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Design of high entropy oxide catalysts for oxidative coupling of methane

Aim

Design of high entropy oxide (HEO) catalysts with high selectivity for methane oxidative coupling (OCM) to ethene via a combination of computational methods and experimental characterizations.

Justification

The catalytic OCM to C_2 hydrocarbons (C_2H_4 and C_2H_6) with oxygen (O_2-OCM) is an interesting route to upgrade C1 molecules to C2 building blocks. The main challenge for this reaction is the low C_2 selectivity at industrially relevant methane conversions. This is because CH_4 and the C_2 products are easily overoxidized to CO_x products, i.e., CO and CO_2. Consequently, catalysts that selectively produce C_2 hydrocarbons instead of CO_2 and CO are highly desired. Various types of oxide-based catalysts have been developed, and important contributions to this field have been made at the LCT[1-3].

CoNiCuMgZnO HEOs will be considered in this project. HEOs are variation on the more established High Entropy Alloys, which are a stable solid solution of at least 5 metals. We postulate that the multi-elemental synergy in HEOs provides a distribution of active adsorption sites, some with optimal affinity for methane activation and other for C-C coupling and water formation. We choose CoNiCuMgZnO HEOs because this type of HEOs has been synthesized and shown promising activity for oxidation reactions[4, 5]. Promising HEO catalysts will be searched via density functional theory (DFT) calculations combined with a machine learning mapping algorithms (**Figure 1**). Promising HEOs from the computational work can be synthesized and tested using the LCT's state-of-the-art reactor infrastructure.

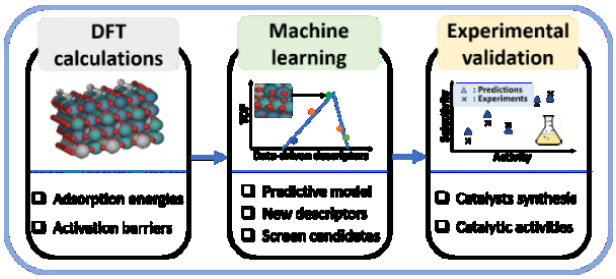

Figure 1 Workflow for finding active HEO catalysts for the O2-OCM

Program

1. Literature survey about high entropy oxides.
2. Model CoNiCuMgZnO-type HEOs with random compositions, screen stable HEOs, and calculate binding energies of intermediates.
3. Build machine learning models using chemical and physical properties of HEOs as descriptors.
4. Predict active and selective CoNiCuMgZnO HEOs with specific atomic configurations.

References

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[3] E. H. Ismailov, D. B. Taghiyev, et al., *Theor. Exp. Chem.*, **2022**, 58, 61-69.
[4] D. Feng, Y. Dong, et al., *Angew. Chem.*, **2020**, 59, 19503-19509.
[5] M. Fracchia, P. Ghigna, et al., *J. Phys. Chem. Lett.*, **2020**, 11, 3589-3593.

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