Table S1: Critical Temperature (T_c), Critical Pressure (P_c) and Acentric Factor (ω) values for the components studied in this work.

Component	T_c(K)	P_c(MPa)	ω	Ref
CH_4	190.564	4.599	0.0115	17
C_3H_8	369.830	4.248	0.1523	17
n-C_4H_{10}	425.120	3.796	0.2002	17
i-C_5H_{12}	460.430	3.381	0.2275	17
n-C_5H_{12}	469.700	3.370	0.2515	17
n-C_6H_{14}	507.600	3.025	0.3013	17
n-C_8H_{18}	568.700	2.490	0.3996	17
n-$C_{10}H_{22}$	617.700	2.110	0.4923	17
n-$C_{12}H_{26}$	658.000	1.820	0.5764	17
n-$C_{16}H_{34}$	723.000	1.400	0.7174	17
n-$C_{20}H_{42}$	768.000	1.160	0.9069	17
n-$C_{24}H_{50}$	804.000	0.980	1.0710	17

Table S2: PC-SAFT EoS parameters for the components studied in this work.

Component	m	σ(Å)	ε/k_B(K)	Ref
CH_4	1.0000	3.7039	150.03	5
C_3H_8	2.0020	3.6184	208.11	5
n-C_4H_{10}	2.3316	3.7086	222.88	5
i-C_5H_{12}	2.5620	3.8296	230.75	5
n-C_5H_{12}	2.6896	3.7729	231.20	5
n-C_6H_{14}	3.0576	3.7983	236.77	5
n-C_8H_{18}	3.8176	3.8373	242.78	5
n-$C_{10}H_{22}$	4.6627	3.8384	243.87	5
n-$C_{12}H_{26}$	5.3060	3.8959	249.21	5
n-$C_{16}H_{34}$	6.6485	3.9552	254.70	5
n-$C_{20}H_{42}$	7.9849	3.9869	257.75	5
n-$C_{24}H_{50}$	9.6836	3.9709	254.69	18

Table S3: CH_4 - n-$C_{10}H_{22}$ mixture Gibbs Ensemble Monte Carlo simulation data. The statistical uncertainty in the last digit is given in parentheses (i.e., 0.503(3) is 0.503±0.003).

Temperature (K)	Pressure (MPa)	CH_4 mole fraction liquid phase	CH_4 mole fraction vapor phase
244	10.01	0.503(3)	0.997(1)
244	15.02	0.59(1)	0.997(1)
244	19.97	0.66(2)	0.992(2)
244	25.05	0.72(3)	0.984(2)
244	30.03	0.76(2)	0.973(3)
244	35.02	0.81(2)	0.95(1)
255	1.00	0.065(3)	0.962(1)
255	9.99	0.464(5)	0.9971(1)
255	25.05	0.72(1)	0.988(3)
255	30.02	0.77(1)	0.976(4)
255	34.98	0.81(1)	0.957(6)
277	0.99	0.054(3)	0.962(1)
277	5.03	0.244(8)	0.9926(1)
277	10.03	0.42(1)	0.9966(2)
277	20.03	0.628(5)	0.992(1)
277	25.00	0.70(1)	0.985(1)
277	30.02	0.75(3)	0.97(1)
277	35.02	0.80(2)	0.96(2)
283	1.00	0.051(2)	0.961(1)
283	5.01	0.242(3)	0.9923(1)
283	10.01	0.40(1)	0.9966(1)
283	13.60	0.502(6)	0.9985(1)
283	17.39	0.577(1)	0.9962(6)
283	24.07	0.68(1)	0.984(5)
283	30.04	0.75(3)	0.96(1)
283	34.97	0.81(4)	0.96(2)
303	1.00	0.048(2)	0.960(1)
303	5.07	0.221(4)	0.9918(1)
303	10.03	0.380(2)	0.9962(1)
303	14.86	0.501(5)	0.9973(2)
303	25.13	0.67(1)	0.98(1)
303	31.49	0.77(4)	0.95(2)

