Supporting Information

**Encapsulation of Dyes in Metal-Organic Frameworks and the Tunable Nonlinear Optical Properties**

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Figure S1. $^1$H NMR spectra (500 MHz) of the dye (a) MBPDC and (b) DMBPDC
Figure S2. TGA curves of bio-MOF-Me and bio-MOF-2Me

Figure S3. The BET surface area of bio-MOF-1 obtained from N\textsubscript{2} adsorption isotherm at 77 K

\[ S_{\text{BET}} = \frac{1}{\text{Slope} + \text{Intercept}} \times 22414 \times 6.023 \times 10^{23} \times 0.162 \times 10^{-18} = 1011.4 \text{ m}^2/\text{g} \]
Figure S4. The BET surface area of bio-MOF-Me obtained from N₂ adsorption isotherm at 77 K

\[
\frac{x}{V(1-x)} \approx 1.40633 \times 10^{-6} x + 0.00501
\]

\[R^2 = 0.9999\]

S\textsubscript{BET} = \frac{1}{(\text{Slope} + \text{Intercept})} \times 22414 \times 6.023 \times 10^{23} \times 0.162 \times 10^{-18} = 868.7 \text{ m}^2/\text{g}

Figure S5. The BET surface area of bio-MOF-2Me obtained from N₂ adsorption isotherm at 77 K

\[
\frac{x}{V(1-x)} \approx 1.47718 \times 10^{-6} x + 0.00584
\]

\[R^2 = 0.9999\]

S\textsubscript{BET} = \frac{1}{(\text{Slope} + \text{Intercept})} \times 22414 \times 6.023 \times 10^{23} \times 0.162 \times 10^{-18} = 745.2 \text{ m}^2/\text{g}
Figure S6. Polarized fluorescence spectra of (a) bio-MOF-1$\supset$DM-1 and (b) bio-MOF-1$\supset$DP-1

Figure S7. The polarized absorption spectra of (a) bio-MOF-1$\supset$DM-1 and (b) bio-MOF-1$\supset$DP-1, and the corresponding dichroic ratio (black dots).
Figure S8. Polarized absorption spectra of (a) bio-MOF-1$\supset$DM-1, (b) bio-MOF-1$\supset$DM-4, (c) bio-MOF-1$\supset$DM-7 and (d) bio-MOF-1$\supset$DM-7. The legend means the angle between the polarizer and the long axis of crystal.
Figure S9. Polarized absorption spectra of (a) bio-MOF-Me$\supset$DM-1, (b) bio-MOF-Me$\supset$DM-4, (c) bio-MOF-Me$\supset$DM-7 and (d) bio-MOF-Me$\supset$DM-7.
Figure S10. Polarized absorption spectra of (a) bio-MOF-2Me⊃DM-1, (b) bio-MOF-2Me⊃DM-4, (c) bio-MOF-2Me⊃DM-7 and (d) bio-MOF-2Me⊃DM-7.
Figure S11. Polarized absorption spectra of (a) bio-MOF-1\(\rightarrow\)DP-1, (b) bio-MOF-1\(\rightarrow\)DP-4, (c) bio-MOF-1\(\rightarrow\)DP-7 and (d) bio-MOF-1\(\rightarrow\)DP-7.
Figure S12. Polarized absorption spectra of (a) bio-MOF-Me\(\rightarrow\)DP-1, (b) bio-MOF-Me\(\rightarrow\)DP-4, (c) bio-MOF-Me\(\rightarrow\)DP-7 and (d) bio-MOF-Me\(\rightarrow\)DP-7.
Figure S13. Effect of reaction time (the right one describes the pseudo-second-order kinetic) on DM-1 adsorption onto (a) bio-MOF-1, (b) bio-MOF-Me and (c) bio-MOF-2Me
Figure S14. SHG spectra of DM-1 crystals