Vapor-liquid equilibria and mixture densities for three different diesel + N_2 mixtures to 535 K and 170 MPa

Supplemental Information

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Experimental Data

The following sections contain tables for bubble-point (BP) transitions and densities for mixtures of three different diesels (ULSD, Highly Paraffinic (HPF), and Highly Aromatic (HAR)) each with N_2.

BP Data

Tables S1 to S3 list experimental BP data for the HPF + N_2, ULSD + N_2, and HAR + N_2 systems respectfully. Table S4 lists experimental vapor pressure data for each diesel fuel. Figures S1a, b, and c show select p-x_{N2} isotherms for HPF + N_2, ULSD + N_2, and HAR + N_2 mixtures using the reported $M_{w,ave}$ for these diesels.

Table S1. Isopleths with BP data obtained in this study for Highly Paraffinic (HPF) ($M_{w,ave}$ = 212.0) + N_2 at varying nitrogen mole fractions (x_{N2}), weight fractions (w_{N2}), pressures, and temperatures. Twenty eight BP temperatures are within ± 0.40 K and one BP temperature at 10.6 wt% N_2 and 357.6 K is within ± 0.73 K.

x_{N2}	w_{N2}	p/MPa	T/K	x_{N2}	w_{N2}	p/MPa	T/K
0.254	0.043	32.3	299.3	0.547	0.137	114.0	338.5
		23.8	356.2			81.2	395.1
		19.7	414.7			62.4	446.4
		17.1	471.9			52.6	490.2
		15.7	519.3			46.7	526.7
0.367	0.071	61.6	302.2	0.700	0.236	136.3	426.2
		45.2	359.0			105.9	464.0
		35.8	420.8			76.1	525.0
		30.1	477.8	0.757	0.292	130.4	466.7
		26.6	522.9			113.8	485.7
0.472	0.106	104.5	299.2			99.2	506.5
		71.3	357.6			87.0	528.7
		54.6	419.5	0.895	0.530	143.6	512.0
		43.0	481.9			126.7	520.8
		38.0	524.7				

Table S2. Isopleths with BP data obtained in this study for ULSD ($M_{w,ave}$ = 199.9) + N_2 at varying nitrogen mole fractions (x_{N2}), weight fractions (w_{N2}), pressures, and temperatures. Twenty four BP temperatures are within ± 0.40 K and one BP temperature at 25.4 wt% N_2 and 494.0 K is within ± 0.71 K.

x_{N2}	w_{N2}	p/MPa	T/K	x_{N2}	w_{N2}	p/MPa	T/K
0.258	0.046	41.1	301.0	0.708	0.254	138.1	434.1
		29.1	372.0			112.6	463.8
		21.6	460.9			92.6	494.0
		18.3	529.5			72.8	534.0
0.445	0.101	99.9	300.4	0.744	0.289	140.9	455.1
		67.6	362.2			117.8	479.0
		51.5	418.8			100.2	503.2
		42.7	476.3			88.0	524.2
		36.4	523.1	0.892	0.536	155.2	495.3
0.564	0.153	99.3	373.5			138.3	513.4
		79.5	413.9			125.7	528.2
		66.3	453.6				
		56.5	493.2				
		49.1	527.4				

Table S3. Isopleths with BP data obtained in this study for Highly Aromatic (HAR) ($M_{w,ave}$ = 194.5) + N_2 at varying nitrogen mole fractions (x_{N2}), weight fractions (w_{N2}), pressures, and temperatures. Twenty one BP temperatures are within ± 0.40 K and one BP temperature at 49.3 wt% N_2 and 490.5 K is within ± 0.52 K.

x_{N2}	w_{N2}	p/MPa	T/K	x_{N2}	w_{N2}	p/MPa	T/K
0.161	0.027	21.9	301.1	0.643	0.206	146.9	393.6
		18.1	366.7			111.2	433.9
		15.3	427.4			88.8	474.3
		13.2	487.5			75.6	504.7
		12.3	528.9			66.1	530.8
0.489	0.121	116.0	302.5	0.748	0.300	145.7	449.8
		77.2	365.3			123.1	470.8
		57.9	425.2			99.2	501.7
		46.9	481.8			82.0	533.8
		39.4	533.1	0.871	0.493	158.2	490.5
						140.0	510.7
						118.4	532.1

Table S4. Vapor pressure of neat HPF, ULSD, and HAR obtained in this study.

T/K	*p*/MPa	T/K	*p*/MPa	T/K	*p*/MPa
HPF		ULSD		HAR	
273.15	$1.1 \cdot 10^{-5}$	273.15	$3.4 \cdot 10^{-5}$	273.15	$4.8 \cdot 10^{-5}$
298.15	$5.5 \cdot 10^{-5}$	298.15	$1.4 \cdot 10^{-5}$	298.15	$1.9 \cdot 10^{-4}$
323.15	$2.3 \cdot 10^{-4}$	323.15	$4.9 \cdot 10^{-4}$	323.15	$6.4 \cdot 10^{-4}$
348.15	$7.4 \cdot 10^{-4}$	348.15	$1.4 \cdot 10^{-4}$	348.15	$1.8 \cdot 10^{-4}$
373.15	$2.1 \cdot 10^{-3}$	373.15	$3.4 \cdot 10^{-3}$	373.15	$4.2 \cdot 10^{-3}$
398.15	$5.0 \cdot 10^{-3}$	398.15	$7.6 \cdot 10^{-3}$	398.15	$9.0 \cdot 10^{-3}$
423.15	$1.1 \cdot 10^{-2}$	423.15	$1.5 \cdot 10^{-2}$	423.15	$1.8 \cdot 10^{-2}$
448.15	$2.3 \cdot 10^{-2}$	448.15	$2.9 \cdot 10^{-2}$	448.15	$3.2 \cdot 10^{-2}$
473.15	$4.2 \cdot 10^{-2}$	473.15	$5.0 \cdot 10^{-2}$	473.15	$5.6 \cdot 10^{-2}$
498.15	$7.4 \cdot 10^{-2}$	498.15	$8.2 \cdot 10^{-2}$	498.15	$9.0 \cdot 10^{-2}$
513.15	$1.0 \cdot 10^{-1}$	509.15	$1.0 \cdot 10^{-1}$	505.15	$1.0 \cdot 10^{-1}$

