Int J Thermophys (2009) 30:1853–1863
DOI 10.1007/s10765-009-0671-6

The Critical Temperature of Aluminum

V. Morel · A. Bultel · B. G. Chéron

Received: 23 January 2009 / Accepted: 19 October 2009 / Published online: 4 November 2009

Abstract The critical temperature, T_c, of metals is a fundamental point when vaporization due to high energy exchanges occurs. Although aluminum is a metal often studied as a benchmark for theories, its critical temperature is not known with a high degree of accuracy. Its determination by experiment is difficult as a result of its high value. This paper reviews the existing data and proposes new ones resulting from recent measurements of particular physical properties and recent theoretical approaches. These new estimates lead to the recommended value of $T_c = (6700 \pm 800)$ K.

Keywords Aluminum · Cohesive enthalpy · Critical point · Critical temperature · Enthalpy of vaporization · Equation of state · Surface tension

1 Introduction

A relevant way for determining the in situ composition of a solid sample is to use laser-induced breakdown spectroscopy (LIBS). This involves the irradiation of the sample by a sufficiently powerful laser, leading to the formation of a plasma, followed by analysis by time-resolved emission spectroscopy. This technique has already been used in the case of tokamaks to probe the edge plasma [1], and developments are currently in progress to verify the carbon balance in fusion machines [2]. LIBS presents an important problem which prevents the technique from being universal: it requires a comparison of the experimental spectra with others performed earlier in controlled conditions with known test sample compositions. The only way to avoid relying on a representative databank is to model the complete situation. The model has to describe

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V. Morel · A. Bultel (✉) · B. G. Chéron
CORIA, UMR CNRS 6614, Université de Rouen, 76821 Saint-Etienne du Rouvray, France
e-mail: arnaud.bultel@coria.fr

the plasma formation, the kinetics mechanism, heating of the sample, its phase change, and the interaction with laser light [3].

In spite of the evaporation of the material and the diffusion of energy inside the sample, the surface temperature can reach the critical point. As soon as the critical temperature, T_c, is reached, the behavior of the surface changes significantly: since the liquid → gas transition does not occur anymore, a description of the edge of the sample by using the concept of a surface with a discontinuity in terms of specific mass is questionable. The critical point is therefore an upper limit of the traditional treatment of the sample in an LIBS situation.

We are currently developing a complete model of the behavior of a sample under nanosecond laser light irradiation based on our experience in modeling the thermal and chemical behavior of non-equilibrium plasmas [4,5]. Aluminum has been chosen as the sample material, since a lot of thermodynamic data have been determined concerning this element up to high-temperature levels. The critical temperature is conversely less well known. As a result, the objectives of the present paper are to review the existing estimates of T_c for aluminium; to propose other estimates resulting from recent data concerning the enthalpy of vaporization, the cohesive enthalpy, the surface tension, and the equation of state; and finally to recommend a value.

2 Review of the Existing Estimates of T_c for Aluminum

The critical temperature, T_c, for aluminum has never been experimentally determined. The different estimates (see Table 1) range between 5400 K and 9500 K and have been obtained using different methods. These methods can be divided into three groups. The first one (denoted as 1 in Table 1) concerns the equation of state. The second one (denoted as 2) is based on the use of rules assumed to be the same, more or less, for all liquid metals. The third one (denoted as 3) consists of extrapolating thermodynamic data experimentally obtained at lower temperature conditions. The related mean values are 8070 K for type 1, 7260 K for type 2, and 5950 K for type 3. The mean-type value is 7090 K whereas the mean value derived from all data of Table 1 is 7310 K.

The plot derived from Table 1 illustrates how T_c has evolved with time (see Fig. 1). This shows that the early estimates belong to three groups: group A (before 1980), group B (between 1990 and 2000), and group C (since 2006). The mean values are 7530 K, 7400 K, and 6550 K, respectively, and therefore, in the vicinity of the global mean value. With time, we can conclude that T_c corresponds to lower and lower values.

