Supplementary Information

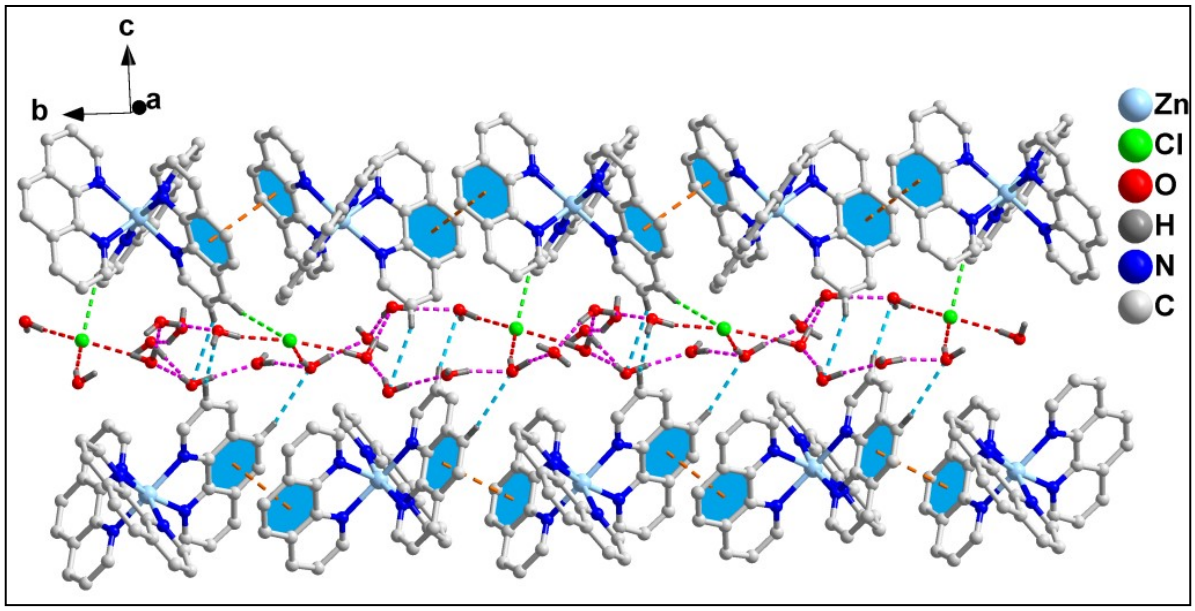


Fig. **S1** Layered assembly of compound **1** along the crystallographic *bc* plane assisted by the infinite Cl-H_2O cluster.

