Reviews

Vapor−Liquid Critical Properties of Elements and Compounds. 6. Unsaturated Aliphatic Hydrocarbons

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This is part 6 of a series of contributions by the critical properties group of the IUPAC Commission I.2 on Thermodynamics, Subcommittee on Thermodynamic Data. It presents all known experimental data for the critical constants of unsaturated aliphatic hydrocarbons, which have been divided into five families: linear alk-1-enes (10 compounds, C_2 to C_{12}); other alkenes (8 compounds, C_4 to C_6); alkadienes (3 compounds, C_3 to C_6); terpenes (3 C_{10} compounds); and alkynes (3 compounds, C_2 to C_4). Recommendations are given together with uncertainties. Critical temperatures have been converted to ITS-90.

The unsaturated aliphatic hydrocarbons have been divided into five families: (a) linear alk-1-enes; (b) other alkenes; (c) alkadienes; (d) terpenes; (e) alkynes. More than one-third of the references are on ethene, commercially the most important organic chemical. Not surprisingly, the critical properties of ethene are known very accurately. On the other hand, the information on alkadienes and terpenes is very limited. The presentation and evaluation of the experimental data follow the guidelines of Ambrose *et al.* in parts 1 and 2 of this series [95-amb/you, 95-amb/tso]. The recommended values are given in Table 1, while all known data have been collected in Table 2. The references follow the format [year-first three letters of first author/first three letters of second author, and, where required, a sequence number].

Linear Alk-1-enes

Experimental critical constants are known for ten linear alk-1-enes. The work of Teja's group has extended our knowledge of the critical properties of linear alk-1-enes (α-olefins) to C_{12}, although no measurements have been made on C_{11}. The alk-1-enes are ethene (ethylene), propene (propylene), but-1-ene, pent-1-ene, hex-1-ene, hept-1-ene, oct-1-ene, non-1-ene, dec-1-ene, and dodec-1-ene. The first five linear alk-1-enes are stable at their critical point, but the heavier ones are progressively more unstable.

Ethene. Ethene (ethylene) is distinguished by having its critical properties measured first by J. D. van der Waals [1880-van]. Although the critical temperature reported by van der Waals, 9.2 °C, is essentially the same as the recommended value, 9.19 °C, it took nearly 100 years for that value to be universally accepted.

The disagreement on the critical temperature of ethene to some extent resulted from the work of Maass and collaborators in the 1930s. They postulated that liquid persists above the temperature at which the meniscus disappears. As late as 1948 [48-mas/maa] they were arguing that maximum opalescence for ethene occurs at 9.20 °C, while the critical temperature (defined from the inflection of the pV isotherm) is 9.90 °C. Today, as noted in part 1 of this series [95-amb/you], it is usual to prefer the view of Kudchadker *et al.* [68-kud/ala] that (a) the inflection point on the pV isotherm and the temperature of the disappearance of the meniscus are essentially identical, and (b) "it is preferable to determine the critical temperature visually".

Ethene also has the distinction that *two* IUPAC volumes [74-ang/arm, 88-jac/jah] have been devoted to its properties. Considerable work was done between 1974 and 1988, primarily in recognition of the commercial importance of ethene, but also owing to the fact that custody transfer takes place at conditions close to its critical point.

The most careful determination of the critical temperature of ethene was probably that of Moldover [74-mol], who used the meniscus disappearance method. For the critical density, Moldover filled four ampules to densities close to the literature value for ρ_c. He then measured the height at which the meniscus disappeared at T_c, and then by using an integrated form of the critical isotherm equation he found the density ($=\rho_c$) at which the meniscus would disappear exactly in the center of the ampule.

Moldover measured t_c = (9.194 ± 0.004) °C and ρ_c = (0.2146 ± 0.0006) g·cm^{-3}. In addition, he interpolated the then unpublished pVT data of Hastings and Levelt Sengers [77-has/lev, 80-has/lev] to determine the value of the critical pressure as p_c = (5.039 ± 0.010) MPa.

Hastings and Levelt Sengers later commented [80-has/lev] that their earlier results used by Moldover were found to be slightly in error.

The 1988 IUPAC volume [88-jac/jah] accepted Moldover's t_c and redetermined p_c and ρ_c from the pVT data of Douslin and Harrison [76-dou/har] and Hastings and Levelt Sengers [80-has/lev]. In addition, Jacobsen *et al.* [88-jac/jah] reported the values used in their critical-region equation and in the main equation of state. The latter values (see Table 2), which correspond to the inflection point on the critical pV isotherm, are outside of the error limits Jacobsen *et al.* set for the selected values. Our recommendations are consistent with these selected values, but are slightly rounded off and have larger uncertainties: T_c = (282.34 ±

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Table 1. Recommended Values of Critical Properties of Unsaturated Aliphatic Hydrocarbons

	molar mass M/g·mol^{-1} [a]	T_c/K[b]	(±)	p_c/MPa	(±)	ρ_c/g·cm^{-3}	(±)	V_c/cm^3·mol^{-1}	Z_c[c]
ethene	28.053 76	282.34	(0.02)	5.041	(0.004)	0.214	(0.002)	131.1	0.2815
propene	42.080 64	364.9	(0.4)	4.60	(0.03)	0.228	(0.005)	184.6	0.2798
but-1-ene	56.107 52	419.5	(0.5)	4.02	(0.05)	0.233	(0.012)	240.8	0.2775
pent-1-ene	70.134 4	464.8	(0.5)	3.56	(0.05)	0.235	(0.005)	298.4	0.275
hex-1-ene	84.161 28	504.0	(0.3)	3.21	(0.03)	0.237	(0.005)	355.1	0.272
hept-1-ene	98.188 16	537.3	(0.4)	2.92	(0.04)	0.24	(0.01)	409	0.267
oct-1-ene	112.215 04	567.0	(0.8)	2.68	(0.08)	0.24	(0.01)	468	0.266
non-1-ene	126.241 92	594.0	(1.0)			0.24	(0.01)	526	
dec-1-ene	140.268 8	617	(2)	2.22	(0.10)	0.24	(0.01)	584	0.253
dodec-1-ene	168.322 56	658	(2)	1.93	(0.20)				
(*Z*)-but-2-ene	56.107 52	435.5	(0.1)	4.21	(0.05)	0.240	(0.010)	233.8	0.272
(*E*)-but-2-ene	56.107 52	428.6	(0.1)	4.10	(0.02)	0.236	(0.005)	237.7	0.2735
2-methylpropene	56.107 52	417.9	(0.1)	4.000	(0.010)	0.235	(0.004)	238.8	0.2749
(*Z*)-pent-2-ene	70.134 4	475	(1)	3.69	(0.10)				
3-methylbut-1-ene	70.134 4	452.7	(0.3)	3.53	(0.03)	0.230	(0.010)	304.9	0.286
2-methylbut-2-ene	70.134 4	470	(1)	3.42	(0.10)				
cyclopentene	68.118 52	506.5	(0.5)	4.80	(0.05)	0.278	(0.010)	245	0.279
cyclohexene	82.145 4	560.4	(0.1)						
propadiene	40.064 76	394	(?)	5.25	(?)				
buta-1,3-diene	54.091 64	425	(1)	4.32	(0.10)	0.245	(0.010)	221	0.270
hexa-1,5-diene	82.145 4	508	(?)						
)-(*R*)-*p*-mentha-1,8-diene	136.237 04	653	(2)			0.29	(0.01)	470	
)-(1*S*)-pin-2-ene	136.237 04	644	(2)			0.30	(0.01)	454	
)-car-3-ene	136.237 04	658	(2)			0.28	(0.01)	487	
ethyne	26.037 88	308.3	(0.1)	6.138	(0.010)	0.232	(0.004)	112.2	0.2687
propyne	40.064 76	402.4	(0.2)	5.63	(0.02)	0.245	(0.005)	163.5	0.275
but-1-yne	54.091 64	440	(2)	4.60	(0.20)	0.26	(0.03)	208	0.262

Molar masses based on carbon = 12.011, hydrogen = 1.00794. [b] Temperatures are expressed in ITS-90. [c] $Z_c = p_c V_c / R T_c$ where R = 8.314 51 J·mol^{-1}·K^{-1}.

