

Supplementary information

X-ray Raman Scattering: A New *In Situ* Probe of Molecular Structure during Nucleation and Crystallization from Liquid Solutions

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Figures S1-S5 show the individual N K-edge spectra of imidazole solid, aqueous solution at 35 °C, aqueous solution at 20 °C, aqueous solution at 13 °C and aqueous solution after crystallization. The individual spectra do not show a sign of time-dependent changes that could be attributed to radiation induced damage. This is expected because spinning of the solid sample and the large stock volume (~ 250 ml) of the aqueous solution accompanied with constant circulation would minimize radiation damage. The same is noticed for the C K-edge individual spectra illustrated in Figures S6-S9

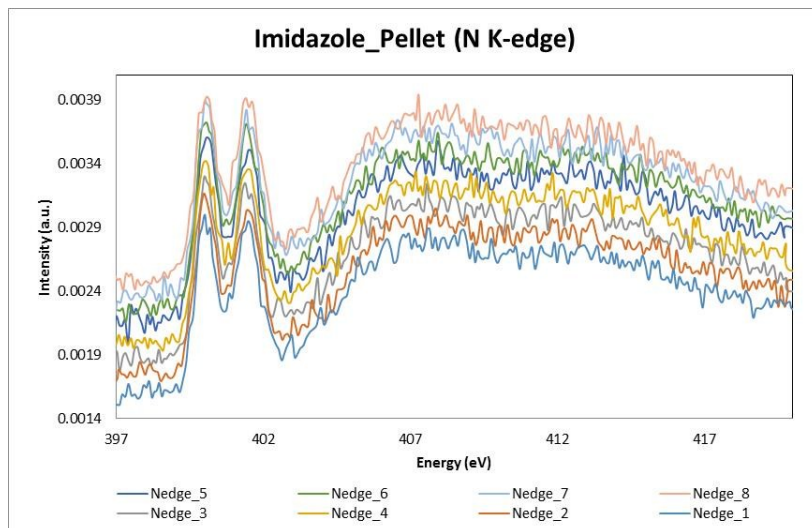


Figure S1: Imidazole solid individual N K-edge spectra

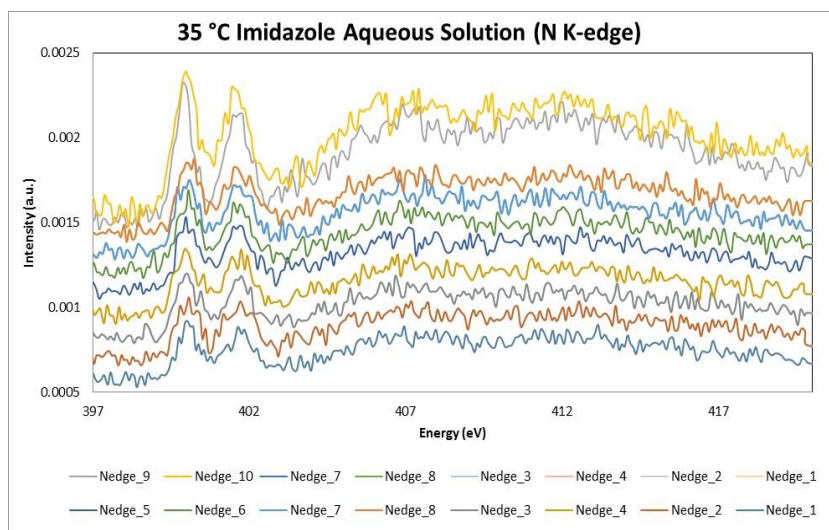


Figure S2: Imidazole aqueous solution at 35°C individual N K-edge spectra

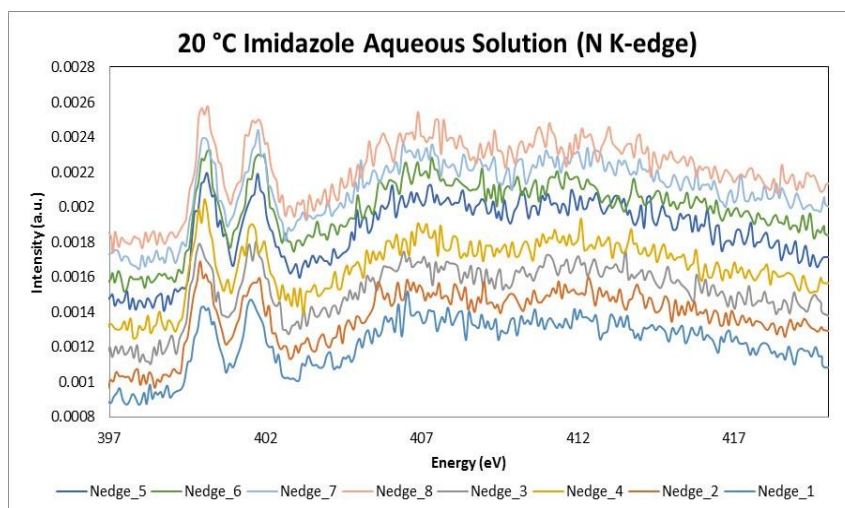


Figure S3: Imidazole aqueous solution at 20°C individual N K-edge spectra

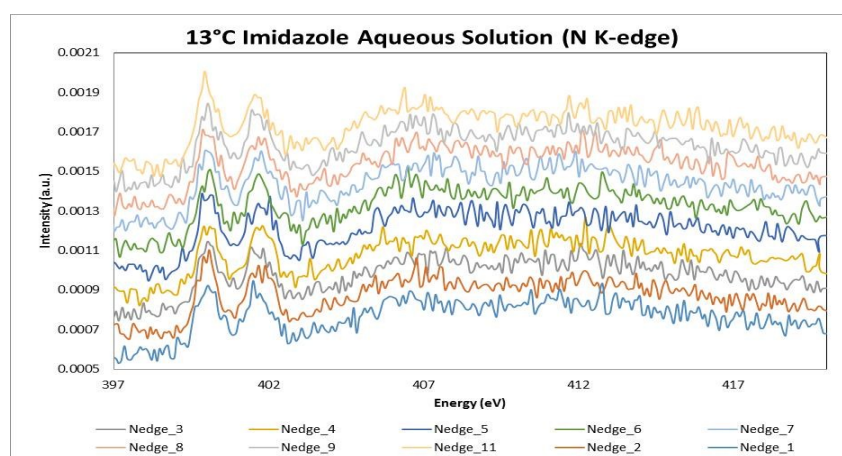


Figure S4: Imidazole aqueous solution at 13°C individual N K-edge spectra

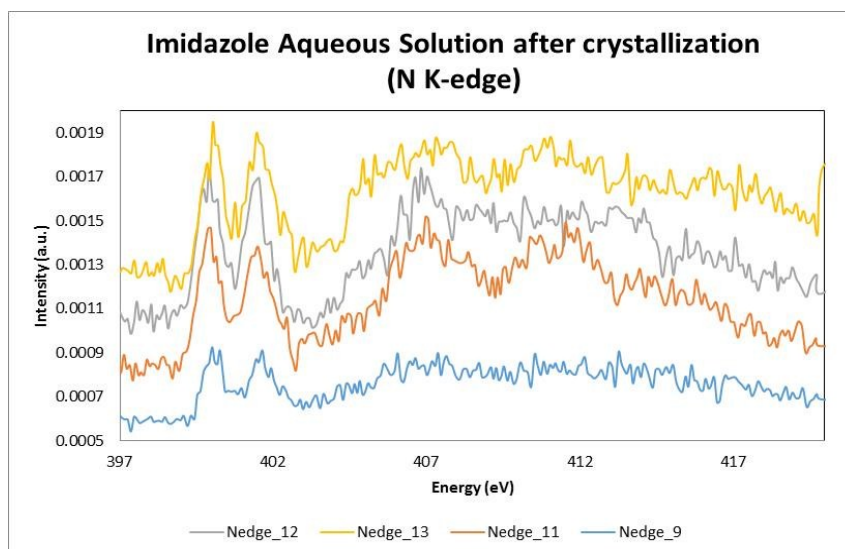


Figure S5: Imidazole aqueous solution after crystallization individual N K-edge spectra

