Table S1 Selected bond lengths (Å) for complexes **1**–**11**

	1	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**
Ln(1)–O(1)	2.486(2)	2.523(3)	2.469(3)	2.481(2)	2.467(3)	2.458(3)	2.446(3)	2.385(3)	2.382(3)	2.365(3)	2.383(5)
Ln(1)–O(2)	2.532(2)	2.479(3)	2.491(3)	2.496(3)	2.465(3)	2.459(3)	2.450(4)	2.387(3)	2.374(3)	2.374(3)	2.399(5)
Ln(1)–O(3)	2.492(2)	2.523(3)	2.514(3)	2.443(2)	2.422(3)	2.405(3)	2.477(4)	2.436(3)	2.464(3)	2.424(3)	2.357(4)
Ln(1)–O(4)	2.541(2)	2.583(3)	2.504(3)	2.424(2)	2.413(3)	2.417(2)	2.443(3)	2.437(3)	2.420(3)	2.406(3)	2.345(4)
Ln(1)–O(5)	2.597(2)	2.519(3)	2.448(3)	2.484(2)	2.469(3)	2.461(3)	2.399(3)	2.467(3)	2.430(3)	2.411(3)	2.353(6)
Ln(1)–O(6)	2.537(2)	2.471(3)	2.469(3)	2.475(2)	2.426(3)	2.418(3)	2.454(3)	2.428(3)	2.421(3)	2.455(3)	2.439(6)
Ln(1)–O(7)	2.572(2)	2.505(3)	2.511(3)	2.472(2)	2.486(3)	2.459(3)	2.404(3)	2.448(3)	2.384(3)	2.448(4)	2.448(5)
Ln(1)–O(8)	2.520(2)	2.548(3)	2.495(3)	2.438(3)	2.464(3)	2.484(3)	2.398(3)	2.390(3)	2.445(4)	2.372(4)	2.395(5)
Ln(1)–O(9)	2.513(2)	2.488(3)	2.550(4)	2.515(3)	2.488(4)	2.486(3)	2.468(4)	2.450(3)	2.443(4)	2.421(4)	2.405(6)

Table S2 Selected bond lengths (Å) for complexes **12**–**16**

	12	**13**	**14**	**15**	**16**
Ln(1)–O(3)	2.340(4)	2.328(4)	2.348(5)	2.363(8)	2.359(5)
Ln(1)–O(2)(4)	2.360(4)	2.396(5)	2.383(5)	2.325(8)	2.320(5)
Ln(1)–O(1)#1	2.395(4)	2.349(5)	2.325(4)	2.297(7)	2.293(4)
Ln(1)–O(9)	2.406(5)	2.401(5)	2.392(5)	2.369(9)	2.360(5)
Ln(1)–O(10)	2.412(5)	2.408(6)	2.387(5)	2.373(8)	2.352(5)
Ln(1)–N(4)(3)	2.431(4)	2.424(5)	2.423(5)	2.405(9)	2.387(6)
Ln(1)–N(1)#2	2.596(5)	2.591(5)	2.576(5)	2.566(10)	2.558(5)
Ln(1)–O(5)(7)	2.641(3)	2.635(4)	2.629(4)	2.631(7)	2.611(4)
Ln(2)–O(7)#3	2.361(4)	2.705(4)	2.344(4)	2.335(8)	2.437(5)
Ln(2)–O(4)(2)	2.402(4)	2.392(4)	2.396(4)	2.357(7)	2.356(4)
Ln(2)–O(11)	2.464(5)	2.453(5)	2.437(5)	2.415(9)	2.410(5)
Ln(2)–O(8)	2.485(4)	2.509(4)	2.467(4)	2.445(8)	2.314(5)
Ln(2)–O(6)	2.517(4)	2.347(4)	2.497(4)	2.480(8)	2.463(5)
Ln(2)–N(3)(4)	2.528(5)	2.520(6)	2.509(5)	2.501(10)	2.480(6)
Ln(2)–O(6)#4(8)	2.547(3)	2.527(4)	2.528(4)	2.518(7)	2.520(4)
Ln(2)–N(2)	2.548(5)	2.531(5)	2.520(5)	2.514(10)	2.483(5)
Ln(2)–O(5)	2.715(4)	2.466(5)	2.710(4)	2.697(7)	2.709(4)

Table S3 Important hydrogen bond interactions in complexes **1**–**11**[#]

