Optimizing PET biorecycling solutions through electronically polarizable simulations

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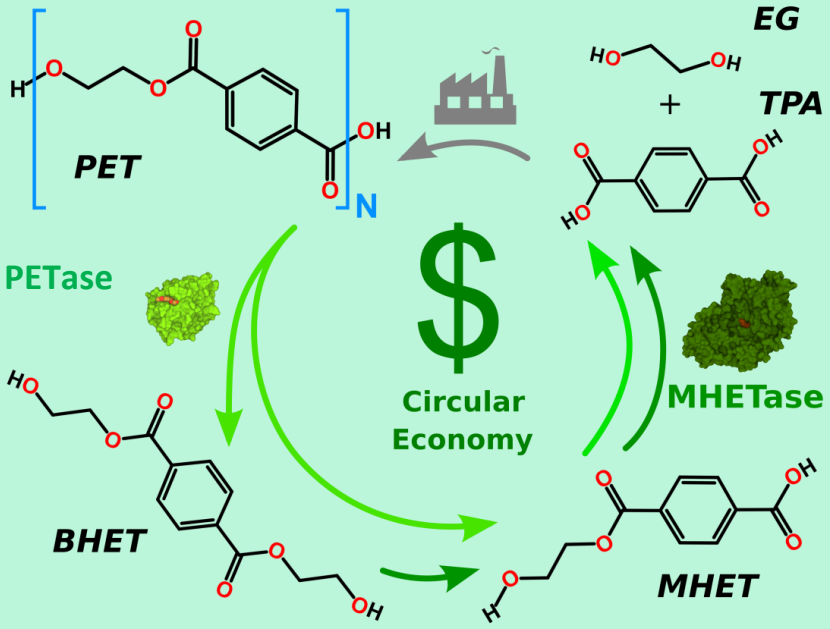
Department of Biotechnology
University of São Paulo (USP)



Oct 8th, 2024

Health & Environmental Threat

Ever increasing accumulation of PET as **microplastic** in the environment is considered a major problem to mankind.

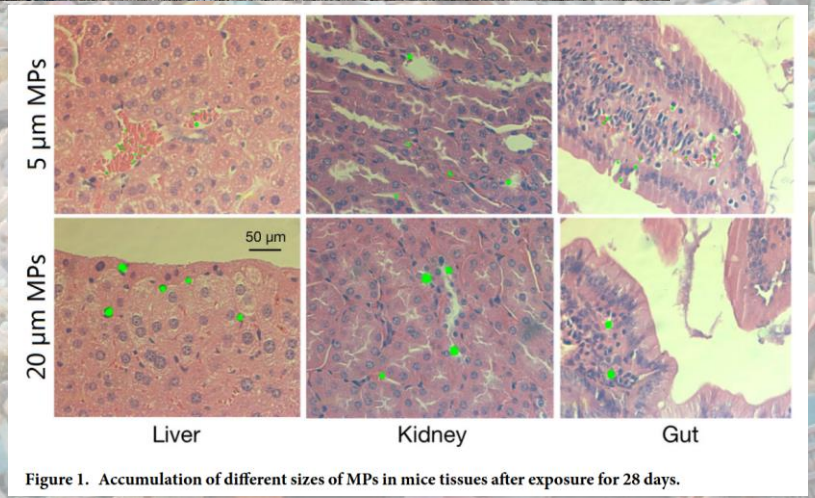
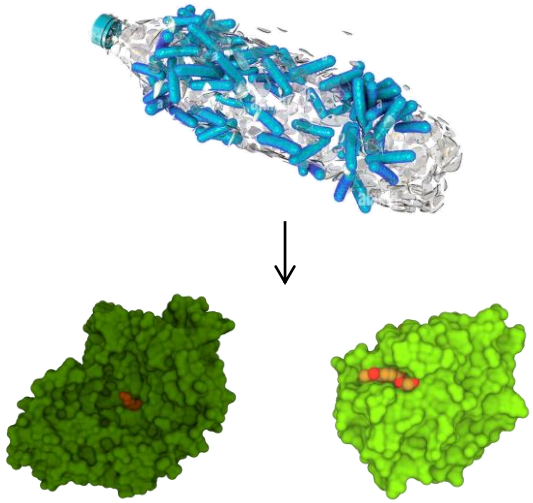


Figure 1. Accumulation of different sizes of MPs in mice tissues after exposure for 28 days.

*Deng, Y., Zhang, Y., Lemos, B. et al. Tissue accumulation of microplastics in mice and biomarker responses suggest widespread health risks of exposure. Sci Rep **7**, 46687 (2017)*

PET-degrading enzymes

I. sakaiensis 201-F6



MHETase **PETase**

Yoshida et. al. (2016). A bacterium that degrades and assimilates poly(ethylene terephthalate). Science 351 (6278), 1196-1199

