Chapter 2

THERMOPHYSICAL AND ELECTRIC PROPERTIES*

2.1 Introduction

Among different heavy liquid metals (HLM), lead (Pb) and three alloys of lead: lead-bismuth eutectic (LBE) 44.5 wt.% Pb + 55.5 wt.% Bi, lead-lithium eutectic 99.32 wt.% Pb + 0.68 wt.% Li, and lead-magnesium eutectic 97.5 wt.% Pb-2.5 wt.% Mg, are considered at present as potential candidates for the coolant of new generation fast reactors (critical and subcritical) and for liquid spallation neutron sources and accelerated driven systems (ADS). LBE is expected to be used in most of ADS projects, mainly due to its low melting temperature of ~397 K (~124°C), resulting in lower corrosion rates and in easier maintenance.

This chapter compiles data on the main thermo-physical properties of molten lead, bismuth and LBE (e.g. density, molar volume, isobaric heat capacity, viscosity, thermal and electrical conductivity, etc.) reported in the open literature. In some cases, significant discrepancies exist among the values given by different sources. Therefore, recommendations based on the "best fit" of data are usually used. Published data on the properties the lead alloys of interest are currently very limited. The main sources are material handbooks published in the 50s and 60s. The first compilations of the main thermophysical properties of Pb and LBE were assembled by [Lyon, 1952], [Kutateladze, 1959], and [Nikolskii, 1959]. In the later handbooks most of data were either reproduced without changes [Mantell, 1958], [Lyon, 1960], [Bonilla, 1964] and [Crean, 1964] or with addition of new results [Friedland, 1966], [Hultgren, 1973, 1974], [Iida, 1988], [Kirillov, 2000], [Gurvich, 1991] and [Cheynet, 1996]. In recent years several review-reports have been published where previous data were reanalysed [Cevolani, 1998], [Imbeni, 1998a, 1998b], [Kyrillov, 2000a, 2000b], [TECDOC-1289, 2002], [Sobolev, 2002, 2005, 2007]. These data and a many of the recommendations and conclusions in this chapter are mainly based on these later reports. Some publications issued in the former Soviet Union and recent Russian compilation reports are not taken into account in this version of Chapter 2 because of difficulties in receiving them.

The reliability of data depends on the method used for production and the care with which the method is used. In general, data concerning metals in the liquid or vapour state show a significant dispersion, with the exception of the melting points. Only a few authors of compilations have paid adequate attention to dispersion and standard deviation of their reported values [Hultgren, 1973, 1974].

The database in this chapter is presented in the form of a set of tables. Each table is devoted to one parameter and contains information about the references used, year of publication, measurement method, precision, temperature range, and composition of a sample. Moreover, values of the parameter from the reference and correlation obtained on the basis of the available data are given. Often it was not possible to access the original sources of data. In this case, the data selected from handbooks and

---

other compilations were used to fill in the database. In the case where information concerning precision or/and method was not available, a question mark (?) is used to indicate that the data should be used with caution. In all recommended correlations, temperature is given in degrees of Kelvin ("kelvins").

2.2   Pb-Bi alloy phase diagram

One of the first more or less complete phase diagrams for the binary Pb-Bi system was published in the handbook of G.O. Hiers [Hiers, 1948] and reproduced later in the well-known Smithells Metal Reference Book [Smithells, 1955]. This phase diagram is presented in Figure 2.2.1 below. (It was reproduced almost without changes in the book of B. Ageron, *et al*. [Ageron, 1959] and in the later editions of the Smithells Metal Reference Book [Smithells, 1983, 2004].)

Figure 2.2.1. Phase diagram of the Pb-Bi system [Smithells, 1955]
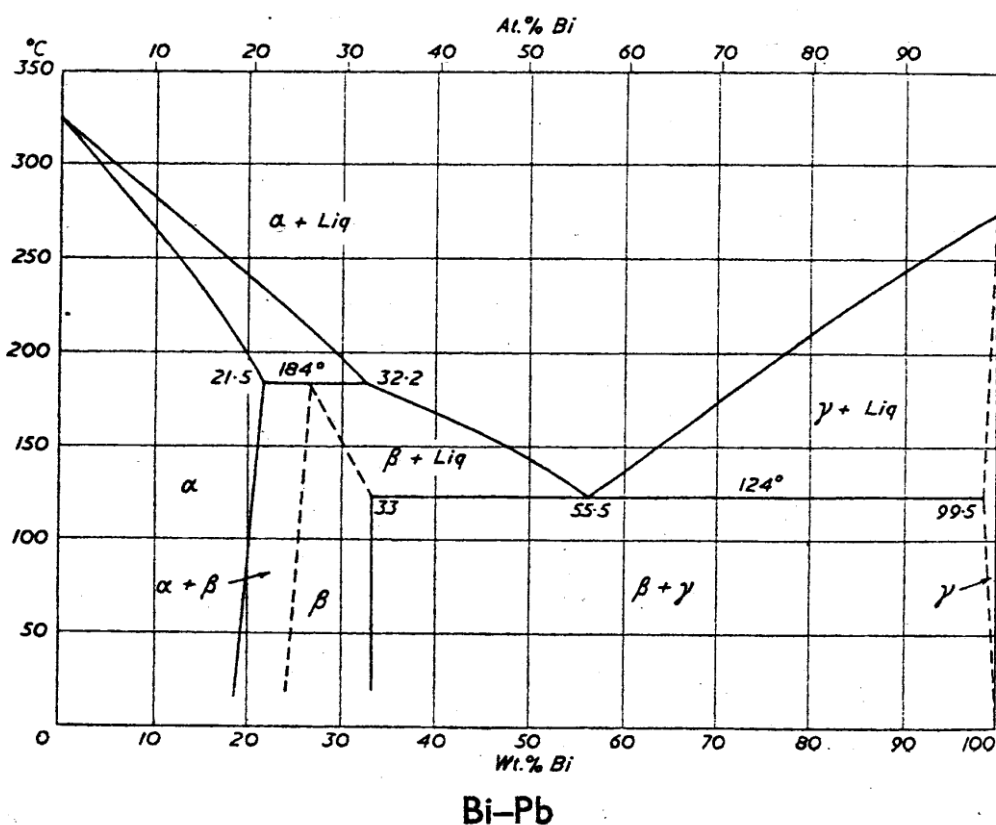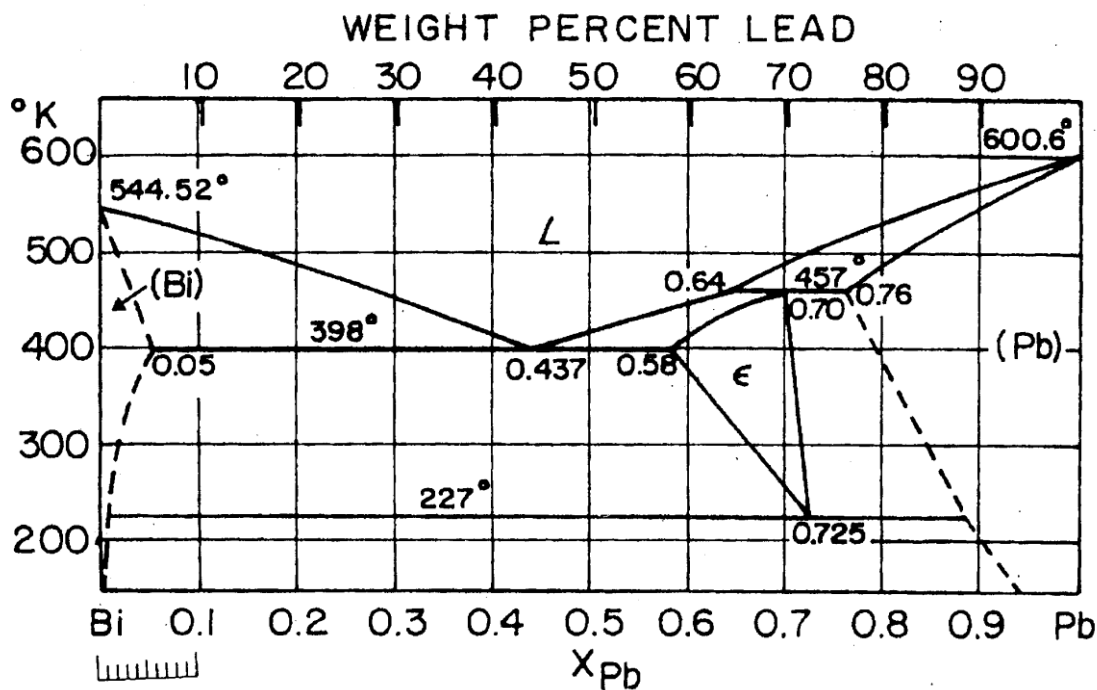

This diagram shows:

- an eutectic point at 55.5 wt.% Bi with a melting temperature of 124°C (397 K);

- a peritectic point at 32.2 wt.% Bi with a melting temperature of 184°C (457 K);

- the solubility limits in solid state: 21.5 wt.% Bi in Pb (α-phase region) and 0.5 wt.% Pb in Bi (γ-phase region);

- intermetallic compound phase (β-phase region);

- liquidus and solidus lines.

M. Hansen and K. Anderko [Hansen 1958] presented the Pb-Bi phase diagram with some new experimental results. This diagram with additional revisions was reported in [Elliott 1965]. Some parameters were changed in comparison with the diagram reproduced in Figure 2.2.1 as follows:

- the eutectic point at 56.7 wt.% Bi (56.3 at.% Bi) with a melting temperature of 124.7°C (398 K);

- the peritectic point at 36.2 wt.% Bi (36 at.% Bi);

- the solubility limits in the solid state are reported to be 23.4 wt.% (23.3 at.%) Bi in Pb.

In 1973, the Pb-Bi phase diagram with refinements of the boundaries of the ε-phase, given by B. Predel and W. Schwerman [Predel, 1967], and boundaries of γ(Bi)-phase, given by M.V. Nosek, *et al.* [Nosek, 1967], was published by R. Hultgren, *et al.* [Hultgren, 1973]. This diagram is reproduced in Figure 2.2.2 below.

Figure 2.2.2. Phase diagram of the Pb-Bi system [Hultgren, 1973]

This diagram provides the same eutectic and peritectic points as those proposed by [Elliott 1965], but gives:

- the melting point of Bi at 271.22°C (544.52 K);

- the melting point of Pb at 327.3°C (600.6 K);

- the solubility limit of Pb in Bi in the solid state – 5 at.%;

- the solubility limit of Bi in Pb in the solid state – 24 at.%;

- an eutectoid point at 72.5 at.% Pb and -46.7°C (227 K);

- ε-phase region.

In 1992 N.A. Gokcen [Gokcen, 1992] proposed a few modifications for some characteristic points (Figure 2.2.3):

- more precise melting points of elements: $T_{melt\ Bi}$ = 271.442°C (544.592 K); $T_{melt\ Pb}$ = 327.502°C (600.652 K);

- the eutectic point at 45.0 at.% Pb and $T_{melt\ LBE}$ = 125.5°C (398.65 K);

- the peritectic point at 71 at.% Pb and 187°C (460.15 K);

- the lower limits of the elements solubility in the solid state – 0.5 at.% Pb in Bi and 22 at.% Bi in Pb.

These modifications were reproduced in a Pb-Bi phase diagram published in the ASM Handbook of 1992 [Baker, 1992].

Figure 2.2.3. Phase diagram of the Pb-Bi system [Gokcen, 1992]
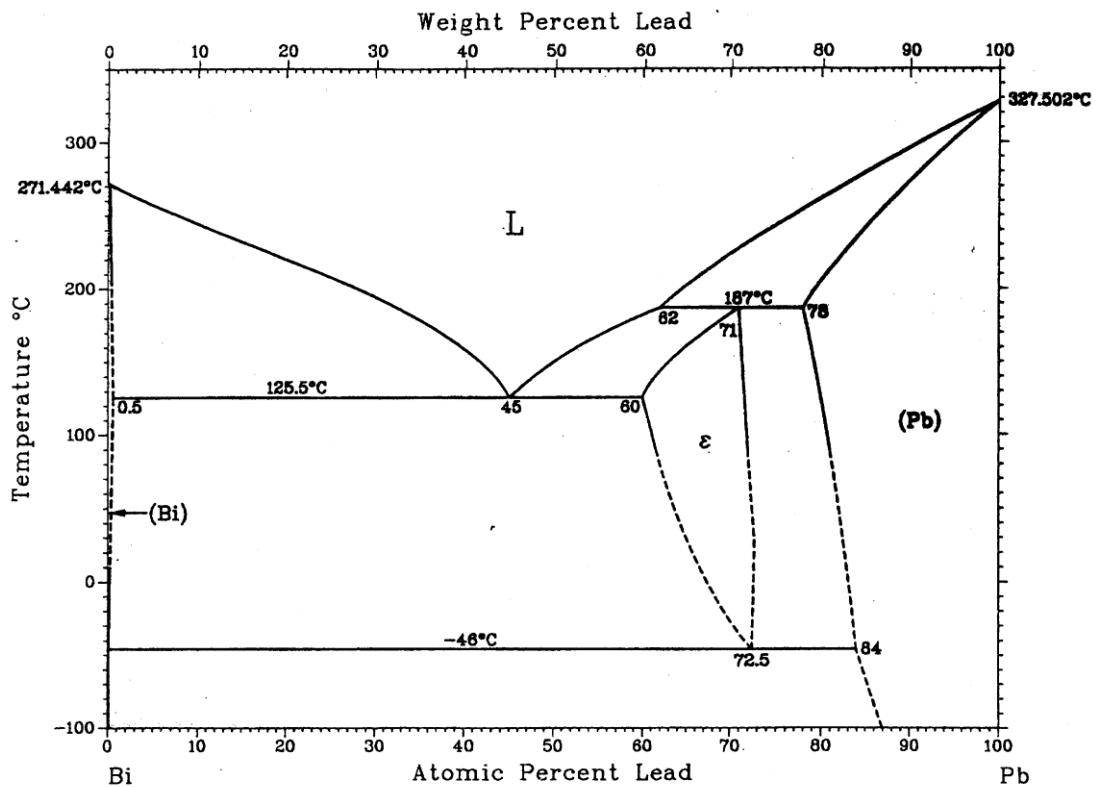
In many Russian publications (e.g. [Orlov 1997, 2003]), followed recently by other authors, a phase-diagram is often presented which gives the LBE eutectic composition at 55.5 wt.% Bi and 44.5 wt.% Pb with the eutectic melting temperature of 123.5°C (396.65 K); the temperature is probably reproduced from [Kutateladze, 1959].

Recommendation

The phase diagram of N.A. Gokcen [Gokcen, 1992] is recommended for use in engineering and design calculations with the exception of the eutectic point which will be considered in the next section.

2.3 Normal melting point

2.3.1 Lead

The values of the lead and bismuth metling temperatures were found in the following handbooks [Lyon, 1954, 1960], [Kutateladze, 1959], [Hofmann, 1970], [Hultgren, 1974], [Lucas, 1984], [Iida, 1988], [Kubaschevski, 1979, 1993], [Gocksen, 1992], [Cheynet, 1996] and [Smithells, 2004].

Friedland [Friedland, 1966] reproduced the values of the melting points from Lyon [Lyon, 1954, 1960]. The compilation [Imbeni, 1998a] presented data on the lead and bismuth melting temperatures from different sources with the conclusion that dispersion is not large. In the handbook [Kyrillov, 2000a] and in the IAEA report [TECDOC-1289, 2002] the lead melting temperature presented in earlier compilations [Kutateladze, 1959] was repeated. These sources have not been included in the database on the melting point.

Hofmann [Hofmann, 1970] refers to an evaluation of the literature data performed by [Kohlraush, 1956]. Hultgren, *et al.* collected data from many earlier sources, and recommended average values and uncertainties [Hultgren, 1974].T. Iida and R.I.L. Guthrie [Iida, 1988] took their data from the Iwanami Dictionary of Physics and Chemistry [Tamamushi, 1981]. O. Kubaschewski, *et al.* [Kubaschewski, 1979, 1993] cited the JANAF Thermochemical Tables [Chase, 1978, 1982], [Knacke, 1991], [Pankratz 1982], and [Hultgren, 1974]. The compilation [Cheynet, 1996] references the JANAF Thermochemical Tables [Chase, 1989], [Barin, 1985], [Cheynet, 1989] and [Knacke, 1991]. The Smithells Metals Reference Book [Smithells, 2004] presented the melting temperatures from the 82nd edition of the CRC Handbook of Chemistry and Physics [CRC Handbook, 2002].

The data for the melting temperature of lead and bismuth, extracted from the above selected sources, are presented in Tables 2.3.1 and 2.3.2 respectively.

All selected data yield approximately the same value within their error limits. The most probable value for the melting temperature of technically pure lead obtained on the basis of the data presented in the table is:

$$T_{melt\,Pb} = 600.6 \pm 0.1 \text{ K} \tag{2.1}$$

The melting point of lead increases by 0.0792 K per 1 MPa when pressure increases from about 15 up to 200 MPa. The increase continues at a lower rate, 0.0671 K per 1 MPa, in the range of 800-1200 MPa, and an increase of 5.4 K for a pressure increase from about 2 to 3 GPa was cited in [Hoffman, 1970].

2.3.2 Bismuth

The database for the melting point of bismuth are extracted from about the same sources as for lead and is presented in Table 2.3.2.

For the melting temperature of bismuth there is uncertainty in the cited data in the first digit after the decimal point. Therefore the recommended mean value is as follows:

$$T_{melt\,Bi} = 544.4 \pm 0.3 \text{ K} \tag{2.2}$$

Table 2.3.1. Database on the normal melting point of lead

Reg. no.	Parameter	Measurement method	Estimated accuracy	Temperature range, K	Pressure range, Pa	Values	Ref.
3.1.1	Melting temperature	Temperature vs. power	0.1°C	n/a	~ 10^5 (?)	327.4°C	[Lyon, 1954]
	Interpolation function: T_{melt} = 600.65 K						
	Comments: Unknown purity						
3.1.2	Melting temperature	Temperature vs. power	0.1°C	n/a	~ 10^5 (?)	327.4°C	[Kutateladze, 1959]
	Interpolation function: T_{melt} = 600.65 K						
	Comments: Unknown purity						
3.1.3	Melting temperature	n/a	0.1°C (?)	n/a	~ 10^5 (?)	327.3°C	[Hofmann 1970]
	Interpolation function: T_{melt} = 600.55 K						
	Comments: Unknown purity						
3.1.4	Melting temperature	Temperature vs. power	0.1°C	n/a	~ 10^5 (?)	600.6 K	[Hultgren, 1974]
	Interpolation function: T_{melt} = 600.6 K						
	Comments: Unknown purity						
3.1.5	Melting temperature	?	0.05 K	n/a	~ 10^5 (?)	600.55 K	[Iida, 1988]
	Interpolation function: T_{melt} = 600.55 K						
	Comments: Unknown purity						
3.1.6	Melting temperature	?	0.005°C (?)	n/a	~ 10^5 (?)	327.502°C	[Gokcen, 1992]
	Interpolation function: T_{melt} = 600.652 K						
	Comments: High purity						
3.1.7	Melting temperature	?	?	n/a	~ 10^5 (?)	601 K	[Kubaschewski, 1993]
	Interpolation function: T_{melt} = 601 K						
	Comments: Unknown purity						
3.1.8	Melting temperature	?	0.1°C (?)	n/a	~ 10^5 (?)	327.4°C	[Cheynet, 1996]
	Interpolation function: T_{melt} = 600.65 K						
	Comments: Unknown purity						
3.1.9	Melting temperature	?	0.005°C (?)	n/a	~ 10^5 (?)	327.462°C	[Smithells, 2004]
	Interpolation function: T_{melt} = 600.612 K						
	Comments: Unknown purity						

Table 2.3.2. Database on the normal melting point of bismuth

Reg. no.	Parameter	Measurement method	Estimated accuracy	Temperature range, K	Pressure range, Pa	Values	Ref.
3.2.1	Melting temperature	Temperature vs. power	0.1°C (?)	n/a	~ 10^5 (?)	271.0°C	[Lyon, 1954]
	Interpolation function: T_{melt} = 544.15 K						
	Comments: Unknown purity						
3.2.2	Melting temperature	Temperature vs. power	1°C (?)	n/a	~ 10^5 (?)	271°C	[Kutateladze, 1959]
	Interpolation function: T_{melt} = 544.15 K						
	Comments: Unknown purity						
3.2.3	Melting temperature	Temperature vs. power	0.05 K	n/a	~ 10^5 (?)	544.52 K	[Hultgren, 1974]
	Interpolation function: T_{melt} = 544.52 K						
	Comments: Unknown purity						
3.2.4	Melting temperature	?	1 K (?)	n/a	~ 10^5 (?)	545 K	[Kubaschewski, 1993]
	Interpolation function: T_{melt} = 545 K						
	Comments: Unknown purity						
3.2.5	Melting temperature	?	0.05 K	n/a	~ 10^5 (?)	544.10 K	[Iida, 1988]
	Interpolation function: T_{melt} = 544.10 K						
	Comments: Unknown purity						
3.2.6	Melting temperature	?	0.005°C (?)	n/a	~ 10^5 (?)	271.442°C	[Gokcen, 1992]
	Interpolation function: T_{melt} = 544.592 K						
	Comments: High purity						
3.2.7	Melting temperature	?	0.05°C (?)	n/a	~ 10^5 (?)	271.40°C	[Smithells, 2004]
	Interpolation function: T_{melt} = 544.55 K						
	Comments: High purity						

2.3.3 LBE

The sources of data for the LBE melting point included in this handbook are [Lyon, 1954], [Kutateladze, 1959], [Hultgren, 1973], [Smithells, 1955], [Eliot, 1965] and the ASM Handbook [Baker, 1992]. These data were reproduced in later handbooks [Lyon, 1960], [Kyrillov, 2000a], [Smithells, 1983] and compilations [Cevolani, 1998], [Imbeni, 1998b], [TECDOC-1289, 2002], [Sobolev, 2002]. The selected sources included in the database (Table 2.3.3) give for the LBE melting temperature T_{melt} = 123.5-125.5°C (396.7-398.7 K) at normal atmospheric pressure.

The mean value of:

$$T_{melt\ LBE} = 397.7 \pm 0.6 \text{ K} \tag{2.3}$$

is recommended on the basis of the data presented in Table 2.3.3.

Table 2.3.3. Database on the LBE normal melting point

Reg. no.	Parameter	Measurement method	Estimated accuracy %	Temperature range, K	Pressure range, Pa	Values	Ref.
3.3.1	Melting temperature	?	?	n/a	$\sim 10^5$ (?)	125°C	[Lyon, 1954]
	Interpolation function: T_{melt} = 398.15 K						
	Comments: 44.5 wt.% Pb + 55.5 wt.% Bi. Unknown purity.						
3.3.2	Melting temperature	?	?	n/a	$\sim 10^5$ (?)	123.5°C	[Kutateladze, 1959]
	Interpolation function: T_{melt} = 396.65 K						
	Comments: 44.5 wt.% Pb + 55.5 wt.% Bi. Unknown purity.						
3.3.3	Melting temperature	?	?	n/a	$\sim 10^5$ (?)	398 K	[Hultgren, 1973]
	Interpolation function: T_{melt} = 398 K						
	Comments: 43.7 at.% Pb + 56.3 at.% Bi. Unknown purity.						
3.3.4	Melting temperature	?	?	n/a	$\sim 10^5$ (?)	124°C	[Smithells, 1955]
	Interpolation function: T_{melt} = 397.15 K						
	Comments: 44.8 wt.% Pb + 55.2 wt.% Bi. Unknown purity.						
3.3.5	Melting temperature	?	?	n/a	$\sim 10^5$ (?)	125°C	[Eliot, 1965]
	Interpolation function: T_{melt} = 398.15 K						
	Comments: 43.7 at.% Pb + 56.3 at.% Bi. Unknown purity.						
3.3.6	Melting temperature	?	?	n/a	$\sim 10^5$ (?)	125.5°C	[Gokcen, 1992]
	Interpolation function: T_{melt} = 398.65 K						
	Comments: 45.0 at.% Pb + 55.0 at.% Bi. Unknown purity.						

2.4 Volume change at melting and solidification

A detailed knowledge of volume changes in metals and alloys at their melting points is of critical importance in the understanding of solidification processes.

- *Solid lead*. Similar to the majority of metals with the FCC crystal structure, lead exhibits a volume increase upon melting. At normal conditions a volume increase of 3.81% has been observed in pure lead [Iida, 1988]. In several engineering handbooks a value of ~3.6% is often given for lead of technical purity [Lyon, 1954, 1960], [Kyrillov, 2002].

- *Solid bismuth*. Solid bismuth shows a volume contraction during melting, similar to other semimetals. The anisotropic rigid bonds are apparently broken on melting, and the neighbouring atoms are packed closer one to another. According to [Iida, 1988], pure bismuth contracts approximately 3.87% upon melting. A contraction of 3.32% was reported by [Lyon, 1954, 1960] and a value of ~3.3% was recommended in an IAEA report [Kyrillov, 2002] for Bi-coolant.

- *LBE*. A negligible volume change on melting of solid LBE at normal atmospheric pressure has been published in the handbook of Lyon [Lyon, 1954]. This recommendation has been repeated in later handbooks and compilations (e.g. [Kyrillov, 2002], [Sobolev, 2002]).

Recommendations for the mean values of the volume change upon melting are summarised for Pb, Bi and LBE in Table 2.4.1. It is recommended that for very slow melting (quasi-equilibrium conditions) the volume change upon melting is close to zero for LBE.