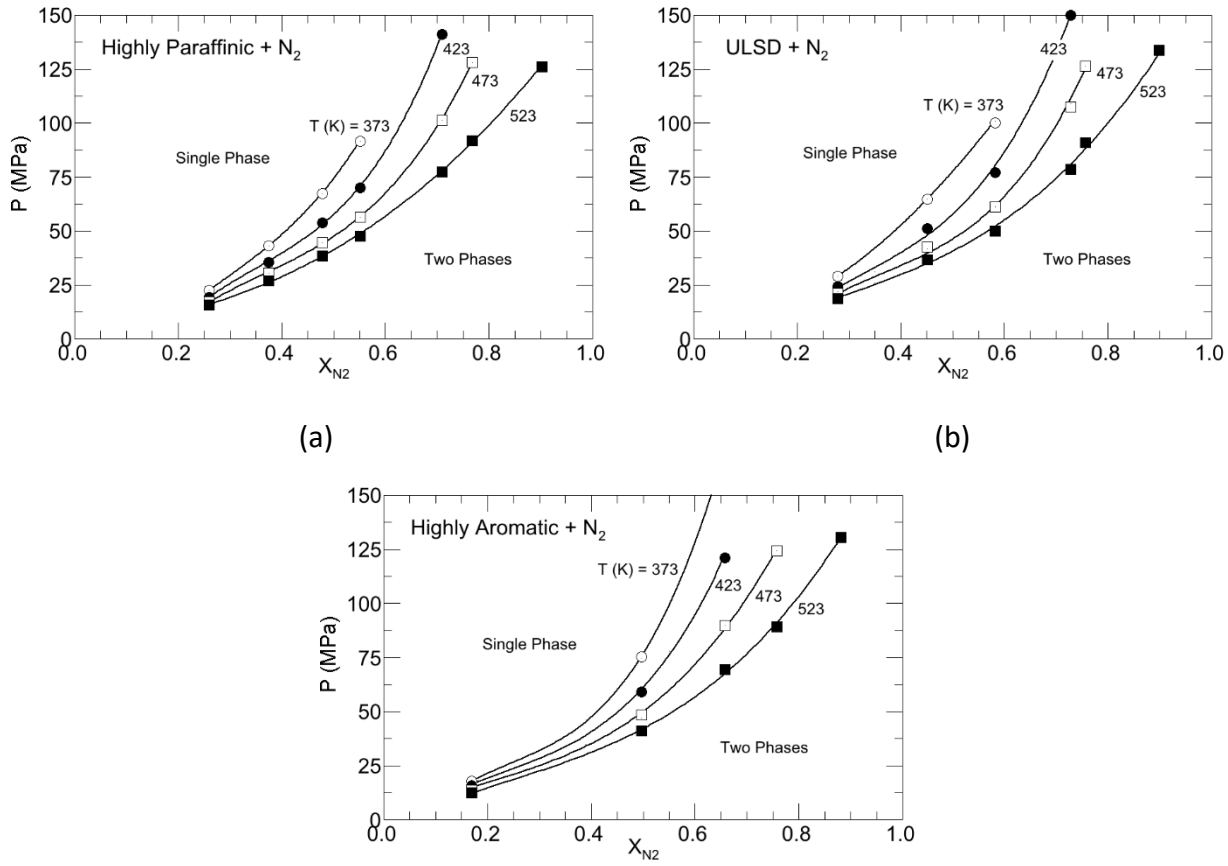

Figure S1. *P*-x_{N2} isotherms at 373, 423, 473, and 523 K for Highly Paraffinic + N_2 mixtures (a), ULSD + N_2 (b), and Highly Aromatic + N_2 mixtures (b). Lines are drawn to guide the eye.

Mixture Density Data

Tables S5, S6, and S7 list densities for Highly Paraffinic (HPF) + N_2, ULSD + N_2, and Highly Aromatic (HAR) + N_2 mixtures, respectfully. The first pressure-density data point at each fixed composition and temperature listed in these tables represents the two-phase boundary for a given isopleth.

Table S5. Densities, ρ, obtained in this study for single-phase, Highly Paraffinic (HPF) ($M_{w,ave}$ = 212.0) + N_2 mixtures at varying nitrogen mole fractions (x_{N2}), weight fractions (w_{N2}), pressures, and temperatures.

p/MPa	ρ/kg·m^{-3}	p/MPa	ρ/kg·m^{-3}	p/MPa	ρ/kg·m^{-3}	p/MPa	ρ/kg·m^{-3}
x_{N2} = 0.254; w_{N2} = 0.043							
T/K = 299.3		T/K = 414.7		T/K = 472.0		T/K = 519.5	
32.3	815	19.7	726	17.1	686	15.7	649
52.6	834	41.3	767	37.9	728	35.8	695
71.6	847	61.9	787	58.6	752	56.1	723
92.2	859	81.3	802	77.7	770	76.5	745
111.8	869	101.5	816	97.3	786	93.7	760
124.2	881	122.3	828	110.9	796	104.3	769
				124.3	805	123.1	783
T/K = 356.2							
23.8	768						
44.4	801						
62.0	816						
81.4	829						
101.9	842						
121.3	852						

Table S5. Cont': Single-phase densities, ρ, Highly Paraffinic (HPF) ($M_{w,ave}$ = 212.0) + N_2 mixtures.