0.02) K, p_c = (5.041 ± 0.004) MPa, and ρ_c = (0.214 ± 0.002) cm^{-3}.

Propene. Next to ethene, propene (propylene) is the most important alkene in commerce. However, the number of investigations of the critical constants of propene is only a small fraction of the investigations on ethene. Furthermore, the critical constants of propene are subject to considerable uncertainty.

Angus *et al.*, in the IUPAC monograph on propene [80-ang/arm], examined six investigations [15-sei/bur, 21-maa/wri, 33-win/maa, 40-vau/gra, 41-lu/new, 49-mar/pre] and concluded that "It is not possible to have complete confidence in any of them". Angus *et al.* suggest that "T_c probably lies in the region (365.15 ± 0.5) K, p_c lies between (4.5 and 5) MPa, and ρ_c is not known". Because of these uncertainties, Angus *et al.* did not constrain their pVT equation to go through a certain critical point, but instead determined the following critical constants by satisfying the critical point criteria, $(\partial p/\partial \rho)_T = 0$ and $(\partial^2 p/\partial \rho^2)_T = 0$: 365.57 K, p_c = 4.6646 MPa, and ρ_c = 0.005 31 mol·cm^{-3} (0.223 g·cm^{-3}). Angus added that their value for T_c may be as much as 0.5 K too high, but still used their values "for the sake of thermodynamic consistency".

Brunner's [88-bru] careful visual observation, the pVT measurements of Ohgaki *et al.* [90-ohg/ume], and the very recent results of Wilson *et al.* [95-wil/wil] have confirmed Angus's supposition that their T_c recommendation [80-ang/arm] is too high. Ohgaki *et al.* made their measurements in the critical region ($0.996 < T_r < 1.004$; $0.6 < \rho_r < 1.4$) and converted them to critical properties with the power law for the vapor−liquid coexistence curve and the $p-\rho$ relation on the critical isotherm (see 96-amb/you for a correction to part 1 of this series). Considering all the experimental measurements that are listed in Table 2, we recommend the following critical constants for propene: T_c = (364.9 ± 0.4) K, p_c = (4.60 ± 0.03) MPa, and ρ_c = (0.228 ± 0.005) g·cm^{-3}. Although the uncertainties for T_c and p_c are larger than is desirable for a substance as important as propene, they are not large enough to include the 1980 IUPAC recommendations [80-ang/arm].

C_4 to C_6 Alk-1-enes. Apparently, no experimental investigation has been made on but-1-ene since 1950. Fortunately, this 45-year old investigation was made by Beattie's group [50-bea/mar], and thus the slightly rounded-off 1950 results can be recommended with confidence.

More recent data have been reported for pent-1-ene and some of the higher alk-1-enes by the groups of Ambrose (C_5 to C_8), Kay (C_5 and C_6), Ma (C_5 and C_6), and Teja (C_5 to C_{10}, C_{12}). In the case of pent-1-ene, there is excellent agreement in the T_c between Ambrose [60-amb/cox] and Teja [91-gud/ros]. In view of this agreement, it appears that a 1972 result from Kay's group [72-mou/kay] is 1 K too low, while a more recent Chinese investigation [91-ma/ma] is 1 K too high. For p_c we recommend the value of Wolfe *et al.* [83-wol/kay], while for ρ_c we averaged the results of Wolfe *et al.*, Ma *et al.* [91-ma/ma], and Gude *et al.* [91-gud/ros].

The modern measurements for hex-1-ene are in good agreement for T_c and p_c, but there is a significant difference for ρ_c, for which we recommend the average of the two available data [91-gud/ros, 91-ma/ma].

C_7 to C_{12} Alk-1-enes. The C_7 and higher alkenes are thermally unstable at their critical points, and this instability increases as the carbon number increases. It is for such unstable compounds that Ambrose had to use a rapid heater in order to reduce the effect of thermal decomposition, while Teja's group developed their special methods [see 95-amb/you and 95-amb/tso].

For hept-1-ene, there is very good agreement in the T_c results of Ambrose *et al.* [60-amb/cox] and Gude *et al.* [91-gud/ros], but we only have Gude's results for p_c and ρ_c. In the latter case, the value of Gude *et al.* was rounded down, in accord with our comment in Table 2 that their results for pent-1-ene and hex-1-ene may be too high; see also 93-ste/chi.

Table 2. Critical Properties from the Literature

year	values reported in nonstandard units	T_{90}/K	p/MPa	ρ/g·cm^{-3}	method[a]	authors
	ETHENE (Ethylene): molar mass = 28.053 76 g; CASRN = 74-85-1					
	$T_{68} - T_{48} = -0.004$ K; $T_{90} - T_{68} = -0.002$ K; $T_{90} - T_{48} = -0.006$ K at 282 K					
1880-van	9.2 °C, 58 atm	282.4	5.88		1	van der Waals
1882-sar	1.5 °C, 43.5 atm, 0.006 739 Amagat (volume)	274.5	4.41	0.186	8	Sarrau
1882-cai	13 °C	286			1	Cailletet
1884-dew	10.1 °C, 51.0 atm	283.2	5.17		1	Dewar
1886-cai/mat	12.5 °C	285.6		0.21	1, 7	Cailletet and Mathias
1895-ols	10 °C, 51.7 atm	283	5.24		1, 5	Olszewski
1897-vil	10 °C	283			1	Villard
12-car/arn	(9.50 ± 0.10) °C, (50.65 ± 0.10) atm	282.6	5.132		1a	Cardoso and Arni
21-maa/wri	(9.9 ± 0.2) °C	283.0			1	Maass and Wright
27-mat/cro, 29-mat/cro		(282.6)		0.21597	7	Mathias *et al.*
29-bri	9.0 °C, 52.0 atm	282.2	5.27		3	Britton
37-maa/ged	(9.50 ± 0.01) °C, (49.98 ± 0.01) atm	282.64	5.064	0.2374 ± 0.0067	1, 7	Maass and Geddes
38-mci/maa	9.50 °C	282.64			1	McIntosh and Maass
39-dac/mci, 39-mci/dac	(9.90 ± 0.01) °C, (50.50 ± 0.01) atm, 4.40 cm^3·g^{-1}	283.04	5.117	0.227	3	Dacey *et al.*, McIntosh *et al.*
40-nal/maa	(9.21 ± 0.015) °C	282.35			1a	Naldrett and Maass
48-kay	48.65 °F, 735.6 psi	282.39	5.072		1a	Kay
53-whi/mas	(9.26 ± 0.01) °C	282.40		0.2095 ± 0.0004	1a, 3	Whiteway and Mason
58-row/sut	9.4 °C, 49.84 atm	282.54	5.050		1a	Rowlinson *et al.*
64-shi/koh	9.54 °C, 50.53 atm	282.68	5.120		1	Shim and Kohn
71-vas	4.69 cm^3·g^{-1}	283.05	5.100	0.213	5, 7, 10	Vashchenko *et al.*
71-zer/kog	123.6 cm^3·mol^{-1}	282.318	4.96	0.2270	1a	Zernov *et al.*
74-ang/arm		282.65 ± 0.25	5.076 ± 0.02	0.218 ± 0.002	7, 10	Angus *et al.*
74-mol	(9.194 ± 0.004) °C	282.342		0.2146 ± 0.0006	1	Moldover
75-ben		282.648	5.075	0.215	8	Bender
76-dou/har	7.635 mol·dm^{-3}	282.35	5.0420	0.2142	3	Douslin and Harrison
76-tra/was	(9.30 ± 0.05) °C, (170.6 ± 1.0) Amagat	282.45		0.2152 ± 0.0013	3	Trappeniers *et al.*
77-goo	7.6 mol·dm^{-3}	282.35	5.04234	0.21	?	Goodwin
77-has/lev	(9.134 ± 0.043) °C	282.282	5.0336 ± 0.0048		3	Hastings and Levelt-Sengers
80-has/lev	(7.6340 ± 0.0009) mol·dm^{-3}	282.3432 ± 0.0017	5.0403 ± 0.0002	0.21416 ± 0.00003	3, 8	Hastings *et al.*
80-tho/zan	(9.22 ± 0.02) °C	282.368			1	Thomas and Zander
81-mcc/jac	7.6340 mol·dm^{-3}	282.341	5.0403	0.21416	8	McCarty and Jacobsen
81-syc/vas		282.348 ± 0.05	5.042 ± 0.002	0.2142 ± 0.0005	8	Sychev *et al.*
82-you	7.650 mol·dm^{-3}	282.341	5.0404	0.2146	8	Younglove
83-neh/hal	(7.635 ± 0.006) mol·dm^{-3}	282.348 ± 0.0105	5.04197 ± 0.0026	0.2142	8	Nehzat *et al.*
84-jah, 86-jah/jac	7.634 mol·dm^{-3}	282.343	5.0401	0.2142	8	Jahangiri, Jahangiri *et al.*
85-bru		282.33 ± 0.1	5.055 ± 0.005		2a	Brunner
86-van/jac	see notes				7	Van Poolen *et al.*
88-jac/jah	selected values	282.342 ± 0.004	5.0408 ± 0.0008	0.2142 ± 0.0004	8	Jacobsen *et al.*
	critical region EOS	282.343	5.0403	0.2142		
	main EOS (7.689 14 mol·dm^{-3})	282.318	5.0373	0.215709		
	recommended values	282.34 ± 0.02	5.041 ± 0.004	0.214 ± 0.002		