Recently, a series of experiments has been performed by Korobenko et al. [24] to determine the electrical resistivity of aluminum passing the critical point. This work is based on the observation of the explosion of a wire suddenly heated by a pulse of current. Underlying the difficulty of performing experiments in very high temperature conditions, this determination necessitates an equation of state: the SESAME library has been used [25]. As expected, the curves given in the paper emphasize an important increase of the resistivity passing the critical point (7000 K to 8000 K). Unfortunately, an equation of state being used, an evaluation of T_c cannot be derived from this experiment.

Table 1 Early estimates of the critical temperature T_c for aluminum (type of method and group: see Sect. 2)

Value (K)	Year	Reference	Method	Type of method	Group
7400	1964	Morris [6]	Corresponding states	1	A
7740	1967	Kopp [7]	Enthalpy of vaporization	2	
7151	1971	Young [8]	Equation of state	1	
6900	1973	Jones [9]	Model of pair potential	2	
8000	1975	Fortov [10]	Kopp-Lang rule	2	
6040	1976	Lang [11]	Surface tension	2	
5410	1976	Martynyuk [12]	Wire explosion	3	
9502	1977	Boissière [13]	Equation of state	1	
7543	1977	Hohenwarter [14]	Planck-Ridel vapour pressure	3	
8560	1977	Lang [15]	Enthalpy of vaporization	2	
8550	1977	Young [16]	Equation of state	1	
5654	1993	Blairs [17]	Sound velocity	2	B
8556	1993	Celliers [18]	Equation of state	1	
5726	1993	Celliers [18]	Isobaric measurements	3	
8860	1996	Likalter [19]	Equation of state	1	
5754	1998	Hess [20]	Guldberg rule	2	
8232	1998	Hess [20]	Kopp-Lang rule	2	
7499	1998	Hess [20]	Goldstein rule	2	
8944	1998	Hess [20]	Vapor pressure curves	2	
5115	2006	Blairs [21]	Surface tension	3	C
6557	2006	Blairs [21]	Guldberg rule	2	
7917	2008	Gordeev [22]	Equation of state	1	
6595	2008	Povarnitsyn [23]	Equation of state	1	

The same remark has to be made concerning the paper of Wu and Shin [26] which treats the determination of the absorption coefficient α_λ near the critical point. Using the Drude model to determine α_λ and starting from the results of Korobenko et al., the use of the equation of state for the determination of the temperature prevents an estimate of T_c.

Finally, another estimate of T_c is given in the work of Mazhukin et al. [27]. The value proposed (8000 K) belongs to the domain of high values, but its source is not cited and has not been taken into account in this review.

3 New Estimates of T_c for Aluminum

In the following, four methods based on recent measurements of physical properties and recent theoretical approaches are used to estimate T_c.