PET-degrading enzymes

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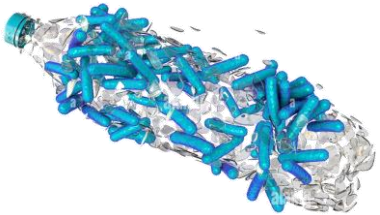
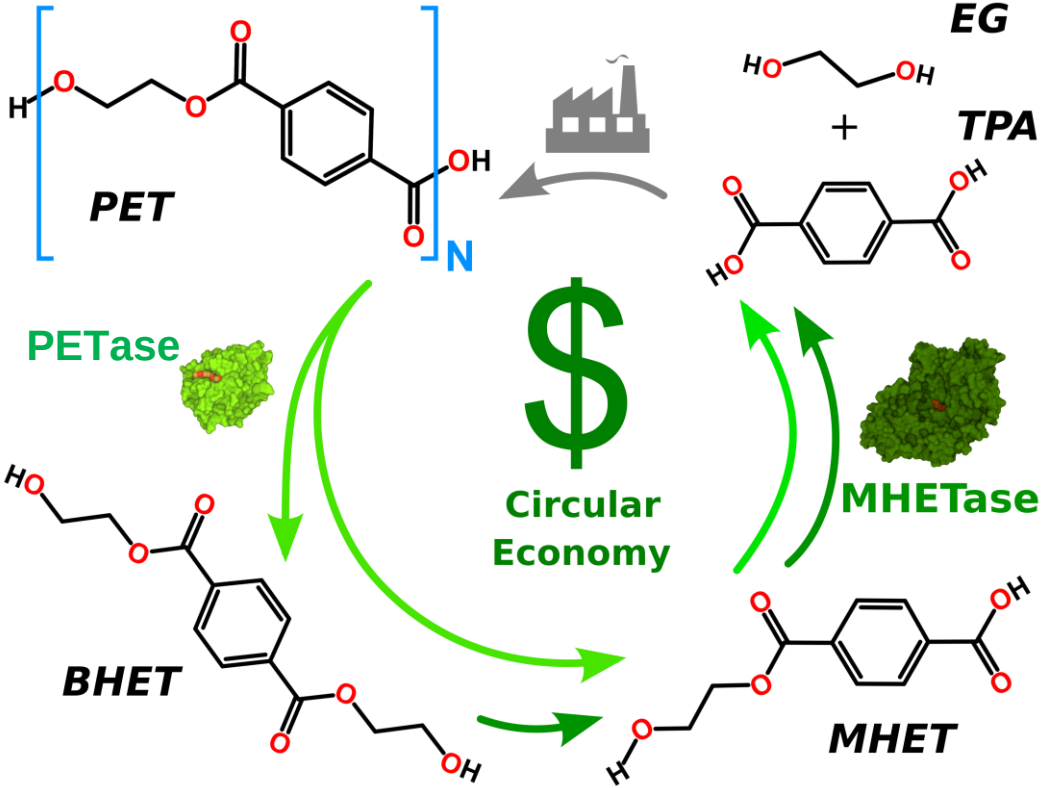
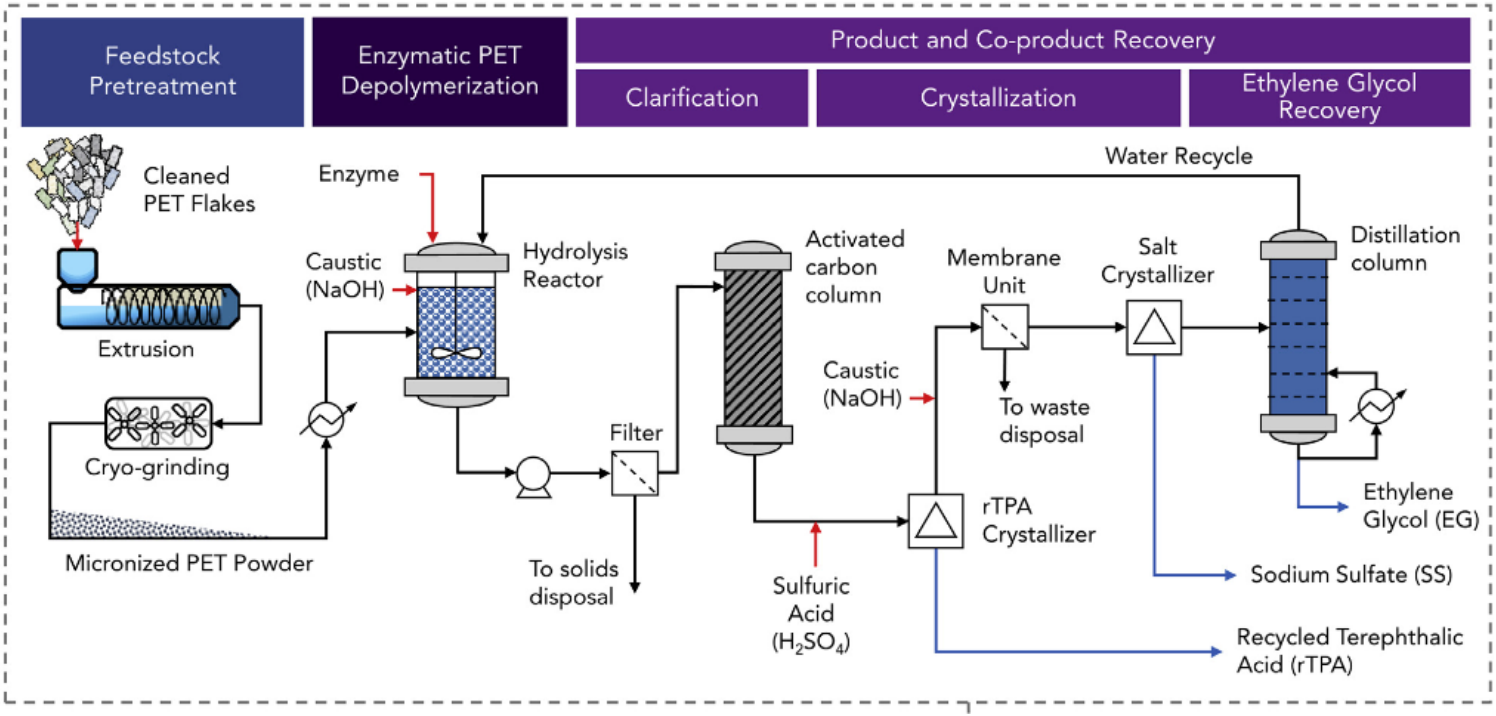


MHETase PETase



Yoshida et. al. (2016). A bacterium that degrades and assimilates poly(ethylene terephthalate). Science 351 (6278), 1196-1199

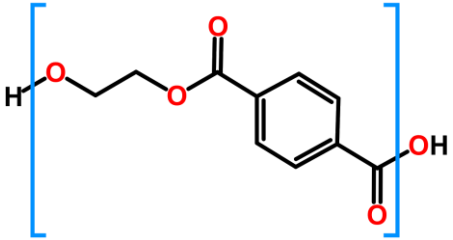
What are the challenges for industrial applications?



