SUPPLEMENTAL MATERIAL for:

Optimizing the parameters of the Gibbs Ensemble Monte Carlo simulation of phase separation: The Role of multiple relaxation times

By Bina Kumari, Pradipta Bandyopadhyay and Subir K. Sarkar

Table 1S: Acceptance probabilities and ranges of data used for calculating a_0, τ_1, α and τ_2 for the parameter set $\lambda = 1.25$, $d_m = 0.0025$, $n_s = 150$ and $n_v = 1$.

Temperature	Run	Low density phase		High density phase		Acceptance probability		
		Fitting range for τ_2 and a_0 (MCC	Fitting range for τ_1 and α (MCC)	Fitting range for τ_2 and a_0 (MCC)	Fitting range for τ_1 and α (MCC)	Particle move-ment	Volume change	Particle transfer
0.66	1	10000-90000	0-5500	10000-70000	0-5500	0.413	0.407	0.121×10^{-2}
	2	10000-75000	0-5500	10000-70000	0-5500	0.413	0.407	0.120×10^{-2}
0.68	1	10000-1.1×10^5	0-4500	10000-90000	0-4800	0.444	0.431	0.221×10^{-2}
	2	10000-1.1×10^5	0-4500	10000-85000	0-4800	0.444	0.431	0.221×10^{-2}
0.70	1	10000-1.5×10^5	0-4200	10000-1.3×10^5	0-4500	0.478	0.460	0.394×10^{-2}
	2	10000-1.5×10^5	0-4200	10000-1.3×10^5	0-4500	0.479	0.460	0.392×10^{-2}
0.72	1	10000-2.5×10^5	0-4000	10000-2.0×10^5	0-4000	0.518	0.498	0.741×10^{-2}
	2	10000-2.5×10^5	0-4000	10000-2.0×10^5	0-4000	0.518	0.497	0.722×10^{-2}

Table 2S: Acceptance probabilities and ranges of data used for calculating a_0, τ_1, α and τ_2 for the parameter set $\lambda = 1.25$, $d_m = 0.005$, $n_s = 150$ and $n_v = 1$.

Temperature	Run	Low density phase		High density phase		Acceptance probability		
		Fitting range for τ_2 and a_0 (MCC)	Fitting range for τ_1 and α (MC)	Fitting range for τ_2 and a_0 (MCC)	Fitting range for τ_1 and α (MCC)	Particle move-ment	Volume change	Particle transfer
0.66	1	10000-38000	0-5500	10000-38000	0-5800	0.413	0.231	0.119×10^{-2}
	2	10000-58000	0-5500	10000-45000	0-5800	0.413	0.231	0.119×10^{-2}
0.68	1	10000-60000	0-4500	10000-50000	0-4800	0.444	0.249	0.218×10^{-2}
	2	10000-70000	0-4500	10000-43000	0-4800	0.444	0.249	0.221×10^{-2}
0.70	1	10000-1.0×10^5	0-4000	10000-90000	0-4200	0.478	0.273	0.394×10^{-2}
	2	10000-1.0×10^5	0-4000	10000-87000	0-4200	0.478	0.273	0.391×10^{-2}
0.72	1	10000-1.0×10^5	0-4000	10000-1.0×10^5	0-4200	0.519	0.305	0.709×10^{-2}
	2	10000-1.5×10^5	0-4000	10000-1.0×10^5	0-4200	0.519	0.305	0.709×10^{-2}

Table 3S: Acceptance probabilities and ranges of data used for calculating a_0 , τ_1 , α and τ_2 for the parameter set $\lambda = 1.25$, $d_m = 0.0025$, $n_s = 150$ and $n_v = 2$.

