Supplementary material for

Hydrogen Bond-Induced Crystallization in Low-Molar-Ratio Urea–Formaldehyde Resins during Synthesis

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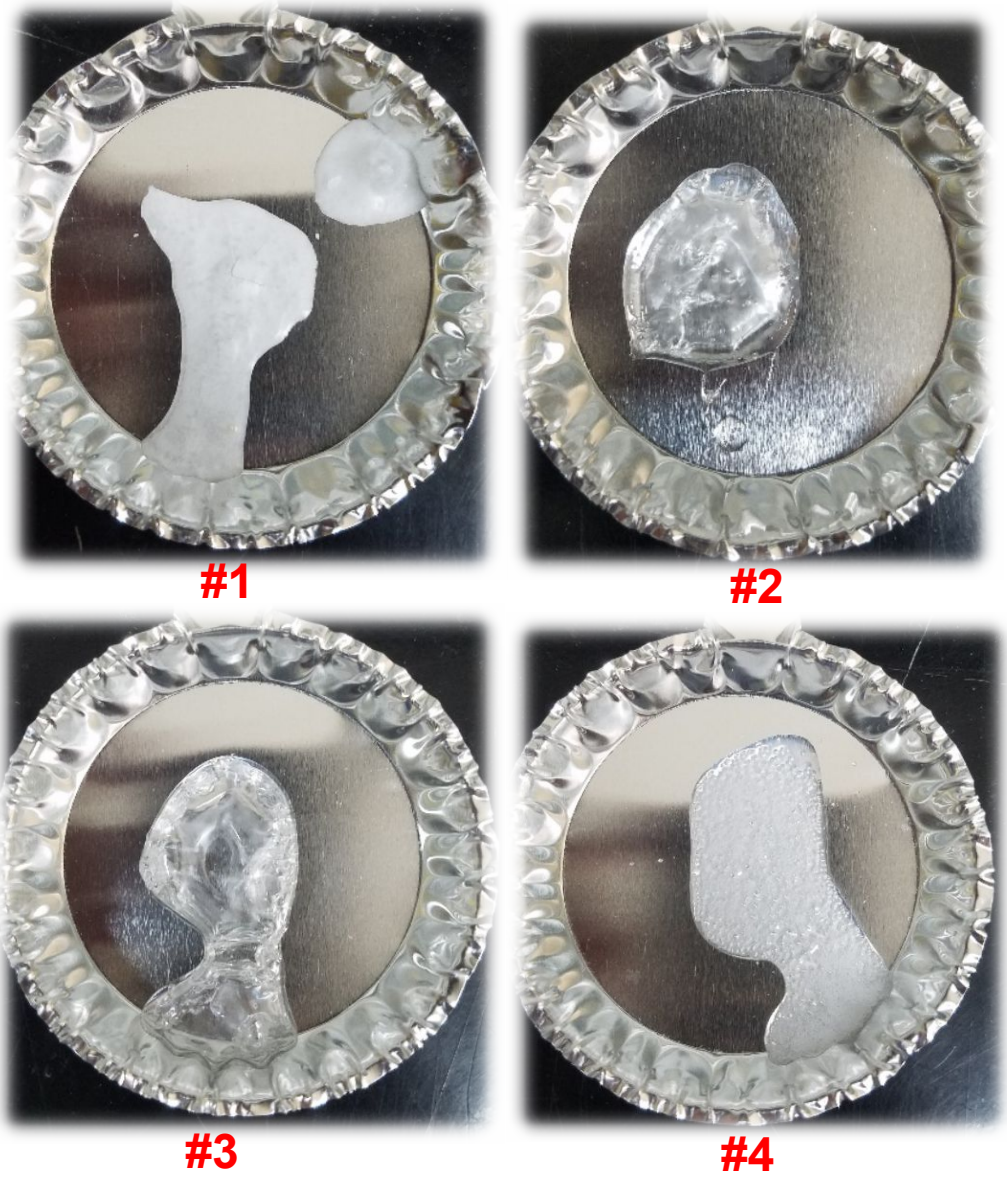


Figure S1. The physical appearance of oven-dried resins for different stages at 105 °C for 3 h.

The crystallinity percentage of each sample was calculated by the following equation:

$$\textbf{Crystallinity percentage (\%)} = 100 \times \textbf{Sc/St.}$$

where Sc represents the area of the crystalline domain, and St is the area of the total domain (crystalline + amorphous), respectively. The deconvolution of XRD patterns is shown in **Figure S2** and the raw data for the Gaussian deconvolution method are listed in **Table S1–2**.