D–H···A	D–H / Å	H···A/ Å	D···A / Å	<D–H···A/ º
		1		
O7–H7A···O8 [a]	0.820	2.229	164.94	3.028
O8–H8A···O6 [b]	0.820	1.940	154.73	2.704
O9–H9C···O4 [c]	0.850	2.306	119.07	2.819
O9–H9C···O3 [d]	0.850	2.553	134.28	3.205
OW2–HW2A···OW1	0.850	1.939	141.50	2.657
OW2–HW2A···OW1 [e]	0.850	2.083	152.50	2.864
OW2–HW2B···O7 [f]	0.850	2.110	160.61	2.925
OW1–HW1A···OW2 [e]	0.850	2.250	129.26	2.864
		2		
O7–H7C···O3 [g]	0.850	1.918	151.65	2.697
O8–H8A···OW1	0.820	2.059	168.63	2.868
O8–H8A···O7 [a]	0.820	2.615	111.73	3.017
O8–H8B···O4 [a]	0.850	2.037	167.70	2.873
O9–H9A···O1 [h]	0.820	2.169	137.08	2.826
O9–H9C···O2 [d]	0.850	2.652	123.30	3.199
OW1–HW1B···O7 [a]	0.850	2.299	132.01	2.937
OW1–HW1C···OW2 [i]	0.850	1.409	125.86	2.028
OW2–HW2A···OW2 [j]	0.850	1.340	149.10	2.115
OW2–HW2A···OW1 [k]	0.850	1.420	124.66	2.028
OW3–HW3B···OW2 [a]	0.850	2.284	144.78	3.019
OW3–HW3C···O8 [i]	0.850	2.204	140.54	2.911
		3		
O9–H9B···OW1 [l]	0.850	2.099	136.19	2.776
O9–H9C···OW1 [m]	0.850	2.284	125.36	2.861
OW1–HW1B···O4 [n]	0.850	2.358	139.04	3.051
OW1–HW1C···OW3 [o]	0.850	2.609	143.96	3.334
OW1–HW1C···O3 [o]	0.850	2.640	112.84	3.072
OW2–HW2B···OW1 [m]	0.850	2.316	153.34	3.099
OW2–HW2B···OW3 [p]	0.850	2.383	114.43	2.842
OW2–HW2C···O8	0.850	2.097	146.37	2.844
OW3–HW3A···OW2 [q]	0.850	2.163	136.66	2.842
OW3–HW3B···O7 [l]	0.850	2.192	157.27	2.994
		4		
O9–H9A···OW1 [m]	0.850	2.460	111.04	2.877
O9–H9C···OW1 [l]	0.850	2.204	122.83	2.759
OW1–HW1A···O2 [o]	0.850	2.237	156.23	3.035
OW1–HW1A···O1 [n]	0.850	2.373	138.54	3.062
OW1–HW1B···OW2 [o]	0.850	2.389	132.38	3.028
OW1–HW1B···O4 [r]	0.850	2.654	148.39	3.407
OW2–HW2A···O6 [q]	0.850	2.038	157.09	2.840
OW2–HW2B···OW2 [s]	0.850	2.623	150.83	3.391

OW3–HW3A···OW2	0.850	2.135	136.40	2.812
OW3–HW3B···O5 [l]	0.850	2.169	157.43	2.972
5				
O9–H9C···OW2	0.850	2.316	122.21	2.861
OW1–HW1A···O2 [t]	0.850	2.430	121.11	2.960
OW2–HW2B···OW3	0.850	2.486	121.73	3.020
OW3–HW3A···O1	0.850	2.227	126.92	2.821
OW3–HW3B···OW1	0.850	1.967	166.36	2.800
6				
O9–H9B···OW3 [j]	0.850	2.276	124.73	2.847
O9–H9C···OW3	0.850	2.073	137.06	2.757
OW1–HW1B···O2 [j]	0.850	2.070	147.21	2.822
OW1–HW1B···O3 [j]	0.850	2.540	132.43	3.176
OW1–HW1C···OW2 [u]	0.850	2.000	161.07	2.818
OW2–HW2B···O1 [l]	0.850	2.129	165.94	2.961
OW3–HW3A···O8 [u]	0.850	2.288	140.37	2.992
OW3–HW3A···O7 [p]	0.850	2.347	147.41	3.097
OW3–HW3B···OW2 [u]	0.850	2.478	157.10	3.278
7				
O9–H9B···OW3 [v]	0.850	2.292	124.73	2.863
O9–H9C···OW3	0.850	2.071	136.94	2.754
OW1–HW1C···O1 [w]	0.850	2.233	142.43	2.953
OW2–HW2B···O8 [x]	0.850	2.322	167.84	3.158
OW2–HW2C···OW2 [y]	0.850	2.542	151.47	3.313
OW3–HW3A···O3 [z]	0.850	2.220	147.05	2.969
OW3–HW3A···O4 [o]	0.850	2.319	153.86	3.104
OW3–HW3B···OW1 [z]	0.850	2.457	157.13	3.257
OW3–HW3B···OW2 [aa]	0.850	2.462	121.62	2.997
8				
O9–H9B···OW1	0.850	2.289	124.52	2.858
O9–H9C···OW1 [l]	0.850	2.062	137.18	2.747
OW1–HW1A···O5 [x]	0.850	2.109	172.92	2.954
OW1–HW1A···O6 [bb]	0.850	2.600	119.86	3.112
OW1–HW1B···OW2	0.850	2.349	130.18	2.969
OW1–HW1B···OW3 [q]	0.850	2.568	139.90	3.264
OW2–HW2A···OW1	0.850	2.330	132.24	2.969
OW2–HW2A···O1	0.850	2.598	123.08	3.144
OW2–HW2B···O4	0.850	2.286	122.83	2.838
OW3–HW3B···OW1 [p]	0.850	2.547	142.75	3.264
9				
O9–H9B···OW3	0.850	2.059	136.73	2.741
O9–H9C···OW3 [l]	0.850	2.294	124.16	2.859
OW1–HW1B···O5	0.850	2.073	148.77	2.835
OW1–HW1C···OW1 [w]	0.850	2.553	157.40	3.354

