

## **Supporting Information for Aging effect on the co-crystallization behavior of the donor and acceptor crystals in aqueous dispersion**

Masaki Takeda,<sup>a</sup> Jun Matsui,<sup>\*b</sup> and Akito Masuhara<sup>\*ac</sup>

<sup>a</sup>*Graduate School of Science and Engineering, Yamagata University, 4-3-16, Jonan, Yonezawa, Yamagata, Japan*

<sup>b</sup>*Faculty of Science, Yamagata University, 1-4-12, Kojirakawa-machi, Yamagata, Yamagata, Japan*

<sup>c</sup>*Research Center for Organic Electronics, Yamagata University, 4-3-16, Jonan, Yonezawa, Yamagata, Japan*

\*E-mail: jun\_m@sci.kj.yamagata-u.ac.jp (J. M.); masuhara@yz.yamagata-u.ac.jp (A. M.)

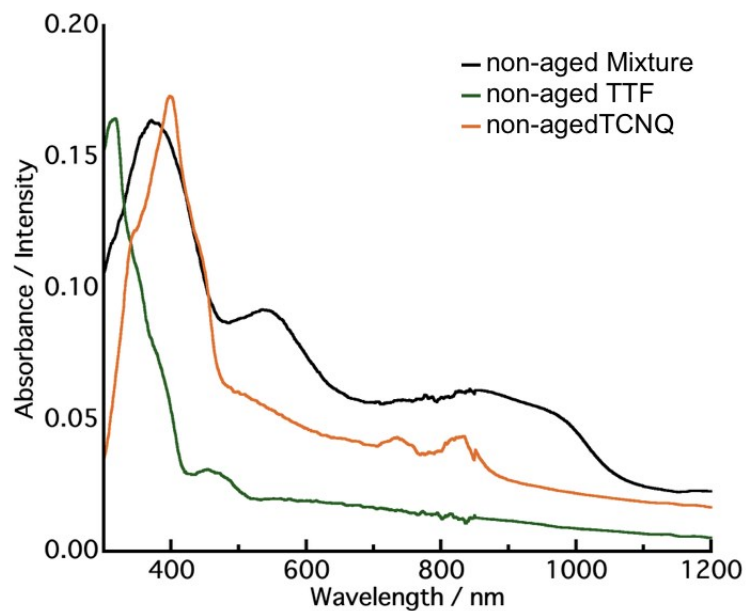


Figure S1. Absorption spectra of the non-aged mixture and their respective constituent crystals.