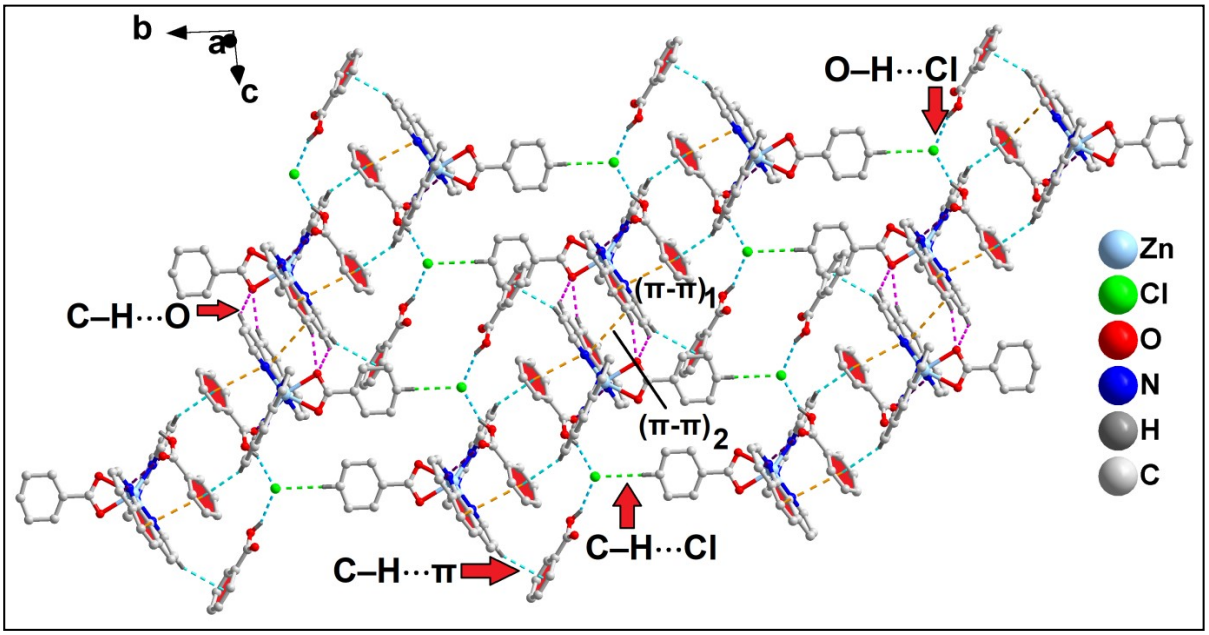


Fig. **S2** Layered assembly of compound **2** along the *bc* plane assisted by the cooperative ternary $(\pi\text{-}\pi)_1/(\pi\text{-}\pi)_2/(\pi\text{-}\pi)_1$ assembly and lattice Cl ions.

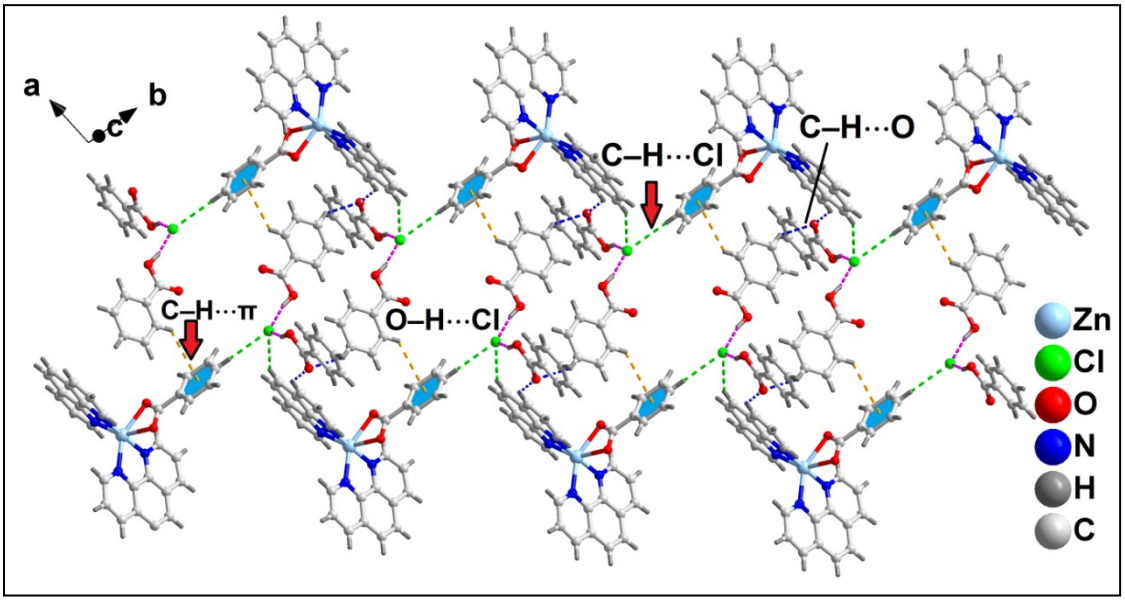


Fig. **S3** 2D assembly of compound **2** along the *ab* plane assisted by the $[(bzH)_4Cl_2]^{2-}$ cores.