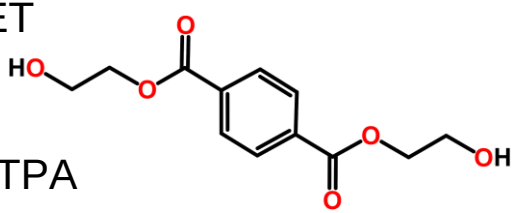
Singh et. al. (2021). Techno-economic, life-cycle, and socioeconomic impact analysis of enzymatic recycling of poly(ethylene terephthalate). Joule 5(9), 2479-2503

Drude Polarizable PET polymer model

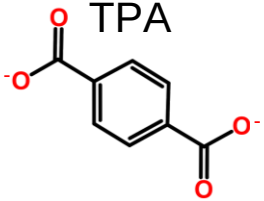
PET



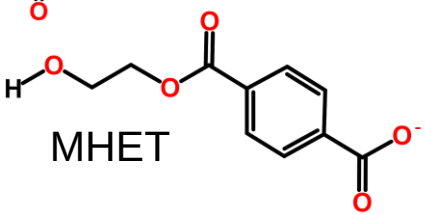
BHET

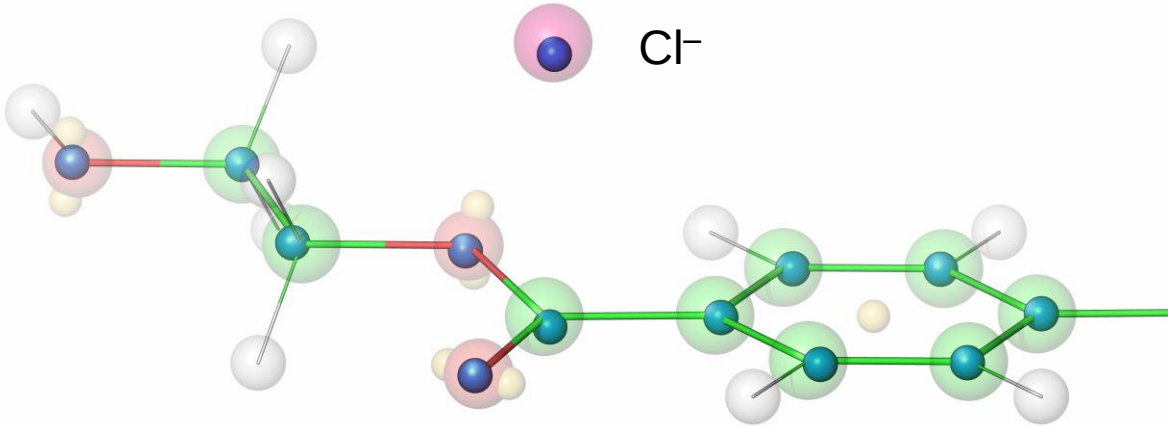


TPA



MHET

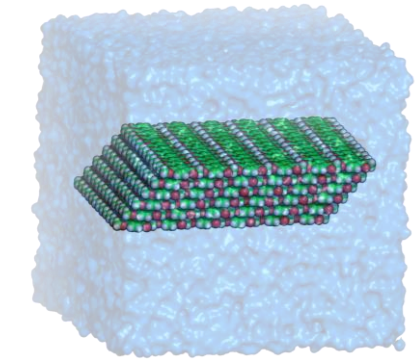


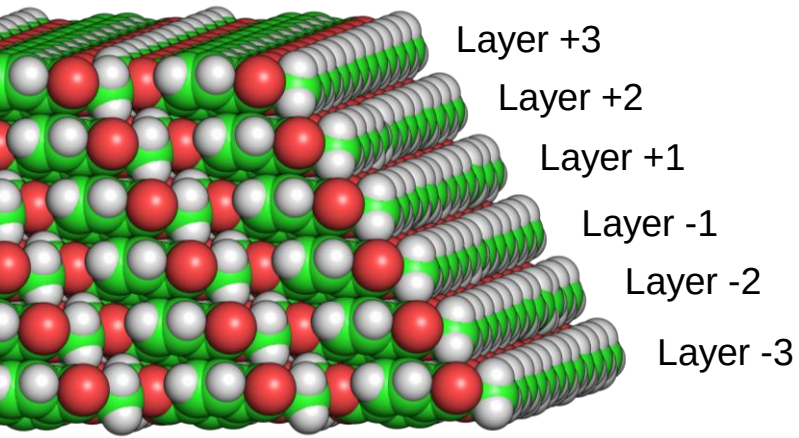


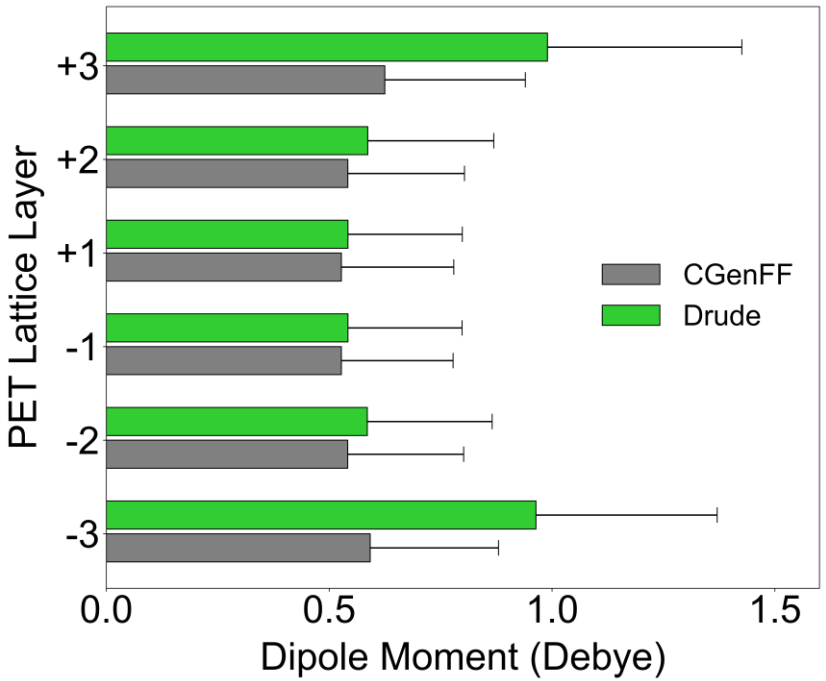
PET monomer
(BHET)