p/MPa	ρ/kg·m^{-3}	p/MPa	ρ/kg·m^{-3}	p/MPa	ρ/kg·m^{-3}	p/MPa	ρ/kg·m^{-3}
x_{N2} = 0.367; w_{N2} = 0.071							
T/K = 302.2		T/K = 420.8		T/K = 478.0		T/K = 522.9	
61.6	822	35.8	727	30.1	676	26.6	637
74.6	833	49.2	747	44.2	706	44.6	678
88.3	842	63.0	762	59.0	727	62.1	703
101.6	850	75.3	774	80.6	750	77.8	724
115.7	859	88.2	785	100.2	768	95.1	742
128.8	866	102.8	796	117.8	781	111.6	756
T/K =	359.1	115.9	805	131.5	790	131.7	771
45.2	775	130.4	814				
59.3	790						
73.6	803						
87.0	813						
101.4	823						
117.2	833						
130.8	841						
x_{N2} = 0.472; w_{N2} = 0.106							
T/K = 299.2		T/K = 419.5		T/K = 482.0		T/K = 524.7	
104.5	848	54.6	730	43.0	666	38.0	631
110.8	853	61.5	741	61.5	702	51.0	659
117.6	858	74.5	756	73.5	718	62.1	677
T/K =	357.6	88.1	771	88.5	736	79.8	701
71.3	785	102.1	784	101.4	749	98.2	722
81.1	795	118.3	798	117.7	764	121.1	745
91.4	805						
101.1	813						
109.3	820						
118.2	827						

Table S5. Cont': Single-phase densities, ρ, Highly Paraffinic (HPF) ($M_{w,ave}$ = 212.0) + N_2 mixtures.

p/MPa	ρ/kg·m^{-3}	p/MPa	ρ/kg·m^{-3}	p/MPa	ρ/kg·m^{-3}	p/MPa	ρ/kg·m^{-3}
x_{N2} = 0.547; w_{N2} = 0.137							
T/K = 338.4		T/K = 446.4		T/K = 490.3		T/K = 526.9	
114.0	829	62.4	699	52.6	656	46.7	616
117.3	832	71.3	717	63.0	675	62.2	648
124.7	839	78.4	726	77.1	697	75.2	670
131.0	844	88.8	740	89.0	713	88.3	689
		98.0	751	98.5	725	101.7	705
T/K = 395.1		108.8	762	109.0	736	115.3	720
81.2	748	118.6	772	118.8	746	130.3	736
89.6	760	131.0	699	130.6	758		
95.4	770						
102.4	779						
109.5	790						
117.2	800						
124.5	806						
131.1	810						
x_{N2} = 0.700; w_{N2} = 0.236							
T/K = 426.2		T/K = 464.0		T/K = 525.0			
136.3	771	105.9	713	76.1	626		
140.9	776	111.2	720	82.8	638		
		117.2	727	95.7	660		
		124.3	736	109.3	680		
		130.8	743	122.1	697		
		139.9	752	137.3	715		

Table S5. Cont': Single-phase densities, ρ, Highly Paraffinic (HPF) ($M_{w,ave}$ = 212.0) + N_2 mixtures.

p/MPa	ρ/kg·m^{-3}	p/MPa	ρ/kg·m^{-3}	p/MPa	ρ/kg·m^{-3}	p/MPa	ρ/kg·m^{-3}
x_{N2} = 0.757; w_{N2} = 0.292							
T/K = 466.7		T/K = 485.7		T/K = 506.5		T/K = 528.9	
130.4	710	113.8	677	99.2	642	87.0	609
137.3	717	120.8	686	104.5	652	99.0	632
		128.6	696	110.8	661	107.5	645
		138.5	707	118.3	672	118.3	661
				124.3	680	128.6	674
				131.3	689	138.6	687
				138.5	697		
x_{N2} = 0.895; w_{N2} = 0.530							
T/K = 512.0		T/K = 520.8					
143.6	615	126.7	586				
147.4	621	134.1	600				
154.1	632	140.3	611				
160.1	641	148.1	622				
		155.1	633				

Table S6. Densities, ρ, obtained in this study for single-phase, ULSD ($M_{w,ave}$ = 199.9) + N_2 mixtures at varying nitrogen mole fractions (x_{N2}), weight fractions (w_{N2}), pressures, and temperatures.

p/MPa	ρ/kg·m^{-3}	p/MPa	ρ/kg·m^{-3}	p/MPa	ρ/kg·m^{-3}	p/MPa	ρ/kg·m^{-3}
x_{N2} = 0.258; w_{N2} = 0.046							
T/K = 301.0		T/K =372.0		T/K = 460.9		T/K = 529.3	
41.1	833	29.1	774	21.6	695	18.3	633
55.2	845	42.6	792	41.1	731	34.6	677
68.6	854	56.5	806	62.0	756	52.1	706
82.0	863	69.9	817	75.8	770	68.6	727
94.4	870	82.6	827	87.9	781	84.6	744
107.8	877	95.7	836	101.3	792	101.6	760
		108.8	844	108.2	797	109.4	767
x_{N2} = 0.445; w_{N2} = 0.101							
T/K = 300.4		T/K = 418.8		T/K = 476.3		T/K = 523.1	
99.9	848	51.2	731	42.7	674	36.4	627
108.1	854	61.6	747	56.0	700	49.3	657
114.1	859	71.3	758	68.7	718	62.0	679
		82.4	771	81.4	734	74.8	698
T/K =	362.2	91.3	779	93.7	748	89.0	716
67.6	785	101.8	789	107.4	761	102.0	730
77.7	796	111.2	797	113.5	767	114.3	742
88.1	806	116.5	801				
98.3	815						
107.7	822						
117.4	830						

Table S6. Continued: Densities, ρ, of single-phase, ULSD ($M_{w,ave}$ = 199.9) + N_2 mixtures.

p/MPa	ρ/kg·m^{-3}	p/MPa	ρ/kg·m^{-3}	p/MPa	ρ/kg·m^{-3}	p/MPa	ρ/kg·m^{-3}
x_{N2} = 0.564; w_{N2} = 0.153							
T/K = 373.5		T/K = 453.6		T/K = 493.2		T/K = 527.6	
99.3	801	66.3	709	56.5	661	49.1	620
104.0	806	77.5	725	65.9	680	62.2	645
		88.7	741	76.6	698	74.8	669
T/K =	413.9	98.2	754	85.7	712	87.7	690
79.5	753	108.6	766	96.4	727	101.0	709
88.3	765			107.4	740	109.2	720
98.2	777						
108.6	788						
x_{N2} = 0.708; w_{N2} = 0.254							
T/K = 434.1		T/K = 463.8		T/K = 494.2		T/K = 534.0	
138.1	773	112.6	725	92.6	674	72.8	610
142.7	778	117.8	731	99.0	684	84.9	636
		124.9	740	105.4	694	97.9	659
		130.8	747	112.2	704	111.4	680
		139.1	757	119.5	714	126.1	700
				126.2	724	138.4	716
				132.7	731	146.0	724
				138.9	739		
				144.1	745		