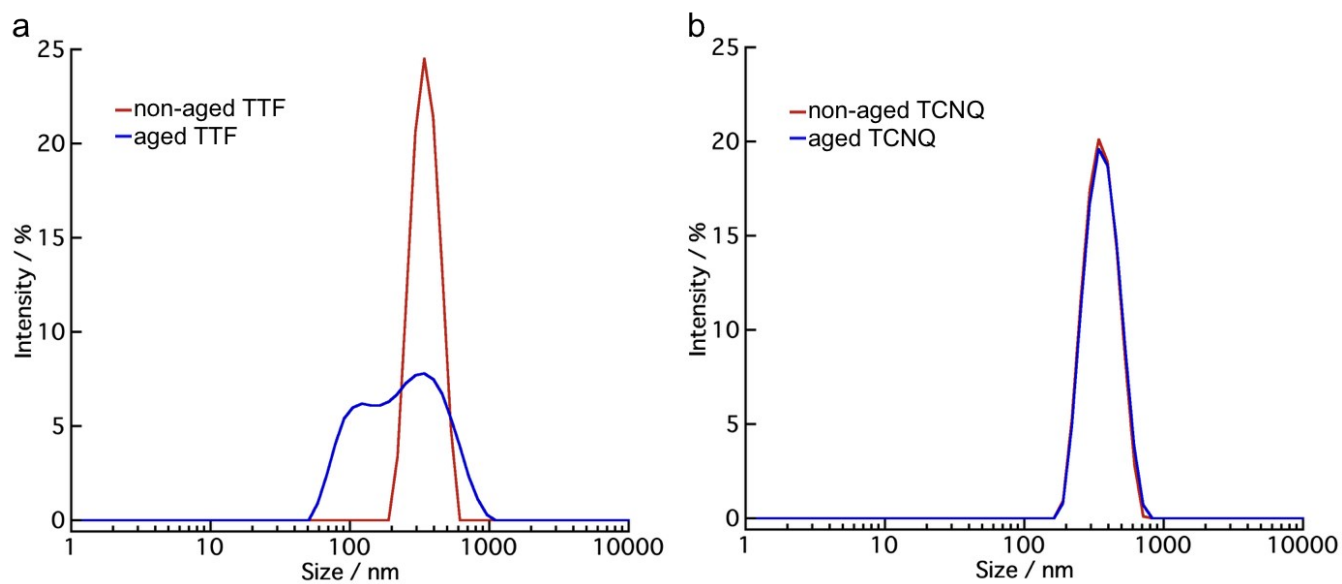
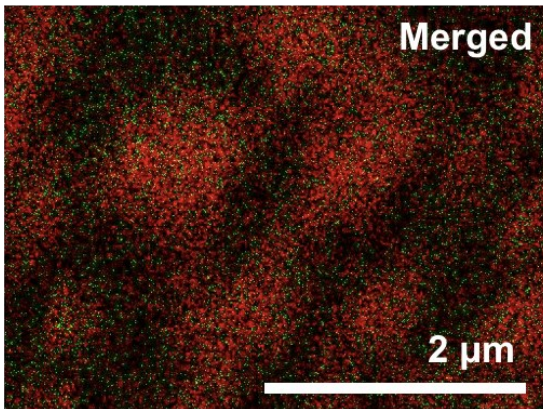
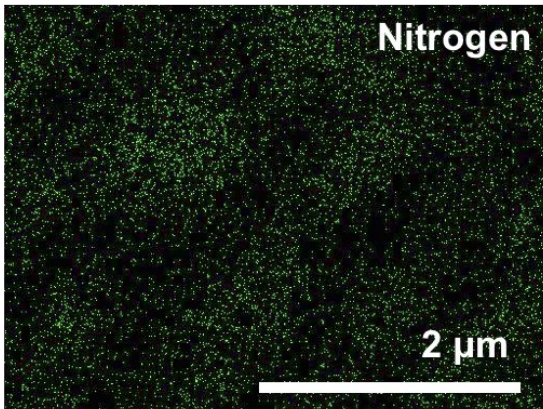
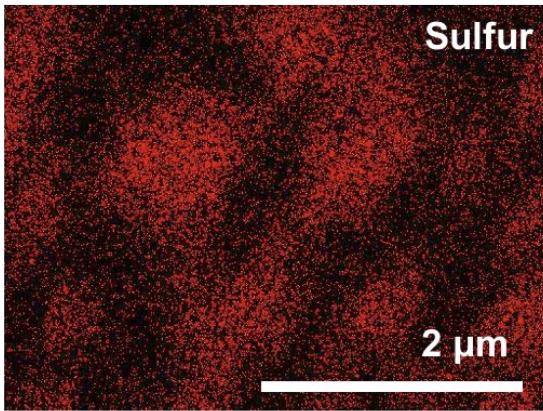
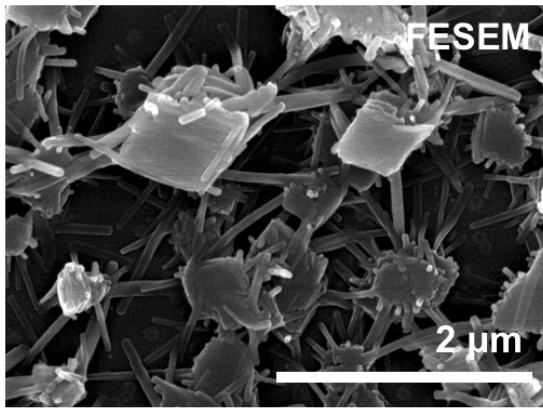


Figure S2. Size distribution data of (a) non-aged and aged TTF crystal or (b) non-aged and aged TCNQ crystal.

a



b

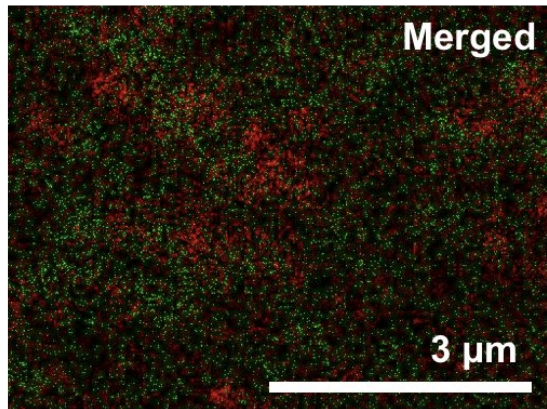
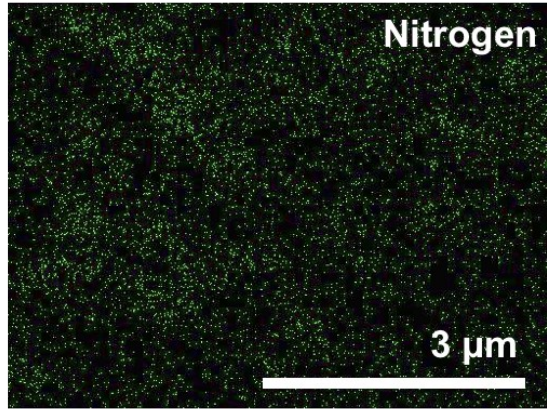
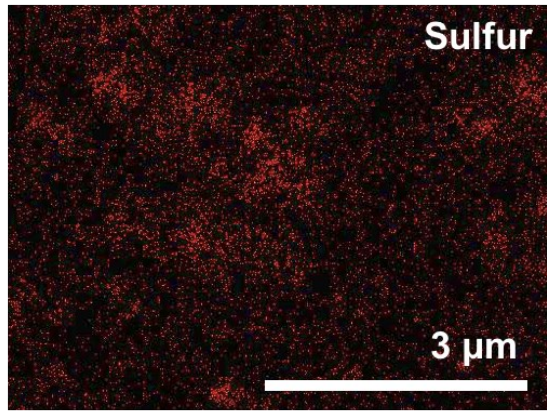
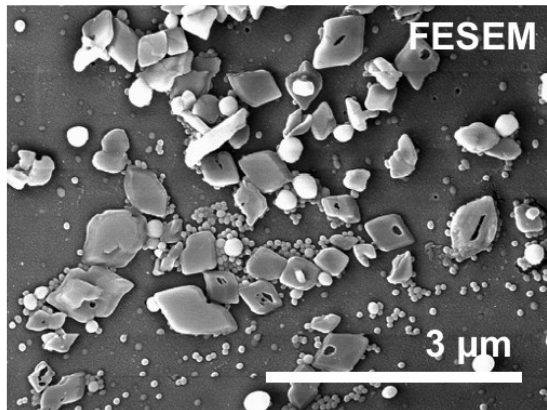