OW2–HW2A···O6 [cc]	0.850	2.379	121.69	2.916
OW3–HW3B···OW1 [l]	0.850	2.398	120.22	2.920
OW3–HW3B···OW2	0.850	2.462	157.26	3.263
OW3–HW3C···O3 [bb]	0.850	2.186	147.95	2.942
OW3–HW3C···O4 [x]	0.850	2.342	152.56	3.122
10				
O9–H9B···OW1	0.850	2.272	125.55	2.851
O9–H9C···OW1 [dd]	0.850	2.082	137.15	2.766
OW1–HW1A···O5 [u]	0.850	2.365	156.42	3.163
OW1–HW1A···O6 [q]	0.850	2.503	113.05	2.942
OW1–HW1B···OW3 [ee]	0.850	2.422	151.33	3.193
OW2–HW2A···OW1	0.850	2.236	140.69	2.943
OW2–HW2A···O2	0.850	2.546	125.16	3.114
OW2–HW2B···O3	0.850	2.194	131.97	2.834
OW3–HW3B···O1 [ff]	0.850	2.370	118.40	2.874
11				
O9–H9A···OW2 [gg]	0.820	2.296	127.13	2.866
OW1–HW1B···O1 [hh]	0.850	2.306	132.60	2.948
OW2–HW2A···OW1 [ii]	0.850	2.373	153.55	3.157
OW2–HW2C···O7 [ii]	0.850	2.225	140.86	2.934
OW2–HW2C···O8 [o]	0.850	2.484	140.06	3.183
OW3–HW3D···O2	0.850	2.082	148.03	2.839
OW3–HW3A···OW1 [x]	0.850	2.213	116.68	2.704
OW3–HW3A···OW2 [gg]	0.850	2.218	149.79	2.983

[#] Symmetry transformations used to generate equivalent atoms: (a) x, –y+1/2, z+1/2; (b) –x+2, y+1/2, –z+3/2; (c) –x+2, y–1/2, –z+3/2; (d) x, –y–1/2, z–1/2; (e) –x+1, –y, –z+2; (f) –x+1, y–1/2, –z+3/2; (g) –x+1, y+1/2, –z+1/2; (h) –x+1, y–1/2, –z+1/2; (i) –x, y–1/2, –z+1/2; (j) –x, –y+1, –z; (k) –x, y+1/2, –z+1/2;(l) –x, –y+1, –z+1; (m) x, y, z–1; (n) x–1/2, –y+1/2, z+1/2; (o) –x+1/2, y–1/2, –z+3/2; (p) –x+1/2, y–1/2, –z+1/2; (q) –x+1/2, y+1/2, –z+1/2; (r) x, y, z+1; (s) –x, –y+2, –z+1; (t) x+1/2, –y+3/2, z–1/2; (u) x–1/2, –y+1/2, z–1/2; (v) –x+1, –y+1, –z+2; (w) –x+1, –y+1, –z+1; (x) –x+1/2, y+1/2, –z+3/2; (y) –x+1, –y+2, –z+1; (z) x+1/2, –y+1/2, z+1/2; (aa) x+1/2, –y+3/2, z+1/2; (bb) x–1/2, –y+3/2, z–1/2; (cc) –x, –y+1, –z+2; (dd) –x, –y, –z; (ee) –x+1, –y, –z; (ff) x+1/2, –y–1/2, z–1/2; (gg) –x, –y, –z+1; (hh) –x, –y, –z+2; (ii) x–1/2, –y–1/2, z–1/2.

