

Two-step metal oxide thermochemical redox has been receiving growing attention in the scientific community over recent years. **They have often been studied in the context of converting heat (typically from concentrated solar power) to chemical energy, which can be further used for the following application: energy storage, renewable fuel production (water and carbon dioxide splitting process for liquid hydrocarbon fuels), air separation, and oxygen pump.**

The development of all of the aforementioned applications depends on the selection of appropriate redox materials for the cycle, which needs to be optimized according to the specific application. Although the ideal choice may vary in each case, they share similarities in terms of redox thermodynamics and beneficial material properties. Therefore, the selection of the correct redox cycle in each case is subject to similar limitations.

Thermodynamics plays a crucial role and should be the primary consideration when attempting to optimize materials or determine conditions for two-step thermochemical redox cycles. If the thermodynamics show promise, the application may still encounter issues such as kinetic constraints or material deterioration. Conversely, if the thermodynamics is not favourable, the practical implementation will certainly be limited by this factor. The wide range of scientific research indicates also that oxides which preserve a complete phase transition during reduction, possess significantly higher specific energy storage compared to materials that undergo partial reduction. On the other hand, partial reduction materials generally exhibit faster kinetics and greater activity at lower temperatures. **Therefore, the selection of materials for different applications often involves a trade-off between the significance of high specific energy storage and the advantages of fast kinetics and operation at low temperatures.**

In this context, our objective is to provide comprehensive knowledge regarding the general mechanism and environmental factors governing thermochemical redox cycles on the exemplary materials i.e. manganese oxide-based catalysts. The project aims at explaining comprehensively the role and the mechanism of redox processes, with particular emphasis on *(i) the temperature-induced reduction of the catalyst (ii) the catalyst's behaviour in variable redox environments, and, (iii) the catalyst's behaviour in variable redox environments after doping the catalyst with elements modelling their redox properties.*

The first objective will allow for a preliminary understanding of the redox behaviour of catalyst during the thermochemical redox cycles. The second objective will explain the role of catalyst redox properties, atomic structures, and morphology, with an emphasis on the role of various oxygen species and vacancies that can be engaged in the thermochemical redox cycles. Finally, the third objective, (iii), by proper selection of dopants, will allow for the design of the manganese oxide-based catalyst with the redox properties tuned towards enhancing efficiency in energy storage, or water and carbon dioxide splitting processes. The research project itself is divided into four tasks. Tasks 1-3 are focused on an in-depth analysis of the inherent morphological, structural, and electronic properties of model systems and a description of behaviour in various redox conditions of model catalytic systems. Task 4 is focused on applying and expanding this knowledge to doped catalytic model systems. The chosen manganese oxides meet all the requirements for catalytic materials and show the required activity in the thermochemical redox process. Their structural and redox versatility, together with a well-defined morphology, provide unique opportunities to study structure-catalytic properties relationships in redox cycles at the molecular level.