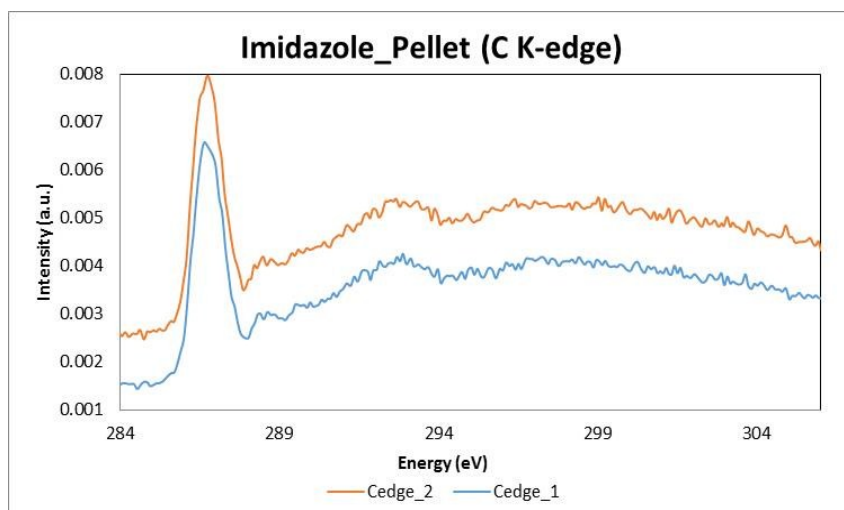


Figure S6: Imidazole solid individual C K-edge spectra

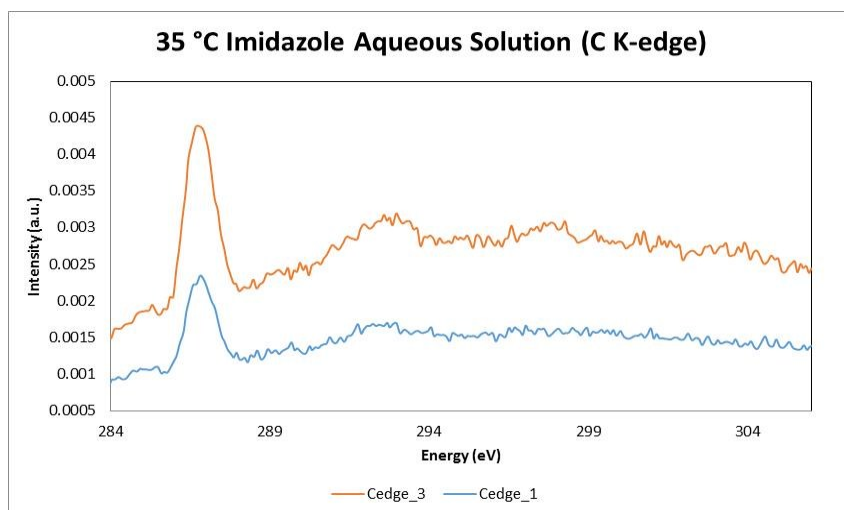


Figure S7: Imidazole aqueous solution at 35°C individual C K-edge spectra

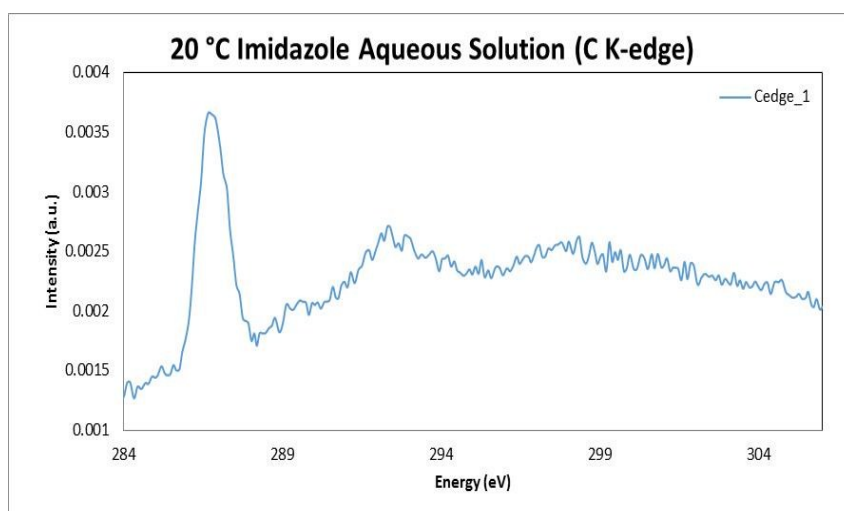


Figure S8: Imidazole aqueous solution at 20°C individual C K-edge spectra

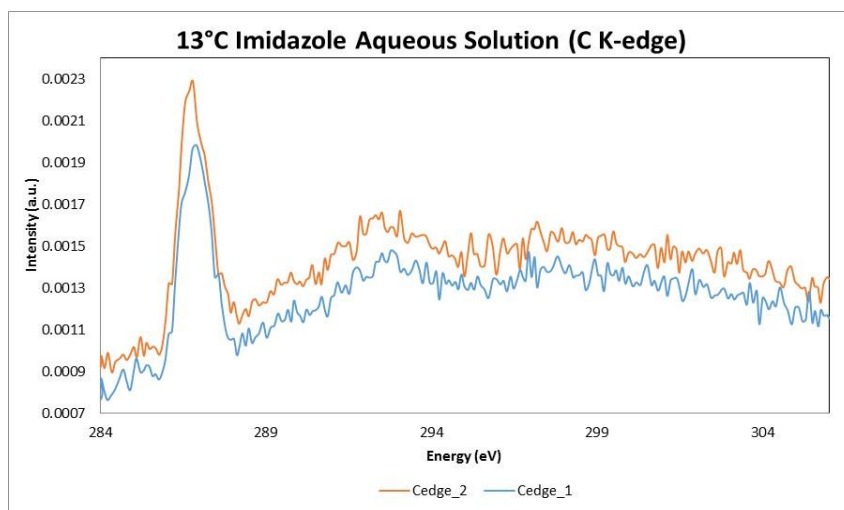
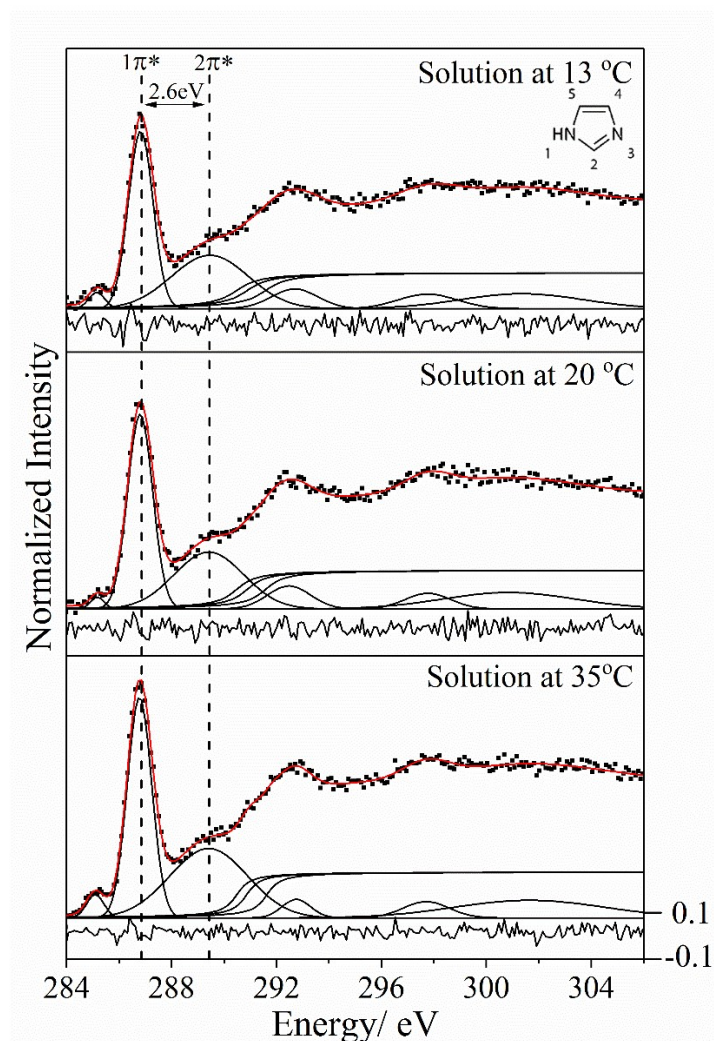


Figure S9: Imidazole aqueous solution at 13°C individual C K-edge spectra

Alternative fit for the imidazole aqueous solution C K-edge is illustrated in Figure S10 that is of comparable quality to the fit presented in the paper. The goodness of the fit for the fit below is comparable to the one reported in the main paper.



S10: Alternative fit for the imidazole aqueous solution C K-edge at 35°C, 20°C and 13°C. Note that the peak at ~ 285 eV is from the Kapton window

Table S1. Peak positions and assignments for the alternative fit to the imidazole aqueous solution C K-edge at 35°C, 20°C and 13°C

	Solution		
	35 °C /eV	20 °C /eV	13 °C /eV
E (From Kapton)	285.095	285.174	285.149
E (C 1s–1n*)	286.782	286.803	286.825
E (C 1s–2n*)	289.418	289.425	289.466
ΔE (C2 2n* -C 1n*)	2.636	2.622	2.641
IP (C2)	291.631	291.580	291.580
IP (C5)	290.860	290.880	290.880
IP (C4)	290.480	290.480	290.480
ΔIP (C2-C5)	0.771	0.7	0.7
ΔIP (C5-C4)	0.380	0.4	0.4
σ (C-H)	292.759	292.483	292.697
σ (C-C)	297.709	297.772	297.769
σ (C-N)	301.641	300.867	301.352

Table S2. Full width half maximum (FWHM) values of the π^* peaks in the N K-edge spectra

	Solid /eV	Solution at 35°C /eV	Solution at 20°C /eV	Solution at 13°C /eV	Solution after crystallization /eV
N3(1s-1n*)	0.857	0.891	0.856	0.876	0.833
N1 (1s-1 n*)	1.161	1.178	1.112	1.147	1.182