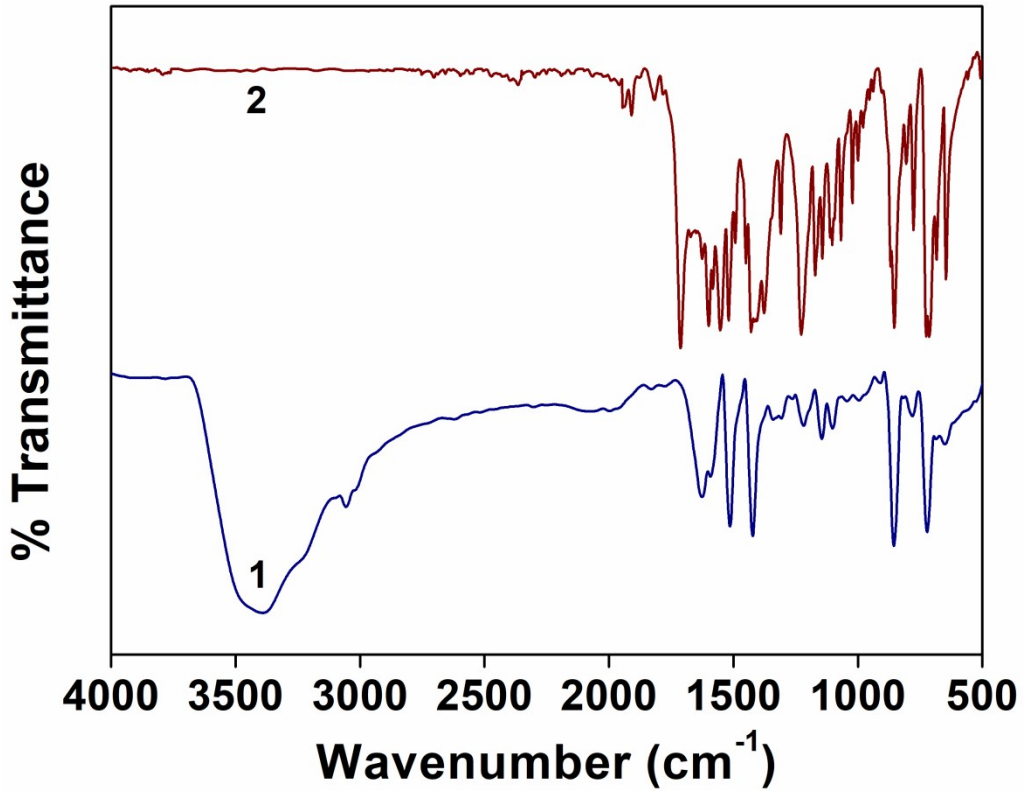
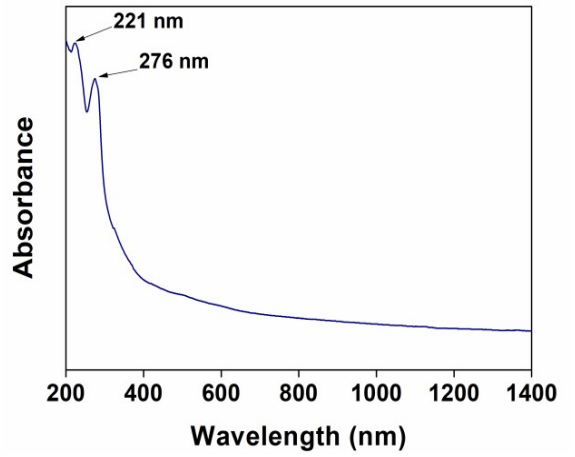


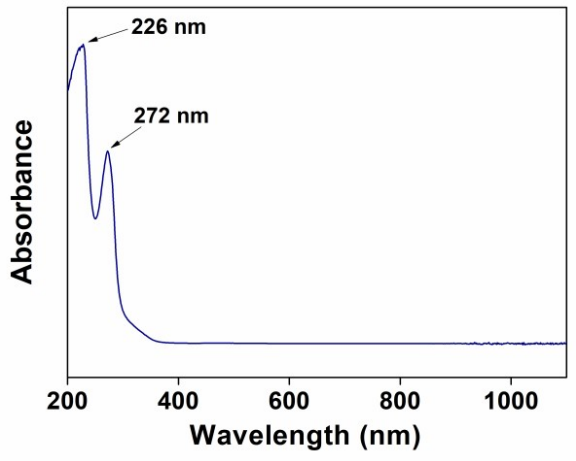
Fig. **S4** FT-IR spectra of the compounds **1** and **2**.