310	1.00	0.045(2)	0.958(1)
310	4.97	0.212(6)	0.9915(2)
310	15.52	0.505(3)	0.9965(4)
310	17.27	0.541(5)	0.9956(7)
310	18.96	0.571(6)	0.9937(6)
310	20.66	0.603(7)	0.991(1)
310	22.47	0.63(1)	0.989(2)
310	24.17	0.65(1)	0.98(1)
310	25.82	0.68(1)	0.979(6)
310	27.53	0.71(1)	0.96(1)
310	29.26	0.73(1)	0.95(1)
310	32.77	0.78(1)	0.94(2)
310	34.45	0.8(1)	0.95(5)
450	4.99	0.167(5)	0.948(4)
450	9.97	0.308(3)	0.956(2)
450	14.99	0.433(3)	0.950(1)
450	19.95	0.544(3)	0.933(4)
450	24.97	0.73(4)	0.84(4)
550	5.02	0.156(3)	0.652(2)
550	9.98	0.40(7)	0.659(8)
550	11.07	0.47(8)	0.655(9)
550	13.01	0.56(6)	0.65(4)

Table S4: CH_4 - n-$C_{12}H_{26}$ mixture Gibbs Ensemble Monte Carlo simulation data. The statistical uncertainty in the last digit is given in parentheses.

Temperature (K)	Pressure (MPa)	CH_4 mole fraction liquid phase	CH_4 mole fraction vapor phase
283	1.00	0.056(4)	0.961(1)
283	5.02	0.242(6)	0.9927(1)
283	9.99	0.398(7)	0.9967(1)
283	15.03	0.51(1)	0.9979(1)
283	21.03	0.606(8)	0.997(2)
283	29.46	0.701(9)	0.98(1)
283	35.39	0.76(3)	0.98(2)
303	1.00	0.051(5)	0.960(2)
303	5.03	0.223(4)	0.9921(1)
303	10.04	0.37(1)	0.9963(1)
303	21.95	0.606(4)	0.996(1)
303	30.03	0.705(8)	0.988(3)
303	35.27	0.75(1)	0.983(4)
303	40.24	0.82(3)	0.971(5)
323	1.00	0.045(3)	0.958(1)
323	4.98	0.206(4)	0.9914(1)
323	9.98	0.354(7)	0.9959(1)
323	20.01	0.560(5)	0.997(2)
323	24.99	0.632(5)	0.994(6)
323	30.04	0.69(1)	0.98(1)
323	34.96	0.75(1)	0.97(2)
323	40.00	0.81(2)	0.96(1)
373	1.01	0.039(1)	0.9523(8)
373	5.02	0.184(2)	0.9900(1)
373	9.98	0.325(4)	0.9951(1)
373	15.01	0.442(3)	0.9962(3)
373	20.00	0.536(3)	0.9935(3)
373	25.02	0.615(4)	0.9892(8)
373	30.05	0.683(6)	0.981(1)
373	35.00	0.75(2)	0.96(1)
373	40.00	0.85(4)	0.92(3)
400	5.00	0.181(3)	0.9955(5)

400	10.00	0.314(5)	0.9948(5)
400	15.00	0.433(2)	0.9934(5)
400	20.00	0.524(5)	0.9895(7)
400	25.00	0.608(7)	0.983(1)
400	30.00	0.68(1)	0.971(5)
400	32.00	0.72(2)	0.964(5)
400	35.00	0.78(2)	0.93(1)
400	37.00	0.82(4)	0.91(3)
450	5.00	0.1717	0.9811
450	10.00	0.3119	0.9830
450	15.00	0.4311	0.9813
450	20.00	0.5241	0.9738
450	25.00	0.6094	0.9591
450	26.00	0.637(2)	0.960(2)
450	28.00	0.669(7)	0.949(4)
500	1.04	0.033(1)	0.79(1)
500	5.00	0.170(3)	0.942(2)
500	10.00	0.312(4)	0.952(2)
500	15.00	0.43(1)	0.947(5)
500	20.00	0.53(1)	0.933(7)
500	21.00	0.573(3)	0.933(2)
500	23.00	0.611(1)	0.923(1)
500	25.00	0.67(7)	0.89(2)
550	0.99	0.021(1)	0.46(2)
550	2.00	0.061(2)	0.70(1)
550	5.00	0.167(2)	0.84(1)
550	8.00	0.263(5)	0.876(5)
550	10.00	0.327(8)	0.88(1)
550	15.00	0.45(7)	0.85(4)
550	16.00	0.50(4)	0.83(2)
550	17.00	0.56(6)	0.79(5)
550	18.00	0.60(2)	0.75(2)
550	19.00	0.64(2)	0.68(4)
600	1.05	0.0056(1)	0.067(2)
600	2.54	0.073(2)	0.45(2)

600	5.11	0.210(5)	0.50(3)

Table S5: CH_4 - *n*-$C_{16}H_{34}$ mixture Gibbs Ensemble Monte Carlo simulation data. The statistical uncertainty in the last digit is given in parentheses.

