A theoretical study on the mechanism of citric acid-driven multi-component nucleation of sulfuric acid-base-water clusters

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Table S1. The number and length of hydrogen bonds in Å in all clusters.

cluster	Number of water molecules				
	0	1	2	3	4
SA·AM·W_n (n = 0 - 4)	1 (**1.539**)	3 (**1.563**,1.719,2.067)	5 (1.725,1.732,1.794,1.799,1.933)	6 (1.708,1.720,1.723,1.794,1.903,1.998)	7 (1.633,1.758,1.796,1.831,1.854,1.961,1.980)
SA·AM·CA·W_n (n = 0 - 4)	4 (**1.450,1.457**,1.913,2.146)	6 (1.659,1.703,1.757,1.789,1.858,1.935)	8 (**1.584**,1.883,1.889,1.917,2.044,2.064,2.073,2.121)	11 (1.686,1.695,1.714,1.739,1.897,1.917,1.926,1.945,1.959,1.961,1.969)	11 (**1.582**,1.666,1.783,1.834,1.857,1.884,1.920,1.929,1.947,1.958,2.002)
SA·DMA·W_n (n = 0 - 4)	1 (**1.392**)	3 (**1.569**,1.803,1.868)	4 (1.710,1.735,1.791,2.021)	6 (1.738,1.785,1.809,1.925,1.939,2.003)	8 (1.694,1.700,1.724,1.748,1.870,1.904,1.928,1.943)
SA·DMA·CA·W_n (n = 0 - 4)	5 (1.643,1.770,2.030,2.041,2.152)	5 (**1.466**,1.676,1.774,1.838,1.983)	7 (1.548,1.770,1.778,1.965,2.003,2.088,2.088)	8 (1.629,1.698,1.738,1.747,1.844,1.849,1.974,2.110)	9 (**1.502**,1.654,1.780,1.853,1.862,1.889,1.929,1.968,1.973)
SA·AM·DMA·W_n (n = 0 - 4)	2 (**1.586**,1.792)	4 (**1.531**,1.684,1.812,1.878)	5 (1.712,1.813,2.024,2.077,2.157)	7 (1.688,1.689,1.698,1.704,1.889,1.966,1.985)	8 (1.659,1.718,1.767,1.849,1.863,1.916,1.938,1.989)
SA·AM·DMA·CA·	4 (**1.45,1.567**,	6 (**1.581**,1.634,1	9 (**1.516**,1.647,1.72	10 (**1.593**,1.670,1.670,1.7	8 (**1.587**,1.602,1.735,1.768,1

W_n (n = 0 - 4)	1.703 , 1.791)	.780,1.785,1.864,1.957)	6,1.791,1.817,1.829,1.854,1.972)	95,1.800,1.905,1.906,1.979,2.077,2.098)	.805,1.857,1.899,1.963)

Table S2. The number of proton transfers in all clusters.

cluster	Number of water molecules				
	0	1	2	3	4
SA·AM·W_n (n = 0 - 4)	0	0	1	1	1
SA·AM·CA·W_n (n = 0 - 4)	1	1	1	1	1
SA·DMA·W_n (n = 0 - 4)	1	1	1	1	1
SA·DMA·CA·W_n (n = 0 - 4)	1	1	1	1	1
SA·AM·DMA·W_n (n = 0 - 4)	1	1	1	1	1
SA·AM·DMA·CA·W_n (n = 0 - 4)	1	1	2	1	1

Table S3. The contribution of CA in the formation of hydrogen bonds in all clusters.

cluster		Donor	Acceptor	Hydrogen bond	Hydrogen bond length/Å
SA·AM·CA·W_n (n = 0 - 4)	n=0	**β-COOH**	SA	C-O-H···O=S	1.457
		SA	**β-COOH**	S=O-H···O=C	1.913
		AM	**α_1-COOH**	N-H···O=C	2.146
	n=1	**α_1-COOH**	SA	C-O-H···O=S	1.757
		AM	**α_1-COOH**	N-H···O=C	1.858
		β-COOH	W	C-O-H···O-H	1.659
		AM	**β-COOH**	N-H···O=C	1.935
	n=2	**α_1-COOH**	W	C-O-H···O-H	1.584
		AM	**α_1-COOH**	N-H···O=C	2.073
		AM	**β-COOH**	N-H···O=C	2.044
	n=3	**β-OH**	**α_1-COOH**	C-O-H···O=C	1.897
		SA	**β-OH**	S=O-H···O-C	1.959
		β-COOH	SA	C-O-H···O=S	1.686
		AM	**β-COOH**	N-H···O=C	1.917
	n=4	**β-COOH**	SA	C-O-H···O=S	1.582

