



# The predictions of the critical point parameters for Al, Cu and W found from the correspondence between the critical point and unit compressibility line (Zeno line) positions

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## ABSTRACT

We make some predictions with respect to the critical parameters of Al, Cu and W (critical parameters of which lie in the phase domain still inaccessible for experiment) on the basis of the correspondence between the critical point and unit compressibility line (Zeno line) positions in the temperature–density plane using two new similarity relations. These ones rely on the fact that the Zeno line must be tangential to the extension of the liquid branch of the binodal at zero temperatures and correctly describe its low-temperature part. We show also that the compressibility factor at the critical point can not exceed 0.32 for the Lennard-Jones like systems.

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For more than a century, researchers have recognized that liquid–gas systems exhibit several regular and scalable volumetric features, like the principle of corresponding states or the law of rectilinear diameter [1,2]. Presently there is considerable experimental evidence confirming one more feature besides the two described above. This feature characterizes the states where the compressibility factor is unity (i.e.  $Z = P/\rho T = 1$ , where  $P$  is the pressure,  $\rho$  is the particle density,  $T$  is the temperature). These states form a straight line on the density–temperature plane.

This empirical regularity is referred to as the Zeno line. Corresponding data are presented for noble gases, hydrocarbons [3], water and other non-metallic materials [4,5]. We studied the experimental data for Hg and Cs and showed that Zeno line is a straight line for these metals as well [6,7] and recently showed [8] that this empirical regularity has a theoretical verification, at least for Lennard-Jones liquid. An important point is that the Zeno line tends asymptotically to the liquid branch of the binodal at low temperatures [7]. This statement was confirmed for model systems and for a wide group of real substances (including metals: Hg, Cs, and Cu). Here we will use this fact to find a relation connecting the Zeno line and the critical point.

The expression, which we will use to describe the liquid branch of binodal is very similar to one used earlier by Guggenheim [1], Reid et al. [9], Filippov [10] and others

$$\rho(T) = \rho_c + \alpha\tau + \beta\tau^{1/3} \quad (1)$$

where subscript 'c' refers to the values at the critical point,  $\tau = 1 - T/T_c$ . There are many ways [9–11] to determine the coefficients

in (1). Here we find the coefficients  $\alpha$  and  $\beta$  using the condition that this equation has to tend to the Zeno line equation at  $T \rightarrow 0$ . The equation for the Zeno line can be written as [4–8]

$$\rho/\rho_B + T/T_B = 1 \quad (2)$$

where  $T_B$  is the Boyle point temperature (second virial coefficient tends to zero when  $T \rightarrow T_B$ ),  $\rho_B$  is the density at the Zeno line at  $T = 0$ . It should be noted that the Boyle temperature is determined by the intermolecular potential and depends significantly on its form. For example, for the Lennard-Jones potential it equals to  $T_B = 3.418$ , whereas for the modified Buckingham potential (6-exp) it depends on the parameter 'a' (see [6,16]). This potential with  $a = 9.8$ –13 was used to describe the properties of Hg. Corresponding Boyle temperatures for 6-exp potential are  $T_B(a = 9.8) = 5.631$ ,  $T_B(a = 13.0) = 3.564$ . (The molecular units are used both for LJ and Buckingham systems.)

Eq. (1) transfers into Eq. (2) at  $T \rightarrow 0$ , if  $\alpha + \beta = \rho_B - \rho_c$  and  $\alpha + \beta/3 = T_c/T_B$ . Substituting these conditions into Eq. (1) we obtain

$$\rho(T) = \rho_c + \frac{1}{2} \left[ \rho_c - \rho_B + 3\rho_B \frac{T_c}{T_B} \right] \tau + \frac{3}{2} \left[ \rho_B - \rho_c - \rho_B \frac{T_c}{T_B} \right] \tau^{1/3} \quad (3)$$

Eq. (3) can also be rewritten as

$$\rho(T)/\rho_B = 1 - T/T_c + \rho_c T/T_c \rho_B - 3S_1 f(T/T_c)/2 \quad (4)$$

where  $f(x) = 1 - x - (1 - x)^{1/3}$ , and the parameter  $S_1$  is determined as

$$S_1 = 1 - T_c/T_B - \rho_c/\rho_B \quad (5)$$

Below we will show that this parameter  $S_1$  can be considered as the first similarity parameter. It allows us to find, for example  $T_B$ , if other three values are known. If  $T/T_c \rightarrow 1$  (critical point), then  $\rho \rightarrow \rho_c$ , and,

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if  $T \rightarrow 0$ , then  $\rho \rightarrow \rho_B$ . Besides, Eq. (4) transforms into Eq. (2) if  $T/T_c \ll 1$ .

