Figure S1 Experimental and theoretical deformation density maps of barbituric acid and urea molecules in the polymorph (I). a) and c) experimental charge density, b) and d) theoretical charge density refined using the multipolar refinement.

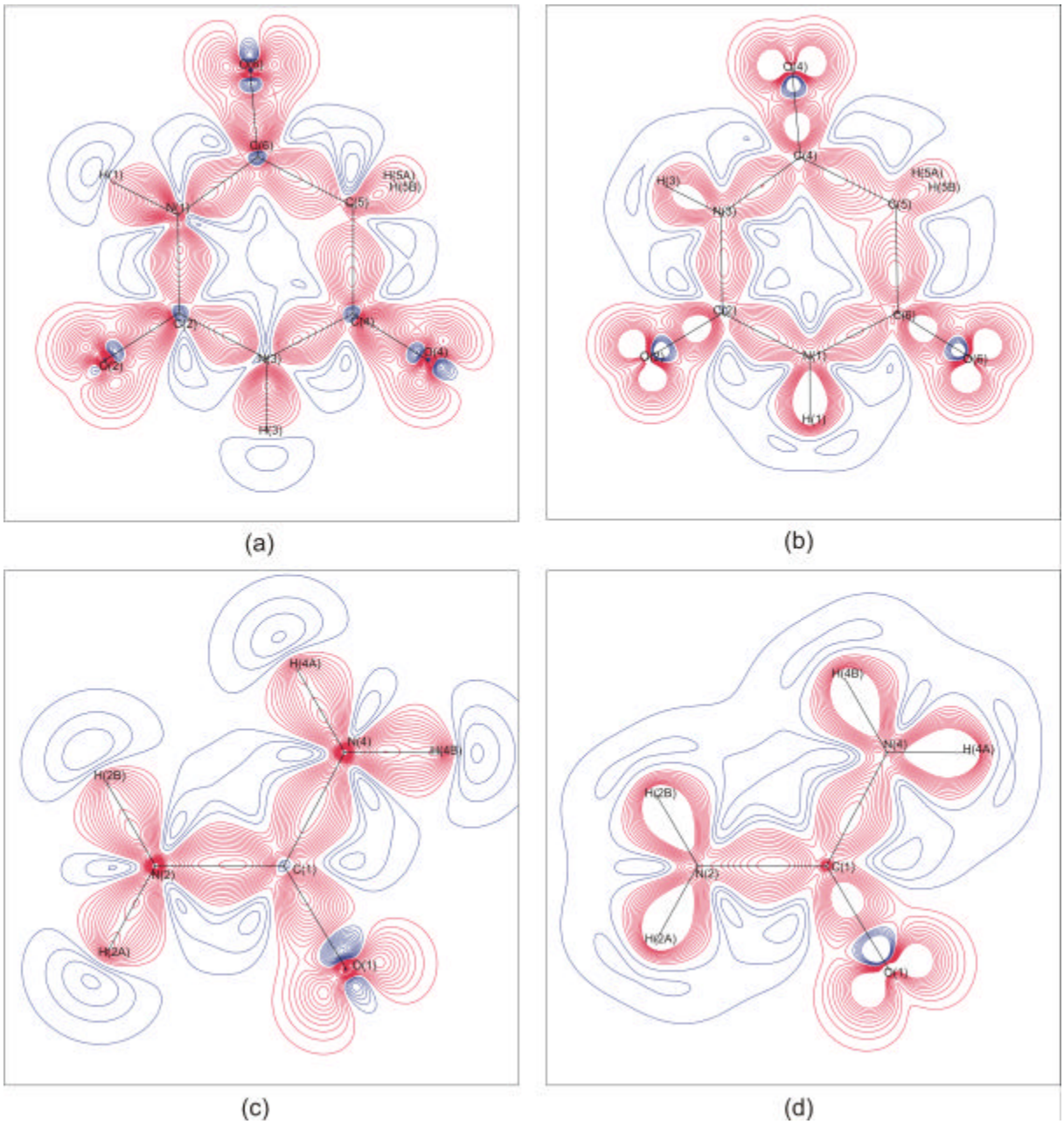

Figure S2 Experimental and theoretical deformation density maps of barbituric acid and urea molecules in the polymorph (II). a) and c) experimental charge density, b) and d) theoretical charge density refined using the multipolar refinement.

