

Figure S1. Powder XRD patterns of compounds **R-Zn** (red line) and **S-Zn** (blue line). The pattern simulated from single crystal data of **S-Zn** is given for comparison.

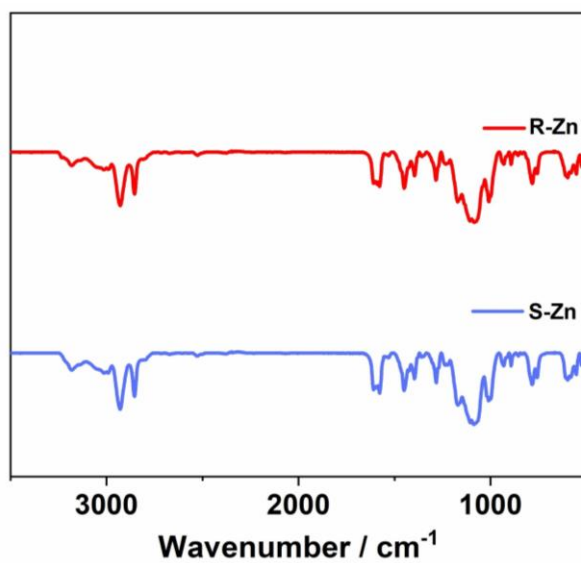


Figure S2. IR spectra of compound **S-Zn** and **R-Zn**.

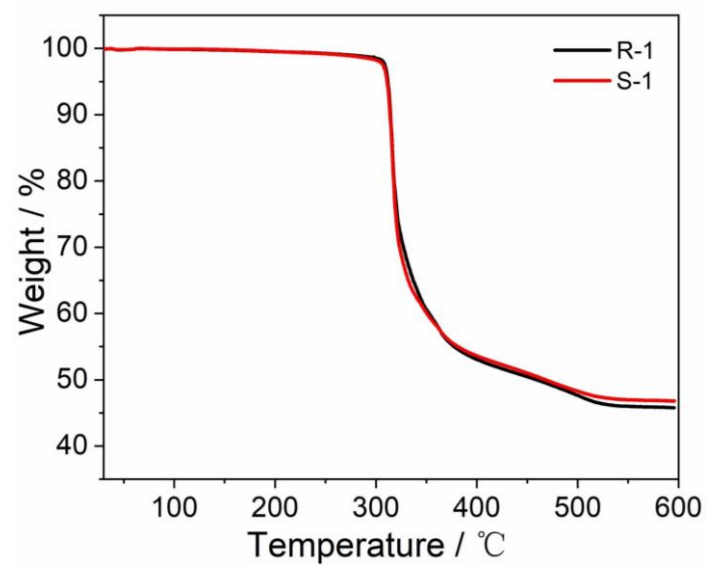


Figure S3. TG analyses of S-, R-Zn₈(S-cyampH)₈Cl₈ (**S-**, **R-Zn**). No weigh loss is observed below 350 °C.

Table S1 Crystallographic data for **S-Zn**.

Empirical formula	C ₇₂ H ₁₅₂ Cl ₈ N ₈ O ₂₄ P ₈ Zn ₈
<i>M</i> _r	2568.31
crystal system	triclinic
space group	<i>P</i> 1
<i>a</i> (Å)	8.6267(9)
<i>b</i> (Å)	15.4971(14)
<i>c</i> (Å)	20.2286(18)
α (°)	87.533(3)
β (°)	89.591(3)
γ (°)	74.367(3)
<i>V</i> (Å ³)	2601.9(4)
<i>Z</i>	1
<i>D</i> _c (g cm ⁻³)	1.638
<i>M</i> (mm ⁻¹)	2.208
<i>F</i> (000)	1326
<i>R</i> (int)	0.060
GoF on <i>F</i> ²	1.046
<i>R</i> ₁ , <i>wR</i> ₂ [<i>I</i> > 2 σ (<i>I</i>)] ^a	0.0862, 0.2216
<i>R</i> ₁ , <i>wR</i> ₂ (all data) ^a	0.0948, 0.2287
Flack parameter	0.15(3)
CCDC	2401522

^a $R_1 = \frac{\sum ||F_0| - |F_c||}{\sum |F_0|}$, $wR_2 = [\frac{\sum w(F_0^2 - F_c^2)^2}{\sum w(F_0^2)^2}]^{1/2}$.

Table S2. Main bond length [Å] and bond angle [°] for compound **S-Zn**.