Table 2.4.1. Volume change of pure lead, bismuth and LBE at melting

	Pb	**Bi**	**LBE**
$\Delta V_m/V_m$	+3.7	-3.7	~0.0

The situation is more complicated for LBE freezing and melting accompanied by rapid temperature change. In the handbook of Lyon [Lyon, 1954] a 1.43 vol.% contraction of LBE on freezing with a subsequent expansion of the solid of 0.77 vol.% at an arbitrary temperature of 65°C has been reported. A contraction of 1.52±0.1 vol.% of the solid phase after solidification of LBE has also been mentioned in [Hofmann, 1970]. The results of measurements of LBE expansion over time at room temperature after solidification and rapid cooling has been reported by H. Glasbrenner *et al.* [Glasbrenner, 2005]. In these results, shown in Figure 2.4.1, one can see that after solidification and cooling the studied material contracted about 0.35%. After about 100 minutes of exposure at room temperature, its volume returns to the initial value, but after one year its linear size increases by about 1.2%.

The problems of freezing and melting of LBE in the reactor circuits were analysed by [Pylchenkov, 1999]. He pointed out that the results of the freezing/de-freezing experiments are very sensitive to the experimental conditions and that very long times (>100 d) are usually required to reach equilibrium. Some of his results are shown in Figure 2.4.2. The volume effect upon freezing/de-freezing depends very strongly on phase-structure of local transformations in the solid state related to the mutual solubility of LBE components (Figure 2.2.1). A negligible volume change has been observed in some experiments. According to E.H. Pylchenkov [Pylchenkov 1999], post-solidification expansion may occur in a metastable alloy as a result of local changes in the composition. An excess in γ-phase precipitation during freezing can result in a volume increase.

Figure 2.4.1. Linear expansion of LBE as a function of time after solidification and cooling down to room temperature [Glasbrenner, 2005]
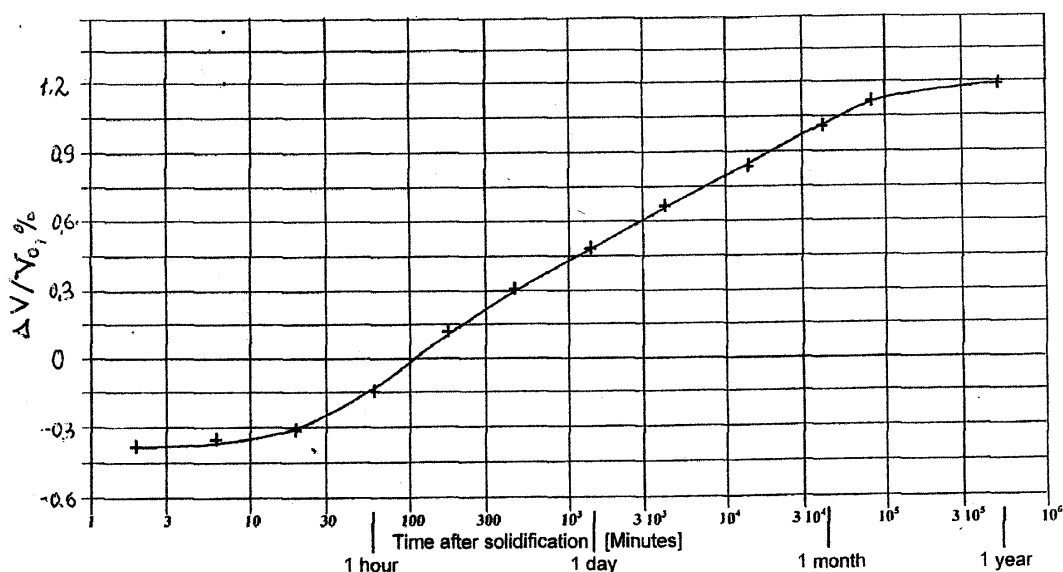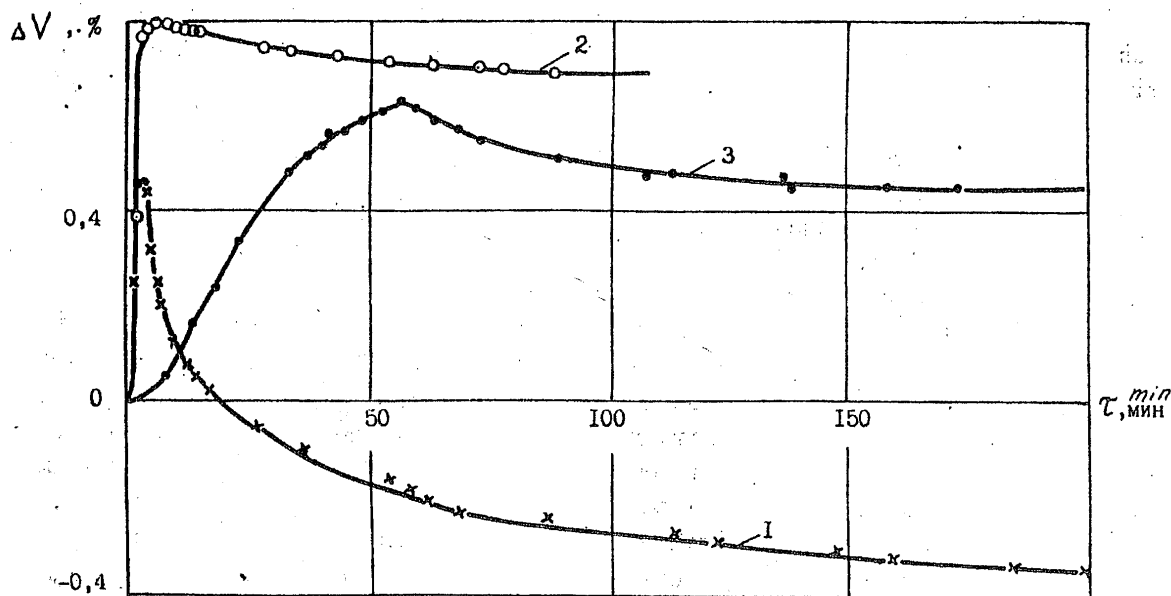
**Figure 2.4.2. Solid LBE volume evolution as a function of time
after heat-up from 25 to 125°C ($< T_{melt}$) [Pylchenkov, 1999]**

1 – Fast heat-up (prehistory: few years at 20-25 °C)
2 – Fast heat-up (prehistory: cooling down to 25 °C after solidification and exposure during 65 hours
3 – Heat-up time is one hour (prehistory: three hours of exposure at 124 °C followed by 15 hours of exposure at 25 °C)

H. Glasbrenner [Glasbrenner, 2005] showed that re-crystallisation at the level of a crystal grain is responsible for the expansion of LBE after freezing. In their experiments, two phases were identified immediately after rapid solidification of LBE:

- β-phase: an intermetallic compound with 42 at.% Bi (at the freezing point) and a hexagonal closed packed structure;

- γ-phase: a solid solution of Pb in Bi (~0.4 at.%Pb) with a rhombohedric structure.

The β-phase formed at freezing is not stable at room temperature. With time it is partly transformed into a β-phase with a lower Bi content (about 35 at.%) and γ-phase [Takeda, 1997], [Agostini, 2004]. As the γ-phase needs much more space, the transformation produces an expansion of the material.

Measurements of this kind of LBE expansion performed at room temperature [Gröschel, 2005] showed that it is reduced by a factor of 6 when slow cooling is used to 60-90°C (333.3-363.3 K) in the place of quenching. Moreover, Takeda's experiments [Takeda, 1997] showed that the first period of a faster expansion [which ranges from 2 h at 110°C (383.3 K) to three months at 20°C (293.3 K)] is followed by a second one with a lower expansion rate. The final increase in volume rarely exceeds 1.3% in one year.

Pylchenkov [Pylchenkov, 1999] pointed out the general tendency: the volume effect of LBE melting-solidification is minimal for pure eutectic materials.

2.5 Latent heat of melting at the normal melting point

2.5.1 *Lead*

The heat of melting of lead and bismuth at the normal melting point (the enthalpy change on melting) was taken from about the same sources as for the melting temperature: [Lyon 1954], [Hultgren, 1974], [Cheynet, 1996], [Kubaschevski, 1993], [Iida, 1988], [Lucas, 1984], and from the IAEA report [TECDOC-1289, 2002]. The selected database is presented in Tables 2.5.1 and 2.5.2 below. The value of the bismuth latent heat of melting from Cheynet [Cheynet, 1996] has not been included in the database because it was extracted from the same source as that of Hultgren [Hultgren, 1974].

Table 2.5.1. Database on the latent heat of melting of lead at the normal melting point

Reg. no.	Parameter	Measurement method	Estimated accuracy	Temperature range, K	Pressure range, Pa	Values	Ref.
5.1.1	Heat of melting	?	?	n/a	$\sim 10^5$ (?)	5.89 cal/g	[Lyon, 1954]
	Interpolation function: $Q_{melt} = 24.7$ kJ/kg = 5.11 kJ/mole						
	Comments: Unknown purity						
5.1.2	Heat of melting	?	?	n/a	$\sim 10^5$ (?)	24.7 kJ/kg	[TECDOC-1289, 2002]
	Interpolation function: $Q_{melt} = 24.7$ kJ/kg = 5.11 kJ/mole						
	Comments: Unknown purity						
5.1.3	Heat of melting		42 J/mole	n/a	$\sim 10^5$ (?)	4799 J/mole	[Hultgren, 1974]
	Interpolation function: $Q_{melt} = 23.2$ kJ/kg = 4.80 kJ/mole						
	Comments: Unknown purity						
5.1.4	Heat of melting	?	0.5 kJ/mole (?)	n/a	$\sim 10^5$ (?)	4.8 kJ/mole	[Kubaschewski, 1993]
	Interpolation function: $Q_{melt} = 23$ kJ/kg = 4.8 kJ/mole						
	Comments: Unknown purity						
5.1.5	Heat of melting	?	?	n/a	$\sim 10^5$ (?)	4.81 kJ/mol	[Iida, 1988]
	Interpolation function: $Q_{melt} = 23.2$ kJ/kg = 4.81 kJ/mole						
	Comments: Unknown purity						
5.1.6	Heat of melting	?	?	n/a	$\sim 10^5$ (?)	5109 J/mol	[Lucas, 1984]
	Interpolation function: $Q_{melt} = 24.7$ kJ/kg = 5.11 kJ/mole						
	Comments: Unknown purity						
5.1.7	Heat of melting	?	?	n/a	$\sim 10^5$ (?)	4.76 kJ/mole	[Cheynet, 1996]
	Interpolation function: $Q_{melt} = 23.0$ kJ/kg = 4.76 kJ/mole						
	Comments: Unknown purity						

The selected values for lead lie in the range of 4.76-5.11 kJ/mole. The mean value and the mean deviation are:

$$Q_{melt\ Pb} = 4.9 \pm 0.2 \text{ kJ/mole} = 23.8 \pm 0.7 \text{ kJ/kg} \tag{2.4}$$

These values are recommended for the latent heat of melting of the pure lead at the normal melting point.

2.5.2 *Bismuth*

The database for the latent heat of melting of bismuth is presented in Table 2.5.2.

Table 2.5.2. Database on the latent heat of melting of bismuth at the normal melting point

Reg. no.	Parameter	Measurement method	Estimated accuracy	Temperature range, K	Pressure range, Pa	Values	Ref.
5.2.1	Heat of melting	?	?	n/a	~ 10^5 (?)	12.0 cal/g	[Lyon, 1954]
	Interpolation function: Q_{melt} = 50.2 kJ/kg = 10.5 kJ/mole						
	Comments: Unknown purity						
5.2.2	Heat of melting	?	?	n/a	~ 10^5 (?)	54.7 kJ/kg	[TECDOC-1289, 2002
	Interpolation function: Q_{melt} = 54.7 kJ/kg = 11.4 kJ/mole						
	Comments: Unknown purity						
5.2.3	Heat of melting	?	209 J/mole	n/a	~ 10^5 (?)	11297 J/mole	[Hultgren, 1974]
	Interpolation function: Q_{melt} = 54.1 kJ/kg = 11.3 kJ/mole						
	Comments: Unknown purity						
5.2.4	Heat of melting	?	0.5 kJ/mole (?)	n/a	~ 10^5 (?)	11.3 kJ/mole	[Kubaschewski, 1993]
	Interpolation function: Q_{melt} = 54.1 kJ/kg = 11.3 kJ/mole						
	Comments: Unknown purity						
5.2.5	Heat of melting	?	?	n/a	~ 10^5 (?)	10.89 kJ/mol	[Iida, 1988]
	Interpolation function: Q_{melt} = 52.1 kJ/kg = 10.9 kJ/mole						
	Comments: Unknown purity						
5.2.6	Heat of melting	?	?	n/a	~ 10^5 (?)	10.5 kJ/mol	[Lucas, 1984]
	Interpolation function: Q_{melt} = 50.2 kJ/kg = 10.5 kJ/mole						
	Comments: Unknown purity						

In the case of Bi, the difference between the maximum and minimum values of the heat of melting is about 0.8 kJ mol^{-1}. The mean value and the mean deviation are:

$$Q_{melt\ Bi} = 11.0 \pm 0.4 \text{ kJ/mole} = 52.6 \pm 1.7 \text{ kJ/kg} \tag{2.5}$$

This value is recommended for the latent heat of melting of the pure bismuth at the normal melting point.

2.5.3 *LBE*

The most cited results for the heat of melting of LBE at normal atmospheric pressure were published in the handbook edited by Friedland [Friedland, 1966]. The recent IAEA compilation [TECDOC-1289, 2002] probably reproduces the value of the melting heat from [Kyrillov, 2000b]. The data from these sources are included in Table 2.5.3.

The mean value for the LBE melting heat of:

$$Q_{melt\ LBE} = 8.01 \pm 0.07 \text{ kJ/mole} = 38.5 \pm 0.3 \text{ kJ/kg} \tag{2.6}$$

is the recommended value at normal atmospheric pressure.

Table 2.5.3. Database on the latent heat of melting of LBE fusion at the normal melting point

Reg. no.	Parameter	Measurement method	Estimated accuracy %	Temperature range, K	Pressure range, Pa	Values	Ref.
5.3.1	Heat of melting	?	?	n/a	$\sim 10^5$ (?)	38134 J kg^{-1}	[Friedland, 1966]
	Interpolation function: $Q_{melt} = 38.134$ kJ/kg = 7.939 kJ/mole						
	Comments: Unknown purity						
5.3.2	Heat of melting	?	?	n/a	$\sim 10^5$ (?)	38.8 kJ kg^{-1}	[TECDOC-1289, 2002]
	Interpolation function: $Q_{melt} = 38.8$ kJ/kg = 8.08 kJ/mole						
	Comments: 44.5 wt.% Pb + 55.5 wt.% Bi. Unknown purity.						

2.6 Normal boiling point

2.6.1 Lead

For the boiling temperature of lead and bismuth at normal pressure, the same sources were used as for their melting temperature: [Lyon, 1954], [Kutateladze, 1959], [Hultgren, 1974], [Lucas, 1984], [Iida, 1988], [Kubaschevski, 1993], [Cheynet, 1996] and [Smithells, 2004], plus, the handbook edited by Howe [Howe, 1961] and the recent IAEA report [TECDOC-1289, 2002].

The data for the boiling temperatures of lead and bismuth extracted from the above-mentioned sources are presented in Tables 2.6.1 and 2.6.2 respectively.

The variation in the lead boiling temperature between the selected sources is more than for the melting temperature and is a large as 25 K. The most probable value of:

$$T_{boil\ Pb} = 2016 \pm 10\ K \tag{2.7}$$

is recommended for the boiling temperature of technically pure lead at normal conditions, based on an analysis of the data in Table 2.6.1.

2.6.2 Bismuth

The database on the normal boiling point of bismuth is presented in Table 2.6.2.

A significant level of uncertainty exists for the boiling point of bismuth with a range of about 150 K between the maximum and the minimum. Therefore the following mean value and the mean deviation, while uncertain, is recommended for the boiling temperature of technically pure bismuth at normal conditions:

$$T_{boil\ Bi} = 1806 \pm 50\ K \tag{2.8}$$

2.6.3 LBE

The boiling point of LBE at normal atmospheric pressure is given in the handbook of Lyon [Lyon, 1954], and by Kutateladze [Kutateladze, 1959]. Both sources give the same value of

Table 2.6.1. Database on the normal boiling point of lead

Reg. no.	Parameter	Measurement method	Estimated accuracy	Temperature range, K	Pressure range, Pa	Values	Ref.
6.1.1	Boiling temp.	Temperature vs. power	10°C (?)	n/a	~ 10^5 (?)	1737°C	[Lyon, 1954]
	Interpolation function: T_{boil} = 2010 K						
	Comments: Unknown purity.						
6.1.2	Boiling temp.	Temperature vs. power	10°C (?)	n/a	~ 10^5 (?)	1740°C	[Kutateladze, 1959]
	Interpolation function: T_{boil} = 2013 K						
	Comments: Unknown purity.						
6.1.3	Boiling temp.	Temperature vs. power	5°C (?)	n/a	~ 10^5 (?)	1749°C	[Smithels, 2004]
	Interpolation function: T_{boil} = 2022 K						
	Comments: Unknown purity.						
6.1.4	Boiling temp.	?	°C (?)	n/a	~ 10^5 (?)	1745°C	[TECDOC-1289, 2002]
	Interpolation function: T_{boil} = 2018 K						
	Comments: Unknown purity.						
6.1.5	Boiling temp.	?	5°C (?)	n/a	~ 10^5 (?)	1750°C	[Iida, 1988]
	Interpolation function: T_{boil} = 2023 K						
	Comments: Unknown purity.						
6.1.6	Boiling temp.	?	5°C (?)	n/a	~ 10^5 (?)	1737°C	[Cheynet, 1996]
	Interpolation function: T_{boil} = 2010 K						
	Comments: Unknown purity.						

Table 2.6.2. Database on the normal boiling point of bismuth

Reg. no.	Parameter	Measurement method	Estimated accuracy	Temperature range, K	Pressure range, Pa	Values	Ref.
6.2.1	Boiling temp.	Temperature vs. power	10°C (?)	n/a	~ 10^5 (?)	1477°C	[Lyon, 1954]
	Interpolation function: T_{boil} = 1750 K						
	Comments: Unknown purity.						
6.2.2	Boiling temp.	Temperature vs. power	10°C (?)	n/a	~ 10^5 (?)	1490°C	[Kutateladze, 1959]
	Interpolation function: T_{boil} = 1763 K						
	Comments: Unknown purity.						
6.2.3	Boiling temp.	Temperature vs. power	10°C	n/a	~ 10^5 (?)	1564°C	[Smithells, 2004]
	Interpolation function: T_{boil} = 1837 K						
	Comments: Unknown purity.						
6.2.4	Boiling temp.	?	°C (?)	n/a	~ 10^5 (?)	1627°C	[Howe, 1961]
	Interpolation function: T_{boil} = 1900 K						
	Comments: Unknown purity.						
6.2.5	Boiling temp.	?	5 K	n/a	~ 10^5 (?)	1833 K	[Iida, 1988]
	Interpolation function: T_{boil} = 1833 K						
	Comments: Unknown purity.						
6.2.6	Boiling temp.	?	5°C (?)	n/a	~ 10^5 (?)	1477°C	[Cheynet, 1996]
	Interpolation function: T_{boil} = 1750 K						
	Comments: Unknown purity.						

$T_{boil\ LBE}$ = 1670°C (1943 K). This value was reproduced later in other publications (e.g. [Friedland, 1966], [Hultgren, 1973], [TECDOC-1289, 2002]). However, in the handbook of Lyon [Lyon, 1954] it is mentioned that in the original source $T_{boil\ LBE}$ = 1677°C (1950 K) was also cited. The recommendations [Lyon, 1954] and [Kutateladze, 1959] have been included in the database presented in Table 2.6.3.

Table 2.6.3. Database on the LBE normal boiling point

Reg. no.	Parameter	Measurement method	Estimated accuracy	Temperature range, K	Pressure range, Pa	Values	Ref.
6.3.1	Boiling temp.	?	?	n/a	~ 10^5 (?)	1670°C	[Lyon, 1954]
	Interpolation function: T_{boil} = 1670°C = 1943 K						
	Comments:						
6.3.2	Boiling temp.	?	?	n/a	~ 10^5 (?)	1670°C	[Kutateladze, 1959]
	Interpolation function: T_{boil} = 1670°C = 1943 K						
	Comments: 44.5 wt.% Pb + 55.5 wt.% Bi. Unknown purity.						

Based on the results of Table 2.6.3, and assuming that uncertainty is the same as for the bismuth boiling temperature, the value of:

$$T_{boil\ LBE} = 1943 \pm 50 \text{ K} \tag{2.9}$$

is recommended for the normal boiling temperature of the technically pure LBE.

2.7   Heat of vaporisation at the normal boiling point

The latent heat (enthalpy) of vaporisation is a measure of the cohesive energy of atoms in a liquid metal. Therefore, it correlates with surface tension and thermal expansion.

2.7.1   Lead

The information on the latent heat of vaporisation of lead and bismuth at the normal boiling point (the enthalpy change on boiling) is not numerous. The following sources were included in the database: [Lyon, 1954], Smithells Metals Reference Book [Smithells, 1983], [Iida, 1988], [Lucas, 1984], and the recommendations of the IAEA report [TECDOC-1289, 2002]. The selected database is presented in Tables 2.7.1 and 2.7.2.

The literature values are very close, with the difference between maximum and minimum values less than 1%. The mean value and the mean deviation are:

$$Q_{boil\ Pb} = 177.8 \pm 0.4 \text{ kJ/mole} = 858.2 \pm 1.9 \text{ kJ/kg} \tag{2.10}$$

These values are recommended for the latent heat of vaporisation for pure lead at the normal boiling point.

2.7.2   Bismuth

The database for the latent heat of vaporisation of molten bismuth at the normal boiling point is presented in Table 2.7.2.

Table 2.7.1. Database on the latent heat of vaporisation of lead at the normal boiling point

Reg. no.	Parameter	Measurement method	Estimated accuracy	Temperature range, K	Pressure range, Pa	Values	Ref.
7.1.1	Heat of boiling	?	?	n/a	$\sim 10^5$ (?)	204.6 cal/g	[Lyon, 1954]
	Interpolation function: Q_{boil} = 856.7 kJ/kg = 177.5 kJ/mole						
	Comments: Unknown purity						
7.1.2	Heat of boiling	?	?	n/a	$\sim 10^5$ (?)	856.8 kJ/kg	[TECDOC-1289, 2002]
	Interpolation function: Q_{boil} = 856.8 kJ/kg = 177.5 kJ/mole						
	Comments: Unknown purity						
7.1.3	Heat of boiling	?	1 kJ/mole (?)	n/a	$\sim 10^5$ (?)	178 kJ/mole	[Iida, 1988]
	Interpolation function: Q_{boil} = 859 kJ/kg = 178 kJ/mole						
	Comments: Unknown purity						
7.1.4	Heat of boiling	?	1 kJ/mole (?)	n/a	$\sim 10^5$ (?)	177.5 kJ/mole	[Lucas,1984]
	Interpolation function: Q_{boil} = 856.8 kJ/kg = 177.5 kJ/mole						
	Comments: Unknown purity						
7.1.5	Heat of boiling	?	?	n/a	$\sim 10^5$ (?)	178.8 kJ/mol	[Smithells, 1983]
	Interpolation function: Q_{boil} = 862.9 kJ/kg = 178.8 kJ/mole						
	Comments: Unknown purity						

Table 2.7.2. Database on the heat of vaporisation of bismuth at the normal boiling point

Reg. no.	Parameter	Measurement method	Estimated accuracy	Temperature range, K	Pressure range, Pa	Values	Ref.
7.2.1	Heat of boiling	?	?	n/a	$\sim 10^5$ (?)	204.3 cal/g	[Lyon,1954]
	Interpolation function: Q_{boil} = 855.4 kJ/kg = 178.8 kJ/mole						
	Comments: Unknown purity.						
7.2.2	Heat of boiling	?	?	n/a	$\sim 10^5$ (?)	852 kJ/kg	[TECDOC-1289, 2002]
	Interpolation function: Q_{boil} = 852 kJ/kg = 178.1 kJ/mole						
	Comments: Unknown purity.						
7.2.3	Heat of boiling	?	?	n/a	$\sim 10^5$ (?)	178.6 J/mole	[Smithells, 1983]
	Interpolation function: Q_{boil} = 857.4 kJ/kg = 179.2 kJ/mole						
	Comments: Unknown purity.						
7.2.4	Heat of boiling	?	0.1 kJ/mole (?)	n/a	$\sim 10^5$ (?)	194 kJ/mole	[Iida, 1988
	Interpolation function: Q_{boil} = 928 kJ/kg = 194 kJ/mole						
	Comments: Unknown purity.						
7.2.5	Heat of boiling	?	?	n/a	$\sim 10^5$ (?)	J/mol	[Lucas, 1984]
	Interpolation function: Q_{boil} = 855.5 kJ/kg = 178.8 kJ/mole						
	Comments: Unknown purity.						

The mean value and the mean deviation are:

$$Q_{boil\ Bi} = 181 \pm 4 \text{ kJ/mole} = 857 \pm 20 \text{ kJ/kg} \tag{2.11}$$

These values are recommended for the latent heat of vaporisation of pure bismuth at the normal boiling point.

2.7.3 LBE

A value of 178.352 kJ/mol was published in the handbook of Friedland [Friedland, 1966] for the latent heat of vaporisation of LBE at the normal boiling point. The more recent IAEA compilation [TECDOC-1289, 2002] recommends a value of 177 kJ/mol. These two sources were included in the database and are listed in Table 2.7.3.

Table 2.7.3. Database on the heat of vaporisation of LBE at the normal boiling point

Reg. no.	Parameter	Measurement method	Estimated accuracy %	Temperature range, K	Pressure range, Pa	Values	Ref.
7.3.1	Heat of boiling	?	?	n/a	$\sim 10^5$ (?)	178352 J/mole	[Friedland, 1966]
	Interpolation function: Q_{boil} = 856722 J/kg = 178352 J/mole						
	Comments: ?						
7.3.2	Heat of boiling	?	?	n/a	$\sim 10^5$ (?)	852 kJ/kg	[TECDOC-1289, 2002]
	Interpolation function: Q_{boil} = 852 J/kg = 177 J/mole						
	Comments: 44.5 wt.% Pb + 55.5 wt.% Bi. Unknown purity.						