First, we check the validity of Eq. (3) for the Lennard-Jones liquid. This model plays a peculiar role in our investigation because it quite adequately describes the thermodynamical properties of a wide group of real substances that satisfy the corresponding states law, and there is a wealth of calculated information on the phase coexistence curves, critical and Zeno line parameters for this model. It should be noted that this model is internally self-consistent and can serve as a test for our theoretical consideration.

The Boyle temperature (in molecular units) is well known and is equal to  $T_B = 3.418$ . The Zeno line and critical parameters according to the results of [12,13] are slightly different and equal to  $P_c = 0.127$ ,  $\rho_B = 1.14$ ,  $T_c = 1.314$ ,  $\rho_c = 0.314$  and  $\rho_B = 1.05$ ,  $T_c = 1.311$ ,  $\rho_c = 0.279$ , respectively. The comparison of the phase coexistence curves according to the data of [12,13] with our calculations according to Eqs. (3) and (4) is presented in Fig. 1. One can see that there is a good agreement.

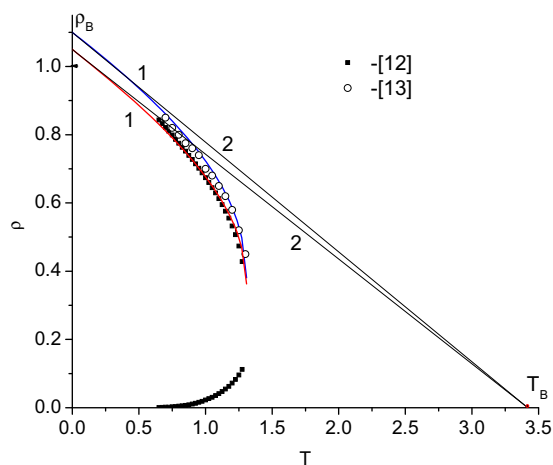
Along with the parameter  $S_1$  we consider the second parameter of similarity  $S_2$  defined as

$$S_2 = (\rho_c T_c - P_c) / \rho_B T_B \quad (6)$$

where  $P_c$  is the critical pressure. This parameter equals to the relation of the typical pressure at the critical point ( $\rho_c T_c - P_c$ , this difference approximately characterizes the potential part of the critical pressure) to the typical pressure along the Zeno line ( $\rho_B T_B$ ). We will show that this factor along with the factor  $S_1$  weakly depends on the substance or model and we assume that it equals to the constant determined from the most reliable data of Lennard-Jones liquid.

Table 1 lists the values for the critical, Zeno line parameters and the similarity factors  $S_1$ ,  $S_2$ : the Lennard-Jones and van der Waals systems; a wide group of real substances that obey the corresponding states law [3]; the metals Hg and Cs, their critical parameters measured in [14,15] and the approximate the Zeno line found in [6,7].

From Table 1 one can see that the values of the similarity factors  $S_1$ ,  $S_2$  depend weakly on the substances or models (at least for this group of materials). They are exact and consecutive for the Lennard-Jones and van der Waals models only. As for metals Hg and Cs, we do not know the exact form of the interaction potential and cannot find the exact values of the Zeno line point parameters. Our earlier work [6,7] used experimental data for that. Nevertheless, the values of the similarity factors presented above for Hg



**Fig. 1.** The phase coexistence curves and Zeno lines for the Lennard-Jones model. The symbols are numerical data on the binodal. Lines 1 and 2 correspond to our calculations (Eqs. (3) and (4)) of the liquid branch of binodal and the Zeno line, respectively.  $T_B$  and  $\rho_B$  are the Zeno line parameters.

**Table 1**

The critical, Zeno line parameters and similarity factors  $S_1$ ,  $S_2$  for different models and substances.

