Abstract

Understanding the relationship between molecular packing, crystal morphology, and surface properties is essential for the rational design of organic crystalline particles. Surface anisotropy plays a key role in determining crystal growth, stability, and interactions with the surrounding environment, ultimately influencing manufacturing efficiency and material performance in applications ranging from pharmaceuticals to functional materials.

In this PhD thesis an integrated experimental and computational approach was developed to investigate the surface properties of selected organic crystalline materials. Particular attention is devoted to the quercetin–dimethylformamide solvate (QDMF), used as a model system to explore facet-specific chemical and mechanical behaviour.

Computational methods, including *Particle Informatics* (PI) tools, were employed to understand the link between molecular structure and crystal lattice properties, analyse intermolecular interactions, predict crystal habit, and estimate surface chemistry across different crystallographic facets. These modelling approaches provided a framework for identifying the dominant intermolecular interactions governing crystal growth and surface stability; thus enabling digital design of crystalline materials with target properties.

Experimental characterization was carried out using complementary spectroscopic and microscopic techniques spanning multiple length scales. Bulk-sensitive techniques such as Infrared and Raman Spectroscopy were combined with high-resolution surface methods including Optical Photothermal Infrared Spectroscopy (OPTIR), scattering-type Scanning Near-field Infrared Microscopy (s-SNIM), and Atomic Force Microscopy (AFM). These techniques enabled the investigation of nanoscale chemical heterogeneity, facet-dependent mechanical properties, and surface transformations associated with desolvation processes.

The results demonstrate that crystal surfaces exhibit pronounced chemical and mechanical anisotropy arising from the underlying intermolecular interaction network. The integration of modelling and experimental characterization provides new insights into the relationship between crystal structure and surface behaviour, highlighting the potential of combined computational–experimental strategies to predict particle quality attributes and guide the design of crystalline materials.