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Critical temperature of the lead–bismuth eutectic (LBE) alloy

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Abstract

Liquid metals such as Bi and Pb and Pb–Bi eutectic alloy are serious contenders for use as coolant in LMFBRs in lieu of sodium due to a number of attractive characteristics (high density, low moderation, low neutron absorption and activation, high boiling point and poor interaction with water and air, etc.). Analysis of hypothetical accidents is of relevance to predict the catastrophe involving loss of coolant accident (LOCA) in LMFBRs. One key parameter to take into account is the critical temperature data of the liquid metals for reactor safety analysis. This communication reports the application of a theoretical model called internal pressure approach to predict the critical temperature (T_c) of the LBE alloy for the first time.

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1. Introduction

Analysis of hypothetical yet highly improbable and undesirable accidents in the water-cooled thermal nuclear reactors as well as the liquid metal-cooled fast breeder nuclear reactors (LMFBRs) is of paramount importance. Such analysis is relevant to predict the catastrophe and ensure the desired safety throughout the reactor operation and during the periodic maintenance and shut down schedules. The possible accidents could involve fuel damage and inadvertent radioactive release during core meltdown and reactor runaway, loss of coolant due to boiling, ignition and reaction with the fuel and structural alloys as well as with the moderator, among others. The likelihood of these mishaps warrants assessing the conditions to reduce the probability of such accidents. An approach by which nuclear safety could be guaranteed and achieved is not by minimizing the probability but by precluding the very reasons leading to the accidents of the kind stated above and observed hitherto (Three Mile Island and Chernobyl, for example). Thus, a new philosophy based on the principles of natural and intrinsic safety ought to concurrently evolve by giving cognizance to the fundamental physical and chemical properties of the fuel, coolant and structural materials constituting the nuclear reactor system.

Liquid sodium had been the choice coolant for the LMFBRs during the 1980s and 1990s. However, due to its violent chemical reactivity towards air and water, an accidental Na-humid air contact could lead to its spontaneous combustion and to the release of high dose of airborne radioactivity and concomitant temperature rise. This in turn could lead to the LOCA (loss of coolant accident), causing further release and fuel damage. Beside the alkali metals, there is only a short list of candidate liquid metals such as Hg, Ga and Sn which have their own inherent shortfalls, such as the high cost, radioactivity, corrosion, toxicity, neutron activation,

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etc. On the other hand metals such as Bi (melting point = 544 K; boiling point = 2022 K) and Pb (melting point = 600 K; boiling point = 1837 K) as well as the Pb–Bi eutectic alloy (LBE, melting point = 396 K; boiling point = 1943 K) have been serious contenders for use as coolant in lieu of sodium due to a number of attractive characteristics (high density, low moderation, low neutron absorption and activation, high boiling point and poor interaction with water and air, etc.).

In the event of an accident, the thermodynamic properties of the fuel and the coolant play an important role [1]. Properties such as isothermal and adiabatic compressibility, isochoric specific heat and sonic velocity provide useful insight into the behavior of liquids under extreme temperatures and pressure excursions. Thus, the development of an equation of state up to the critical point for these systems becomes quite relevant.

Due to the current interest in the low melting Pb–Bi eutectic alloy (56.3 at.% or 56.5 wt% Bi) for its use in the generation IV liquid-metal cooled fast reactors (GEN IV-LFRs) [2], it becomes necessary to take into account the critical temperature data in view of the safe design of reactors and safety analysis under accident conditions. Direct determination of the critical parameters could be extremely difficult from the experimental point of view. On the other hand, theoretical models based on sound thermodynamic principles do provide such data with reasonable accuracy. This paper reports the application of such a theoretical model called internal pressure approach to predict the critical temperature (T_c) of the LBE alloy for the first time.