Temperature	Run	Low density phase		High density phase		Acceptance probability		
		Fitting range for τ_2 and a_0 (MCC)	Fitting range for τ_1 and α (MCC)	Fitting range for τ_2 and a_0 (MCC)	Fitting range for τ_1 and α (MCC)	Particle move-ment	Volume change	Particle transfer
0.66	1	10000-60000	0-5500	10000-60000	0-5500	0.413	0.407	0.120×10^{-2}
	2	10000-58000	0-5500	10000-56000	0-5800	0.413	0.407	0.118×10^{-2}
0.68	1	10000-68000	0-4500	10000-75000	0-4800	0.444	0.431	0.219×10^{-2}
	2	10000-85000	0-4500	10000-85000	0-4800	0.444	0.431	0.222×10^{-2}
0.70	1	10000-1.0×10^5	0-4000	10000-1.0×10^5	0-4000	0.478	0.461	0.402×10^{-2}
	2	10000-1.0×10^5	0-4000	10000-1.0×10^5	0-4200	0.478	0.460	0.391×10^{-2}
0.72	1	10000-1.3×10^5	0-4000	10000-1.4×10^5	0-4000	0.518	0.497	0.707×10^{-2}
	2	10000-1.4×10^5	0-4000	10000-1.5×10^5	0-4000	0.518	0.497	0.713×10^{-2}

Table 4S: Acceptance probabilities and ranges of data used for calculating a_0 , τ_1 , α and τ_2 for the parameter set $\lambda = 1.25$, $d_m = 0.0025$, $n_s = 1000$ and $n_v = 1$.

Temperature	Run	Low density phase		High density phase		Acceptance probability		
		Fitting range for τ_2 and a_0 (MCC)	Fitting range for τ_1 and α (MCC)	Fitting range for τ_2 and a_0 (MCC)	Fitting range for τ_1 and α (MCC)	Particle move-ment	Volume change	Particle transfer
0.66	1	5000-1.0×10^5	0-2000	5000-75000	0-2000	0.413	0.407	0.121×10^{-2}
	2	5000-1.0×10^5	0-2000	5000-75000	0-2000	0.413	0.407	0.12×10^{-2}
0.68	1	5000-1.5×10^5	0-1500	5000-85000	0-1500	0.444	0.431	0.219×10^{-2}
	2	5000-1.5×10^5	0-1500	5000-85000	0-1500	0.444	0.431	0.217×10^{-2}
0.70	1	5000-1.8×10^5	0-1200	5000-1.5×10^5	0-1300	0.478	0.460	0.381×10^{-2}
	2	5000-1.5×10^5	0-1200	5000-1.8×10^5	0-1300	0.478	0.460	0.393×10^{-2}
0.72	1	5000-2.0×10^5	0-1000	5000-2.0×10^5	0-1200	0.518	0.497	0.728×10^{-2}
	2	5000-2.2×10^5	0-1000	5000-2.4×10^5	0-1200	0.518	0.497	0.712×10^{-2}

Table 5S: Acceptance probabilities and ranges of data used for calculating a_0 , τ_1 , α and τ_2 for the parameter set $\lambda = 1.5$, $d_m = 0.005$, $n_s = 150$ and $n_v = 1$.

Temperature	Run	Low density phase		High density phase		Acceptance probability		
		Fitting range for τ_2 and a_0 (MCC)	Fitting range for τ_1 and α (MCC)	Fitting range for τ_2 and a_0 (MCC)	Fitting range for τ_1 and α (MCC)	Particle move-ment	Volume change	Particle transfer
1.0	1	1500-25000	0-650	1500-30000	0-650	0.409	0.409	0.414×10^{-2}
	2	1500-30000	0-650	1500-20000	0-700	0.409	0.409	0.414×10^{-2}
1.05	1	1500-40000	0-550	1500-40000	0-600	0.443	0.443	0.764×10^{-2}
	2	1500-40000	0-550	1500-40000	0-600	0.443	0.443	0.762×10^{-2}
1.08	1	1500-50000	0-520	1500-50000	0-550	0.466	0.467	0.110×10^{-1}
	2	1500-35000	0-520	1500-50000	0-550	0.466	0.467	0.110×10^{-1}
1.10	1	1500-60000	0-520	1500-60000	0-550	0.481	0.484	0.139×10^{-1}
	2	1500-60000	0-520	1500-60000	0-550	0.481	0.484	0.139×10^{-1}
1.12	1	1500-65000	0-520	1500-65000	0-550	0.498	0.502	0.175×10^{-1}
	2	1500-65000	0-520	1500-60000	0-550	0.498	0.502	0.175×10^{-1}

Table 6S: Acceptance probabilities and ranges of data used for calculating a_0 , τ_1 , α and τ_2 for the parameter set $\lambda = 1.5$, $d_m = 0.01$, $n_s = 150$ and $n_v = 1$.

