1. Experimental detail:

Compound **1** here is described as an example:

2.5mg compound **1** was dissolved in a saturated solution of NaOH, and the solution was heated to 80ºC for 30min, then the solution was cooled to room temperature, about 1.5ml concentrated nitric acid was added into the solution dropwise, now you can obtain a clear solution. Finally, transfer the solution to a 25ml volumetric flask for elemental analysis.

2. Table s1. Hydrogen bonds in compounds **2** and **4**.

Compound **2**				
		D···A bond length(Å)	D-H···A angle(º)	Symmetry code
1	N(1)-H(1)···O(6_a)	3.0169(2)	137.714(5)	a: 0.33333-y, 0.66667+x-y, -0.33333+z
2	N(1)-H(1)···O(14_b)	3.0326(3)	145.499(11)	b: 0.33333-x+y, 0.66667-x, -0.33333+z
3	N(2)-H(2)···O(4W)	2.9704(2)	144.985(6)	
4	O(4W)···O(1_c)	2.7959(2)		c: -y, x-y, z
5	O(4W)···O(8_d)	3.0354(1)		d: -x+y, 1-x, z
6	O(3W)···O(2_e)	3.0957(2)		e: 0.33333-y, -0.33333+x-y, -0.33333+z
Compound **4**				
1	O(4W)···O(2)	3.1096(1)		
2	O(4W)···O($3W_f$)	3.0380(1)		f: 1.5-x, -0.5+y, 0.5-z
3	O(2W)···O(1_f)	3.0003(2)		
4	O(3W)···O(2_g)	3.1170(3)		g: 1.5-x, 0.5+y, 0.5-z
5	O(3W)···O(3_h)	3.0925(2)		h: x, 1+y, z
6	O(1W)···O(4_g)	2.9952(1)		
7	O(1W)···O(13_g)	3.0954(2)		
8	O(1W)···O(10_i)	2.9501(1)		i: x, 1+y, 1+z

