Electronic supplementary information

High-efficiency coupling of Ni^{2+} coordinated/uncoordinated pyridine N-COF self-supporting nanofilm for asymmetric supercapacitor

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The structure of $COF_{TAPB-BPY}$ by A-A stacking arrangement was determined by MS simulation. In Fig. S1, the $COF_{TAPB-BPY}$ by A-A stacking arrangement exhibits the pore size of 3.946 nm, and layer spacing of 0.351 nm.

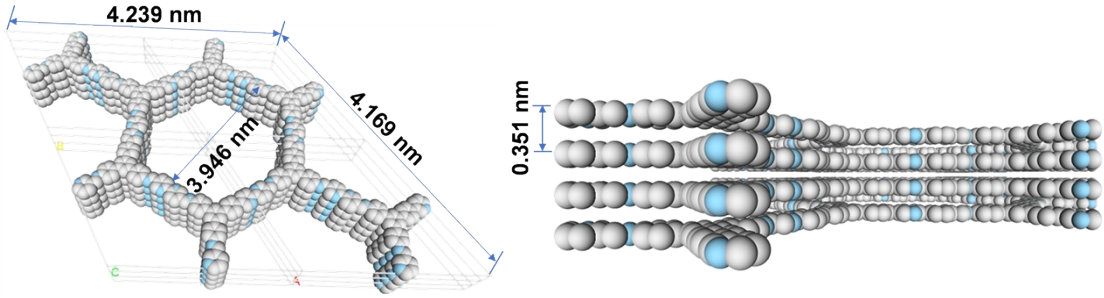

Fig. S1. AA stacking models of $COF_{TAPB-BPY}$

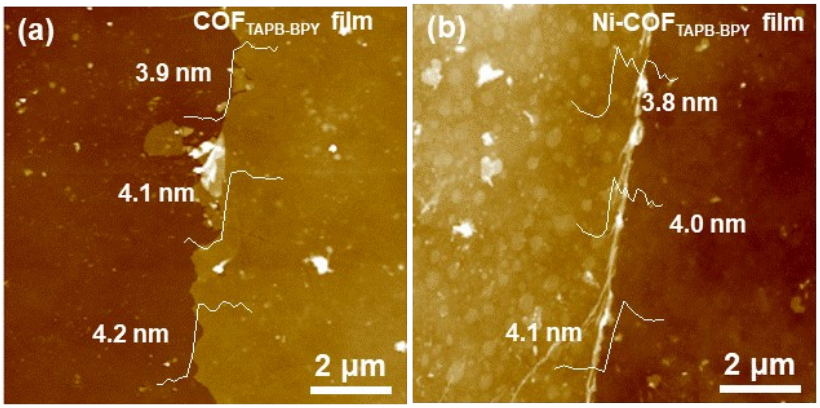

Fig. S2. AFM images of $COF_{TAPB-BPY}$ and Ni-$COF_{TAPB-BPY}$ film

In the experimental characterization, the pore size of $COF_{TAPB-BPY}$ and Ni-$COF_{TAPB-BPY}$ films are attempted to confirm by film XRD measurement. However, the diffraction signal of these COF nanofilms is weak to detect, owing to the ultra-low mass and ultra-thin thickness (only 4 nm), *that is*, these films are not appropriate for the traditional powder XRD test. Refer to other reported works, XRD result of counterpart bulk COF can be used to verify the crystal structure of COF nanofilms in reticular chemistry [S1-3]. Therefore, XRD result of bulk $COF_{TAPB-BPY}$ can be used to reflect the $COF_{TAPB-BPY}$ nanofilm. In Fig. S3, the diffraction peak at $2\theta = 2.2^{o}$ belongs to (100) crystal plane, which can be further calculate the pore size of $COF_{TAPB-BPY}$ about 3.9 nm.

