

Electronic Supplementary Information (ESI)

ESI 1 AAAA-stacked layers structures containing decanuclear connecting nods with the dimension of $21.602(4) \times 16.973(5)$ Å² for **1**. (pydc and bpy ligands are both substituted by a connection between metals)



ESI 2 The ladder-like chains with a same mesh size in 1



ESI 3 AAAA-stacked layers structures interlinked by the ladder-like chains for **1** (pydc and bpy ligands are both substituted by a connection between metals)



ESI 4 AAAA-stacked layers structures containing decanuclear connecting nods with the dimension of $21.757(5) \times 17.163(6)$ Å² for **2** (pydc and bpy ligands are both substituted by a connection between metals)



ESI 5 The ladder-like chains with a same mesh size in 2



ESI 6 AAAA-stacked layers structures interlinked by the ladder-like chains for **2** (pydc and bpy ligands are both substituted by a connection between metals)



ESI 7 ABAB-stacked bilayers structures containing decanuclear connecting nods with the dimension of $21.587(4) \times 16.993(3)$ Å² for **3** (pydc and bpy ligands are both substituted by a connection between metals)



ESI 8 The ladder-like chains with a different mesh size in 3



ESI 9 ABAB-stacked bilayers structures interlinked by the ladder-like chains for **3**. (pydc and bpy ligands are both substituted by a connection between metals)

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ESI 10 ABAB-stacked bilayers structures containing decanuclear connecting nods with the dimension of $21.570(3) \times 16.976(3)$ Å² for **4** (pydc and bpy ligands are both substituted by a connection between metals)



ESI 11 The ladder-like chains with a different mesh size in 4



ESI 12 ABAB-stacked bilayers structures interlinked by the ladder-like chains for **4**. (pydc and bpy ligands are both substituted by a connection between metals)

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ESI 13 ABAB-stacked bilayers structures containing decanuclear connecting nods with the dimension of $21.618(8) \times 16.968(7)$ Å² for **5** (pydc and bpy ligands are both substituted by a connection between metals)



ESI 14 The ladder-like chains with a different mesh size in 5

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ESI 15 ABAB-stacked bilayers structures interlinked by the ladder-like chains for **5**. (pydc and bpy ligands are both substituted by a connection between metals)



ESI 16 FT-IR spectra for 1



ESI 17 Solid-state excitation spectrum for 2 at room temperature



ESI 18 Solid-state excitation spectrum for **4** at room temperature



ESI 19 Simulated and experimental powder X-ray diffraction patterns for 2.