Supplementary Information

Anion and pH induced spontaneous resolution of Δ - and Λ -[M(H₂Biim)₃]SO₄

 $(M=Ru^{2+}, Co^{2+}, Ni^{2+}, Mn^{2+}, Fe^{2+}, and Zn^{2+})$ enantiomers

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Fig. S1. Photographs showing the shapes of Λ -2 (left) and Δ -2 (right) under a microscope.



Fig. S2. TGA of *rac*-2·5H₂O.



Fig. S3. Solid state CD spectra of a large single crystal of *Λ*-2 (blue line), *Δ*-2 (red line), *rac*-2 (black line), and UV-vis spectra of 2 (green line) diluted with KCl.



Fig. S4. Powder XRD patterns of the simulations based on the single-crystal analyses and assynthesized samples of 2.



Fig. S5. Powder XRD patterns of the simulations based on the single-crystal analyses and assynthesized samples of 3 form different synthesis methods.



Fig. S6. Powder XRD patterns of the simulations based on the single-crystal analyses and assynthesized samples of 4 form different synthesis methods.



Fig. S7. Powder XRD patterns of the simulations based on the single-crystal analyses and assynthesized samples of **5** form different synthesis methods.



Fig. S8. Powder XRD patterns of the simulations based on the single-crystal analyses and assynthesized samples of 6 form different synthesis methods.



Fig. S9. Solid state CD spectra of a large single crystal of Λ -3 (blue line), Δ -3 (red line), 3 (black line),

and UV-vis spectra of 3 (green line) diluted with KCl.



Fig. S10. Solid state CD spectra of a large single crystal of Λ -4 (blue line), Δ -4 (red line), 4 (black line), and UV-vis spectra of 4 (green line) diluted with KCl.



Fig. S11. Solid state CD spectra of a large single crystal of Λ -5 (blue line), Δ -5 (red line), 5 (black line),





Fig. S12. Solid state CD spectra of a large single crystal of Λ -6 (blue line), Δ -6 (red line), 6 (black line), and UV-vis spectra of 6 (green line) diluted with KCl.