2. Methodology

The internal pressure approach (IPA) used in this work to compute the critical temperature is based on the following equation of state which interconnects the sonic velocity, (C, m s^{-1}), density (ρ, kg m^{-3}), volume expansivity (α, K^{-1}) and heat capacity (C_p, J kg^{-1} K^{-1}):

$$(P + P_i)/T = C^2\alpha\rho C_p/(C_p + TC^2\alpha^2), \tag{1}$$

where T is temperature in Kelvin and P and P_i are the pressure (Pa) and internal pressure (Pa), respectively. In an alternate form, Eq. (1) could also be written as:

$$(P + P_i)/T = C^2\alpha\rho\gamma, \tag{2}$$

where, γ is the ratio of the specific heat capacity at constant pressure to that at constant volume (C_p/C_v).

The methodology of IPA has been described in ample details by Azad and Ganesan [3]. The equivalence shown in Eqs. (1) and (2) could be derived easily by invoking the first law of thermodynamics, and the well known Maxwell relationships [4]. The validity of the proposed method has successfully been demonstrated in the case of a wide range of fluids including inert gases, pure liquid alkali metals, and Na–K and Pb–Li alloys [5–8].

Rowlinson [9] has observed that the variation of $(P + P_i)/T$ vs T is almost linear for most of the substances (liquids and liquid mixtures). This is illustrated schematically in Fig. 1. As expected, the internal pressure (P_i) tends to zero for vapors under ideal gas approximation, so that $(P + P_i)/T$ approaches P/T which in turn generally approaches zero for gases. On the other hand, Eq. (1) predicts that it is a non-zero quantity for liquids. Thus, it is evident that $(P + P_i)/T$ vs T line in

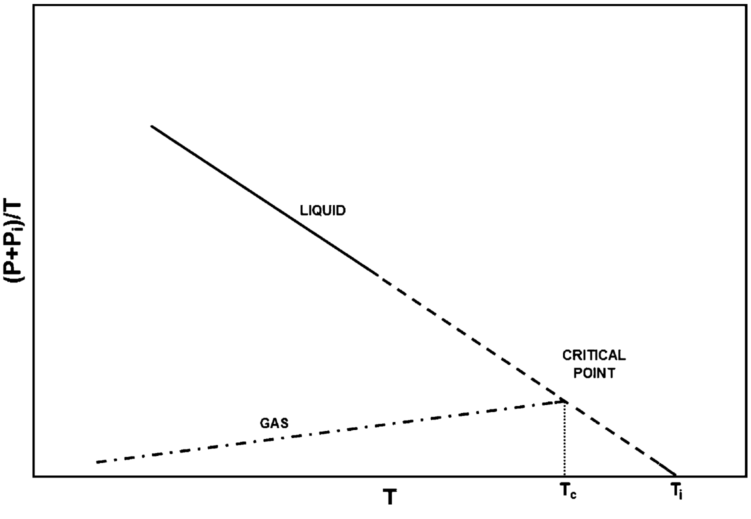

Fig. 1. Rowlinson plot of $(P + P_i)/T$ vs temperature for a typical liquid and gas.

Fig. 1 for a given liquid should intersect the corresponding line for the gaseous phase at their consulate temperature – the critical temperature, T_c. Though exaggerated for the sake of clarity, the scale of the ordinate in Fig. 1 is such that the ideal gas line could be identical with the abscissa as a good first approximation. Hence, the x-axis intercept of the liquid line, T_i, gives a value close to the critical temperature, the latter being computed by refined iteration after incorporating ideal gas approximation.

It has been found [5–8] that the value of $(P + P_i)/T$ computed as per Eq. (1) or (2) could be fitted into a linear expression of the type:

$$(P+P_i)/T = A - BT. \tag{3}$$

According to the schematic shown in Fig. 1, as $(P + P_i)/T \approx 0$, $T \rightarrow T_i$, so that:

$$0 = A - BT_i \quad \text{or} \quad A = BT_i. \tag{4}$$

Similarly, at the critical temperature, $(P + P_i)/T \approx P_c/T_c$, so that:

$$P_c/T_c = A - BT_c \quad \text{or} \quad A = P_c/T_c + BT_c. \tag{5}$$

By solving Eqs. (4) and (5), one gets:

$$T_i - T_c = \frac{P_c}{BT_c}, \tag{6}$$

from where

$$T_c = \frac{BT_i + \sqrt{(BT_i)^2 - 4BP_c}}{2B}. \tag{7}$$

Accordingly, the computation of T_c value from the slope (T_i) and intercept (B) of the $(P + P_i)/T$ vs T line, however, necessitates the a priori knowledge of the critical pressure of the system, P_c – a parameter which may sometime turn out to be as difficult to measure or predict as T_c itself. As drawn in Fig. 1, T_i is larger than T_c. Assuming the ideal gas behavior of the vapor, the difference between the intercept of the line $(P + P_i)/T$ vs T and the theoretical critical temperature can be deduced approximately as:

$$T_i - T_c \simeq (P_c/T_c)/|\text{slope}|. \tag{8}$$

The validity of this model has been verified in the case of liquid sodium [3–5], where Eq. (1) yields $T_i = 2652$ K and from Eq. (8) $T_i - T_c \sim 18$ K. However, at the critical point, for a Van der Waals gas, $(P + P_i)/T = 4P_c/T_c$ is a more reasonable correction. Incorporating this, the value of T_c then works out to be 2581 K ($4P_c/T_c \sim 71$ K) which compares well with a T_c vale of 2634 K assuming a Van der Waals gas behavior. It is interesting to note that in the case of the first five alkali metals (Li to Cs) the scatter ($T_i - T_c$) was found to be 21 ± 2 K [8]. The theoretically estimated values of T_c for sodium are in excellent agreement with the experimental values as well. Fink and Leibowitz [10] have recently tabulated the critical temperature values for sodium that range from 2485 to 2573 K; the recommended T_c value for sodium is 2503.7 ± 12 K.

There is no experimental or theoretical data in the literature on the critical parameters of bismuth or LBE alloy. The theoretically estimated critical temperature of liquid lead has been reported earlier by Azad et al. [7]. Hence, the IPA was employed to compute the critical temperature of pure bismuth and LBE alloy using the available thermophysical parameters needed for the calculation of $(P + P_i)/T$ values according to Eq. (1). Thermophysical data from more recent sources were also used to compute the T_c of molten lead again and compared with that reported earlier [7] to verify the consistency of the methodology.

It should be pointed out that in the case of a pure one-component system (such as, argon or n-hexane) both liquid and the gaseous phase consist of the same pure component. Furthermore, it is well established that in metals the liquid–vapor interface is different from the simple non-metallic liquid–vapor interface because of the variation of the local electronic structure with electron density in the interfacial region. This leads to a strong variation of the effective ion–ion interactions and the binding energy per ion with position in the interface [11,12]. Hence, while the critical temperature in non-metallic and simple metallic systems is a uniquely defined point of phase transition in the p–T coordinates, same might not be valid if the eutectic in the liquid phase or at the liquid–vapor interface at sufficiently high temperature (in the vicinity of boiling) has different electron density or, dissociates into two liquids (pure components or any other composition) prior to boiling. It is also likely that the eutectic experiences incongruent vaporization with concomitant ionic polymerization in the vapor phase. The thermodynamic analysis of such a situation becomes complicated, especially, in the absence of even the most primitive data that takes cognizance of such a possibility. For example, earlier measurements on the vapor pressure of liquid bismuth by Knudsen torsion–effusion, vacuum microbalance gravimetry and quasi-static methods [13–17] in a wide range of temperatures, have indicated that the vapor phase consists of ∼55–65 mol.% of Bi_2 species and 35–45 mol.% of Bi monomer. On the other hand, the vapors of lead were found to be monoatomic [17]. Shpil'rain et al. [18] have recently reported saturation vapor pressures in binary and ternary alloys (including eutectic compositions) of alkali metals, viz., Na–K, Na–Cs, Na–Rb, K–Cs, K–Rb and Na–K–Cs. Their results agree very well within the permissible limit of experimental and computational error with those derived by assuming the additive rule of the form: $p = p_1 \cdot x_1 + p_2 \cdot x_2$ (for a binary alloy), indicating that the constitution of the vapor phase remains essentially the same as that of the condensed phase. For example, at 1200 K, the saturated vapor pressure of the eutectic Na–0.821Rb was found to be 0.56585 MPa which is in excellent agreement with 0.55484 MPa (=0.179 ∗ 0.1482 + 0.821 ∗ 0.6435) obtained from the Gibbs–Konovalov additive law within 2% [19]. It was also shown that the error in the calculated values of the pressure of saturated vapors of a molten Na–Rb system was about ±5%. This confirms that the saturation pressure of a liquid A–A′ alkali metal alloy of a given composition and the results of the experimental investigation of the $p_s - T_s$ dependence of the same melt, differ from each other by not more than ±5%.