Table S6. Continued: Densities, ρ, of single-phase, ULSD ($M_{w,ave}$ = 199.9) + N_2 mixtures.

p/MPa	ρ/kg·m^{-3}	p/MPa	ρ/kg·m^{-3}	p/MPa	ρ/kg·m^{-3}	p/MPa	ρ/kg·m^{-3}
x_{N2} = 0.744; w_{N2} = 0.289							
T/K = 455.1		T/K = 479.0		T/K = 503.2		T/K = 524.5	
140.9	734	117.8	692	100.2	653	88.0	618
145.2	739	124.3	701	108.6	665	94.7	631
		130.8	709	115.3	675	101.2	641
		137.7	717	120.9	682	108.6	654
		145.0	725	129.0	693	114.3	662
				135.0	701	124.6	676
				145.0	712	128.8	682
						136.2	691
						144.9	702
x_{N2} = 0.892; w_{N2} = 0.536							
T/K = 495.3		T/K = 513.4		T/K = 528.2			
155.2	649	138.3	617	125.7	588		
157.2	651	144.7	628	131.4	598		
160.5	657	151.5	638	139.3	612		
165.2	664	157.0	647	145.1	622		
				152.6	634		
				157.2	641		
				164.5	652		

Table S7. Densities, ρ, obtained in this study for single-phase, Highly Aromatic (HAR) ($M_{w,ave}$ = 194.5) + N_2 mixtures at varying nitrogen mole fractions (x_{N2}), weight fractions (w_{N2}), pressures, and temperatures.

p/MPa	ρ/kg·m^{-3}	p/MPa	ρ/kg·m^{-3}	p/MPa	ρ/kg·m^{-3}	p/MPa	ρ/kg·m^{-3}
x_{N2} = 0.161; w_{N2} = 0.027							
T/K = 301.1		T/K = 427.4		T/K = 487.5		T/K = 528.5	
21.9	826	15.3	728	13.2	684	12.3	650
41.0	847	34.5	767	34.6	728	33.4	701
61.1	862	55.7	790	55.4	754	54.4	731
82.7	874	75.0	807	74.6	774	75.7	755
102.7	885	95.5	822	95.4	791	96.1	774
122.1	894	112.1	832	110.7	803	114.7	789
		128.9	842	129.4	816	133.6	800
T/K = 366.7							
18.1	779						
37.6	807						
57.7	825						
72.7	836						
85.5	844						
98.8	852						
110.5	859						
127.8	868						

Table S7. Cont': Single-phase densities, ρ, Highly Aromatic (HAR) ($M_{w,ave}$ = 194.5) + N_2 mixtures.

p/MPa	ρ/kg·m^{-3}	p/MPa	ρ/kg·m^{-3}	p/MPa	ρ/kg·m^{-3}	p/MPa	ρ/kg·m^{-3}
x_{N2} = 0.489; w_{N2} = 0.121							
T/K = 302.5		T/K = 425.2		T/K = 481.8		T/K = 533.1	
116.0	887	57.9	749	46.9	690	39.4	634
123.0	893	71.5	770	61.7	718	59.9	681
129.6	897	84.9	787	77.7	743	81.0	716
136.3	903	97.7	801	95.4	765	102.4	743
		110.2	815	110.6	782	122.1	766
T/K =	365.3	123.7	828	127.6	800	139.0	783
77.2	813	137.7	840	140.0	811		
97.8	835						
110.9	848						
124.2	861						
137.3	871						
x_{N2} = 0.643; w_{N2} = 0.206							
T/K = 393.6		T/K = 474.4		T/K = 504.7		T/K = 530.8	
146.9	816	88.8	702	75.6	660	66.1	624
151.1	819	102.0	720	88.8	682	88.4	664
		115.6	736	101.8	700	108.4	693
T/K =	433.9	128.2	750	117.0	721	124.3	712
111.2	756	142.1	766	129.0	734	141.7	730
117.0	763	148.6	772	141.7	748	150.7	739
124.1	769			151.5	758		
130.6	776						
137.2	783						
143.9	790						
150.5	795						

Table S7. Cont': Single-phase densities, ρ, Highly Aromatic (HAR) ($M_{w,ave}$ = 194.5) + N_2 mixtures.

p/MPa	ρ/kg·m^{-3}	p/MPa	ρ/kg·m^{-3}	p/MPa	ρ/kg·m^{-3}	p/MPa	ρ/kg·m^{-3}
x_{N2} = 0.748; w_{N2} = 0.300							
T/K = 449.8		T/K = 470.8		T/K = 501.7		T/K = 533.5	
145.7	774	123.1	732	99.2	677	82.0	622
154.3	784	129.4	741	106.4	689	95.6	651
		135.7	749	118.0	707	108.8	673
		143.7	759	131.9	726	121.9	693
		154.6	773	142.9	741	134.1	711
				155.4	757	146.6	727
						155.3	740
x_{N2} = 0.871; w_{N2} = 0.493							
T/K = 490.5		T/K = 510.7		T/K = 532.1			
158.2	668	140.0	630	118.4	581		
160.5	672	144.1	636	124.7	592		
162.3	675	150.5	646	131.1	603		
165.0	678	157.8	656	137.9	615		
		164.1	664	144.7	625		
		140.0	630	151.2	635		
				158.4	646		
				164.0	653		

Correlation of Experimental Data

The following sections provide parameters for correlations to interpolate both the BP and mixture density data sets. Here the Antoine's equation is used to interpolate the BP data and the Tait equation is used to interpolate the mixture density data.