Temperature (K)	Pressure (MPa)	CH_4 mole fraction liquid phase	CH_4 mole fraction vapor phase
340	0.99	0.049(5)	1.0000(1)
340	4.98	0.210(5)	1.0000(1)
340	10.01	0.359(2)	1.0000(1)
340	15.03	0.463(8)	0.9971(1)
340	19.97	0.545(5)	0.9978(1)
340	24.56	0.602(2)	0.9984(1)
340	36.42	0.719(4)	0.9943(2)
340	53.30	0.85(1)	0.9697(5)
400	1.00	0.042(3)	0.9490(4)
400	4.98	0.186(3)	0.9890(1)
400	10.01	0.329(3)	0.9945(5)
400	15.02	0.436(5)	0.9964(3)
400	20.04	0.5243(6)	0.9973(2)
400	30.06	0.659(5)	0.9947(1)
462	2.08	0.083(1)	0.9934(1)
462	5.09	0.186(2)	0.9960(1)
462	10.29	0.331(2)	0.9960(1)
462	14.96	0.433(4)	0.9949(1)
462	20.08	0.526(7)	0.9928(4)
462	25.58	0.61(2)	0.989(2)
500	1.00	0.039(2)	0.952(1)
500	4.98	0.182(3)	0.9896(1)
500	9.95	0.323(3)	0.9900(1)
500	15.09	0.440(4)	0.988(1)
500	19.96	0.532(3)	0.9855(1)
500	29.95	0.683(4)	0.972(1)
500	35.00	0.77(2)	0.94(1)
550	1.00	0.0384(3)	0.878(6)

550	4.98	0.184(1)	0.963(1)
550	9.97	0.331(3)	0.971(1)
550	15.07	0.451(6)	0.968(3)
550	20.05	0.556(5)	0.963(3)
600	1.00	0.0323(7)	0.65(1)
600	4.98	0.190(3)	0.901(4)
600	9.96	0.348(1)	0.927(3)
600	14.97	0.48(1)	0.91(2)
600	20.02	0.69(7)	0.86(4)
623	2.14	0.079(1)	0.731(2)
623	3.23	0.126(2)	0.81(1)
623	5.13	0.207(3)	0.86(1)
623	10.03	0.38(2)	0.87(2)
670	1.00	0.012(2)	0.14(1)
670	3.00	0.107(1)	0.531(9)
670	5.02	0.202(3)	0.66(2)
670	6.98	0.30(3)	0.71(3)

Table S6: CH_4 - *n*-$C_{20}H_{42}$ mixture Gibbs Ensemble Monte Carlo simulation data. The statistical uncertainty in the last digit is given in parentheses.

Temperature (K)	Pressure (MPa)	CH_4 mole fraction liquid phase	CH_4 mole fraction vapor phase
323	1.01	0.06(1)	1.0000(1)
323	5.02	0.25(1)	1.0000(1)
323	10.00	0.399(8)	0.9954(1)
323	15.02	0.50(1)	0.9970(1)
323	20.02	0.575(1)	0.9978(1)
323	25.01	0.62(1)	0.9982(1)
323	34.95	0.70(1)	0.9978(1)
323	39.99	0.73(1)	0.9980(1)
323	44.94	0.76(1)	0.9963(1)
323	62.76	0.83(1)	0.9892(4)
323	71.42	0.863(8)	0.9839(4)
323	79.87	0.91(2)	0.9674(4)