Model or substance	L-J <sup>a</sup>	VdW	Corresp. states law <sup>a</sup>	Hg	Cs
$T_c$	1.314 <sup>a</sup> 1.311 <sup>b</sup>	1	1.05	1751	1938
$\rho_c$	0.314 <sup>a</sup> 0.279 <sup>b</sup>	1	0.3	5.8	0.41
$T_B$	3.418	27/8	3	6500	4000
$\rho_B$	1.1 <sup>a</sup> 1.05 <sup>b</sup>	3	1	14	1.96
$S_1$	0.332 <sup>a</sup> 0.328 <sup>b</sup>	10/27	0.35	0.317	0.315
$S_2$	0.076	5/81	0.078	0.065	0.075

<sup>a</sup> Data Ref. [12].

<sup>b</sup> data Ref. [13].

<sup>a</sup> The values are given in dimensionless units, for Hg and Cs the density and temperature are expressed in g/cm<sup>3</sup> and K, respectively.

are close to van der Waals model. These factors for Cs and real substances that obey the corresponding states law are close to the prediction of the Lennard-Jones model. Below we will call this group of materials as the Lennard-Jones like systems. The mercury and van der Waals model form another group of substances.

This circumstance induces a suggestion that for the Lennard-Jones like systems these factors can be (in some approximation) set equal to the value following from the Lennard-Jones model as the one closest to the real substances, i.e.  $S_1 = 0.33$  and  $S_2 = 0.076$ . In this case Eqs. (1) and (2) can be rewritten in the form

$$\frac{\rho_c}{\rho_B} + \frac{T_c}{T_B} = 1 - S_1; \quad S_1 = 0.33 \quad (7)$$

$$S_2 = (\rho_c T_c - P_c) / \rho_B T_B = 0.076 \quad (8)$$

Eqs. (7) and (8) give rise to the universal expression for the critical pressure

$$P_c = \rho_c T_c \left( 1 - \frac{S_2}{\frac{\rho_c}{\rho_B} \left( 1 - 0.67 \frac{\rho_c}{\rho_B} \right)} \right) \quad (9)$$

One can see from Eq. (9) that the compressibility factor at the critical point  $P_c / \rho_c T_c$  as a function of  $\rho_c / \rho_B$  has a maximum when  $\rho_c / \rho_B = 0.335$ . In this case we find the greatest possible value of the compressibility factor at the critical point. This value is

$$P_c / \rho_c T_c = 0.32 \quad (10)$$

For  $\rho_c / \rho_B \neq 0.335$  the compressibility factor becomes lesser than 0.32 only. So according to our consideration the compressibility factor at the critical point can not be larger than 0.32 (at least, for the substances which are similar to the Lennard-Jones system). Eq. (7) can be treated as an equation describing the line of the critical points on the density-temperature plane. This line is parallel to the Zeno line and cuts off the segments on the density  $\rho_B(1 - S_1)$  and temperature  $T_B(1 - S_1)$  axes. We should note once more that the value  $S_1 = 0.33$  and  $S_2 = 0.076$  do not correspond to the van der Waals equation and experimental data for mercury. (For this equation the Boyle point parameters [8] are equal to  $T_B = 27/8$  and  $\rho_B = 3$ . The exact values of  $S_1$  and  $S_2$  are  $S_1 = 10/27 = 0.37$  and  $S_2 = 5/81 = 0.062$ , respectively.)

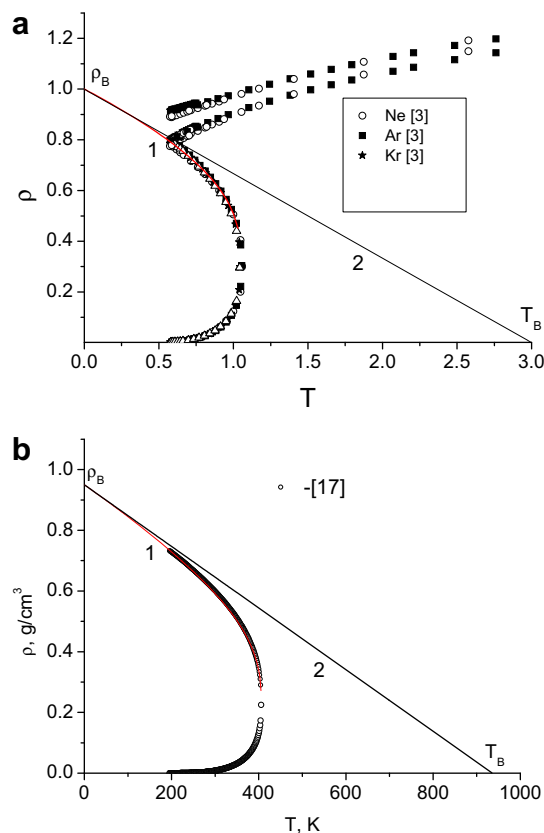
The binodal of a substance can be calculated according to Eqs. (3)–(5) if the critical and Zeno line parameters are known. The measurements and calculations of critical parameters have received much attention in literature [16]. The Zeno line parameters –  $T_B$  and  $\rho_B$  – can be found in several ways. First, the Zeno line can be calculated if the interaction potential corresponding to the substance of interest is known. This approach is applicable to the model system (see above), but this potential as a rule is unknown for the real substance. Another approach is based on the fact that the Zeno line is an asymptote relative to the binodal at  $T \rightarrow 0$  and has to tend to the tangent to an imaginary extension of this line

