Supplementary Information

High-throughput growth of organic scintillating single crystal fibers

for neutron/gamma pulse shape discrimination

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1. Materials

The *p*-terphenyl (analytical grade), 9, 10-diphenylanthrance (purified by sublimation) were purchased from TCI. The octadecyltrichlorosilane (OTS, 90%) was purchased from Sigma-Aldrich. Acetone, toluene and isopropanol (analytical grade) came from Tianjin Fuyu Fine Chemical Co., Ltd. H_2O_2 and H_2SO_4 were purchase from China National Pharmaceutical Group Co., Ltd and Yantai Yuandong Fine Chemical Co., Ltd, repectively. All the chemicals were used without further purification. The quartz capillaries with various inner diameter are commercially available.

2. Experimental procedures

2.1 Purification of *p*-terphenyl

In order to improve the purity of *p*-terphenyl, its raw material was purified by different methods. Firstly, the powder was purified by recrystallization from toluene. Then, the *p*-terphenyl was sublimation deposition in a double temperature zone tube furnace with N_2 gas flow. Finally, the *p*-terphenyl powder was purified by zone refining.

2.2 Cleaning of molds

Firstly, the quartz mold was emersed in acetone for five days. After that, it was subjected to ultrasonic cleaning in ultrapure water for three times with each session lasting five minutes. Then, the mold was washed by hot $H_2SO_4:H_2O_2$ (2:1) for 30 minutes. After the solution cooled, the mold was cleaned with ultrapure water to eliminate any residual solution. Finally, the quartz mold was dried in an oven at 100 °C, ensuring it was free from any contaminants before reuse. The quartz molds were treated by O_2 plasma/OTS vapor.

2.3 Preparation of organic scintillating single crystal fiber

The organic scintillating SCFs were prepared using *p*-terphenyl as host, 9, 10dipenylanthrance as guest with different doping concentrations of 0.01 wt%, 0.05 wt%, 0.1 wt% and 1 wt%. Specifically, the high-temperature zone was set at 245 °C, while the low-temperature zone was set at 185 °C.

2.4 Fabrication of the SCF array

Initially, two parts of epoxy resin were mixed in a mass ratio of 1:2. This mixture was then poured into a template containing the arranged SCFs. Then, the composite was left

to cure for 12 hours in an oven at 80 °C to ensure the polymer solidified effectively. Finally, the SCFs were segmented into an array using a diamond wire sawing machine.

3. Characterization

The refractive indexes of doped crystal and quartz were obtained from Horiba UVISEL Plus. Optical and cross-polarized optical microscopy images were obtained using a Zeiss Axiolab 5 microscope. The water contact angle tests were performed using JC2000C1, POWEREACH. The distribution of molten *p*-terphenyl liquid was observed by real time observation under a hot stage microscope system consisting of a Linkam THMS600 heating stage and a Leica DM2700P microscope. Scanning electron microscopy (SEM) images and elemental mapping analysis were finished using a Phenom Pharos benchtop scanning electron microscope. The Laue diffraction patterns were collected with a real-time back-reflection Laue camera system (MWL 120, Multiwire Laboratories, Ltd.). The X-ray diffraction pattern of single crystal fiber was collected by Bruker AXS D2 Phaser X-ray powder diffractometer using Cu Ka radiation ($\lambda = 1.5418$ Å).

The calculation of texture coefficient of scintillating single crystal fiber is according to the following equation:

$$TC_{(hkl)} = \frac{I_{(hkl)}}{I_{0(hkl)}} / (\frac{1}{N} \sum_{i=1}^{N} \frac{I_{(h_{i}k_{i}l_{i})}}{I_{0(h_{i}k_{i}l_{i})}})$$

where $I_{(hkl)}$ is the observed relative intensity of (hkl) peak in XRD pattern, $I_{0(hkl)}$ is the diffraction intensity of (hkl) peak in standard XRD pattern.

The Raman spectra of crystal samples were carried out on a Horiba confocal Raman spectrometer equipped with a laser ($\lambda = 633$ nm) under ambient condition. The steady-state photoluminescence emission (PL) spectra of single crystal fiber were collected on an Edinburgh Instruments FLS 1000 spectrometer equipped with a Xenon lamp. The lifetime measurements of the doped organic single crystal fibers were conducted on Edinburgh FLS 1000 spectrometer equipped with a pulsed laser source (EPL-375, excitation wavelength: 375 nm). The emission decay was recorded at 387 nm. The photoluminescence quantum yield (PLQY) was determined by using an integrating sphere on FLS 980 spectrometer. The theoretical calculations were finished

by the Gaussian program at B3LYP/6-31G(d, p) level. The absorption spectra were conducted on Cary 50 spectrophotometer at medium scan rate in the range of 200–800 nm. The radioluminescence (RL) spectrum was collected using an Edinburgh Instruments FLS 1000 spectrometer coupled with an X-ray tube (Tungsten target, TUB00154-9I-W06, Moxtek Inc.) operated at 50 kV and 50 μ A.