Polêto, M.D., Lemkul, J.A. Structural and electronic properties of polyethylene terephthalate (PET) from polarizable molecular dynamics simulations. ChemRxiv (2024)

01 - Electronic properties of crystal PET







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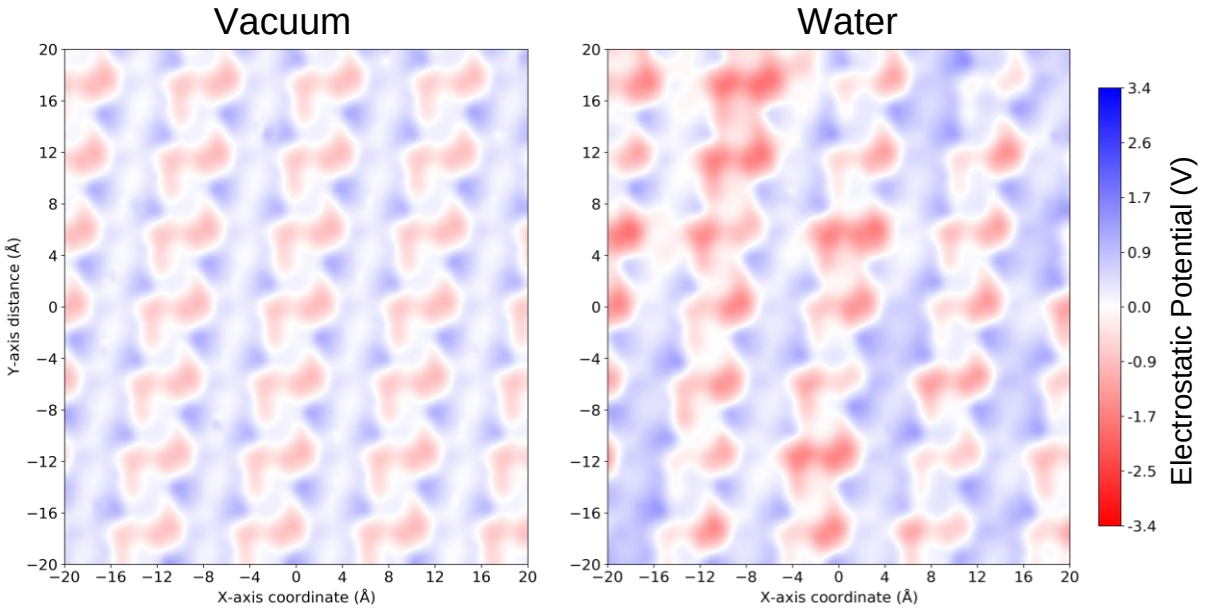