Zn1-Cl1	2.248(64)	P6-O17	1.542(13)	Cl8-Zn8-O9B	109.7(4)
Zn1-O1	1.919(15)	P6-O18	1.544(13)	O21-Zn8-O22	110.0(6)
Zn1-O4	1.956(16)	P7-C55	1.833(17)	O21-Zn8-O9B	111.2(6)
Zn1-O12A	1.975(13)	P7-O19	1.514(16)	O22-Zn8-O9B	98.3(6)
Zn2-Cl2	2.238(59)	P7-O20	1.508(15)	C1-P1-O1	104(1)
Zn2-O2	1.925(17)	P7-O21	1.532(13)	C1-P1-O2	112(1)
Zn2-O5	1.931(14)	P8-C64	1.803(21)	C1-P1-O3	105(1)
Zn2-O18C	1.968(12)	P8-O22	1.488(16)	O1-P1-O2	114.1(9)
Zn3-Cl3	2.238(6)	P8-O23	1.518(16)	O1-P1-O3	109.5(8)
Zn3-O6	1.929(13)	P8-O24	1.522(13)	O2-P1-O3	111.8(8)
Zn3-O7	1.980(17)	Cl1-Zn1-O1	109.5(5)	C10-P2-O4	103(1)
Zn3-O10	1.953(13)	Cl1-Zn1-O4	122.8(5)	C10-P2-O5	106(1)
Zn4-Cl4	2.203(65)	Cl1-Zn1-O12A	105.3(5)	C10-P2-O6	106(1)
Zn4-O8	1.941(13)	O1-Zn1-O4	101.3(6)	C37-P5-O13	106.4(9)
Zn4-O11	1.920(17)	O1-Zn1-O12A	111.0(6)	C37-P5-O14	103.6(9)
Zn4-O23D	1.992(12)	O4-Zn1-O12A	106.8(6)	C37-P5-O15	105.9(9)
Zn5-Cl5	2.228(63)	Cl2-Zn2-O2	107.4(5)	O13-P5-O14	114.0(8)
Zn5-O3	1.945(13)	Cl2-Zn2-O5	115.4(5)	O13-P5-O15	108.8(7)
Zn5-O13	1.955(12)	Cl2-Zn2-O18C	113.8(4)	O14-P5-O15	117.0(8)
Zn5-O16	1.901(17)	O2-Zn2-O5	105.7(6)	C46-P6-O16	105.1(9)
Zn6-Cl6	2.247(49)	O2-Zn2-O18C	98.2(6)	C46-P6-O17	104.2(9)
Zn6-O14	1.961(16)	O5-Zn2-O18C	114.3(6)	C46-P6-O18	107.1(9)
Zn6-O17	1.945(14)	Cl3-Zn3-O6	112.2(5)	O16-P6-O17	114.1(8)
Zn6-O20	1.952(13)	Cl3-Zn3-O7	111.8(5)	O16-P6-O18	115.0(8)
Zn7-Cl7	2.246(55)	Cl3-Zn3-O10	114.4(5)	O17-P6-O18	110.5(8)
Zn7-O19	1.938(13)	O6-Zn3-O7	105.7(6)	C55-P7-O19	105.3(8)
Zn7-O24	1.974(16)	O6-Zn3-O10	102.5(6)	C55-P7-O20	106.1(8)
Zn7-O15	1.956(13)	O7-Zn3-O10	109.6(6)	C55-P7-O21	106.9(8)
Zn8-Cl8	2.232(64)	Cl4-Zn4-O8	111.7(5)	O19-P7-O20	115.4(8)
Zn8-O21	1.980(15)	Cl4-Zn4-O11	112.4(5)	O19-P7-O21	113.7(8)
Zn8-O22	1.942(13)	Cl4-Zn4-O23D	115.1(4)	O20-P7-O21	108.7(8)
Zn8-O9B	1.961(13)	O8-Zn4-O11	105.5(6)	C64-P8-O22	104.2(9)
P1-C1	1.778(22)	O8-Zn4-O23D	99.6(6)	C64-P8-O23	107.0(9)
P1-O1	1.532(16)	O11-Zn4-O23D	111.4(6)	C64-P8-O24	106.4(9)
P1-O2	1.500(16)	Cl5-Zn5-O3	111.4(4)	O22-P8-O23	115.6(8)
P1-O3	1.536(15)	Cl5-Zn5-O13	107.4(4)	O22-P8-O24	113.5(8)
P2-C10	1.802(23)	Cl5-Zn5-O16	116.0(5)	O23-P8-O24	109.4(8)
P2-O4	1.532(17)	O3-Zn5-O13	112.3(5)	O4-P2-O5	115.2(8)
P2-O5	1.497(16)	O3-Zn5-O16	96.7(6)	O4-P2-O6	115.2(8)
P2-O6	1.522(17)	O13-Zn5-O16	112.8(6)	O5-P2-O6	110.2(9)
P3-C19	1.924(25)	Cl6-Zn6-O14	112.7(4)	C19-P3-O7	107.9(9)
P3-O7	1.511(14)	Cl6-Zn6-O17	113.0(5)	C19-P3-O8	106.6(9)

P3-O8	1.512(16)	Cl6-Zn6-O20	108.7(4)	C19-P3-O9	103.9(9)
P3-O9	1.510(14)	O14-Zn6-O17	110.7(6)	O7-P3-O8	114.9(8)
P4-C28	1.818(25)	O14-Zn6-O20	104.2(6)	O7-P3-O9	109.0(8)
P4-O10	1.487(15)	O17-Zn6-O20	106.9(6)	O8-P3-O9	113.7(8)
P4-O11	1.518(14)	Cl7-Zn7-O19	112.6(4)	C28-P4-O10	105(1)
P4-O12	1.518(14)	Cl7-Zn7-O24	112.6(4)	C28-P4-O11	106(1)
P5-C37	1.769(24)	Cl7-Zn7-O15	108.3(4)	C28-P4-O12	105(1)
P5-O13	1.531(11)	O19-Zn7-O24	111.9(6)	O10-P4-O11	116.7(8)
P5-O14	1.480(15)	O19-Zn7-O15	103.3(6)	O10-P4-O12	113.6(8)
P5-O15	1.518(13)	O24-Zn7-O15	107.5(6)	O11-P4-O12	109.7(8)
P6-C46	1.821(25)	Cl8-Zn8-O21	109.6(4)		
P6-O16	1.510(16)	Cl8-Zn8-O22	117.6(4)		

Symmetric codes for **S-Zn**: A: -1+x, y, z; B: -1+x, y, z; C: 1+x, y, z; D: 2+x, y, -1+z.