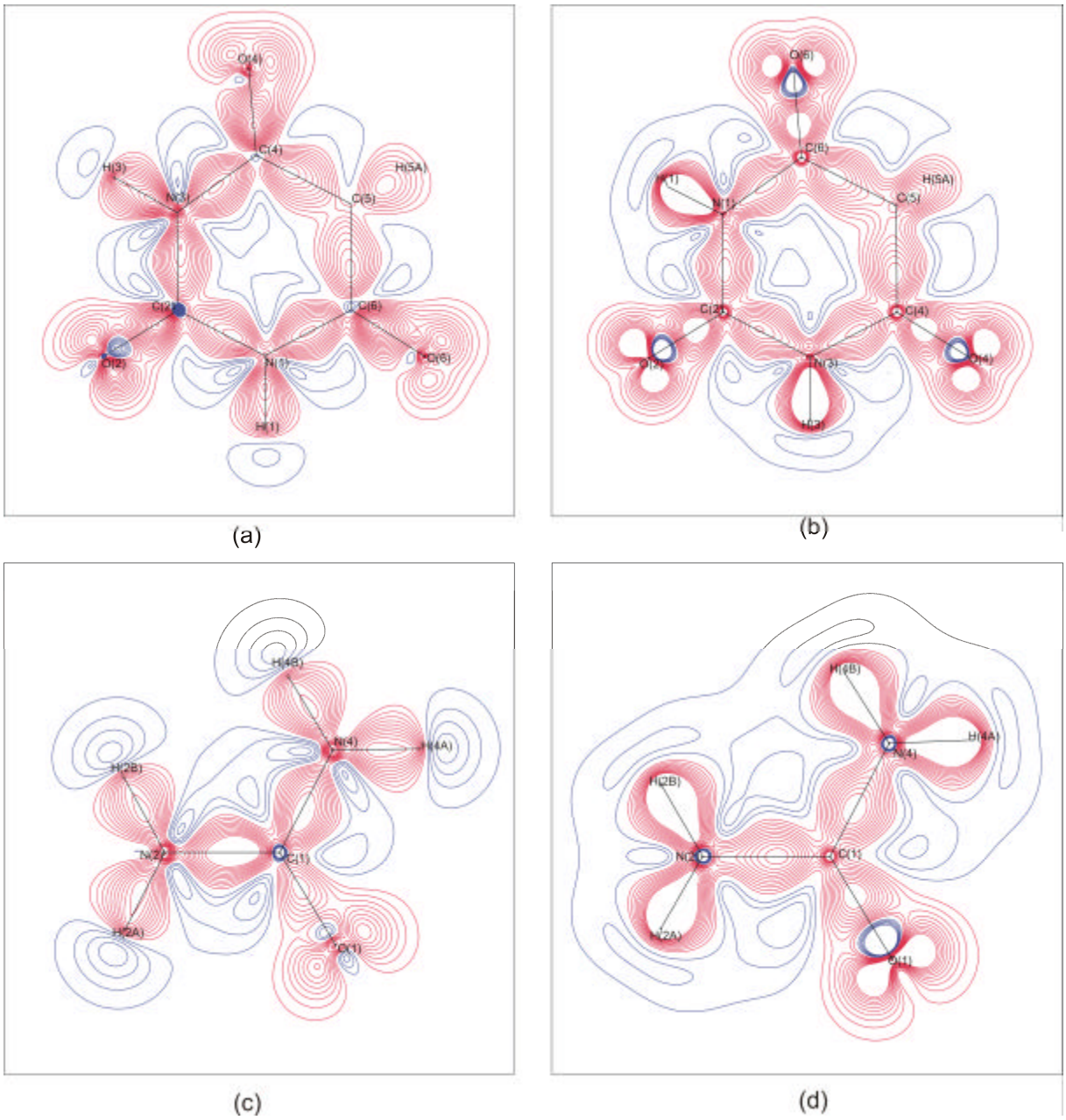

Figure S3 Exponential fitting of local kinetic energy density G(rCP) /eÅ^{-5} and local potential energy density V(rCP) /eÅ^{-5} on Rij /Å values.