Sarrau [1882-sar] determined the critical constants by fitting Amagat's pVT data with the equation of state of Clausius. Since he assumed that the absolute temperature is 273.00 + t/°C, Sarrau's T_c should be 274.5 K (and t_c = 1.35 °C). Kobe and Lynn [53-kob/lyn] report the critical density incorrectly as 0.148 g·cm^{-3}.

Dewar [1884-dew] notes that the data for ethylene are not new, but Dewar's values could not be found in an 1884 or earlier paper.

Cailletet and Mathias [1886-cai/mat] report 12 °C and 0.22 g·cm^{-3} in the text, but the values interpolated from their graph are 12.5 °C and 0.21 g·cm^{-3}.

Mathias *et al.* [27-, 29-mat/cro] determined ρ_c at the t_c of Maass and Wright [21-maa/wri]; at 9.19 °C, ρ_c would have been 0.216 16 g·cm^{-3}.

Whiteway and Mason [53-whi/mas] used a vertical Pyrex bomb whose contents were vigorously stirred. However, no stirring was used in determining ρ_c.

F. Din (*Thermodynamic Functions of Gases*, Vol. 2, Butterworths, London, 1962) selected the T_c and ρ_c of Maass and Geddes [37-maa/ged] and the ρ_c of Mathias *et al.* [29-mat/cro] extrapolated to 9.5 °C.

Vashchenko *et al.* [71-vas] took T_c from the literature and determined p_c by graphically extrapolating their vapor pressure data; ρ_c by extrapolating the rectilinear diameter.

Angus *et al.* [74-ang/arm] took T_c and p_c from the literature (after some analysis) and determined ρ_c by extrapolating the rectilinear diameter of Mathias *et al.* [27-mat/cro].

Bender [75-ben] selected the critical data and constrained his equation of state to these values and the criticality conditions.

Table 2 (continued)

Sychev *et al*. [81-syc/vas] may have selected the critical constants from the literature and constrained their equation of state to these values and the criticality conditions. The p_c and ρ_c uncertainties are those on p 37; different values are given on p 113.
Nehzat *et al*. [83-neh/hal] correlated the data of Douslin and Harrison [76-dou/har] and adopted their values, but provided estimates for the uncertainties (on p 205).
The T_c and p_c values in 85-bru are also reported by E. Brunner (*J. Chem. Thermodyn.* **1987**, *19*, 823−835; **1988**, *20*, 1397−1409).
Van Poolen *et al*. [86-van/jac] determined ρ_c at the T_c of Jahangiri *et al*. [86-jah/jac] by straight rectilinear diameter (7.624 mol·dm^{-3} or 0.2139 g·cm^{-3}), by rectilinear diameter with a "hook" (7.639 or 0.2143), and by the critical liquid mole fraction (7.633 or 0.2141).

year	values reported in nonstandard units	T_{90}/K	p/MPa	ρ/g·cm^{-3}	method[a]	authors
	PROPENE (Propylene): molar mass 42.080 64 g; CASRN 115-07-1					
	$T_{68} - T_{48} = -0.002$ K; $T_{90} - T_{68} = -0.024$ K; $T_{90} - T_{48} = -0.026$ K at 365 K					
1882-nad	90 °C	363			1	Nadezhdin
1883-nad	91.6 °C	364.8			1	Nadezhdin
15-sei/bur	92.6 °C, 34 463 mmHg	365.8	4.5947		1	Seibert and Burrell
21-maa/wri	(92.1 ± 0.2) °C	365.2			1	Maass and Wright
33-win/maa	92	365		0.225	1, 7	Winkler and Maass
40-gil/sch	203 °F, 675 psi	368	4.654		1(?)	Gilliland and Scheeline
40-sou	91.9 °C, 44.6 atm, 179.7 cm^3·mol^{-1}	365.0	4.519	0.234	10	Souders
40-vau/gra	91.4 °C, 45.4 atm, 180 cm^3·mol^{-1}	364.6	4.600	0.233	1, 3, 7	Vaughan and Graves
41-lu/new	92.1 °C, 45.4 atm	365.2	4.600		1	Lu *et al*.
43-cra		(364.6)		0.2277	7	Cragoe
49-far/sag	197.0 °F, 668 psi, 0.06944 ft^3·lb^{-1}	364.8	4.606	0.2307	5, 7, 10	Farrington and Sage
49-mar/pre	(91.76 ± 0.015) °C, (45.61 ± 0.02) atm, (0.191 ± 1%) L·mol^{-1}	364.88	4.6214	0.220	3	Marchman *et al*.
55-kre	92.05 °C	365.17			1	Kreglewski
71-vas	4.34 cm^3·g^{-1}	365.03	4.64	0.230	5, 7, 10	Vashchenko *et al*.
75-ben		364.88	4.613	0.227	8	Bender
78-juz/sif		364.96	4.60	0.230	5, 8	Juza *et al*.
80-ang/arm	0.005 31 mol·cm^{-3}	365.55	4.6646	0.223	8	Angus *et al*.
88-bru		364.83 ± 0.2	4.600 ± 0.02		2a	Brunner
90-ohg/ume	0.005 55 mol·cm^{-3}	364.57	4.579	0.2335	3	Ohgaki *et al*.
95-wil/wil		365.16 ± 0.05	4.594 ± 0.005		2c	Wilson *et al*.
	recommended values	364.9 ± 0.4	4.60 ± 0.03	0.228 ± 0.005		

