Table S1. Experimental details

For all structures: $CoH_{15}N_6O_2^+ \cdot 2(Br^-)$, $M_r = 349.93$, $Z = 4$. Experiments were carried out at 295 K with Mo *K*a radiation using a Xcalibur, Ruby, Gemini diffractometer. H-atom parameters were constrained.

	0.0001 GPa	0.15 GPa	0.58 GPa	0.97 GPa
Crystal data				
Crystal system, space group	Monoclinic, *C*2/*c*	Monoclinic, *C*2/*c*	Monoclinic, *C*2/*c*	Monoclinic, *C*2/*c*
a, *b*, *c* (Å)	10.6807 (6), 8.8299 (8), 10.9759 (5)	10.6575 (5), 8.7649 (9), 10.9567 (5)	10.6184 (4), 8.6082 (8), 10.9307 (4)	10.6243 (5), 8.4030 (8), 10.9113 (5)
α, β, γ (°)	90, 94.604 (5), 90	90, 94.589 (5), 90	90, 94.591 (4), 90	90, 94.483 (4), 90
V (Å^3)	1031.79 (12)	1020.21 (12)	995.92 (11)	971.14 (11)
No. of reflections for cell measurement	1571	1795	1799	1835
θ range (°) for cell measurement	2.8–27.4	2.8–27.8	2.8–28.1	2.8–27.6
μ (mm^{-1})	9.38	9.49	9.72	9.97
Crystal size (mm)	0.20 × 0.20 × 0.05	0.20 × 0.20 × 0.05	0.20 × 0.20 × 0.05	0.20 × 0.20 × 0.05
Data collection				
T_{min}, T_{max}	0.233, 0.302	0.231, 0.300	0.228, 0.297	0.225, 0.293
No. of measured, independent and observed [$I > 2\sigma(I)$] reflections	2634, 605, 532	2719, 588, 541	2610, 569, 518	2651, 562, 522
R_{int}	0.059	0.056	0.054	0.051
$(\sin\theta/\lambda)_{max}$ (Å^{-1})	0.625	0.625	0.625	0.624
Range of *h*, *k*, *l*	*h* = -12→12, *k* = -8→8, *l* = -13→13	*h* = -12→12, *k* = -8→8, *l* = -13→13	*h* = -12→12, *k* = -8→8, *l* = -13→13	*h* = -12→12, *k* = -8→8, *l* = -13→13
Refinement				
$R[F^2 > 2\sigma(F^2)]$, $wR(F^2)$, *S*	0.029, 0.068, 1.11	0.027, 0.058, 1.08	0.027, 0.063, 1.06	0.024, 0.056, 1.12
No. of reflections	605	588	569	562
No. of parameters	52	52	52	52
No. of restraints	0	0	0	0
$\Delta\rho_{max}$, $\Delta\rho_{min}$ (e Å^{-3})	0.40, -0.36	0.50, -0.30	0.31, -0.34	0.33, -0.32

	1.61 GPa	2.60 GPa	3.30 GPa	3.80 GPa
Crystal data				
Crystal system, space group	Monoclinic, *C*2/*c*	Monoclinic, *C*2/*c*	Monoclinic, *C*2/*c*	Monoclinic, *C*2/*c*
a, *b*, *c* (Å)	10.6792 (4), 8.0977 (7), 10.9004 (4)	10.6750 (5), 7.8148 (8), 10.8204 (4)	10.6557 (4), 7.7006 (7), 10.7594 (4)	10.6379 (4), 7.6361 (6), 10.7112 (3)
α, β, γ (°)	90, 94.192 (4), 90	90, 93.829 (4), 90	90, 93.693 (4), 90	90, 93.601 (3), 90
V (Å^3)	940.11 (10)	900.66 (11)	881.03 (9)	868.38 (8)
No. of reflections for cell measurement	1813	1806	1745	1743
θ range (°) for cell measurement	3.2–27.8	2.8–27.7	2.8–28.0	2.8–28.2
μ (mm^{-1})	10.30	10.75	10.99	11.15
Crystal size (mm)	0.20 × 0.20 × 0.05	0.20 × 0.20 × 0.05	0.20 × 0.20 × 0.05	0.20 × 0.20 × 0.05
Data collection				
T_{min}, T_{max}	0.219, 0.288	0.215, 0.282	0.212, 0.279	0.210, 0.277
No. of measured, independent and observed [$I > 2\sigma(I)$] reflections	2485, 528, 490	2359, 502, 457	1398, 484, 439	1833, 478, 443
R_{int}	0.055	0.056	0.041	0.049
$(\sin\theta/\lambda)_{max}$ (Å^{-1})	0.624	0.625	0.624	0.625
Range of *h*, *k*, *l*	*h* = -12→12, *k* = -7→7, *l* = -13→13	*h* = -12→12, *k* = -7→7, *l* = -13→13	*h* = -12→12, *k* = -7→7, *l* = -13→12	*h* = -12→12, *k* = -7→7, *l* = -13→13
Refinement				
$R[F^2 > 2\sigma(F^2)]$, $wR(F^2)$, *S*	0.024, 0.053, 1.06	0.023, 0.055, 1.11	0.025, 0.051, 1.12	0.022, 0.050, 1.08
No. of reflections	528	502	484	478
No. of parameters	52	52	52	52
No. of restraints	0	0	0	42
$\Delta\rho_{max}$, $\Delta\rho_{min}$ (e Å^{-3})	0.31, -0.45	0.30, -0.35	0.32, -0.32	0.33, -0.26