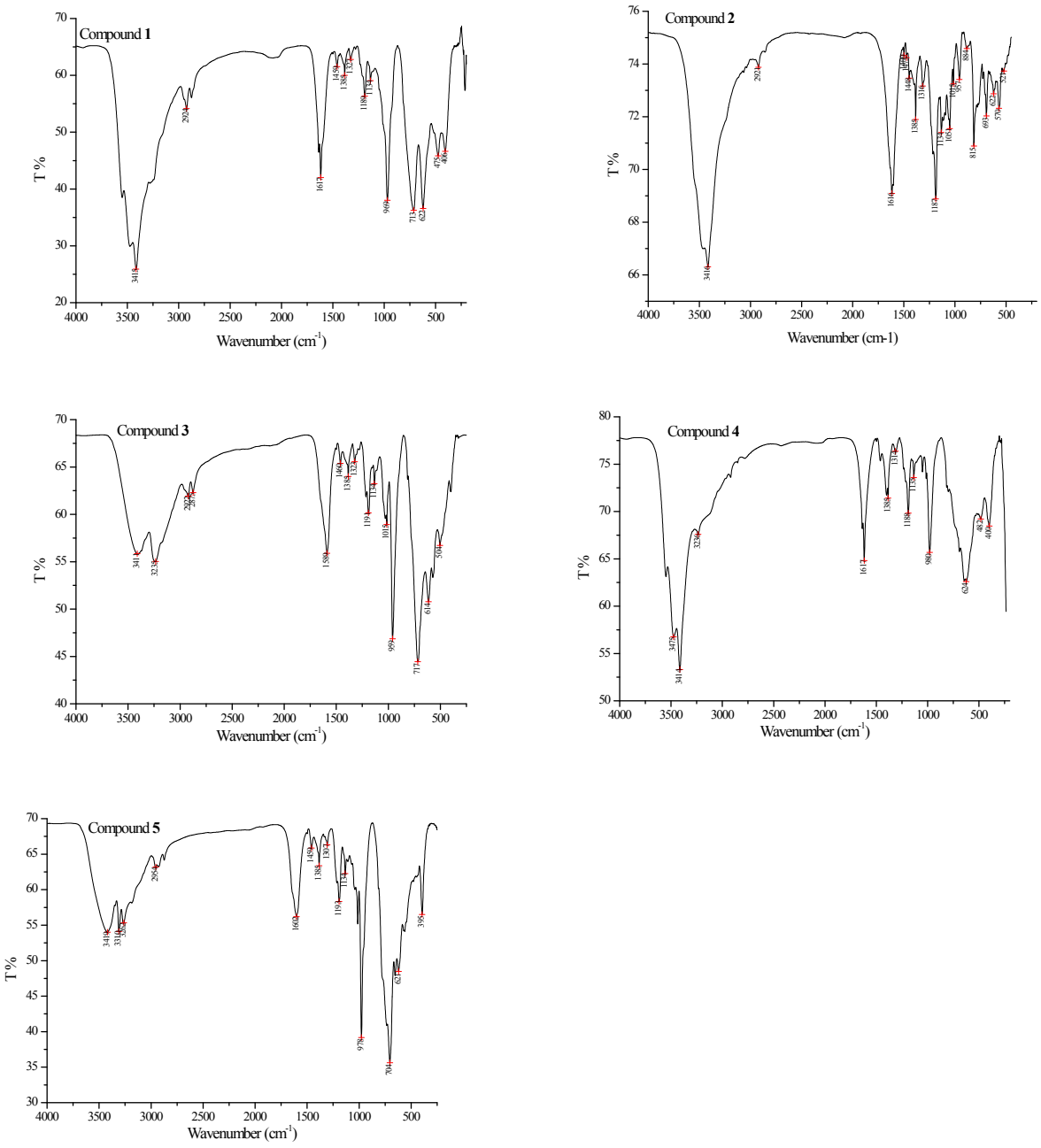
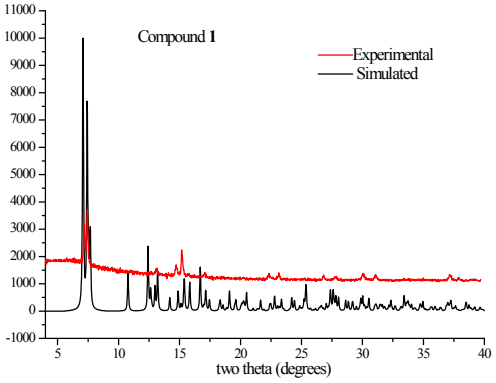
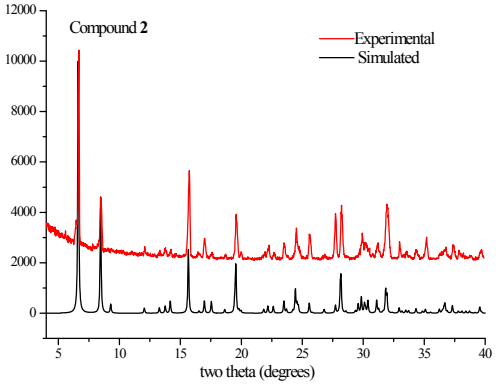