In the absence of reliable experimental data in the literature on the saturated vapor pressure over the molten LBE alloy and regarding the constitution of the vapor phase, we assume that the LBE composition exhibits absolute congruence in its melting as well as the vaporization. This assumption in conjunction with that of Van der Waals behavior in the gas phase is reasonable and predicts a trend very similar to that shown in Fig. 1. This is a crucial and significant assumption whose experimental validation is warranted.

3. Results and discussion

The input data on the heat capacity, density and sonic velocity in molten Bi and Pb used in the construction of the Rowlinson plots (akin to Fig. 1) are summa-

Table 1
Heat capacity, density and ultrasonic velocity for liquid Bi and Pb

Metal	$C_p = a + bT + c/T^2$			Temperature range T (K)	Refs.
	a	b	c		
(a) Heat capacity ($J\,kg^{-1}\,K^{-1}$)					
Bi	95.7	3.484×10^{-2}	1.011×10^{7}	544–1093	[20]
	150.206	–	–	578–715	[21]
	134.57	9.06	–	544–1273	[24][a]
Pb	156.50	-1.494×10^{-2}	–	600–1473	[20]
	155.47	-1.331×10^{-2}	–		[22]
	$\rho = a - bT$				
	a		b		
(b) Density ($kg\,m^{-3}$)					
Bi	10744		1.2549	578–715	[21][a]
	10394		1.2361	673–1073	[23]
	10809		1.4428	573–1235	[24][a]
Pb	11290.8		1.16466	600–1473	[24][a]
	$C = a + bT + cT^2$				
	a	b	c		
(c) Ultrasonic velocity ($m\,s^{-1}$)					
Bi	2111.3	−0.7971	–	578–715	[21][a]
Pb	1951.75	−0.343	7.635×10^{-5}	600–2000	[25]
	1790 ± 15	–	–	601	[26]
	1866.86	−0.277	–	601–643	[27]

[a] Parametric equations were derived by using the tabulated numerical data.

Table 2
Input data for the LBE alloy

Property	Parametric equation	Refs.
Heat capacity ($J\,Kg^{-1}\,K^{-1}$) ± 14	$121.82 + 3.04513 \times 10^{-2}T + 5.6923 \times 10^{6}/T^2$	[20]
Density ($kg\,m^{-3}$) ± 164	$10981.6835 - 1.1369T$	[32]
Ultrasonic velocity ($m\,s^{-1}$) ± 80	$2041.58 - 0.5987T + 3.3387 \times 10^{-5}T^2$	[21,25]
	$2004.37 - 0.5480T$	[21,27]

rized in Table 1. There are only sporadic experimental data on the density of the LBE in the past 55 years. For example, using the method of maximum pressure in a gas bubble, Been et al. [28] reported the density in the liquid LBE (56.5 wt% Bi) in the range 480–1271 K. Nikol'skii et al. [29,30] used a dilatometric technique