3.3.2 Electronic spectroscopy

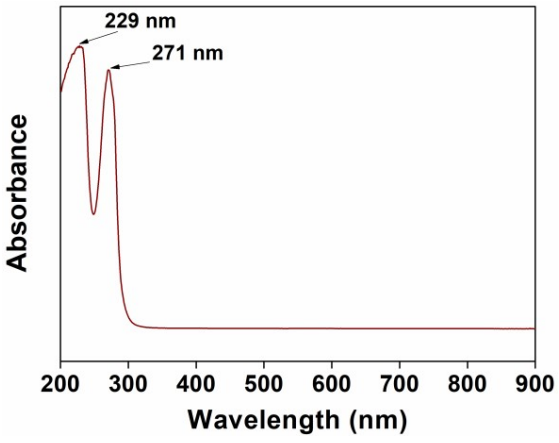
The electronic spectra of the compounds (Figs. S5 and S6) have been recorded in both solid as well as in aqueous phases (0, 6, 12, 24 and 48 hours). The spectra of the diamagnetic Zn(II) compounds in both the phases do not exhibit any absorption bands in the visible region.[1] The peaks obtained in the UV region can be attributed to the π-π* transition of the aromatic ligands.[2] The similar absorption peaks obtained in the solid as well as aqueous phase spectra of the compounds imply that the bonding as well as the geometries of the compounds does not undergo any structural distortion in the solution phase.[3]



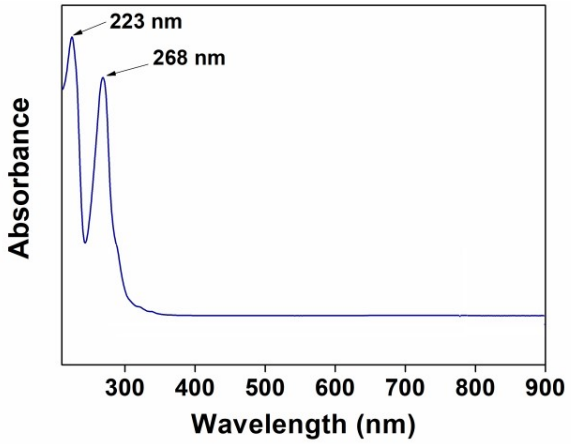
(**a**)



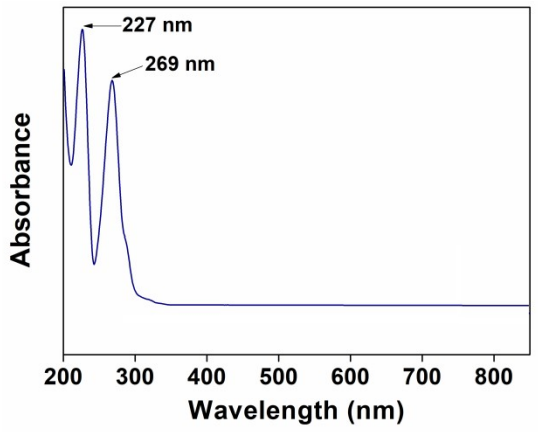
(**b**)



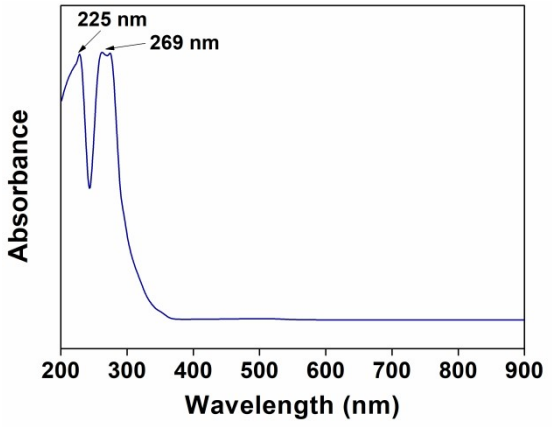
(**c**)