up to zero temperature domain (as we noted above). But the liquid branch of the binodal at  $T \rightarrow 0$  does not exist because the substance attains solid state, so all the measurements of binodals exist for finite temperatures  $T > 0$ . Nevertheless we can construct the liquid branch of the binodal using Eq. (5) and extend it till the zero temperature. Then a tangent to this line at  $T \rightarrow 0$  will be the Zeno line. Therefore we will require that the deviations of the binodal thus constructed from the experimental part of the binodal must be minimal. Consequently we can use the least-squares method in our calculations. It should be noted that Eq. (3) was checked for 107 different substances [11] (with  $\alpha$  and  $\beta$  different from ours). There were in [11] also additional considerations allowing for a description of the gas branch of the binodal. Here we will use only the fact that Eq. (3) can successfully describe the liquid branch for a wide range of substances.

First, let us consider the substances with known critical parameters, where measurements of the binodal exist. We set  $\rho_B$  in the range obtained by the least-squares method. The range of possible values  $\rho_B$  was found by the following way. We considered the straight lines, constructed by the least-squares method on the basis of experimental binodal points. The line drawn through two experimental points with the lowest temperatures gives the minimum possible value of  $\rho_B$ . The line drawn through all experimental points gives the maximum possible value of  $\rho_B$ . Then we find  $T_B$ , using criterion  $S_1$  – Eq. (5). After that, we calculate the dependence of density upon temperature along the binodal  $\rho(T)$  according to (3). Then we compare  $\rho(T)$  with the measured data  $\rho_{\text{exp}}(T)$ , i.e. calculate the value  $\delta$ :  $\delta(\rho_B) = \sum_i \left| 1 - \frac{\rho(T_i, \rho_B)}{\rho_{\text{exp}}(T_i, \rho_B)} \right|$ . Then we vary  $\rho_B$  in the range found above. If  $\delta < 10^{-3}$ , we can consider that the calculated  $\rho(T)$  is in agreement with the measured data  $\rho_{\text{exp}}(T)$ . Otherwise we change  $\rho_B$  and repeat the procedure until  $\delta < 10^{-3}$ . Then the parameter similarity  $S_2$  can be calculated according to Eq. (8).

The Zeno line parameters and parameter  $S_2$  obtained this way are presented in Table 2. We considered 17 gases, water, and 5 metals. The data for gases and Cs, Hg are presented in Table 2. (The critical parameters of gases were taken from the NIST database [17], the ones for Cs and Hg [14,15], the ones for water [5,18,19], the water Zeno line was obtained for the first time in [5]).

For the gases considered here, the procedure above was always successful. Below in Fig. 2 we present the results of the binodal calculation for the substances which satisfy the corresponding states law (a) and ammonia (b). One can see how for these substances the



**Fig. 2.** The phase coexistence curves and Zeno line for the substances, obeying to the corresponding states law – (a), ammonia – (b). The symbols are experimental data on the binodal. Curve 1 is the results of our calculations of the liquid branch of binodal according to Eq. (5). Straight line 2 is the Zeno line.  $T_B$  and  $\rho_B$  are the Zeno-line parameters.

similarity formulated here is satisfied and the condition of coincidence of the Zeno line and the tangent to the binodal at  $T \rightarrow 0$ .