323	83.20	0.931(1)	0.956(6)
323	83.41	0.934(4)	0.953(3)
353	1.00	0.051(2)	0.9505(3)
353	4.99	0.22(1)	0.9896(1)
353	10.01	0.369(8)	0.9950(1)
353	14.98	0.47(1)	0.9967(1)
353	20.00	0.550(6)	0.9975(1)
353	24.99	0.611(7)	0.9980(1)
353	29.98	0.659(9)	0.9974(1)
353	35.04	0.701(3)	0.9977(1)
353	40.08	0.730(2)	0.9959(1)
353	45.11	0.765(8)	0.9945(4)
353	58.92	0.84(2)	0.987(1)
353	66.07	0.86(1)	0.982(1)
353	72.88	0.90(2)	0.968(6)
353	75.40	0.92(1)	0.960(4)
423	15.01	0.447(2)	0.9959(1)
423	20.05	0.533(2)	0.9969(1)
423	25.02	0.598(3)	0.9975(2)
423	35.08	0.706(5)	0.9973(4)
423	44.94	0.779(6)	0.992(2)
423	50.08	0.81(1)	0.987(3)
423	55.00	0.84(1)	0.978(5)
423	60.01	0.87(1)	0.970(1)
500	0.99	0.042(2)	0.9391(1)
500	5.04	0.195(4)	0.985(1)
500	10.07	0.336(6)	0.9928(2)
500	14.96	0.449(5)	0.9968(4)
500	20.05	0.53(1)	0.9956(6)
500	25.01	0.612(4)	0.9940(6)
500	30.07	0.67(1)	0.991(1)
500	34.96	0.728(8)	0.987(3)
500	40.05	0.780(8)	0.983(2)
500	45.04	0.83(2)	0.95(1)
550	1.00	0.043(2)	0.926(2)

550	5.05	0.199(5)	0.9848(1)
550	10.10	0.346(4)	0.9921(2)
550	14.93	0.462(5)	0.9908(4)
550	20.07	0.55(1)	0.9875(6)
550	25.07	0.629(4)	0.9847(6)
550	30.04	0.69(1)	0.977(1)
550	35.03	0.788(7)	0.952(3)
600	0.99	0.043(1)	0.890(8)
600	5.04	0.207(2)	0.969(8)
600	10.08	0.361(5)	0.975(2)
600	15.11	0.485(6)	0.975(1)
600	20.07	0.579(5)	0.970(1)
600	24.98	0.666(3)	0.962(4)
650	5.00	0.212(1)	0.917(2)
650	8.00	0.316(3)	0.935(1)
650	10.00	0.373(2)	0.935(1)
650	12.00	0.428(7)	0.94(1)
650	15.00	0.49(3)	0.93(1)
700	1.00	0.027(1)	0.35(1)
700	5.11	0.227(3)	0.78(1)
700	10.05	0.416(5)	0.83(1)
700	12.04	0.57(9)	0.79(1)

Table S7: CH_4 - *n*-$C_{24}H_{50}$ mixture Gibbs Ensemble Monte Carlo simulation data. The statistical uncertainty in the last digit is given in the parentheses.

Temperature (K)	Pressure (MPa)	CH_4 mole fraction liquid phase	CH_4 mole fraction vapor phase
330	10.02	0.43(1)	1.0000
330	15.01	0.52(2)	1.0000
330	20.00	0.597(5)	1.0000
330	30.03	0.68(1)	1.0000
330	35.01	0.71(1)	1.0000
330	40.04	0.73(1)	1.0000
330	44.97	0.76(1)	1.0000

