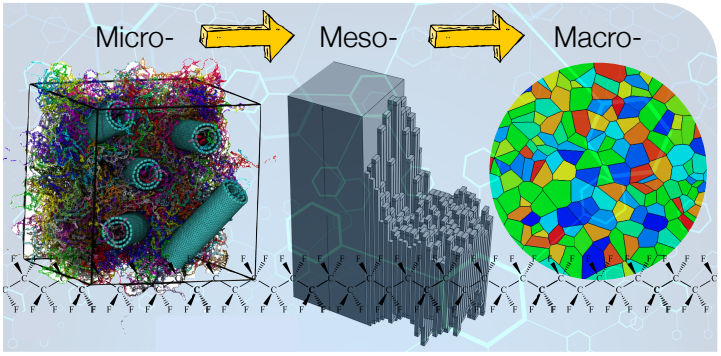
(Truly) Multiscale Simulations of Polymer Crystallization: from molecular dynamics to kinetic modelling

Supervisors:
Dr. Gabriele C. Sosso (Warwick Chemistry) and Dr. Lukasz Figiel (WMG)

The project:
The crystallization of long-chain macromolecules in the presence of nanoparticles is an intriguing physical phenomenon, which is of high practical relevance to optimum fabrication and end-use properties of environmentally sustainable composite materials and their products [1] . The theoretical understanding of the phenomenon is limited by its complexity, caused by the inherent dynamic nature of the problem and the hierarchy of processes across different temporal and spatial length scales [2]. Thus, given an ever-increasing need for sustainable polymer composites in various industrial sectors (from packaging to energy, and biomedical applications), the accurate and reliable modelling of polymer crystallization with nanoparticles [3,4] is essential to: (1.) improve our fundamental understanding of the phenomenon down to the nanoscopic length scale, and (2.) enable design of the next-generation sustainable composite materials, and associated manufacturing techniques.

A standard approach to model the crystallization kinetics of polymers is the Avrami equation [5] (or one of its extensions [6,7]), an empirical modelling framework that can quantify crystal nucleation and growth. However, this approach suffers from severe limitations when applied to polymer crystallization, especially under non-isothermal conditions and/or in the presence of nucleating agents. Those aspects are particularly important for industrially relevant manufacturing processes for composites of polymers and functional nanofillers.



Instead of blindly seeking yet another modification to the Avrami framework via trial and error, this project aims to predict crystallisation kinetics via a systematic bottom-up approach based on multiscale simulations. Specifically, we will employ coarse-grained molecular dynamic simulations to unravel the nanoscopic interactions involved with the crystallization of polymers with nanoparticles, taking into account both interaction strength and directionality, and nanoparticle type. Their results will then be used to build lattice models [8] that will allow us to investigate polymer crystallization on much larger length scales, and subsequently produce a data-driven macroscopic modelling framework exploiting Gaussian Processes [9] for predicting the non-isothermal crystallization of heterogeneous polymer mixtures.

The outcomes:

- Molecular-level understanding of the crystallization kinetics of polymers
- A data-driven novel macroscopic model, capable of describing polymer crystallization in composite material systems.
- A theoretical framework, delivered by means of a transformative bottom-up approach, to predict the crystallization behaviour of polymers in the presence of nanoparticles.
- A suite of general-purpose software connecting coarse-grained molecular dynamics simulations with lattice models

References:
[1] S. Chen, E. Olson, S. Jiang, X. Yong, Nanoscale 12 (2020) 14560–14572.
[2] T. Verho, A. Paajanen, J. Vaari, A. Laukkanen, Macromolecules 51 (2018) 4865–4873.
[3] T. Yamamoto, Polymer 50 (2009) 1975–1985.
[4] J.-U. Sommer, G. Reiter, J. Chem. Phys. 112 (2000) 4384–4393.
[5] M. Fanfoni, M. Tomellini, Il Nuovo Cimento D 20 (1998) 1171–1182.
[6] R.J. Kirkpatrick, J. Geophys. Res. 1896-1977 81 (1976) 2565–2571.
[7] J.W. Cahn, Acta Metall. 4 (1956) 449–459.
[8] Y. Lifanov, B. Vorselaars, D. Quigley, J. Chem. Phys. 145 (2016) 211912.
[9] C.E. Rasmussen, in: Adv. Lect. Mach. Learn., Springer, Berlin, Heidelberg, 2004, pp. 63–71.