(**d**)

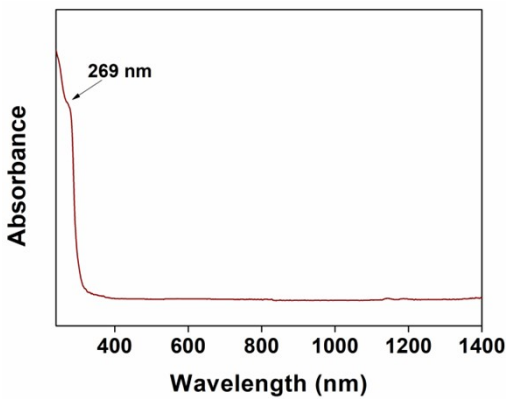


(**e**)

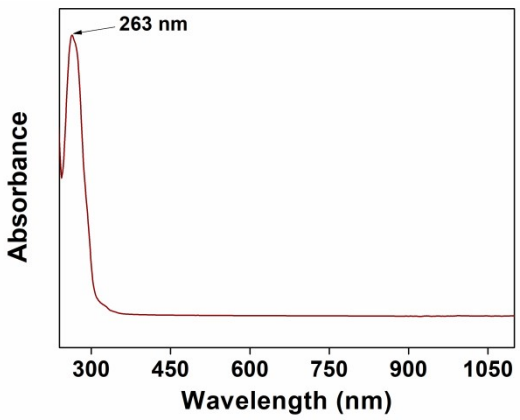


(**f**)

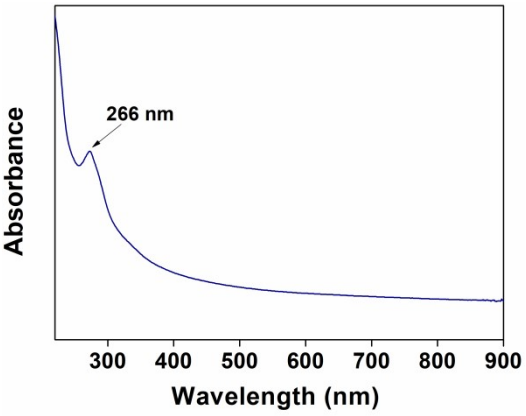
Fig. **S5**(**a**) UV-Vis-NIR spectrum of compound **1**; (**b**) UV-Vis spectrum of **1** (0 hour); (**c**) UV-Vis spectrum of compound **1** (6 hours); (**d**) UV-Vis spectrum of compound **1** (12 hours); (**e**) UV-Vis spectrum of compound **1** (24 hours); (**f**) UV-Vis spectrum of compound **1** (48 hours).



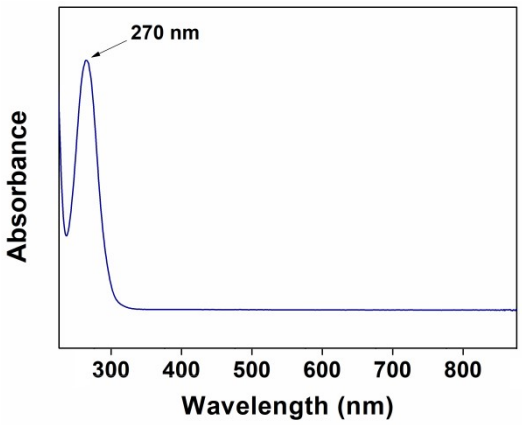
(**a**)