330	49.96	0.78(2)	1.0000
330	59.55	0.817(4)	0.9951(1)
330	69.51	0.84(2)	0.9924(7)
330	83.10	0.88(1)	0.9870(6)
330	97.09	0.93(1)	0.965(5)
350	5.01	0.26(2)	1.0000
350	10.02	0.41(1)	1.0000
350	15.02	0.50(1)	1.0000
350	19.99	0.57(1)	1.0000
350	25.01	0.62(1)	1.0000
350	29.96	0.67(1)	1.0000
350	35.05	0.71(1)	1.0000
350	39.96	0.740(6)	1.0000
350	45.00	0.76(1)	1.0000
350	57.29	0.81(1)	0.9949(2)
350	66.08	0.84(1)	0.9920(6)
350	78.04	0.88(1)	0.986(1)
350	90.14	0.94(1)	0.965(13)
374	0.90	0.060(1)	1.0000
374	5.05	0.242(6)	1.0000
374	10.02	0.38(1)	1.0000
374	15.01	0.48(1)	1.0000
374	19.98	0.56(1)	1.0000
374	25.02	0.62(1)	1.0000
374	30.00	0.67(1)	1.0000
374	35.00	0.70(1)	1.0000
374	39.97	0.736(8)	1.0000
374	45.04	0.765(5)	1.0000
374	49.98	0.79(1)	1.0000
374	55.15	0.815(6)	0.9946(4)
374	60.17	0.837(7)	0.9930(1)
374	71.07	0.873(7)	0.9874(3)
374	80.21	0.914(8)	0.976(6)
374	84.30	0.94(1)	0.963(4)
400	4.99	0.22(1)	0.9130(1)

400	10.04	0.393(6)	0.9323(2)
400	15.02	0.50(1)	0.9336(2)
400	19.97	0.570(6)	0.9504(4)
400	25.04	0.622(6)	0.962(1)
400	30.02	0.673(3)	0.9658(5)
400	35.03	0.71(1)	0.972(1)
400	40.07	0.74(1)	0.979(4)
400	45.04	0.77(1)	0.994(3)
400	50.01	0.797(6)	0.992(1)
400	55.06	0.835(4)	0.9898(6)
450	10.00	0.343(1)	0.9950(5)
450	20.00	0.544(3)	0.9983(1)
450	30.00	0.652(4)	0.9972(2)
450	40.00	0.74(1)	0.9911(1)
450	50.00	0.83(2)	0.9575(1)
500	0.98	0.048(2)	0.9983(1)
500	10.05	0.35(1)	0.9992(1)
500	15.03	0.46(1)	0.9990(1)
500	20.12	0.55(1)	0.9983(2)
500	25.03	0.62(1)	0.9977(3)
500	30.08	0.677(7)	0.996(1)
500	35.03	0.72(1)	0.995(1)
500	40.10	0.77(1)	0.992(3)
500	45.02	0.806(7)	0.987(5)
500	50.02	0.84(2)	0.97(1)
550	5.00	0.213(2)	0.9971(1)
550	10.00	0.36(1)	0.9973(1)
550	20.00	0.564(7)	0.9581(2)
550	30.00	0.694(4)	0.9932(4)
550	35.00	0.74(1)	0.989(1)
600	1.04	0.050(7)	0.963(7)
600	10.21	0.377(6)	0.990(1)
600	15.05	0.494(4)	0.990(1)
600	19.99	0.588(6)	0.988(2)
600	25.07	0.661(4)	0.985(2)

600	30.05	0.733(8)	0.981(3)
700	1.03	0.047(1)	0.71(1)
700	5.22	0.249(6)	0.916(8)
700	10.17	0.423(3)	0.937(3)
700	15.01	0.549(7)	0.93(1)
750	1.08	0.038(3)	0.38(4)
750	5.01	0.255(4)	0.78(2)
750	7.40	0.37(2)	0.80(4)
750	9.98	0.52(9)	0.82(3)

