

Heterogenization of molecular catalysts in layered hydroxide host materials towards CO₂ electrochemical reduction application

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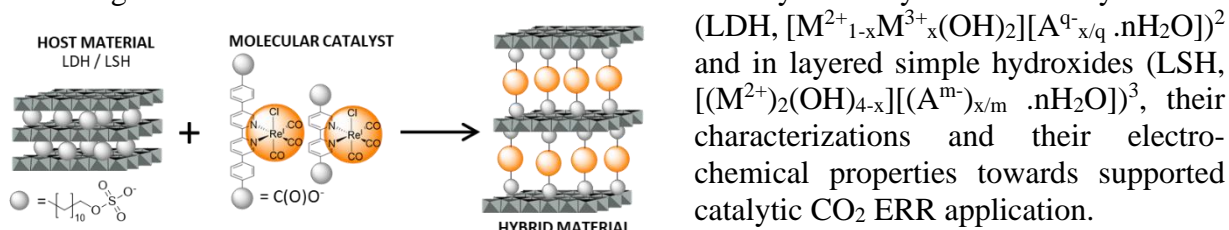
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The reduction of CO₂ is a domain of intense activity with the aim to use CO₂ as a source of carbon for fuels or, more broadly, for commodity molecules. Efforts are thus made to develop selective catalysts for CO₂ electroreduction reaction (CO₂ERR). In that endeavour, molecular catalysts have an advantage which is the fine control of their structure and properties. Molecular catalysts are usually first studied as homogeneous catalysts to evaluate their activity however, to move toward applications, it is often better to design devices in which the molecular catalyst is heterogenized on electrode surface. Among the various strategies to immobilize molecular catalysts, chemical bonding to electrode conductive materials presents some advantages because of the robustness of the linkage. When the catalyst is a transition metal complex, the anchoring group has to be connected to the complex through the ligands, and an important question is thus raised on the influence of this function on the redox and on the catalytic properties of the complex.

We illustrate this issue through an investigation of the effect of the chemical anchoring groups used to immobilize molecular catalysts on host materials for supported CO₂ electroreduction.¹ For rhenium bipyridyl carbonyl complexes, we shown that if the bipyridine ligand is directly linked with an anchoring function, such as carboxylate, there is a drastic effect on the catalytic activity of the corresponding Re complex toward CO₂ERR in solution. In that case, the reducing equivalents are mainly accumulated on the ligand and prevent the formation of Re(0) to interact with CO₂. Alternatively, phenyl-carboxylate substituents on the bpy ligand preserves the catalytic activity of the Re complex. We present preliminary results with hybrid materials resulting from immobilization of such Re-molecular catalysts in layered double hydroxides



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References

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