4. Neutron detection

Neutron/gamma pulse shape discrimination (PSD) properties were studied using multichannel analyzer of CAEN-N6730SB, photomultiplier tube (PMT, Hamamatsu CR173-01) and radiation excitation source ²⁵²Cf. The polished crystal and fiber array were wrapped with Teflon film except for one face coupled to the PMT entrance window that was placed in a closed aluminum box light shield. Silicone oil was used as a coupling agent. The PMT operating voltage was set at 800 V during the measurement. And the signals collected at the PMT anode were recorded using a CAEN N6730SB 12-bit digitizer at a sampling rate of 500 MHz for offline analysis. The PSD performance was quantitative analysis by comparison of figure of merit (FOM). The FOM value was optimized by adjusting the time of short gate and cutting different energy range of neutron/gamma.

The optimization of pulse shape discrimination (PSD) performance was primarily achieved through careful tuning of the signal integration windows used to distinguish between neutron and gamma events. Specifically, the prompt and delayed integration windows were systematically adjusted to maximize the figure of merit (FOM), defined as the separation between the neutron and gamma peaks divided by the sum of their full widths at half maximum (FWHM). This process ensures optimal separation and minimizes event misclassification.

Additionally, we selected a suitable photomultiplier tube (PMT) and a suitable multi-channel analyzer to faithfully capture the fast scintillation decay characteristics of the organic single crystal. This is critical since the differences in pulse shapes for neutron and gamma interactions are subtle and require high fidelity signal acquisition.

In our experiments, the PMT was operated within the manufacturer's recommended linear dynamic range to balance between sufficient gain and signal-to-

noise ratio, and to avoid saturation effects. Although some degree of bending is observed in the PSD plot, we have checked that the pulse amplitude remains within the regime where the PMT response is sufficiently linear for discrimination purposes. Moreover, despite the slight bending, the neutron and gamma bands remain well separated across the entire energy range of interest, and the figure of merit (FOM) for PSD remains stable. This indicates that the observed curvature does not significantly degrade the accuracy of neutron/gamma discrimination in our case.

Indeed operating at a lower voltage reduce the risk of non-linearity, while it may also reduce the signal amplitude and lead to poor signal-to-noise ratio, particularly for lower-energy events, thus causing negative impact on timing resolution and PSD performance. Based on our experimental experience, the selected voltage offers the best compromise between linearity, gain, and discrimination performance. In future studies, systematic investigations of PMT voltage effects will be carried out to further refine the PSD performance.



Fig. S1 The water contact angles for different treated surfaces (the upper panel) and the distribution of molten *p*-terphenyl liquid on the different treated surfaces (the lower panel).

We modified the surfaces of growth molds by grafting self-assembled monolayers, including octadecyltrichlorosilane (OTS) and O_2 plasma to tailor the wettability. The wettability of modified surfaces was characterized through the measurement of their water contact angles. The results revealed the following order of wettability: OTS <

bare < O_2 plasma, with corresponding contact angles of 90.50°, 42.50°, and 0°, respectively. At 250 °C, the molten *p*-terphenyl liquid exhibited unfavorable wetting condition on the hydrophobic OTS surfaces, shrinking into hemispherical droplets to minimize contact area. In contrast, the molten *p*-terphenyl liquid demonstrated better spreading behavior on the O_2 plasma-treated surface. The enhanced wettability facilitated capillary action and maintained the stability of a planar solid-liquid interface during the crystal growth process. Consequently, O_2 plasma-treated quartz capillary molds were selected for the growth of organic scintillating single crystal fibers.



Fig. S2 The optical microscopy (left) and cross-polarized optical microscopy (right) images of SCFs with inner diameter of 500 µm at different growth rates.



Fig. S3 The excitation spectrum of doped crystal with doping concentration of 0.01 wt%.



Fig. S4 The time-resolved photoluminescence spectra of a scintillating single crystal fiber at 411 nm.



Fig. S5 The time-resolved photoluminescence spectra of a scintillating single crystal fiber at 428 nm.



Fig. S6 The time-resolved photoluminescence spectra of scintillating single crystal fiber array at 411 nm.



Fig. S7 The time-resolved photoluminescence spectra of scintillating single crystal fiber array at 428 nm.



Fig. S8 Energy transfer diagram within the doped crystal.



Fig. S9 The lifetime of a scintillating single crystal fiber with doping concentration of 0.01 wt% at 411 nm.



Fig. S10 The lifetime of a scintillating single crystal fiber with doping concentration of 0.01 wt% at 428 nm.

Sample Short gate time/ns	Single crystal fiber array	Monolithic bulk crystal
70	2.039	1.998
80	3.684	2.824
90	3.502	1.862

Table S1. The neutron/gamma pulse shape discrimination performance of single crystal

 fiber array and monolithic bulk crystal at other gate times.



Fig. S11 (a) The variation of FOM value with energy selection range for single crystal fiber array; (b) The variation of FOM value with energy selection range for monolithic organic single crystal.

It can be clearly seen that the FOM values of single crystal fiber array and monolithic organic single crystal were highest within the 300–700 channel energy selection range, with corresponding values of 3.684 and 2.824, respectively.