Nadehzdin reported the t_c as 90 °C in 1882-nad and 91.6 °C in 1883-nad, which is the average of two values reported earlier in the paper: 90.2 °C and 93 °C. The German summary [1883-nad-1] gives the value 97 °C, but no such value could be found in the original papers.
Winkler and Maass [33-win/maa] measured the saturated vapor and liquid densities up to 91.6 °C. Critical temperature and density were estimated from their Figure 1.
Souders [40-sou] is quoted in 40-vau/gra as having "critically chosen" the critical constants.
Cragoe [43-cra] determined ρ_c at the T_c (and p_c) of Vaughan and Graves [40-vau/gra].
Farrington and Sage [49-far/sag] determined their t_c from a review of available data and p_c by extrapolating their vapor pressure data from 190 °F to t_c.
Vashchenko *et al*. [71-vas] took T_c from the literature and determined p_c by graphically extrapolating their vapor pressure data and ρ_c by extrapolating the rectilinear diameter.
Bender [75-ben] selected the critical data and constrained his equation of state to these values and the criticality conditions.
Juza *et al*. [78-juz/sif] claim that their equation of state is valid from 180 K to 460 K, but also report a T_c and ρ_c; their vapor pressure equation was extrapolated from 360 K to T_c.
The T_c of Ohgaki *et al*. [90-ohg/ume] is presumed to be consistent with the IPTS-68 scale.
Wilson *et al*. [95-wil/wil] made their measurements with both a static and a flow method that employed a visual cell. "There was no statistical difference between the values measured" by the two methods.

year	values reported in nonstandard units	T_{90}/K	p/MPa	ρ_c/g·cm^{-3}	method[a]	authors
	BUT-1-ENE: molar mass 56.107 52 g; CASRN 106-98-9					
	$T_{68} - T_{48} = 0.0019$ K; $T_{90} - T_{68} = -0.035$ K; $T_{90} - T_{48} = -0.016$ K at 420 K					
28-cof/maa	144 °C	417			1	Coffin and Maass
43-cra	39.2 atm	(417)	3.97	0.2354	6, 7	Cragoe
46-old/sag	297 °F, 588 psi, 3.85 ft^3·lbmol^{-1}	420	4.05	0.2334	3	Olds *et al*.
50-bea/mar	(146.4 ± 0.3) °C, (39.7 ± 0.3) atm, (4.15 ± 5%) mol·L^{-1}	419.53	4.023	0.233	3	Beattie and Marple
	recommended values	419.5 ± 0.5	4.02 ± 0.05	0.233 ± 0.012		

Cragoe [43-cra] determined the ρ_c at the T_c of Coffin and Maass [28-cof/maa]. He also estimated p_c by extrapolating available vapor pressure data.
Beattie and Marple [50-bea/mar] reported the uncertainty in T_c as 0.3 °C on pp 1450 and 1452, but as 0.03 °C on p 1451 (caption for Figure 2).

Table 2 (continued)

year	values reported in nonstandard units	T_{90}/K	p/MPa	ρ/g·cm^{-3}	method[a]	authors
	PENT-1-ENE: molar mass 70.1344 g; CASRN 109-67-1					
	$T_{68} - T_{48} = 0.039$ K; $T_{90} - T_{68} = -0.040$ K; $T_{90} - T_{48} = -0.001$ K at 465 K					
1883-paw	201.0 °C	474.2			1	Pawlewski
1893-alt	208.0 °C	481.2			1	Altschul
1895-pic	201.2 °C	474.4			1	Pictet
1895-pic/alt	201.0 °C	474.2			1	Pictet and Altschul
03-ves	202.6 °C, 40.4 atm	475.8	4.094		1	Vespignani
48-day/nic	201 °C, 30 700 mmHg	474	4.093		3, 5	Day *et al.*
60-amb/cox	(191.59 ± 0.03) °C	464.74			1	Ambrose *et al.*
72-mou/kay	515.03 psi	463.73	3.551		1a	Mousa *et al.*
83-wol/kay		465.1	3.55	0.233	1a, 7	Wolfe *et al.*
91-gud/ros		464.7 ± 0.3		0.239 ± 0.005	1c	Gude *et al.*
91-ma/ma	(192.59 ± 0.17) °C	465.70	3.592 ± 0.026	0.2334 ± 0.0009	1, 7	Ma *et al.*
	recommended values	464.8 ± 0.5	3.56 ± 0.05	0.235 ± 0.005		

Pawlewski [1883-paw] measured the t_c of "amylene"; however, he also reported the t_b = 38 °C, which is close to that of 2-methylbut-2-ene (38.55 °C).

Altschul [1893-alt] also measured the t_c of "amylene"; see previous note.

Pictet *et al.* [1895-pic, 1895-pic/alt] reported the t_c of "pental", which may be 2-methyl-2-butene. However, the compound is given as pent-1-ene in 23-lan/boe and 53-kob/lyn.

Vespignani [03-ves] identified his sample as "amilene-n"; however, he reported a t_b (37.5 °C at 761.5 mmHg) that is close to that of 2-methylbut-2-ene or of the two but-2-enes, (36 to 37) °C. On the other hand, the reported density (0.6360 g·cm^{-3} at 21/4 °C) is closer to that of pent-1-ene.

Day *et al.* [48-day/nic] mention that t_c is in the neighborhood of 201 °C and add that the critical region will be considered in a future paper, but such a paper could not be located. They measured the vapor pressure up to 200 °C. We calculated p_c with their equation for the (170 to 200) °C range. (The 10^{-3} in the last term of the equation should be 10^{-9}.)

et al. [91-ma/ma] report 192.59 °C in Tables 4 and 8, but 192.42 °C in Table 9.

year	values reported in nonstandard units	T_{90}/K	p/MPa	ρ/g·cm^{-3}	method[a]	authors
	HEX-1-ENE: molar mass 84.161 28 g; CASRN 592-41-6					
	$T_{68} - T_{48} = 0.055$ K; $T_{90} - T_{68} = -0.040$ K; $T_{90} - T_{48} = 0.015$ K at 504 K					
1893-alt	243.5 °C (?)	516.6			1	Altschul
60-amb/cox	(230.83 ± 0.03) °C	504.00			1	Ambrose *et al.*
74-kay/you		503.2 ± 0.1	3.212 ± 0.002		1a	Kay and Young
91-gud/ros		504.2	3.206 ± 0.02	0.242 ± 0.005	1c	Gude *et al.*
91-ma/ma	(230.57 ± 0.25) °C	503.68	3.200 ± 0.030	0.2320 ± 0.0009	1, 7	Ma *et al.*
94-gud/tej	gas furnace	504.2 ± 0.2		0.241 ± 0.005	1c	Gude and Teja
	platinum furnace	504.0 ± 0.3		0.242 ± 0.03		
	flow method	504.8 ± 0.8	3.206 ± 0.02			
	recommended values	504.0 ± 0.3	3.21 ± 0.03	0.237 ± 0.005		

Altschul [1893-alt] reported the t_c of "hexylene" with a question mark; he noted that the liquid became brown and the temperature started increasing.

Kay and Young [74-kay/you] reported the unpublished data of W. B. Kay and S. Pak, API "Critical Properties of Hydrocarbon Mixtures", Progress Report No. 5, 1970−71.

Gude *et al.* [91-gud/ros] measured (504.0 ± 0.3) K with the sealed ampule method and (504.8 ± 0.8) K with the flow method.

In 94-gud/tej, "gas furnace" and "platinum furnace" refer to the static (sealed ampule) method.

year	values reported in nonstandard units	T_{90}/K	p/MPa	ρ/g·cm^{-3}	method[a]	authors
	HEPT-1-ENE: molar mass 98.188 16 g; CASRN 592-76-7					
	$T_{68} - T_{48} = 0.064$ K; $T_{90} - T_{68} = -0.040$ K; $T_{90} - T_{48} = 0.024$ K at 537 K					
60-amb/cox	(264.08 ± 0.05) °C	537.25			1c	Ambrose *et al.*
91-gud/ros		537.4	2.921 ± 0.02	0.244 ± 0.005	1c	Gude *et al.*
	recommended values	537.3 ± 0.4	2.92 ± 0.04	0.24 ± 0.01		

Ambrose *et al.* [60-amb/cox] measured T_c in a rapid heater; the rate of change of the apparent T_c was +0.5 K·h^{-1}.

