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***In situ* environmental transmission electron microscopy investigation of core-shell supported co-catalyst system for optimized visible-light water splitting**

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As a result of diminishing fossil fuel reserves, there is an increasing need to switch energy dependence to renewable resources such as sunlight. Photocatalysts provide a viable route for converting solar energy into chemical bonds. In order to optimize the performance of such materials, a fundamental understanding of their reaction mechanisms, chemical behavior, structure and morphology before, during and after reaction using *in situ* investigations is necessary. Here, we focus on the *in situ* characterization of photocatalysts [1] in an environmental transmission electron microscope (ETEM) [2,3]. Such fundamental insight can be used for further material optimization with respect to performance and stability [4].

The activity of photocatalysts for water splitting under visible light exposure can be enhanced by adding co-catalysts for the hydrogen evolution reaction (HER) on their surface, Pt being one of the most commonly used. Pt is a reversible catalyst for HER, meaning that unless precautions are taken it will also efficiently catalyze the reverse reaction, resulting in a loss of net activity. The addition of a shell on top of the co-catalyst has proven an efficient method to dramatically reduce the reverse reaction in both catalytic [5] and photocatalytic systems [6]. Encapsulated Pt is obtained by substrate reduction in H₂ and is a novel promising system for enhanced water splitting using visible light.

In this work the encapsulation of supported co-catalysts on a number of semiconductor substrates including SrTiO₃ and TiO₂ is studied using ETEM techniques (Fig.1). The shell formation mechanism is studied *in situ* as a function of reaction conditions such as temperature, pressure, time, and atmosphere composition. Catalytic efficiency of the encapsulated samples is tested in parallel using micro-reactors [7] allowing for direct correlation of reaction conditions and catalyst performance.

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Figure 1. Bright field TEM images of STO-supported Pt nanoparticles before (left) and after (right) high-temperature in situ reduction in H_2 .