Table S4 Important hydrogen bond interactions in complexes **12**–**16**[#]

D–H···A	D–H / Å	H···A/ Å	D···A / Å	<D–H···A/ º
		12		
O9–H9B···O4	0.820	2.256	119.72	2.756
O9–H9B···O3 [a]	0.820	2.628	121.84	3.139
O10–H10A···O8	0.820	1.954	147.52	2.682
O11–H11A···OW2 [b]	0.820	2.401	163.35	3.195
O11–H11A···OW2 [c]	0.820	2.559	117.64	3.028
O11–H11B···N3 [d]	0.850	2.469	160.92	3.284
OW1–HW1A···O10 [b]	0.850	2.415	161.93	3.234
OW1–HW1A···O2 [b]	0.850	2.560	116.15	3.032
OW2–HW2B···O11 [e]	0.850	2.401	131.04	3.028
OW2–HW2C···O7 [f]	0.850	2.422	160.11	3.234
OW3–HW3A···O9 [e]	0.850	2.135	123.95	2.704
		13		
O9–H9B···O5	0.960	2.030	121.21	2.658
O10–H10A···O2 [g]	0.820	2.468	167.94	3.274
O10–H10B···O4	0.960	1.820	164.54	2.757
O10–H10B···O3 [g]	0.960	2.536	117.25	3.096
O11–H11A···OW2 [h]	0.820	2.326	167.78	3.132
O11–H11A···OW2	0.820	2.435	115.95	2.889
O11–H11B···N4 [i]	0.960	2.332	159.74	3.250
OW1–HW1B···O10 [h]	0.850	2.045	125.12	2.628
OW2–HW2A···OW2 [h]	0.850	1.395	141.35	2.126
OW2–HW2A···OW2 [j]	0.850	2.380	121.13	2.912
OW2–HW2A···OW2 [k]	0.850	2.502	110.64	2.912
OW3–HW3A···O4 [h]	0.850	2.485	155.37	3.276
		14		
O9–H9A···O4 [l]	0.850	2.457	161.35	3.274
O9–H9B···O2 [l]	0.850	1.914	162.84	2.738
O10–H10A···OW1 [m]	0.850	2.544	145.17	3.278
O10–H10B···O8	0.850	1.977	137.70	2.667
O11–H11A···OW1	0.850	2.208	126.16	2.795
O11–H11A···O5	0.850	2.620	113.26	3.057
O11–H11B···OW2 [n]	0.850	2.373	139.58	3.070
OW1–HW1A···OW2	0.850	2.634	123.45	3.183
OW1–HW1B···O2 [o]	0.850	2.564	140.75	3.267
OW1–HW1B···O1 [o]	0.850	2.637	157.17	3.436
OW2–HW2A···O7 [n]	0.850	2.576	133.77	3.223
OW2–HW2A···OW1	0.850	2.653	121.67	3.183
OW3–HW3A···O9 [l]	0.850	2.245	110.55	2.665
		15		
O9–H9A···OW2	0.820	2.226	115.80	2.687
O9–H9B···O2 [o]	0.960	2.175	116.19	2.737

O9–H9B···O1 [o]	0.960	2.548	118.37	3.120
O11–H11A···OW1 [p]	0.820	2.219	141.73	2.908
O11–H11B···N3 [p]	0.960	2.286	160.59	3.207
OW2–HW2A···O9 [l]	0.850	2.232	113.48	2.687
OW2–HW2B···O9 [n]	0.850	2.145	121.28	2.687
OW3–HW3B···O11 [o]	0.850	2.323	113.93	2.778
OW3–HW3B···O3 [o]	0.850	2.441	112.032.870	
16				
O9–H9B···O7	0.850	2.125	122.78	2.682
O10–H10A···O3 [q]	0.820	2.478	166.54	3.281
O10–H10B···O2	0.850	1.925	162.48	2.747
O10–H10B···O1 [q]	0.850	2.647	117.20	3.128
O11–H11B···N3 [r]	0.820	2.416	160.98	3.203
O11–H11C···OW2	0.850	2.034	142.93	2.760
OW1–HW1B···O10 [q]	0.850	2.194	117.96	2.699
OW2–HW2A···O2 [h]	0.850	2.518	154.41	3.305
OW3–HW3B···O8 [h]	0.850	2.638	126.17	3.214
OW3–HW3C···OW3 [h]	0.850	1.404	114.26	1.917
OW3–HW3C···O11 [h]	0.850	2.407	137.93	3.091

[#] Symmetry transformations used to generate equivalent atoms: (a) x–1/4, –y+7/4, –z–1/4; (b) x–1/4, –y+5/4, z–1/4; (c) –x+7/4, y+3/4, z–1/4; (d) –x+2, –y+1, –z–1; (e) x–3/4, –y+7/4, z+1/4; (f) –x+2, –y+1/2, z; (g) –x+3/4, y–3/4, –z+3/4; (h) –x, –y+3/2, z; (i) –x, –y+2, –z; (j) –x+3/4, y–3/4, –z–1/4; (k) x+3/4, –y+3/4, –z–1/4;(l) x+1/4, –y+5/4, –z+1/4; (m) –x+5/4, y+1/4, z+1/4; (n) –x+1, –y+3/2, z; (o) y–1/4, –x+5/4, –z+1/4; (p) –x+1/2, –y+3/2, –z–1/2; (q) –y+3/4, x+3/4, –z+3/4; (r) –x, –y+1, –z.