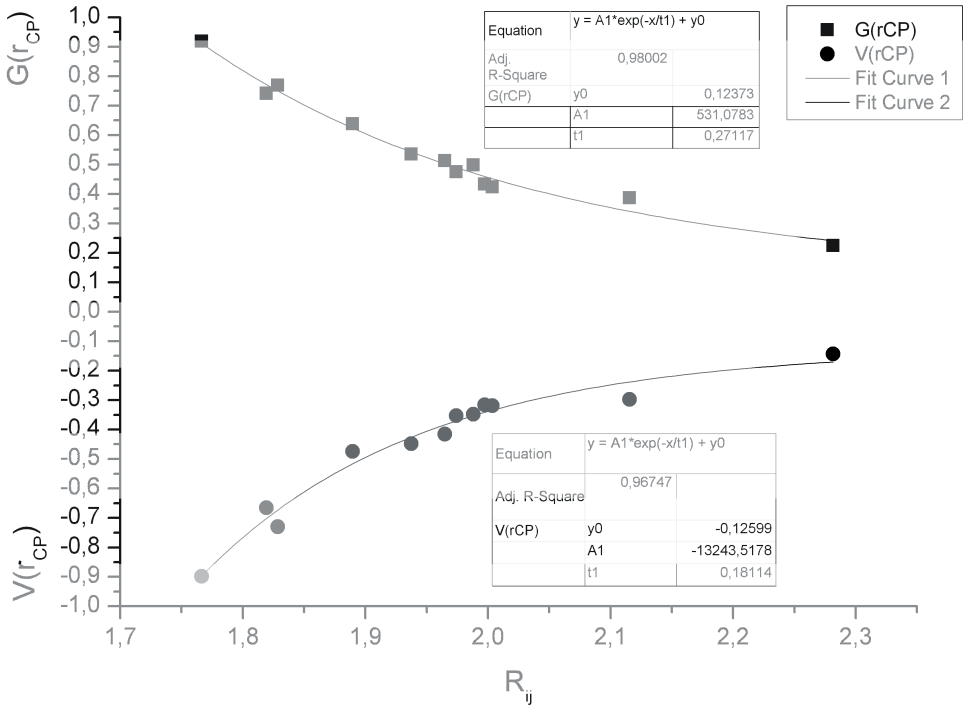

Table S1 Bond lengths and angles for (I) and (II) after multipolar refinement.

	(I)	(II)
Barbituric acid		
O(2)–C(2)	1.2237(6)	1.2292(1)
O(4)–C(4)	1.2315(5)	1.2247(9)
O(6)–C(6)	1.2188(5)	1.2229(9)
N(1)–C(2)	1.3706(5)	1.3757(9)
N(1)–C(6)	1.3777(5)	1.3726(9)
N(3)–C(2)	1.3828(5)	1.3758(1)
N(3)–C(4)	1.3633(5)	1.3729(9)
C(4)–C(5)	1.4952(5)	1.5043(9)
C(5)–C(6)	1.5018(5)	1.5010(1)
Urea		
O(1)–C(1)	1.2579(6)	1.2649(1)
N(2)–C(1)	1.3483(5)	1.3447(1)
N(4)–C(1)	1.3453(5)	1.3457(1)
Barbituric acid		
C(2) –N(1) – C(6)	125.37(3)	124.93(6)
C(2) –N(3) – C(4)	124.79(3)	125.16(6)
O(2) –C(2) – N(1)	121.78(4)	121.04(7)
O(2) –C(2) – N(3)	120.42(4)	120.94(7)
N(1) –C(2) – N(3)	117.80(4)	118.02(7)
O(4) –C(4) – N(3)	120.75(4)	120.70(7)
O(4) –C(4) – C(5)	121.86(4)	122.26(7)
N(3) –C(4) – C(5)	117.39(3)	117.01(6)
C(4) –C(5) – C(6)	117.25(3)	116.35(6)
O(6) –C(6) – N(1)	120.97(4)	120.10(7)
O(6) –C(6) – C(5)	122.40(4)	122.74(7)
N(1) –C(6) – C(5)	116.63(3)	117.15(6)
Urea		
O(1) –C(1) – N(2)	121.07(4)	121.22(7)
O(1) –C(1) – N(4)	120.82(4)	117.49(7)
N(2) –C(1) – N(4)	118.11(3)	117.49(7)