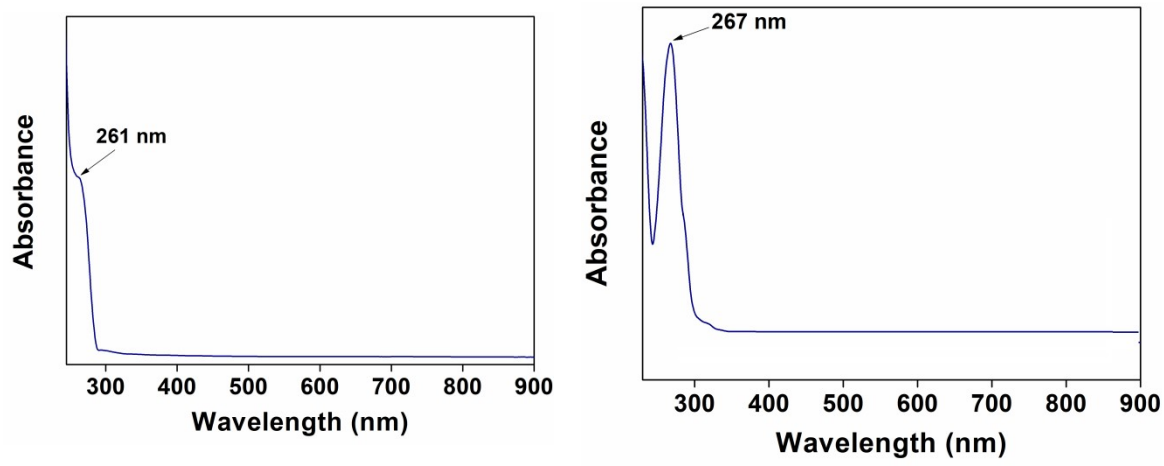
(**b**)



(**c**)



(**d**)



(**e**) (**f**)

Fig. **S6**(**a**) UV-Vis-NIR spectrum of compound **2**; (**b**) UV-Vis spectrum of **2** (0 hour); (**c**) UV-Vis spectrum of compound **2** (6 hours); (**d**) UV-Vis spectrum of compound **2** (12 hours); (**e**) UV-Vis spectrum of compound **2** (24 hours); (**f**) UV-Vis spectrum of compound **2** (48 hours).