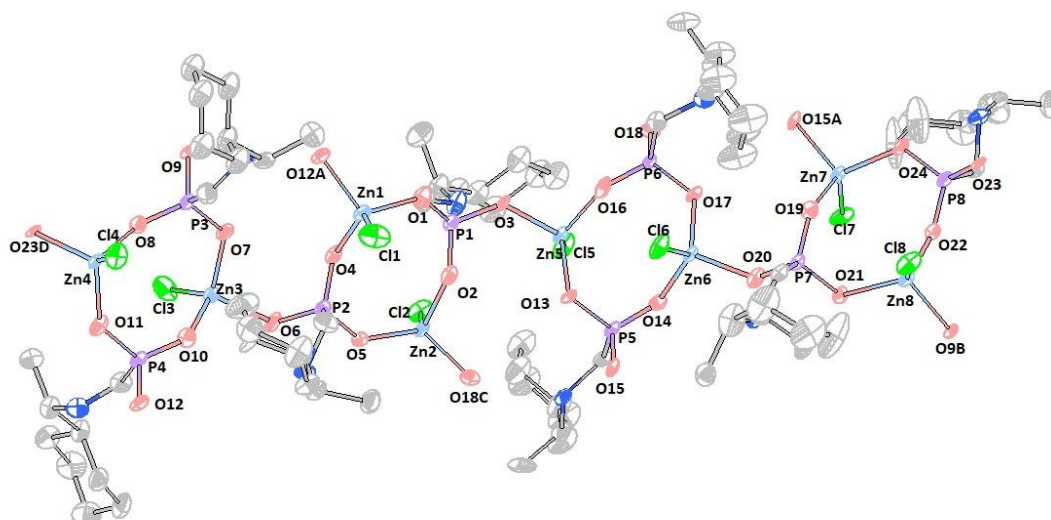


Figure S4. Asymmetrical unit of ***S*-Zn** with an atomic labeling scheme. All hydrogen atoms are omitted for clarity. Symmetric codes for ***S*-Zn**: A: $-1+x, y, z$; B: $-1+x, y, z$; C: $1+x, y, z$; D: $2+x, y, -1+z$.

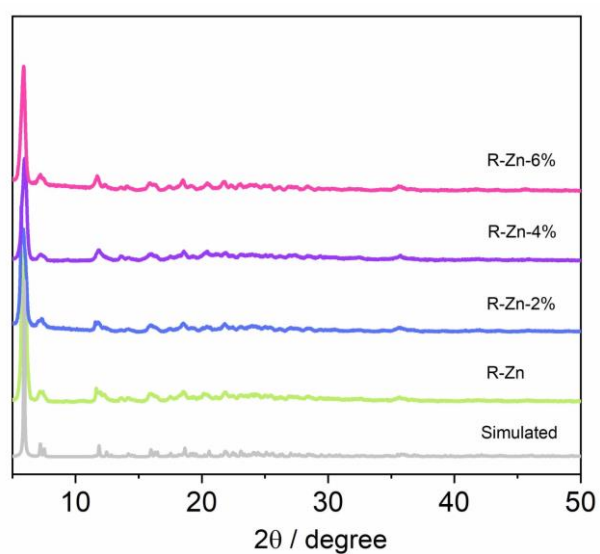


Figure S5. PXRD patterns of ***R*-Zn** and ***R*-Zn-*x*%** ($x = 2, 4, 6$). The pattern simulated from single crystal data of ***S*-Zn** is given for comparison.

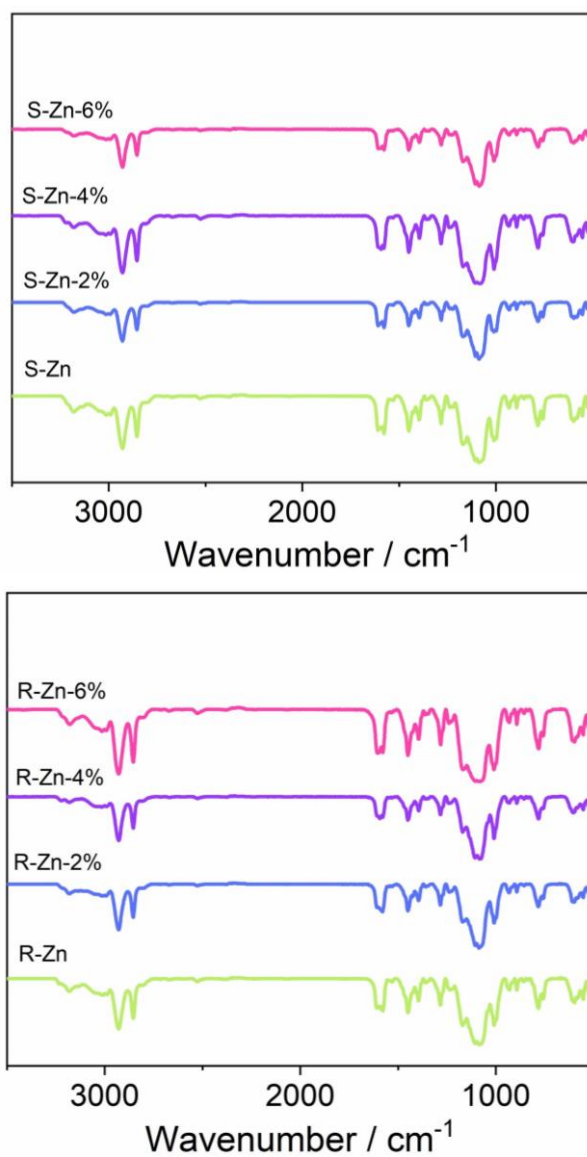


Figure S6. IR spectra of *S*-, *R*-Zn and *S*-, *R*-Zn-*x*% (*x* = 2, 4, 6).