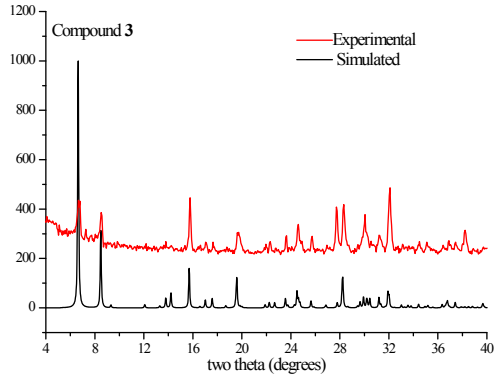
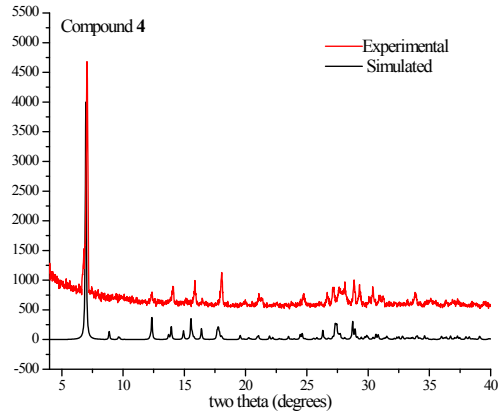