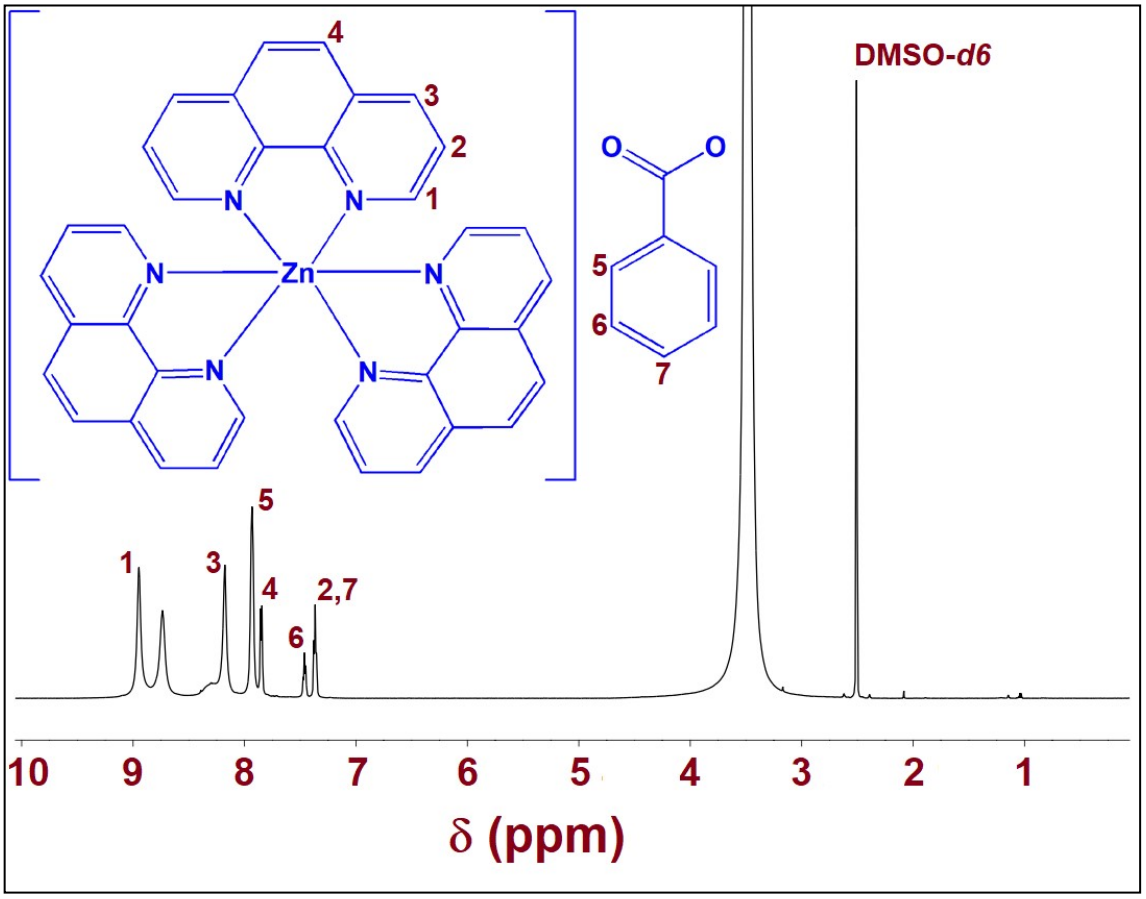


Fig. **S7** ^{1}H-NMR spectrum of compound **1** in DMSO-*d6*.

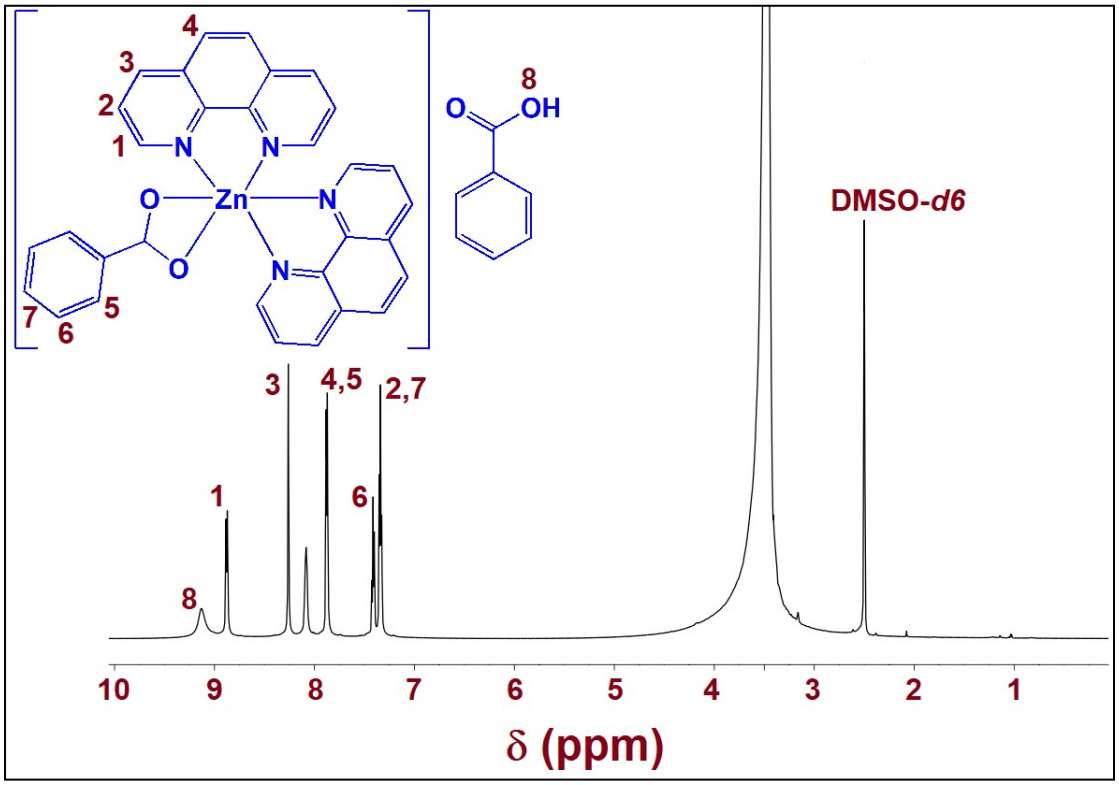


Fig. **S8** [1]H-NMR spectrum of compound **2** in DMSO-*d6*.