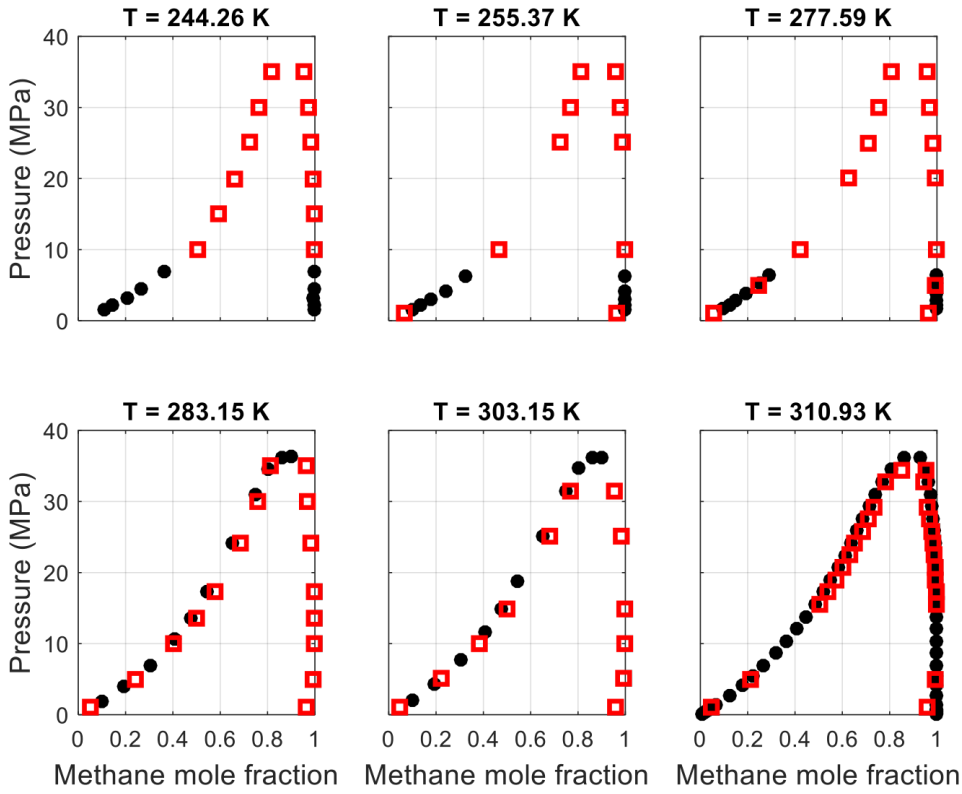

Figure S1: Pressure – composition VLE for the CH_4 - *n*-$C_{10}H_{22}$ mixture at various temperatures. Experimental data[19-21] are represented by black data points. GEMC simulation data are represented by red squares.

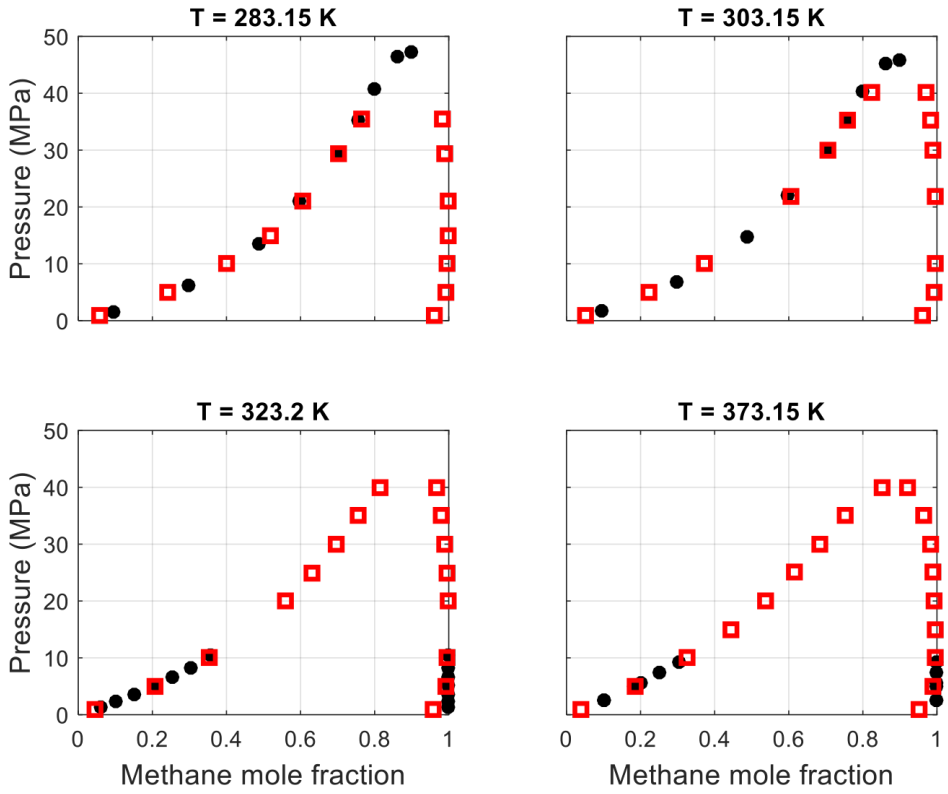

Figure S2: Pressure – composition VLE for the CH_4 - n-$C_{12}H_{26}$ mixture at various temperatures. Experimental data[22,23] are represented by black data points. GEMC simulation data are represented by red squares.