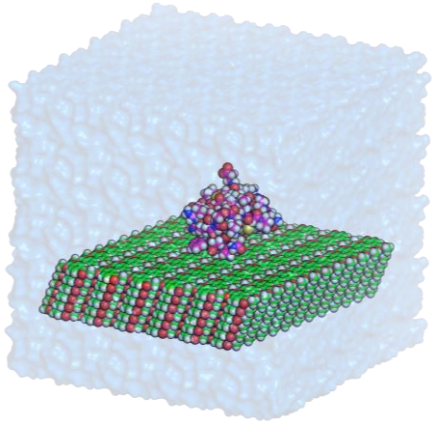

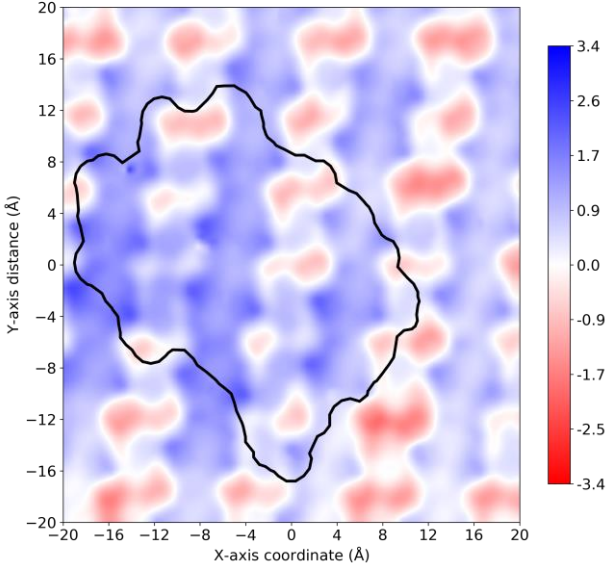

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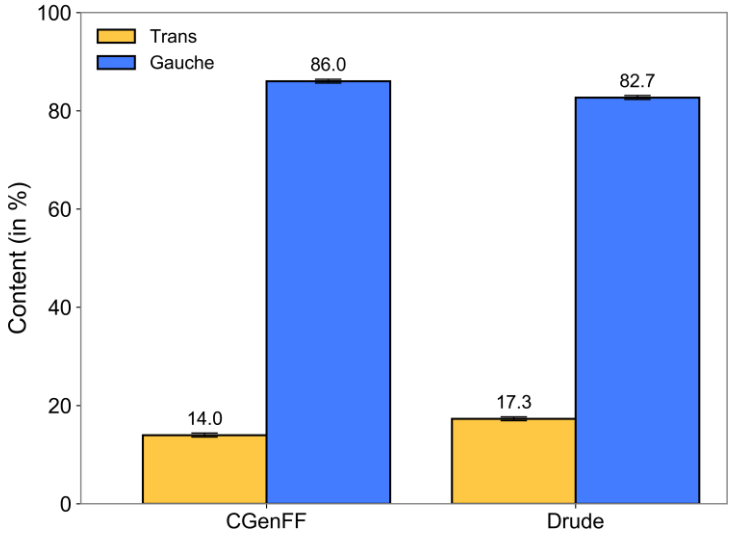
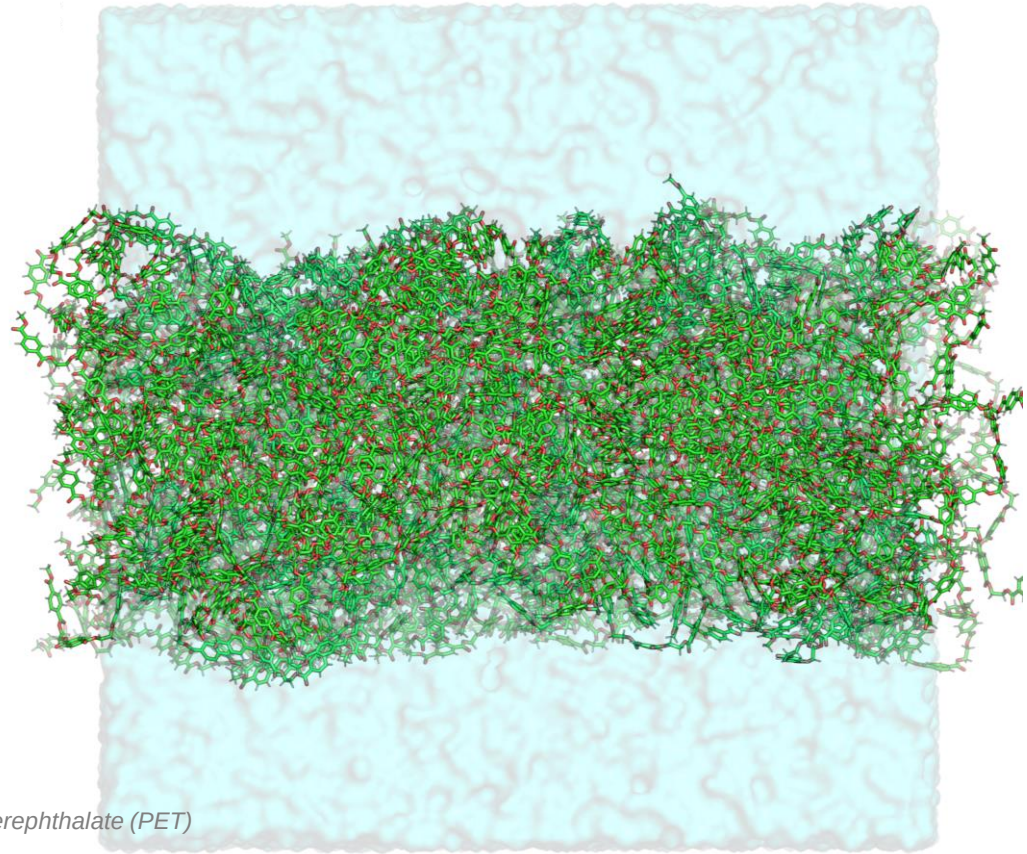
02 - Electronic properties of amorphous PET

