Supporting Information

Exploiting the Synergy of Powder X-ray Diffraction and Solid-State NMR Spectroscopy in Structure Determination of Organic Molecular Solids

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Definitions Relating to GIPAW Calculated Shielding Tensors and Chemical Shifts

Tables S1 and S2 list the chemical shift tensor components calculated by the GIPAW approach. The principal components are labelled according to the Haeberlen convention:

$$\left|\sigma_{zz} - \sigma_{iso}\right| \geq \left|\sigma_{xx} - \sigma_{iso}\right| \geq \left|\sigma_{yy} - \sigma_{iso}\right|$$

The isotropic chemical shielding is given by:

$$\sigma_{iso} = \tfrac{1}{3}(\sigma_{xx} + \sigma_{yy} + \sigma_{zz}) \quad \text{(S1)}$$

while the reduced chemical shift anisotropy is defined as:

$$\delta_{aniso} = \sigma_{zz} - \sigma_{iso} = \tfrac{2}{3}\sigma_{zz} - \tfrac{1}{3}(\sigma_{xx} + \sigma_{yy}) \quad \text{(S2)}$$

The asymmetry is defined as:

$$\eta = (\sigma_{yy} - \sigma_{xx})/(\sigma_{zz} - \sigma_{iso})\,. \quad \text{(S3)}$$

Table S1 Calculated (GIPAW) ^{1}H Chemical Shift Tensor Properties for the IND-NIC cocrystal.

IND	σ_{xx}	σ_{yy}	σ_{zz}	σ_{iso}	δ_{iso} [a]	σ_{aniso}	η
H4	22.34	24.03	25.99	24.12	6.78	2.80	0.90
H6	22.96	24.37	28.05	25.13	5.77	4.39	0.48
H7	26.57	24.49	19.82	23.62	7.28	–5.71	0.55
H9a	30.26	27.22	24.01	27.17	3.73	–4.73	0.97
H9b	31.85	28.44	23.24	27.84	3.06	–6.91	0.74
H11a	24.22	28.24	36.60	29.69	1.21	10.37	0.58
H11b	22.86	24.46	33.24	26.85	4.05	9.58	0.25
H11c	22.52	25.12	33.01	26.89	4.01	9.19	0.42
H12a	23.26	27.27	37.26	29.26	1.64	11.99	0.50
H12b	25.65	27.81	34.89	29.45	1.45	8.16	0.40
H12c	26.86	31.13	35.75	31.25	–0.35	6.75	0.95
H15	22.82	24.79	28.37	25.33	5.57	4.56	0.65
H16	26.78	24.46	19.75	23.66	7.24	–5.87	0.59
H18	22.20	24.70	28.13	25.01	5.89	4.68	0.80
H19	22.46	22.89	28.86	24.74	6.16	6.18	0.10
OH	1.83	4.21	31.10	12.38	18.52	28.09	0.13
NIC	σ_{xx}	σ_{yy}	σ_{zz}	σ_{iso}	δ_{iso} [a]	σ_{aniso}	η
H1	18.66	19.84	25.06	21.19	9.71	5.81	0.30
H3	24.53	23.87	21.65	23.35	7.55	–2.54	0.39
H4	20.66	22.22	24.48	22.45	8.45	3.04	0.77
H5	18.13	20.93	24.46	21.17	9.73	4.93	0.85
NH_2a	13.45	17.10	30.64	20.40	10.50	15.37	0.36
NH_2b	14.25	19.16	33.04	22.15	8.75	16.33	0.45

[a] $\delta_{iso} = -(\sigma_{iso} - \sigma_{ref})$, with $\sigma_{ref} = 30.9$ ppm

Table S2 Calculated (GIPAW) ^{13}C Chemical Shift Tensor Properties for the IND–NIC cocrystal.

IND	σ_{xx}	σ_{yy}	σ_{zz}	σ_{iso}	δ_{iso} [a]	σ_{aniso}	η
C1	106.0	41.5	–51.9	31.9	135.4	–125.6	0.77
C2	–20.4	41.4	136.0	52.3	115.0	125.5	0.74
C3	–30.6	–7.2	147.7	36.6	130.7	166.6	0.21
C4	–9.8	52.8	155.5	66.2	101.1	134.0	0.70
C5	–65.1	–7.4	100.8	9.4	157.9	137.0	0.63
C6	–23.8	40.0	173.2	63.1	104.2	165.1	0.58
C7	–27.0	22.8	170.3	55.4	111.9	172.4	0.43
C8	–40.9	26.7	128.6	38.1	129.2	135.7	0.75
C9	116.4	135.7	168.8	140.3	27.0	42.8	0.68
C10	65.3	–6.8	–95.7	–12.4	179.7	–124.9	0.87
C11	85.2	99.0	161.4	115.2	52.1	69.3	0.30
C12	149.5	157.2	169.4	158.7	8.6	16.1	0.72
C13	69.9	22.7	–93.0	–0.1	167.4	–139.3	0.51
C14	–53.2	4.1	150.9	33.9	133.4	175.5	0.49
C15	–64.5	14.6	164.1	38.1	129.2	189.1	0.63
C16	–60.7	13.9	155.3	36.2	131.1	178.7	0.63
C17	111.8	31.4	–74.6	22.9	144.4	–146.2	0.83
C18	–53.9	27.4	146.6	40.0	127.3	159.8	0.76
C19	–59.6	8.1	158.7	35.7	131.6	184.5	0.55
NIC	σ_{xx}	σ_{yy}	σ_{zz}	σ_{iso}	δ_{iso} [a]	σ_{aniso}	η
C1	–76.3	–9.2	145.8	20.1	147.2	188.5	0.53
C2	–64.0	10.5	153.1	33.2	134.1	179.9	0.62
C3	–82.0	–10.7	174.3	27.2	140.1	220.7	0.48
C4	–71.0	21.0	169.9	39.9	127.4	194.9	0.71
C5	–91.3	1.1	141.8	17.2	150.1	186.9	0.74
C6	–77.0	–3.0	81.8	0.6	166.7	121.8	0.91

[a] $\delta_{iso} = -[\sigma_{iso} - \sigma_{ref}]$, with $\sigma_{ref} = 167.3$ ppm.

Table S3 Relation between the atom labelling schemes used for the crystal structure determined from powder XRD data (PXRD) and for the solid-state ^{13}C and ^{1}H NMR data.

IND

^{13}C NMR	PXRD	^{1}H NMR	PXRD
C1	C11	H4	H8
C2	C10	H6	H5
C3	C9	H7	H3
C4	C7	H9	H23,24
C5	C6	H11	H19,20,21
C6	C4	H12	H13,14,15
C7	C2	H15	H41
C8	C1	H16	H39
C9	C22	H18	H35
C10	C25	H19	H33
C11	C18		
C12	C12		
C13	C29		
C14	C31		
C15	C40		
C16	C38		
C17	C36		
C18	C34		
C19	C32		

NIC

^{13}C NMR	PXRD	^{1}H NMR	PXRD
C1	C43	H1	H44
C2	C42	H3	H51
C3	C50	H4	H49
C4	C48	H5	H47
C5	C46	NH_2a	H56
C6	C52	NH_2b	H55