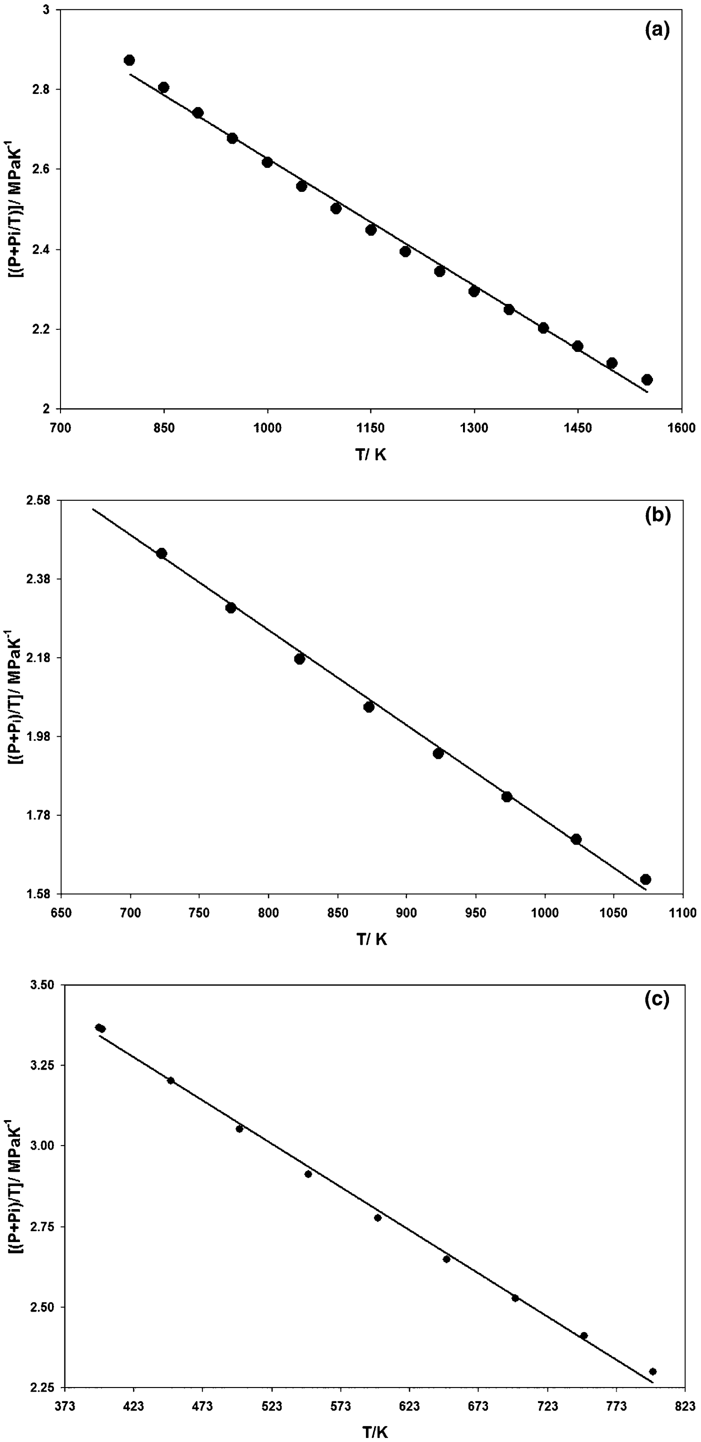

Fig. 2. Temperature dependence of $(P + P_i)/T$ in molten Pb (a), Bi (b) and LBE alloy (c).

for the determination of density in LBE (55.5 wt% Bi) in the temperature range of 420–860 K. Similarly, Kazakova et al. [31] measured the density of LBE (56.9 wt% Bi) in the range 400–1070 K using a sessile drop method. Alchagirov et al. [32] have recently assessed these data and caution their use since these measurements were reported in the form of graphs and approximating equations that make it difficult to estimate their error. They have also reported the density of molten LBE measured by pycnometry in the temperature range of 410–726 K and have given a parametric equation for the same that is valid in the range 398–800 K.

No reliable sonic velocity or heat capacity data (from experimental measurement or computational estimation) is available for molten LBE in the published literature. Hence, for the computation of internal pressure from Eq. (1), these were calculated by the law of additivity, viz., $X_{\mathrm{LBE}} = 0.563 * X_{(\mathrm{Bi})} + (1 - 0.563) * X_{(\mathrm{Pb})}$, where X is the property and 0.563 is the atom fraction of Bi in the LBE. The input data obtained in this fashion for the calculation of T_c of the LBE alloy are summarized in Table 2 along with the computed uncertainties.