Figure S3. EDS mapping images of the (a) urchin-like morphology in non-aged mixture and (b) spherical/rhombic morphology in aged mixture. The sulfur and nitrogen atom which comes from TTF and TCNQ are mapped with red and green in the mapping images, respectively.

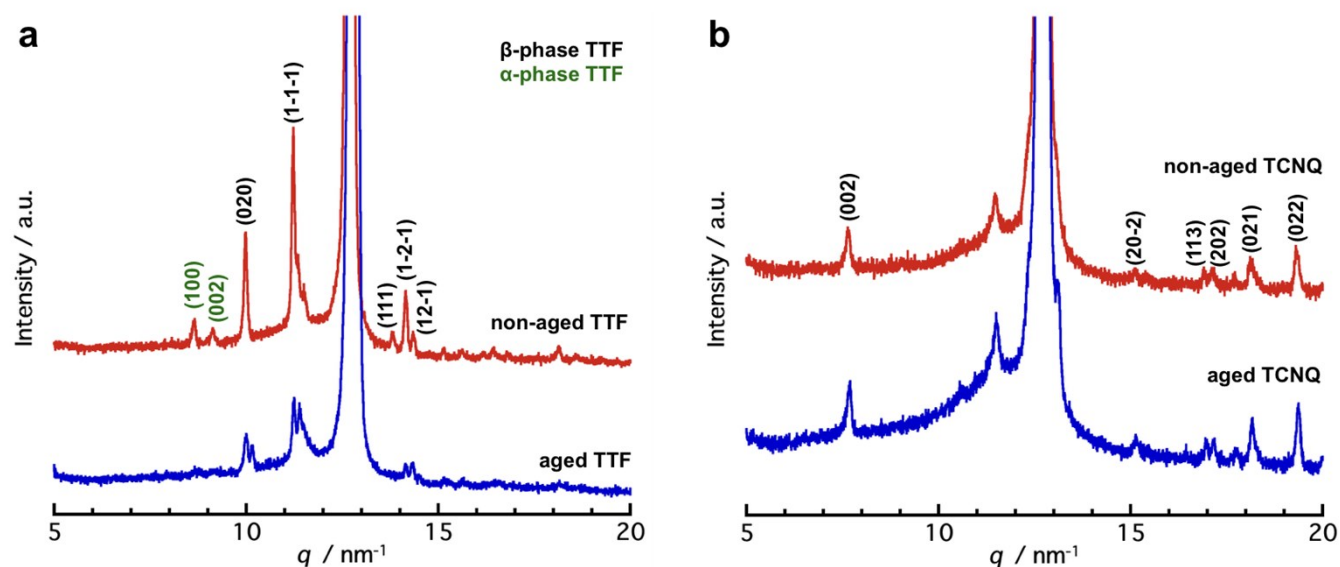


Figure S4. XRD patterns of (a) non-aged and aged TTF crystal or (b) non-aged and aged TCNQ crystal. The diffraction peaks at  $8.7 \text{ nm}^{-1}$  and  $9.1 \text{ nm}^{-1}$  in non-aged TTF crystals can be indexed to (100) and (002) plane of  $\alpha$ -phase TTF<sup>[1]</sup> which formed during the filtration process. The diffraction peaks are approximately  $11.5 \text{ nm}^{-1}$  and  $12.8 \text{ nm}^{-1}$  indicating the membrane filter.