Gude *et al.* [91-gud/ros] measured (537.4 ± 0.3) K with the sealed ampule method and (537.6 ± 0.8) K with the flow method; 537.4 K is their recommendation. Because their ρ_c values for pent-1-ene and hex-1-ene were higher than other literature values, their results for C_7+ (which are unstable at the critical point) have been adjusted to lower values with one less significant figure.

year	values reported in nonstandard units	T_{90}/K	p/MPa	ρ/g·cm^{-3}	method[a]	authors
	OCT-1-ENE: molar mass 112.215 04 g; CASRN 111-66-0					
1883-paw	298.6 °C	571.8			1	Pawlewski
1893-alt	304.8 °C	578.0			1	Altschul
60-amb/cox	(293.4 ± 0.1) °C	566.6			1c	Ambrose *et al.*
90-smi/neg		569.8 ± 1		0.270	1c	Smith *et al.*
91-gud/ros		567.4	2.675 ± 0.02	0.242 ± 0.005	1c	Gude *et al.*
	recommended values	567.0 ± 0.8	2.68 ± 0.08	0.24 ± 0.01		

Table 2 (continued)

Ambrose *et al*. [60-amb/cox] measured T_c in a rapid heater; the rate of change of the apparent T_c was +3.0 K·h^{-1}.
Smith *et al*. [90-smi/neg] noted that their T_c was the first point observed, and therefore the accuracy would not be better than ±1 K. Their ρ_c is clearly an approximate value.
Gude *et al*. [91-gud/ros] measured (567.3 ± 0.3) K with the sealed ampule method and (567.8 ± 0.8) K with the flow method; 567.4 K is their recommendation. See also note for Gude *et al*. under hept-1-ene.

year	values reported in nonstandard units	T_{90}/K	p/MPa	ρ/g·cm^{-3}	method[a]	authors
	NON-1-ENE: molar mass 126.241 92 g; CASRN 124-11-8					
91-gud/ros		593.7 ± 0.3		0.241 ± 0.005	1c	Gude *et al*.
	recommended values	594.0 ± 1.0		0.24 ± 0.01		

Gude *et al*. [91-gud/ros] measured T_c only with the sealed ampule method. See also note for Gude *et al*. under hept-1-ene.

year	values reported in nonstandard units	T_{90}/K	p/MPa	ρ/g·cm^{-3}	method[a]	authors
	DEC-1-ENE: molar mass 140.2688 g; CASRN 872-05-9					
91-gud/ros		616.4	2.218 ± 0.02	0.240 ± 0.005	1c	Gude *et al*.
94-gud/tej	gas furnace	617.2 ± 1.1		0.239 ± 0.005	1c	Gude and Teja
	platinum furnace	616.9 ± 1.2		0.240 ± 0.003		
	flow method	616.0 ± 0.8	2.218 ± 0.02			
	recommended values	617 ± 2	2.22 ± 0.10	0.24 ± 0.01		

Gude *et al*. [91-gud/ros] measured (616.9 ± 1.2) K with the sealed ampule method and (616.0 ± 0.8) K with the flow method; 616.4 K is their recommendation. See also note for Gude *et al*. under hept-1-ene.
In 94-gud/tej, "gas furnace" and "platinum furnace" refer to the static (sealed ampule) method.

year	values reported in nonstandard units	T_{90}/K	p/MPa	ρ/g·cm^{-3}	method[a]	authors
	DODEC-1-ENE: molar mass 168.322 56 g; CASRN 112-41-4					
91-gud/ros		657.6 ± 0.9	1.930 ± 0.02		1c	Gude *et al*.
	recommended values	658 ± 2	1.93 ± 0.20			

Gude *et al*. [91-gud/ros] measured T_c only with the flow method.

year	values reported in nonstandard units	T_{90}/K	p/MPa	ρ/g·cm^{-3}	method[a]	authors
	(*Z*)-BUT-2-ENE (*cis*-But-2-ene): molar mass 56.107 52 g; CASRN 590-18-1					
	$T_{68} - T_{48} = 0.026$ K; $T_{90} - T_{68} = -0.037$ K; $T_{90} - T_{48} = -0.011$ K at 436 K					
28-cof/maa	155 °C	428			1	Coffin and Maass
43-cra	(320 °F), 41.5 atm	(433)	4.205	0.2398	6, 7, 9	Cragoe
60-amb/cox	(162.40 ± 0.02) °C	435.54			1	Ambrose *et al*.
	recommended values	435.5 ± 0.1	4.21 ± 0.05	0.240 ± 0.010		

Coffin and Maass [28-cof/maa] prepared a β-butylene sample, a mixture of (*Z*)- and (*E*)-but-2-enes; see text.
Cragoe [43-cra] estimated t_c and then determined p_c by extrapolating literature vapor-pressure data.

year	values reported in nonstandard units	T_{90}/K	p/MPa	ρ/g·cm^{-3}	method[a]	authors
	(*E*)-BUT-2-ENE (*trans*-But-2-ene): molar mass 56.107 52 g; CASRN 624-64-6					
	$T_{68} - T_{48} = 0.024$ K; $T_{90} - T_{68} = -0.037$ K; $T_{90} - T_{48} = -0.013$ K at 429 K					
28-cof/maa	155 °C	428			1	Coffin and Maass
43-cra	(311 °F), 40.5 atm	(428)	4.104	0.2359	6, 7, 9	Cragoe
60-amb/cox	(155.46 ± 0.02) °C	428.60			1	Ambrose *et al*.
	recommended values	428.6 ± 0.1	4.10 ± 0.02	0.236 ± 0.005		

Coffin and Maass [28-cof/maa] prepared a β-butylene sample, a mixture of (*Z*)- and (*E*)-but-2-enes; see text.
Cragoe [43-cra] estimated t_c and then determined p_c by extrapolating literature vapor pressure data.

year	values reported in nonstandard units	T_{90}/K	p/MPa	ρ/g·cm^{-3}	method[a]	authors
	2-METHYLPROPENE (Isobutylene): molar mass 56.107 52 g; CASRN 115-11-7					
	$T_{68} - T_{48} = 0.018$ K; $T_{90} - T_{68} = -0.034$ K; $T_{90} - T_{48} = -0.016$ K at 418 K					
1883-nad, -nad-1	150.7 °C	423.8			1	Nadezhdin, Nadejdine (sic)
27-cof/maa, 28-cof/maa	143.5 °C	416.6			1	Coffin and Maass
39-sch/gil	(300 ± 2) °F, (583 ± 4) psi	422	4.02		1	Scheeline and Gilliland
42-bea/ing	(144.73 ± 0.05) °C, (39.48 ± 0.05) atm	417.86	4.000	0.234 ± 0.002	3	Beattie *et al*.
42-ben	(144.6 ± 0.6) °C, (39.5 ± 0.2) atm	417.8	4.002		1	Benedict
43-cra		(417.86)		0.2352	7	Cragoe
	recommended values	417.9 ± 0.1	4.000 ± 0.010	0.235 ± 0.004		

Nadezhdin's [1883-nad] value for the t_c was the average of 11 observations; his name was spelled Nadejdine in 1883-nad-1.
Coffin and Maass first reported the t_c in [27-cof/maa], although the details of their measurements were given in their later paper [28-cof/maa].