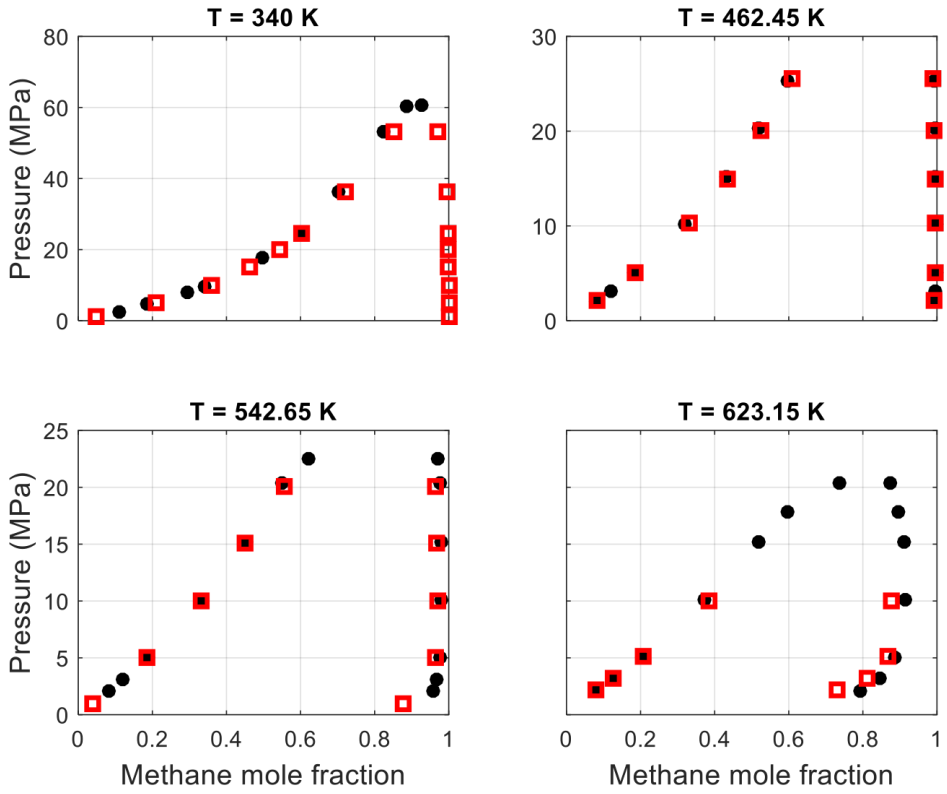

Figure S3: Pressure – composition VLE for the CH_4 - n-$C_{16}H_{34}$ mixture at various temperatures. Experimental data[24,25] are represented by black data points. GEMC simulation data are represented by red squares.