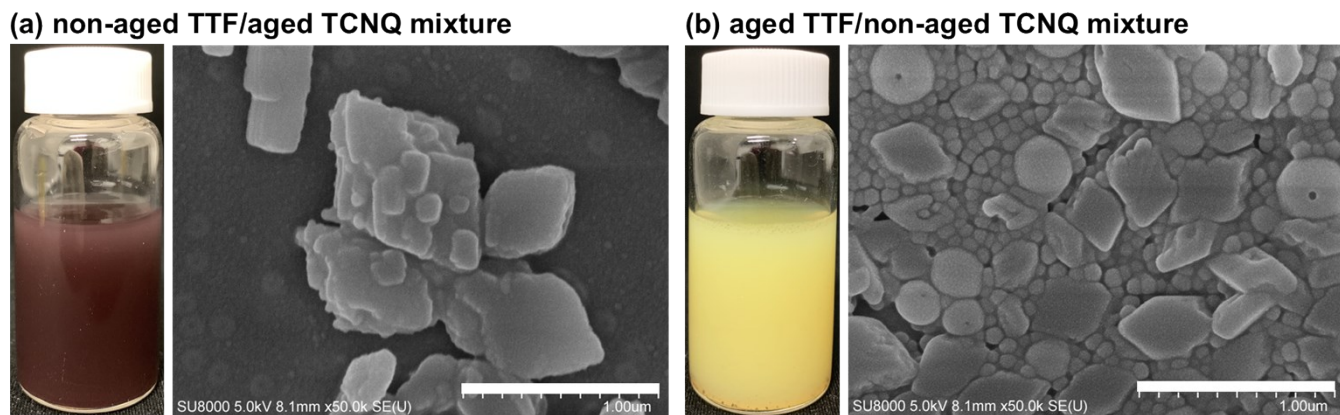


Figure S5. Photographs and FESEM images of the (a) non-aged TTF/aged TCNQ mixture and (b) aged TTF/non-aged TCNQ mixture.

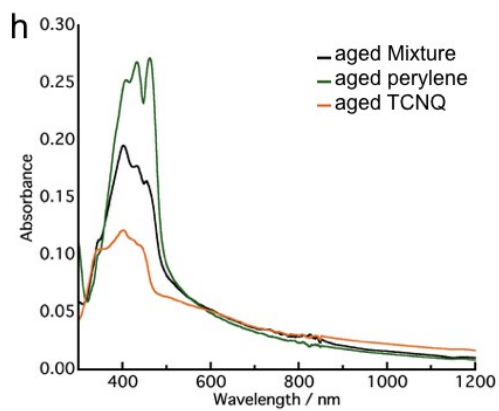
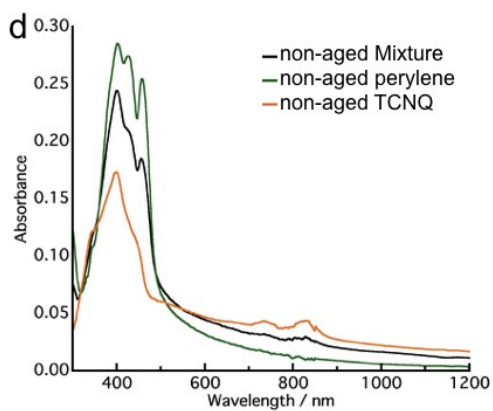
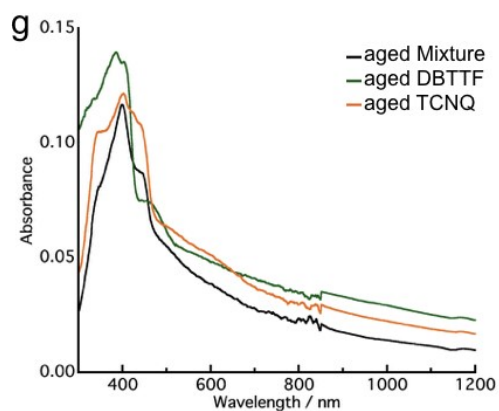
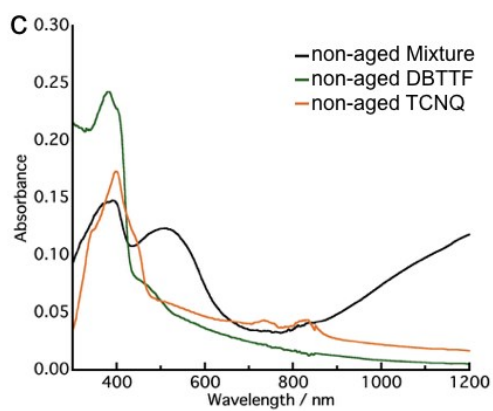
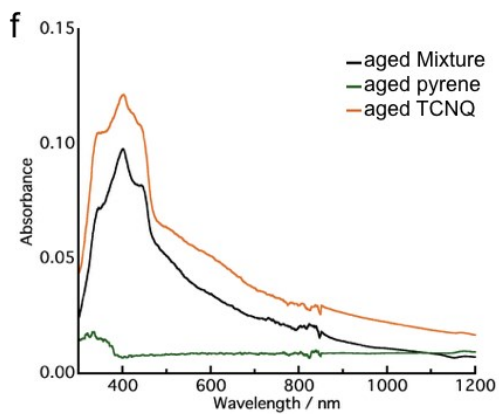
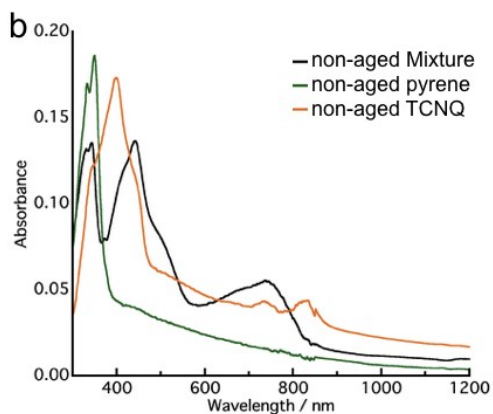
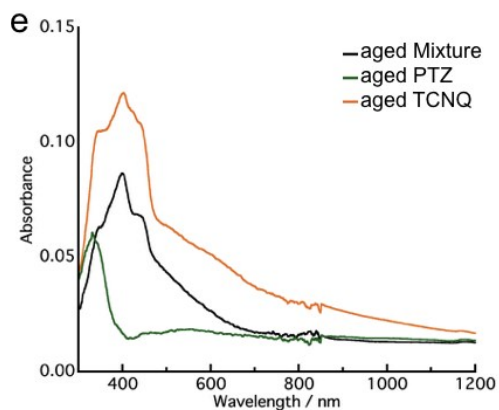
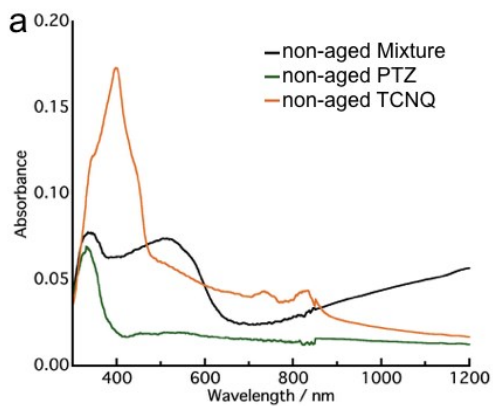