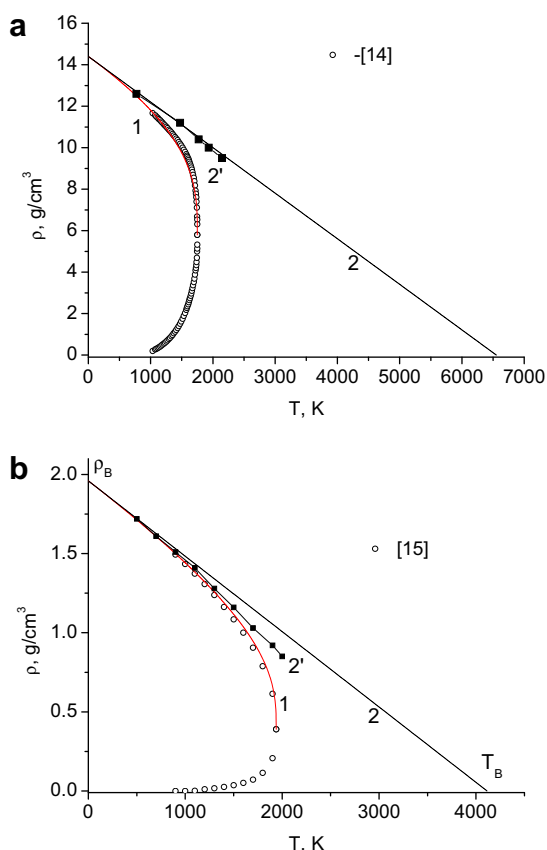
The situation for the metals is in many ways similar to that for the gases. The analysis of the measurements carried out in our previous work [6,7] showed the existence of the straight Zeno line for Hg and for Cs. But the construction of the binodal according to Eq. (3) was somewhat less exact in comparison with gases. In Fig. 3 we present the corresponding results for Cs. Here the best fit of the binodal corresponds to  $\delta = 1.1 \times 10^{-3}$  (this value is somewhat greater than for gases). We also draw the Zeno line calculated in our previous work [7] on the basis of the corresponding measurements. This line has a different slope than the one calculated here. The reason for this discrepancy, apparently, is the uncertainty in the ionic composition of Cs used in the calculation of [7]. Here we do not use any considerations about the Cs composition. So the new Zeno line should be more accurate.

For Hg, there is a discrepancy between calculated and measured binodals for  $T > 1000$  K (see Fig. 3). The best fit corresponds to the parameter  $\delta \sim 10^{-2}$ . So it seems that Eq. (5) has a lower accuracy for Hg in comparison with other substances. This is not a surprise since Hg has different properties in comparison with many other metals [14]. Consequently the binodal equation for Hg can also be different. But the Zeno line regularity also likely applies to Hg. Our previous results of the Zeno line calculation [7] are in good agreement with the present one. Thus the relation (7) can be also possibly true for Hg.

From Table 2 it follows also that for the majority of substances considered in Table 2 the values of the similarity parameter  $S_2$  are close to the corresponding value for the Lennard-Jones liquid. The

**Table 2**  
The critical and Zeno line parameters for different substances.

	$T_c$ (K)	$\rho_c$ (g/cm <sup>3</sup> )	$T_B$ (K)	$\rho_B$ (g/cm <sup>3</sup> )	$P_c$ (atm)	$S_2$
Ar	150.69	0.536	392.84	1.87	48.63	0.078
Ne	44.49	0.482	118.85	1.63	26.79	0.077
Kr	209.48	0.909	537.98	3.24	55.25	0.077
Xe	289.73	1.100	740.02	3.95	58.42	0.077
Ammonia	405.40	0.225	935.92	0.95	113.30	0.077
CO <sub>2</sub>	304.13	0.468	741.37	1.80	73.77	0.077
Ethane	305.33	0.207	779.35	0.74	48.72	0.079
Ethene	282.35	0.214	714.12	0.78	50.42	0.078
Fluorine	144.41	0.593	385.06	2.01	51.72	0.080
Hexane	507.82	0.233	1235.84	0.90	30.34	0.078
Methane	190.56	0.163	498.06	0.57	45.99	0.078
N <sub>2</sub>	126.19	0.313	327.62	1.10	33.96	0.078
O <sub>2</sub>	154.58	0.436	401.14	1.53	50.43	0.078
Propene	365.57	0.223	894.42	0.86	46.65	0.075
R13	302.00	0.583	763.31	2.13	38.79	0.078
R22	369.30	0.524	907.89	1.99	49.9	0.078
R32	351.26	0.424	823.92	1.74	57.82	0.079
Cs	1924	0.39	4114.47	1.96	94	0.075
Hg	1751	5.8	6552.60	14.40	1650	0.065
Water	647.3	0.32	1268.0	1.2	221.2	0.105