The mean values of:

$$Q_{boil\ LBE} = 178.0 \pm 0.4 \text{ kJ/mole} = 854 \pm 2 \text{ kJ/kg} \tag{2.12}$$

are recommended for the latent heat of vaporisation of the pure LBE at the normal boiling point.

2.8 Saturation vapour pressure

The vapour pressure of a liquid metal is an important property which is directly related to the latent heat of evaporation (cohesive energy). For the equilibrium between liquid ($_l$) and vapour ($_v$) phases, the Clausius-Clapeyron equation states that:

$$\frac{dp}{dT} = \frac{H_v - H_l}{T\left(V_v - V_l\right)} \tag{2.13}$$

Therefore, assuming that the vapour behaves as a perfect gas and neglecting the volume of liquid in comparison with that of the gas, it can be obtained that:

$$p = A \cdot \exp\left(-\frac{\Delta H}{RT}\right) \tag{2.14}$$

where A is a constant of integration and ΔH is the heat (enthalpy) of evaporation.

The above equation can provide approximate values for equilibrium vapour pressures over a wide range of temperature due to the relatively small variation of ΔH with temperature. Closer fits of the experimental results can sometimes be obtained by adding supplementary temperature dependent terms.

2.8.1 Lead

The data for the saturation vapour pressure of lead at different temperatures were found in the following publications: [Lyon, 1954, 1960], [Friedland, 1966], [Hultgren, 1974], in the Smithells Metals Reference Book [Smithells, 1893, 2004], in the compilations of B. Cheynet [Cheynet, 1996] and of O. Kubaschewski, *et al.* [Kubaschewski, 1993] reproduced in [Iida, 1988].

O. Kubaschewski, *et al.* [Kubaschewski, 1993] collected the data from [Gross, 1948], [Eucken, 1934] and [Kelley, 1935]. B. Cheynet, *et al.* [Cheynet, 1996] recompiled the results communicated by himself earlier [Cheynet, 1989] and by M.W. Chase *et al.* [Chase, 1985], I. Barin, *et al.* [Barin, 1989], and O. Knacke, *et al.* [Knacke, 1991].

Data reported by J.R. Weeks [Weeks, 1971] were taken from [Lyon, 1954, 1960] and [Smithells, 1955], therefore they were not included in the database.

Figures 2.8.1 and 2.8.2 show the values of vapour pressure and deviation of molten lead, respectively, taken from the above cited literature and plotted as a function of temperature. It can be seen that, at first view, the agreement among the different sources is satisfactory. Using the selected data and Eq. (2.14) as a first approach, the following correlation was obtained for the saturated vapour pressure of molten lead where temperature is in kelvins:

$$p_s = 6.189 \times 10^9 \cdot \exp\left(-22216/T\right) \qquad (2.15)$$

The deviation of the selected literature data on the saturated vapour pressure of lead from the recommended correlation (2.15) is plotted in Figure 2.8.2. One can see that agreement is rather good and the deviation is about 12-15% (except the low temperature points of [Hultgren 1974]). Under theses circumstances, there is no need for a more precise correlation. Therefore relationship (2.15) is recommended for estimation of the saturated vapour pressure of lead from the melting temperatures up to the normal boiling point.

2.8.2 Bismuth

The data on the saturation vapour pressure for bismuth as a function of temperature were found in the publications of Lyon [Lyon, 1954, 1960], Hove [Hove, 1961], Hultgren [Hultgren, 1974], in the Smithells Metals Reference Book [Smithlls, 1983, 2004], in the compilations of Kubaschewski [Kubaschewski, 1993] and Chynet *et al.* [Cheynet, 1996]. Hove [Hove, 1961] based his compilation on the data presented in [Lyon, 1954, 1960], [Smithells, 1955], [Hansen, 1958], and other older sources. R. Hultgren [Hultgren, 1974] reported values selected on the basis of a critical assessment of many previously published data.

Figures 2.8.3 shows the values for the saturated vapour pressure of molten bismuth from the above cited sources plotted as a function temperature.

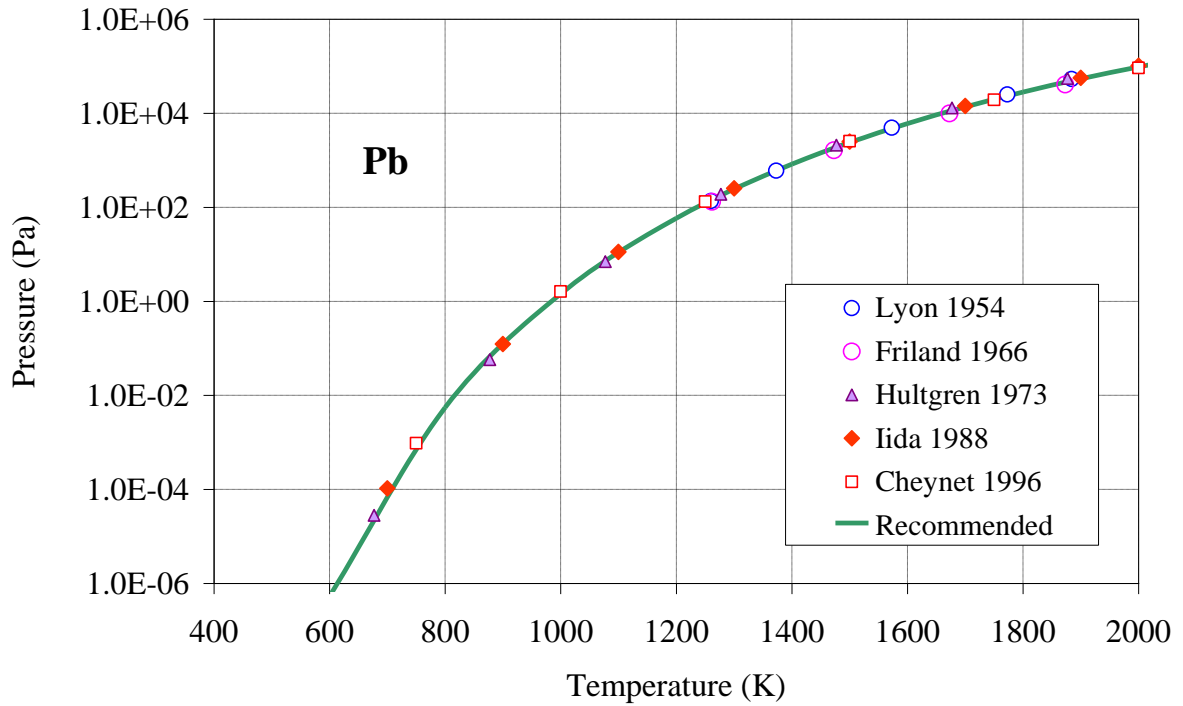**Figure 2.8.1. Saturated vapour pressure of molten lead versus temperature**

Figure 2.8.2. Deviation of the selected literature data on the saturated vapour pressure of lead from the recommended correlation (2.15)
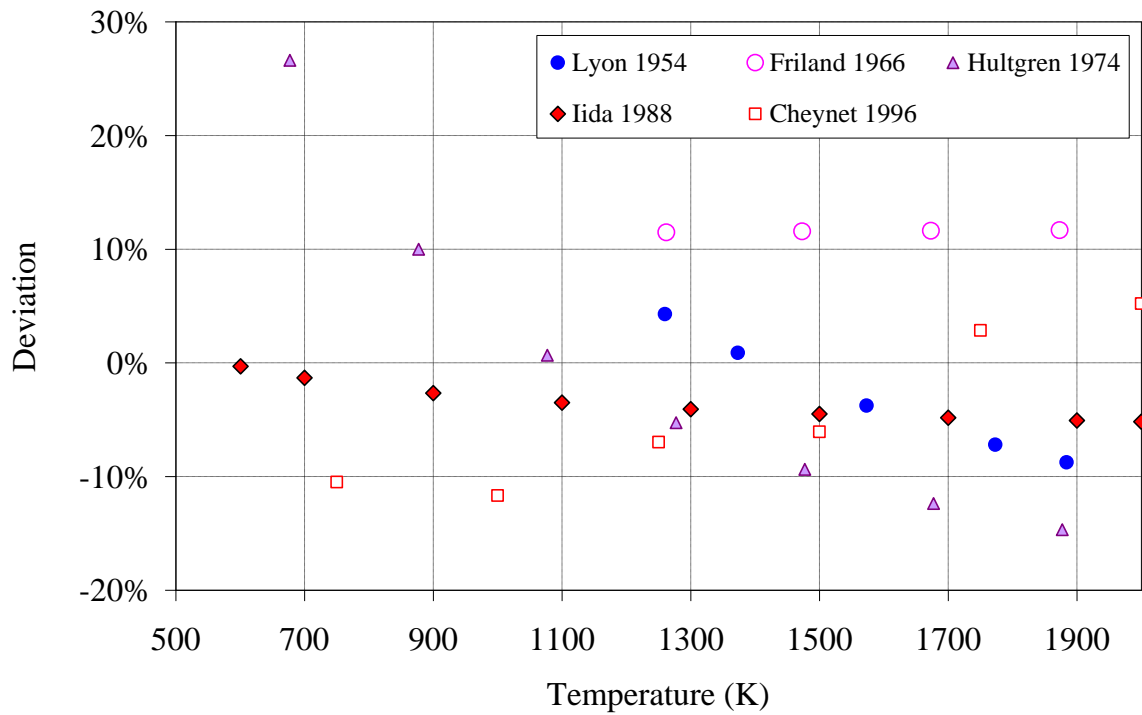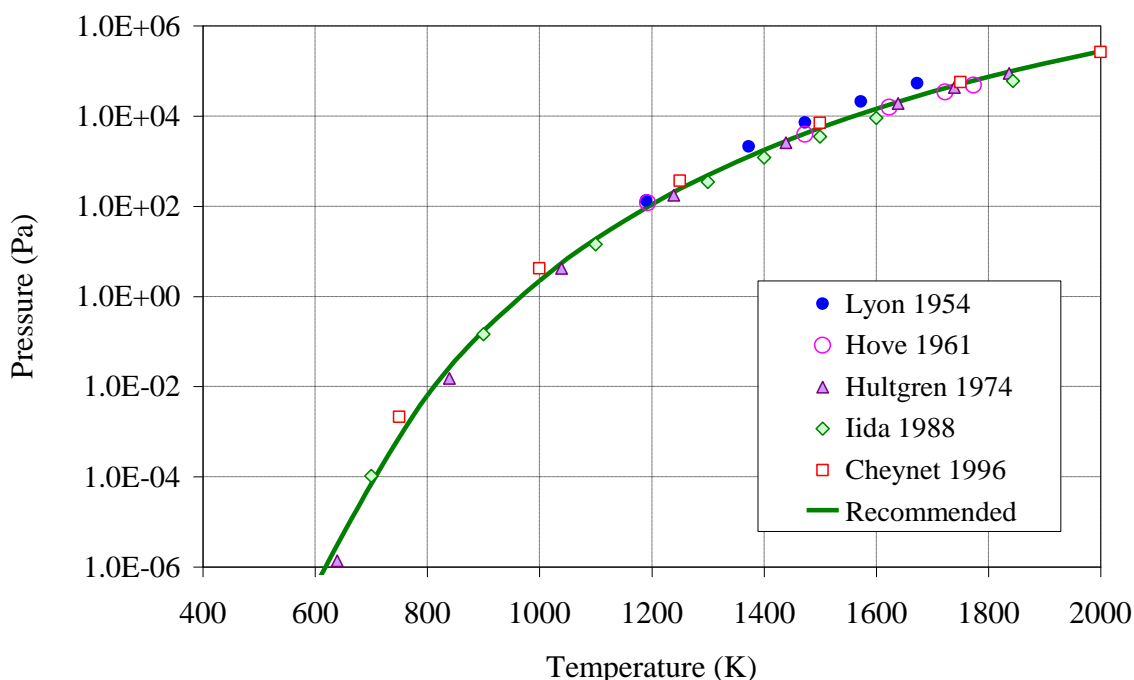
Figure 2.8.3. Saturated vapour pressure of bismuth versus temperature

It can be seen that the agreement in temperature dependence given by the different sources is satisfactory. However a significant scatter exists in values. Therefore Eq. (2.14) was chosen to construct an empirical correlation allowing one to calculate the vapour pressure of liquid bismuth as a function of temperature. Using the selected data, the following correlation was obtained for the saturated vapour pressure of molten bismuth where temperature is in kelvins:

$$p_{sBi}[Pa] = 2.4723 \cdot 10^{10} \cdot \exp(-22858/T) \tag{2.16}$$

This correlation is recommended for temperatures up to the normal boiling point.

The deviation of the selected literature data for the saturated vapour pressure of bismuth from the recommended correlation (2.16) is illustrated in Figure 2.8.4. One can see that very large differences exist between different sources. The recommended correlation (2.16) gives "the best estimate" with a maximum error of about 70%.

2.8.3 LBE

R.B. Tupper, *et al*. [Tupper, 1991] studied polonium evaporation from LBE and published some experimental results on the LBE (saturation) vapour pressure at temperatures 235-268 and 520-550°C (508-541, 793-823 K). Later Yu. Orlov, *et al*. [Orlov, 1997] published experimental values for the saturation pressure of the LBE vapour at equilibrium with the liquid phase at five different temperatures. However, both publications mentioned that a very large uncertainty in the pressure measurement exists at low temperatures (<500°C = 773 K). The results of [Orlov, 1997] were reproduced later in [Gromov, 1997]. New results have recently been communicated by P. Michelato, *et al*. [Michelato, 2003], P. Schuurmans, *et al*. [Schuurmans, 2005], and published by S. Ohno, *et al*. [Ohno 2005]. All these sources were included in the database and their results are plotted in Figure 2.8.5.

Figure 2.8.4. Deviation of the selected literature data on the saturated vapour pressure of bismuth from the recommended correlation (2.16)
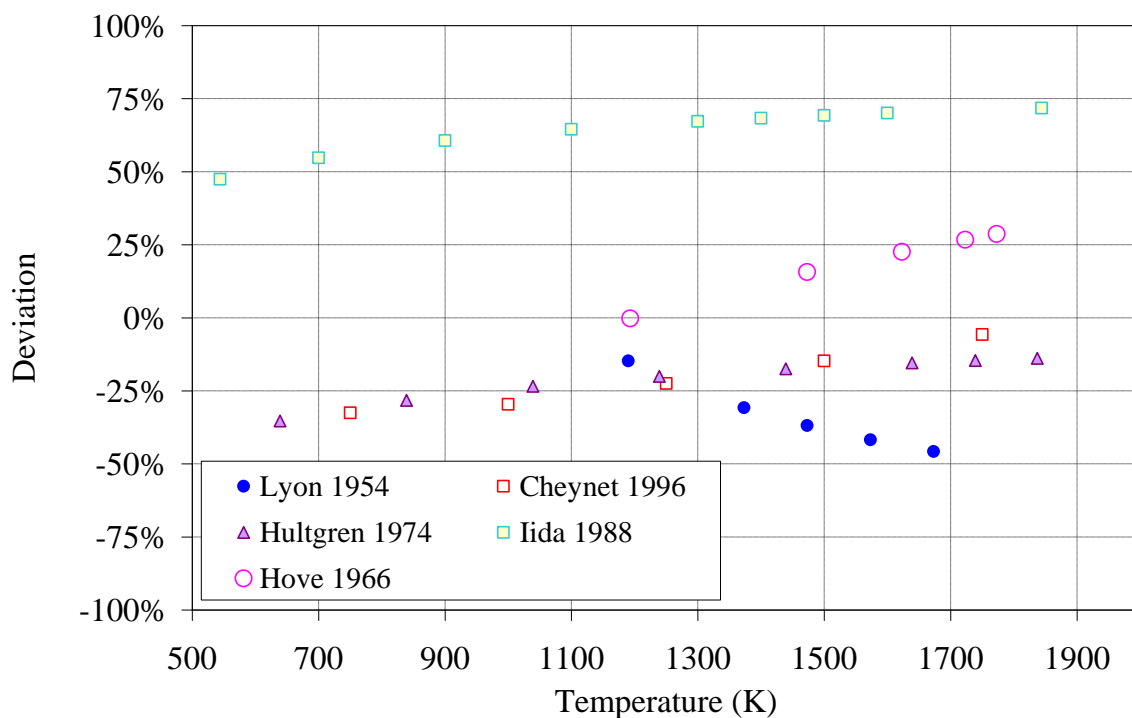

Figure 2.8.5. Saturated vapour pressures of LBE versus temperature
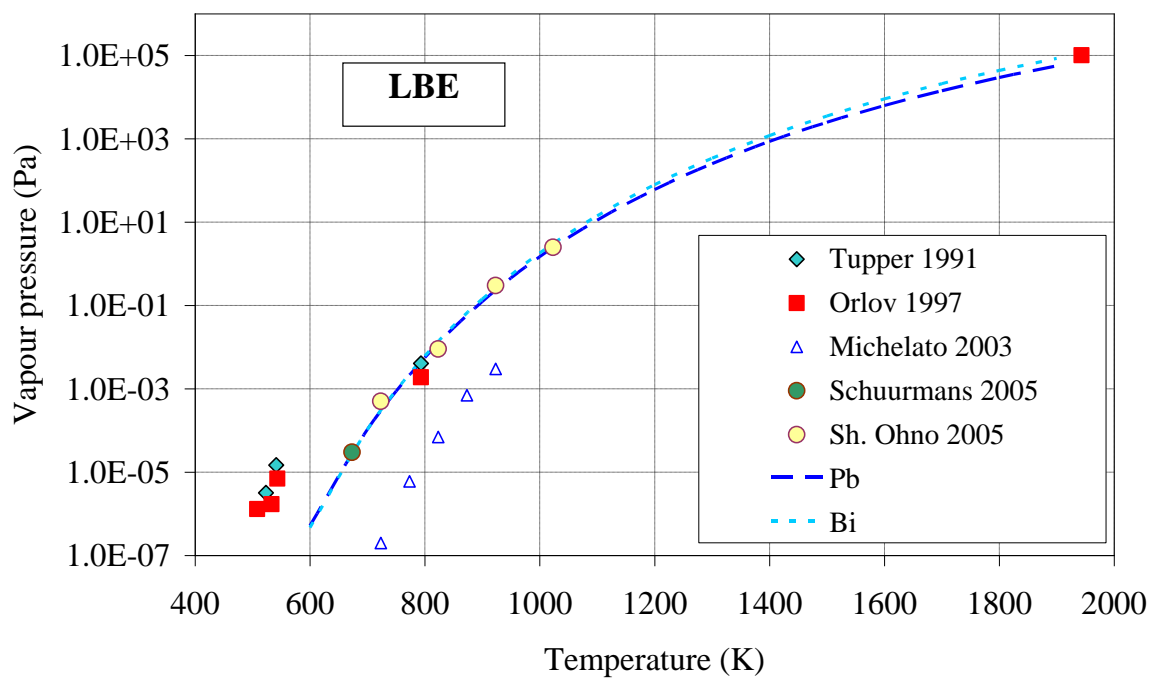
Figure 2.8.5 illustrates that in the temperature range 650-1940 K, the vapour pressure of both lead and bismuth are very close to the experimental values for LBE available from [Tupper, 1991], [Orlov, 1997], [Schuurmans, 2005] and [Ohno, 2005]. The results of [Michelato, 2003] are systematically lower than the body of data, therefore they are not used. At temperatures of 500-550 K (227-277°C), Tupper, *et al*. [Tupper, 1991], and Orlov, *et al*. [Orlov, 1997], give a LBE vapour pressure that is an order of magnitude higher than the liquid-vapour equilibrium lines of lead and bismuth extrapolated to the lower temperatures. However, it should be noticed that at the expected level of saturated pressure (~10^{-8} Pa) it is very difficult to measure the pressure correctly, and overestimation by several orders of magnitude can often be obtained due to contamination of the liquid metal or of the experimental rig. Therefore it is proposed that these points not be considered at present.

The selected data set was used to determine the coefficients for a simplified equilibrium equation (2.14). The values of ΔH and A thus obtained were: $\Delta H = 187.5$ kJ/mol and $A = 11.1 \cdot 10^9$ Pa. The obtained value of ΔH is in a good agreement with the value of the latent heat of LBE evaporation at the normal boiling point recommended in Section 2.7 (178 kJ/mol), taking into account the large uncertainty in the experimental results used for its calculation. The following simplified correlation for the saturation vapour pressure of molten LBE is recommended where temperature is in kelvins:

$$p_{LBE}[Pa] = 1.11 \cdot 10^{10} \cdot \exp(-22552/T) \tag{2.17}$$

This correlation similar to those proposed above for lead and bismuth. The deviation of the selected experimental values from this correlation is illustrated in Figure 2.8.6. The maximum deviation in the temperature range of 673-1943 K (400-1670°C) is about ±60%. (There is no agreement at all at the lower temperatures.) In spite of this significant deviation it was decided to not search for a more sophisticated formula given the very large dispersion of data obtained from different sources.

**Figure 2.8.6. Deviation of the selected data on the saturated
vapour pressures of LBE from the recommended Eq. (2.17)**
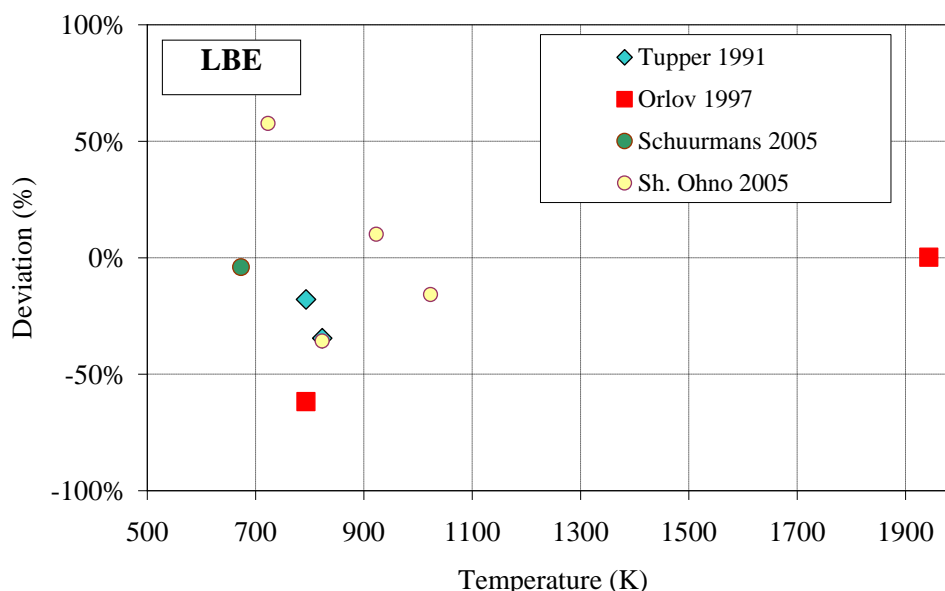
2.9 Surface tension

A surface tension of liquid surfaces (σ) is related to tendency to minimise their surface energy. It decreases with temperature and reduces to zero at the critical temperature (T_c) where difference disappears between liquid and gas phases. According to Eötvös' law [Imbeni, 1998a] for normal liquids this reduction can be described by formula:

$$\sigma = K_\sigma \cdot V_a^{2/3} \cdot \left(T_c - T \right) \tag{2.18}$$

where V_a is the molar volume. The average value of the coefficient K_σ for normal liquid metals is $6.4 \cdot 10^{-8}$ J m^{-2} K^{-1} mol$^{-2/3}$.

B.J. Keene [Keene, 1993] reviewed many data on the surface tension of pure metals and also concluded that the temperature dependence of the surface tension is linear for most liquid metals:

$$\sigma = a + b \cdot T \tag{2.19}$$

2.9.1 Lead

Based on the results of analysis of V. Imbeni, *et al*. [1998a] the following sources were included in the database for the surface tension of molten lead as a function of temperature: [Lyon, 1954], [Friedland, 1966], [Smithells, 1983, 2004], [Lucas, 1984], [Gmelins, 1986], [Jauch, 1986], [Iida, 1988], [Keene, 1993]. Moreover, later data of P.L. Kyrillov and G.P. Bogoslovslaya [Kyrillov, 2000a] and of the IAEA report [TECDOC-1289, 2002] were also included.

The Smithells Metals Reference Book [Smithells, 1983] reproduced a relationship from the book of B.C. Allen [Allen, 1972]. L.D. Lucas, *et al*. [Lucas, 1984] use the correlation proposed by J.C. Joud, *et al*. [Joud, 1972]. Iida, *et al*. [Iida, 1988] reproduce a correlation from the Iida's previous publication [Kasama, 1976]. B.J. Keene [Keene, 1993] reviewed many previous works to obtain values of the coefficients in his linear empirical correlation. The coefficients obtained by him on the basis of all considered sources are very close to those recommended by U. Jauch and B. Schulz [Jauch, 1996]. (However, in his final recommendation B.J. Keene [Keene, 1993] based only on seven sources.) P.L. Kyrillov and G.P. Bogoslovslaya [Kyrillov, 2000a] give a table of the recommended data obtained from different sources. The data recommended by [TECDOC-1289, 2002] are very similar to those presented in [Kyrillov, 2000a] but have slightly higher values.

Figure 2.9.1 shows the values of surface tension of lead taken from the selected sources. Most sources report only empirical coefficients (except [Lyon, 1954], [Kyrillov, 2000a] and [TECDOC-1289, 2002]), with the experimental values being limited to the surface tension in the region of the melting point. A difference of ±5% exists between the surface tension values given by different sources at T < 1000 K (727°C). The lowest values are given by Friedland [Friedland, 1966] and the highest by Smithells [Smithells, 2004]. The values calculated using the correlation given in [Iida, 1988], [Lucas, 1984] and [Jauch, 1986] are very close in this temperature range. At higher temperatures, the scatter increases. The temperature slopes of the different correlations can be to up to ±50%.

