

Pyridine vs imidazole axial ligation on cobaloxime grafted graphene: Hydrogen evolution reaction insights

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Cobaloximes have been protagonists in the (photo)electrocatalytic H₂ production research scene for a while now, owing to their redox-active metal center and their closely lying polar oximes coordinated in a square-planar geometry. However, there have been insights that the outer coordination sphere of the cobaloxime catalytic center e.g. the axial and equatorial ligation, drastically affects the hydrogen evolution reaction (HER) performance,[1] being a worth-studying topic appealing to researchers engaged in catalyst design. Aspiring to bring this study in a hydrogen fuel cell realistic set-up, we envisioned a rationally designed heterogeneous electrocatalytic system, compatible with practical aqueous acidic conditions, in order to study the HER performance of two twin electrocatalysts with sole difference their axial ligation to the cobaloxime complex. Covalent functionalization of graphene was selected as an ideal route to ensure the required stability for cobaloxime under such conditions, while pyridine and imidazole functionalities were selected as the axial ligands, based on their versatility, occurrence and different electron-donating ability. Interestingly, while pyridine axial ligation mirrors a drastically superior electrocatalytic performance, imidazole exhibits a remarkable long-term stability.[2]

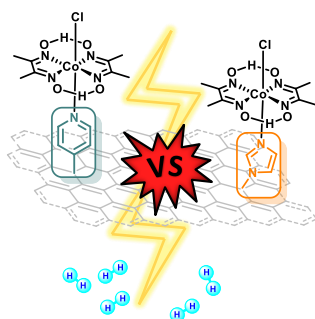


Figure 1: Pyridine vs imidazole axial ligation of a cobaloxime complex covalently grafted on graphene and their effect on hydrogen evolution reaction (HER) performance

References

[1] A. Panagiotopoulos, K. Ladomenou, D. Sun, V. Artero and A. G. Coutsolelos, Dalton Trans. **45**, 6732 (2016).

[2] I. K. Sideri, G. Charalambidis, A. G. Coutsolelos, R. Arenal and N. Tagmatarchis, Submitted (2022).

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