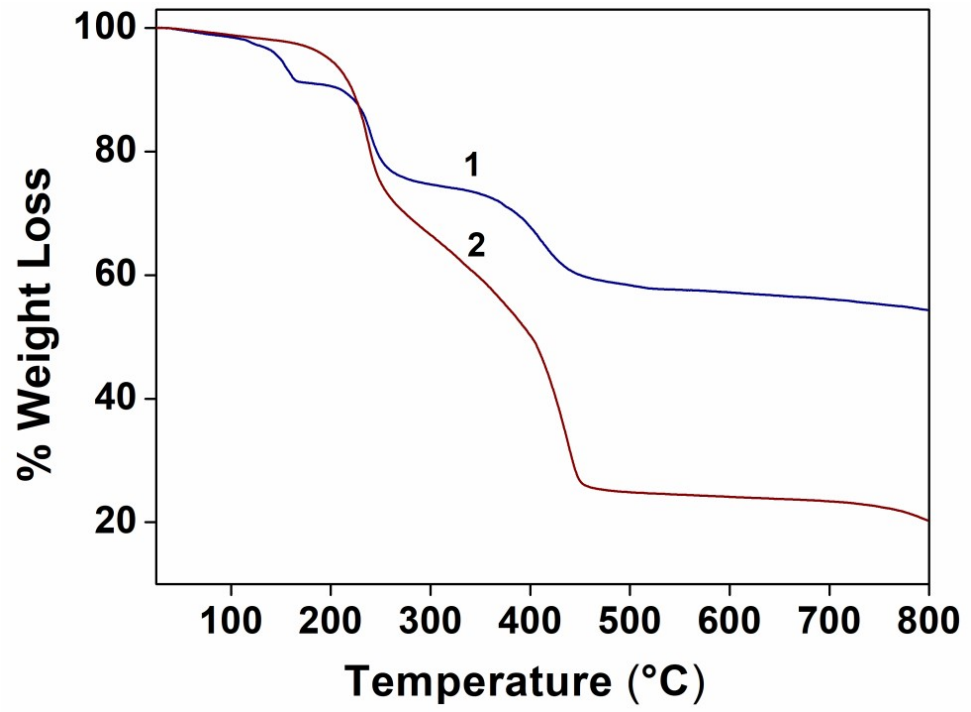


Fig. **S9** Thermogravimetric curves of the compounds **1** and **2**.

Supplementary References

1 (a) E. L. Araújo, H. F. G. Barbosa, E. R. Dockal and E. T. G. Cavalheiro, *Int. J. Biol. Macromol*., 2017, **95**, 168-176;

(b) A. C. Ekennia, D. C. Onwudiwe, A. A. Osowole, L. O. Olasunkanmi and E. E. Ebenso, *J. Chem*., 2016, **2016**, 1-12.

2 U. Saha, D. Dutta, H. Nath, A. Franconetti, A. Frontera and M. K. Bhattacharyya, *Inorg. Chim. Acta*, 2019, **488**, 159-169.

3 (a) H. Nath, D. Dutta, P. Sharma, A. Frontera, A. K. Verma, M. Barceló-Oliver, M. Devi and M. K. Bhattacharyya, *Dalton Trans*., 2020, **49**, 9863-9881;

(b) C. Yenikaya, M. Poyraz, M. Sari, F. Demirci, H. Ilkimen and O. Buyukgungor, *Polyhedron*, 2009, **28**, 3526-3532.