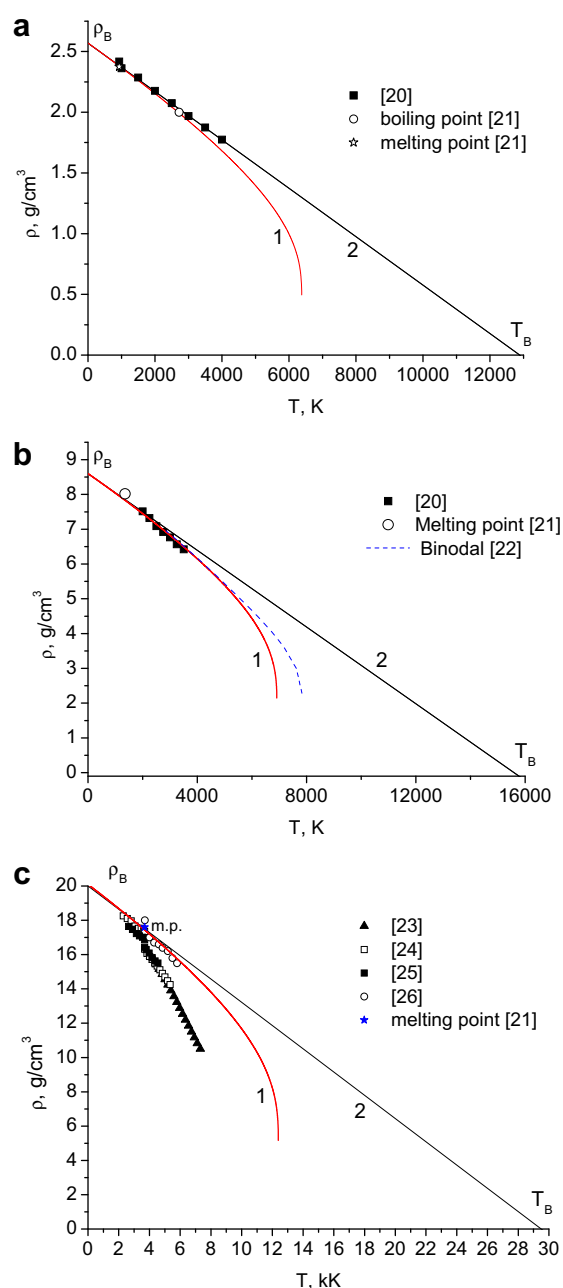


**Fig. 3.** The phase coexistence curves and Zeno line for metals: Hg (a) and Cs (b). The symbols are experimental data on the binodal. Line 1 is the results of our calculations of the liquid branch of binodal according to Eq. (3). Straight line 2 is the Zeno line of the present work, 2'-the previous one presented in [6,7].  $T_B$  and  $\rho_B$  are the Zeno line parameters.

exception is Hg and water. We have called attention to the peculiarities of Hg and they are developing under the calculation of  $S_2$  also. The value of  $S_2$  for Hg most likely corresponds to one following from van der Waals model. It has been noted in [5] that the Zeno line for water is a straight line in some approximation only. So it is not surprise that the similarity parameter  $S_2$  for water differs from other substances.

It is a challenge to use the method suggested above when we know the low-temperature branch of the binodal only and have no information concerning the critical and Zeno line parameters. This is current the state of knowledge for many metals, which have the critical parameters in the phase diagram domain still inaccessible for experiment. Let us consider, for example, metals Al, Cu and W. The low-temperature branches of binodals found on the basis of the experimental data for these substances are presented in Fig. 4a–c. The tangent to the low-temperature part of the binodal allows us to find the Zeno line parameters (as it is shown in Fig. 4). After that we again can suppose that full binodal can be described by Eq. (3).