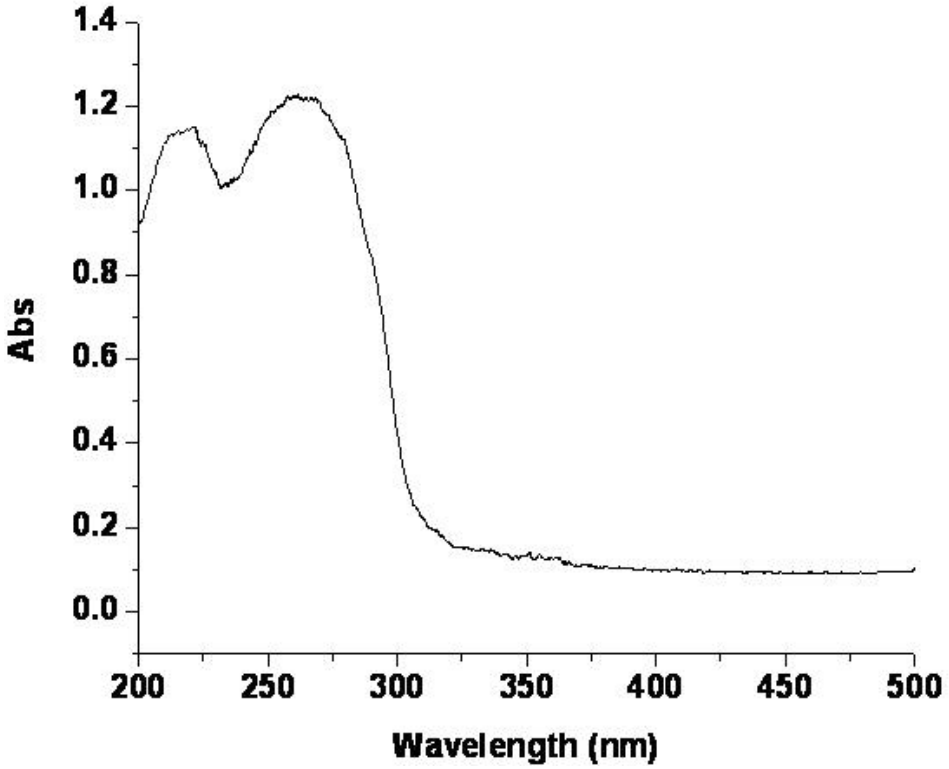

Fig. S1 UV – Vis absorption spectrum of the oxalic acid ligand in solid–state.