Correlation for Bubble Point Data

Equation S1 is Antoine's equation used to interpolate BP data and table S8 lists values for parameters *A*, *B,* and *C* needed in equation S1.

$$ln\left(\frac{p}{MPa}\right) = A - \frac{B}{T+C} \quad \text{(S1)}$$

Table S8. Constants for Antoine's equation used to interpolate BP isopleth data for ULSD, Highly Paraffinic, and Highly Aromatic each with N_2 at x_{N2} and w_{N2}. Refer to tables S1, S2, and S3 for the applicable temperature range of application for Antoine's equation for each isopleth. The average, absolute percent deviation, Δ_{AAD}, and maximum deviation, Δ_{max} are also listed.

x_{N2}	w_{N2}	A	B	C	Δ_{AAD}	Δ_{max}
			ULSD + N_2			
0.258	0.046	1.7181	$-6.7292 \cdot 10^2$	$3.5899 \cdot 10$	0.1	0.2
0.445	0.101	2.1233	$-8.1527 \cdot 10^2$	$2.8118 \cdot 10$	0.6	1.4
0.564	0.153	$3.9294 \cdot 10^{-1}$	$-3.2410 \cdot 10^3$	$3.9820 \cdot 10^2$	0.4	0.8
0.708	0.254	-7.2152	$-2.1871 \cdot 10^4$	$1.3672 \cdot 10^3$	0.3	0.5
0.744	0.289	2.1219	$-9.8516 \cdot 10^2$	$-1.0682 \cdot 10^2$	0.2	0.2
0.892	0.536	$-6.7147E \cdot 10^5$	$-7.0412 \cdot 10^{13}$	$1.0486 \cdot 10^8$	0.02	0.03
			Highly Paraffinic + N_2			
0.254	0.043	2.0793	$-2.8780 \cdot 10^2$	$-9.2950 \cdot 10$	0.2	0.5
0.367	0.071	1.9391	$-7.7583 \cdot 10^2$	$5.3707 \cdot 10$	0.5	0.7
0.472	0.106	2.1356	$-8.4717 \cdot 10^2$	$3.7919 \cdot 10$	0.6	1.3
0.547	0.127	2.0196	$-1.0395 \cdot 10^3$	$4.3927 \cdot 10$	0.5	0.8
0.700	0.236	1.8325	$-1.3072 \cdot 10^3$	-2.1226	0.01	0.02
0.757	0.292	2.1091	$-9.9459 \cdot 10^2$	$-1.0682 \cdot 10^2$	0.1	0.2
0.895	0.530	$-5.9281 \cdot 10$	$-2.8988 \cdot 10^5$	$4.0000 \cdot 10^3$	0.0	0.0

Table S8. Continued: Constants for Antoine's equation.

x_{N2}	w_{N2}	A	B	C	Δ_{AAD}	Δ_{max}
			Highly Aromatic + N_2			
0.161	0.027	$2.5701 \cdot 10^{-1}$	$-2.4606 \cdot 10^{3}$	$5.6826 \cdot 10^{2}$	0.6	1.0
0.489	0.121	1.8879	-1.0951E•10^{3}	7.8944E•10	0.4	0.7
0.643	0.206	1.6408	-1.4875E•10^{3}	5.0229E•10	0.6	1.1
0.748	0.300	2.0625	-1.0009E•10^{3}	-1.0682E•10^{2}	0.0	0.1
0.871	0.493	$-2.2623 \cdot 10^{6}$	-7.3400E•10^{3}	3.2444E•10^{8}	0.8	1.2

Single phase, mixture density data are correlated with the Tait equation, equations S2-S4 for the HPF, ULSD, and HAR diesels respectfully. This application of the Tait equation uses a reference density, $\rho_0(T)$, calculated at p_0 equal to the BP pressure at each T-w_{N2} value calculated with Antoine's equation. Values for parameters C, a_0, a_1, a_2, b_0, b_1, and b_2 are found in tables S9-S11.

$$\frac{\rho-\rho_0(T)}{\rho} = C \log_{10}\left(\frac{P+B(T)}{P_0+B(T)}\right) \quad \text{(S2)}$$

$$\rho_0(T)/kg \bullet m^{-3} = \sum_{i=0}^{2} a_i T^i \quad \text{(S3)}$$

$$B(\mathrm{T})/MPa = \sum_{i=0}^{2} b_i T^i \quad \text{(S4)}$$

Table S9. Tait parameters for mixture density calculations for Highly Paraffinic (HPF) + N_2 mixtures for each isopleth listed with the Δ_{AAD}, Δ_{max}, and the temperature-pressure ranges fit to the equation. The Tait parameters for neat HPF fuel are obtained from Rowane et al.[1].

x_{N2}:	0	0.254	0.367	0.472	0.55	0.700	0.76	0.89
w_{N2}:	0	0.043	0.071	0.106	0.14	0.236	0.29	0.53
C:	$2.2021 \cdot 10^{-1}$	$2.0004 \cdot 10^{-1}$	$3.0000 \cdot 10^{-1}$	$4.0000 \cdot 10^{-1}$	$3.0346 \cdot 10^{-1}$	$3.5104 \cdot 10^{-1}$	$4.5451 \cdot 10^{-1}$	$8.0676 \cdot 10^{-1}$
a2	$-4.3527 \cdot 10^{-4}$	$-3.0485 \cdot 10^{-4}$	$-2.5676 \cdot 10^{-4}$	$3.4132 \cdot 10^{-4}$	$1.9686 \cdot 10^{-3}$	$1.2336 \cdot 10^{-3}$	$2.4405 \cdot 10^{-3}$	$2.6736 \cdot 10^{-4}$
a1	$-3.8341 \cdot 10^{-1}$	$-4.8534 \cdot 10^{-1}$	$-6.0549 \cdot 10^{-1}$	-1.2077	-2.7193	-2.6413	-3.9985	-3.5054
a0	$9.7749 \cdot 10^{2}$	$9.8808 \cdot 10^{2}$	$1.0286 \cdot 10^{3}$	$1.1758 \cdot 10^{3}$	$1.5193 \cdot 10^{3}$	$1.6728 \cdot 10^{3}$	$2.0424 \cdot 10^{3}$	$2.3418 \cdot 10^{3}$
b2	$6.7987 \cdot 10^{-4}$	$1.8333 \cdot 10^{-4}$	$4.3377 \cdot 10^{-4}$	$1.4290 \cdot 10^{-4}$	$1.5971 \cdot 10^{-4}$	$1.4089 \cdot 10^{-4}$	$-6.2298 \cdot 10^{3}$	0.0000
b1	$-9.2733 \cdot 10^{-1}$	$-3.3700 \cdot 10^{-1}$	$-7.2665 \cdot 10^{-1}$	$-4.6374 \cdot 10^{-1}$	$-1.8012 \cdot 10^{-1}$	$-2.0276 \cdot 10^{-1}$	6.4225	-1.7440
b0	$3.2524 \cdot 10^{2}$	$1.2771 \cdot 10^{2}$	$2.6717 \cdot 10^{2}$	$2.1524 \cdot 10^{2}$	$3.9886 \cdot 10$	$4.0209 \cdot 10$	$-1.6733 \cdot 10^{3}$	$9.1286 \cdot 10^{2}$
Δ_{AAD}	0.1	0.21	0.30	0.40	0.44	0.01	0.12	0.19
Δ_{MAX}	0.6	1.12	0.27	1.07	0.77	0.04	0.42	0.41
T/K	298-530	299-519	302-523	299-525	338 - 527	426 - 525	467 - 529	512 - 521
p/MPa	4 - 300	BP* - 124	BP* - 132	BP* - 121	BP* - 131	BP* - 141	BP* - 141	BP* - 160

