## ESI

## Hydrogen Bonded Coassembly of Aromatic Amino Acids and Bipyridines that Serves as Sacrificial Templates in Superstructure Formation

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Figure S1. Optical microscopy images. a) Asp-BPE coassembly in MeOH ( $C_{Asp}$  =

66.7mM). b) Gly-BP coassembly in acetone ( $C_{Gly} = 100 \text{ mM}$ ). c) Gly-BPE coassembly in acetone ( $C_{Gly} = 100 \text{ mM}$ ). d) Ala-bpe coassembly in acetone ( $C_{Ala} = 100 \text{ mM}$ ). e) Asp-BPE coassembly in acetone ( $C_{Asp} = 100 \text{ mM}$ ). f) Gly-BP coassembly in EtOH ( $C_{Gly} = 66.7 \text{ mM}$ ). g) Gly-BPE coassembly in EtOH ( $C_{Gly} = 66.7 \text{ mM}$ ). h) Asp-BPE coassembly in EtOH ( $C_{Asp} = 66.7 \text{ mM}$ ). i) Gly-BPE coassembly in MeCN ( $C_{Gly} = 15 \text{ mM}$ ). j) Asp-BPE coassembly in MeCN ( $C_{Asp} = 66.7 \text{ mM}$ ). k) Gly-BPE coassembly in THF ( $C_{Gly} = 66.7 \text{ mM}$ ). Molar ratio between aromatic amino acids and bipyridine was fixed at 2:1.



**Figure S2.** Polarized optical microscopic images. a) Asp-BPE coassembly in MeOH  $(C_{Asp} = 66.7 \text{mM})$ . b) Gly-BP coassembly in acetone  $(C_{Gly} = 100 \text{ mM})$ . c) Gly-BPE coassembly in acetone  $(C_{Gly} = 100 \text{ mM})$ . d) Ala-bpe coassembly in acetone  $(C_{Ala} = 100 \text{ mM})$ . e) Asp-BPE coassembly in acetone  $(C_{Asp} = 100 \text{ mM})$ . f) Gly-BP coassembly in EtOH  $(C_{Gly} = 66.7 \text{ mM})$ . g) Gly-BPE coassembly in EtOH  $(C_{Gly} = 66.7 \text{ mM})$ . g) Gly-BPE coassembly in EtOH  $(C_{Gly} = 66.7 \text{ mM})$ . i) Gly-BPE coassembly in MeCN  $(C_{Gly} = 15 \text{ mM})$ . j) Asp-BPE coassembly in MeCN  $(C_{Asp} = 66.7 \text{ mM})$ . k) Gly-BPE coassembly in THF  $(C_{Gly} = 66.7 \text{ mM})$ . Molar ratio between aromatic amino

acids and bipyridine was fixed at 2:1.



Figure S3. SEM image of Asp-BPE coassembly in MeOH (66.7mM:33.3mM).



Figure S4. Stacking of Gly-BP in crystal structure.



**Figure S5.** XRD pattern comparison of Gly-BP in MeOH ( $C_{Gly} = 66.7$ mM), acetone ( $C_{Gly} = 200$  mM), EtOH ( $C_{Gly} = 66.7$ mM). Molar ratio between aromatic amino acids and bipyridine was fixed at 2:1.



Figure S6. XRD patterns of Asp-BPE coassemblies in MeOH ( $C_{Asp} = 66.7$  mM),

acetone ( $C_{Asp} = 100 \text{ mM}$ ), EtOH ( $C_{Asp} = 66.7 \text{ mM}$ ) and MeCN ( $C_{Asp} = 66.7 \text{ mM}$ ). Molar ratio between aromatic amino acids and bipyridine was fixed at 2:1.



Figure S7. SEM images of Co-BP coassembly (50mM:50mM) in methanol.



Figure S8. SEM images of Ni-BP coassembly (50mM:50mM) in methanol.



Figure S9. SEM images of Cu-BP coassembly (50mM:50mM) in methanol.



Figure S10. SEM images of Zn-BP coassembly (50mM:50mM) in methanol.



Figure S11. SEM images of Ni-BPE coassembly (50mM:50mM) in methanol.



Figure S12. SEM images of Zn-BPE coassembly (50mM:50mM) in methanol.



Figure S13. SEM image of Ni(II) treated Gly-BP (66.7 mM:33.3 mM) coassemblies  $(C_{Ni} = 25 \text{ mM}).$ 



Figure S14. SEM image of Ni(II) treated Gly-BPE (66.7 mM:33.3 mM) coassemblies  $(C_{Ni} = 11.8 \text{ mM for a-c}; C_{Ni} = 6.25 \text{ mM for d-f}).$ 



Figure S15. SEM image of Cu(II) treated Gly-BP (66.7 mM:33.3 mM) coassemblies

 $(C_{Ni} = 11.8 \text{ mM}).$ 



Figure S16. SEM image of Co(II) treated Gly-BP (66.7 mM:33.3 mM) coassemblies  $(C_{Co} = 11.8 \text{ mM for a-c}; C_{Co} = 9.1 \text{ mM for d-f}).$ 



**Figure S17.** FT-IR spectra of Zn-treated Gly-BP samples with different  $Zn^{2+}$  concentrations. (Zn-BP 50 mM:50 mM; Gly-BP 66.7 mM:33.3 mM)