Electronic Supplementary Information

A solid-state synthetic strategy toward nickel-based bimetallic interstitial compounds

$(MNi_3C_x, M = Zn, In, Ga)$

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Fig. S1. The PXRD pattern of the as-prepared Ni-Zn precipitate from the co-precipitation method. Vertical lines are the reference standards.



Fig. S2. The PXRD patterns of products synthesized by the solid-state reaction between the metal hydroxides of Ni and In, and melamine under different synthetic parameters: **a**, Weight ratios of melamine:Ni-In precursors = 3:1-7:1, 973 K, 4 h in H₂, **b**. Annealing temperatures of 573-1073 K, weight ratios of melamine:Ni-In precursors = 5:1, 4 h in H₂. Inset in **b** shows the zoom in at 28- $35^{\circ} 2\theta$. Vertical lines are the reference standards.



Fig. S3. The PXRD patterns of products synthesized by the solid-state reaction between the metal hydroxides of Ni and Ga, and melamine under different synthetic parameters: **a**, Weight ratios of melamine:Ni-Ga precursors = 3:1-15:1, 973 K, 4 h in H₂, **b**. Annealing temperatures of 573-973 K, weight ratios of melamine:Ni-In precursors = 15:1, 4 h in H₂. Vertical lines are the reference standards.



Fig. S4. The PXRD patterns of products synthesized by annealing the co-precipitation-derived precursors in H_2 for 4 h at 773 and 973 K. Vertical lines are the reference standards.



Fig. S5. a, The N_2 sorption isotherms of the developed catalysts and b, the key porosity parameters.



Fig. S6. The PXRD patterns of the catalysts after the testing in DMO hydrogenation.



Fig. S7. The PXRD patterns of the as-prepared $ZnNi_3C_{0.7}$ and after oxidation in air at different temperatures for 3 h.



Fig. S8. Catalytic performance of the as-prepared and oxidized $ZnNi_3C_{0.7}$ in the selective hydrogenation of DMO. Conditions: catalyst weight 0.5 g, liquid hourly space velocity based on DMO of 0.55 h⁻¹, molar H₂:DMO of 70, and P = 2.5 MPa. **a**, DMO conversion, **b**. MG selectivity.