BP* -- The lowest pressure is the BP for each mixture.

Table S10. Tait parameters for mixture density calculations for ULSD + N_2 mixtures for each isopleth listed with the Δ_{AAD}, Δ_{max}, and the temperature-pressure ranges fit to the equation. The Tait parameters for neat ULSD fuel are obtained from Rowane et al.[1].

x_{N2}:	0	0.258	0.445	0.564	0.708	0.744	0.892
w_{N2}:	0	0.046	0.101	0.153	0.254	0.289	0.536
C:	$2.1022 \cdot 10^{-1}$	$1.9982 \cdot 10^{-1}$	$3.9860 \cdot 10^{-1}$	$8.4699 \cdot 10^{-1}$	$7.9795 \cdot 10^{-1}$	$4.4701 \cdot 10^{-1}$	$7.5327 \cdot 10^{-1}$
a_2:	$-1.2769 \cdot 10^{-4}$	$-5.4096 \cdot 10^{-4}$	$4.2990 \cdot 10^{-4}$	$9.8195 \cdot 10^{-4}$	$1.5949 \cdot 10^{-3}$	$2.6445 \cdot 10^{-3}$	$-4.0289 \cdot 10^{-3}$
a_1:	$-5.9072 \cdot 10^{-1}$	$-4.2308 \cdot 10^{-1}$	-1.2987	-2.0523	-3.1834	-4.2318	2.3246
a_0:	$1.0209 \cdot 10^{3}$	$1.0087 \cdot 10^{3}$	$1.1998 \cdot 10^{-3}$	$1.4342 \cdot 10^{3}$	$1.8586 \cdot 10^{3}$	$2.1114 \cdot 10^{3}$	$4.8546 \cdot 10^{2}$
b_2:	$9.6105 \cdot 10^{-4}$	$2.3293 \cdot 10^{-4}$	$1.2413 \cdot 10^{-4}$	$4.5725 \cdot 10^{-3}$	$2.1585 \cdot 10^{-2}$	$-6.2298 \cdot 10^{-3}$	$-1.9889 \cdot 10^{-3}$
b_1:	-1.1654	$-3.8671 \cdot 10^{-1}$	$-4.6413 \cdot 10^{-1}$	-4.4600	$-2.2427 \cdot 10$	6.4225	2.1474
b_0:	$3.7662 \cdot 10^{2}$	$1.3572 \cdot 10^{2}$	$2.2419 \cdot 10^{2}$	$1.1756 \cdot 10^{3}$	$5.8741 \cdot 10^{3}$	$-1.6733 \cdot 10^{3}$	$-5.9085 \cdot 10^{2}$
Δ_{AAD}:	0.1	0.1	0.3	0.4	0.3	0.1	0.1
Δ_{MAX}:	0.6	0.5	0.8	0.8	0.6	0.4	0.1
T_{range}/K	298 - 522	301 - 527	300 - 527	374 - 519	434 - 527	455 - 529	495 - 531
p_{range}/MPa	4 - 300	BP* - 99	BP* - 100	BP* - 100	BP* - 127	BP* - 129	BP* - 99

BP* -- The lowest pressure is the BP for each mixture.

Table S11. Tait parameters for mixture density calculations for Highly Aromatic (HAR) + N_2 mixtures for each isopleth listed with the Δ_{AAD}, Δ_{max}, and the temperature-pressure ranges fit to the equation. The Tait parameters for neat HAR fuel are obtained from Rowane et al. [1]

x_{N2}:	0	0.161	0.489	0.643	0.748	0.871
w_{N2}:	0	0.027	0.121	0.206	0.3	0.493
C:	$2.1022 \cdot 10^{-1}$	$3.9948 \cdot 10^{-1}$	$3.9771 \cdot 10^{-1}$	$3.1133 \cdot 10^{-1}$	$3.8333 \cdot 10^{-1}$	$9.9996 \cdot 10^{-1}$
a_2:	$-1.2769 \cdot 10^{-4}$	$-6.1463 \cdot 10^{-4}$	$5.8921 \cdot 10^{-4}$	$5.2057 \cdot 10^{-4}$	$3.4835 \cdot 10^{-3}$	$4.4688 \cdot 10^{-4}$
a_1:	$-5.9072 \cdot 10^{-1}$	$-2.6167 \cdot 10^{-1}$	-1.5401	-1.8821	-5.2151	-2.5482
a_0:	$1.0209 \cdot 10^{3}$	$9.6016 \cdot 10^{2}$	$1.2974 \cdot 10^{3}$	$1.4761 \cdot 10^{3}$	$2.4146 \cdot 10^{3}$	$1.8124 \cdot 10^{3}$
b_2:	$9.6105 \cdot 10^{-4}$	$1.4594 \cdot 10^{-4}$	$7.5701 \cdot 10^{-4}$	$2.7285 \cdot 10^{-3}$	$-9.1359 \cdot 10^{-4}$	$-7.3945 \cdot 10^{-3}$
b_1:	-1.1654	$-3.1331 \cdot 10^{-1}$	$-8.9403 \cdot 10^{-1}$	-2.7377	1.0282	7.2315
b_0:	$3.7662 \cdot 10^{2}$	$1.2718 \cdot 10^{2}$	$2.7655 \cdot 10^{2}$	$6.5713 \cdot 10^{2}$	-3.2350•102	$-1.7181 \cdot 10^{3}$
Δ_{AAD}:	0.2	0.2	0.2	0.2	0.1	0.1
Δ_{MAX}:	0.5	1	1.5	0.3	0.3	0.4
T_{range}/K	323 - 526	301 - 529	304 - 533	394 - 531	450 - 534	491 - 532
p_{range}/MPa	4 - 262	BP* - 134	BP* - 140	BP* - 152	BP* - 155	BP* - 165

BP* -- The lowest pressure is the BP for each mixture.