		W	**β-COOH**	O-H···O=C	2.002
		W	**α_1-COOH**	O-H···O=C	1.929
SA·DMA·CA·W_n (n = 0 - 4)	n=0	**α_1-COOH**	SA	C-O-H···O=S	1.643
		DMA	**α_1-COOH**	N-H···O=C	2.152
		β-COOH	SA	C-O-H···O=S	2.030
		DMA	**β-COOH**	N-H···O=C	2.041
	n=1	**α_1-COOH**	SA	C-O-H···O=S	1.466
		DMA	**α_1-COOH**	N-H···O=C	1.774
		W	**β-OH**	O-H···O-C	1.983
	n=2	**α_1-COOH**	W	C-O-H···O-H	1.778
		W	**α_1-COOH**	O-H···O=C	2.003
		W	**α_2-COOH**	O-H···O=C	1.965
		β-COOH	SA	C-O-H···O=S	1.548
		DMA	**β-COOH**	N-H···O=C	2.088
	n=3	**α_1-COOH**	W	C-O-H···O-H	1.698
		DMA	**α_1-COOH**	N-H···O=C	2.110
		β-COOH	W	C-O-H···O-H	1.629
		W	**β-COOH**	O-H···O=C	1.849
	n=4	**α_1-COOH**	SA	C-O-H···O=S	1.502
		SA	**α_1-COOH**	S=O-H···O=C	1.862
		W	**β-OH**	O-H···O-C	1.889
SA·AM·DMA·CA·W_n (n = 0 - 4)	n=0	**α_1-COOH**	SA	C-O-H···O=S	1.454
		DMA	**α_1-COOH**	N-H···O=C	1.791
	n=1	**α_1-COOH**	SA	C-O-H···O=S	1.581
		DMA	**α_1-COOH**	N-H···O-C	1.957
		α_2-COOH	SA	C-O-H···O=S	1.780
		β-OH	SA	C-O-H···O=S	1.785
	n=2	DMA	**α_1-COOH**	N-H···O-C	1.516
		β-OH	**α_1-COOH**	C-O-H···O-C	1.647
		AM	**β-OH**	N-H···O-C	1.829
		α_2-COOH	SA	C-O-H···O=S	1.726
		W	**α_1-COOH**	O-H···O=C	1.972

	n=3	**α_1-COOH**	AM	C-O-H···N-H	1.670
		W	**α_1-COOH**	O-H···O=C	1.905
		α_2-COOH	W	C-O-H···O-H	1.800
		W	**α_2-COOH**	O-H···O=C	1.979
		β-COOH	SA	C-O-H···O=S	1.593
		SA	**β-COOH**	S=O-H···O=C	1.670
	n=4	**α_1-COOH**	W	C-O-H···O-H	1.587
		W	**α_2-COOH**	O-H···O=C	1.963

Table S4 The reaction Gibbs free energies (Kcal mol^{-1}) of $SA \cdot AM \cdot W_n \cdot CA_m$ (n = 0 - 4, m=0-1), $SA \cdot DMA \cdot W_n \cdot CA_m$ (n = 0 - 4, m=0-1) and $SA \cdot AM \cdot DMA \cdot W_n \cdot CA_m$ (n = 0 - 4, m=0-1) clusters through various formation manners using M06-2X/6-311++G (2d, 2p) at 298.15K and 101.3KPa.