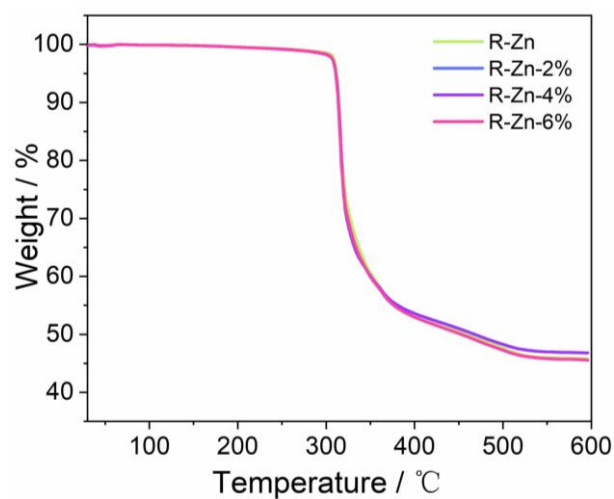
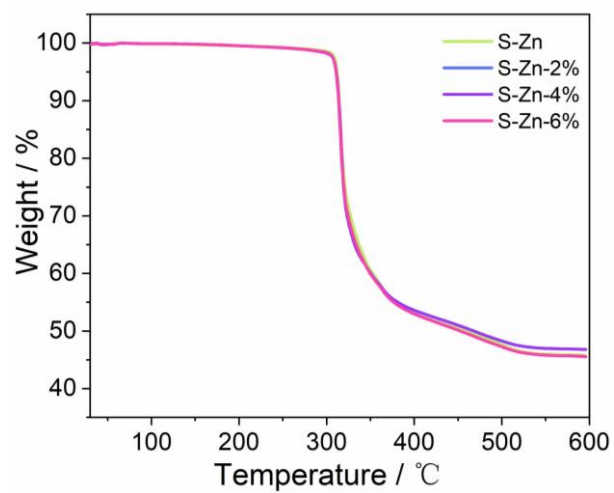


Figure S7. TG curves of *S*-, *R*-Zn and *S*-, *R*-Zn-*x*% (*x* = 2, 4, 6).

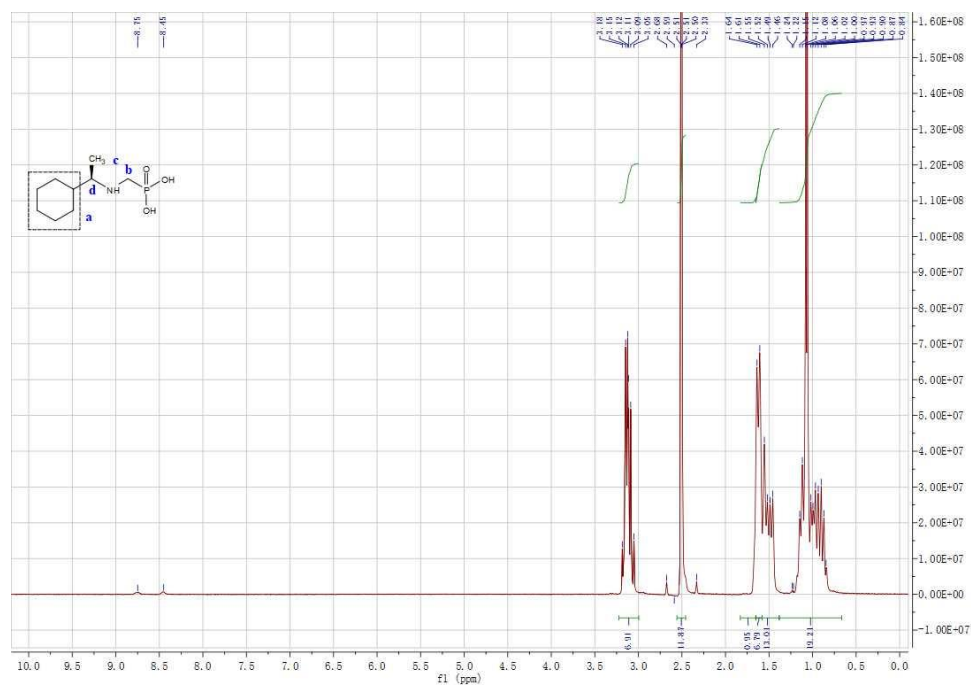


Figure S8. ¹H NMR spectrum of *S*-Zn in a DMSO-d₆ containing 1M D₂SO₄.

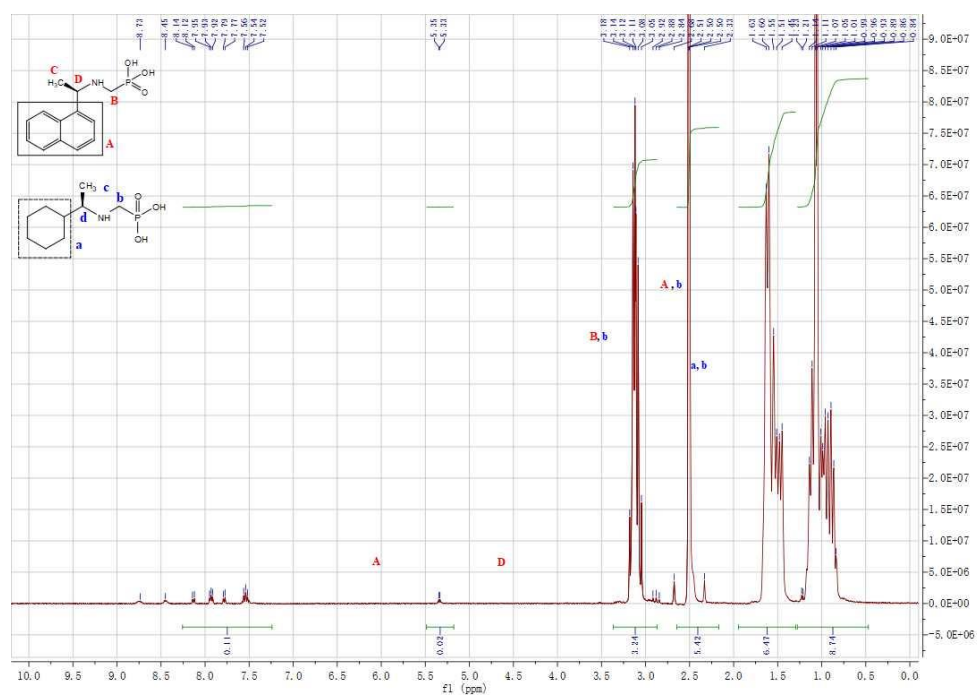


Figure S9. ¹H NMR spectrum of *S*-Zn-2% in a DMSO-d₆ containing 1M D₂SO₄.