Experimental:
Trans: 14% / Gauche: 86%





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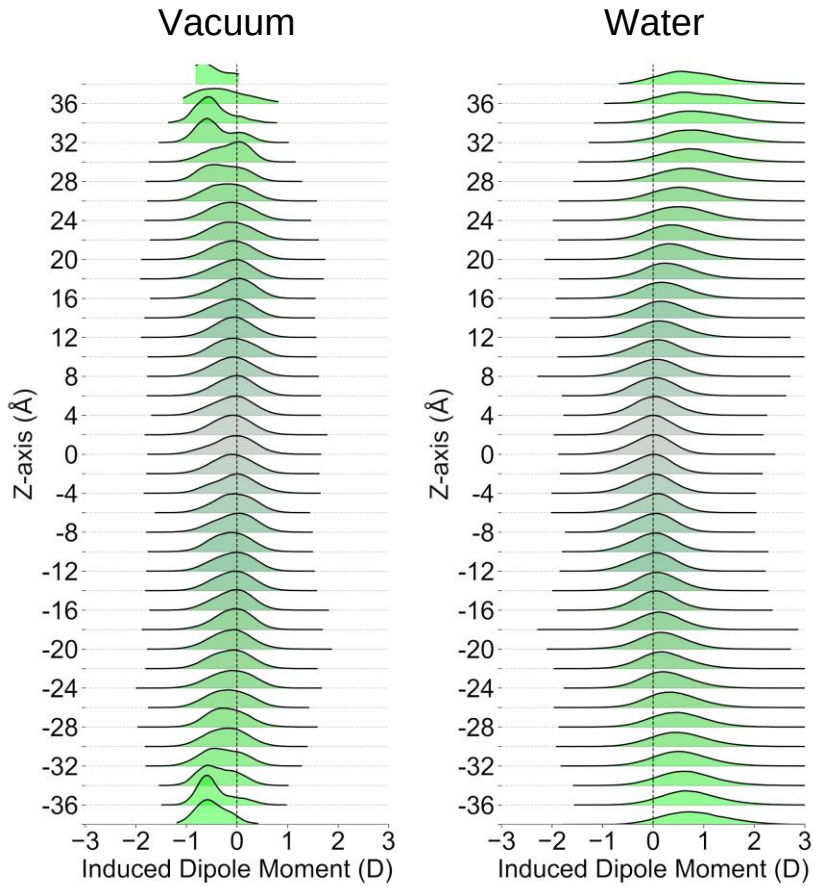
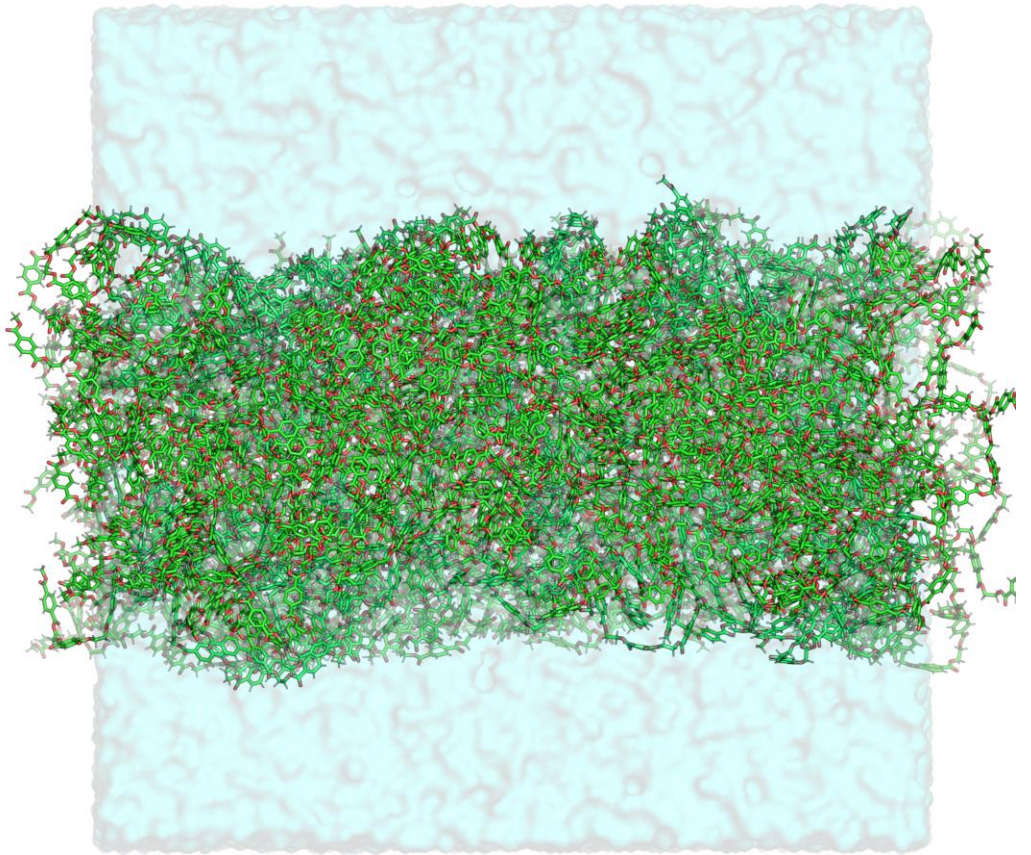
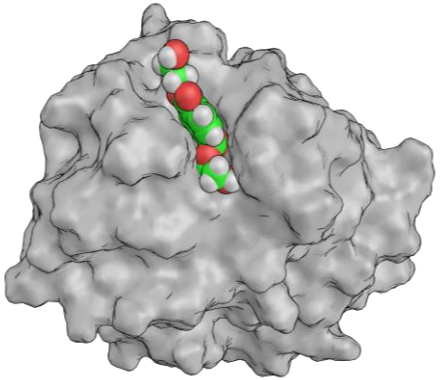
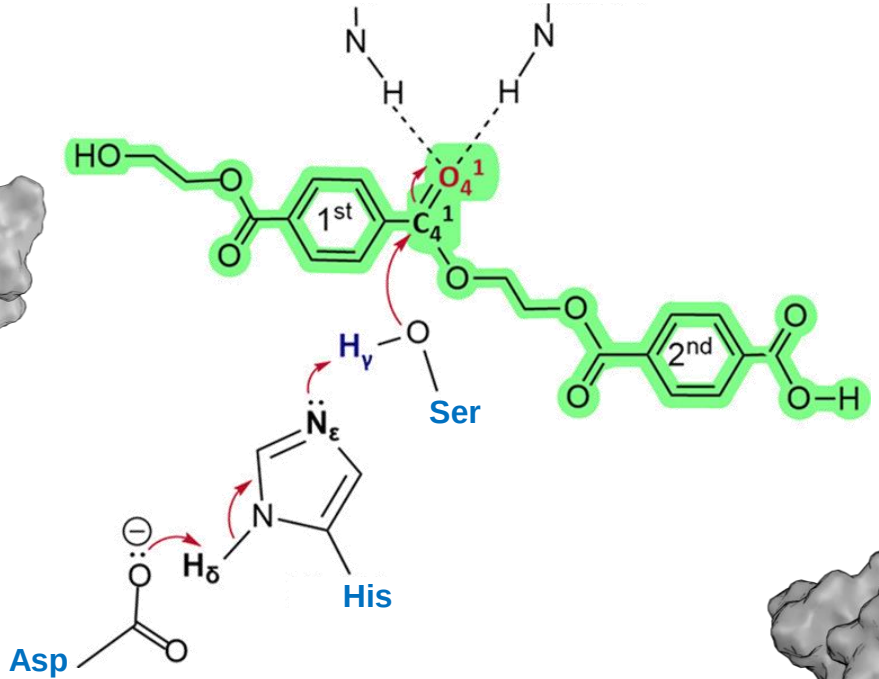