Modeling

In this section all modelling of the diesel + N_2 systems is performed using the PC-SAFT EoS. Here the diesel is treated as a single pseudo-component using the correlations of Rokni et al.[2, 3] to calculate the diesel PC-SAFT parameters m, σ, and ε/k_b. Rokni et al. provide two correlations where the first is developed using the GC data base of Burgess et al.[4] (B-GC) an the second using the GC database of Sauer et al.[5]

PC-SAFT (Density Predictions)

The following sections provide more PC-SAFT EoS modeling results for diesel density data. Tables S12 to S14 provide results when the B-GC (a) and S-GC (b) methods are used at each composition for HPF, ULSD, and HAR respectfully. In all cases for density modelling $k_{ij} = 0$.

Table S12. Results for PC-SAFT predicted mixture densities assuming the multi-component diesel can be represented as a single, pseudo-component with GC-calculated pure component parameters and with $k_{ij} = 0$ for HPF + N_2 mixtures using (a) the B-GC method and (b) the S-GC method.

(a) HAR + N_2 (B-GC Method)

	w_{N2}							
	0.043	0.071	0.106	0.137	0.236	0.292	0.530	Overall
Δ_{AAD}	1.4	1.0	1.3	1.9	1.7	3.6	3.9	1.8
Δ_{bias}	0.6	-0.6	-1.3	-1.8	-1.7	-3.6	-3.9	-1.3
Δ_{stdev}	0.7	1.0	1.4	1.7	1.3	0.8	0.5	1.5
Δ_{max}	2.4	4.5	5.1	6.7	4.5	5.6	4.6	6.7

(b) HAR + N_2 (S-GC Method)

	w_{N2}							
	0.043	0.071	0.106	0.137	0.236	0.292	0.530	Overall
Δ_{AAD}	7.5	6.1	5.2	4.3	3.4	1.1	1.4	4.8
Δ_{bias}	7.5	6.1	5.2	4.3	3.4	1.0	-1.4	4.7
Δ_{stdev}	0.7	0.4	0.6	1.1	0.9	0.5	0.4	2.2
Δ_{max}	8.5	6.6	6.1	6.2	4.6	1.7	1.9	8.5

Table S13. Results for PC-SAFT predicted mixture densities assuming the multi-component diesel can be represented as a single, pseudo-component with GC-calculated pure component parameters and with k_{ij} = 0 for ULSD + N_2 mixtures using (a) the B-GC method and (b) the the S-GC method.

(a) ULSD + N_2 (B-GC Method)

	w_{N2}						
	0.046	0.101	0.153	0.254	0.289	0.536	Overall
Δ_{AAD}	1.6	1.1	1.3	1.0	2.5	1.9	1.4
Δ_{bias}	1.1	-0.7	-0.7	-0.2	-2.5	-1.9	-0.4
Δ_{stdev}	0.9	1.3	1.2	0.8	0.9	0.6	1.1
Δ_{max}	4.1	5.3	4.8	3.6	4.5	2.6	5.3

(b) ULSD + N_2 (S-GC Method)

	w_{N2}						
	0.046	0.101	0.153	0.254	0.289	0.536	Overall
Δ_{AAD}	8.0	5.8	5.4	4.6	2.0	0.5	5.4
Δ_{bias}	8.0	5.8	5.4	4.6	2.0	0.5	5.4
Δ_{stdev}	0.4	0.7	1.0	0.8	0.5	0.5	2.2
Δ_{max}	8.6	6.7	6.8	5.7	2.8	1.5	8.9

Table S14. Results for PC-SAFT predicted mixture densities assuming the multi-component diesel can be represented as a single, pseudo-component with GC-calculated pure component parameters and with $k_{ij} = 0$ for HAR + N_2 mixtures using (a) the B-GC method and (b) the the S-GC method.

(a) HAR + N_2 (B-GC Method)

	w_{N2}					
	0.027	0.121	0.206	0.300	0.493	Overall
Δ_{AAD}	1.5	2.2	1.4	1.5	3.4	1.9
Δ_{bias}	0.9	1.7	-1.3	1.2	-3.4	0.1
Δ_{stdev}	0.8	1.1	1.3	0.8	0.6	1.2
Δ_{max}	2.9	3.8	5.1	2.7	4.7	5.1

(b) HAR + N_2 (S-GC Method)

	w_{N2}					
	0.027	0.121	0.206	0.300	0.493	Overall
Δ_{AAD}	7.9	7.7	3.9	5.5	0.7	5.7
Δ_{bias}	7.9	7.7	3.9	5.5	-0.7	5.6
Δ_{stdev}	0.4	1.0	0.9	0.7	0.5	2.5
Δ_{max}	8.6	8.9	5.3	6.4	1.7	8.9

PC-SAFT EoS (BP Predictions)

The following sections provide additional statistical measures at each temperature modeled and for the overall data set for the PC-SAFT EoS modeling results for BP data. Tables S15 to S17 provide modelling results when $k_{ij} = 0$ for both the B-GC (a) and S-GC (b) methods for the HPF, ULSD, and HAR diesels respectfully. Tables S18 to S20 provide modelling results when the best fit value for k_{ij} is used for both the B-GC (a) and S-GC (b) methods for HPF, ULSD, and HAR diesels respectfully.

Table S15. Results for PC-SAFT predicted p-w_{N2} isotherms for HPF + N_2 mixtures assuming the multi-component diesel can be represented as a single, pseudo-component with GC-calculated pure component parameters calculated with the B-GC and S-GC methods and with $k_{ij} = 0$.