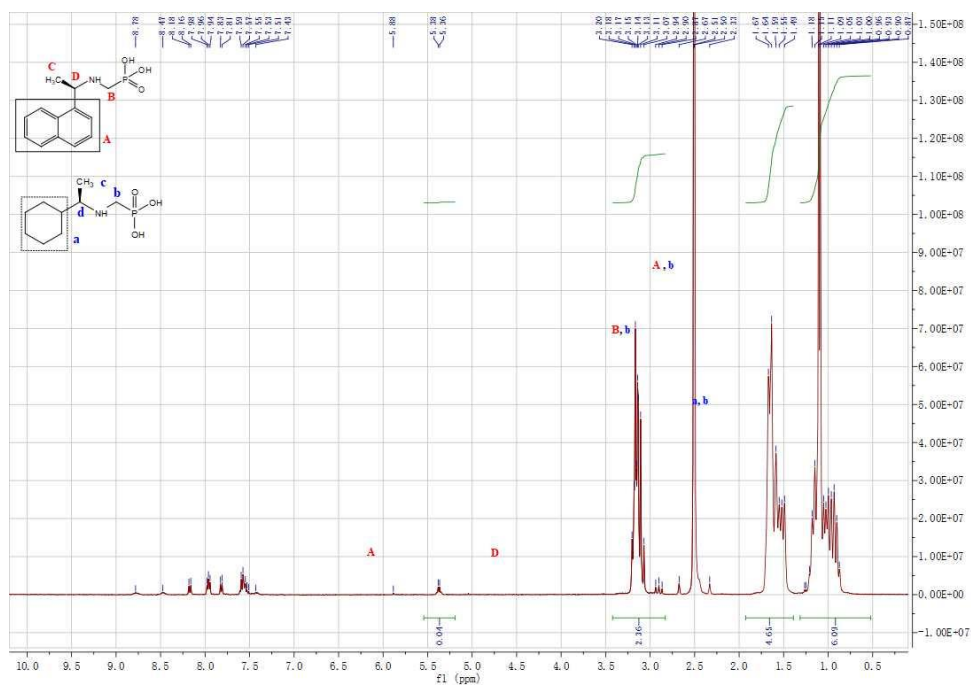


Figure S10. ¹H NMR spectrum of *S*-Zn-4% in a DMSO-d₆ containing 1M D₂SO₄.

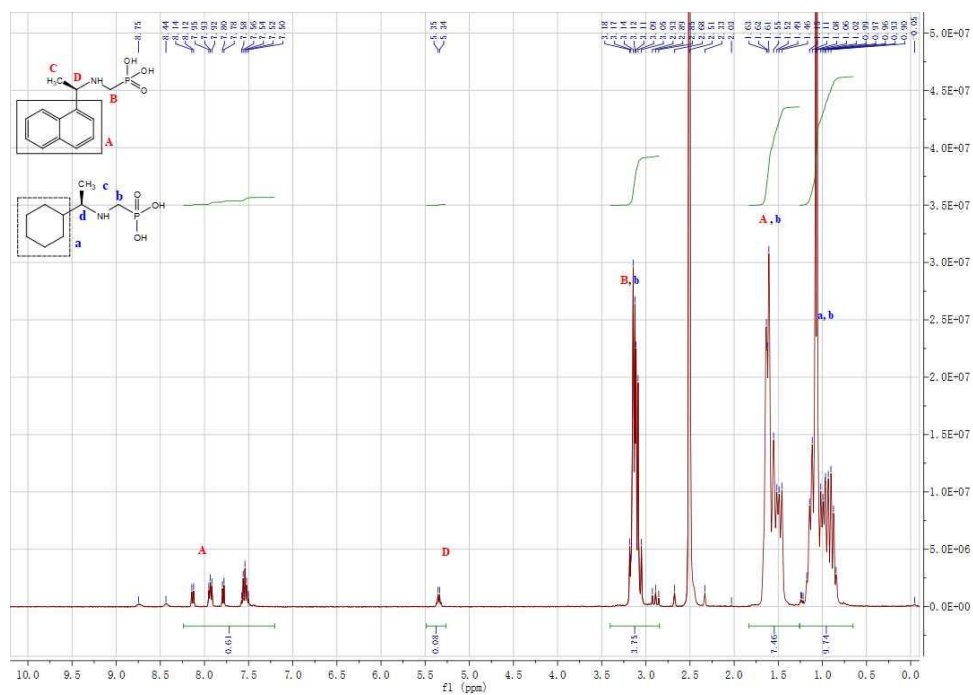


Figure S11. ¹H NMR spectrum of *S*-Zn-6% in a DMSO-d₆ containing 1M D₂SO₄.

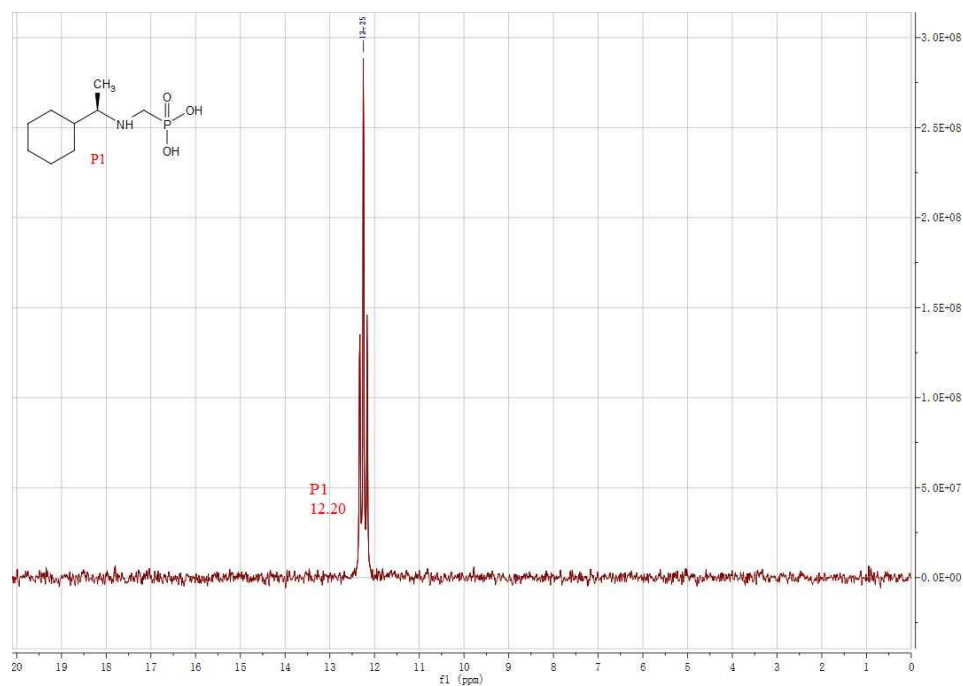


Figure S12. ^{31}P NMR spectrum of *S*-Zn in a DMSO- d_6 containing 1M D_2SO_4 .

