied in dependence of the surface structure, with the aim to control or suppress the formation of residuals and dopants. Finally, we will transfer the knowledge from the UHV studies to realistic conditions and real materials, i.e. we will bridge the pressure and materials gap between surface science and applied material synthesis. Here, high-resolution microscopy and chemical analysis of real nanoparticles will allow correlating the interfacial chemistry with the chemical, structural, and morphological properties of the synthesized materials. Based on a molecular-level understanding of the interfacial chemistry, we will make use of the unique chemical flexibility of ILs to design novel routes to tailor-made materials. Combining appropriate reaction conditions and ILs, we will aim at controlling and minimizing the influence of contaminants. New synthesis strategies will be explored, using ILs that are thermally converted to volatile products for easy solvent removal. Finally, we aim at integrating precursors directly into these self-volatizing IL materials. Particle synthesis from these new ILs may provide as efficient as simple low-temperature pathways to ultra-clean, doped, or mixed oxide materials.