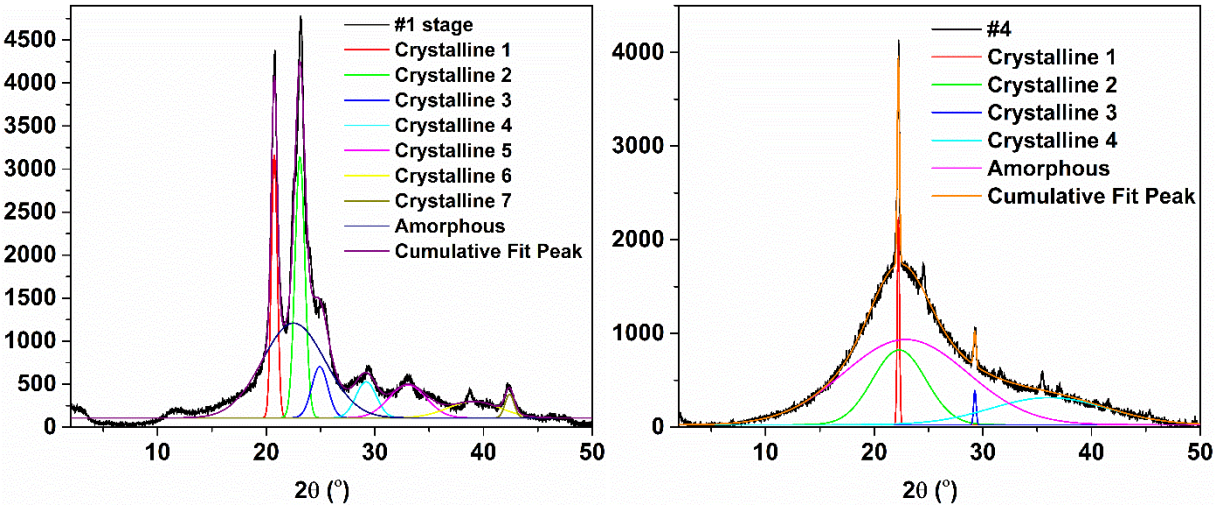


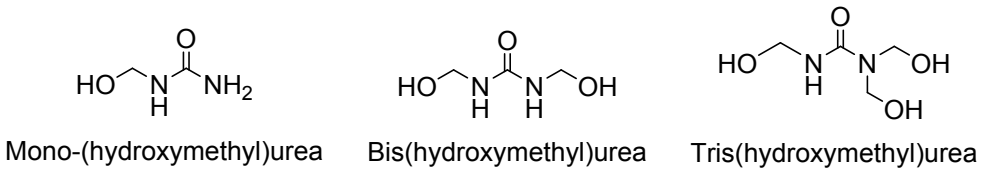
Figure S2. The deconvolution of XRD patterns using the Gaussian method for resins samples at different synthesis stages: (a) addition, and (d) final.

Table S1. Gaussian curve-fitting results for resins at the **addition stage (#1)**

Model	Gaussian		
Equation	y = y0 + A/(w*sqrt(PI/(4*ln(2)))) * exp(-4*ln(2)*(x-xc)^2/w^2)		
Reduced Chi-Sqr	8239.03202		
Adj. R-Square	0.98502		
		Value	Standard Error
Crystalline 1	y0	106.95688	3.10861
Crystalline 1	xc	20.74323	0.00239
Crystalline 1	**A (Area)**	**2105.50481**	23.4072
Crystalline 1	W (FWHM)	0.64697	0.00653
Crystalline 2	y0	106.95688	3.10861
Crystalline 2	xc	23.10244	0.00442
Crystalline 2	**A (Area)**	**3451.00489**	47.98355
Crystalline 2	W (FWHM)	1.06784	0.01113
Crystalline 3	y0	106.95688	3.10861
Crystalline 3	xc	24.92954	0.0284
Crystalline 3	**A (Area)**	**1112.56338**	103.52915
Crystalline 3	W (FWHM)	1.75748	0.10707
Crystalline 4	y0	106.95688	3.10861
Crystalline 4	xc	29.19907	0.05139
Crystalline 4	**A (Area)**	**991.96244**	89.55171
Crystalline 4	W (FWHM)	2.21823	0.14264
Crystalline 5	y0	106.95688	3.10861
Crystalline 5	xc	33.01686	0.08263
Crystalline 5	**A (Area)**	**1655.17189**	116.47046
Crystalline 5	W (FWHM)	4.0428	0.2843
Crystalline 6	y0	106.95688	3.10861
Crystalline 6	xc	39.12262	0.20577
Crystalline 6	**A (Area)**	**1149.74007**	116.39897
Crystalline 6	W (FWHM)	5.6535	0.59958
Crystalline 7	y0	106.95688	3.10861
Crystalline 7	xc	42.40184	0.03254
Crystalline 7	**A (Area)**	**287.08988**	32.45579
Crystalline 7	W (FWHM)	0.98098	0.09325
Amorphous	y0	106.95688	3.10861
Amorphous	xc	22.5238	0.06973
Amorphous	**A (Area)**	**7984.55961**	178.46777
Amorphous	W (FWHM)	6.80755	0.10946

Table S2. Gaussian curve-fitting results for resins at the **final stage (#4)**

Model	Gaussian		
Equation	y = y0 + A/(w*sqrt(PI/(4*ln(2)))) * exp(-4*ln(2)*(x-xc)^2/w^2)		
Reduced Chi-Sqr	1800.7665		
Adj. R-Square	0.99387		
		Value	Standard Error
Crystalline 1	y0	24.67848	2.61994
Crystalline 1	xc	22.22231	9.14E-04
Crystalline 1	**A (Area)**	**563.14734**	**4.95143**
Crystalline 1	W (FWHM)	0.23831	0.00224
Crystalline 2	y0	24.67848	2.61994
Crystalline 2	xc	22.29868	0.02229
Crystalline 2	**A (Area)**	**4917.00147**	**221.75515**
Crystalline 2	W (FWHM)	5.78688	0.09715
Crystalline 3	y0	24.67848	2.61994
Crystalline 3	xc	29.26586	0.00574
Crystalline 3	**A (Area)**	**105.18755**	**5.0518**
Crystalline 3	W (FWHM)	0.265	0.01391
Crystalline 4	y0	24.67848	2.61994
Crystalline 4	xc	36.10706	0.25697
Crystalline 4	**A (Area)**	**3898.15844**	**186.82948**
Crystalline 4	W (FWHM)	12.60141	0.35691
Amorphous	y0	24.67848	2.61994
Amorphous	xc	22.90268	0.10397
Amorphous	**A (Area)**	**13071.93134**	**235.42469**
Amorphous	W (FWHM)	13.48242	0.25121



Mono-(hydroxymethyl)urea Bis(hydroxymethyl)urea Tris(hydroxymethyl)urea

Figure S3. Structures of model compounds used in comparison to XRD analysis.