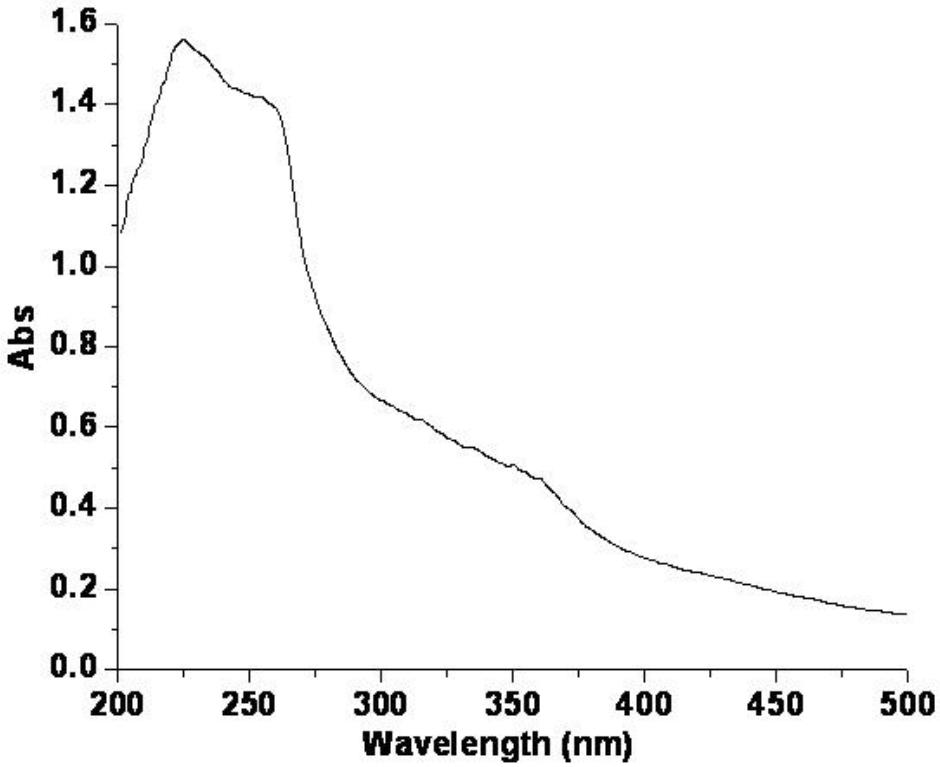

Fig. S2 UV–Vis absorption spectrum of the imidazole–4,5–dicarboxylate ligand in solid–state.

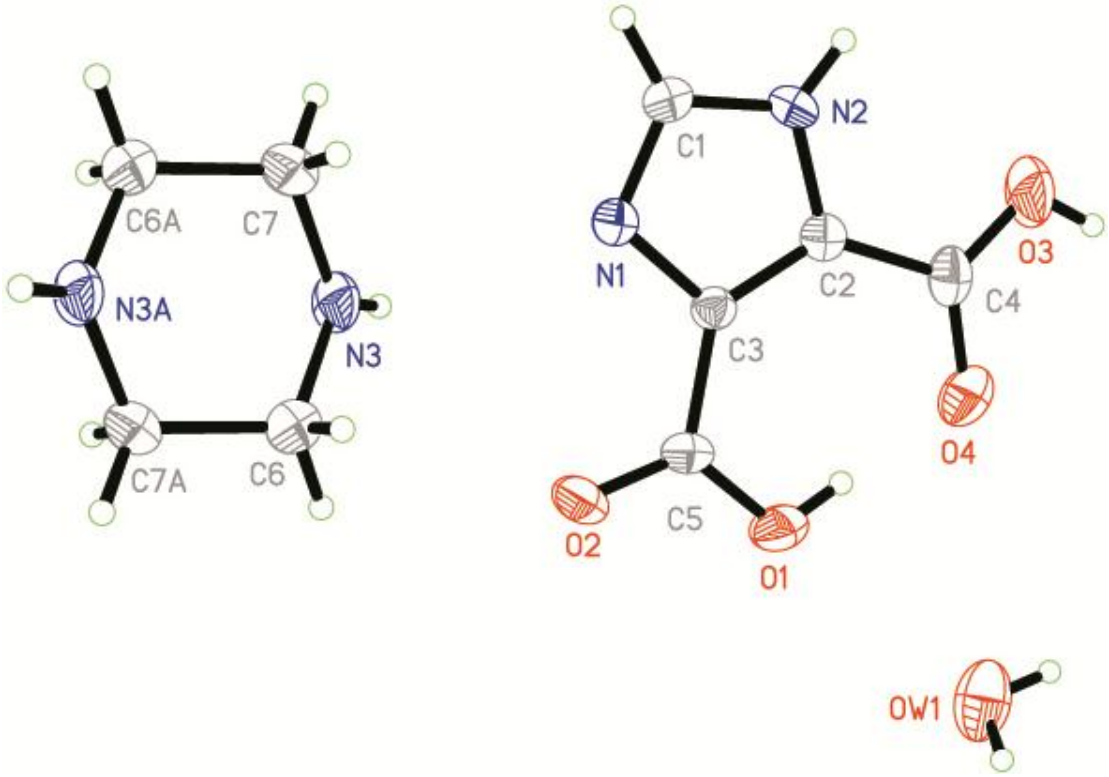

Fig. S3 The structural unit of $[(H_3IMDC)_2 \cdot (PZ)_{0.5} \cdot (H_2O)]$ with labeling scheme and 50% thermal ellipsoids.