The correlation proposed by U. Jauch and B. Schultz [Jauch, 1986] allows a compromise represented by the following formula where temperature is in kelvins:

$$\sigma_{Pb} \left[Nm^{-1} \right] = 0.519 - 1.13 \cdot 10^{-4} \cdot T \tag{2.20}$$

Unfortunately, the temperature of validity is not reported in the literature source. In light of this a conservative validity range of from the melting temperature of 600.6 K to 1200 K (327.6-927°C) is considered acceptable.

Figure 2.9.1. Lead surface tension as a function of temperature
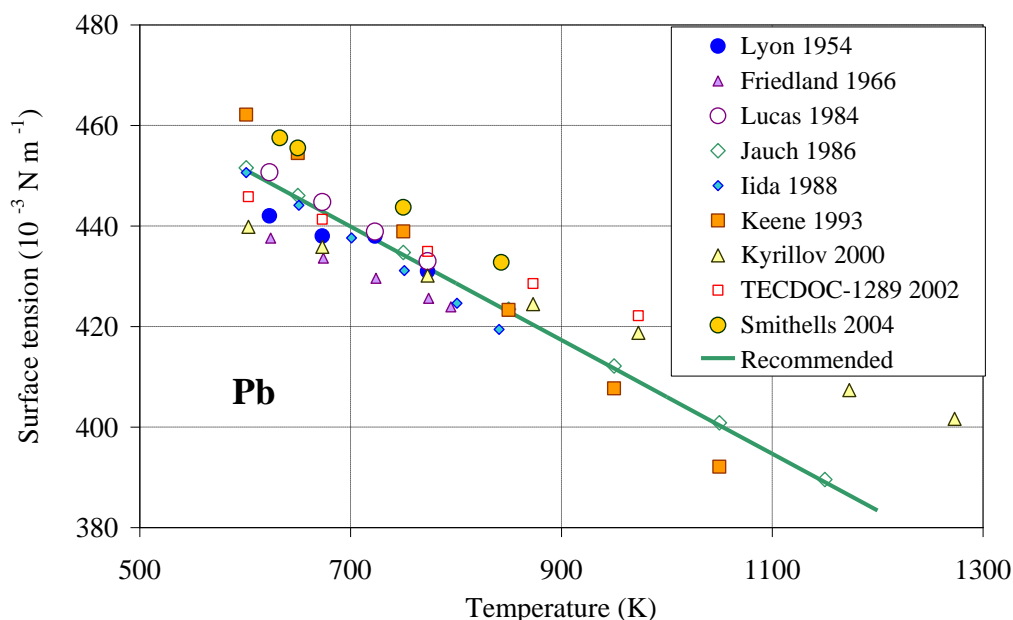
2.9.2 Bismuth

As in the case of lead, the results of analysis of V. Imbeni, *et al*. [1998a] were used as starting point. The data for bismuth surface tension as a function of temperature were taken from [Lyon, 1954], [Howe, 1961], [Friedland, 1966], the Smithells Metals Reference Book [Smithells, 2004], [Lucas, 1984], [Iida, 1988] and [Keene, 1993].

H.E. Howe [Howe, 1961] cited [Lyon, 1954, 1960] and [Smithells, 1955] as main source along with a few older complementary sources. In the last edition of the Smithells Metals Reference Book [Smithells, 2004] a linear correlation from the book of B.C. Allen [Allen, 1972] was reproduced (the same correlation was also reproduced by Iida [Iida, 1988]). Lucas [Lucas, 1984] used the correlation taken from G. Lang, *et al*. [Lang, 1977]. As with lead, B.J. Keene [Keene, 1993] recommended a linear correlation with the coefficients obtained from an analysis of many previous sources (however, in his final recommendation B.J. Keene based only on seven sources). Figure 2.9.2 shows the selected values of surface tension of the molten bismuth in the range of temperature.

The empirical correlations reported by the Smithells Metals Reference Book [Smithells, 1983] and by T. Iida and R.I.L. Guthrie [Iida, 1988] give practically the same values as those of Lyon [Lyon, 1954] and of Howe [Howe, 1961]. The correlation, reported by Lucas [Lucas, 1984] gives 2-3% lower values of the surface tension than other correlations. A good compromise is a correlation obtained by Keene [Keene, 1993] on the basis of all sources analysed by him:

$$\sigma_{Bi}\left[N\,m^{-1}\right] = 0.4255 - 8.0 \cdot 10^{-5} \cdot T \qquad (2.21)$$

where temperature is in kelvins. The expected validity range is T_{melt} -1300 K (1027°C).

Figure 2.9.2. Bismuth surface tension as a function of temperature
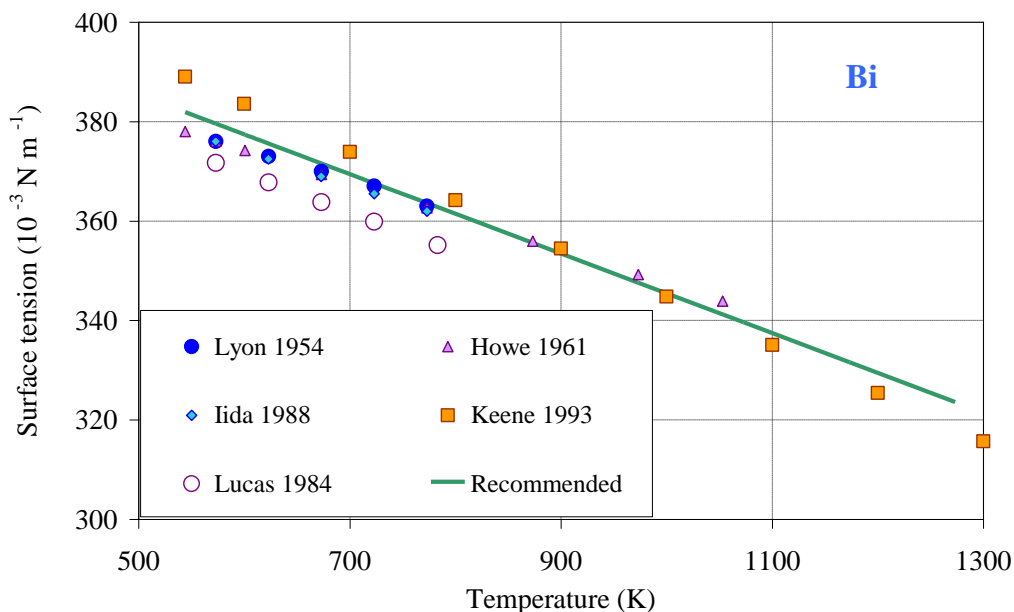
2.9.3 LBE

The sources of data for surface tension of the molten LBE, found in the open literature and included in the present database (Table 2.9.3), are from R.N. Lyon [Lyon, 1954], R.R. Miller [Miller, 1951], V.K. Semenchenko [Semenchenko, 1961], I.V. Kazakova, *et al.* [Kazakova, 1984], R. Novacovic, *et al.* [Novacovic, 2002], D. Giuranno, *et al.* [Giuranno, 2002], and F.C. Pastor Torres [Pastor Torres, 2003]. In the handbook of P.L. Kyrillov and G.P. Bogoslovskaya [Kyrillov, 2002a] incorrect data on the surface tension of molten LBE were given. These data have been corrected by Kyrillov later in [TECDOC-1289]. The last results were also included in the database.

R.R. Miller [Miller, 1951] presented LBE surface tension values at 200, 300 and 400°C. R.N. Lyon [Lyon, 1954] reported only two values: σ = 367 dynes cm^{-1} at T = 800°C, and σ = 356 dynes·cm^{-1} at 1000°C. I.V. Kazakova, *et al.* [Kazakova, 1984] measured surface tension of Pb-Bi melts using the sessile drop method at temperatures up to 800°C (1073 K). R. Novacovic, *et al.* [Novacovic, 2002] and Giuranno [Giuranno, 2002] also used the sessile-drop method to study a molten Pb-Bi alloy with almost eutectic composition (43.9 wt.% Pb) over the temperature range of 623-773 K (350-500°C). F.C. Pastor Torres [Pastor Torres, 2003] recently measured LBE surface tension with the pendent drop method in an inert atmosphere at temperatures from 423 to 573 K (150-300°C). However, the results obtained at T < 473 K (200°C) were discarded because of difficulties related to oxidation. The data from the above cited sources are presented in Table 2.9.1 and Figure 2.9.3.

The deviations for the LBE surface tension data are shown in Figure 2.9.4. The LBE data can be described with an uncertainty of less than 3% by the following liner correlation:

$$\sigma_{LBE}[N\,m^{-1}] = 0.437 - 6.6 \cdot 10^{-5} \cdot T \tag{2.22}$$

where temperature is in kelvins. The above correlation is recommended for estimation of the surface tension of molten LBE at normal pressure up to 1400 K (1127°C).

Table 2.9.1. LBE surface tension database

Reg. no.	Parameter	Measurement method	Estimated accuracy %	Temperature range, K	Pressure range, Pa	Values	Ref.
9.3.1	Surface tension	?	?	1073, 1273	~ 10^5 (?)		[Lyon, 1954]
	Interpolation function: $\sigma = (426.02 - 0.055 \cdot T) \cdot 10^{-3} \, Nm^{-1}$						
	Comments:						
9.3.2	Surface tension	Sessile drop	?	623-773	~ 10^5 (?)		[Novacovic, 2002]
	Interpolation function: $\sigma = (445.419 - 0.0801 \cdot T) \cdot 10^{-3} \, Nm^{-1}$						
	Comments: 43.9 wt.% Pb + 56.1 wt.% Bi. Unknown purity.						
9.3.3	Surface tension	Pendent drop	?	423-573	~ 10^5 (?)		[Pastor Torres, 2003]
	Interpolation function: $\sigma = (450.86 - 0.0873 \cdot T) \cdot 10^{-3} \, Nm^{-1}$						
	Comments: 44.5 wt.% Pb + 55.5 wt.% Bi. Unknown purity.						
9.3.4	Surface tension	?	?	1073-1373	~ 10^5 (?)		[Semenchenko, 1961]
	Interpolation function: $\sigma = (421.26 - 0.0665 \cdot T) \cdot 10^{-3} \, Nm^{-1}$						
	Comments: ?						
9.3.5	Surface tension	?	?	473?-673	~ 10^5(?)		[Miller, 1951]
	Interpolation function: $\sigma = (421.26 - 0.0665 \cdot T) \cdot 10^{-3} \, Nm^{-1}$						
	Comments: ?						
9.3.6	Surface tension	Sessile drop	?	???-1073	~ 10^5(?)		[Kazakova, 1984]
	Interpolation function: $\sigma = (415.7 - 0.047 \cdot T) \cdot 10^{-3} \, Nm^{-1}$						
	Comments: 43.3 wt.% Pb + 56.7 wt.% Bi. Unknown purity.						
9.3.7	Surface tension	?	?	403-1073	~ 10^5(?)		[TECDOC-1289, 2002]
	Interpolation function: $\sigma = (444.0 - 0.0703 \cdot T) \cdot 10^{-3} \, Nm^{-1}$						
	Comments: 44.5 wt.% Pb + 55.5 wt.% Bi.						

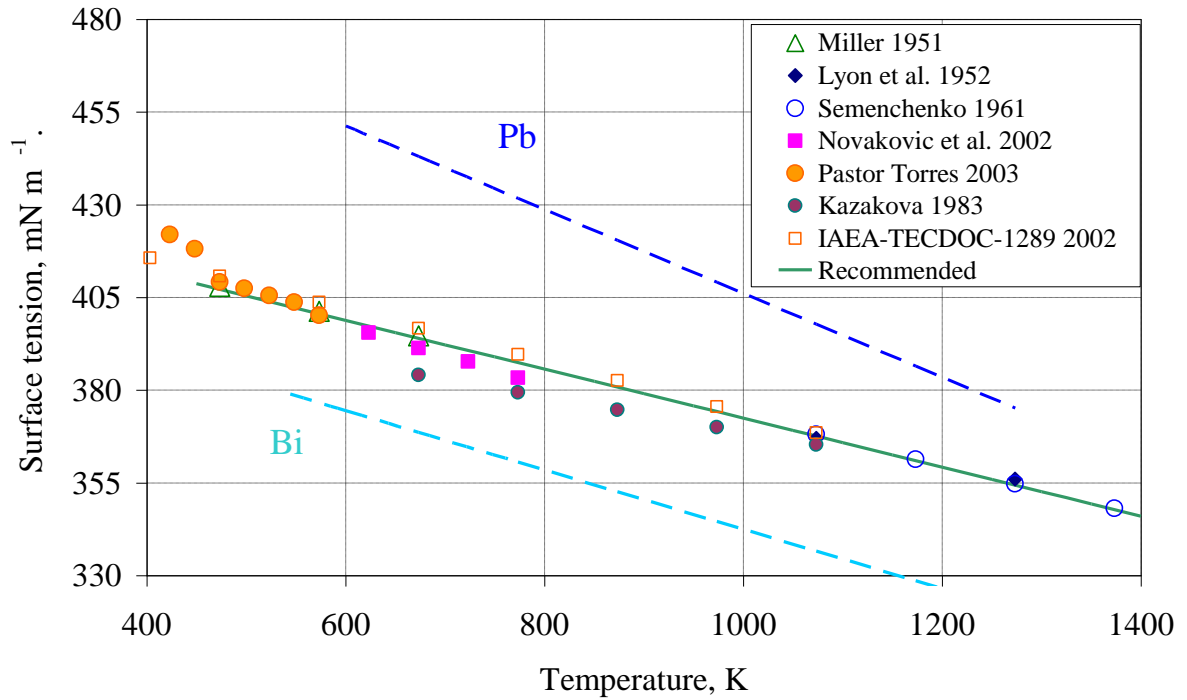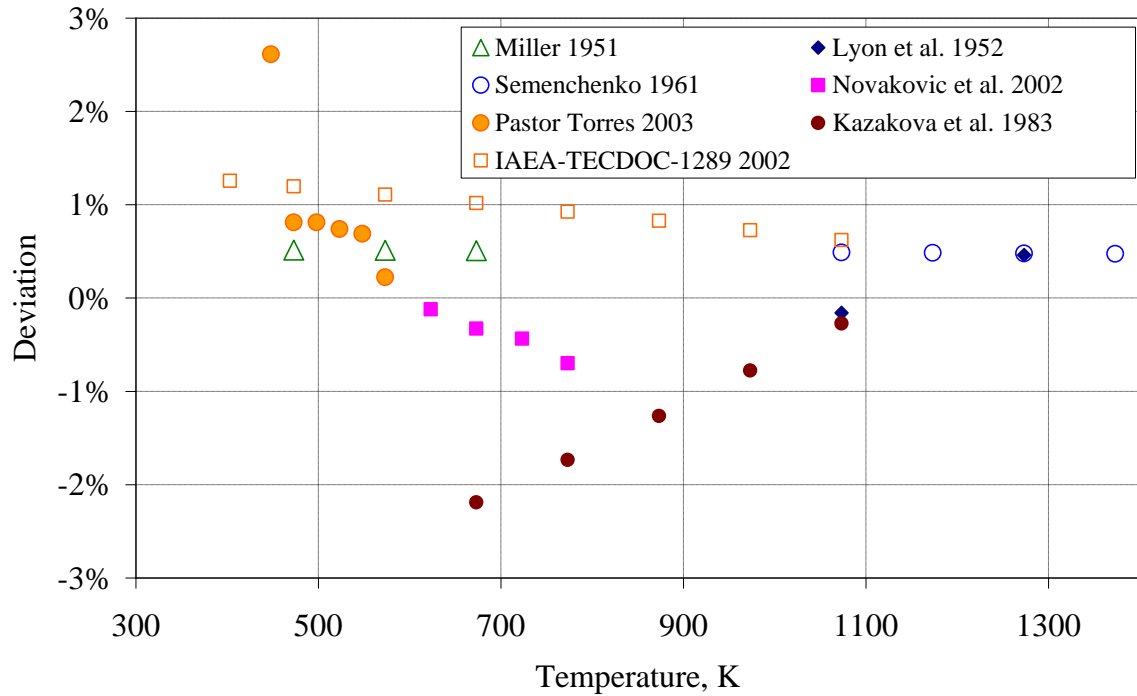**Figure 2.9.3. Surface tension of the molten LBE**

**Figure 2.9.4. Deviation of the source data on surface tension
of molten LBE from the recommended correlation (2.21)**

2.10 Density

The temperature dependence of density provides essential information for the development of an equation of state (EOS). In engineering practice it is used to determine the concentration of atoms in unit volume and hydraulic parameters in installations. Also, the measurement or calculation of basic physical properties, e.g. viscosity, surface tension, thermal diffusivity, requires knowledge of density.

2.10.1 Lead

The data for the density of the molten lead from R.N. Lyon [Lyon, 1954], S.S. Kutateladze, *et al*. [Kutateladze, 1959], A.D. Kirshenbaum, *et al*. [Kirshenbaum, 1961], L.E. Crean and W.E. Parkins [Crean, 1964], Ruppersberg and Speicher [Ruppersberg, 1976], [Smithells, 1983, 2004], L.D. Lucas [Lucas, 1984a], T. Iida and R.I.L. Guthrie [Iida, 1988] (analysed also by V. Imbeni, *et al*. [Imbeni, 1998a]), and more recent compilations of P.I. Kyrillov, *et al*. [Kyrillov, 2000a], [TECDOC-1289, 2002] were included in the present handbook.

A.D. Kirshenbaum, *et al*. [Kirshenbaum, 1961] presented the experimental results on the molten lead density up to the melting temperature. Smithells [Smithells, 1983] provides recommendations on the molten lead density based on the data taken from [Kirshenbaum, 1961] and from [Strauss, 1960]. Lucas [Lucas, 1984a] reported values found by the author experimentally earlier [Lucas, 1969]. Iida [Iida, 1988] uses a correlation recommended by Steinberg [Steinberg, 1974].

The recommendations of Friedland [Friedland, 1966] and Weeks [Weeks, 1971] have not been taken into account because they repeat the results of [Smithells, 1955] and [Lyon, 1954] already included in the database.

The selected database for density of molten lead is presented in Table 2.10.1.

Figure 2.10.1 shows the molten lead density values from the selected sources versus temperature.

A set of the selected data can be described by linear temperature dependence where temperature is in kelvins:

$$\rho_{Pb}\left[kg\ m^{-3}\right] = 11367 - 1.1944 \cdot T \tag{2.23}$$

The deviation of the selected source values from Eq. (2.23) is illustrated in Figure 2.10.2. One can see that maximum difference does not exceed 0.7%.

Table 2.10.1. Database on the density of molten lead at the normal pressure

Reg. no.	Parameter	Measurement method	Estimated accuracy	Temperature range, K	Pressure range, Pa	Values	Ref.
10.1.1	Density	?	?	673-1273	$\sim 10^5$ (?)		[Lyon, 1954]
	Interpolation function: $\rho = 11291 - 1.165 \cdot T$ kg m^{-3}						
	Comments: Unknown purity.						
10.1.2	Density	?	?	673-1073	$\sim 10^5$ (?)		[Kutateladze, 1959]
	Interpolation function: $\rho = 11380 - 1.169 \cdot T$ kg m^{-3}						
	Comments: Unknown purity.						

Table 2.10.1. Database on the density of molten lead at the normal pressure (*cont.*)

Reg. no.	Parameter	Measurement method	Estimated accuracy	Temperature range, K	Pressure range, Pa	Values	Ref.
10.1.3	Density	?	?	601-1345	~ 10^5 (?)		[Crean, 1964]
	Interpolation function: $\rho = 11307 - 1.177 \cdot T$ kg m^{-3}						
	Comments: Unknown purity.						
10.1.4	Density	?	?	?	~ 10^5 (?)		[Smithells, 1983]
	Interpolation function: $\rho = 11470 - 1.3174 \cdot T$ kg m^{-3}						
	Comments: Unknown purity.						
10.1.5	Density	?	?	601-1823	~ 10^5 (?)		[Lucas, 1984]
	Interpolation function: $\rho = 11418 - 1.301 \cdot T + 6.278 \cdot 10^{-5} \cdot T^2$ kg m^{-3}						
	Comments: Unknown purity.						
10.1.6	Density	?	?	601-2000	~ 10^5 (?)		[Iida, 1988]
	Interpolation function: $\rho = 10670 - 1.32 \cdot (T - T_{melt})$ kg m^{-3}						
	Comments: Unknown purity.						
10.1.7	Density	?	?	603-1273	~ 10^5 (?)		[TECDOC-1289, 2002]
	Interpolation function: $\rho = 11417 - 1.239 \cdot T$ kg m^{-3}						
	Comments: Unknown purity.						
10.1.8	Density	?	?	636-829	~ 10^5 (?)		[Ruppersber, 1976]
	Interpolation function: $\rho = 11417 - 1.350 \cdot T$ kg m^{-3}						
	Comments: Unknown purity.						

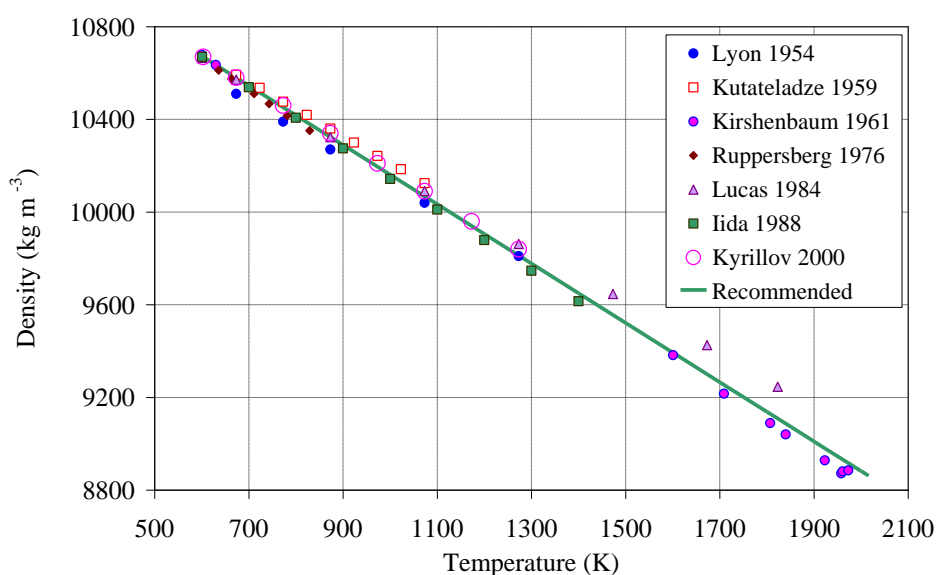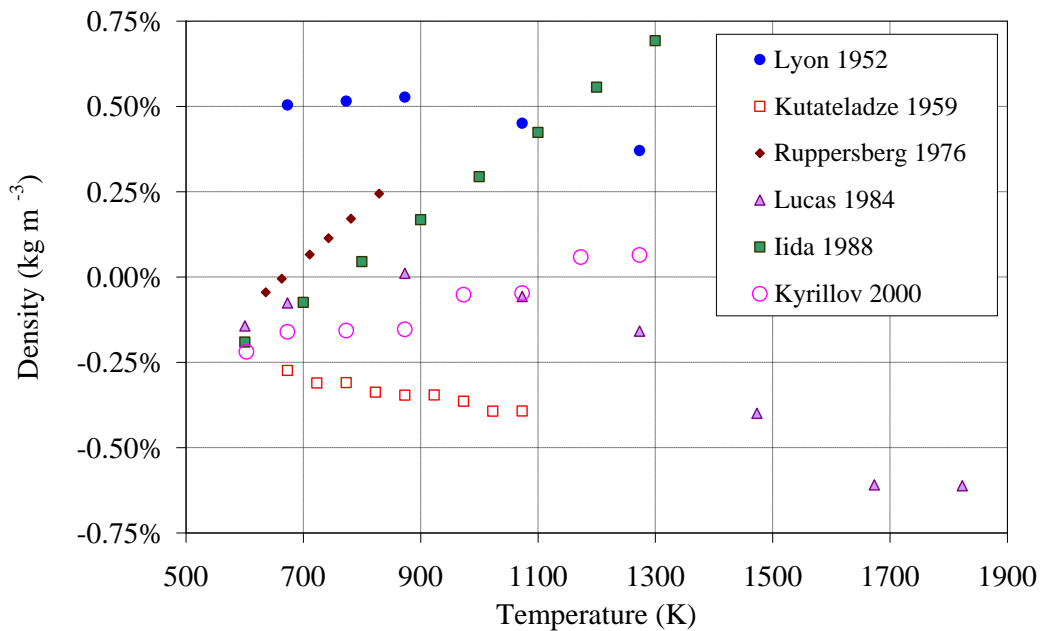**Figure 2.10.1. Density of molten lead as function of temperature at normal pressure**

Figure 2.10.2. Deviation of the database values on the lead density from Eq. (2.23)

2.10.2 Bismuth

The data for molten bismuth density that have been included in the database, were taken from [Lyon, 1954], [Kutateladze, 1959], [Crean, 1964], [Cahil, 1963], [Bonilla, 1964], [Lucas, 1984] and [Iida, 1988].

Data from Smithells Metal Reference Book [Smithells, 1983], Friedland [Friedland, 1966] and Weeks [Weeks, 1971] have not been included in the database because they reproduce the results already presented in the above cited sources. Bonilla [Bonilla, 1964] reported values that were obtained by interpolation of data collected from a number of sources. Iida and Guthrie [Iida, 1988] reproduce the recommendations of Steinberg [Steinberg, 1974].

The selected database on the density for molten bismuth is presented in Table 2.10.2.

Figure 2.10.3 shows the molten bismuth density values from the selected sources versus temperature.

All sets of the selected data can be described with linear temperature dependence recommended by V. Imbeni, *et al.* [Imbeni, 1998a]:

$$\rho_{Bi}\left[kg\ m^{-3}\right] = 10726 - 1.2208 \cdot T \tag{2.24}$$

where temperature is in kelvins.