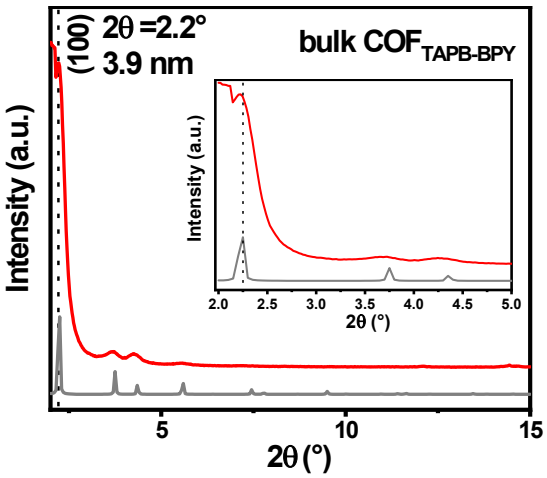

Fig. S3. XRD of bulk $COF_{TAPB-BPY}$

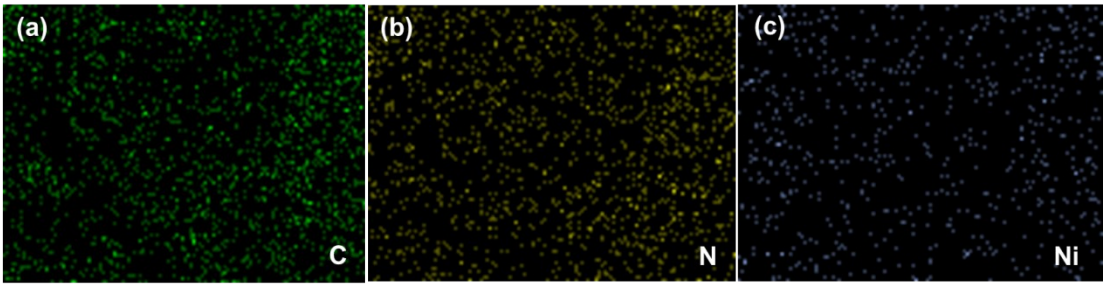

Fig. S4. Element mapping images of $COF_{TAPB-BPY}$ film

In the EIS plots of Fig. S5, at the low frequency region, the Ni-$COF_{TAPB-BPY}$ exhibits the obviously higher slope than $COF_{TAPB-BPY}$, indicating the enhanced attraction with electrolyte ions by adding Ni^{2+}.

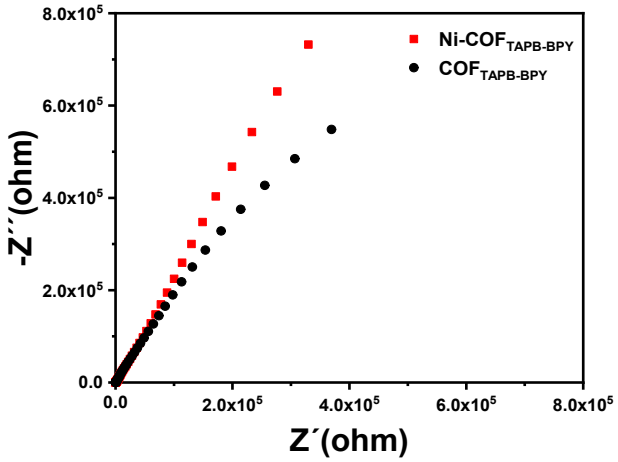

Fig. S5. EIS of $COF_{TAPB-BPY}$ and Ni-$COF_{TAPB-BPY}$ film

The $COF_{TAPB-BPY}$ film with 8 nm thickness was prepared by increasing monomer concentration as shown in the corresponding AFM image (Fig. S6a). Though electrochemical test, the 8 nm $COF_{TAPB-BPY}$ film owns the higher areal capacitance, but lower volumetric capacitance, compared to original 4 nm $COF_{TAPB-BPY}$ film (Fig. S6b).

There is no remarkable gap in these capacitance values at a current density, which may be owing to the $COF_{TAPB-BPY}$ skeleton dominated active sites for energy storage.

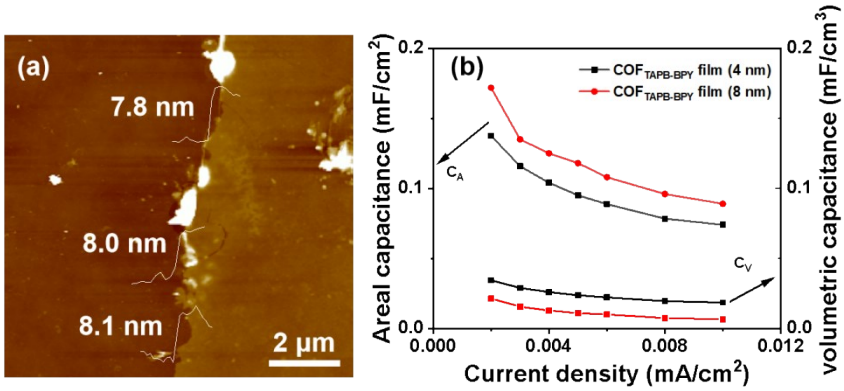

Fig. S6. AFM images of $COF_{TAPB-BPY}$ film with 8 nm thickness (a), capacitance performances of $COF_{TAPB-BPY}$ film with different thickness (b)

References

[S1] Z. Ou, B. Liang, Z. Liang, F. Tan, X. Dong, L. Gong, P. Zhao, H. Wang, Y. Zou, Y. Xia, X. Chen, W. Liu, H. Qi, U. Kaiser, Z. Zheng, *Journal of the American Chemical Society*, 2022, 144, 3233–3241.

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