Figure S6. Absorption spectra of the non-aged mixture, aged mixture, and their respective constituent crystals. (a, e) PTZ/TCNQ mixture, (b, f) pyrene/TCNQ mixture, (c, g) DBTTF/TCNQ mixture, and (d, h) perylene/TCNQ mixture.

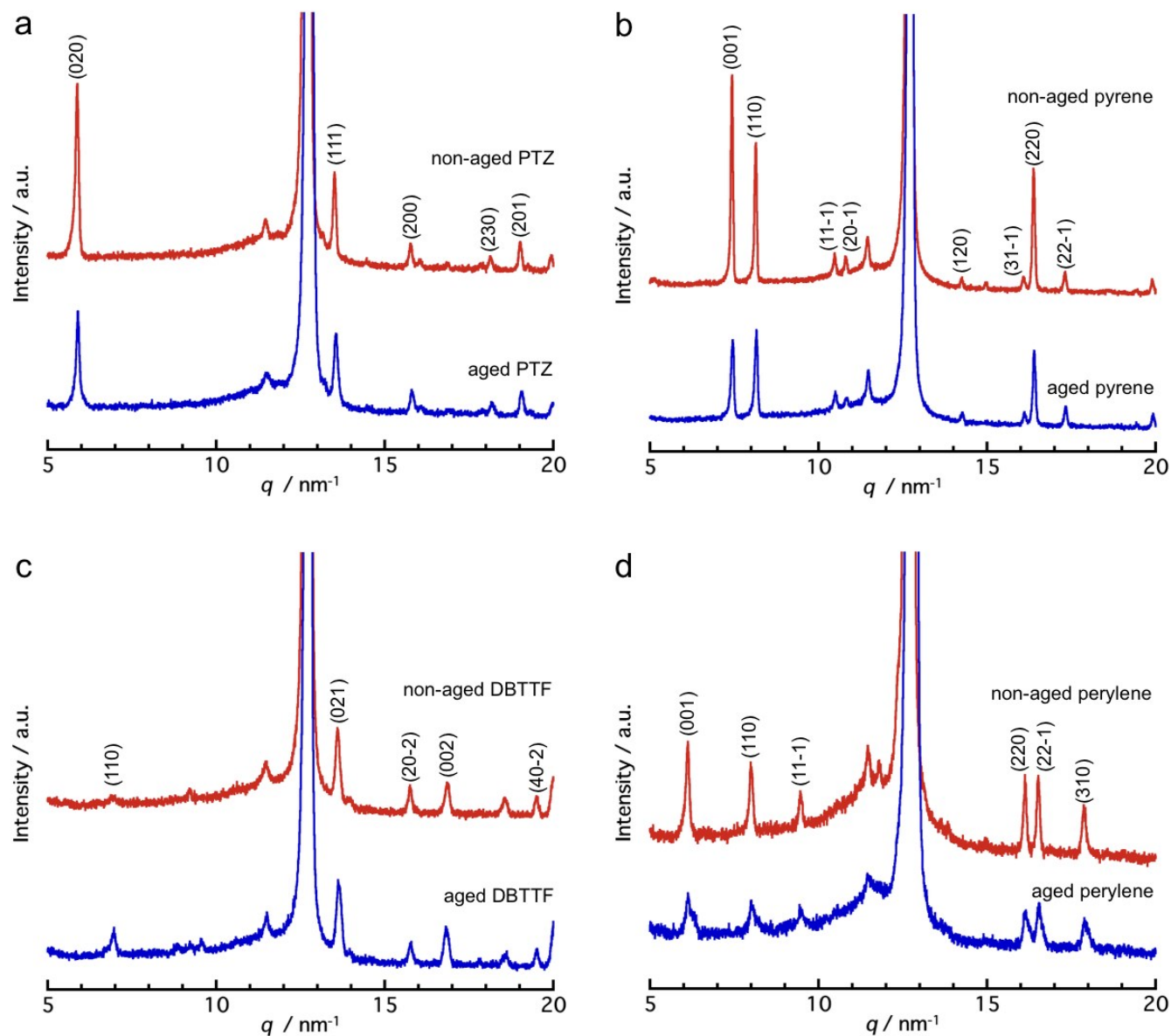


Figure S7. XRD patterns of (a) non-aged and aged PTZ crystal, (b) non-aged and aged pyrene crystal, (c) non-aged and aged DBTTF crystal, or (b) non-aged and aged perylene crystal. The diffraction peaks are approximately  $11.5 \text{ nm}^{-1}$  and  $12.8 \text{ nm}^{-1}$  indicating the membrane filter.

Table S1. Space group and cell parameter of PTZ, pyrene, DBTTF, and perylene crystal prepared by the reprecipitation method identified from the powder XRD pattern.

	Phenothiazine (PTZ)	Pyrene	Dibenzotetrathiafulvalene (DBTTF)	Perylene
Space group	<i>Pnma</i>	<i>C<sub>2</sub><sub>v</sub>/a</i>	<i>Cc</i>	<i>C<sub>2</sub><sub>v</sub>/a</i>
<i>a</i> / Å	7.916	13.649	15.154	11.277
<i>b</i> / Å	20.974	9.256	11.571	10.826
<i>c</i> / Å	5.894	8.470	8.027	10.263
<i>α</i> / deg	90	90	90	90
<i>β</i> / deg	90	100.28	111.637	100.55
<i>γ</i> / deg	90	90	90	90
Reference	2	3	4	5

