STRUCTURAL SCIENCE
CRYSTAL ENGINEERING
MATERIALS

Volume 78 (2022)

Supporting information for article:

Dynamics and disorder: on the stability of pyrazinamide polymorphs

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Figure S1. Contributions of Coulomb and dispersion energies for pyrazinamide polymorphs.

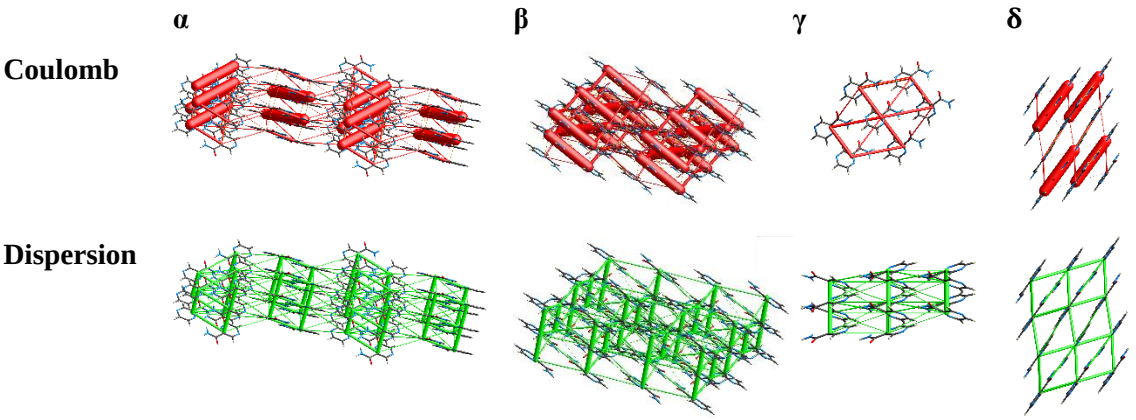

Fig S2. wR2 and ADPs after NoMoRe refinement against 120 K data, 6 frequencies refined. Ellipsoids are shown at the 90 % probability level. Hydrogen atoms are not shown.

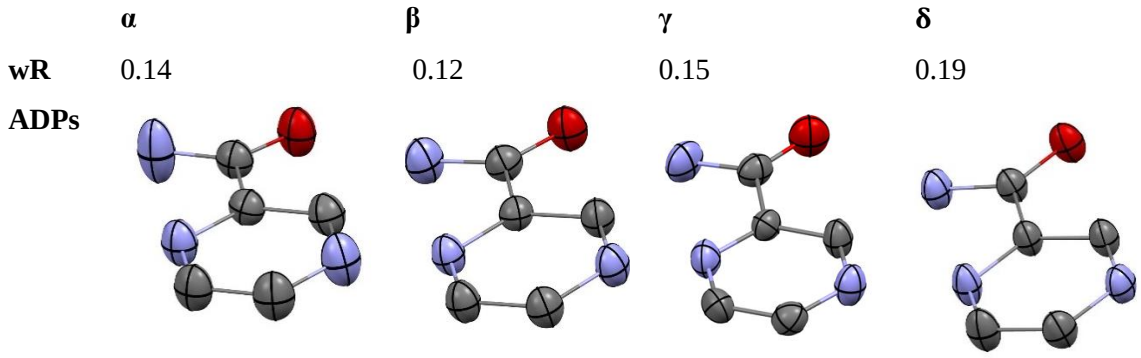

Table S2. Vibrational contributions to total energy (kJ/mol) directly from periodic *ab-initio* calculations B3LYP, pVTZ, ZPE- zero point energy, Hvib-vibrational contributions to enthalpy, S- entropy, Z- number of molecules in the unit cell. Top value – frequencies calculated at unit cell from 120 K, bottom – frequencies calculated at unit cell from room temperature. Thermodynamic values are calculated at 298 K.

	α	β	γ	δ
	1116.2	1121.0	559.6	561.6
ZPE	*1115.1*	*1120.3*	*559.0*	*560.2*
	82.2	81.8	38.2	36.8
Hvib	*82.5*	*82.3*	*38.4*	*37.3*
	172.3	163.3	74.0	68.5
TS	*176.1*	*166.0*	*75.1*	*71.1*
	1026.0	1039.6	523.7	529.8
Hvib+ZPE-S	*1021.5*	*1036.6*	*522.3*	*526.4*
	256.5	259.9	261.9	264.9
(Hvib+ZPE-S)/Z	*255.4*	*259.1*	*261.1*	*263.2*

Table S3. Contributions to the free energy from vibrations (Hvib +ZPE-TS) for four polymorphic forms as a function of temperature obtained after NoMoRe. Temperature is in Kelvin, energies in kJ/mol.

Temperature	Alpha	Beta	Gamma	Delta
10	280	281	280	281
25	279	281	280	281
50	279	280	280	281
75	278	280	279	280
100	277	278	277	279
125	275	277	276	278
150	273	275	274	276
175	270	273	271	274
200	268	270	268	271
250	261	265	262	265
300	254	258	255	259
350	245	249	246	251
400	235	240	237	242
450	225	230	226	232
500	213	219	214	221
550	200	207	201	209

Table S4. Comparison of vibrational contributions to the free energy calculated from frequencies after NoMoRe and purely from DFT calculations at 298K. All values in kJ/mol.

	α	**β**	**γ**	**δ**
DFT (CRYSTAL17)	256,5	259,9	261,9	264,9
NoMoRe	254	258	255	259

Table S5. Differences of enthalpies, which were used for G-T diagrams construction (kJ/mol)

	α	**β**	**γ**	**δ**
Model 1:				
periodic DFT (B3LYP, fixed cell, TZP, D3)	2.12	-1.79	0.53	0
Model 2:				
periodic DFT results from Wahlberg[1] et al., (model B3LYP-D/VTZ-mod)	2.65	0.76	3.57	0
Model 3:				
average values of experimentally observed enthalpy differences, as observed from DSC by Cherukuvada[2] et al.	0.36	0.88	1.66	0

[1]*Wahlberg, N., Ciochoń, P., Petriĉek, V. & Madsen, A. Ø. (2014). Crystal Growth & Design* ***14****, 381*
[2]*Cherukuvada, S., Thakuria, R. & Nangia, A. (2010). Crystal Growth & Design* ***10****, 3931*