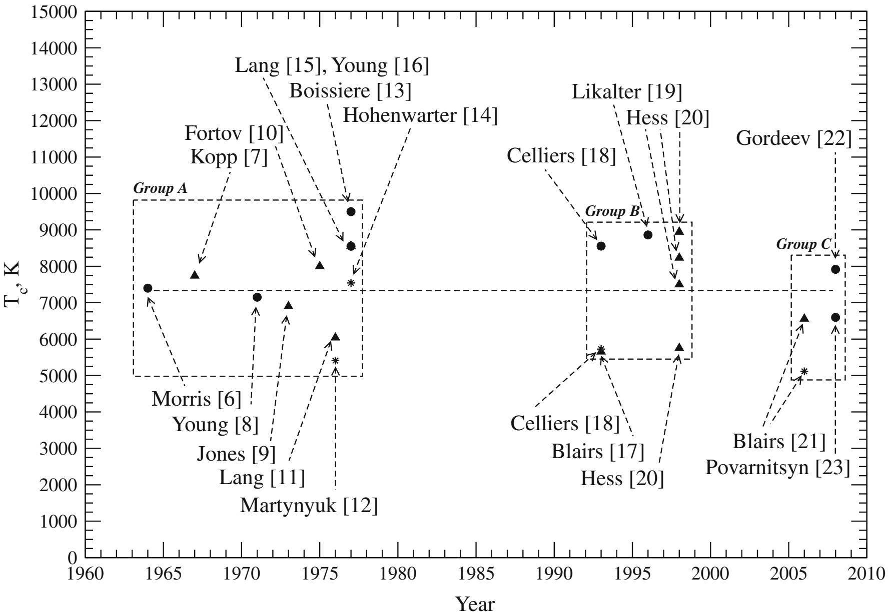

Fig. 1 Evolution in time of estimates of the critical temperature T_c of aluminum. The *dashed line* is located at the mean value 7310 K. *Circles*, *triangles*, and *stars* are related to the results obtained from methods 1 (mean value 8070 K), 2 (mean value 7260 K), and 3 (mean value 5950 K), respectively

3.1 Enthalpy of Vaporization

Considering the equilibrium vapor pressure of aluminum, Hess [20] discusses the validity of the interpolations allowing the matching of the experimental data up to the boiling point. The general equation of Dupré is adopted:

$$\ln(p_v/p_0) = A + B/T + C\ln(T/T_0) \tag{1}$$

with $p_0 = 1$ bar and $T_0 = 1$ K. The constants A, B, and C are listed in Table 2.

From this equality, we can derive the boiling point T_b, the enthalpy of ebullition Δh_b (i.e., the enthalpy of vaporization under atmospheric pressure) as well as its derivative with respect to temperature in the form,

$$\mathrm{d}\Delta h_b/\mathrm{d}T = CR_g \tag{2}$$

by using the Clausius–Clapeyron equation, R_g being the universal gas constant. These data are also given in Table 2.

T_c, T_b, Δh_b, and $\mathrm{d}\Delta h_b/\mathrm{d}T$ are linked together by the Meyra et al. [30] or Watson [31] equations, leading to the following form:

$$T_c = T_b - Z_c\Delta h_b/(\mathrm{d}\Delta h_b/\mathrm{d}T) \tag{3}$$

Table 2 Critical temperatures derived from the treatment of the enthalpy of vaporization.

Constants of Eq. 1	Reference	T_b (K)	Δh_b ($J\cdot mol^{-1}$)	$d\Delta h_b/dT$ ($J\cdot mol^{-1}\cdot K^{-1}$)	T_c (K) Eq. 3	T_c (K) Eq. 4
$A = 21.75$	[28]	2730	290.88	–8.314	12946	7626
$B = -37716$ K					16025	
$C = -1$						
$A = 13.62$	[29]	2740	310.34	0	∞	8899
$B = -37327$ K					∞	
$C = 0$						
$A = 24.37$	[29]	2800	293.84	–10.919	10658	7504
$B = -39020$ K					13026	
$C = -1.3133$						

The two values given for T_c correspond to the use of Eq. 3 with Z_c = 0.292 (Meyra et al. [30]) and Z_c = 0.38 (Watson [31])

with Z_c = 0.292 for Meyra et al. and Z_c = 0.38 for Watson.

On the other hand, Fish and Lielmezs [32] have worked on liquid metals and have extracted from experimental results a correlation which can be presented as follows, after some algebra:

$$T_c = T_b\{2\,[1 - T_b(d\Delta h_b/dT)/\Delta h_b]\}/\{1 - 2T_b(d\Delta h_b/dT)/\Delta h_b - n + m\} \quad (4)$$

where n = 0.20957 and m = – 0.17464.

We observe that T_b, Δh_b, and $d\Delta h_b/dT$ derived from Eqs. 1 and 2 and the Clausius–Clapeyron equation can be used to calculate T_c with Eqs. 3 and 4. Table 2 presents the values of T_c obtained by this way. It is clear that Eq. 3 overestimates the critical temperatures obtained in the past and displayed in Table 1. Therefore, Eq. 3 has to be rejected. Conversely, the Fish and Lielmezs equation is better: the values obtained (7500 K to 8900 K) belong to the range of high values and are in good accord with previous determinations. Alcock et al. [29] indicate that the most precise set in Table 2 is the third one; therefore, the value of 7504 K has to be particularly considered. The first set is less precise, but the value derived for T_c is very close to the previous one. The second set leads to a constant Δh_b near T_b: this behavior does not obey the usual law concerning the enthalpy of vaporization. This set has to be therefore also rejected as well as the related critical temperature.