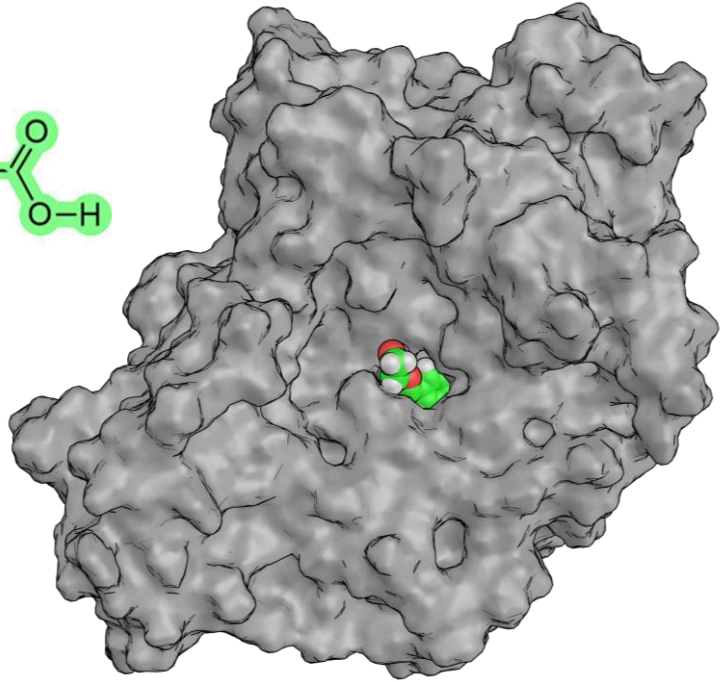


03 - Electronic properties of PET ligands

Catalytic triad: **Ser - His-Asp**

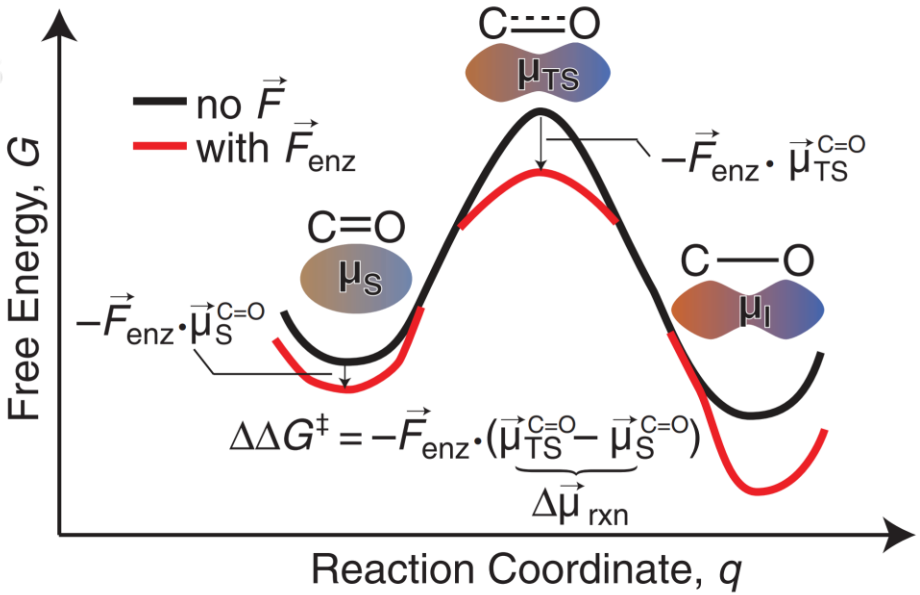


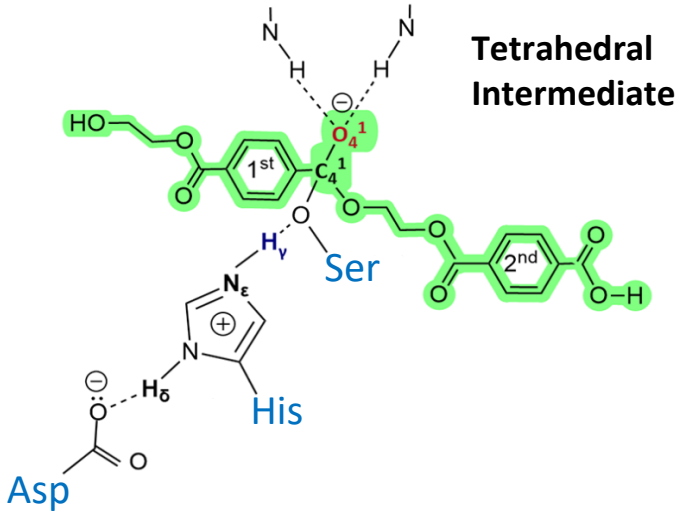




Polêto, M.D., Lemkul, J.A. Exploring enzyme-mediated electronic polarization in PET catalysis using a Drude polarizable model. In Preparation

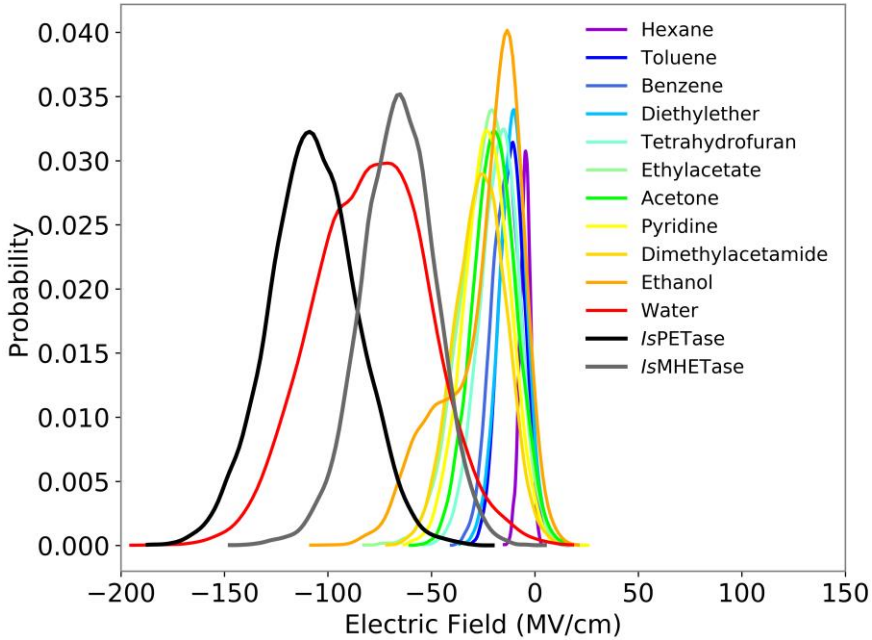
03 - Electronic properties of PET ligands

