Molecular simulation of Methanol Valorisation to Sustainable Aviation Fuels over Zeolites.

Alessandro Morittu

Abstract:
Aviation is crucial for global transportation, connecting people and goods across vast distances. However, its environmental impact, particularly its carbon footprint, has become a significant concern in the face of climate change. As the sector moves towards decarbonization, sustainable aviation fuel (SAF) represents a key solution to achieve net-zero emissions; in fact, SAF can reduce emissions by up to 80%, leading to a substantial decrease in the carbon intensity of aviation.
Light olefins, such as ethylene and propylene, are fundamental building blocks in organic synthesis and are extensively used in the petrochemical industry. They also show promise for generating sustainable aviation fuels, making it essential to research viable production routes. Olefins can be obtained by converting methanol over acidic zeolites through the methanol-to-olefin process. However, this catalytic process is quite complex due to the various intermediate reactions that constitute the overall process that have not been fully understood yet. Because of the importance of clarifying these processes, this project aims to achieve a systematic understanding of the catalytic mechanisms involved in the MTO process over acidic zeolite catalysts, focusing on its application in sustainable aviation fuel production. Molecular simulations will be employed to gain mechanistic insights with atomistic detail at operando conditions and to determine the thermodynamic and kinetic parameters. The thermodynamic and kinetic parameters, such as free energy and reaction rates, will be obtained through the synergy of three techniques: molecular dynamics, enhanced sampling and machine learning potentials.
The project is divided into several phases to be addressed throughout the entire PhD course. In the first year, a significant amount of time will be dedicated to a literature review of methanol-to-hydrocarbons, methanol-to-aromatics, and sustainable aviation fuel production over zeolites to clarify the processes involved. Following this, the development of a simulation model will commence, which includes the preparation of the surface model and the calculation of acidic zeolite structures. Once the model is ready, the remainder of the first year will focus on understanding the mechanism of dimethyl ether to olefins and dimethyl ether to methanol processes. The first part of the second year will be dedicated to preparing and executing metadynamics simulations of methanol conversion to hydrocarbons and aromatics. These simulations will be analyzed to determine the free energies and reaction rates of the considered processes, providing a complete picture of the global reaction, including both thermodynamic and kinetic data. The last part of the year will involve repeating the simulation processes in various known zeolites to create a library that contains the catalytic performances for each structure analysed.
In the third year a machine learning model will be built and trained with the results obtained from the investigated zeolites reported in the library previously built, with the aim to correlate the material structure with reactivity, aiming to select a better performing zeolite, from known and hypothetical zeolites, that guarantees maximal catalytic performance.