

The quest for correlations



Correlating structure and activity is a very important research goal in catalysis. This Editorial reflects on this topic, taking inspiration from examples in the current issue.

As with many areas of physical science, catalysis is deeply involved in establishing correlations. Deciphering the interplay between structure and activity represents a crucial endeavour not only to deepen the understanding of fundamental physical principles, but also to inform the development of more effective catalytic systems and support the transfer of technologies from the lab into industry. Many approaches to establish correlations start by developing qualitative insights. The Sabatier principle is one such example: it links the strength of interaction of a reactant to a catalytic material to the reactivity of the latter, highlighting the importance of balancing reactant adsorption and product desorption for catalysis to occur.

Qualitative knowledge can be further refined and eventually may adopt mathematical formulations that impact the study of reaction mechanisms and our ability to make quantitative predictions about the outcome of a reaction. Correlations can be empirical as in the case of the Hammett equation, which was developed to formalize the relationship between reaction rates and equilibrium constants for compounds containing substituted phenyl groups and turned out to be a powerful tool in physical organic chemistry¹. Others can be derived from theoretical frameworks as in the case of the Eyring equation², which links reaction rates and temperature, and together with its empirical counterpart – the Arrhenius equation – belongs to the indispensable toolbox for the study of reaction kinetics.

In the case of complex catalytic systems, such as supported nanoparticulate catalysts, establishing the interdependence between specific structural features and reactivity

may be particularly challenging and requires a high level of control of the synthesis of the composite material. In this context, the [Article](#) by Aizenberg and co-authors featured in our current issue showcases how to leverage colloidal synthesis approaches to disentangle the effect of nanoparticle size and proximity in thermal catalysis. Thanks to the ability to control loading and placement of particles with narrow size distributions, the study demonstrates how interparticle distance can affect the selectivity of benzaldehyde hydrogenation on PdAu alloys.

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While traditional structure–activity studies have been based on trial-and-error approaches, it is important to highlight how recent advances in modelling and data science have prompted researchers to explore the relationships existing between reactivity and specific physico-chemical properties of the catalytic system³. The resulting reactivity descriptors – such as the electronic states of metals or the adsorption energies of reactants – are extremely powerful in predicting reactivity trends and therefore very useful in the context of catalyst design. Against this background, we published an [Article](#) by

Haoxiang Xu, Daojian Cheng, Dapeng Cao and Xiao Cheng Zeng in 2018 that introduced a descriptor to predict the electrocatalytic activity of single-atom catalysts for the hydrogen evolution, oxygen evolution and oxygen reduction reactions using a combination of accessible structural and electronic features. Although the study received much attention in the years following publication, the editorial office was alerted in 2022 that the original dataset of adsorption energies contained erroneous results, as some of the structural models used to obtain such adsorption energies were incorrect⁴. After informing the journal’s readership through an Editor’s Note we have corresponded with the authors, requesting that they clarify the issues. An in-depth investigation followed and the emerging analysis – resulting in the development of a modified ϕ' descriptor – has undergone scrutiny by external independent experts. At the end of this complex process, the authors decided to [retract the article](#) as the revised ϕ' descriptor turns out to be not as universal as the original one intended to be, impacting the original message of the work. At the same time, the current issue of *Nature Catalysis* also features an [Article](#) detailing the properties of the revised ϕ' descriptor – based on the correct dataset. This Article preserves some of the features and the design principle of the initial descriptor, which have been a significant source of inspiration to the single-atom catalysis field. We are convinced that this represents the best course of action to correct the scientific record while retaining the legacy of the initial investigation as the quest for correlations goes on.

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References

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