Temperature	Run	Low density phase		High density phase		Acceptance probability		
		Fitting range for τ_2 and a_0 (MCC)	Fitting range for τ_1 and α (MCC)	Fitting range for τ_2 and a_0 (MCC)	Fitting range for τ_1 and α (MCC)	Particle move-ment	Volume change	Particle transfer
1.0	1	1500-15000	0-650	1500-15000	0-700	0.409	0.235	0.411×10^{-2}
	2	1500-15000	0-650	1500-15000	0-700	0.409	0.235	0.414×10^{-2}
1.05	1	1500-20000	0-550	1500-20000	0-600	0.443	0.264	0.761×10^{-2}
	2	1500-20000	0-550	1500-20000	0-600	0.443	0.264	0.762×10^{-2}
1.08	1	1500-27000	0-520	1500-27000	0-550	0.466	0.284	0.109×10^{-1}
	2	1500-27000	0-520	1500-27000	0-550	0.466	0.284	0.109×10^{-1}
1.10	1	1500-32000	0-520	1500-32000	0-550	0.481	0.299	0.139×10^{-1}
	2	1500-32000	0-520	1500-32000	0-550	0.481	0.299	0.139×10^{-1}
1.12	1	1500-40000	0-520	1500-35000	0-550	0.498	0.316	0.176×10^{-1}
	2	1500-40000	0-520	1500-35000	0-550	0.498	0.316	0.175×10^{-1}

Table 7S: Acceptance probabilities and ranges of data used for calculating a_0 , τ_1 , α and τ_2 for the parameter set $\lambda = 1.5$, $d_m = 0.005$, $n_s = 150$ and $n_v = 2$.

Temperature	Run	Low density phase		High density phase		Acceptance probability		
		Fitting range for τ_2 and a_0 (MCC)	Fitting range for τ_1 and α (MCC)	Fitting range for τ_2 and a_0 (MCC)	Fitting range for τ_1 and α (MCC)	Particle move-ment	Volume change	Particle transfer
1.0	1	1500-15000	0-650	1500-15000	0-700	0.409	0.409	0.411×10^{-2}
	2	1500-15000	0-650	1500-15000	0-700	0.409	0.409	0.414×10^{-2}
1.05	1	1500-20000	0-550	1500-18000	0-600	0.443	0.443	0.762×10^{-2}
	2	1500-22000	0-550	1500-20000	0-600	0.443	0.443	0.767×10^{-2}
1.08	1	1500-28000	0-520	1500-28000	0-550	0.466	0.467	0.110×10^{-1}
	2	1500-28000	0-520	1500-25000	0-550	0.466	0.467	0.109×10^{-1}
1.10	1	1500-19000	0-520	1500-35000	0-550	0.481	0.484	0.140×10^{-1}
	2	1500-30000	0-520	1500-35000	0-550	0.481	0.484	0.139×10^{-1}
1.12	1	1500-40000	0-520	1500-40000	0-550	0.498	0.502	0.176×10^{-1}
	2	1500-40000	0-520	1500-40000	0-550	0.498	0.502	0.176×10^{-1}

Table 8S: Acceptance probabilities and ranges of data used for calculating a_0 , τ_1 , α and τ_2 for the parameter set $\lambda = 1.5$, $d_m = 0.005$, $n_s = 1000$ and $n_v = 1$.

