A100-02-270823 The Arsenal of Supramolecular Interactions for Crystal Engineering

Full-colour palette from one chromogen – modular design strategy revealed by quantum crystallography

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Technological applications of *colour chemistry* are enormous. For example, *chromic materials* can be found in photochromic systems or smart self-dimming windows, paints and indicators, thermal papers and visual displays or in biosensors [1].

Many known crystals can change colour selectively. [2] However, not many chromogens can create multi-coloured systems in the presence of a particular colourless coformer. [3] Even fewer show high performance, sufficient flexibility of the chromic response to applied stimuli and reversibility of the process.

It is vital to understand molecular aggregation to obtain materials with enhanced properties. Each structure can be divided into subsections (modules), which can be modified, removed or exchanged with other molecules or between different systems. Understanding how those modules interact with each other and how they change depending on the in-crystal environment is necessary to design smart devices.

In this work, we have used *quantum crystallography tools* [4,5] in conjunction with H^1 *NMR spectroscopy* to formulate a general mechanism of *chromic effects* in violuric acid salts and co-crystals. The obtained results proved that we could influence the light absorption properties of a material using a reformulated reverse crystal engineering concept. The *modular approach* led to distinctly coloured multicomponent crystalline products (Fig. 1) based on colourless starting components (e.g. pyridine, aliphatic and aromatic amine derivatives). Chromic effects were examined using solid-state UV-VIS spectroscopy, XRPD and hot-stage microscopy. Our research will contribute to a more conscious design of *chromic multicomponent materials*.

Figure 1. Full-colour palette from one chromogen – multicomponent chromic materials based on violuric acid.

[1] Vik, M. Periyasamy, A. P. ed. Viková M. (2021) *Chromic Materials. Fundamentals, Measurements and Applications*, CRC Press.
[2] Bamfield, P., Hutchings, M. (2018) *Chromic Phenomena: Technological Applications of Colour Chemistry*, Royal Society of Chemistry.
[3] Gryl, M., Rydz, A., Wojnarska, J., Krawczuk, A., Koziel, M., Seidler, T., Ostrowska, K., Marzec, M. & Stadnicka, K. M. (2019). *IUCrJ* 6, 226-237.
[4] Bader, R. F. W. (2003). *Atoms in Molecules: A Quantum Theory*, International Series of Monographs on Chemistry, Vol. 22. Oxford: Clarendon Press.
[5] AIMAll (Version 19.10.12), Todd A. Keith, TK Gristmill Software, Overland Park KS, USA, 2019 (aim.tkgristmill.com); Dovesi, R.; Erba, A.; Orlando, R.; Zicovich-Wilson, C. M.; Civalleri, B.; Maschio, L.; Rerat, M.; Casassa, S.; Baima, J.; Salustro, S.; Kirtman, B. *WIREs Comput Mol Sci*., 2018, 8, e1360.

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