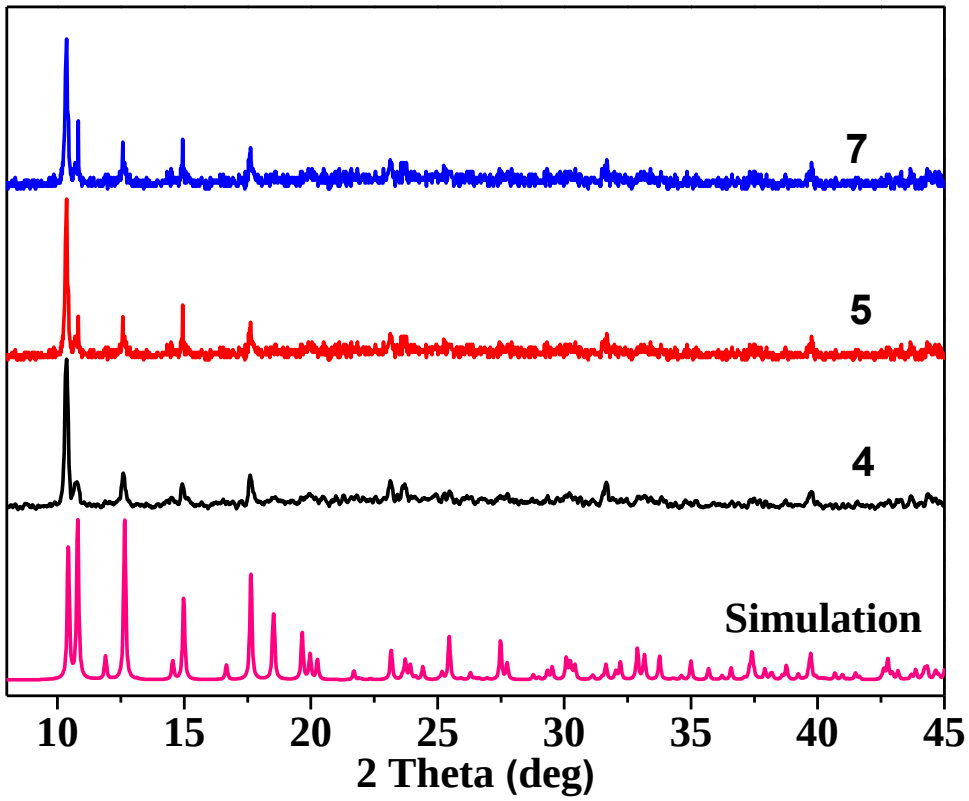

Fig. S4 PXRD patterns of **4**, **5** and **7** compared with a simulated pattern.

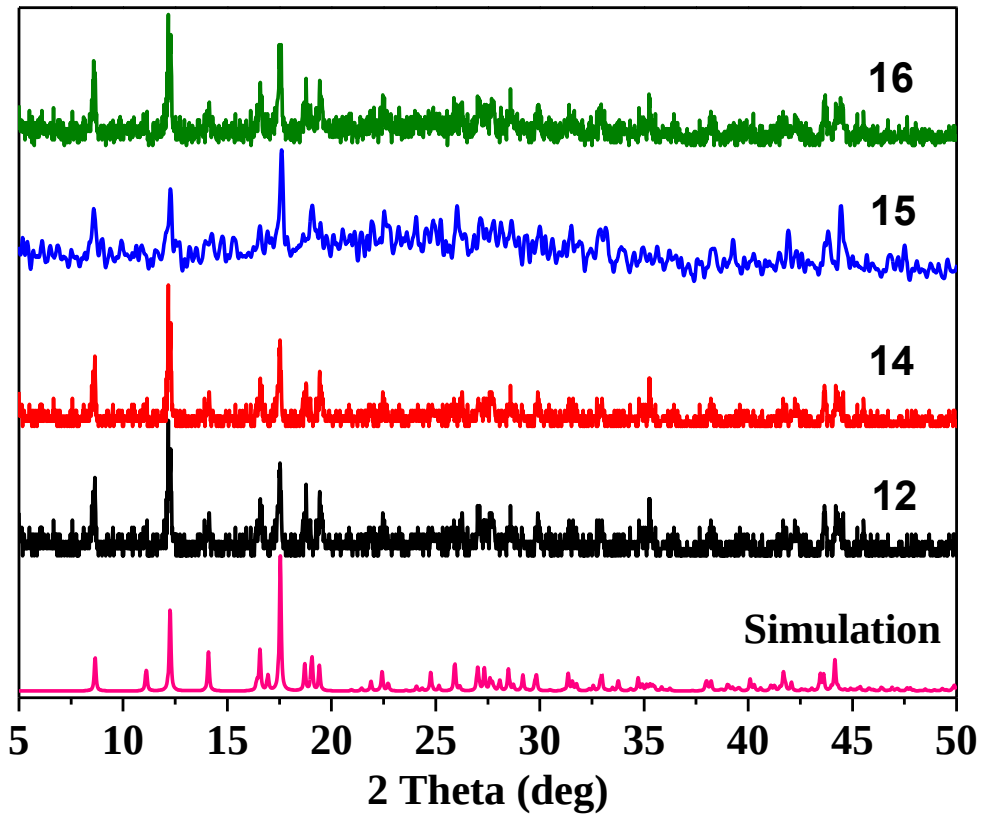

Fig. S5 PXRD patterns of **12**, **14**, **15** and **16** compared with a simulated pattern.