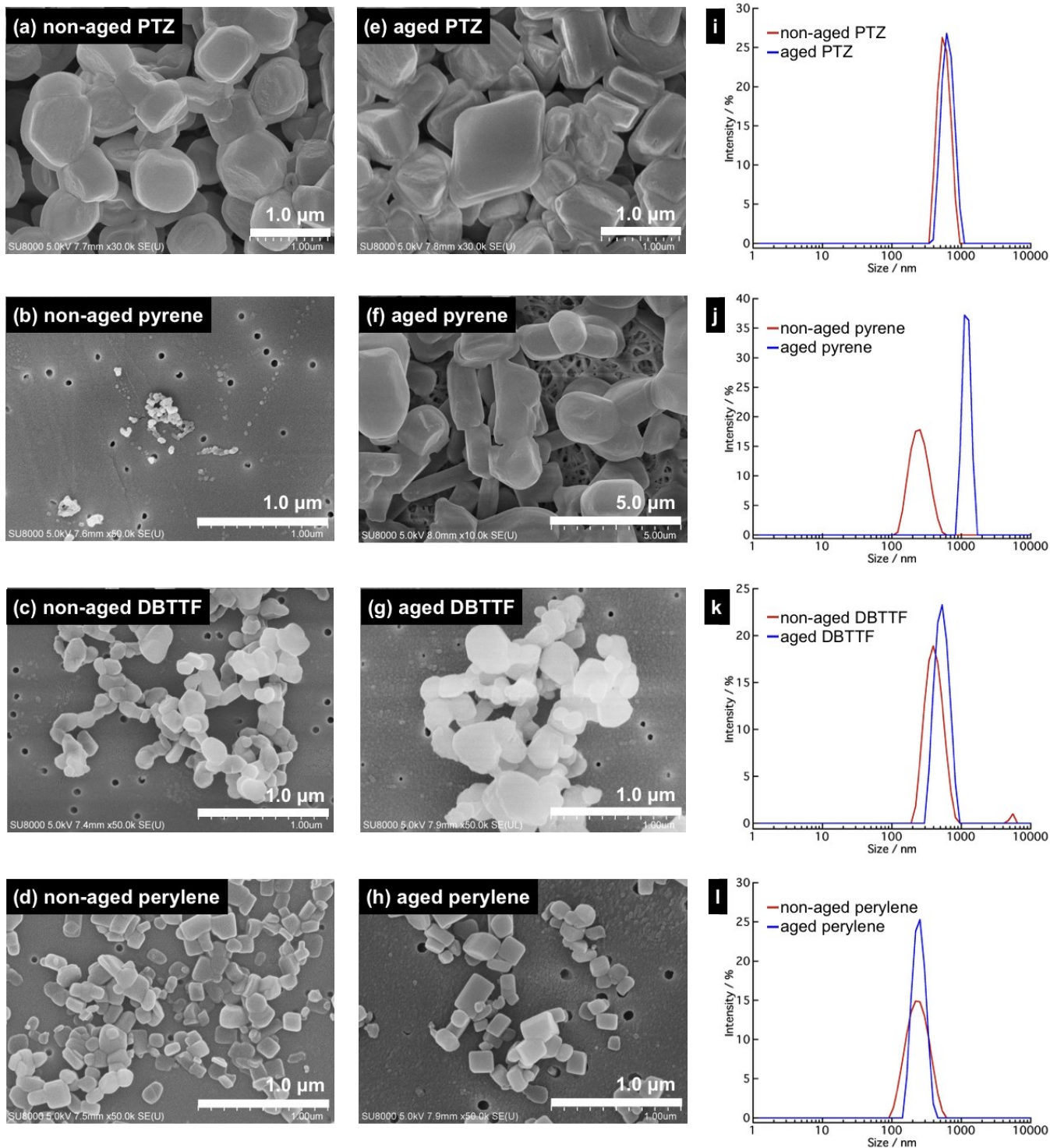


Figure S8. FESEM images of (a) non-aged PTZ crystal, (b) non-aged pyrene crystal, (c) non-aged DBTTF crystal, (d) non-aged perylene crystal, (e) aged PTZ crystal, (f) aged pyrene crystal, (g) aged DBTTF crystal, and (h) aged perylene crystal. (i-l) Size distribution data measured by DLS.