Notice that Roman et al. [33] modified by Velasco et al. [34] give a correlation formula different from those of Meyra et al. and Watson, but it leads to the same form given by Eq. 3 for T_c: the resulting values are therefore the same as those of Table 2.

3.2 Cohesive Enthalpy

Recently, Kaptay [35,36] has given a unified model linking the cohesive enthalpy of a liquid $h_l^c(T)$ with the melting point T_m of alkali metals. This model also yields fairly good results for other metals. The cohesive enthalpy is defined as the energy existing in the liquid resulting from the mutual atomic attraction, which vanishes when the critical temperature is reached. As a result,

$$h_{\mathrm{l}}^{\mathrm{c}}(T) = h_{\mathrm{l}}(T) - h_{\mathrm{l}}(T_{\mathrm{c}}) \tag{5}$$

Kaptay gives its general evolution towards T_{m};

$$h_{\mathrm{l}}^{\mathrm{c}}(T_{\mathrm{m}}) = -q_1 R_{\mathrm{g}} T_{\mathrm{m}} - q_2 \left(R_{\mathrm{g}} T_{\mathrm{m}}\right)^2 \tag{6}$$

with $q_1 = 26.3 \times 10^{-4}$ and $q_2 = -2.62 \times 10^{-4} \mathrm{mol} \cdot \mathrm{J}^{-1}$.

Over the range $933\,\mathrm{K} \leq T \leq 4000\,\mathrm{K}$, Gathers [37] has derived the evolution of the specific enthalpy with temperature from experiments. It leads to an evolution of h_l according to

$$h_{\mathrm{l}}(T) = 1319.7 + 28.881T + 6.2284 \times 10^{-4} T^2 \tag{7}$$

The measurements of Gathers were performed under rather high-pressure conditions ($p = 0.3$ GPa). Nevertheless, the same measurements performed under lower pressure conditions ($p = 0.2$ GPa) have given similar results [38]: the pressure seems to have a negligible influence on h_{l}. As a result, Eq. 7 has been used directly for calculating $h_{\mathrm{l}}^{\mathrm{c}}$ (T_{m}) from Eqs. 5 and 6, the latter having been established under atmospheric pressure. Moreover, assuming that Eq. 7 is correct for a range of temperature extending up to T_{c}, the resulting value for the critical temperature is

$$T_{\mathrm{c}} = 6548\,\mathrm{K} \tag{8}$$

This value is remarkably close to that of Povarnitsyn [23].

3.3 Surface Tension

Molina et al. [39] have performed measurements of the surface tension of molten aluminum in particularly clean conditions of the surface, i.e., by minimizing the influence of adsorbed oxygen by working at high temperature. They have brought to light an increase of the surface tension when the influence of oxygen is reduced. The values obtained are $\gamma(1375\,\mathrm{K}) = 850\,\mathrm{mN} \cdot \mathrm{m}^{-1}$ with a slope $\mathrm{d}\gamma/\mathrm{d}T = -0.185\,\mathrm{mN} \cdot \mathrm{m}^{-1} \cdot \mathrm{K}^{-1}$. Assuming a constant slope over the range $[T_{\mathrm{m}}, 1375\,\mathrm{K}]$, they have deduced $\gamma\ (T_{\mathrm{m}}) = 940\,\mathrm{mN} \cdot \mathrm{m}^{-1}$.

The surface tension being zero at the critical temperature T_{c}, linking the surface tension and T_{c} can provide an estimate of the latter temperature. A lot of work has been devoted to the problem of the link between γ and T_{c}. Since the work of Binder [40], a general trend,

$$\gamma\ (T) = \gamma_0 \left(1 - T/T_{\mathrm{c}}\right)^n \tag{9}$$

with $n = 1.26$ is more or less accepted. In the case of hydrocarbons, the exponent is 1.244 [41] and for nitrogen, $n = 1.27$ [42]. For quantum fluids such as helium, $n = 1.289$ [43]. In the case of liquid metals, Eustathopoulos et al. [44] have proposed a model of $\mathrm{d}\gamma/\mathrm{d}T$: their results are very well correlated with the experimental ones obtained by Molina et al. Unfortunately, their model is based on assumptions which are valid only for temperatures around the melting point: consequently, their model cannot be applied up to T_c.