	4.40 GPa	4.80 GPa	5.50 GPa	6.01 GPa
Crystal data				

Crystal system, space group	Monoclinic, *C*2/*c*	Monoclinic, *C*2/*c*	Monoclinic, *C*2/*c*	Monoclinic, *C*2/*c*
a, *b*, *c* (Å)	10.6148 (4), 7.5680 (7), 10.6476 (4)	10.6023 (5), 7.5308 (7), 10.6049 (4)	10.5931 (4), 7.4783 (7), 10.5357 (4)	10.5914 (5), 7.4435 (7), 10.4601 (4)
α, β, γ (°)	90, 93.508 (3), 90	90, 93.439 (4), 90	90, 93.300 (4), 90	90, 93.121 (4), 90
V (Å^3)	853.75 (9)	845.21 (9)	833.24 (9)	823.42 (9)
No. of reflections for cell measurement	1712	1704	1697	1683
θ range (°) for cell measurement	2.6–28.2	2.6–27.8	2.8–27.9	2.8–28.0
μ (mm^{-1})	11.34	11.45	11.62	11.76
Crystal size (mm)	0.20 × 0.20 × 0.05	0.20 × 0.20 × 0.05	0.20 × 0.20 × 0.05	0.20 × 0.20 × 0.05
Data collection				
T_{min}, T_{max}	0.208, 0.274	0.207, 0.272	0.205, 0.270	0.205, 0.268
No. of measured, independent and observed [$I > 2\sigma(I)$] reflections	2262, 488, 455	2236, 482, 451	2182, 475, 441	2170, 469, 438
R_{int}	0.055	0.055	0.050	0.056
$(\sin\theta/\lambda)_{max}$ (Å^{-1})	0.625	0.625	0.625	0.625
Range of *h*, *k*, *l*	*h* = -12→12, *k* = -7→7, *l* = -13→13	*h* = -12→12, *k* = -7→7, *l* = -13→13	*h* = -12→12, *k* = -7→7, *l* = -12→12	*h* = -12→12, *k* = -7→7, *l* = -12→12
Refinement				
$R[F^2 > 2\sigma(F^2)]$, $wR(F^2)$, *S*	0.024, 0.058, 1.08	0.025, 0.060, 1.07	0.023, 0.056, 1.10	0.026, 0.062, 1.09
No. of reflections	488	482	475	469
No. of parameters	52	52	52	52
No. of restraints	0	0	0	0
$\Delta\rho_{max}$, $\Delta\rho_{min}$ (e Å^{-3})	0.33, -0.30	0.39, -0.38	0.44, -0.36	0.46, -0.39

	6.85 GPa_HP_phase
Crystal data	
Crystal system, space group	Orthorhombic, *Cmcm*
a, *b*, *c* (Å)	10.7595 (5), 7.4390 (6), 10.0890 (4)

α, β, γ (°)	90, 90, 90
V (Å^3)	807.52 (8)
No. of reflections for cell measurement	1696
θ range (°) for cell measurement	2.8–27.6
μ (mm^{-1})	11.99
Crystal size (mm)	0.20 × 0.20 × 0.05
Data collection	
T_{min}, T_{max}	0.202, 0.265
No. of measured, independent and observed [$I > 2\sigma(I)$] reflections	2137, 333, 310
R_{int}	0.062
$(\sin \theta/\lambda)_{max}$ (Å^{-1})	0.625
Range of h, k, l	h = -12→12, k = -7→7, l = -12→12
Refinement	
$R[F^2 > 2\sigma(F^2)]$, $wR(F^2)$, S	0.027, 0.064, 1.05
No. of reflections	333
No. of parameters	36
No. of restraints	42
$\Delta\rho_{max}$, $\Delta\rho_{min}$ (e Å^{-3})	0.64, -0.52