SCIENCES & BIOENGINEERING SCIENCES

The Research Group
General Chemistry

has the honor to invite you to the public defense of the PhD thesis of

Mats Denayer

to obtain the degree of Doctor of Sciences

Title of the PhD thesis:

Towards a Reliable Computational Protocol to Predict Polymer Solubility through Molecular Dynamics Simulations: Development and Application to Polymer Recycling

Promotors:

Prof. dr. Frank De Proft (VUB)
Prof. dr. Frederik Tielens (VUB)
Dr. Jelle Vekeman (UGent)

The defense will take place on

Friday, February 23, 2024 at 16h00 in auditorium I.2.02

The defense can be followed through a live stream: https://bit.ly/3HVsQNl

Members of the jury

Prof. dr. Ulrich Hennecke (VUB, chair)
Prof. dr. Mercedes Alonso (VUB, secretary)
Prof. dr. Niko Van den Brande (VUB)
Dr. Julia Contreras-Garcia (Sorbonne Université and CNRS, France)
Prof. dr. ir. Dirk De Vos (KULeuven)
Prof. dr Farnaz Heidar-Zadeh (Queen's University, Canada)

Curriculum vitae

After earning the title of Master of Sciences in Chemistry at the Vrije Universiteit Brussel (VUB) in 2019, Mats joined the General Chemistry research group as a predoctoral student in October of the same year. His research focused on the theoretical prediction of polymer solubility with the aim of contributing to a circular economy. He has (co-) authored four scientific papers published in international peer-reviewed scientific journals, and four more manuscripts are currently in preparation. In addition to his research activities, Mats actively engaged in teaching both Bachelor and Master courses.

Abstract of the PhD research

Concerned by the environmental impact of polymer use, the European Union and the United Nations desire a transition from the current linear "use and dispose" plastic economy towards a circular model, limiting the accumulation of plastic waste and reducing the exploitation of fossil recourses. Hereto, recycling processes must be developed in which the original polymer can be recovered without losing product quality (down-cycling) while the solvents utilized in the process are preferably "green" and usable in reduced volumes. A key component in the development of novel recycling processes is the selection of efficient solvents that can be used effectively and sustainably. In this effort, predictive tools are indispensable to optimize existing solvents as well as to screen for viable new alternatives. Current approaches rely heavily on experimental input and sometimes lack accuracy and chemical insight. Moreover, they cannot account for polymer crystallinity, and features such as polymer swelling are not easily captured.

In this thesis, a novel computational method to predict polymer solubility is proposed, combining classical Molecular Dynamics (MD) simulations with a conformational and a Non-Covalent Interaction (NCI) analysis. It allows to gain insight in the dissolution process (polymer swelling and chain disentanglement) at a molecular level, while delivering detailed information on the chemical interactions governing the solubility behaviour. A bottom-up approach was adopted, whereby a single polymer chain was brought into solution and an NCI-based index, labelled the integrated NCI density on one hand, and the Solvent Accessible Surface Area (SASA) on the other, were taken as the fundamental quantities to express solubility. In a next step of the progressive approach, an amorphous bundle consisting of multiple polymer chains was immersed into a solvent and the breakup of the assembly was assessed. A dissociation propensity was defined as the ratio of the equilibrated to the initial SASA, effectively capturing the polymer's solubility in a single number. The developed protocols were successfully applied to real-life problems encountered by our experimental partners in the context of polymer recycling and green alternatives.