Fig. s1 IR spectra of compounds **1**-**5**. Infrared spectra for compounds **1**-**5** were recorded as KBr pellets on a Perkin-Elmer SPECTRUM ONE FTIR spectrophotometer.









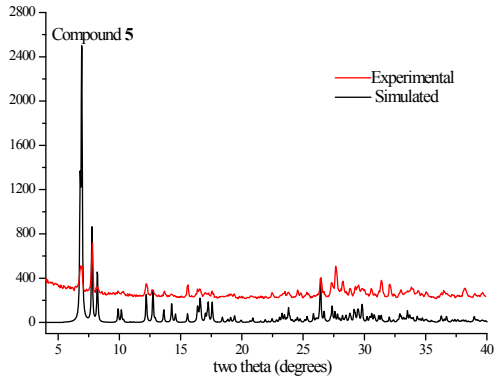


Fig. s2 experimental and simulated XRD patterns of compounds **1**-**5**. Powder XRD patterns for compounds **1**-**5** were obtained with a Scintag X1 powder diffractometer system using Cu Kα radiation with a variable divergent slit and a solid-state detector.

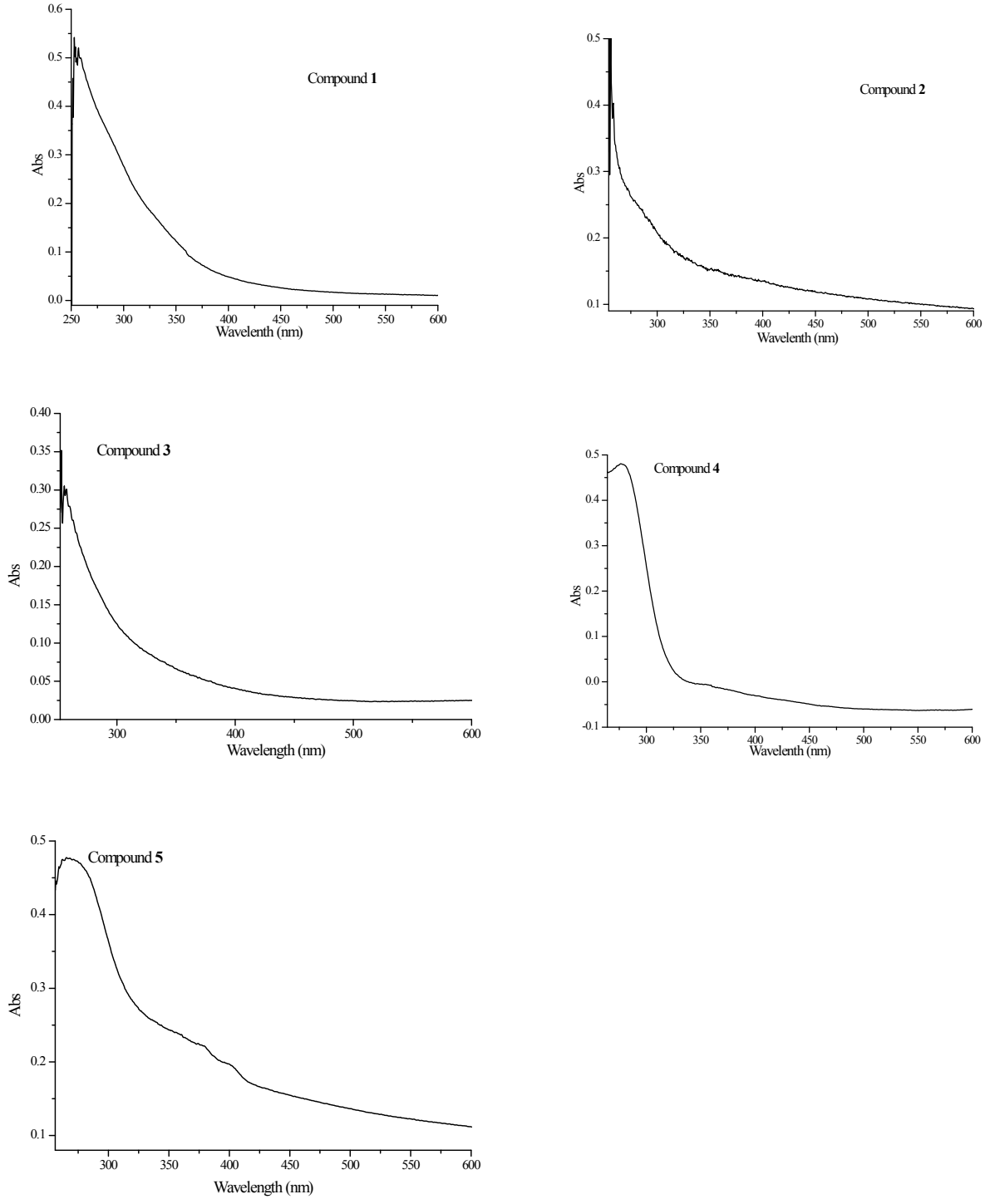


Fig. s3 UV-Vis spectra of compounds **1**-**5**. UV-vis spectra for compounds **1**-**5** were recorded on a Shimadzu UV-3100 spectrophotometer.