In more recent work, Digilov [45] has developed another model of the evolution with temperature of the surface tension γ leading to the following equation:

$$\gamma(T) = \gamma(T_\mathrm{m})\,[1 + 0.13(1 - T/T_\mathrm{m})]^{1.67} \tag{10}$$

Therefore,

$$(\mathrm{d}\gamma/\mathrm{d}T)\,T_\mathrm{m} = -0.217\gamma(T_\mathrm{m})/T_\mathrm{m} \tag{11}$$

Considering the reference value $\gamma(T_\mathrm{m}) = 940\,\mathrm{mN \cdot m^{-1}}$, Eq. 11 leads to $\mathrm{d}\gamma/\mathrm{d}T = -0.219\,\mathrm{mN \cdot m^{-1} \cdot K^{-1}}$ which is in good agreement with the work of Molina et al. By equating the surface tension to zero when $T = T_\mathrm{c}$, the critical temperature $T_\mathrm{c} = 8.69\ T_\mathrm{m} = 8114$ K can be deduced from Eq. 10. This value belongs to the high range of critical temperatures displayed in Fig. 1. Digilov adopts assumptions which are valid only in the vicinity of the melting point: these assumptions explain why the slope $\mathrm{d}\gamma/\mathrm{d}T$ is so well correlated with the experiment and cannot be used to derive T_c.

Assuming that $n = 1.26$ in Eq. 9, a critical temperature of

$$T_\mathrm{c} = 7164\ \mathrm{K} \tag{12}$$

is deduced from the results of Molina et al. obtained for $T = 1375$ K.

3.4 Equation of State

Different equations of state for aluminum have been established recently, and we have tested the more reliable among them for the purpose of evaluating the critical temperature. The equation of Chiew et al. [46], initially developed for the case of chain-like fluids and polymers, has to be mentioned. It is based on a first-order variational perturbation theory applied to molecules considered as chains of atoms interacting with each other according to a Lennard–Jones potential (called the PLJC method for the *perturbed Lennard–Jones chain* method). The resulting equation of state is

$$p/(nk_\mathrm{B}T) = f_1(\eta) + f_2(\eta) \tag{13}$$

where n is the number density of atoms, f_1 is the hard-sphere reference part, f_2 is the attractive perturbation part (proportional to n^3), and η is the hard-segment packing fraction depending on the number density n and the temperature T. The parts f_1 and f_2 depend on the usual two Lennard–Jones parameters σ and ε.

Table 3 Early estimates of the critical density ρ_c and pressure p_c for aluminum

ρ_c (kg · m^{-3})	p_c (GPa)	Reference	Year	Type of method
600	0.415	Morris [6]	1964	1
690	0.546	Young [8]	1971	1
1030	–	Jones [9]	1973	2
640	0.447	Fortov [10]	1975	2
895	0.938	Boissière [13]	1977	1
–	0.246	Hohenwarter [14]	1977	3
420	0.182	Young [16]	1977	1
685	–	Celliers [18]	1993	1
280	0.468	Likalter [19]	1996	1
430	0.473	Hess [20]	1998	2
660	0.467	Gordeev [22]	2008	1
698	0.399	Povarnitsyn [23]	2008	1

This approach gives results in good agreement with the experiment and has been consequently applied recently by Mousazadeh and Ghanadi Marageh [47] to the case of liquid metals. At critical conditions, the first and second derivatives of the pressure with respect to the density at constant temperature are equal to zero. For aluminum, we have therefore deduced

$$T_c = 5698\,\mathrm{K} \tag{14}$$

and $\rho_c = 566\,\mathrm{kg} \cdot \mathrm{m}^{-3}$ and $p_c = 0.393$ GPa. These values for critical density and pressure are in good agreement with those reported in Table 3.

