Functional materials exploration through evolutionary searching and large-scale crystal structure prediction

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The design of molecular crystals with targeted properties is the goal of crystal engineering. However, our predictive understanding of how a crystal's properties relate to its structure, and how crystal structure in turn relates to molecular structure, are not yet sufficiently reliable to confidently design functional materials. Computational methods for crystal structure prediction (CSP) have been developed to help anticipate the crystal structure that a molecule will form. These methods are based on a global search of the lattice energy surface and a ranking of local energy minima according to their calculated relative stabilities. Thus, each molecule is associated with a list of potential crystal structures, each of which then leads to a set of predicted properties. The resulting ensemble of structures, their relative energies and associated properties can be interpreted to judge a molecule's promise for a target function. These methods have been demonstrated to be valuable in guiding experimental materials discovery programmes. A remaining challenge is the best choice of molecules that should be assessed, given the enormous chemical space of possible molecules. To address this, we have combined evolutionary searching of chemical space with large scale crystal structure and property prediction as a route to the discovery of novel molecules with high likelihood of yielding good properties [1]. The approach will be discussed with example studies in the area of organic semiconductor discovery.

[1] Cheng, C. Y., Campbell, J. E. and Day, G. M. (2020) *Chem. Sci.*,**11**, 4922-4933.

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