The Zeno line parameters are unknown as well as the critical ones. We have to find the Zeno line parameters independently, using the least-squares method. We again draw the straight lines through 2, 3, ...,  $N$  points. Now we will consider the points, where these lines cross the axes, as Boyle parameters, but depending on  $N$ - $\rho_B(N)$  and  $T_B(N)$ . Now for given Zeno line parameters we set some value of  $\rho_c$ . The range of  $\rho_c$  was chosen from 0.5 g/cm<sup>3</sup> to 20 g/cm<sup>3</sup>. Then, we can calculate  $T_c$  with help of Eq. (7). After that we can calculate  $\delta$  like in previous section. Farther we obtain the



**Fig. 4.** The phase coexistence curves, Zeno line and critical parameters for metals: Al (a), Cu (b) and W (c). The symbols are experimental data on the binodal. Line 1 is the results of our calculations of the liquid branch of binodal according to Eq. (3). Straight line 2 is the Zeno line.

necessary value of  $\delta$  by the variation of  $\rho_c$  and  $N$ . It should be noted that the experimental data for Al, Cu and W are really do not belong to the binodals, they belong to the part of isobar curves. It is followed from thermodynamical consideration [30] that the isobar curves for liquid state go very close to the binodals. Therefore the measured binodals [20–26] practically coincide with the corresponding binodals. But it is also possible that the experimental points can lie at the Zeno line, as it takes place for Al. To take into account possible difference we somewhat increase the value of the criterion  $\delta$ , i.e.  $\delta < 0.01$ . After that we can calculate the critical pressure using Eq. (8).

The results of our calculations of the critical and Zeno line parameters are collected in Table 3. Besides, in this table, one can find the values of the critical parameters as obtained by other



**Table 3**

The critical and Zeno line parameters for metal liquids Al, Cu and W.

Metal	$T_c$ (K)	$\rho_c$ (g/cm <sup>3</sup> )	Method	Ref.	$T_B$ (K)	$\rho_B$ (g/cm <sup>3</sup> )	$P_c$ (atm)
Al	6378	0.45	This work	–	12888	2.57	1074
	8860	0.28	Scaling	[27]			4680
		0.64	Extrapolation	[28]			4470
Cu	7093	1.95	This work	–	15593	8.6	450
	7620	1.4	Scaling	[27]			
	8390	2.4	Extrapolation	[28]			7460
W	12387	4.92	This work	–	29131	20.1	7448
	12500	4.52		[29]			11000
	14000	4.71		[23]			5000

authors. We also present the corresponding Fig. 4a–c with the binodals and the Zeno lines for these substances. It should be noted that there is substantial scatter in the experimental data for W. Among them we choose data from [23,26], since they give the most reasonable values of the critical parameters. For Cu we also present the binodal obtained by means semi-empirical equation of state [22]. Our binodal seems to be more exact, because the data [22] are located higher than the experimental data.

Our findings are consistent with earlier known results. Moreover, they correspond to the theoretical and experimental information on the phase coexistence curves, critical and Boyle point parameters. This suggests that these findings are well justified.

Finally we note that the assumption of the constancy of similarly factors ( $S_1$ ,  $S_2$ ) was checked for a wide group of real substances for which the critical parameters are known. We developed a self-consistent method of determination of the Zeno line parameters providing the description of the liquid branch of the coexistence curve. For the substances with unknown critical parameters, this similarity makes it possible to find the critical density, temperature and pressure. To apply it to a particular substance, it is enough to know (for example, from experiment) the low-temperature part of the liquid binodal. We construct the procedure to find the position of such a Zeno line that provides the best agreement between the calculated binodal and the experimental one. Thus we have calculated the critical parameters for several metals (Al, Cu and W). Note that these parameters are still inaccessible for direct measurements. Finally, we have compared our present results with previously known ones and with the data of other researchers.

We have shown also that the idea of construction of a new similarity based on the correspondence of the critical and Zeno line parameters turns out to be highly fruitful. It is applied to a wider group of substances in comparison with those ones satisfying the corresponding states principle. The condition that the Zeno line must be tangential to the extension of the binodal liquid branch at  $T \rightarrow 0$  allows us to avoid difficulties connected with the uncertainty of intermolecular potentials. Due to this new similarity we can find the critical density, temperature and pressure using only the experimental data on the low-temperature part of the liquid

binodal branch. Consequently, it is possible to find the critical parameters of the substances that have the critical point in the phase diagram domain still inaccessible for experiment.

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