Table 1S Final atomic coordinates for bicyclo[3.3.1]nonane-2,6-dione at 80 K after restrained refinement in the monoclinic space group *C*2/*c* by the Rietveld method. The atom numbering scheme is shown in Fig. 2a. Uiso*100 = 0.732(2) Å^2; *a* = 7.38032(4) Å, *b* = 10.38202(5) Å, *c* = 9.75077(7) Å, β =95.359(1)º, *V* = 743.86(2)Å^3; ρ_{calc} = 1.358 g cm^{-3}; *Z* = 4.

Atom	x/a	y/b	z/c
O1	0.5943(2)	0.1352(1)	0.0522(1)
C1	0.4295(2)	0.2404(2)	-0.1426(2)
C2	0.5775(2)	0.1515(2)	-0.0714(1)
C3	0.6962(2)	0.0797(1)	-0.1660(2)
C4	0.7355(2)	0.1582(2)	-0.2968(2)
C9	0.5	0.3287(4)	-0.25
H1	0.377(1)	0.2966(5)	-0.0617(5)
H2	0.63287(7)	-0.0114(4)	-0.1931(5)
H3	0.8205(5)	0.0557(5)	-0.1052(4)
H4	0.793(1)	0.0945(5)	-0.3692(8)
H5	0.836(1)	0.231(1)	-0.2690(5)
H6	0.3898(4)	0.3853(2)	0.2983(3)

Table 2S. Final atomic coordinates for bicyclo[3.3.1]nonane-3,7-dione at 100 K after restrained refinement in the tetragonal space group $P4_12_12$ by the Rietveld method. The atom numbering scheme is shown in Fig. 2b. Biso = 2.16(1) Å^2; *a* = 6.8558(1) Å, *c* = 16.9375(1) Å, *V* = 796.10(1) Å^3; ρ_{calc} = 1.268 g cm^{-3}; *Z* = 4.

Atom	x/a	y/b	z/c
O1	0.5505(2)	0.1570(2)	0.3279(1)
C1	0.3270(2)	0.4997(2)	0.1951(1)
C2	0.3169(2)	0.3119(2)	0.2450(1)
C3	0.4679(3)	0.3045(2)	0.3115(1)
C4	0.4932(3)	0.4900(2)	0.3605(1)
C9	0.3253(7)	0.6783(3)	0.25
H1	0.1994(5)	0.5029(4)	0.1578(2)
H2	0.1754(3)	0.3033(5)	0.2728(2)
H3	0.3333(5)	0.1823(3)	0.2095(1)
H4	0.396(1)	0.477(1)	0.4103(4)
H5	0.6398(6)	0.492(1)	0.3837(6)
H6	0.193(2)	0.736(2)	0.276(1)