The deviation of the selected source values from Eq. (2.24) is illustrated in Figure 2.10.4. One can see that maximum difference does not exceed 0.4%.

Table 2.10.2. Database on the density of the molten bismuth at the normal pressure

Reg. no.	Parameter	Measurement method	Estimated accuracy	Temperature range, K	Pressure range, Pa	Values	Ref.
10.2.1	Density	?	?	573-1235	$\sim 10^5$ (?)		[Lyon, 1954]
	Interpolation function: $\rho = 10755 - 1.258 \cdot T$ kg m^{-3}						
	Comments: Unknown purity.						
10.2.2	Density	?	?	553-973	$\sim 10^5$ (?)		[Kutateladze, 1959]
	Interpolation function: $\rho = 10744 - 1.243 \cdot T$ kg m^{-3}						
	Comments: Unknown purity.						
10.2.3	Density	?	?	589-1033	$\sim 10^5$ (?)		[Bonilla, 1964]
	Interpolation function: $\rho = 10722 - 1.211 \cdot T$ kg m^{-3}						
	Comments: Unknown purity.						
10.2.4	Density	?	?	566-1266	$\sim 10^5$ (?)		[Crean, 1964]
	Interpolation function: $\rho = 10700 - 1.3174 \cdot T$ kg m^{-3}						
	Comments: Unknown purity.						
10.2.5	Density	?	?	673-1273	$\sim 10^5$ (?)		[Lucas, 1984]
	Interpolation function: $\rho = 10714 - 1.1986 \cdot T$ kg m^{-3}						
	Comments: Unknown purity.						
10.2.6	Density	?	?	601-2000	$\sim 10^5$ (?)		[Iida,1988]
	Interpolation function: $\rho = 10050 - 1.18 \cdot \left(T - T_{melt}\right)$ kg m^{-3}						
	Comments: Unknown purity.						

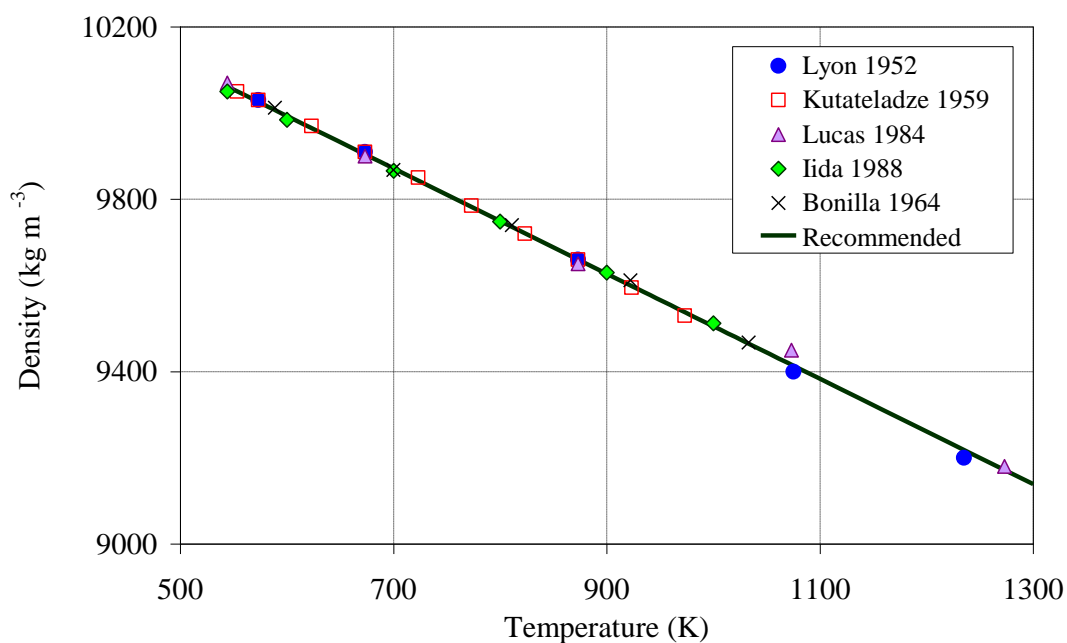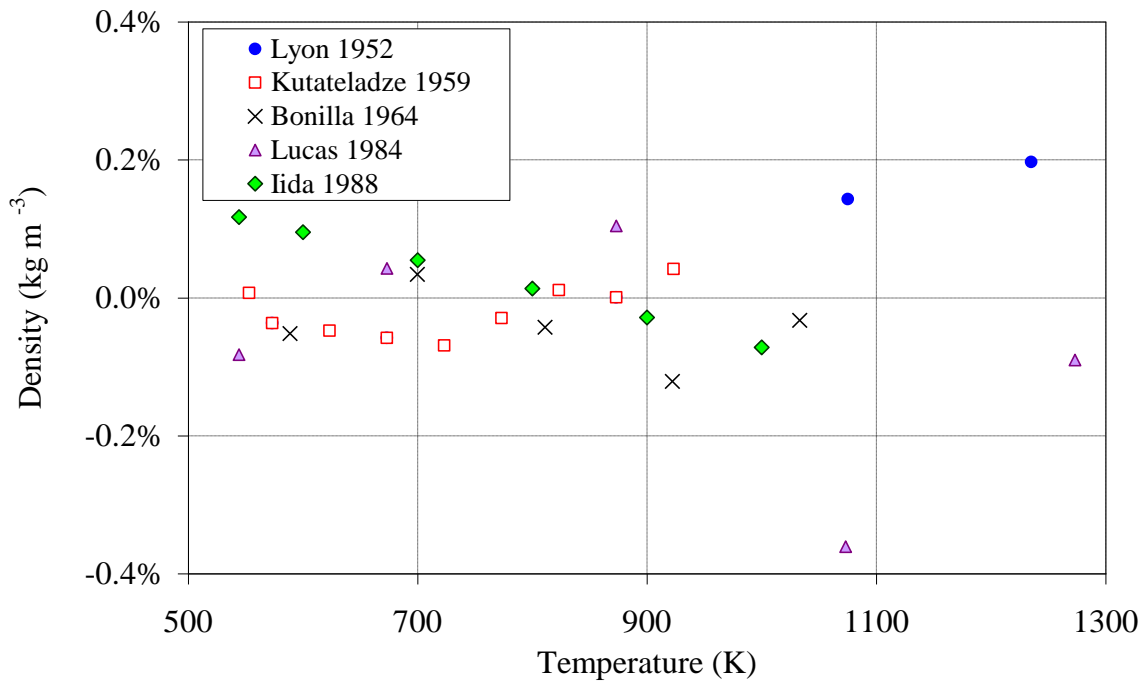**Figure 2.10.3. Density of molten bismuth as a function of temperature at normal pressure**

Figure 2.10.4. Deviation of the database values on the bismuth density from Eq. (2.24)

2.10.3 LBE

Only data that do not repeat one another have been included in the database for molten LBE presented in Table 2.10.3. The data included are from R.N. Lyon [Lyon, 1954], S.S. Kutateladze [Kutateladze, 1959], the IAEA report [TECDOC-1289, 2002] and B.B. Alchagirov [Alchagirov, 2003].

The data presented by C.F. Bonilla [Bonilla, 1964], Crean [Crean, 1964] and Hofman [Hofmann, 1970] were obtained by the interpolation of the results of Lyon [Lyon, 1954, 1956] and were therefore not included in the database.

The extracted density values as a function of temperature at normal atmospheric pressure are presented in Figure 2.10.5. The temperature dependence of the densities of pure molten lead and pure molten bismuth, calculated with the empirical correlations recommended in preceding sections are also reproduced for comparison.

The linear regression of values presented in Figure 2.10.5 yields for the LBE density:

$$\rho_{LBE}\left[kg\ m^{-3}\right] = 11096 - 1.3236 \cdot T \tag{2.25}$$

where the temperature is in kelvins.

Figure 2.10.6 shows that the deviation of the selected literature data for molten LBE density from Eq. (2.25) does not exceed 0.8%.

Table 2.10.3. LBE density database

Reg. no.	Parameter	Measurement method	Estimated accuracy	Temperature range, K	Pressure range, Pa	Values	Ref.
10.3.1	Density	?	?	473-1273	$\sim 10^5$ (?)		[Lyon, 1954]
	Interpolation function: $\rho = 11113 - 1.375 \cdot T$ kg m^{-3}						
	Comments: Unknown data on eutectic composition and purity.						
10.3.2	Density	Picnometer	2%	403-973	$\sim 10^5$ (?)		[Kutateladze, 1959]
	Interpolation function: $\rho = 11062 - 1.2175 \cdot T$ kg m^{-3}						
	Comments: 44.5 wt.% Pb + 55.5 wt.%Bi. Unknown purity.						
10.3.3	Density	Picnometer	?	403-1073	$\sim 10^5$ (?)		[TECDOC-1289, 2002]
	Interpolation function: $\rho = 11047 - 1.2564 \cdot T$ kg m^{-3}						
	Comments: 44.5 wt.% Pb + 55.5 wt.% Bi. Unknown purity.						
10.3.4	Density	Picnometer	0.1%	410-726	$\sim 10^5$ (?)		[Alchagirov, 2003]
	Interpolation function: $\rho = 10981.7 - 1.1369 \cdot T$ kg m^{-3}						
	Comments: 44.6 wt.% Pb + 55.4 wt.% Bi. Purity Pb – 99.9985 wt.%. Purity Bi – 99.98 wt.%						

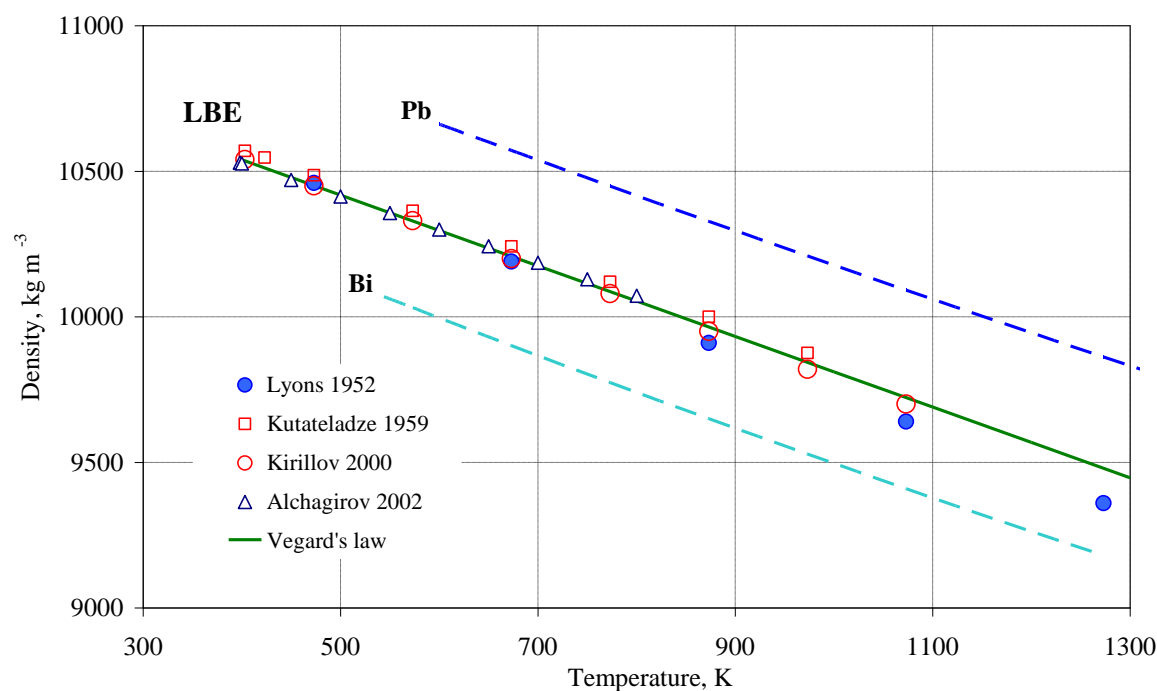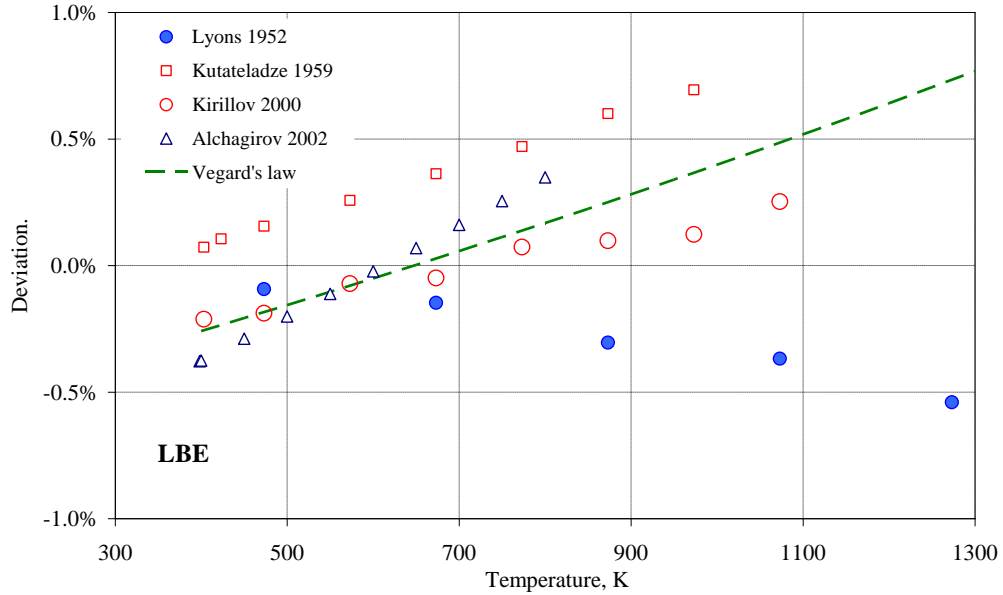**Figure 2.10.5. Density of the melted LBE versus temperature**

**Figure 2.10.6. Deviation of the literature recommendations
for melted LBE density from the Eq. (2.25)**

In Figures 2.10.5 and 2.10.6, the results of calculation of the LBE density using Vegard's law which expresses the molar volume of LBE through the molar fractions ($x_{\mu\,Pb}$ and $x_{\mu\,Bi}$) and the molar volumes ($v_{\mu\,Pb}$ and $v_{\mu\,Bi}$) of lead and bismuth as follows:

$$V_{\mu\,LBE} = x_{\mu\,Pb} \cdot V_{\mu\,Pb} + x_{\mu\,Bi} \cdot V_{\mu\,Bi} \tag{2.26}$$

are also presented.

Eq. (2.26), transformed to the following form:

$$\rho_{LBE} = \frac{\mu_{LBE}}{\dfrac{x_{\mu Pb} \cdot \mu_{Pb}}{\rho_{Pb}} + \dfrac{\left(1 - x_{\mu Pb}\right) \cdot \mu_{Bi}}{\rho_{Bi}}} \tag{2.27}$$

(where μ_{Pb}, μ_{Bi} and μ_{LBE} are the molar masses of Pb, Bi and LBE respectively) allows one to calculate the LBE density using as input the densities of pure molten lead and pure molten bismuth which can be predicted with better precision than that of LBE.

From Figures 2.10.5 and 2.10.6 one can see that the Vegard's law can be used for calculation of the LBE density as a good approximation. In spite the fact that the Vegard's law can not be applied to LBE at temperatures lower than highest melting point of the component, it gives very good results from the LBE melting temperature to at least 1300 K (1027°C) if one extrapolates the correlations for the densities of the melted Pb and Bi down to $T_{melt\,LBE}$.

2.11 Thermal expansion

The density of liquid metals changes with temperature due to thermal expansion related to anharmonicity of interatomic forces. In a general case, a link between the density and the coefficient

of volumetric thermal expansion at any constant thermodynamic parameter x is defined by the following formula:

$$\beta_x(T) \equiv \frac{1}{V} \cdot \left(\frac{\partial V}{\partial T}\right)_x = -\frac{1}{\rho} \cdot \left(\frac{\partial \rho}{\partial T}\right)_x \qquad (2.28)$$

Literature sources describing a direct measurement of the coefficient of thermal expansion (CTE) of molten Pb, Bi and LBE were not found. Therefore, available information on the temperature dependence of their density (see Section 2.10) was used for deduction of the isobaric CTE. Substituting the correlation for density recommended in Section 2.10 into definition (2.28) yields the following formulae for the isobaric volumetric CTE of molten lead, bismuth and LBE respectively:

a) Lead:

$$\beta_{pPb}\left[K^{-1}\right] = \frac{1}{9516.9 - T} \qquad (2.29)$$

b) Bismuth:

$$\beta_{pBi}\left[K^{-1}\right] = \frac{1}{8786.2 - T} \qquad (2.30)$$

c) LBE:

$$\beta_{pLBE}\left[K^{-1}\right] = \frac{1}{8383.2 - T} \qquad (2.31)$$

The results obtained are presented in Figure 2.11.1. Comparison of the isobaric volumetric CTE of molten lead, bismuth and LBE, obtained with the formulae above shows that LBE has the highest CTE. In principle, this can be explained by the lower attractive forces between Pb and Bi atoms in LBE than in pure lead and in pure bismuth. However, it could also be due to a large uncertainty of the determination of the thermal expansion coefficient by using the procedure of differentiation of the density correlations.

Eqs. (2.29), (2.30) and (2.31) are recommended for practical applications as "the best choice" (because of their consistency with the correlations recommended above in Section 2.10) for calculation of the CTE of molten lead, bismuth and LBE respectively.

2.12 Sound velocity and compressibility

There are no results for direct measurement of the compressibility for molten lead, bismuth and LBE in the literature. Usually, the adiabatic compressibility K_S (or the adiabatic elastic modulus B_S) is found from the results of measurement of the sound velocity u_{sound} and density ρ using the following thermodynamic relationship [Dreyfus, 1971]:

$$B_S \equiv -V\left(\frac{\partial p}{\partial V}\right)_S \equiv \frac{1}{K_S} = \frac{\rho}{\left(\frac{\partial \rho}{\partial p}\right)_S} = \rho \cdot u_{sound}^2 \qquad (2.32)$$

**Figure 2.11.1. Volumetric thermal expansion coefficients of
molten lead, bismuth and LBE in function of temperature**
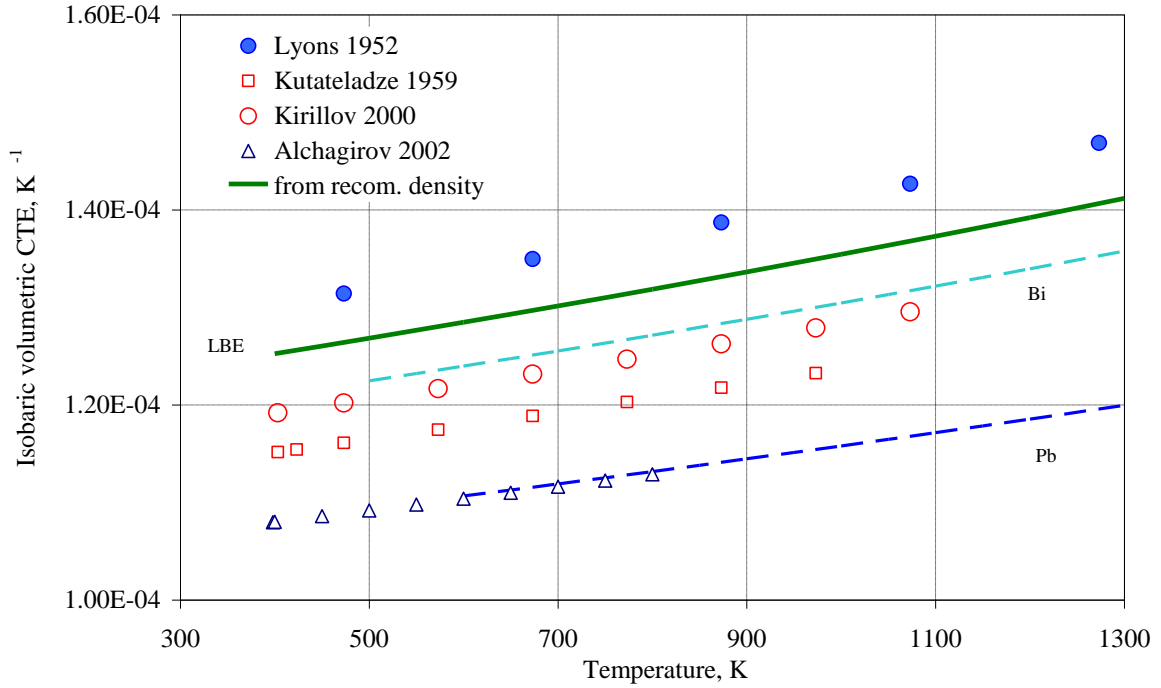
2.12.1 Lead

The velocity of propagation of longitudinal sound waves in molten lead was measured by
O.J. Kleppa [Kleppa, 1950] close to the melting point, by R.B. Gordon [Gordon, 1959] over a
temperature interval from the melting temperature to 643 K (370°C), by G.M. Mustafin and
G.F. Shailhiev [Mustafin, 1983] over the temperature range 601-2000 K and by G.V. Konyuchenko
[Konyuchenko, 1969] over the temperature range 673-973 K (400-700°C). Rather good agreement is
observed among the first three sources in the region of the melting temperature. The sound velocity
values presented by Konyuchenko [Konyuchenko, 1969] are 3% higher. No information was given on
Pb purity and on the measurement method used. Therefore this source was not taken into account in
the database presented in Table 2.12.1 below. The data are plotted in Figure 2.12.1.

The most complete and reliable data seems to be that from Mustafin and [Mustafin, 1983],
therefore the correlation recommended for the estimation of the sound velocity in the molten lead is:

$$u_{sound\,Pb}\left[m\,s^{-1}\right]=1951.75 - 0.3423\cdot T + 7.635\cdot 10^{-5}\cdot T^2 \qquad (2.33)$$

where temperature is in kelvins.

2.12.2 Bismuth

The sound velocity in molten bismuth was measured by J. Jarzynski [Jarzynski, 1963] from the
melting temperature up to 603 K (330°C). This work has been included in the database shown in
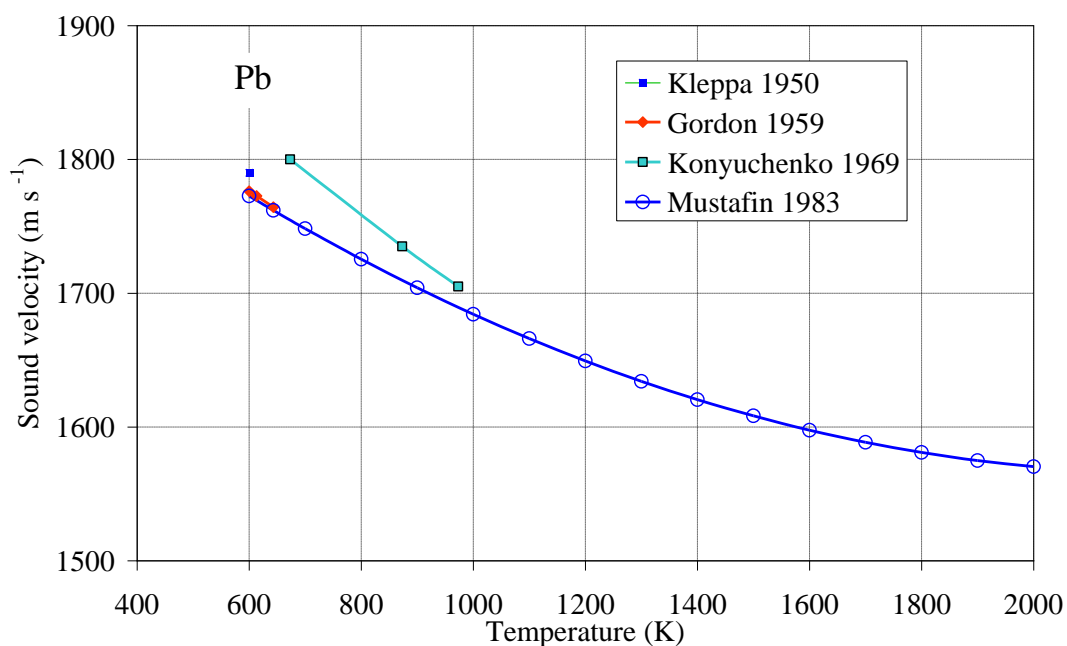Table 2.12.2.

Table 2.12.1. Sound velocity in the molten lead

Reg. no.	Parameter	Measurement method	Estimated accuracy %	Temperature range, K	Pressure range, Pa	Values	Reference
12.1.1	Sound velocity	Pulse propagation	0.01	601-643	$\sim 10^5$ (?)		[Gordon, 1959]
	Interpolation function: $u_{sound}(T) = 1776 - 0.277 \cdot (T - T_{melt})$ m s^{-1}						
	Comments: Lead of technical purity.						
12.1.2	Sound velocity	Pulse propagation	0.005	601-2000	$\sim 10^5$ (?)		[Mustafin, 1983]
	Interpolation function: $u_{sound}(T) = 1951.75 - 0.3423 \cdot T + 7.635 \cdot 10^{-5} \cdot T^2$ m s^{-1}						
	Comments: Pure lead.						
12.1.3	Sound velocity	?	?	600.2 ?	$\sim 10^5$ (?)	1790 m/s	[Kleppa, 1950]
	Interpolation function: n/a						
	Comments: Unknown purity.						

Figure 2.12.1. Velocity of ultrasound in the molten lead in function of temperature

Table 2.12.2. Sound velocity in the molten bismuth

Reg. no.	Parameter	Measurement method	Estimated accuracy %	Temperature range, K	Pressure range, Pa	Values	Reference
12.2.1	Sound velocity	Pulse propagation	?	545-603	$\sim 10^5$ (?)		[Jarzynski, 1963]
	Interpolation function: $u_{sound} = 2111.3 - 0.7971 \cdot T$ m s^{-1}						
	Comments: Lead of technical purity.						

