SGVC Innovation Price 2022: Master Thesis

Abstract

The ability of carbon to generate highly performing metal-based catalysts for acetylene hydrochlorination, a key industrial process for production of vinyl chloride (VCM), has long been recognized and made it the support of choice for this reaction. Nevetheless, despite the superior performance of carbon-supported systems, the origin of its unique suitability for this application remains unclear. Accordingly, this project aims to investigate the role of this carrier in determining the activity and stability of the hydrochlorination catalysts. To this end, a platform of platinum single atoms hosted on carbons with varying porous, structural and chemical properties is derived. Micropore volume and pore accessibility of the support are revealed to control the acetylene adsorption capacity, which in turn governs the catalytic activity. This observation is further validated for gold- and ruthenium-based systems. The density of acidic oxygen functionalities on the support surface, responsible for the formation of coke deposits, is found to be the descriptor of catalyst stability. At the same time, the presence of oxygen-containing groups also has a beneficial role, aiding in the support's ability to adsorb acetylene, providing anchoring sites for stabilization of isolated gold and platinum atoms, and boosting the initial activity of ruthenium-based catalysts. This highlights the importance of the optimal carbon design depending on the nature of the metal. With this understanding, a platinum single-atom catalyst is developed, exhibiting unparalleled stability over 50 h on stream at a higher productivity rate than the previously reported benchmark system. These findings mark a step forward in the understanding of the role of carbon in acetylene hydrochlorination and provide guidelines for rational catalyst design, both for sustainable VCM production and for other applications, where the inherent properties of the carrier affect the catalytic performance of the system.