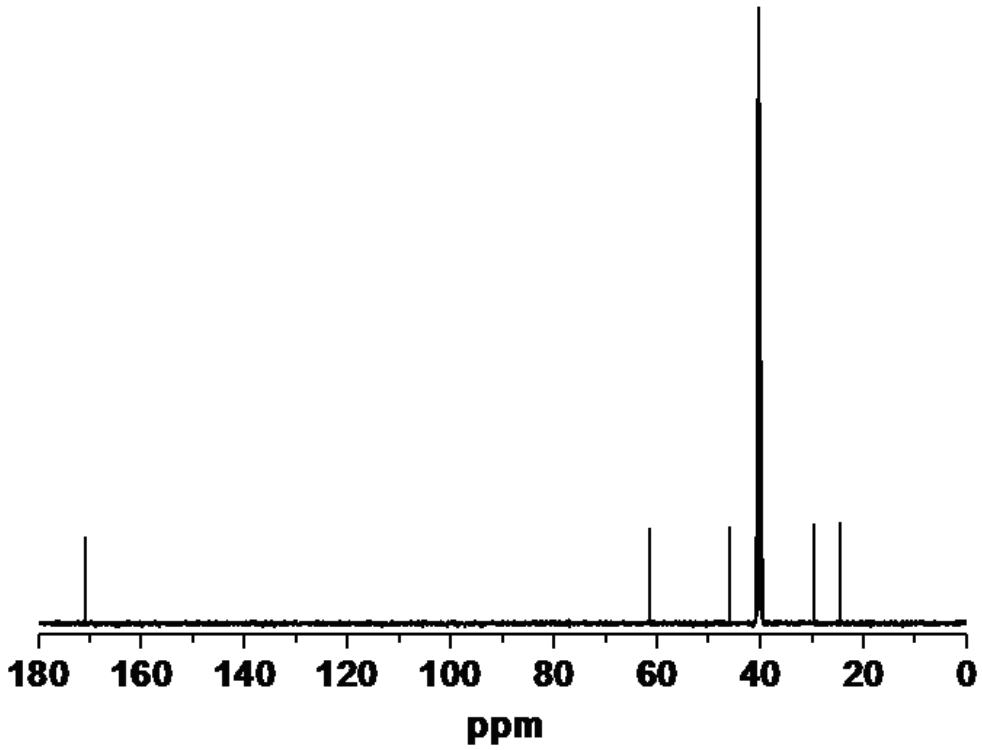

Fig. S6 The ^{13}C NMR spectrum of L-proline.

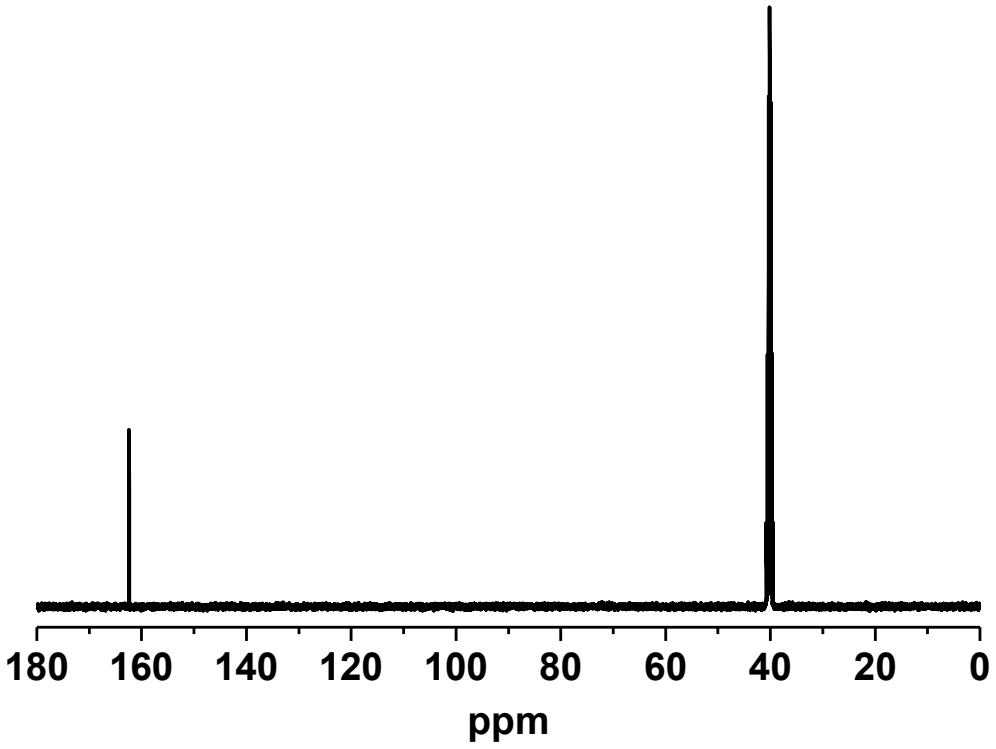

Fig. S7 The ^{13}C NMR spectrum of the reaction product.