The $(P + P_i)/T$ values calculated from Eq. (1) for molten lead, bismuth and the lead–bismuth eutectic alloy are plotted as a function of temperature in Fig. 2 (a through c). The linearity of these plots confirms the validity of IPA as a reliable tool for the estimation of T_c values of these systems. It should be pointed out that the accuracy of IPA depends intrinsically on the accuracy of the input parameters. This is illustrated for lead and bismuth in Fig. 3(a) and (b), respectively, when input parameters for a given system from different sources were employed. The accuracy also depends on the linearity of the Rowlinson plot; any deviation from linearity could lead to very large errors in the extrapolation. The least-squares expressions obtained via iteration of the computed data for molten bismuth, lead and LBE alloy are given in Table 3. Critical pressure values for

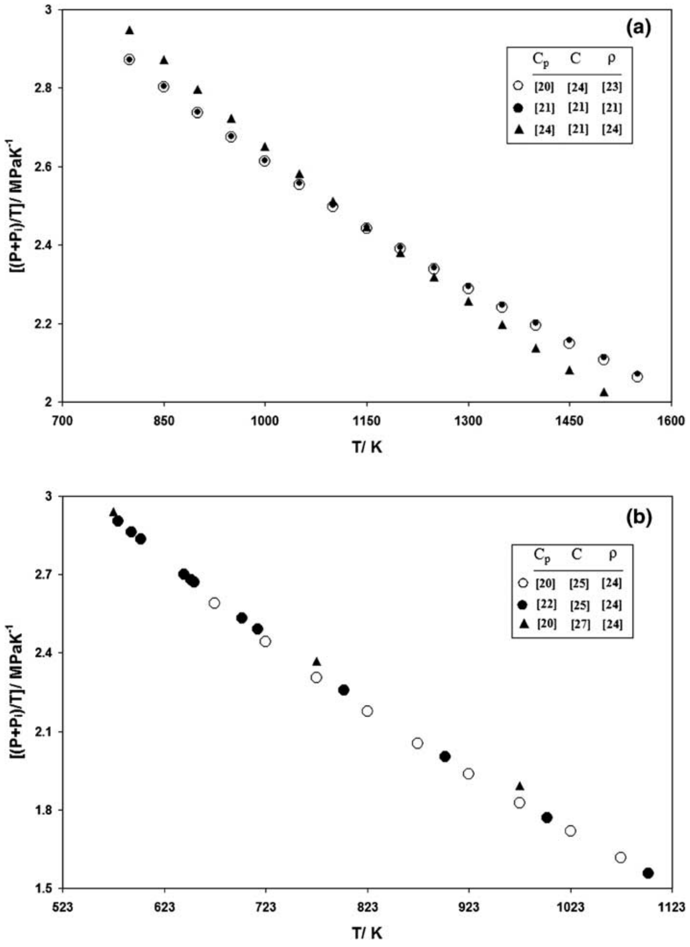

Fig. 3. Effect of the choice of input data (see Table 3) on the temperature dependence of $(P + P_i)/T$ in molten Pb (a) and, Bi (b). The source of input data is also indicated.

Table 3
Least-squares expressions derived via iteration, for the Rowlinson plots for Pb(*l*), Bi(*l*) and LBE alloy (shown in Figs. 2 and 3)

System	$[(P + P_i)/T]$/MPa $K^{-1} = A + BT/K$		Intercept T_i (K)	Critical temperature (K)[a]
	A	$B \times 10^3$		
Pb	3.6942	−1.0710	3449	3369
	3.6883	−1.0626	3471	3391
	3.6796	−1.0659	3452	3372
Bi	2.8927	−1.3554	2134	2054
	2.8505	−1.3042	2186	2106
	3.2611	−1.5365	2122	2042
LBE	3.1542	−1.2663	2491	2411

[a] Using $T_i - T_c \simeq 80$ K.

bismuth, lead or LBE alloy are not readily available in the literature. Therefore, it was difficult to calculate the critical temperatures by employing Eq. (7) or (8). However, using the observed trend of scatter between T_i and T_c in the case of alkali metals [8], the critical temperatures of Bi, Pb and LBE were calculated by treating the vapors to be Van der Waals real gases. These values are also listed in Table 3.