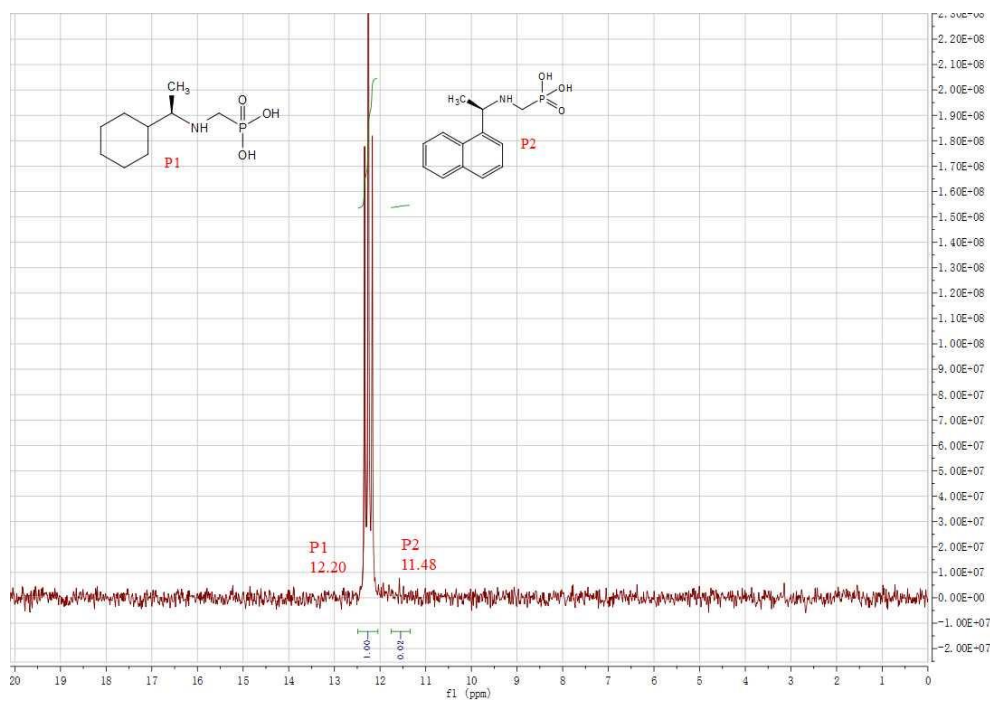


Figure S13. ^{31}P NMR spectrum of *S*-Zn-2% in a DMSO- d_6 containing 1M D_2SO_4 .

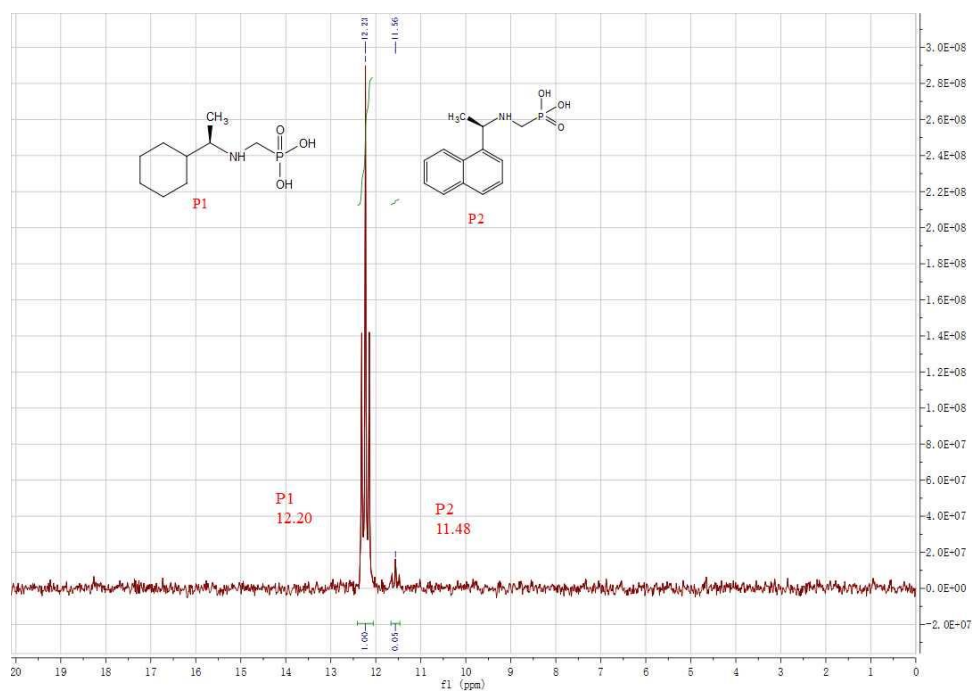


Figure S14. ^{31}P NMR spectrum of *S*-Zn-4% in a DMSO- d_6 containing 1M D_2SO_4 .