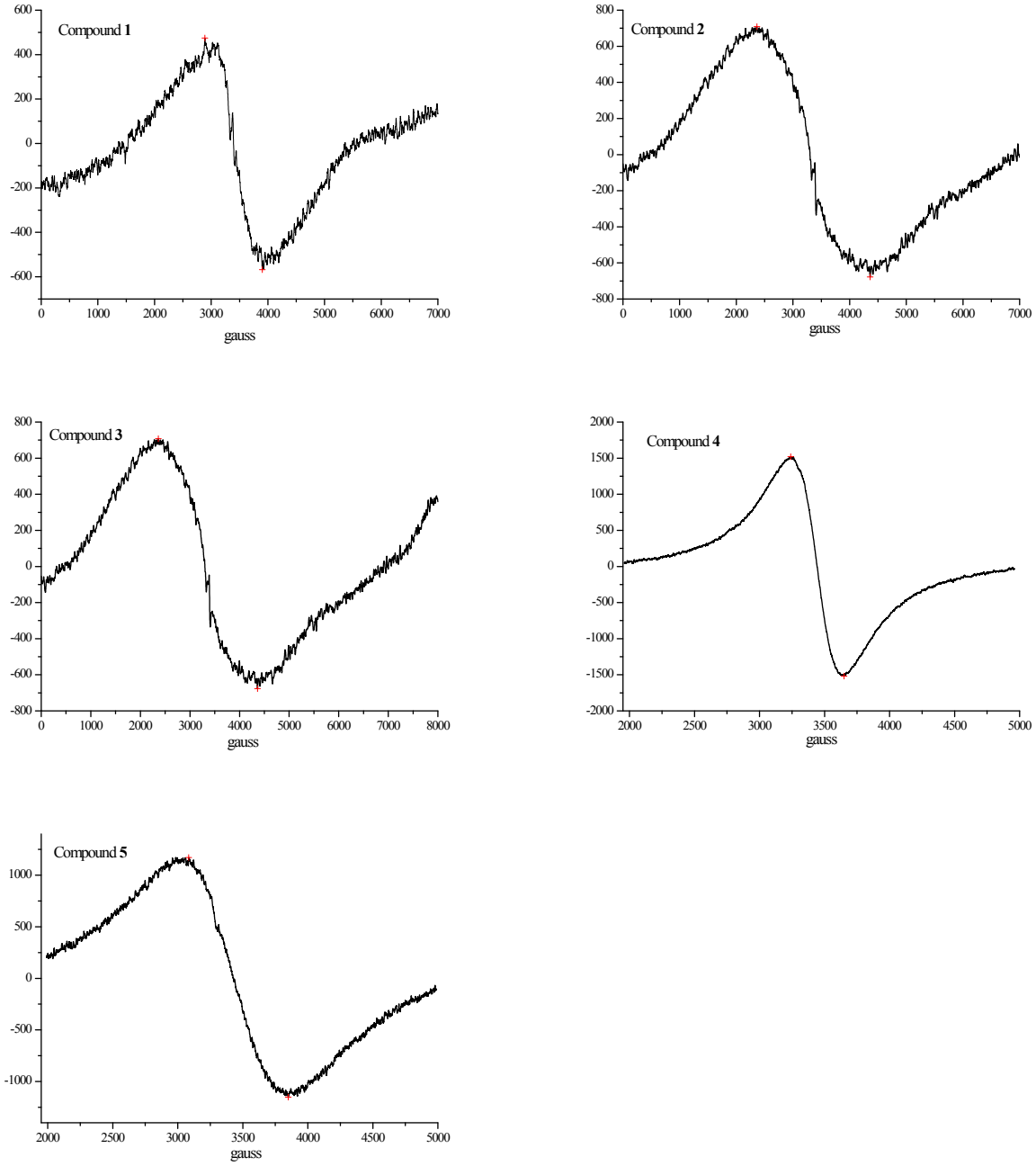


Fig. s4 ESR spectra of compounds **1**-**5**. Electron spin resonance (ESR) spectra for compounds **1**-**5** were performed on a JEOL JES-FA200 spectrometer operating in the X-band mode. The g value was calculated by comparison with the spectrum of 1,1-diphenyl-2-picrylhydrazyl (DPPH), whereas the spin concentrations were determined by comparing the recorded spectra with that of a Mn marker and DPPH, using the built-in software of the spectrometer.

