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Electrocatalysts for Sustainable Hydrogen Energy

Disordered and Heterogeneous Nanomaterials

Joakim Ekspong

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Fakultetsopponent: Prof. Tomas Edvinsson,
Institutionen for materialvetenskap, Uppsala Universitet,
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Author
Joakim Ekspong

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Abstract
With the current global greenhouse gas emissions, our remaining carbon budget is depleted in only 7 years. After that, several biophysical systems are predicted to collapse such as the arctic ice, coral reefs and the permafrost, leading to potentially irreversible consequences. Our emissions are strongly correlated to access of energy and even if we are aware of the planetary emergency today, our emissions still continue to grow. Electrical vehicles have the possibility to reduce the emissions in the transportation sector significantly. However, these vehicles are still expensive and impractical for long-distance or heavy transportation. While political actions and technological development are essential to keep prices down, the driving distance can be increased by replacing the batteries for onboard electricity production.

In hydrogen fuel cells, electricity is produced by combining hydrogen gas (H_2) and oxygen with only water as the by-product and if employed in electrical vehicles, distances of 500 km are enabled with a refueling time in 5 minutes. For other uses than in vehicles, H_2 is also promising for large-scale electricity storage and for several industrial processes such as manufacturing CO_2-free steel, ammonia and synthetic fuels. However, today most H_2 production methods relies on fossil fuels and releases huge amounts of CO_2.
Electrolysis of water is an alternative production method where H_2, along with oxygen are produced from water. To split the water, electricity has to be added and if renewable energy sources are used, the method has zero emissions and is considered most promising for a sustainable hydrogen energy economy. The technique is relatively expensive compared to the fossil fuel-based methods and relies on rare noble metals such as platinum as catalysts for decreasing the required energy to split water. For large scale productions, these metals need to be replaced by more sustainable and abundant catalysts to lower the cost and minimize the environmental impacts.

In this thesis we have investigated such candidates for the water splitting reaction but also to some extent for the oxygen reduction reaction in fuel cells. By combining theory and experiments we hope to aid in the development and facilitate a transition to clean hydrogen energy. We find among other things that i) defects in catalytic materials plays a significant role the performance and efficiency, and that ii) heterogeneity influence the adsorption energies of reaction intermediates and hence the catalytic efficiency and iii) while defects are not often studied for electrocatalytic reactions, these may inspire for novel materials in the future.

Keywords
Water splitting, electrolysis, electrochemistry, hydrogen evolution reaction, oxygen evolution reaction, fuel cells, hydrogen production, nanomaterials, density functional theory.

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