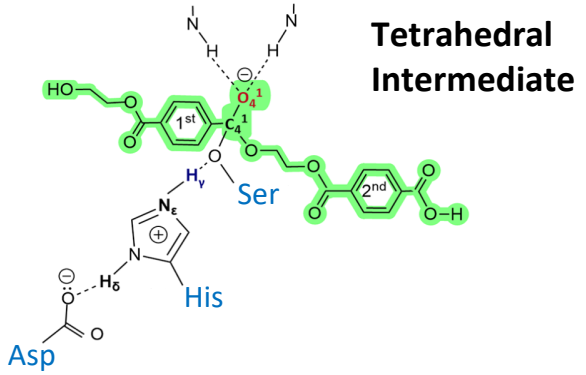


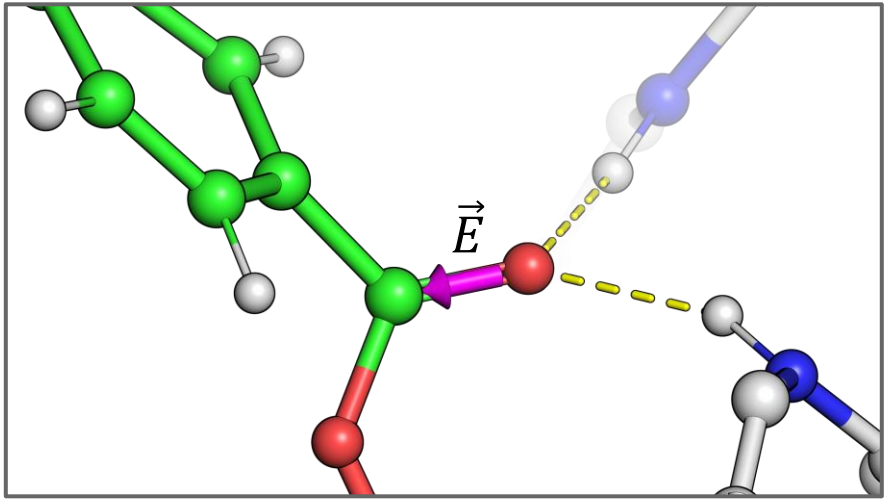


Fried, S.; Bagchi, S.; Boxer, S. Extreme electric fields power catalysis in the active site of ketosteroid isomerase. Science, 346, 1510-1514 (2014)

03 - Electronic properties of PET ligands







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Takeaway points

- Our model works! =D
- Electronic properties at the polymer-solvent interface are an important aspect of these systems and should not be neglected.
- PETase and MHETase active sites modulate the substrate in different ways. How? We want to further test this.
- Can we use the electric field data to guide enzyme engineering efforts?

Thank you!

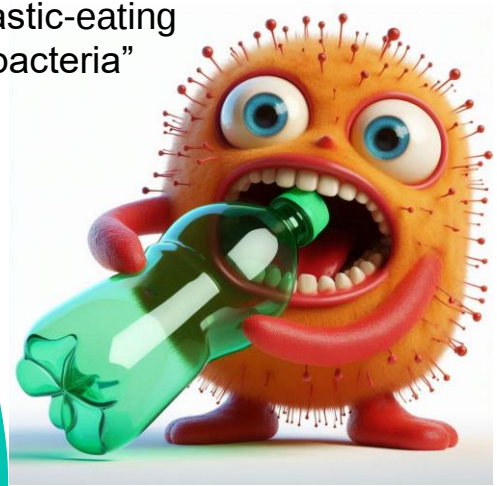
I would be happy to take any questions.

https://mdpoleto.github.io/tupa/

TUPÃ

Electric field analyses for molecular simulations

AI trying to draw a “plastic-eating bacteria”

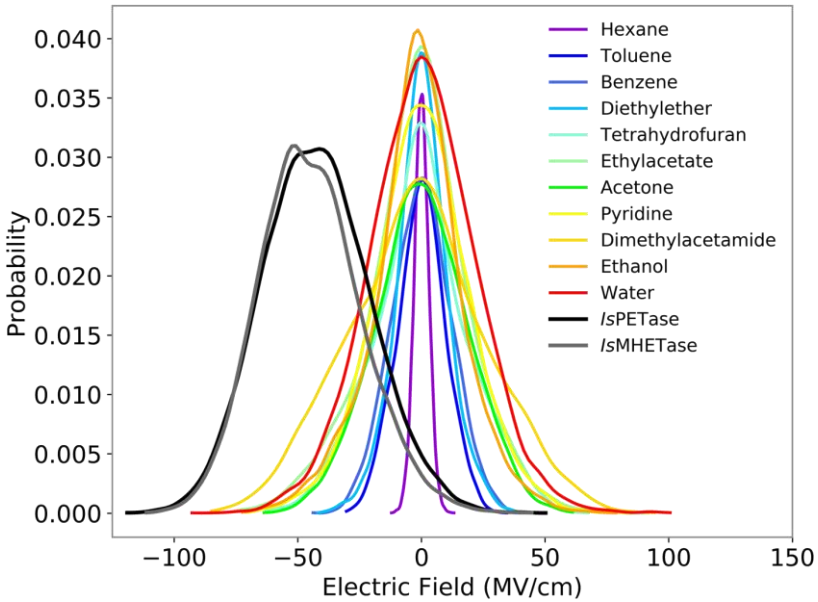


marcelodepolo@gmail.com

https://mdpoleto.github.io

03 - Electronic properties of PET ligands

Electric Field at CX2 (Z-axis contribution)



Drude particle displacement