In the publication of Jarzynski [Jarzynski, 1969] it was proposed that the temperature dependence of the sound velocity in molten bismuth followed a linear correlation:

$$u_{sound\,Bi}\left[m\ s^{-1}\right] = 2111.3 - 0.7971 \cdot T \qquad (2.34)$$

where temperature is in kelvins.

2.12.3 LBE

The ultrasound velocity in molten LBE has been measured by R. Kažys [Kažys, 2002] over the temperature range 433-603 K (160-330°C). The results of this publication have been included in the database in Table 2.12.3. A-M. Azad [Azad, 2005] calculated the sound velocity in molten LBE based on Vegard's law and using the experimental data on the sound velocity in lead [Mustafin, 1969] and in bismuth [Jarzynski, 1963]. The correlation recommended by Azad [Azad, 2005] has also been included in the database.

Table 2.12.3. Sound velocity of the melted LBE

Reg. no.	Parameter	Measurement method	Estimated accuracy %	Temperature range, K	Pressure range, Pa	Values	Ref.
12.3.1	Sound velocity	Direct propagation	0.1	433-603	$\sim 10^5$ (?)		[Kažys, 2002]
	Interpolation function: $u_{sound\,LBE}(T) = 1773 + 0.1049 \cdot T - 2.873 \cdot 10^{-4} \cdot T^2$ m s^{-1}						
	Comments: 44.5 wt.% Pb + 55.5 wt.% Bi. Unknown purity.						
12.3.2	Sound velocity	–	?		$\sim 10^5$ (?)		[Azad, 2005]
	Interpolation function: $u_{sound\,LBE}(T) = 2041.58 - 0.5987 \cdot T + 3.3387 \cdot 10^{-5} \cdot T^2$ m s^{-1}						
	Comments: 44.5 wt.% Pb + 55.5 wt.% Bi. Unknown purity.						

The values for the sound velocity in LBE are plotted in Figure 2.12.2 together with plots for the recommended correlations for lead and bismuth.

The Vegard's law points are extrapolated to the melting temperature of LBE. From Figure 2.12.2 one can see that difference between the experimental data on the sound velocity and those obtained with the Vegard's law is less then 2% in the region of the melting temperature of lead. However, the slope of the temperature dependence differs significantly (more than twice).

While the present data is very scattered and, the following correlation for the LBE sound velocity is based on the experimental results of Kazys [Kazys, 2002]:

$$u_{sound\,LBE}\left[m\ s^{-1}\right] = 1773 + 0.1049 \cdot T - 2.873 \cdot 10^{-4} \cdot T^2 \qquad (2.35)$$

where temperature is in kelvins.

Figure 2.12.2. Sound velocity in molten LBE, Pb and Bi versus temperature
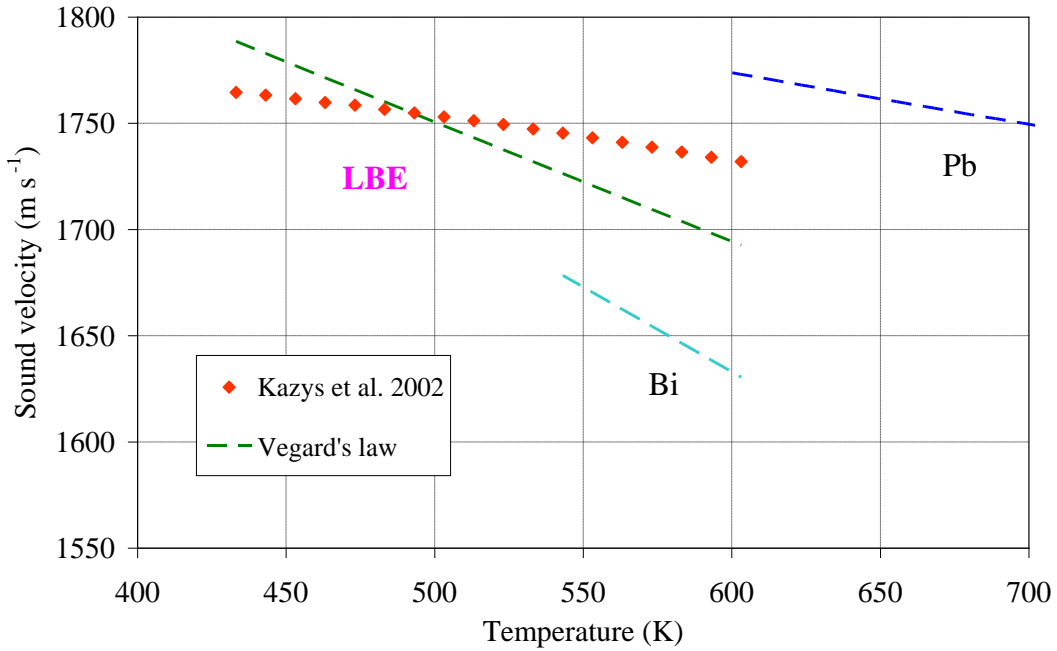
The results of the calculations for elastic modulus of molten lead, bismuth and LBE obtained with Eq. (2.32) are presented in Figure 2.12.3.

In the respective temperature range and at normal atmospheric pressure, the temperature dependence of the elastic modulus can be described with the help of parabolic and linear functions.

a) Lead:

$$B_{s\,Pb}[Pa] = \left(42.15 - 1.652 \cdot 10^{-2} \cdot T + 3.273 \cdot 10^{-6} \cdot T^2\right) \cdot 10^9 \tag{2.36}$$

b) Bismuth:

$$B_{s\,Bi}[Pa] = \left(44.67 - 0.0299 \cdot T\right) \cdot 10^9 \tag{2.37}$$

c) LBE:

$$B_{s\,LBE}[Pa] = \left(35.18 - 1.541 \cdot 10^{-3} \cdot T - 9.191 \cdot 10^{-6} \cdot T^2\right) \cdot 10^9 \tag{2.38}$$

where temperatures are in kelvins.

2.13 Heat capacity

Available experimental data on heat capacity of heavy liquid metals are even less numerous than those for density. The theoretical calculation of heat capacity of liquids is restricted by the extreme complexity of the motion of atoms in liquids. Often, the same types of empirical correlations as for solids are used to fit the experimental results. An extensive review of the existing experimental data on the heat capacity of lead, bismuth and LBE was performed by V. Imbeni, *et al*. [Imbeni, 1998a, 1998b].

Figure 2.12.3. Bulk elastic modulus of molten LBE versus temperature
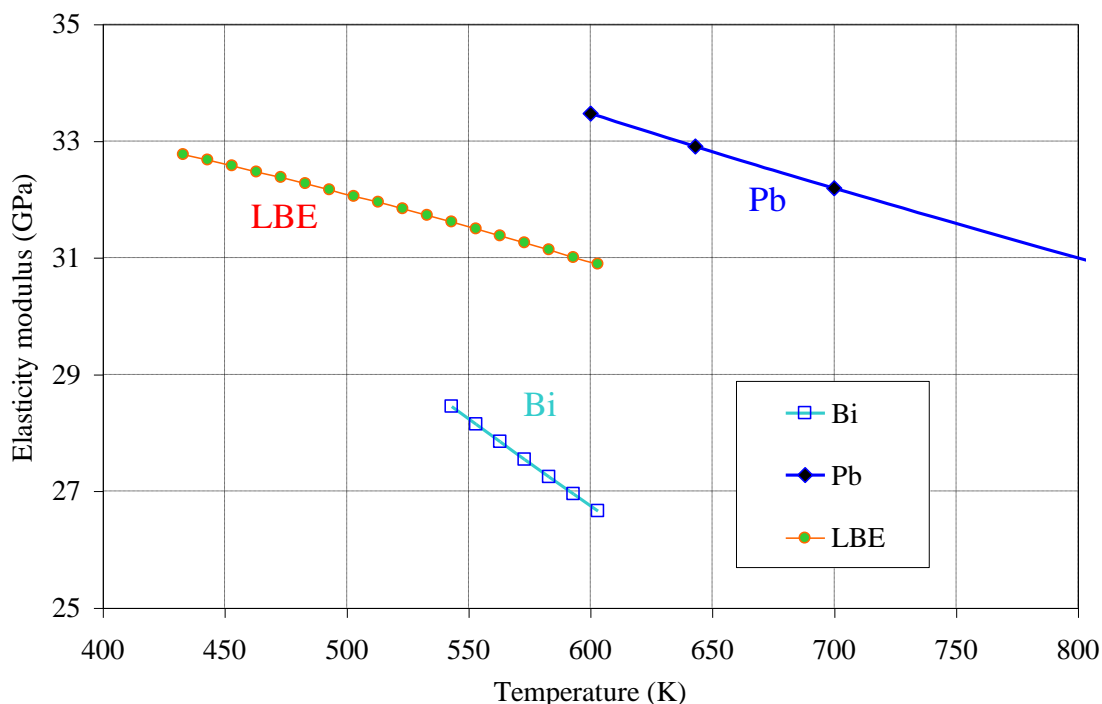
2.13.1 Lead

The heat capacity data of lead taken from [Lyon, 1954], [Kutateladze, 1959], [Friedland, 1966], [Hultgren, 1974], [Kubashewski, 1979, 1993], [Iida, 1988] and [Gurvich, 1991] have been analysed to include in the current database.

Friedland [Friedland, 1966] used data of T.B. Douglas, J.L. Dever [Douglas, 1950] and D.R. Stull, *et al*. [Stull, 1956]. R. Hultgren, *et al*. [Hultgren, 1974] presented values selected from many previous publications. T. Iida and R.I.L. Guthrie [Iida, 1988] reproduced an analytical equation taken from Kubaschewski and Alcock [Kubaschewski, 1979] who reference many previous sources ([Kelley, 1960], [Stull, 1971], [Chase, 1978, 1982], [Glushko, 1981], [Pankratz, 1982], [Knacke, 1991]). L.V. Gurvich, *et al*. [Gurvich, 1991] gave tables with data obtained by benchmarked models. Smithells Material Reference Book [Smithells, 2004] reported values taken from [Grosse, 1966], [Stull, 1956] and [Hultgren, 1963].

Data presented in [Kyrillov, 200b] and [TECDOC-1289, 2002] repeat those of [Kutateladze, 1959], therefore they were not included in the database.

Figure 2.13.1 shows the values of heat capacity taken from the selected literature for lead.

R.N. Lyon [Lyon, 1954] gives higher values for the heat capacity of lead than those reported by other sources. The values recommended by S.S. Kutateladze [Kutateladze, 1959] are independent of temperature. At temperatures higher than 1300 K (1027°C) two tendencies are observed. According to Friedland [Friedland, 1966], Iida and Guthrie [Iida, 1988], the specific heat of lead decreases linearly with temperature, while according to R. Hultgren, *et al*. [Hultgren, 1974] it remains constant.

Figure 2.13.1. Heat capacity of molten lead as a function of temperature
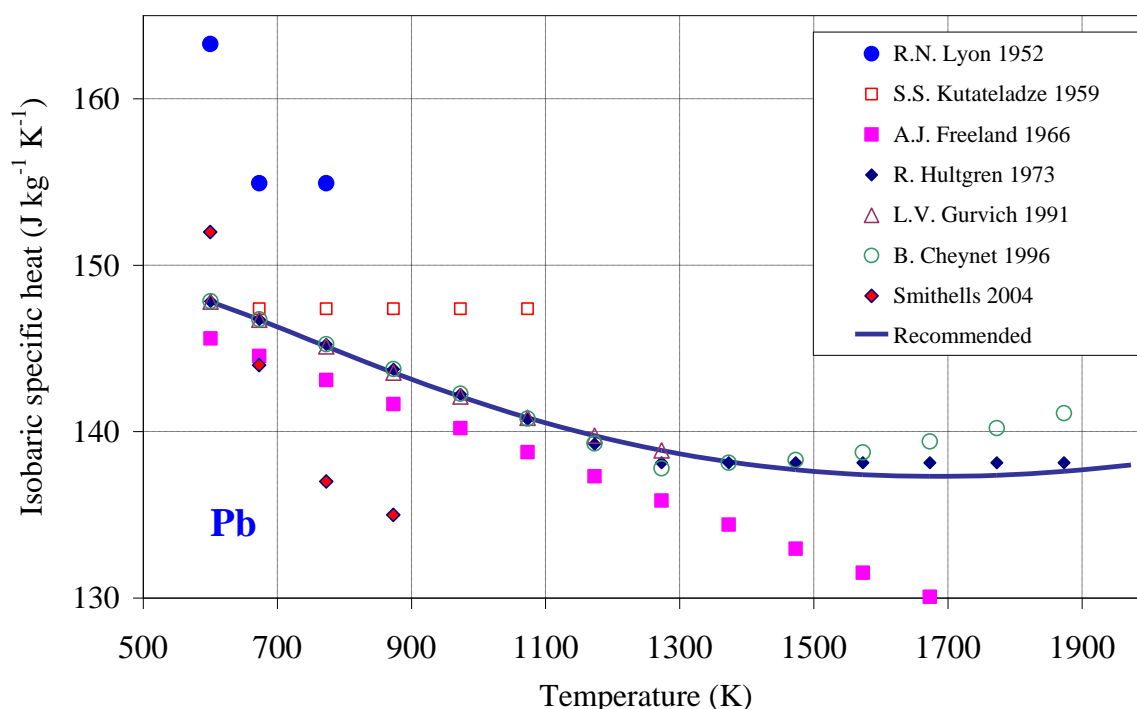
A reasonable choice for the heat capacity of molten lead in the temperature range of T_{melt} to 1300 K (1027°C) is the recommendation of [Gurvich, 1991] where temperature is in kelvins:

$$c_{p\,Pb}\left[J\ kg^{-1}\ K^{-1}\right] = 175.1 - 4.961\times10^{-2}\cdot T + 1.985\times10^{-5}\cdot T^2 - 2.099\times10^{-9}\cdot T^3 - 1.524\times10^6\cdot T^{-2} \quad (2.39)$$

2.13.2 Bismuth

The data for the heat capacity of bismuth as a function of temperature analysed in this handbook have been taken from [Lyon, 1954], [Kutateladze, 1959], [Hultgren, 1974], [Iida, 1988], [Kubaschewski, 1993], [Cheynet, 1996] and Smithells Metal Reference Book [Smithells, 2004].

Howe [Howe, 1961] and Crean [Crean, 1964] reported values given earlier by Lyon [Lyon, 1954, 1960], and were not included in the database.

R. Hultgren, *et al*. [Hultgren, 1974] presented values selected on the basis of a critical assessment of previously published data. T. Iida and R.I.L. Guthrie [Iida, 1988] reported an analytical equation taken from [Kubaschewski, 1979]. O. Kubaschewski, *et al*. [Kubaschewski, 1993] reproduced recommendations of [Knacke, 1991]. (In the earlier compilation [Kubaschewski, 1979] the results of Wartenberg [Wartenberg, 1933] were cited.) Cheynet [Cheynet, 1996] recommended different correlations for different temperature ranges. Smithells Metal Reference Book [Smithells, 2004] presented a set of values with a reference to [Grosse, 1966], [Stull, 1956] and [Hultgren, 1963]. (In the previous edition [Smithells, 1983], an empirical correlation and a set of values based on the results of [Samsonov, 1969], [Viswanath, 1972], [Bush, 1963] and [Stull, 1956] were given.)

Figure 2.13.2 shows the values of heat capacity for molten bismuth extracted from the selected literature sources in the temperature range of interest. As it can be seen, the values reported by R.N. Lyon [Lyon, 1954] increase with temperature and those of S.S. Kutateladze [Kutateladze, 1959] remain constant, in contrast with the results published by other authors. These data were not used in the development of the recommended correlation.

Figure 2.13.2. Heat capacity of bismuth versus temperature
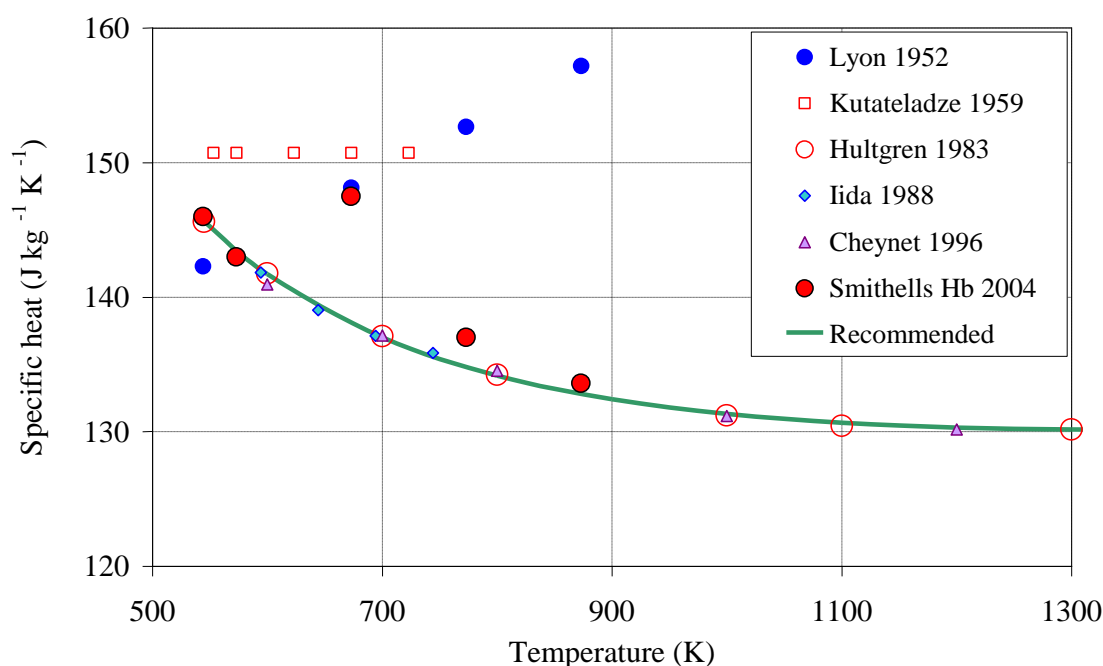
Basing on analogy with other liquid metals the data were chosen with the heat capacity values decreasing with temperature, and the correlation proposed by V. Imbeni, *et al.* [Imbeni 1998a] (transformed to other units) is recommended to use for the isobaric specific heat of molten bismuth:

$$c_{pBi}\left[J\ kg^{-1}\ K^{-1} \right] = 118.2 + 5.934\cdot 10^{-3}\cdot T + 71.83\cdot 10^{5}\cdot T^{-2} \qquad (2.40)$$

Eq. (2.40) is applicable in the temperature range of T_{melt} to 1300 K (1027°C).

2.13.3 LBE

Available data on the heat capacity of LBE are very limited. The authors have found only three independent sources where heat capacity of molten LBE is given at different temperatures: [Lyon, 1954], [Kutateladze, 1959], and [Hultgren, 1973]. The two older publications, [Lyon, 1954] and [Kutateladze, 1959], recommend the same value of $c_{p\,LBE} = 0.035$ cal g^{-1} °C^{-1} for a range of temperatures. In Chapter 2 of the original handbook of Lyon [Lyon, 1954] it was only indicated that this value has been obtained in the temperature interval from 144 to 358°C. However, in Chapter 5 of the same handbook this value was presented as interpolation for the interval of 300 to 700°F (149-371°C or 422-644 K). In [Kutateladze, 1959] this value was recommended for temperatures from 130°C to 700°C (403-973 K). This recommendation was reproduced in later handbooks [Mantell, 1958], [Lyon, 1960], [Bonilla, 1964], [Crean, 1964], [Friedland, 1966], and some of them even extended it to higher

temperatures without any explanation. This value (but in SI units – 146 J kg^{-1} K^{-1}) was recently repeated in [Kyrillov, 2000a] and then reproduced in [TECDOC-1289, 2002]. The only source which reports the LBE heat capacity as a function of temperature is that of R. Hultgren, *et al*. [Hultgren, 1973], which refers to a report of T.B. Douglas and J.L. Dever [Douglas, 1953] as the original source. The data from [Hultgren, 1973] were reprinted in later compilations and a linear interpolation was proposed in [Imbeni, 1998b] and in [Morita, 2004]. Based on an analysis of the available data, the information from three sources: [Lyon, 1954], [Kutateladze, 1959], and [Hultgren, 1973] is included in the database shown in Table 2.13.1. Figure 2.13.3 shows plots of the heat capacity of molten LBE as a function of temperature.

Table 2.13.1. Heat capacity database of LBE

Reg. no.	Parameter	Measurement method	Estimated accuracy %	Temperature range, K	Pressure range, Pa	Values	Ref.
13.1.1	Specific heat	?	?	473-1273	~ 10^5 (?)		[Lyon, 1954]
	Interpolation function: $c_{p\,LBE}(T) = 146.5$ J kg^{-1} K^{-1}						
	Comments: 44.5 wt.% Pb + 55.5 wt.% Bi. Unknown purity.						
13.1.2	Specific heat	Direct heating	?	403-973	~ 10^5 (?)	Annex	[Kutateladze, 1959]
	Interpolation function: $c_{p\,LBE}(T) = 146.5$ J kg^{-1} K^{-1}						
	Comments: 44.5 wt.% Pb + 55.5 wt.% Bi. Unknown purity.						
13.1.3	Specific heat	Heat evolution	5 %	400-1100	~ 10^5 (?)	Annex	[Hultgren, 1973]
	Interpolation function: $c_{p\,LBE}(T) = 160 - 0.0239 \cdot T$ J kg^{-1} K^{-1}						
	Comments: 45.3 wt.% Pb + 54.7 wt.% Bi. Unknown purity.						

Figure 2.13.3. Specific heat of molten LBE versus temperature
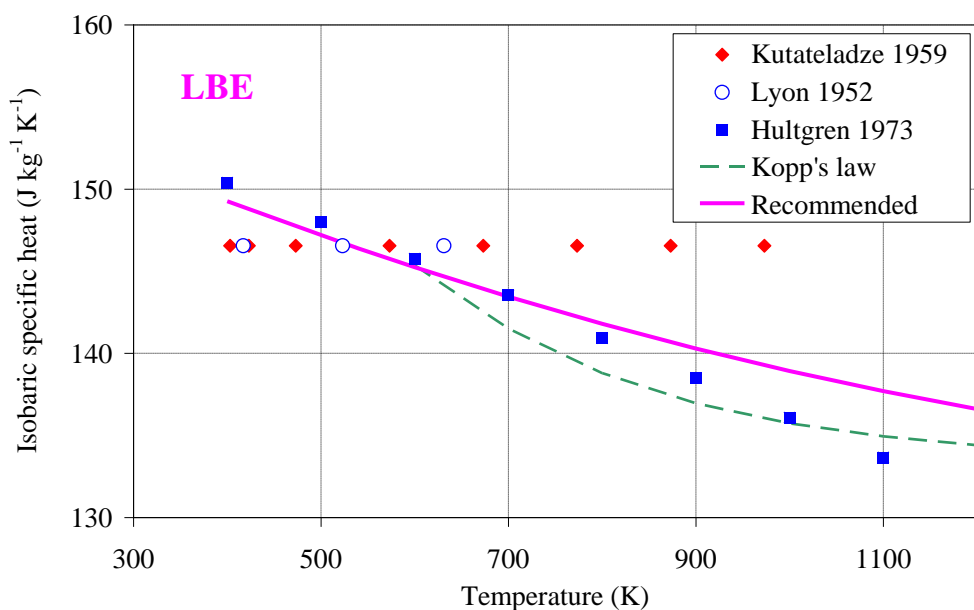
A line obtained with the additive Kopp's law (often used for calculation of the heat capacities of the binary systems) is as follows:

$$c_{p\,LBE} = \frac{x_{Pb} \cdot \mu_{Pb} \cdot c_{p\,Pb} + \left(1 - x_{Pb}\right) \cdot \mu_{Bi} \cdot c_{p\,Bi}}{\mu_{LBE}} \qquad (2.41)$$

where $c_{p\,Pb}$ and $c_{p\,Bi}$ are the specific heat capacities, x_{Pb} and x_{Bi} are the weight contents and μ_{Pb} and μ_{Bi} are molecular masses of lead and bismuth in LBE.

In spite of a good agreement between the values of the selected sources and with the Kopp's law in the temperature interval between the melting temperatures of lead and bismuth 267-327°C (540-600 K), a disagreement exists in the temperature dependence, leading to a larger difference at higher temperatures.

Attempting, on the one hand, to take into account the similarity between LBE and its components (Pb and LB) and, on other hand, to use the available experimental data on LBE specific heat, a fitting of the data set was developed using values given above with a parabolic polynomial (also taking into account the values obtained with the Kopp's law) in the temperature range of 400-1100 K (127-827°C) with the results as follows:

$$c_{p\,LBE}\left[J\ kg^{-1}\ K^{-1}\right] = 159 - 2.72 \cdot 10^{-2} \cdot T + 7.12 \cdot 10^{-6} \cdot T^2 \qquad (2.42)$$

where temperature is in kelvins.

The deviation of the data included in the data set from the recommended Eq. (2.42) is illustrated by Figure 2.13.4.

