Heterogeneous catalysis in space: CH_3OH formation via Fischer-Tropsch reactions. A computational approach.

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Gas-phase chemistry under extreme conditions, characterized by low densities and temperatures, poses significant challenges. The presence of interstellar grains plays a crucial role in facilitating the formation of molecules that cannot naturally emerge in the gaseous phase. Interstellar grains are believed to enhance the interaction between reactive species, promoting reactions on their surfaces, and mitigate the energy excess of highly exothermic reactions. However, their function as chemical catalysts, offering low activation energy pathways to accelerate reaction rates, remains less explored.

Nevertheless, various materials with catalytic properties can be found in interstellar environments, including refractory grains containing transition metals abundant in space. In this study, we present novel mechanistic insights into the synthesis of methanol (CH_3OH) via the Fischer-Tropsch process under astrophysical conditions. We employ single-atom iron (Fe)-containing silica surfaces as heterogeneous catalysts in an interstellar context. Our approach involves quantum chemical calculations considering extended periodic surfaces to identify stationary points and transition states, ultimately constructing reaction potential energy surfaces. Additionally, we conduct binding energy and kinetic calculations based on the Rice–Ramsperger–Kassel–Marcus (RRKM) scheme to assess the catalytic capability of these grains and place these reaction processes within the framework of astrochemistry.

Our mechanistic investigations demonstrate the feasibility of astrocatalysis in astrophysical environments. While the proposed process is thermodynamically highly exergonic, it exhibits kinetic barriers necessitating an activation energy to proceed. Furthermore, our kinetic calculations underscore the strong temperature dependence of this reaction process, with tunnelling effects being insignificant across the involved energetic barriers. These findings shed light on the presence of CH_3OH in various regions where current models struggle to explain its observed abundance. Moreover, the revelation of astrocatalysis introduces an entirely new spectrum of synthetic pathways, sparking chemical evolution in space.

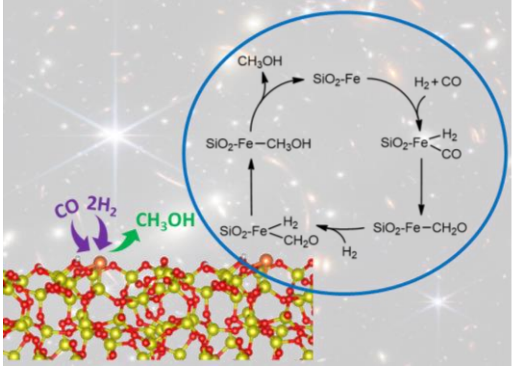

Figure 1. Representation of the surface model and the catalytic cycle under study.