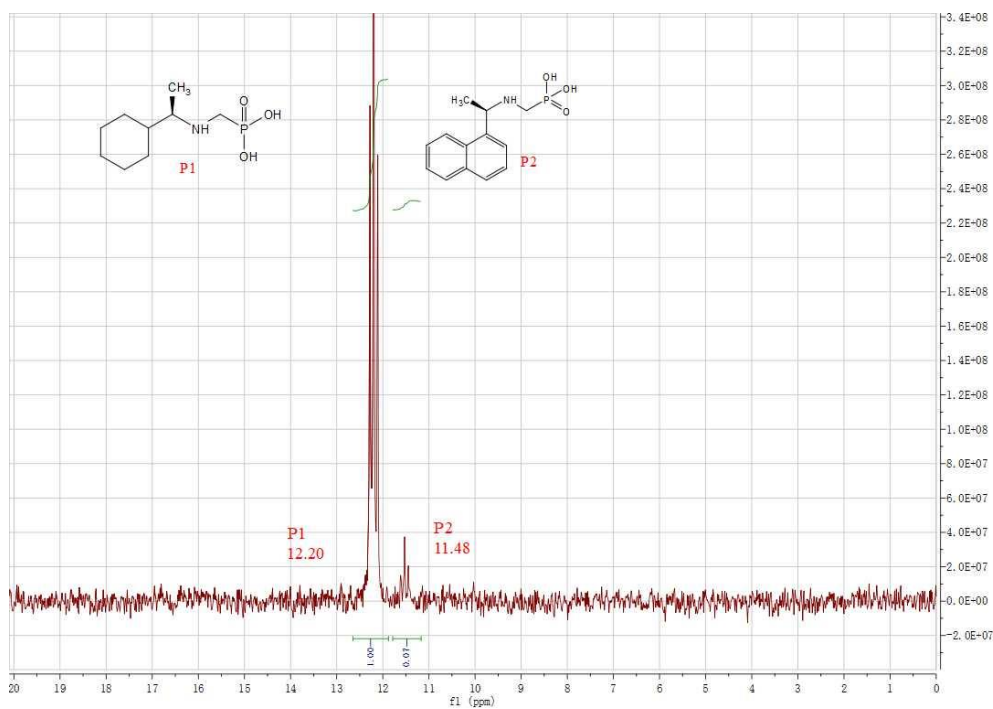


Figure S15. ^{31}P NMR spectrum of *S*-Zn-6% in a DMSO- d_6 containing 1M D_2SO_4 .

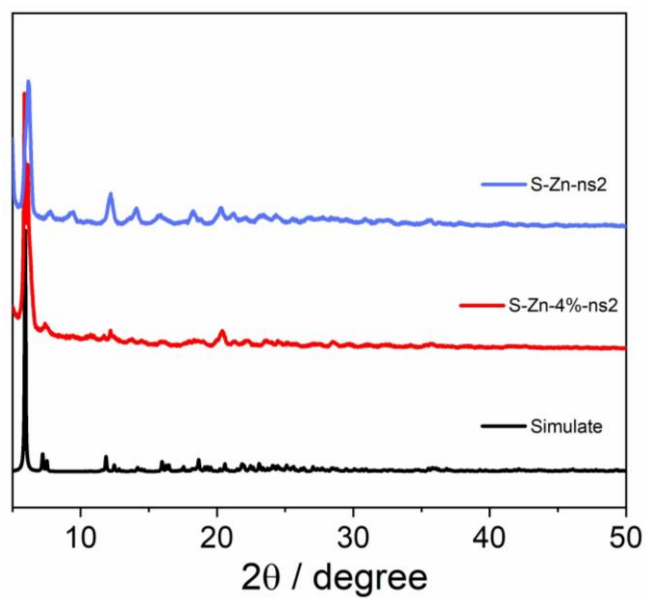


Figure S16. PXRD patterns of nanosheets of **S-Zn-ns2** (red line) and **S-Zn-4%-ns2** (blue line) after centrifuging. The pattern simulated from single crystal data of **S-Zn** is given for comparison.

