Poster

Multicomponent crystals of sulfamic acid with amino acids as alternative piezoelectric / NLO materials

Suman Bhattacharya[1], Matthew Gleeson[2], Emma Kiely[1], Pierre A. Cazade[1], Krishna Hari[1], Tara Ryan[1], Sarah Guerin*[1]

[1]Department of Physics and Bernal Institute, University of Limerick, Limerick, Republic of Ireland, V94 D9W8 [2] Laboratoire d'optique et biosciences, École Polytechnique, Route de Saclay, 91128 Palaiseau Cedex, France

Suman.S.Bhattacharya@ul.ie ; Sarah.Guerin@ul.ie

Multicomponent crystals [1], *viz.*, cocrystals and salts, designed and synthesised following crystal engineering [2] principles, are alternative solid forms of a parent material with altered structures and properties of interest. The number of reported cocrystals has been increasing exponentially in the Cambridge Structural Database every year. In the previous century, designing multicomponent crystals was an exercise aimed at understanding the science of self-assembly, followed by the impactful emergence of pharmaceutical cocrystals [1,3]. A paradigm shift has occurred in the last two decades, where multicomponent crystals are being studied for understanding mechanical, thermal and electrical properties [4], enabling future design of materials with targeted properties of interest and structures.

Piezoelectricity is the phenomenon of conversion of mechanical energy to electric energy and *vice versa*. Piezoelectric materials when exposed to mechanical stimuli generate voltage, and conversely, applying voltage to a piezoelectric material generates a mechanical force. Traditional piezomaterials, *e.g.*, lead zirconium nitrate (PZT), lithium niobate ($LiNbO_3$), and potassium sodium niobate (KNN), although efficient in terms of performance, are toxic and lack the possibility of further design and development. Hence, our recent focus has been on exploring organic piezoelectric materials which are biocompatible and environmentally friendly[5-7]. Cocrystallisation of organic materials further provides the opportunity to expand the material library, enabling further design of materials with higher structure and property control.

Amino acids are non-toxic, biocompatible, inherently chiral materials and have been explored as alternative piezoelectric and NLO materials [6]. Amino acid crystals harbour few disadvantages, *e.g.*, limited solubility in other solvents except water and a high degree of crystallinity, which creates problems in using them for practical purposes in the form of films or stand-alone assemblies. Guerin *et al.* reported the cocrystal of glycine with sulfamic acid in 2017 [7]. While the report focused on generating a non-centrosymmetric piezoelectric system using achiral coformers, it also revealed sulfamic acid as a potential coformer for producing cocrystals with amino acids.

Herein, we discuss the piezoelectric and non-linear optical (NLO) performance of a series of sulfamic acid multicomponent crystals with diverse amino acids *viz.*, glycine, L-arginine, L-methionine, L-valine, L-histidine and L-phenylalanine. All of these multicomponent crystals could be scaled up with considerable phase purity and have been systematically characterised using diffraction, thermal analysis, and infrared spectroscopy. Importantly, the cocrystal phases could be developed as moulded polycrystalline mould assemblies without a substrate. Theoretical calculations and experimental efforts have validated the performance of all the multicomponent crystals.

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