Figure 2.13.4. Deviation of the available data on heat capacity of molten LBE from Eq. (2.42)
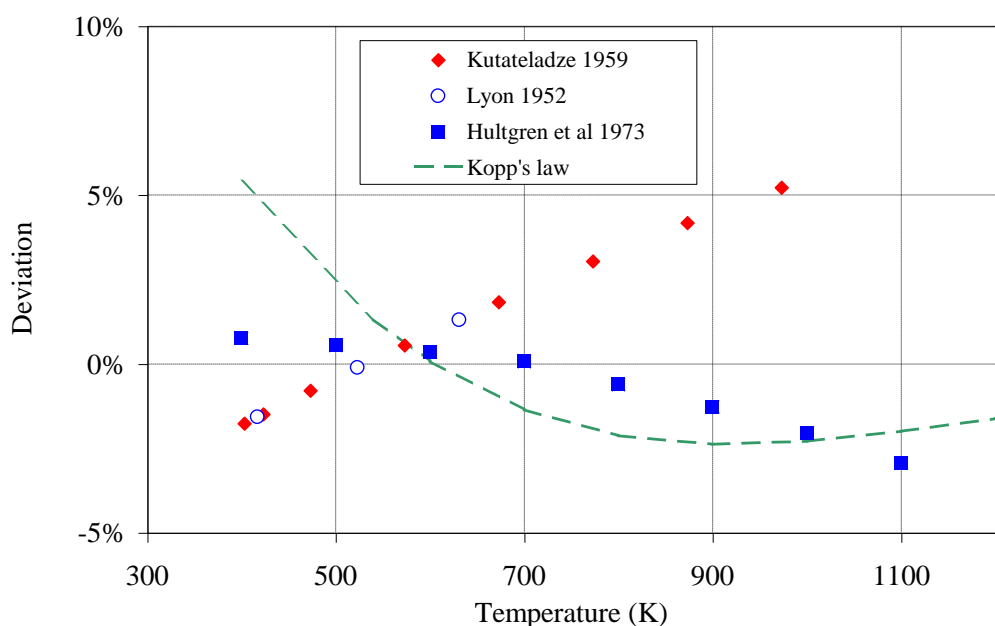
From Figure 2.13.4 one can see that the deviation of the selected values of the LBE specific heats from the recommended Eq. (2.42) does not exceed 5% in the temperature range of 400-1100 K (127-827°C). The deviation is about the same using Kopp's law.

2.14 Critical constants and equation of state

2.14.1 Critical parameters

2.14.1.1 Lead

G. Pottlacher [Pottlacher, 1990] published a summary of experimentally determined and theoretically estimated parameters for the critical point of lead available in the literature as of this date. They considered 16 sets of data as well as their own estimations based on experimental results obtained using the pulse-heating technique. Their summary shows that the experimental and theoretical data on the critical temperature of lead ($T_{c\,Pb}$) lie within the range of 3584-6000 K (3311-5727°C), while their own estimation yields $T_{c\,Pb} = 5400 \pm 400$ K (5127 ± 400°C).

M.M. Martynyuk [Martynyuk, 1998] estimated the critical points of metals by determining parameters of the generalised Van der Waals (VdW) equation using experimental data on liquid phase density and heat of vaporisation in the temperature range from the triple point to the normal boiling point. The calculated results for lead were compared with the experimental data obtained by himself [Martynyuk, 1983]. Poor agreement between the theoretical and experimental values on the critical pressure and density was explained by low accuracy (~50%) in the experiment. Values of $T_{c\,Pb} = 4766$ K (4493°C), $p_{c\,Pb} = 90$ MPa, $\rho_{c\,Pb} = 2259$ kg/m^3 were recommended for the critical point of lead.

K.M. Watson [Watson, 1933] proposed a theoretical method for estimating the critical temperature of non-polar or a slightly polar material and deduced the following formula:

$$\frac{T_e}{T_c} = 0.283 \cdot \left(\frac{M}{\rho_l(T_b)} \right)^{0.18} \tag{2.43}$$

where M is in g/mol, $\rho_l(T_b)$ is the liquid density in g/cm^3 at the normal boiling point T_b and T_e is the temperature at which the molar volume of vapour is $22.4 \cdot 10^{-3}$ m^3/mol. The following relation was used to calculate T_e based on the work of T.C. Chawla, *et al*. [Chawla, 1981]:

$$\ln T_e = \frac{9.8 T_e}{T_b} - 4.2 \tag{2.44}$$

The critical density can be estimated from the semi-empirical "law of rectilinear diameter" [Cailletet, 1986]. With a reported value for the critical compressibility, Z_c, of 0.21 for lead [Pottlacher, 1998] the following values were obtained: $T_{c\,Pb} = 4961$ K, $p_{c\,Pb} = 114$ MPa and $\rho_{c\,Pb} = 2720$ kg/m^3.

Recently, A-M. Azad [Azad, 2005] estimated the critical temperatures of lead, bismuth and LBE with a theoretical model called "the internal pressure approach", which is described below in Section 2.14.1.3. The estimation for lead indicates a quite low value of the lead critical temperature ($T_{c\,Pb} = 3096\text{-}3118$°C (3369-3391 K)) in comparison with most of other reported theoretical and experimental values. This can be explained by errors in extrapolation of the experimental data to the high temperatures.

A summary of data on the critical point of lead extracted from the above-cited literature is presented in Table 2.14.1 (the data reviewed in [Pottlacher, 1990] are not included – only their recommendation is reproduced in the table). The critical compressibility, Z_c, is determined in the table as follows:

$$Z_c \equiv \frac{p_c V_c}{RT_c} = \frac{p_c M}{RT_c \rho_c} \tag{2.45}$$

where M is the molecular mass and R = 8.314 J/mol is the universal gas constant.

Table 2.14.1. Summary of the critical point data for lead

Reference	T_c [K]	p_c [MPa]	ρ_c [kg/m^3]	Z_c
[Martynyuk, 1983]	4460	160	2950	0.30
[Pottlacher, 1990]	5400 ± 400	250 ± 30	3200 ± 300	0.36
[Martynyuk, 1998]	4776	90	2259	0.21
[Pottlacher, 1998]	4961	114	2720	0.21
[Azad, 2005]	3369-3391	–	–	–

From Table 2.14.1 one can see that the later more accurate results of M.M. Martinuk [Martynyuk, 1998] and G. Pottlacher [Pottlacher, 1998] are in a very good agreement. Taking into account a large uncertainty in the estimations, the mean rounded values of these two sources are recommended for the critical temperature, pressure and density of lead:

$$T_{c\,Pb} = 4870 \text{ K } (4597°C), \; p_{c\,Pb} = 100 \text{ MPa}, \; \rho_{c\,Pb} = 2490 \text{ kg/m}^3 \tag{2.46}$$

2.14.1.2 Bismuth

Only a few sets of the critical point data are available for bismuth. A.V. Grosse [Grosse, 1961] estimated the critical temperature of bismuth ($T_{c\,Bi} = 4620 \text{ K} \approx 4347°C$) by application of the principle of corresponding states to the enthalpy of vaporisation. J.A. Cahill, *et al*. [Cahill, 1968] compared this value with one estimated by the method of rectilinear diameter ($T_{c\,Bi} \sim 5000 \text{ K} \approx 4727°C$), which assumes that the average density of the liquid and the saturated vapor changes linearly with temperature. V.E. Fortov, *et al*. [Fortov, 1975] calculated the theoretical values of the critical temperature ($T_{c\,Bi} = 4200 \text{ K} \approx 3927°C$), pressure ($p_{c\,Bi} = 126$ MPa) and density ($\rho_{c\,Bi} = 2660 \text{ kg/m}^3$) of bismuth, based on traditional approaches such as the Grosse method [Grosse, 1961] and on the method of rectilinear diameter [Pottlacher, 1998]. Martynyuk [Martynyuk, 1983] estimated the parameters of the critical point of bismuth from the pulse heating experiment and obtained $T_{c\,Bi} = 3780 \text{ K } (3507°C)$, $p_{c\,Bi} = 132$ MPa and $\rho_{c\,Bi} = 3010 \text{ kg/m}^3$. The estimation of the critical parameters using the Watson method [Watson, 1931] and the law of rectilinear diameter yields: $T_{c\,Bi} = 4354 \text{ K } (4081°C)$ and $\rho_{c\,Bi} = 2705 \text{ kg/m}^3$. The critical temperature of bismuth estimated by Azad [Azad, 2005] ($T_{c\,Bi} = 2042\text{-}2106 \text{ K} \approx 1769\text{-}1883°C$) is lower than recommended by other authors.

A summary of the critical point parameters for bismuth, available in the literature, is presented in Table 2.14.2.

Table 2.14.2. Summary of the critical point data of bismuth

Reference	T_c [K]	p_c [MPa]	ρ_c [kg/m^3]	Z_c
[Grosse, 1961]	4620			
[Cahill, 1968]	5000			
[Fortov, 1975]	4200	126	2660	0.28
[Martynyuk, 1998]	3780	132	3010	0.29
[Azad, 2005]	2042-2106			

There is still a very large uncertainty in the available data. For the time being, we recommend the mean rounded values reported by Fortov [Fortov, 1975] and by Martynyuk [Martynyuk, 1998] as the more complete and consistent data set:

$$T_{c\,Bi} = 3990 \text{ K } (3717°\text{C}), \ p_{c\,Bi} = 130 \text{ MPa}, \ \rho_{c\,Bi} = 2890 \text{ kg/m}^3 \qquad (2.47)$$

The recommended above values for the critical parameters of lead and bismuth are reasonable in comparison with the experimental data [Martynyuk, 1983] if we consider the experimental uncertainty estimated as ±50% for p_c, ±15% for T_c, and ±20% for ρ_c.

2.14.1.3 LBE

Only two sets of the critical parameters of LBE are available in the published literature. One is proposed by K. Morita, *et al*. [Morita, 2004, 2005] and another by A-M. Azad [Azad, 2005]. Both were estimated theoretically on the basis of known properties for LBE. No publications are currently available with the results of measurement of the LBE critical parameters.

K. Morita, *et al*. [Morita, 2004, 2005] used the method proposed by Martynyuk [Martynyuk, 1998], which is based on the following generalised relationship:

$$p = \frac{RT}{V-b} - \frac{a}{V^n} \qquad (2.48)$$

where a, b and n are constants that must be determined. In [Martynyuk, 1998] these constants are expressed through the melting and boiling temperatures, the latent heats of melting and boiling, and the liquid molar volume in the melting and boiling points. In their calculations, Morita [Morita, 2004, 2005] took the liquid density from [Alchagirov, 2003] and calculated the heat of vaporisation from the Clapeyron equation using their estimated vapour pressure curve. The critical parameters of LBE obtained by them are $T_{c\,LBE} = 4890$ K (4617°C), $\rho_{c\,LBE} = 2170$ kg/m^3 and $p_{c\,LBE} = 87.8$ MPa.

M-A. Azad [Azad, 2005] estimated the LBE critical temperatures using a simple equation of state (EOS) for the liquid phase based on the concept of internal pressure of liquids and assuming that data on density, heat capacity and sound velocity are known:

$$\frac{p + p_i}{T} = u_{sound}\, \alpha_p \rho \frac{C_p}{C_V} \qquad (2.49)$$

where p_i is the internal pressure, u_{sound} is the sound velocity, α_p is the volumetric thermal expansion coefficient and C_p and C_V are the isobaric and isochoric heat capacities, respectively. The right side of Eq. (2.49) has been described with a linear empirical function of the form:

$$\frac{p + p_i}{T} = A - BT \tag{2.50}$$

The gas phase was described with the perfect gas equation. In order to determine the constants A and B, the LBE melt density was taken from the measurements of B.B. Alchagirov [Alchagirov, 2003]. The LBE heat capacity and the sound velocity were computed with Vegard's law using the literature data on heat capacities [Kubaschewski, 1979], [Hultgren, 1974] and sound velocities [Mustafin, 1983], [Jarzynski, 1963] of lead and bismuth. The calculated values for the LBE critical temperature and pressure are $T_{c\ LBE}$ = 2411 K (2138°C) and $p_{c\ LBE}$ = 244 MPa. It should be noted that the critical temperature of LBE obtained by Azad [Azad 2005] is more than by factor 2 lower and the critical pressure is 3 times higher than those calculated by Morita, *et al.* [Morita, 2004, 2005]. A summary of the critical parameters of LBE is given in Table 2.14.3.

Table 2.14.3. Summary of the critical point data for LBE

Reference	T_c [K]	p_c [MPa]	ρ_c [kg/m^3]	Z_c
[Morita, 2004]	4890	87.8	2170	
[Azad, 2005]	2411	244	–	–

A very large uncertainty still exists in the estimation of the LBE critical parameters. More detailed analysis and comparison with other heavy metals is needed in order to provide recommendations. One of the reasons for a lower critical temperature obtained in the calculations of Azad may be the use of an incorrect temperature dependence of the sound velocity. In the absence of better data, the rounded values obtained by K. Morita, *et al.* [Morita, 2004] are recommended for very approximate estimations:

$$T_{c\ LBE} = 4890 \text{ K } (4617°C), \ p_{c\ LBE} = 88 \text{ MPa}, \ \rho_{c\ LBE} = 2170 \text{ kg/m}^3 \tag{2.51}$$

2.14.2 Equation of state

The vapour EOS for LBE was developed by K. Morita, *et al.* [Morita, 2004, 2005] for use in reactor safety analysis. They used the modified Redlich-Kwong (MRK) equation extended to a reacting system [Morita, 1998a], where the dimmer and monomer components consist of vapour. The functional form of the MRK equation extended to a reacting system is as follows:

$$p = \frac{RT}{M(1 + y_2)(v - a_1)} - \frac{a(T)}{v(v + a_3)} \tag{2.52}$$

with:

$$a(T) = a_2 \left(\frac{T}{T_c}\right)^{a_4}, \qquad T \leq T_c \tag{2.53}$$

$$a(T) = a_2 + \left.\frac{da}{dT}\right|_{T_c} (T - T_c), \quad T > T_c \tag{2.54}$$

where p is the pressure, T is the temperature, v is the specific volume, y_2 is the dimer fraction, and a_1, a_2, a_3 and a_4 are the model parameters. From the definition, y_2 is related to the equilibrium constant k_2 given by:

$$k_2 = \frac{p_2}{(p_1)^2} = \frac{y_2}{(1-y_2)^2 p} \qquad (2.55)$$

where the total pressure p is the sum of the monomer pressure p_1 and the dimer pressure p_2:

$$p = p_1 + p_2 \qquad (2.56)$$

Although Eq. (2.52) was originally developed by Morita [Morita, 1998b] for sodium with dimerisation, it was applied to LBE assuming that LBE vapour consists of monatomic and diatomic components, representing the diatomic component of the LBE vapour as the diatomic bismuth. Therefore, Eq. (2.56) was given by:

$$p_1 = p_{Pb} + p_{Bi} \qquad (2.57)$$

$$p_2 = p_{Bi_2} \qquad (2.58)$$

where p_{Pb}, p_{Bi} and p_{Bi_2} are the partial pressure of monatomic lead, monatomic bismuth and diatomic bismuth, respectively.

The equilibrium constant of LBE, Eq. (2.55), was evaluated based on the vapour pressure data taken from the "best" values tabulated by R. Hultgren, *et al.* [Hultgren, 1974] for lead and bismuth as used for the estimation of the LBE vapour pressure. The results in the temperature range 700-2000 K (427-1727°C) were fitted to the following equation:

$$k_2 = \exp\left(-24.611 + \frac{23511}{T}\right) \qquad (2.59)$$

where k_2 is in Pa^{-1} and T is in K.

The EOS parameters a_1, a_2 and a_3 in Eq. (2.52) were determined from the estimated values of critical constants [Morita, 2004, 2005], and the fact that the critical isotherm on a pressure-volume *p-v* diagram has an inflection point at the critical point. The parameter a_4 was fitted to the slope of the vapour pressure curve [Morita, 2004, 2005] at the critical temperature of 4617°C (4890 K). The resultant parameters are:

$$\begin{aligned} a_1 &= 6.26824 \times 10^{-5}, \ a_2 = 1.59328 \times 10^{2}, \\ a_3 &= 8.11866 \times 10^{-4}, \ a_4 = 3.78359 \times 10^{-1} \end{aligned} \qquad (2.60)$$

A similar approach can be used for construction of the EOS for lead and bismuth.

2.15 Viscosity

Accurate and reliable data on viscosity of liquid metals are not abundant. Some discrepancies between experimental data can be attributed to the high reactivity of metallic liquids, to the difficulty of taking precise measurements at elevated temperatures, and to a lack of a rigorous formula for calculations. All liquid metals are believed to be Newtonian liquids. The temperature dependence of viscosity of liquid metals are usually described by an Arrhenius type formulae:

$$\eta = \eta_0 \cdot \exp\left(E_\eta / RT\right) \tag{2.61}$$

where E_η is activation energy of motion for viscous flow.

In engineering hydrodynamics, the kinematic viscosity ν is often used, which is a ratio of the dynamic viscosity η to the liquid density ρ:

$$\nu = \eta/\rho \tag{2.62}$$

2.15.1 Lead

The viscosity data for lead as a function of temperature included in the present handbook were taken from R.N. Lyon [Lyon, 1954, 1960], S.S. Kutateladze, *et al*. [Kutateladze, 1959], W. Hofmann [Hofmann, 1970], L.D. Lucas [Lucas, 1984b], T. Iida and R.I.L. Guthrie [Iida, 1988], P.L. Kyrillov and G.P. Bogoslovskaya [Kyrillov, 2000a] (reproduced in [TECDOC-1289, 2002]) and Smithells Metal Reference Book [Smithells, 2004].

W. Hofmann [Hofmann, 1970] analysed data of many previous authors and found a satisfactory agreement. Lucas [Lucas ,1984b] used the lead viscosity values from Ofte [Ofte, 1967]. Iida and Guthrie [Iida, 1988] reference [Iida, 1975]. Data from [Kyrillov, 2000a] and [TECDOC-1289, 2002] are close to those of [Kutateladze, 1959] but extended to higher temperature (1273 K $\approx$ 1000°C indicated in [Kyrillov, 2000a]). Smithells Metals Handbook [Smithells, 2004] took the parameters of Eq. (2.61) from [Beyer, 1972] and [Wittenberg, 1970].

Figure 2.15.1 shows the values of molten lead viscosity taken from the cited literature.

Figure 2.15.1. Lead dynamic viscosity as a function of temperature
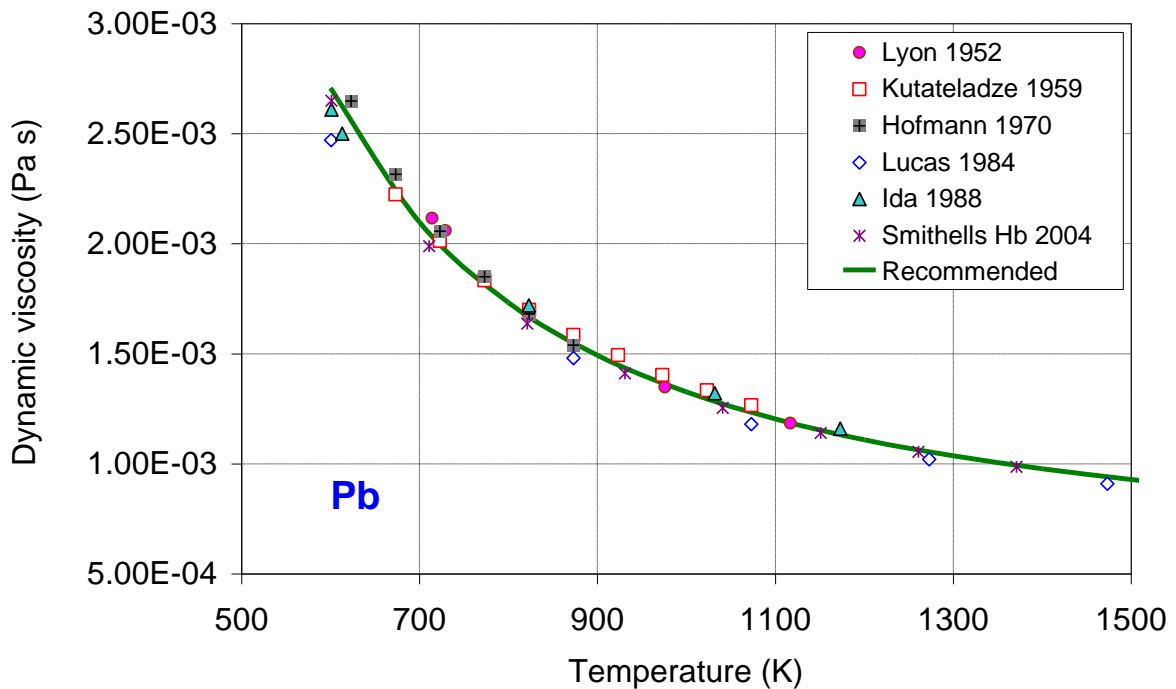
Good agreement exists among the different sets of experimental data and the values calculated by means of the empirical equations. A reliable choice of an empirical equation to describe the temperature dependence of the dynamic viscosity of molten lead can be obtained by fitting the selected values into an Arrehenius type equation of the form:

$$\eta_{Pb}[Pa\ s] = 4.55 \cdot 10^{-4} \cdot \exp\left(\frac{1069}{T}\right) \tag{2.63}$$

where temperature is in kelvins. This correlation is valid in the temperature range from T_{melt} to 1470 K (1197°C).

2.15.2 Bismuth

Most of analysed viscosity data for bismuth as a function of temperature are from about the same sources as for lead: [Lyon, 1954], [Kutateladze, 1959], [Bonilla, 1964], [Lucas, 1984b], [Iida, 1988] and Smithells Metal Reference Book [Smithells, 2004].

The values reported by Bonilla [Bonilla, 1964], are very high as compared with those given by the other sources, and those reported by Iida [Iida, 1988] are significantly lower. These data should be disregarded.

Smithells Metal Reference Book [Smithells, 2004] gives the parameters of Eq. (2.61) based on the experimental results from [Wittenberg, 1970] and [Beyer, 1972]. Lucas [Lucas, 1984b] presented viscosity experimentally determined by Ofte [Ofte, 1963]. Iida [Iida, 1988] references [Iida, 1975].

Figure 2.15.2 shows the values of viscosity taken from the cited literature for bismuth.

Figure 2.15.2. Bismuth viscosity as a function of temperature
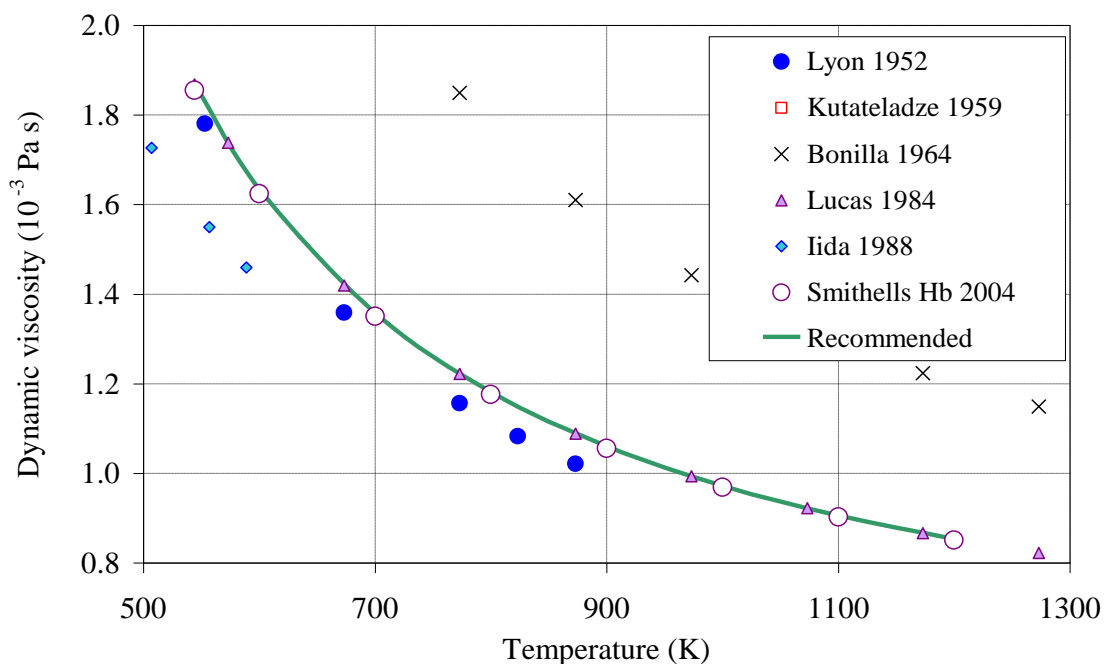
A reasonable choice for an empirical correlation to calculate the bismuth viscosity as a function of temperature is that proposed by Smithells Metal Reference Book [Smithells, 2004]:

$$\eta_{Bi}[Pa\ s] = 4.458 \cdot 10^{-4} \cdot \exp\left(\frac{775.8}{T}\right) \tag{2.64}$$

where temperature is in kelvins. This correlation is valid in the temperature range from T_{melt} to 1300 K (1027°C).

2.15.3 LBE

In the old Western sources, the recommendations for LBE viscosity are given for the temperature range of 300-700°C (673-973 K) by R.N Lyon [Lyon, 1954, 1960], C.L. Mantell [Mantell, 1958], and C.F. Bonilla [Bonilla, 1964]. At temperatures below 300°C (573 K) the data for the LBE kinematic viscosity are found in the publication of Kutateladze [Kutateladze, 1959]. New data were reported by J.P. Holman [Holman, 1968] and detailed measurements were performed later by B. Kaplun, *et al*. [Kaplun, 1979]. Recent data from [Kyrillov, 2000a] and [TECDOC-1289, 2002] are very close to those of [Kutateladze, 1959] up to about 900 K (627°C). At higher temperatures they deviate to higher values, probably, due to the presence of oxides or other impurities. The sources included in database are described in Table 2.15.1.

The data extracted from the selected sources are presented in Figure 2.15.3.

