## Controlled Crystallization of Centimeter-Size 4HCB Organic Single Crystalline Semiconductors for Novel Radiation Sensor Applications

Dou Zhao<sup>1</sup>\*, Donghao Ma<sup>1</sup>, Wanqi Jie<sup>1</sup>, Yadong Xu<sup>1</sup>, Wenbao Jia<sup>2</sup>, Xiaoping Ouyang<sup>3</sup> \*lead presenter: z.dou@mail.nwpu.edu.cn

- 1 Northwestern Polytechnical University, China
- 2 Nanjing University of Aeronautics and Astronautics, China
- 3 Northwest Institute of Nuclear Technology, China

4-hydroxycyanobenzene (C<sub>7</sub>H<sub>5</sub>NO, 4HCB) organic single crystalline semiconductors (OSCS) have raised significant interest in next-generation radiation detection applications due to their high hydrogen density, tissue equivalence, and excellent charge transport properties [1-3]. However, their performances suffer from the uncontrollable solution growth process, leading to issues such as dense surface defects, serious crystals adhesion and small crystals size [4-6]. To improve the quality of the as-grown 4HCB crystals, the physical origin behind the above issues should be clarified first.

Here, the two-dimensional (2D) nucleation and layer-growth mechanism for solution-grown 4HCB OSCS is clarified by analyzing the 2D anisotropic intermolecular force and characterizing the multiscale surface layer edges. In this mechanism, the small increase of solution supersaturation leads to a dense crystallization nucleus, resulting in the growth issues mentioned above. Then, the controlled crystallization method in terms of the solution supersaturation (by adjusting growth temperature) and the nucleation position (by adjusting the anti-solvent) is utilized to optimize growth of 4HCB OSCS, with obtaining 4HCB OSCS in the dimension of  $18\times15\times1.2$  mm<sup>3</sup>, high bulk resistivity of  $(1.12\pm0.05)\times10^{12}$   $\Omega\cdot\text{cm}$ , and excellent charge mobility of  $3.40\pm0.10$  cm<sup>2</sup>·s<sup>-1</sup>·V<sup>-1</sup>.

As the result, high-performance sensors are fabricated based on the as-grown 4HCB OSCS, achieving direct detection of low-dose X-rays [7], solar-blind UV lights [8], and fast neutrons [9] with superior performances, fast response and low-cost.

## References

- [1] Fraboni, B. et al. Solution-grown, macroscopic organic single crystals exhibiting three-dimensional anisotropic charge-transport properties. *Adv. Mater.* **2009**, *21* (18), 1835-1839.
- [2] Fraboni, B. et al. Three-dimensional anisotropic density of states distribution and intrinsic-like mobility in organic single crystals. *Org. Electron.* **2010,** *11* (1), 10-15.
- [3] Fraboni, B. et al. Organic semiconducting single crystals as next generation of low-cost, room-temperature electrical X-ray detectors. *Adv. Mater.* **2012**, *24* (17), 2289-2293.
- [4] Fraleoni-Morgera, A. et al. Solution growth of single crystals of 4-hydroxycyanobenzene (4HCB) suitable for electronic applications. *J. Cryst. Growth* **2010**, *312* (23), 3466-3472.
- [5] Ciavatti, A. et al. Toward low-voltage and bendable X-Ray direct detectors based on organic semiconducting single crystals. *Adv. Mater.* **2015**, *27* (44), 7213-7220.
- [6] Ciavatti, A. et al. Charged-particle spectroscopy in organic semiconducting single crystals. *Appl. Phys. Lett.* **2016**, *108* (15), 153301.
- [7] Zhao, D. et al. Purely organic 4HCB single crystals exhibiting high hole mobility for direct detection of ultralow-dose X-radiation. *J. Mater. Chem. A.* **2020**, 8 (10), 5217-5226.
- [8] Zhao, D. et al. Solar-blind UV detection by ultra-wide-bandgap 4HCB organic single crystal semiconductor. *Appl. Phys. Lett.* **2022**, *120* (1), 013301.
- [9] Zhao, D. et al. Direct Detection of Fast Neutrons by Organic Semiconducting Single Crystal Detectors. *Adv. Funct. Mater.* **2022**, *32* (7), 2108857.