Temperature	Run	Low density phase		High density phase		Acceptance probability		
		Fitting range for τ_2 and a_0 (MCC)	Fitting range for τ_1 and α (MCC)	Fitting range for τ_2 and a_0 (MCC)	Fitting range for τ_1 and α (MCC)	Particle move-ment	Volume change	Particle transfer
1.0	1	400-22000	0-140	400-15000	0-150	0.409	0.409	0.412×10^{-2}
	2	400-20000	0-140	400-16000	0-150	0.409	0.409	0.412×10^{-2}
1.05	1	400-32000	0-120	400-25000	0-130	0.443	0.443	0.766×10^{-2}
	2	400-18000	0-120	400-22000	0-130	0.443	0.443	0.764×10^{-2}
1.08	1	400-35000	0-120	400-30000	0-130	0.466	0.467	0.109×10^{-1}
	2	400-25000	0-120	400-29000	0-130	0.466	0.467	0.110×10^{-1}
1.10	1	400-30000	0-110	400-32000	0-120	0.481	0.484	0.139×10^{-1}
	2	400-40000	0-110	400-40000	0-120	0.481	0.484	0.140×10^{-1}
1.12	1	400-55000	0-110	400-35000	0-120	0.498	0.502	0.175×10^{-1}
	2	400-50000	0-110	400-35000	0-120	0.498	0.501	0.174×10^{-1}

Table 9S: Acceptance probabilities and ranges of data used for calculating a_0 , τ_1 ,α and τ_2 for the parameter set $\lambda = 1.75$, $d_m = 0.0085$, $n_s = 150$ and $n_v = 1$.

Temperature	Run	Low density phase		High density phase		Acceptance probability		
		Fitting range for τ_2 and a_0 (MCC)	Fitting range for τ_1 and α (MCC)	Fitting range for τ_2 and a_0 (MCC)	Fitting range for τ_1 and α (MCC)	Particle move-ment	Volume change	Particle transfer
1.55	1 2	600-24000 600-24000	0-220 0-220	600-24000 600-24000	0-250 0-250	0.414 0.414	0.396 0.395	0.160×10^{-1} 0.160×10^{-1}
1.57	1 2	600-24000 600-24000	0-220 0-220	600-24000 600-24000	0-250 0-250	0.426 0.426	0.411 0.412	0.185×10^{-1} 0.186×10^{-1}
1.60	1 2	600-25000 600-21000	0-220 0-220	600-24000 600-25000	0-250 0-250	0.443 0.443	0.436 0.435	0.231×10^{-1} 0.228×10^{-1}
1.62	1 2	600-25000 600-25000	0-220 0-220	600-25000 600-25000	0-250 0-250	0.455 0.455	0.454 0.453	0.265×10^{-1} 0.264×10^{-1}

Table 10S: Acceptance probabilities and ranges of data used for calculating a_0 , τ_1 , α and τ_2 for the parameter set $\lambda = 1.75$, $d_m = 0.017$, $n_s = 150$ and $n_v = 1$.

Temperature	Run	Low density phase		High density phase		Acceptance probability		
		Fitting range for τ_2 and a_0 (MCC)	Fitting range for τ_1 and α (MCC)	Fitting range for τ_2 and a_0 (MCC)	Fitting range for τ_1 and α (MCC)	Particle move-ment	Volume change	Particle transfer
1.55	1 2	600-25000 600-25000	0-220 0-220	600-25000 600-25000	0-250 0-250	0.414 0.414	0.223 0.223	0.159×10^{-1} 0.159×10^{-1}
1.57	1 2	600-28000 600-28000	0-220 0-220	600-28000 600-28000	0-250 0-250	0.425 0.425	0.236 0.235	0.185×10^{-1} 0.184×10^{-1}
1.60	1 2	600-32000 600-32000	0-220 0-220	600-32000 600-32000	0-250 0-250	0.443 0.443	0.256 0.256	0.230×10^{-1} 0.229×10^{-1}
1.62	1 2	600-35000 600-35000	0-220 0-220	600-35000 600-35000	0-250 0-250	0.455 0.455	0.271 0.270	0.264×10^{-1} 0.263×10^{-1}

Table 11S: Acceptance probabilities and ranges of data used for calculating a_0 , τ_1 , α and τ_2 for the parameter set $\lambda = 1.75$, $d_m = 0.0085$, $n_s = 150$ and $n_v = 2$.