Other approaches have been proposed combining a hard-sphere chain equation of state perturbed by a van der Waals attraction term and applied to some liquid metals, for instance, by Eslami [48]. In this case, the equation of state still presents the form of Eq. 13, but with a repulsive part proportional to n. The parts f_1 and f_2 also depend on two parameters: a reference radius and a reference energy denoted σ and ε, respectively, as in the case of the Lennard–Jones approach.

The metals Li, Na, K, Ru, Ce, Hg, Sn, Pb, and Bi have been treated by either the PLJC method or the perturbed van der Waals method. The comparison of σ and ε between both methods shows that the reference energy ε is approximately the same, whereas the reference radius σ is systematically lower when using the van der Waals method. The latter has not yet been tested on aluminum: we have consequently chosen to adopt the value of ε given in [47]. The value of σ has been evaluated by using the equation of state (Eq. 13) such that the specific mass of liquid aluminum at the melting point derived from the equation of state is equal to the reference value ($2377\,\mathrm{kg} \cdot \mathrm{m}^{-3}$) recommended by Assael et al. [49]. We have obtained $\sigma = 2.52 \times 10^{-10}$ m which is lower than the value proposed by [47] as for the other metals.

Table 4 New estimates of the critical temperature T_c for aluminum

T_c (K)	Method	Type of method
5698	Equation of state	1
6063	Equation of state	1
6548	Cohesive enthalpy	2
7164	Surface tension	3
7504	Enthalpy of vaporization	3
7626	Enthalpy of vaporization	3

Finally, the critical point estimated with the perturbed van der Waals method corresponds to the following conditions:

$$T_c = 6063\,\mathrm{K} \tag{15}$$

$\rho_c = 556\ \mathrm{kg \cdot m^{-3}}$ and $p_c = 0.373$ GPa. The coordinates of the critical point are also well correlated with the data given in Table 3.

4 Discussion

In Sect. 3, we have estimated T_c for aluminum with different methods. Table 4 summarizes these estimates ordered in increasing values. We see that passing successively from a method of type 1 to type 3 leads to an increase of T_c, contrary to the case of the early estimates. Therefore, we cannot conclude that one method is more reliable than another: all methods have consequently to be developed in order to refine the estimate of T_c. This conclusion is reinforced by the analysis of Table 3: except for the results of Jones [9], Boissière and Fiorese [13], Hohenwarter et al. [14], Young [16], Celliers and Ng [18], and Likalter [19], our estimates of ρ_c and p_c are in good agreement with the existing values derived not only from the use of method 1 (equation of state), but also from method 2.

The mean value obtained in this work is 6770 K, confirming the estimates of the recent group C and the decrease of the estimates of T_c with time. Our work is based globally on recent data: we can consequently combine our results with those of group C and deduce a value of 6680 K. For lack of other estimates, and waiting for a direct measurement of T_c, the recommended value is therefore $T_c = 6700\,\mathrm{K}$.

The uncertainty of the previous value has to be given. The standard deviation of the group of values obtained in Table 4 and in group C is 800 K. We assume that this standard deviation is a good estimate of the uncertainty even if these values are not related statistically. As a conclusion, we can state that $T_c = (6700 \pm 800)$ K.

5 Conclusion

The critical temperature of aluminum has not been measured directly so far, and it has been the subject of a lot of estimates in the past. In this work, we have reviewed the existing values and shown that the later the estimate, the smaller the value. By analyzing recent data concerning aluminum's enthalpy of vaporization, cohesive enthalpy, surface tension, and equation of state, we have deduced six new estimates of T_c confirming the global evolution with time put forward in our review. In addition, they are in good agreement with the values obtained for the last 3 years. Finally, the recommended critical temperature of aluminum, obtained as the average of our own results and the latter, is

$$T_c = (6700 \pm 800)\,\mathrm{K}$$

Acknowledgment The authors wish to thank the "Région Haute-Normandie" in France for its financial support (contract no 922/2000A/00112).

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