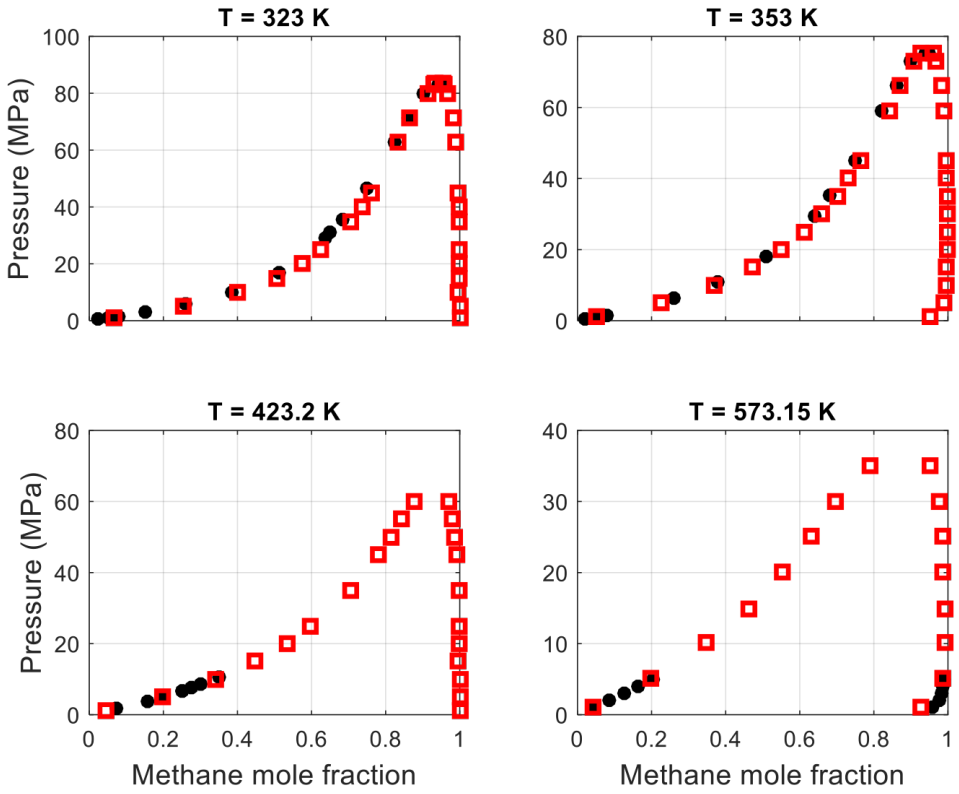

Figure S4: Pressure – composition VLE for the CH_4 - n-$C_{20}H_{42}$ mixture at various temperatures. Experimental data[26-28] are represented by black data points. GEMC simulation data are represented by red squares.

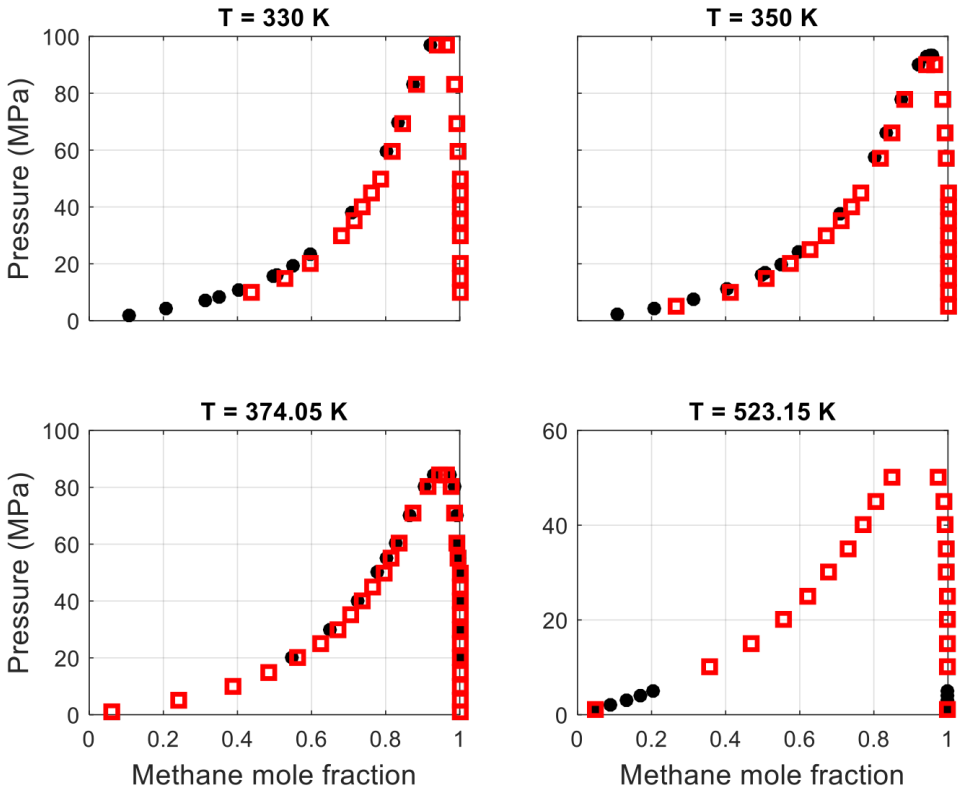

Figure S5: Pressure – composition VLE for the CH_4 - n-$C_{24}H_{50}$ mixture at various temperatures. Experimental data[29-31] are represented by black data points. GEMC simulation data are represented by red squares.