Table 2 (continued)

year	values reported in nonstandard units	T_{90}/K	p/MPa	ρ/g·cm^{-3}	method[a]	authors
	(*Z*)-PENT-2-ENE (*cis*-Pent-2-ene): molar mass 70.1344 g; CASRN 627-20-3					
71-len/reb	395.2 °F, 535 psi	474.9	3.69		1a	Lenoir *et al.*
	recommended values	475 ± 1	3.69 ± 0.10			

The critical constants were measured in a Kay-type phase boundary apparatus. The sample was 96.3 mol % (*Z*)- and 3.5 mol % (*E*)-pent-2-ene; 0.2 mol % of other impurities.

year	values reported in nonstandard units	T_{90}/K	p/MPa	ρ/g·cm^{-3}	method[a]	authors
	3-METHYLBUT-1-ENE: molar mass 70.1344 g; CASRN 563-45-1					
	$T_{68} - T_{48} = 0.034$ K; $T_{90} - T_{68} = -0.039$ K; $T_{90} - T_{48} = -0.005$ K at 452.5 K					
91-ma/ma	(179.54 ± 0.14) °C	452.65	3.527 ± 0.025	0.2300 ± 0.0009	1, 7	Ma *et al.*
	recommended values	452.7 ± 0.3	3.53 ± 0.03	0.230 ± 0.010		
	2-METHYLBUT-2-ENE (β-Isoamylene): molar mass 70.1344 g; CASRN 513-35-9					
1882-zhu, 1883-nad	191.6 °C, 33.93 atm	464.8	3.438		1	Zhuk, Nadezhdin
51-kiy/suz	197.2 °C, 33.7 atm	470.4	3.415		1	Kiyama *et al.*
	recommended values	470 ± 1	3.42 ± 0.10			

Zhuk [1882-zhu], Nadezhdin's student, reported the data for "amylene"; Nadezhdin [1883-nad] repeated the t_c, but identified the compound as "isoamylene", which may be a mixture of 3-methylbut-1-ene and 2-methylbut-2-ene. The reported t_c is remarkably close to the modern value for pent-1-ene.

Kiyama *et al.* [51-kiy/suz] also determined via their pVT measurements that t_c = (195 to 200) °C. Their sample had a t_b = 37.8 °C, 0.8 deg below the accepted value. It should also be noted that the data reported by Kiyama's group [51-kiy/ike] for ethyne are very poor: 3 K and 0.2 MPa higher than the recommended values for T_c and p_c.

year	values reported in nonstandard units	T_{90}/K	p/MPa	ρ/g·cm^{-3}	method[a]	authors
	CYCLOPENTENE: molar mass 68.118 52 g; CASRN 142-29-0					
	$T_{68} - T_{48} = 0.055$ K; $T_{90} - T_{68} = -0.040$ K; $T_{90} - T_{48} = 0.015$ K at 506 K					
57-amb/gra	(232.9 ± 0.05) °C	506.06			1	Ambrose and Grant
90-tej/ans		507.6 ± 0.3		0.278 ± 0.005	1c	Teja and Anselme
90-tej/ros		507.0 ± 0.6	4.802 ± 0.02		1c	Teja and Rosenthal
	recommended values	506.5 ± 0.5	4.80 ± 0.05	0.278 ± 0.010		
	CYCLOHEXENE: molar mass 82.1454 g; CASRN 110-83-8					
	$T_{68} - T_{48} = 0.070$ K; $T_{90} - T_{68} = -0.039$ K; $T_{90} - T_{48} = 0.031$ K at 560 K					
60-amb/cox	(287.27 ± 0.02) °C	560.45			1	Ambrose *et al.*
62-che/mcc		560.43			1	Cheng *et al.*
	recommended values	560.4 ± 0.1				

Ambrose *et al.* [60-amb/cox] reported that the compound was only slightly unstable (+0.03 K·h^{-1}).

year	values reported in nonstandard units	T_{90}/K	p/MPa	ρ/g·cm^{-3}	method[a]	authors
	PROPADIENE (Allene): molar mass 40.064 76 g; CASRN 463-49-0					
05-les/cha	120.75 °C	393.9			1	Lespieau and Chavanne
47-stu	51.8 atm	(393.9)	5.25		6	Stull
	recommended values	394 ± ?	5.25 ± ?			

Stull [47-stu] apparently extrapolated vapor pressure data to the t_c of [05-les/cha].

year	values reported in nonstandard units	T_{90}/K	p/MPa	ρ/g·cm^{-3}	method[a]	authors
	BUTA-1,3-DIENE: molar mass 54.091 64 g; CASRN 106-99-0					
42-gar/ada	325.7 °F	436.3			?	Garner *et al.*
43-cra		(425.2)		0.2430	7	Cragoe
45-sco/mey	152 °C, 32 420 mmHg	425.2	4.322	0.245	1, 5, 7	Scott *et al.*
	recommended values	425 ± 1	4.32 ± 0.10	0.245 ± 0.010		

Garner *et al.* [42-gar/ada] gave "private communication" for the source of their t_c.

Cragoe [43-cra] calculated ρ_c at the then unpublished T_c (and p_c) of Scott *et al.* [45-sco/mey].

Scott *et al.* [45-sco/mey] state that "the true critical pressure may be slightly lower" than the value they give.

year	values reported in nonstandard units	T_{90}/K	p/MPa	ρ/g·cm^{-3}	method[a]	authors
	HEXA-1,5-DIENE (Diallylene): molar mass 82.1454 g; CASRN 592-42-7					
1883-paw	234.4 °C	507.6			1	Pawlewski
	recommended values	508 ± ?				

Table 2 (Continued)

year	values reported in nonstandard units	T_{90}/K	p/MPa	ρ/g·cm^{-3}	method[a]	authors
	TERPENES: molar mass 136.237 04 g					
	(+)-(*R*)-*p*-MENTHA-1,8-DIENE [(+)-D-LIMONENE]; CASRN 5989-27-5					
90-smi/neg		653 ± 2		0.29 ± 0.01	1c	Smith *et al.*
	(−)-(1*S*)-PIN-2-ENE: CASRN 7785-26-4					
90-smi/neg		644 ± 2		0.30 ± 0.01	1c	Smith *et al.*
	(+)-CAR-3-ENE: CASRN 13466-78-9					
90-smi/neg		658 ± 2		0.28 ± 0.01	1c	Smith *et al.*

Landolt-Börnstein [23-lan/boe] report for turpentine oil a calculated (?) t_c attributed to Guldberg (*Beibl. Ann. Phys.* **1883**, *7*, 350): 376.0 °C or 649 K, which is in close agreement with the results of Smith *et al.* [90-smi/neg].
The sample of (+)-car-3-ene used by 90-smi/neg was only 92.4% pure. (The menthadiene and pinene samples were 99% pure.)

year	values reported in nonstandard units	T_{90}/K	p/MPa	ρ/g·cm^{-3}	method[a]	authors
	ETHYNE (Acetylene): molar mass 26.037 88 g; CASRN 74-86-2					
	$T_{68} - T_{48} = -0.010$ K; $T_{90} - T_{68} = -0.009$ K; $T_{90} - T_{48} = -0.019$ K at 308 K					
1879-ans	37.05 °C, 68 atm	310.2	6.9	0.36	1, 5, 7	Ansdell
1897-kue	35.25 °C, 61.02 atm	308.4	6.183		1a	Kuenen
07-mci	36.5 °C, 61.6 atm	309.6	6.24	0.314	1	McIntosh
09-mat	37.05 °C	310.2		0.2306	1, 7	Mathias
10-car/bau	35.5 °C, 61.6 atm	308.6	6.24		1	Cardoso and Baume
12-car/bau	(35.50 ± 0.10) °C, (61.65 ± 0.10) atm	308.63	6.247		1	Cardoso and Baume
51-kiy/ike	38.5 °C, 64.0 atm	311.6	6.48		3 (?)	Kiyama *et al.*
56-amb	(35.18 ± 0.05) °C	308.31			1	Ambrose
64-amb/tow	35.18 °C, (60.58 ± 01) atm	308.31	6.138		1	Ambrose and Townsend
66-kho	0.1126 L·mol^{-1}	308.64		0.231	1	Khodeeva
72-gol/kho	35.20 °C	308.32			1	Goloborod'ko and Khodeeva
	recommended values	308.3 ± 0.1	6.138 ± 0.010	0.232 ± 0.004		