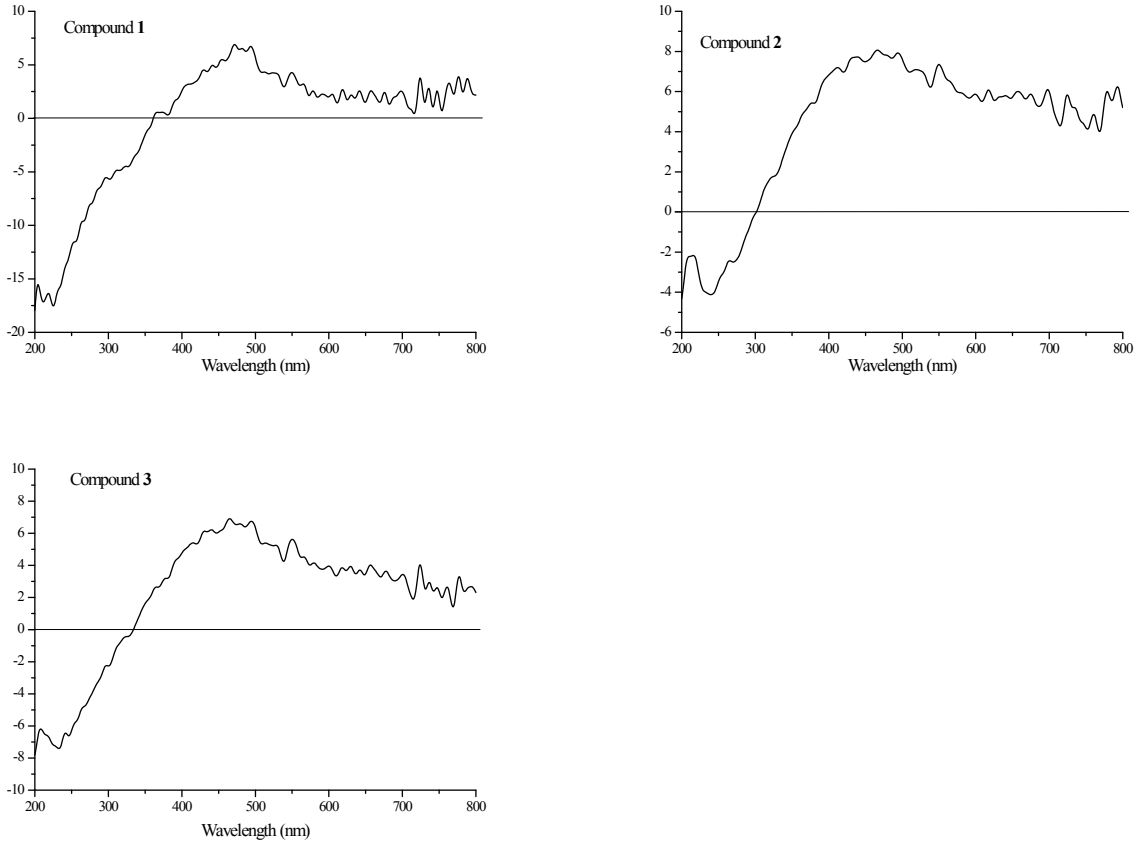


Fig. s5 solid state CD spectra of compounds **1**-**3**. Solid state circular dichroism (CD) spectra for compounds **1**-**3** were recorded at room temperature with a Jasco J-810(S) spectropolarimeter.

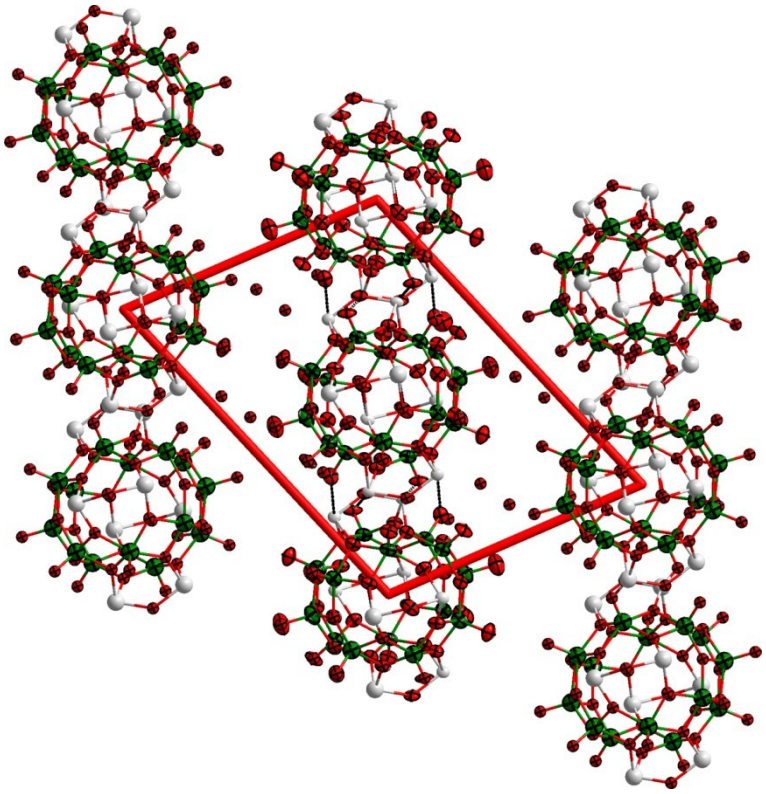


Fig. s6 ball-and-stick representation of the stacking of the inorganic layers in compound **4**.