Reactions/n/$\triangle G$		
$SA \cdot AM \cdot W_{n-1}+W \rightarrow SA \cdot AM \cdot W_n$	$SA+AM+nW \rightarrow SA \cdot AM \cdot (W)_n$	
	n =0, $\triangle G$ =-7.98	
n =1, $\triangle G$ =-2.6	n =1, $\triangle G$ =-10.61	
n =2, $\triangle G$ =-2.2	n =2, $\triangle G$ =-12.80	
n =3, $\triangle G$ =-1.9	n =3, $\triangle G$ =-14.70	
n =4, $\triangle G$ =-3.3	n =4, $\triangle G$ =-18.02	
$SA \cdot AM \cdot CA \cdot W_{n-1}+W \rightarrow SA \cdot AM \cdot CA \cdot W_n$	$SA \cdot AM \cdot W_n+CA \rightarrow SA \cdot AM \cdot CA \cdot W_n$	$SA+AM+CA+nW \rightarrow SA \cdot AM \cdot CA \cdot W_n$
	n =0, $\triangle G$ =-8.45	n =0, $\triangle G$ =-16.42
n =1, $\triangle G$ =-2.7	n =1, $\triangle G$ =-8.52	n =1, $\triangle G$ =-19.13
n =2, $\triangle G$ =-1.4	n =2, $\triangle G$ =-7.74	n =2, $\triangle G$ =-20.54
n =3, $\triangle G$ =-1.6	n =3, $\triangle G$ =-7.47	n =3, $\triangle G$ =-22.17
n =4, $\triangle G$ =-1.1	n =4, $\triangle G$ =-5.26	n =4, $\triangle G$ =-23.29
$SA \cdot DMA \cdot W_{n-1}+W \rightarrow SA \cdot DMA \cdot W_n$	$SA+DMA+nW \rightarrow SA \cdot DMA \cdot (W)_n$	
	n =0, $\triangle G$ =-11.22	
n =1, $\triangle G$ =-4.9	n =1, $\triangle G$ =-16.07	
n =2, $\triangle G$ =-3.3	n =2, $\triangle G$ =-19.34	
n =3, $\triangle G$ =-2.0	n =3, $\triangle G$ =-21.34	

n =4, $\triangle G$ =-1.7	n =4, $\triangle G$ =-23.02	
SA·DMA·CA·W_{n-1}+W→SA·DMA·CA·W_n	SA·DMA·W_n+CA→SA·DMA·CA·W_n	SA+ DMA +CA+nW→SA·DMA·CA·$(W)_n$
	n =0, $\triangle G$ =-13.45	n =0, $\triangle G$ =-24.67
n =1, $\triangle G$ =**0.83**	n =1, $\triangle G$ =-7.77	n =1, $\triangle G$ =-23.84
n =2, $\triangle G$ =-1.7	n =2, $\triangle G$ =-6.18	n =2, $\triangle G$ =-25.52
n =3, $\triangle G$ =-3.4	n =3, $\triangle G$ =-7.55	n =3, $\triangle G$ =-28.89
n =4, $\triangle G$ =**0.26**	n =4, $\triangle G$ =-5.61	n =4, $\triangle G$ =-28.63
SA·AM·DMA·W_{n-1}+W→SA·AM·DMA·W_n	SA+ AM + DMA +nW→SA·AM·DMA·$(W)_n$	
	n =0, $\triangle G$ =-15.49	
n =1, $\triangle G$ =-3.4	n =1, $\triangle G$ =-18.86	
n =2, $\triangle G$ =-2.0	n =2, $\triangle G$ =-20.83	
n =3, $\triangle G$ =-3.2	n =3, $\triangle G$ =-23.98	
n =4, $\triangle G$ =-3.0	n =4, $\triangle G$ =-26.98	
SA·AM·DMA·CA·W_{n-1}+W→SA·AM·DMA·CA·W_n	SA·AM·DMA·W_n+CA→SA·AM·DMA·CA·W_n	SA+ AM + DMA +CA+nW→SA·AM·DMA·CA·$(W)_n$
	n =0, $\triangle G$ =-10.24	n =0, $\triangle G$ =-25.73
n =1, $\triangle G$ =**1.81**	n =1, $\triangle G$ =-5.06	n =1, $\triangle G$ =-23.91
n =2, $\triangle G$ =-4.4	n =2, $\triangle G$ =-7.49	n =2, $\triangle G$ =-28.32
n =3, $\triangle G$ =**1.61**	n =3, $\triangle G$ =-2.74	n =3, $\triangle G$ =-26.72
n =4, $\triangle G$ =-2.6	n =4, $\triangle G$ =-2.33	n =4, $\triangle G$ =-29.31