Ansdell [1879-ans] measured the vapor pressure up to 36.9 °C (67.96 atm) and the liquid density up to 35.8 °C (0.364 g·cm^{-3}); his values were rounded off.
Kobe and Lynn [53-kob/lyn] cite two papers by J. Dewar. The first one (*Proc. R. Soc. London* **1880**, *30*, 538−546) is a qualitative investigation of mixtures of CO_2, including the CO_2/acetylene binary. In the second paper (*Philos. Mag.* **1884**, *18*, 210−216) Dewar reported the T_c and p_c values of Ansdell (who investigated ethyne at Dewar's suggestion), but also proposed, by analogy with CO_2, that $\rho_c = 0.32$ g·cm^{-3} rather than 0.36 g·cm^{-3}.
McIntosh [07-mci] determined the ρ_c by weighing samples at the critical point; he concluded that he could "hardly recommend" his method.
Cardoso and Baume [10-car/bau] report four individual measurements of p_c as 61.6 atm, but give the average as 61.5 atm.
F. Din (*Thermodynamic Functions of Gases*; Butterworths: London, 1962; Vol. 2) selected the T_c and p_c of 12-car/bau and the ρ_c of 09-mat; the t_c from the latter reference was reported incorrectly as 35.4 °C.
Khodeeva [66-kho] may only have measured T_c, because the method involves filling up a tube of known volume with a liquid "such that its density has the critical value, and the critical temperature ... is measured".

year	values reported in nonstandard units	T_{90}/K	p/MPa	ρ/g·cm^{-3}	method[a]	authors
	PROPYNE (Methylacetylene, allylene): molar mass 40.064 76 g; CASRN 74-99-7					
	$T_{68} - T_{48} = 0.012$ K; $T_{90} - T_{68} = -0.032$ K; $T_{90} - T_{48} = -0.020$ K at 402 K					
05-les/cha	129.5 °C	402.6			1	Lespieau and Chavanne
21-maa/wri	127.9 °C	401.0			1	Maass and Wright
47-stu	52.8 atm	(401.0)	5.35		6	Stull
62-voh/kan	(129.23 ± 0.02) °C, (55.54 ± 0.02) atm	402.36	5.628	0.2449 ± 0.002	3	Vohra *et al.*
	recommended values	402.4 ± 0.2	5.63 ± 0.02	0.245 ± 0.005		

Stull [47-stu] extrapolated vapor pressure data to the t_c of [21-maa/wri].

year	values reported in nonstandard units	T_{90}/K	p/MPa	ρ/g·cm^{-3}	method[a]	authors
	BUT-1-YNE (Ethylacetylene): molar mass 54.091 64 g; CASRN 107-00-6					
95-ste		440 ± 1	4.586 ± 0.10	0.257 ± 0.03	3	Steele
	recommended values	440 ± 2	4.60 ± 0.20	0.26 ± 0.03		

[a] For methods see Table 3.

What was said for hept-1-ene can be repeated for oct-1-ene, except that the agreement in T_c is not as good. The results of Smith *et al.* [90-smi/neg] are of lower quality: 3 K and 0.03 g·cm^{-3} higher than the recommended values for T_c and ρ_c.

Finally, only Gude's measurements [91-gud/ros] are available for C_9, C_{10}, and C_{12} alk-1-enes. No p_c was measured for C_9, and no ρ_c for C_{12}.

Other Alkenes

Only the three butenes, cyclopentene, and cyclohexene have been investigated by several groups, but the critical pressure of the two but-2-enes has not yet been determined experimentally. The following compounds have experimental data: (*Z*)-but-2-ene (*cis*-but-2-ene), (*E*)-but-2-ene (*trans*-but-2-ene), 2-methylpropene (isobutylene), (*Z*)-pent-2-ene (*cis*-pent-2-ene), 3-methylbut-1-ene, 2-methylbut-2-

Table 3. Key to Methods of Critical Point Determination (Reprinted with Permission from 95-amb/you; Copyright 1995 American Chemical Society)

1.	visual—in glass tube
2.	visual—in cell with windows
3.	nonvisual—pVT measurement
4.	other nonvisual measurement
5.	critical pressure measurement combined with vapor pressure measurement up to the critical point
6.	critical pressure by extrapolation of vapor pressure curve
7.	orthobaric density measurements
8.	equation of state, thermodynamic study
9.	calculation from another physical property
10.	literature survey
(a)	with stirring
(b)	instrumental detection of critical point
(c)	special feature of apparatus

ene, cyclopentene, and cyclohexene.

(Z)- and (E)-But-2-ene. Coffin and Maass [28-cof/maa] measured the critical temperature of a "β-butylene" sample that they synthesized. They reported a melting point (−127 °C) that is between that of (*Z*)-(−138.9 °C) and (*E*)-but-2-ene (−105.57 °C), but a normal boiling point (1.0 °C) that is much closer to that of (*E*)-but-2-ene (0.88 °C) than that of (*Z*)-but-2-ene (3.72 °C).

Gershinowitz and Wilson [38-ger/wil] showed that (*Z*)-but-2-ene is the high-boiling isomer, whereas previously it had been thought to be the low-boiling isomer. Cragoe [43-cra] identified the two isomers properly, but Kobe and Lynn switched Cragoe's results. However, Kobe and Lynn only made recommendations for the (*Z*−*E*) mixture, for which they averaged Cragoe's estimates for T_c, p_c, and ρ_c (which Cragoe calculated by applying the law of rectilinear diameters).

The critical temperature of both isomers was subsequently measured by Ambrose *et al.* [60-amb/cox], but for the critical pressure we still have only Cragoe's 1943 estimates. In view of the closer agreement of Cragoe's value with Ambrose's T_c for (*E*)-but-2-ene, the p_c of this isomer has been assumed to have a lower uncertainty than that for (*Z*)-but-2-ene.

2-Methylpropene. No experimental measurements have been reported on the industrially important 2-methylpropene since 1942, but fortunately, as in the case of but-1-ene, those measurements were made by Beattie's group [42-bea/ing] and were confirmed by Benedict [42-ben].

C_5 Alkenes. Relatively recent experimental data are available for (*Z*)-pent-2-ene [71-len/reb], 3-methylbut-1-ene [91-ma/ma], and 2-methylbut-2-ene [51-kiy/suz]. Additional data on C_5 alkenes were measured from 1882 to 1903, but the identity of the C_5 alkenes is unclear. Zhuk [1882-zhu] called his sample amylene, but the 1883 paper by Nadezhdin, Zhuk's professor, and the German summary [1883-nad, -nad-1] called it isoamylene, which is either 3-methylbut-1-ene or 2-methylbut-2-ene. Pawlewski [1883-paw] and Altschul [1893-alt] referred to amylene, which can be pent-1-ene (α-*n*-amylene) or pent-2-ene (*β*-*n*-amylene), although Pawlewski's t_b (38 °C) is close to that of 2-methylbut-2-ene (38.55 °C). There may even be a question about Pictet's [1895-pic, -pic/alt] and Vespignani's [03-ves] samples; see notes in Table 2.

The identity of the samples used in the 1882 to 1903 investigations is of interest only because more recent reviews do not agree on their identification. For example, Kobe and Lynn [53-kob/lyn] give the results of Pawlewski, Altschul, Pictet and Altschul, and Vespignani under pent-1-ene; Vespignani's values are also given for pent-2-ene; and Nadejdine's (sic) results are listed under "isoamylene".

Also questionable is the "diamylene" sample used by Altschul [1893-alt], which most likely was a mixture of isomers, and for this reason the critical temperature reported by Altschul, (341 to 342) °C, is not included in Tables 1 and 2. In a 1943 compilation of physical constants [43-dos], Altschul's diamylene was assumed to be 2,6,6-trimethylhept-2-ene.

Finally, the values reported by Ambrose [80-amb] for (*E*)-pent-2-ene, 2-methylbut-1-ene, and 2-methylbut-2-ene are estimates.