Temperature	Run	Low density phase		High density phase		Acceptance probability		
		Fitting range for τ_2 and a_0 (MCC)	Fitting range for τ_1 and α (MCC)	Fitting range for τ_2 and a_0 (MCC)	Fitting range for τ_1 and α (MCC)	Particle move-ment	Volume change	Particle transfer
1.55	1	600-16000	0-220	600-16000	0-250	0.414	0.396	0.160×10^{-1}
	2	600-16000	0-220	600-16000	0-250	0.414	0.395	0.159×10^{-1}
1.57	1	600-18000	0-220	600-18000	0-250	0.425	0.411	0.185×10^{-1}
	2	600-18000	0-220	600-18000	0-250	0.425	0.411	0.185×10^{-1}
1.60	1	600-20000	0-220	600-20000	0-250	0.443	0.435	0.228×10^{-1}
	2	600-20000	0-220	600-20000	0-250	0.443	0.436	0.229×10^{-1}
1.62	1	600-25000	0-220	600-25000	0-250	0.455	0.453	0.264×10^{-1}
	2	600-25000	0-220	600-25000	0-250	0.455	0.453	0.263×10^{-1}

Table 12S: Acceptance probabilities and ranges of data used for calculating a_0 , τ_1 , α and τ_2 for the parameter set $\lambda = 1.75$, $d_m = 0.0085$, $n_s = 1000$ and $n_v = 1$.

Temperature	Run	Low density phase		High density phase		Acceptance probability		
		Fitting range for τ_2 and a_0 (MCC)	Fitting range for τ_1 and α (MCC)	Fitting range for τ_2 and a_0 (MCC)	Fitting range for τ_1 and α (MCC)	Particle move-ment	Volume change	Particle transfer
1.55	1	400-22000	0-45	400-20000	0-45	0.414	0.396	0.160×10^{-1}
	2	400-20000	0-45	400-15000	0-45	0.414	0.396	0.160×10^{-1}
1.57	1	400-16000	0-45	400-16000	0-45	0.425	0.411	0.185×10^{-1}
	2	400-21000	0-45	400-18000	0-45	0.425	0.411	0.184×10^{-1}
1.60	1	400-25000	0-40	400-20000	0-45	0.443	0.436	0.230×10^{-1}
	2	400-23000	0-40	400-23000	0-45	0.443	0.435	0.228×10^{-1}
1.62	1	400-26000	0-40	400-22000	0-45	0.455	0.453	0.264×10^{-1}
	2	400-26000	0-40	400-26000	0-45	0.455	0.453	0.265×10^{-1}

Table 13S: Comparison of the vapour-liquid equilibrium number density computed by us and those reported in L. Vega, E. de Miguel, L. F. Rull, G. Jackson, and I. A. Mclure, J. Chem. Phys. **96**, 2296 (1992) for the case $\lambda = 1.25$, $d_m = 0.0025$, $n_s = 150$ and $n_v = 1$.

Temperature	Low density phase		High density phase	
	This work	Vega et al.	This work	Vega et al.
0.66	0.043	0.062	0.816	0.823
0.68	0.059	0.060	0.788	0.792
0.70	0.080	0.085	0.754	0.761
0.72	0.110	0.103	0.707	0.708

Table 14S: Comparison of the vapour-liquid equilibrium number density computed by us and those reported in L. Vega, E. de Miguel, L. F. Rull, G. Jackson, and I. A. Mclure, J. Chem. Phys. **96**, 2296 (1992) for the case $\lambda = 1.50$, $d_m = 0.005$, $n_s = 150$ and $n_v = 1$.

Temperature	Low density phase		High density phase	
	This work	Vega et al.	This work	Vega et al.
1.0	0.031	0.038	0.647	0.659
1.05	0.045	0.053	0.619	0.632
1.08	0.056	0.051	0.599	0.599
1.10	0.065	0.058	0.584	0.579
1.12	0.076	0.078	0.568	0.567

Table 15S: Comparison of the vapour-liquid equilibrium number density computed by us and those reported in L. Vega, E. de Miguel, L. F. Rull, G. Jackson, and I. A. Mclure, J. Chem. Phys. **96**, 2296 (1992) for the case $\lambda = 1.75$, $d_m = 0.005$, $n_s = 150$ and $n_v = 1$.

Temperature	Low density phase		High density phase	
	This work	Vega et al.	This work	Vega et al.
1.55	0.046	0.048	0.534	0.537
1.57	0.051	0.053	0.525	0.529
1.60	0.058	0.059	0.511	0.514
1.62	0.063	0.062	0.501	0.503