(a) HPF + N_2 (B-GC Method, $k_{ij} = 0$)

T/K	300	373	423	473	530	Overall
Δ_{AAD}	38	21	17	16	18	19
Δ_{bias}	38	21	17	16	18	19
Δ_{stdev}	5	2	4	2	9	8
Δ_{max}	41	23	20	19	34	41
Data Points	2	4	5	6	7	24

(b) HPF + N_2 (S-GC Method, $k_{ij} = 0$)

T/K	300	373	423	473	530	Overall
Δ_{AAD}	175	124	107	92	86	106
Δ_{bias}	-191	-124	-107	-92	-86	-110
Δ_{stdev}	26	25	32	22	17	33
Δ_{max}	193	150	159	125	117	193
Data Points	2	4	5	6	7	24

Table S16. Results for PC-SAFT predicted p-w_{N2} isotherms for ULSD + N_2 mixtures assuming the multi-component diesel can be represented as a single, pseudo-component with GC-calculated pure component parameters calculated with the B-GC and S-GC methods and with $k_{ij} = 0$.

(a) ULSD + N_2 (B-GC Method, $k_{ij} = 0$)

T/K	300	373	423	473	530	Overall
Δ_{AAD}	57	17	12	13	18	19
Δ_{bias}	-57	-17	-12	-13	-17	-19
Δ_{stdev}	49	11	8	7	10	19
Δ_{max}	92	28	21	21	26	92
Data Points	2	3	4	5	6	20

(b) ULSD + N_2 (S-GC Method, $k_{ij} = 0$)

T/K	300	373	423	473	530	Overall
Δ_{AAD}	69	59	55	51	50	54
Δ_{bias}	69	59	55	51	50	54
Δ_{stdev}	2	1	5	5	6	8
Δ_{max}	71	60	63	57	62	71
Data Points	2	3	4	5	6	20

Table S17. Results for PC-SAFT predicted p-w_{N2} isotherms for ULSD + N_2 mixtures assuming the multi-component diesel can be represented as a single, pseudo-component with GC-calculated pure component parameters calculated with the B-GC and S-GC methods and with $k_{ij} = 0$.

(a) HAR + N_2 (B-GC Method, $k_{ij} = 0$)

T/K	300	373	423	473	530	Overall
Δ_{AAD}	19	15	14	18	20	18
Δ_{bias}	19	15	12	16	17	15
Δ_{stdev}	-	19	11	7	7	9
Δ_{max}	19	29	24	22	28	29
Data Points	1	2	3	5	5	16

(b) HAR+ N_2 (S-GC Method, $k_{ij} = 0$)

T/K	300	373	423	473	530	Overall
Δ_{AAD}	198	132	120	111	88	120
Δ_{bias}	-198	-132	-120	-111	-88	-120
Δ_{stdev}	26	7	27	26	14	39
Δ_{max}	217	137	147	150	104	217
Data Points	2	2	3	5	4	16

Table S18. Results for PC-SAFT predicted p-w_{N2} isotherms for HPF + N_2 mixtures assuming the multi-component diesel can be represented as a single, pseudo-component with GC-calculated pure component parameters calculated with the B-GC and S-GC methods and with a nonzero k_{ij}

(a) HPF + N_2 (B-GC Method, k_{ij} = -0.0350)

T/K	300	373	423	473	530	Overall
Δ_{AAD}	2	6	7	4	5	5
Δ_{bias}	2	-5	-5	-1	4	-1
Δ_{stdev}	1	3	8	3	5	5
Δ_{max}	3	10	21	9	14	21
Data Points	3	4	5	6	7	25

(b) HPF + N_2 (S-GC Method, k_{ij} = 0.1375)

T/K	300	373	423	473	530	Overall
Δ_{AAD}	23	5	5	8	18	11
Δ_{bias}	23	3	-2	-7	-10	-3
Δ_{stdev}	12	4	4	5	9	9
Δ_{max}	32	11	10	15	30	32
Data Points	2	4	5	6	7	24

Table S19. Results for PC-SAFT predicted p-w_{N2} isotherms for ULSD + N_2 mixtures assuming the multi-component diesel can be represented as a single, pseudo-component with GC-calculated pure component parameters calculated with the B-GC and S-GC methods and with $k_{ij} = 0$.

(a) ULSD + N_2 (B-GC Method, k_{ij} = - 0.0238)

T/K	300	373	423	473	530	Overall
Δ_{AAD}	9	5	6	4	7	6
Δ_{bias}	-3	5	5	1	-4	0
Δ_{stdev}	4	5	6	3	4	4
Δ_{max}	12	10	14	7	11	14
Data Points	2	3	4	5	6	20

(b) ULSD + N_2 (S-GC Method, k_{ij} = 0.1413)

T/K	300	373	423	473	530	Overall
Δ_{AAD}	1	12	10	8	16	11
Δ_{bias}	-1	-2	3	7	9	5
Δ_{stdev}	-	11	7	8	8	8
Δ_{max}	1	22	18	21	27	27
Data Points	1	3	4	5	6	19

Table S20. Results for PC-SAFT predicted p-w_{N2} isotherms for ULSD + N_2 mixtures assuming the multi-component diesel can be represented as a single, pseudo-component with GC-calculated pure component parameters calculated with the B-GC and S-GC methods and with $k_{ij} = 0$.

(a) HAR + N_2 (B-GC Method, k_{ij} = - 0.0313)

T/K	300	373	423	473	530	Overall
Δ_{AAD}	15	12	12	7	11	11
Δ_{bias}	5	-8	-8	-3	3	-2
Δ_{stdev}	8	11	6	7	6	7
Δ_{max}	21	19	19	19	20	21
Data Points	2	2	3	5	5	17

(b) HAR+ N_2 (S-GC Method, k_{ij} = 0.1363)

T/K	300	373	423	473	530	Overall
Δ_{AAD}	8	20	12	21	18	17
Δ_{bias}	-8	-8	-10	1	-13	-7
Δ_{stdev}	-	11	19	19	18	16
Δ_{max}	8	28	34	43	49	49
Data Points	1	2	3	5	5	16

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