Cyclopentene and Cyclohexene. Teja's group [90-tej/ans; 90-tej/ros] measured a significantly higher critical temperature for cyclopentene than Ambrose and Grant [57-amb/gra]. On the other hand, for cyclohexene, which is slightly unstable at its critical point, the T_c of Ambrose and Grant has been confirmed by Cheng *et al.* [62-che/mcc].

Alkadienes

Experimental data have been reported for propadiene, buta-1,3-diene, and hexa-1,5-diene. All three are unstable at their critical points, and only for the commercially very important buta-1,3-diene, which polymerizes rapidly above 385 K, do we have a relatively recent determination [45-sco/mey]. Although there may be considerable uncertainty in the critical temperature of propadiene [05-les/cha] and hexa-1,5-diene [1883-paw], it should be noted that the critical temperature reported by Lespieau and Chavanne [05-les/cha] for propyne agrees with the modern value.

Terpenes

Smith *et al.* [90-smi/neg] measured the critical temperature and density of (+)-(*R*)-*p*-mentha-1,8-diene [(+)-D-limonene], (−)-(1*S*)-pin-2-ene, and (+)-car-3-ene. The results may be considerably uncertain, because the terpenes are unstable at their critical point. It has already been noted that the results for oct-1-ene reported in the same paper were of low quality.

Alkynes (Acetylenes)

Experimental data could be found in the open literature only for ethyne (acetylene) and propyne (methylacetylene). In addition, unpublished data are available for but-1-yne (ethylacetylene). Calculated values for the critical temperature of higher alkynes have been presented by Morehouse and Maas [31-mor/maa, 34-mor/maa], but it will be shown later that these values are rough estimates.

Ethyne. Kobe and Lynn [53-kob/lyn] reviewed the measurements of McIntosh [07-mci], Mathias [09-mat], Cardoso and Baume [12-car/bau], and Kiyama *et al.* [51-kiy/ike]. They also presented the data of Ansdell [1879-ans] and Kuenen [1897-kue] "for historical reasons". However, Kuenen's results, especially for T_c, are in excellent agreement with the recommended values.

The recommended values for T_c and p_c are based on the measurements of Ambrose [56-amb, 64-amb/tow]. In the case of T_c, there is excellent agreement with both more recent [72-gol/kho] and very old measurements [1897-kue]. Even the p_c reported in the latter reference is only 0.7% higher than the recommended value.

Mathias [09-mat] measured the saturated liquid and vapor densities up to 32.93 °C and determined the mean of the densities as a function of t: $\rho/\mathrm{g\cdot cm^{-3}} = 0.25431 - 0.00064t/°C$. At the recommended t_c (35.16 °C), this equation gives $\rho_c = 0.2318\ \mathrm{g\cdot cm^{-3}}$, which is the value we recommend (rounded off).

Propyne. The T_c's in the oldest [05-les/cha] and most recent sources [62-voh/kan] agree very well, while the extrapolated value for p_c reported by Stull [47-stu] is 5% lower than the only other available value [62-voh/kan].

Table 4. Critical Temperature of Alkynes[a]

	calcd t_c/°C		exptl	
	34-mor/maa	this work	t_c/°C	ΔT/K
ethyne	39.7	39.3	35.2	4.1
propyne	121.6	141.6	129.2	12.4
but-1-yne	190.5	189.1	167	22
but-2-yne	215.5	215.5		
pent-1-yne	220.3	220.0		

[a] The value for ethyne was calculated with $k = 2.02$, as given in 34-mor/maa; with the value $k = 2.09$ given in 31-mor/maa, we calculate 35.5 °C, although even that reference gives 39.7 °C.

But-1-yne. Unpublished measurements on but-1-yne have been made by Steele's group [95-ste]. A 95%-pure sample was purchased from Scott Specialty Gases and was purified by GLC to 99.15%. The vapor pressure was measured with a dead-weight gauge from 308 K to 443 K (no decomposition was observed until 433 K), and the critical temperature was inferred to be (440 ± 1) K by the corner in the curve when pressure was plotted against temperature. Because this is an insensitive method, the uncertainty in T_c may be closer to 2 K. Correspondingly, the uncertainty in the reported p_c, 4.586 MPa, may be 0.2 MPa. Plotting pressure against the density, which was also measured, led to the estimate $\rho_c = 0.257$ g·cm^{-3}, with an uncertainty of 10%.

Calculations with the Ramsay−Shields Equation

In addition to the experimental measurements on ethyne and propyne, several reviews (e.g., ref 53-kob/lyn) report critical temperature values for C_4 to C_5 alkynes that have been determined with the Ramsay−Shields equation. This is at best an approximation, because it is based on surface tension measurements and the relation between surface tension, σ, and critical temperature that was proposed by Ramsay and Shields [1893-ram/shi]:

$$\sigma V^{2/3} = k(t_c - t - 6) \qquad (1)$$

In eq 1, V is the molar liquid volume and k is a constant that is equal to 2.12 for "normal" substances. As noted in ref 53-kob/lyn, k is not a true constant and eq 1 is of limited accuracy.

Morehouse and Maass [31-mor/maa, 34-mor/maa] measured the surface tension of propyne, but-1-yne, but-2-yne (dimethylacetylene), and pent-1-yne and converted them (and the surface tension of ethyne) to critical temperatures with eq 1. We recalculated the values after noting that Kobe and Lynn [53-kob/lyn] had questioned the result for propyne. Both sets of calculations, the experimental values, and the differences (from our calculations) are given in Table 4.

The comparison given in Table 4 clearly raises questions about the results of Morehouse and Maass with eq 1. The difference between calculated and experimental values apparently increases with increasing carbon number (the calculated value for propyne agrees with the result given in ref 53-kob/lyn), and therefore the uncertainty in the calculated values for but-2-yne and pent-1-yne may exceed 20 K. For this reason, we have not included any T_c values calculated with eq 1 in Table 2.

Overview of Recommendations (Table 1)

Table 1 summarizes the recommended critical values given in Table 2. It also includes, where appropriate, the critical volume, V_c, and the critical compressibility factor, $Z_c = p_c V_c / RT_c$ ($R = 8.314\,51$ J·mol^{-1}·K^{-1}). A superficial comparison of the recommendations for linear alk-1-enes with those for normal alkanes from ref 95-amb/tso suggests that the ratio of the T_c's reduces very close to unity by C_{12}, but the p_c ratio does not decrease with increasing carbon number. At the same carbon number, it appears that the p_c of linear alk-1-enes is 6% larger than that of normal alkanes. Steele and Chirico [93-ste/chi], in their review of the properties of C_5 + alkenes, have pointed out the similar trends in the ρ_c of linear alk-1-enes and normal alkanes.

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Registry No. Supplied by the Author: Ethene, 74-85-1; propene, 115-07-1; but-1-ene, 106-98-9; pent-1-ene, 109-67-1; hex-1-ene, 592-41-6; hept-1-ene, 592-76-7; oct-1-ene, 111-66-0; non-1-ene, 124-11-8; dec-1-ene, 872-05-09; dodec-1-ene, 112-41-4; (*Z*)-but-2-ene, 590-18-1; (*E*)-but-2-ene, 624-64-6; 2-methylpropene, 115-11-7; (*Z*)-pent-2-ene, 627-20-3; 3-methylbut-1-ene, 563-45-1; 2-methylbut-2-ene, 513-35-9; cyclopentene, 142-29-0; cyclohexene, 110-83-8; propadiene, 463-49-0; buta-1,3-diene, 106-99-0; hexa-1,5-diene, 592-42-7; (+)-(*R*)-*p*-mentha-1,8-diene [(+)-D-limonene], 5989-27-5; (−)-(1*S*)-pin-2-ene, 7785-26-4; (+)-car-3-ene, 13466-78-9; ethyne, 74-86-2; propyne, 74-99-7; but-1-yne, 107-00-6.

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