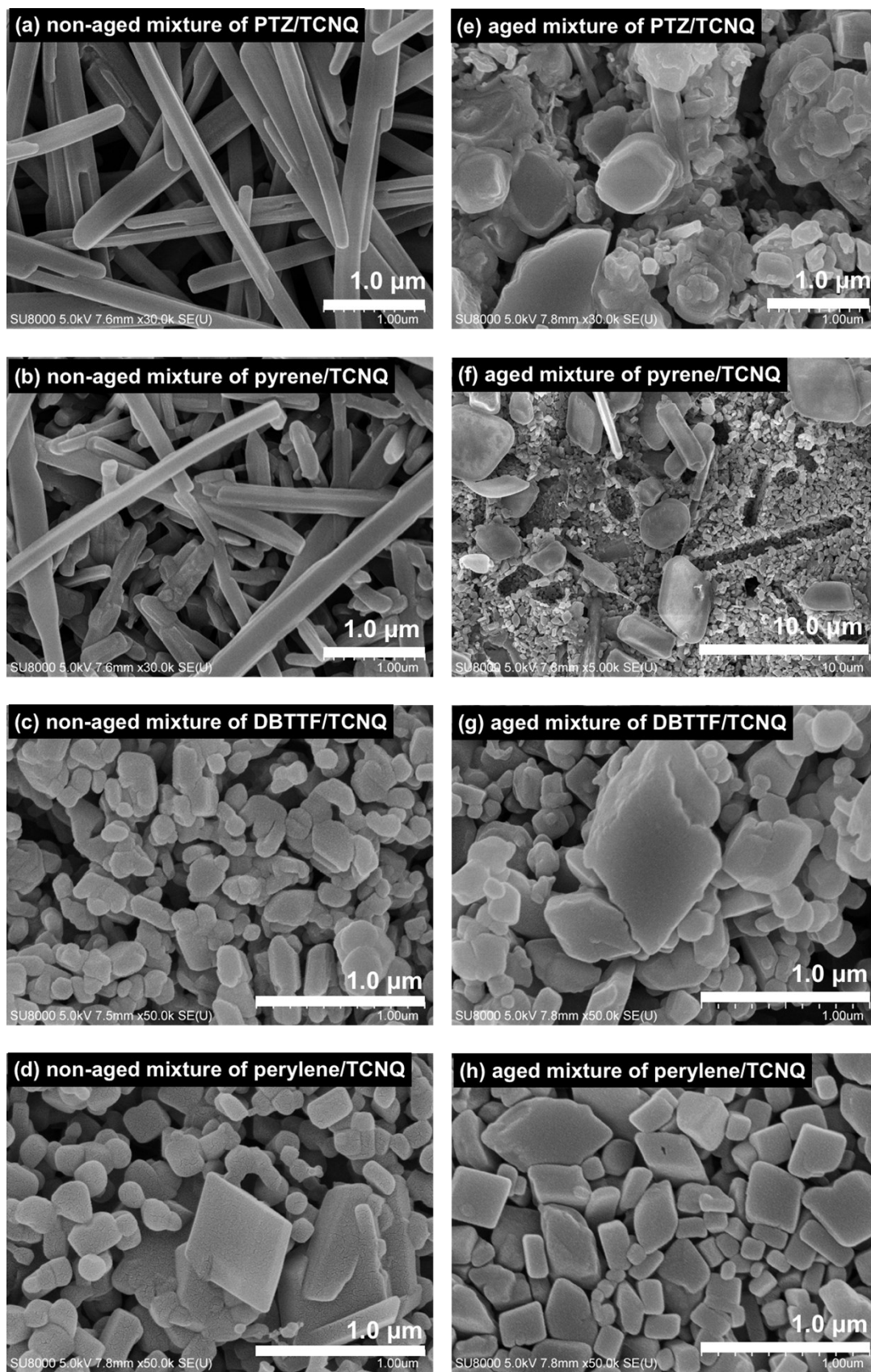


Figure S9. FESEM images of (a) non-aged mixture of PTZ/TCNQ, (b) non-aged mixture of pyrene/TCNQ, (c) non-aged mixture of DBTTF/TCNQ, (d) non-aged mixture of perylene/TCNQ, (e)

aged mixture of PTZTCNQ, (f) aged mixture of pyrene/TCNQ, (g) aged mixture of DBTTF/TCNQ, and (h) aged mixture of perylene/TCNQ.

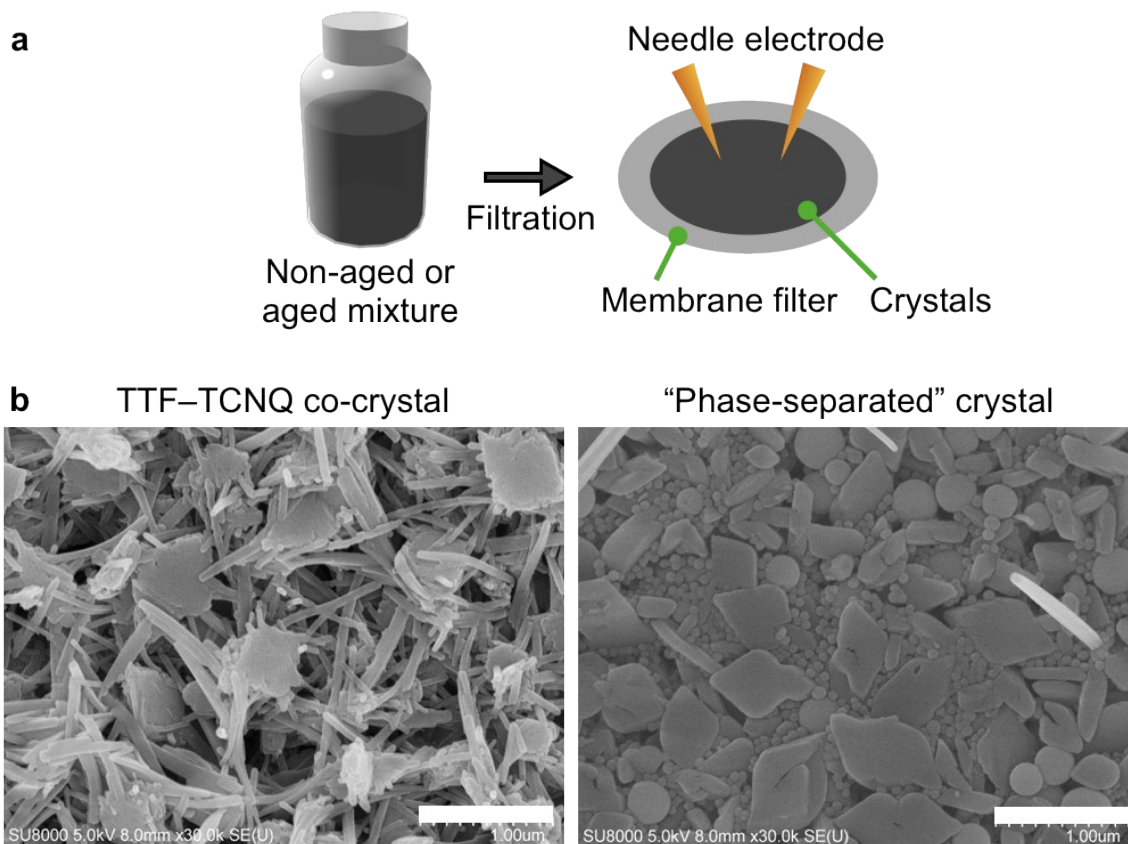


Figure S10. (a) Scheme of the preparation and measurement procedure. (b) FESEM images of the TTF-TCNQ co-crystal and “phase-separated” crystal on a filter. Scale bar, 1  $\mu\text{m}$ .

## REFERENCES

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