

Support information

In Situ template Generation via *N*-alkylation in the Syntheses of Open-framework Zinc

Phosphites and Phosphate

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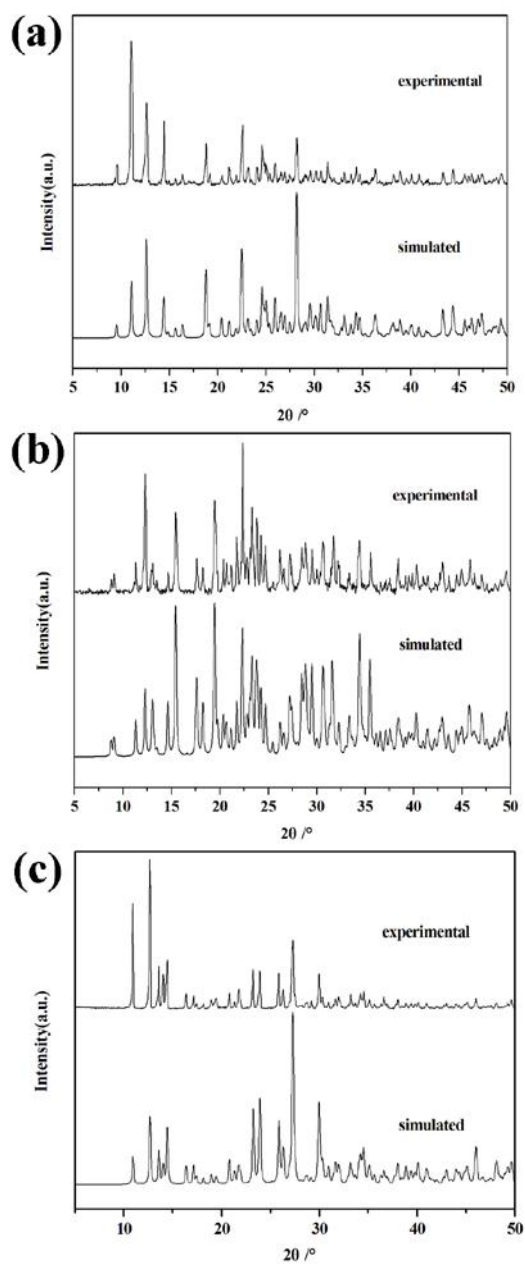


Figure S1. Simulated and experimental X-ray powder diffraction patterns of compounds I (a), II (b) and III (c).

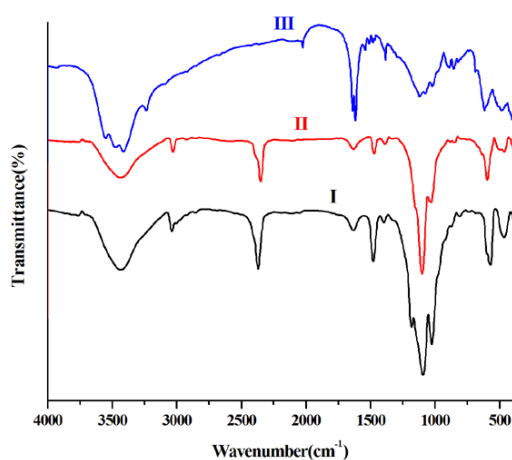


Figure S2. IR spectra of compounds I-III.

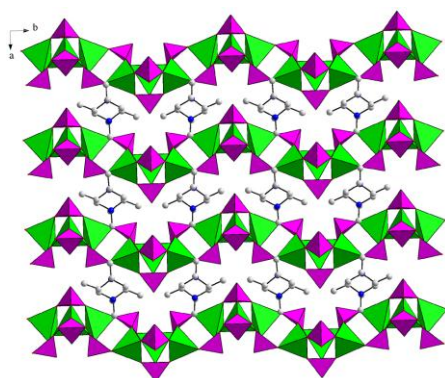


Figure S3. View of the structure of I along the [001] direction, showing the inorganic layers intercalated with organic pmpip²⁺ cations. Color code: ZnO₄ tetrahedra, green; HPO₃ pseudopyramids, purple.

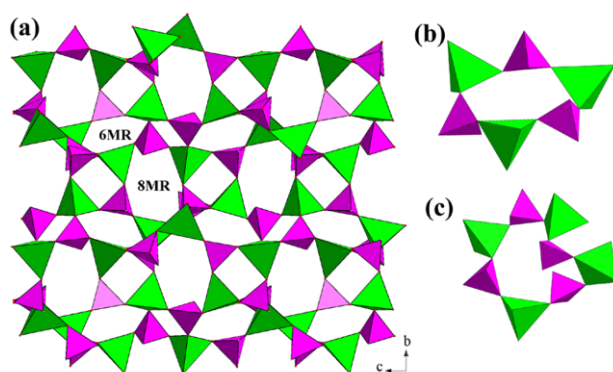


Figure S4. View of the structure of compound II along the [100] direction, showing the 6- and 8-ring channels. Color code: ZnO₄ tetrahedra, green; HPO₃ pseudopyramids, purple.

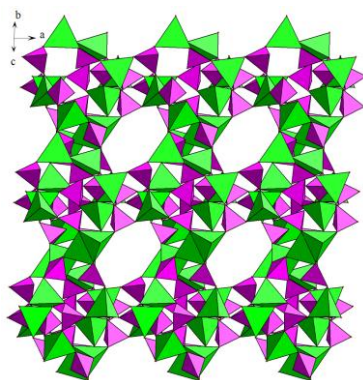


Figure S5. View of the structure of **II** along the [011] direction, showing the puckered 12-ring channels. Color code: ZnO₄ tetrahedra, green; HPO₃ pseudopyramids, purple.

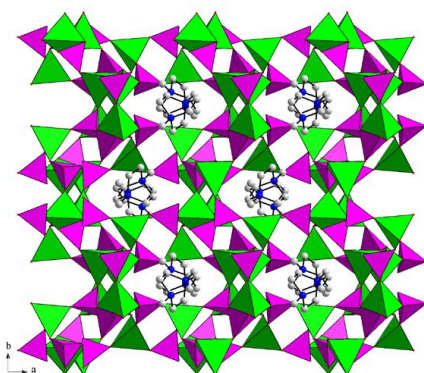


Figure S6. View of the structure of **II** along the [001] direction with dmdabco²⁺ cations residing in the 10-ring channels. Color code: ZnO₄ tetrahedra, green; HPO₃ pseudopyramids, purple.

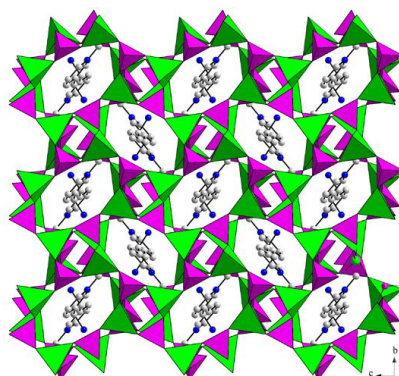


Figure S7. View of the structure of **III** along the [100] direction with mampy²⁺ cations residing in the 8-ring channels. Color code: ZnO₄ tetrahedra, green; HPO₃ pseudopyramids, purple.