Minicolloque n° CPR3 **Oral**

Atomistic modeling of the formation of dynamic molecular polymers

Romain Dupuis[a,b*], Guillaume Maurin[a], et Mihail Barboiu[b]

a. ICGM, 8 Rue de l'École Normale, 34090 Montpellier
b. IEM, 300 Avenue du Professeur Emile Jeanbrau, 34090 Montpellier
* email : romain.dupuis@umontpellier.fr

Capturing CO_2 is a major societal issue. Recently synthesized covalent molecular dynamic polymers (dynamers) show promising performances for the selective adsorption of CO2 towards different gas molecules[1]. These polymers formed by self-assembly of bi- and tri-functionalized macromonomers by amine groups and connector agents carrying aldehyde functions, are dynamic due to the reversibility of their covalent bonds, which gives them a unique adaptive character. The free enthalpy of reaction to form a bond within these dynamers is of the order of a few K_bT, the association kinetics of the molecules is therefore governed by entropy and steric hindrance. The mechanisms of formation of this family of polymers and their structural properties remain unknown to this day due to their complexity and their evolutionary nature. To allow us to describe structural models of these assemblies for the first time, we have developed a molecular simulation method integrating quantum and force-field approaches that make it possible to account for the chemical reactivity of these systems, a crucial step in studying the formation of these assemblies in solution.

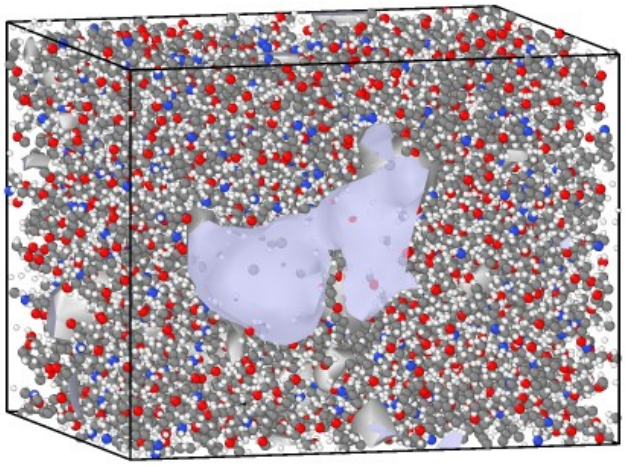

Figure 1 : example of a porous dynamer (the pores are shown in transparent blue)

During this presentation, I will outline the method that we have developed as well as the results it achieves. It is a method of molecular dynamics coupled with a Monte Carlo. The energy surface of the system is determined by quantum calculations to determine the probabilities of binding and dissociation of the dynamers. I will detail its interest in relation to recent developments of other methods, carried out with the aim of reproducing the formation of solids or colloids in solution[2], in which I have participated. These models allow us to study the formation of porosity and compare it with experimental data. On the other hand, the constructed atomic model makes it possible to simulate gas adsorption isotherms essential for understanding the mechanisms of selectivity and capture. Dynamers are of particular interest because the dynamic aspect could be exploited to control porosity and selectivity of adsorption during membrane use.

[1] M. Barboiu, Constitutional Dynameric Networks for Membranes, Membrane Materials, Characterization, and Module Design, 2013, https://doi.org/10.1002/9781118522318.emst054

[2] R. Dupuis et al. Molecular simulation of silica gels: Formation, dilution, and drying, Physical Review Materials, 2019, https://doi.org/10.1103/PhysRevMaterials.3.075603