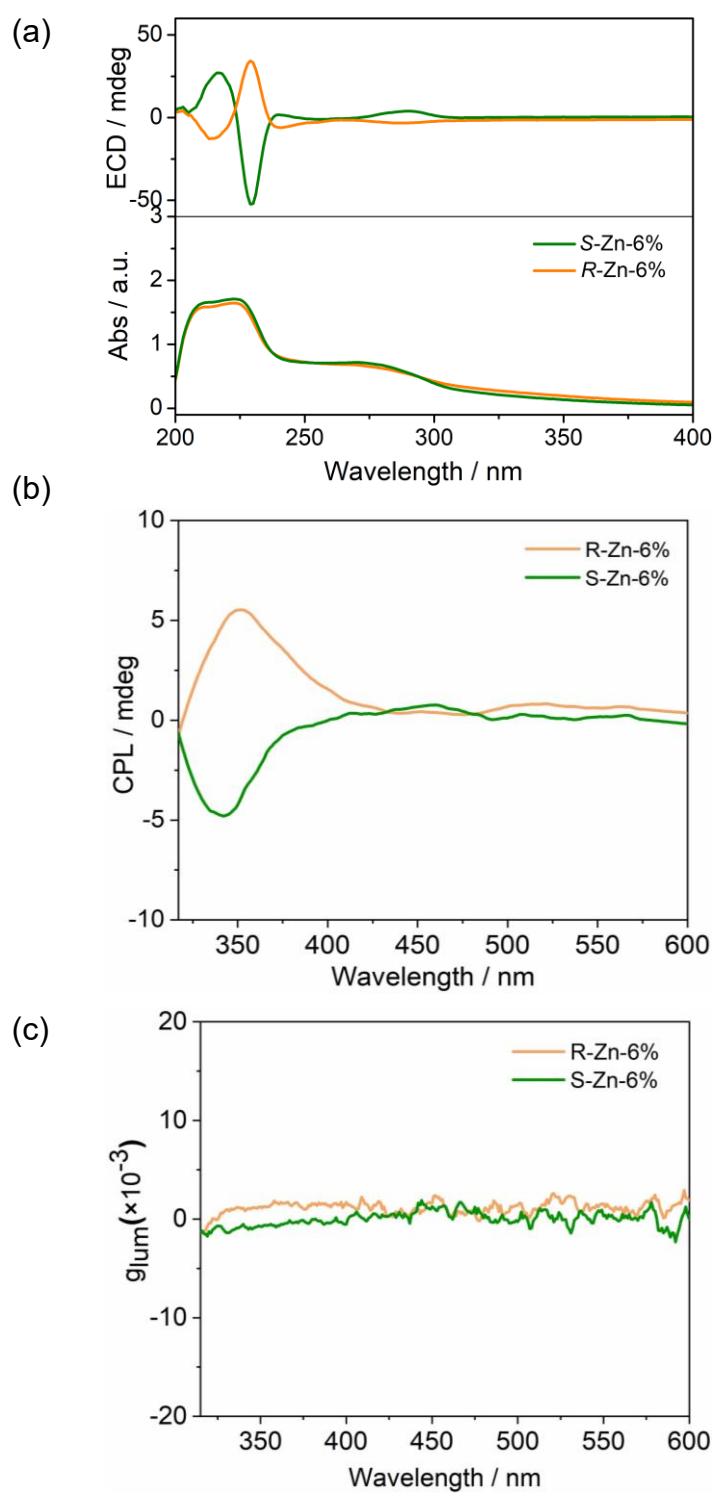


Figure S17. (a) UV-vis absorption and ECD spectra, (b) CPL spectra, and (c) $|g_{lum}|$ values of the CPL spectra of *S*-, *R*-Zn-6%.

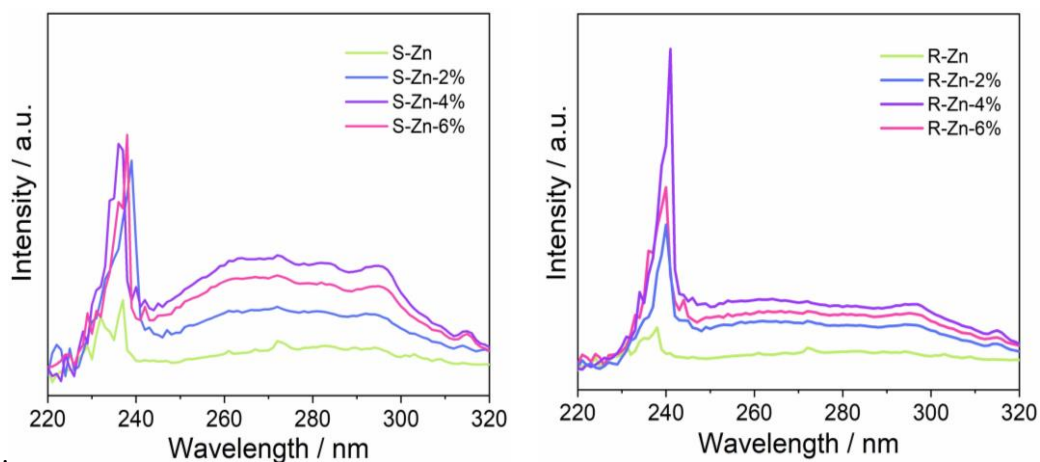


Figure S18. The excitation spectra of *S*-, *R*-Zn and *S*-, *R*-Zn-*x*% (*x* = 2, 4, 6) in the solid state at room temperature ($\lambda_{\text{em}} = 345$ nm).

Table S3. Luminescence Quantum Yields (Φ_{lum}), Lifetimes (τ_{obs}), and Dissymmetry Factors (g_{lum}) of Compounds *S*-, *R*-Zn-*x*% (*x* = 2, 4, 6) Measured in the Solid State at room temperature.

compound	Φ_{lum} (%)	τ_{obs} (ns)	$ g_{\text{lum}} $ ($\pi^* \rightarrow \pi$)
<i>R</i> -Zn-2%	8.04	37.594	6.51×10^{-4}
<i>R</i> -Zn-4%	9.54	44.762	1.588×10^{-3}
<i>R</i> -Zn-6%	6.74	47.545	8.8×10^{-4}
<i>S</i> -Zn-2%	7.95	39.645	1.193×10^{-3}
<i>S</i> -Zn-4%	9.12	42.916	1.845×10^{-3}
<i>S</i> -Zn-6%	7.77	47.762	9.68×10^{-4}

Table S4. Elemental analysis of superhelices of *S*-, *R*-Zn and *S*-, *R*-Zn-*n*% (*n* = 0-6%).

		<i>C</i> /%	<i>H</i> /%	<i>N</i> /%	Yield/%
<i>S</i>-Zn	<i>Calculated</i>	33.67	5.97	4.36	/
	<i>Experiment</i>	34.02	6.13	4.43	45.0
<i>S</i>- Zn-2%	<i>Calculated</i>	33.87	5.93	4.35	/
	<i>Experiment</i>	34.12	5.97	4.37	43.5
<i>S</i>- Zn-4%	<i>Calculated</i>	34.07	5.89	4.33	/
	<i>Experiment</i>	34.32	6.02	4.36	44.5
<i>S</i>- Zn-6%	<i>Calculated</i>	34.27	5.86	4.32	/
	<i>Experiment</i>	34.64	5.89	4.35	47.0
<i>R</i>- Zn	<i>Calculated</i>	33.67	5.97	4.36	/
	<i>Experiment</i>	33.25	5.64	4.34	49.5
<i>R</i>- Zn-2%	<i>Calculated</i>	33.87	5.93	4.35	/
	<i>Experiment</i>	33.80	5.73	4.20	45.1
<i>R</i>- Zn-4%	<i>Calculated</i>	34.07	5.89	4.33	/
	<i>Experiment</i>	33.79	5.56	4.19	47.5
<i>R</i>- Zn-6%	<i>Calculated</i>	34.27	5.86	4.32	/
	<i>Experiment</i>	34.56	5.95	4.38	42.2