From the foregoing discussion it is clear that the intercepts (T_i) obtained via iteration in each case are consistent within the acceptable range of computational error. It is also worth mentioning that in the case of bismuth, even though slightly different parametric equation was obtained when employing the density and heat capacity data reported by Lyon [24], the intercept value (2122 K) was in good agreement with those arrived at by using the input parameters from other sources for the calculation of $(P + P_i)/T$. This evaluation of T_c for LBE does not take into account the deviation from ideal behavior in computing density, sonic velocity and heat capacity from those of the constituent elements. A more reliable estimation of the critical temperature in each of these systems should have been possible if more accurate and reliable experimental data on the thermophysical properties of these liquid systems were available. An error of ±76 was estimated for the T_c value of the LBE alloy based on the standard deviation in the computed $(P + P_i)/T$ values. This value is very close to the difference $T_i - T_c$ (~80). The theoretical approach reported in this work, therefore, underlines the need of direct and accurate determination of crucial parameters such as heat capacity and sonic velocity, isothermal compressibility and the critical parameters, in order to describe a more realistic equation of state for the coolants of fast breeder nuclear reactors.

The critical temperature in a single component phase diagram (P–T) is the terminal point of the liquid–gas phase boundary and as such does not enjoy the uniqueness of the triple point in a true thermodynamic sense (degree of freedom $\neq 0$). Though there appears to be no report on the critical temperature of LBE alloy or that of the terminal elements, it is worthwhile to make an internal comparison of the values of T_c obtained in this work for the pure elements and the eutectic alloy. This is shown in Fig. 4, along with their respective melting and boiling points.

The critical temperature values derived in this work show a rather monotonic variation with atom fraction of Bi. Interestingly, boiling points also follow a similar but shallower trend while the melting behavior of the

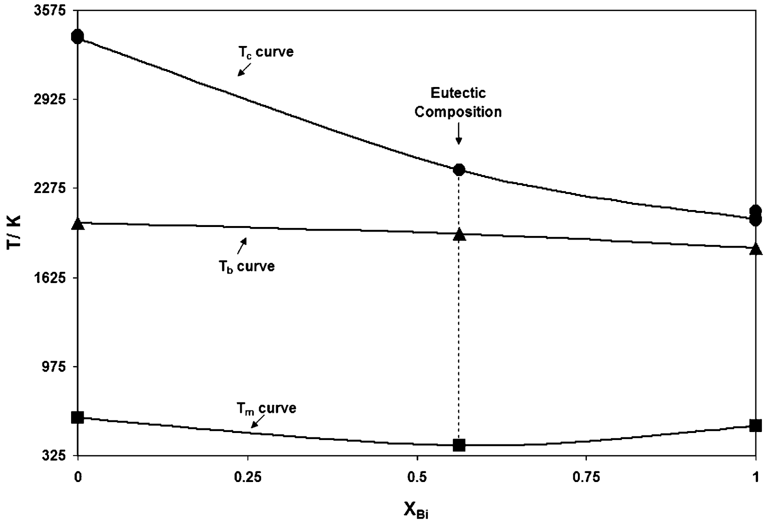

Fig. 4. Variation of critical temperature in the liquid Pb–Bi binary system as a function of atom fraction of bismuth.

system is in line with that expected in a binary alloy system with a eutectic. As such, the locus of the critical temperature along the composition axis resembles in its concavity the liquidus of the liquidus–vaporous curve of a continuously miscible two-component system.

4. Conclusion

In the event of a nuclear accident, the thermodynamic properties of the fuel and the coolant play an important role. Parameters such as isothermal and adiabatic compressibility, isochoric specific heat and sonic velocity provide useful insight into the behavior of liquids under extreme temperatures and pressure excursions. Thus, the development of an equation of state up to the critical point for these systems becomes relevant. For the first time, a simple equation of state based on the concept of internal pressure in liquids has been used to estimate the critical temperatures of bismuth and lead–bismuth eutectic (potential coolants in fast nuclear reactors). It should, however, be emphasized that while this work establishes a theoretical basis for computing the critical temperature of the LBE alloy, there is a need for direct and accurate determination of the crucial thermophysical properties that could verify these results.

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