Table 2.15.1. Viscosity database for molten LBE

Reg. no.	Parameter	Measurement method	Estimated accuracy %	Temperature range, K	Pressure range, Pa	Values	Reference
15.3.1	Dynamic viscosity	?	?	605-873	~ 10^5 (?)		[Lyon, 1954]
	Interpolation function: $\eta_{LBE}(T) = \left(5.37 - 8.92 \cdot 10^{-3} \cdot T + 4.71 \cdot 10^{-6} \cdot T^2\right) \cdot 10^{-3}$ Pa s						
	Comments: 44.5 wt.% Pb + 55.5 wt.% Bi. Unknown purity.						
15.3.2	Kinematic viscosity	?	?	403-973	~ 10^5 (?)		[Kutateladze, 1959]
	Interpolation function: $\eta_{LBE}(T) = \left(2.077 - 8.983 \cdot 10^{-3} \cdot T + 1.629 \cdot 10^{-5} \cdot T^2 - 1.352 \cdot 10^{-8} \cdot T^3 + 4.25 \cdot 10^{-12} \cdot T^4\right) \cdot 10^{-2}$ Pa s						
	Comments: 44.5 wt.% Pb + 55.5 wt.% Bi. Unknown purity.						
15.3.3	Dynamic viscosity	?	?	403-1073	~ 10^5 (?)		[Holman, 1968]
	Interpolation function: $\eta_{LBE}(T) = \left(4.56 - 7.03 \cdot 10^{-3} \cdot T + 3.61 \cdot 10^{-6} \cdot T^2\right) \cdot 10^{-3}$ Pa s						
	Comments:						
15.3.4	Dynamic viscosity	?	?	410-726	~ 10^5 (?)		[Kaplun, 1979]
	Interpolation function: $\eta_{LBE}(T) = \left(1.702 - 6.612 \cdot 10^{-3} \cdot T + 1.064 \cdot 10^{-5} \cdot T^2 - 7.751 \cdot 10^{-9} \cdot T^3 + 2.118 \cdot 10^{-12} \cdot T^4\right) \cdot 10^{-2}$ Pa s						
	Comments:						

* The data on the LBE dynamic viscosity presented in Table 2.15.1 from [Kutateladze, 1959] have been calculated with Eq. (2.62) using the data on the kinematic viscosity and density presented in the same source.

Figure 2.15.3. Dynamic viscosity of the molten LBE versus temperature
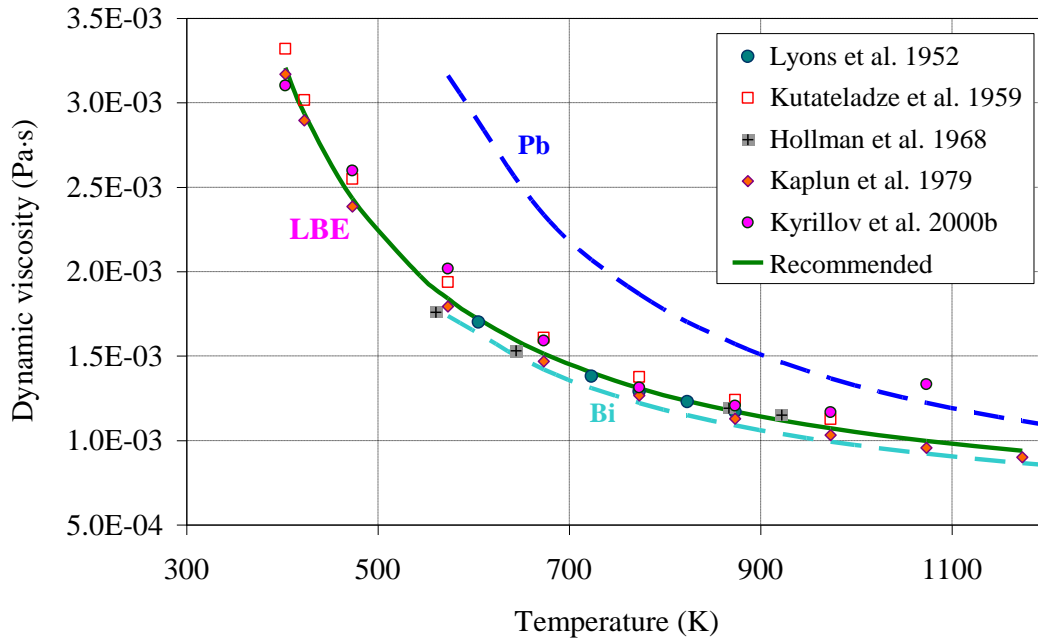
Most of the discrepancies in the experimental data on LBE viscosity can be attributed to its high reactivity. This physical parameter is never additive for mixtures or alloys. Fitting the parameters of Eq. (2.61) to the viscosity database presented above yields the following correlation:

$$\eta_{\text{LBE}}[\text{Pa s}] = 4.94 \cdot 10^{-4} \cdot \exp\left(\frac{754.1}{T}\right) \tag{2.65}$$

where temperature is in kelvins.

Eq. (2.65), along with the recommended correlations for the dynamic viscosity of molten lead and of molten bismuth, are also presented in Figure 2.15.3. It can be seen that viscosity of LBE is significantly lower than that of lead (especially at lower temperatures) and very close to the viscosity of bismuth. The maximum difference between the LBE viscosity given by (2.65) and the database set is about 6% are shown in Figure 2.15.4 in the temperature range of 400-1100 K (127-827°C).

The kinematic viscosity can be determined from Eq. (2.62) using the recommended correlations for the dynamic viscosity. Its dependence on temperature is illustrated by Figure 2.15.5. The kinematic viscosity of LBE is 1.2-1.5 times smaller than that of lead in the temperature range of 600-1100 K (327-827°C), but it is very close to the kinematic viscosity of bismuth.

2.16 Electrical resistivity

Metals in general are characterised by a very low electrical resistance, that increases with temperature and about doubles as a consequence of melting. The electrical resistivity of liquid metals with rare exceptions increases linearly with temperature (in the temperature region of interest) and therefore can be represented as follows:

$$r = r_0 + b_e \cdot T$$

77

Figure 2.15.4. Dispersion of the database set on the LBE dynamic viscosity from Eq. (2.65)
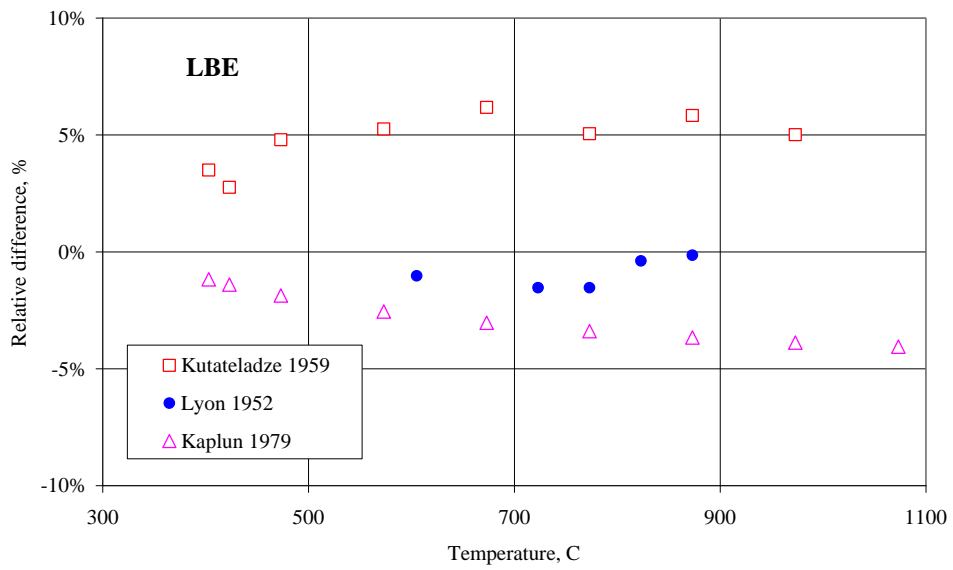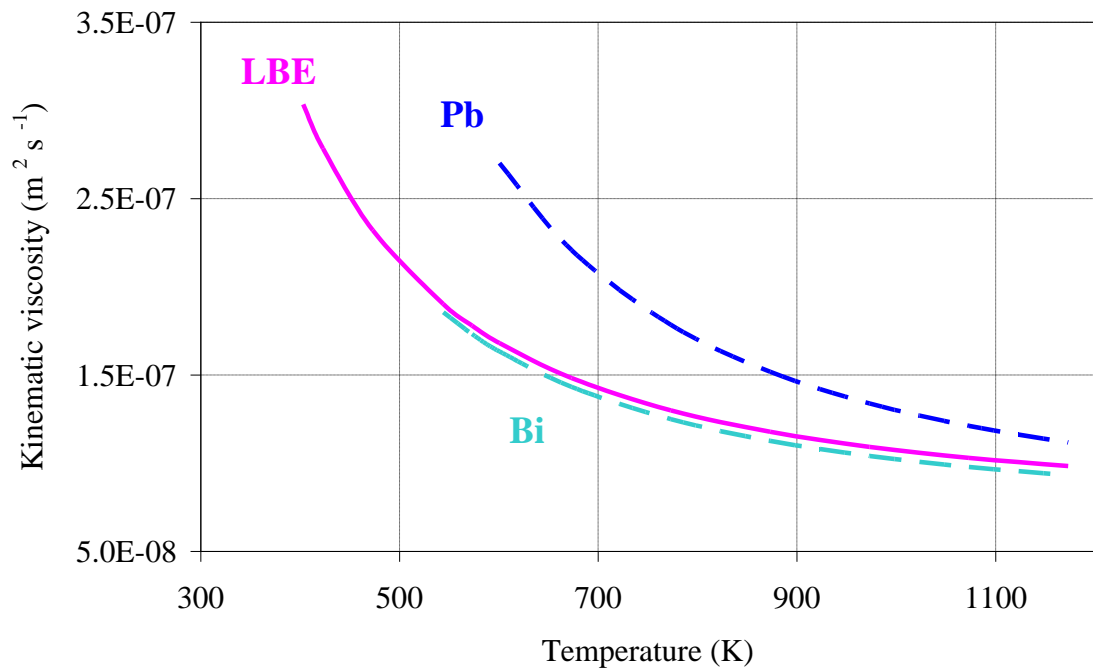

Figure 2.15.5. Kinematic viscosity of the melted lead, bismuth and LBE versus temperature

In general, the electrical resistivity of liquid metals increases, when impurities enter the melt. However, in the case of a liquid alloy system that is composed of polyvalent component, the resistivity sometimes shows a negative deviation from additivity of component resistivities.

2.16.1 Lead

The data on lead electrical resistivity as a function of temperature included in this handbook were taken from [Lyon, 1954, 1960], [Hofman, 1970], [Smithells, 1983, 2004], T. Iida and R.I.L. Guthrie [Iida, 1988], [Bretonnet, 1988], [Kyrillov, 2000a] and [TECDOC-1289, 2002].

Smithells Metal Reference Book [Smithells, 1983, 2004] reported the lead electric resistivity taken from C.Y. Ho, *et al*. [Ho, 1968] and H.A. Davies and J.S.L. Leach [Davies, 1970]. T Iida and R.I.L. Guthrie [Iida, 1988] reported a correlation taken from N. Cusack and J.E. Enderby [Cusack, 1960]. J.L. Bretonnet, *et al.* [Bretonnet, 1988] treated different sources to obtain his correlation for lead electric resistivity: [Tschirner, 1969], [Davies, 1970] and [Mera, 1972].

A.J. Friedland [Friedland, 1966] reproduced data from [Lyon, 1960] and therefore was not included in the database.

Figure 2.16.1 shows the values of the electrical resistivity for molten lead taken from the literature cited above.

Figure 2.16.1. Electrical resistivity of molten lead versus temperature
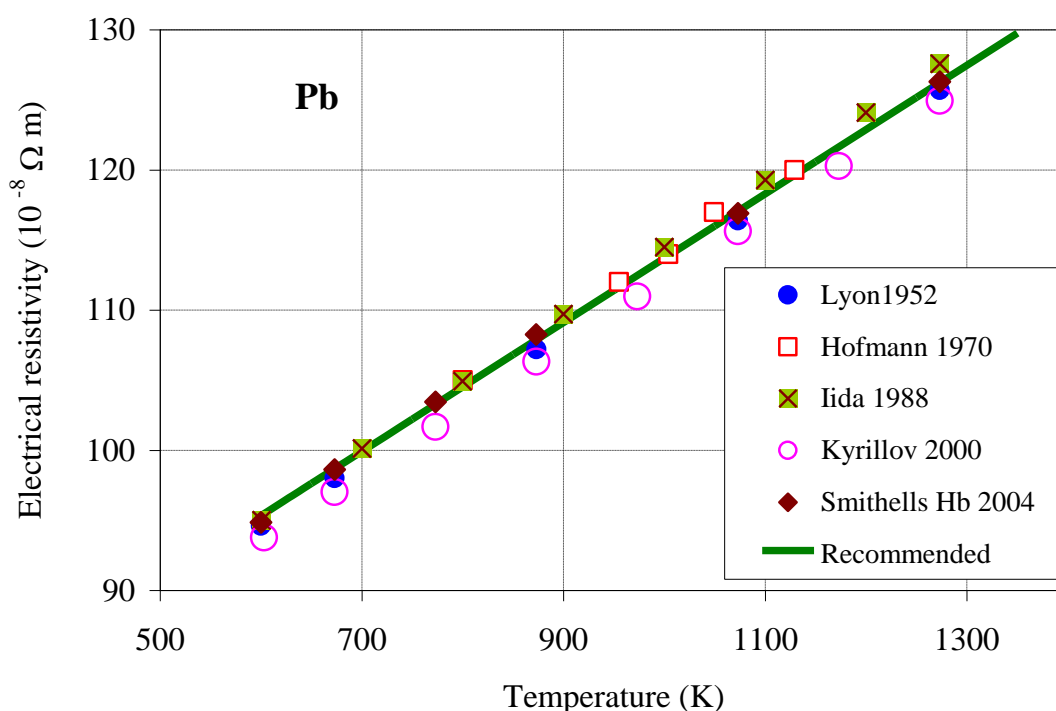
The agreement between different sources is very good. Therefore, a reasonable choice of an empirical equation suitable for the calculation of the electrical resistivity of liquid lead as a function of temperature can be based on the coefficients given by Iida and Guthrie [Iida, 1988], i.e.:

$$r_{Pb}[\Omega \ m] = 0.666 \cdot 10^{-6} + 4.79 \cdot 10^{-10} \cdot T \qquad (2.66)$$

which is valid in the temperature range of 601-1273 K (328-1000°C). The deviation of the selected data from this correlation is less than 1%.


2.16.2 Bismuth

The data for the electrical resistivity of molten bismuth included in this handbook were taken from [Lyon, 1954], [Iida, 1988] and [Bretonnet, 1988].

Smithells Metal Reference Book [Smithells, 2004] only reported the resistivity value at the melting point. T. Iida and R.I.L. Guthrie [Iida, 1988] reproduced a correlation from [Cusack, 1960]. J.L. Bretonnet, *et al.* [Bretonnet, 1988] obtained his formula using the data of J.G. Gasser, *et al.* [Gasser, 1984] and S. Ohno, S. Tamaki [Ohno, 1975].

Figure 2.16.2 shows the values of electrical resistivity taken from literature for liquid bismuth, in the range of temperature of interest.

Figure 2.16.2. Electrical resistivity of liquid bismuth versus temperature
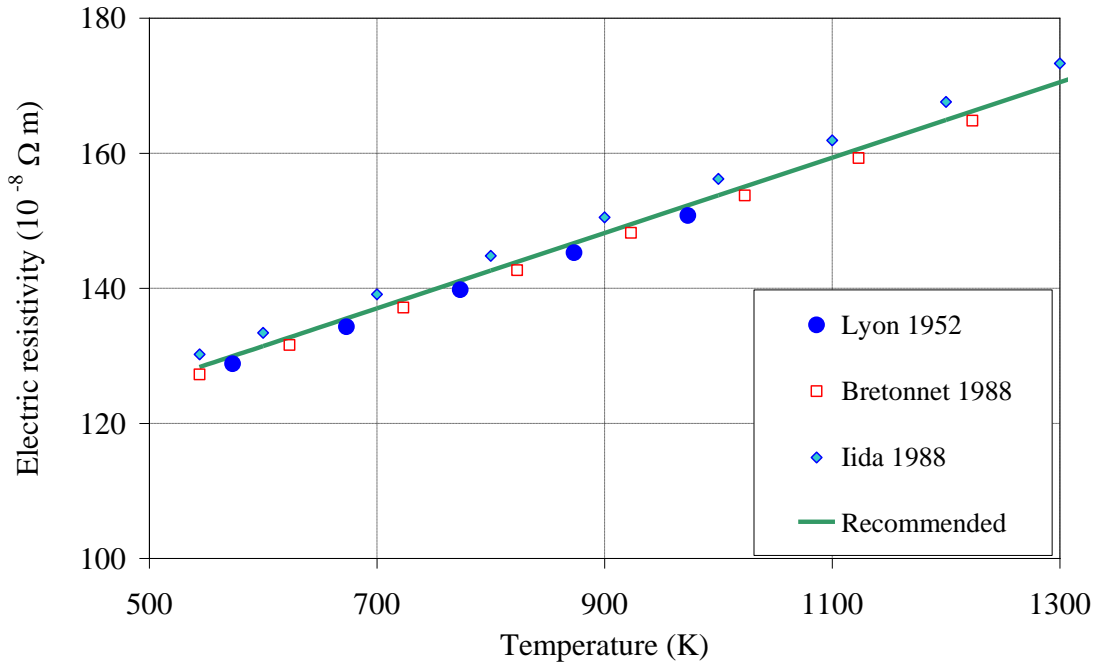
The amount of the available data on the electric resistivity of molten bismuth is limited, but the agreement between the three sources is good. Therefore, it is proposed to use an empirical correlation based on the coefficients given by J.L. Bretonnet [Bretonnet, 1988], i.e.:

$$r_{Bi}[\Omega \ m] = 0.971 \cdot 10^{-6} + 5.534 \cdot 10^{-10} \cdot T \qquad (2.67)$$

where temperature is in kelvins. The estimated deviation of other data from this correlation is about of 0.8% in the temperature range of 545-1420 K (272-1147°C).

2.16.3 LBE

Only two reliable data sources for the electric resistivity of molten LBE were found in the literature: [Lyon, 1954] (probably repeated by [Kutateladze, 1959]) and [TECDOC-1289, 2002]. A.J. Friedland [Friedland, 1966] reported only one value at 538°C (811 K). The data presented in Kyrillov and Bogoslovskaya [Kyrillov, 2000a] have been discarded because they give a very low electrical resistivity which is almost independent of temperature (later these data were corrected by P.L. Kyrillov in [TECDOC-1289, 2002]). The data from [Lyon, 1954] and [TECDOC-1289, 2002] were included in the database shown in Table 2.16.1.

Table 2.16.1. Electrical resistivity database for the melted LBE

Reg. no.	Parameter	Measurement method	Estimated accuracy %	Temperature range, K	Pressure range, Pa	Values	Ref.
16.3.1	Electrical resistivity	?	?	473-773	~ 10^5 (?)		[Lyon, 1954]
	Interpolation function: $r_{LBE}(T) = (89.343 + 0.05 \cdot T) \cdot 10^{-8}$ Ωm						
	Comments: 44.5 wt.% Pb + 55.5 wt.% Bi. Unknown purity.						
16.3.2	Electrical resistivity	?	?	403-1073	~ 10^5 (?)		[TECDOC-1289, 2002]
	Interpolation function: $r_{LBE}(T) = (83.325 + 0.0523 \cdot T) \cdot 10^{-8}$ Ωm						
	Comments: 44.5 wt.% Pb + 55.5 wt.% Bi. Unknown purity.						

The experimental data are presented in Figure 2.16.3 together with those recommended for the electric resistivity of molten lead and of molten bismuth.

Figure 2.16.3. Electric resistivity of molten lead, bismuth and LBE versus temperature
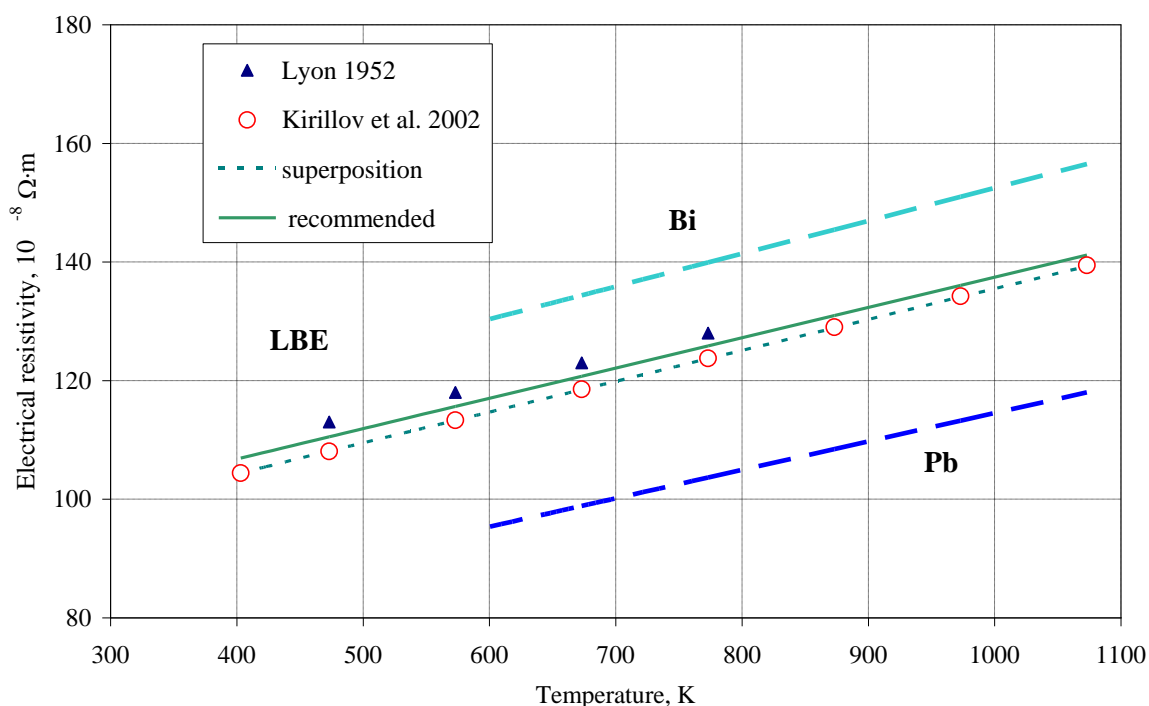
In Figure 2.16.3 one can see that the LBE resistivity data recommended in [TECDOC-1289, 2002] fit very well the line calculated with the Vegard-Kopp law using the resistivities of lead and bismuth. The recommendations of Lyon [Lyon, 1954] yield higher values. Taking into account that the Vegard-Kopp law is not applicable in the general case to transport properties of binary systems, and that only two sources are available, all data points were included to in the development of the recommended linear interpolation correlation as follows:

$$r_{LBE}[\Omega \text{ m}] = (86.334 + 0.0511 \cdot T) \cdot 10^{-8} \qquad (2.68)$$

where temperature is in kelvins.

The deviation of the data recommended in [Lyon, 1954] and [TECDOC-1289, 2002] for the electric resistivity of molten LBE at normal conditions is illustrated in Figure 2.16.4. The maximum difference does not exceed 2.5% in the temperature range of 400-1100 K (127-823°C).

Figure 2.16.4. Deviation of the experimental data on the electric resistivity of LBE from the recommended Eq. (2.68)
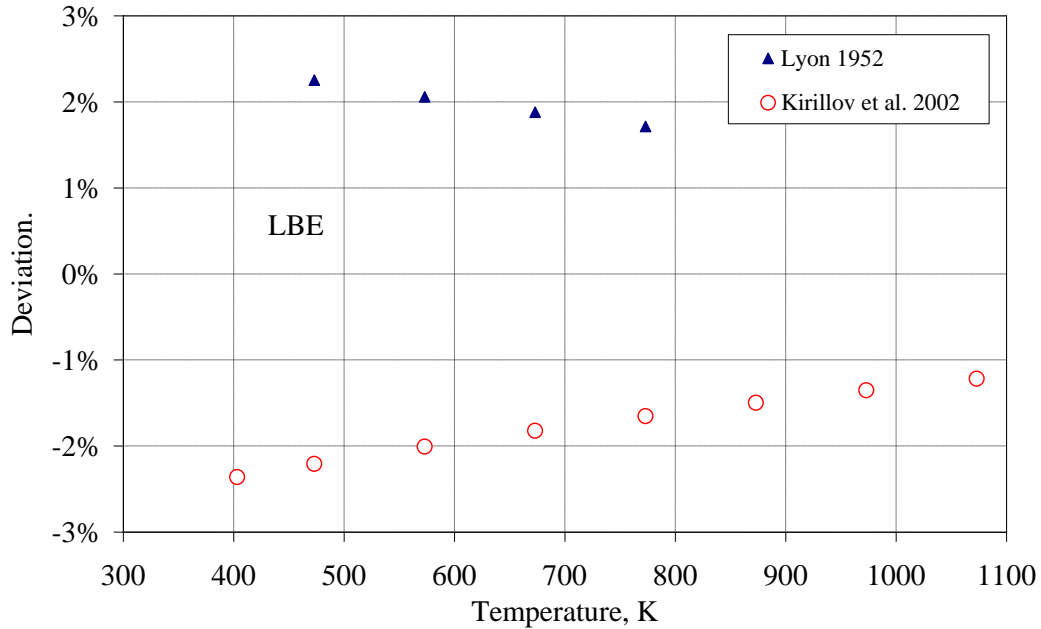
2.17 Thermal conductivity and thermal diffusivity

Experimental determination of thermal conductivity of liquid metals is difficult because of the problems related to convection and to wetting. At present, few experimental data are available. Moreover, large discrepancies can exist between different sets of data. The high thermal conductivity of liquid metals is mainly due to free electrons. A simple theoretical relation exists for pure metals between electrical and thermal conductivities known as Wiedemann-Franz-Lorenz law [Kittel, 1956]:

$$\lambda_e = L_0 \cdot T / r \qquad (2.69)$$

where λ_e is the electronic thermal conductivity, r is the electrical resistivity and $L_0 = 2.45 \cdot 10^{-8}$ W Ω K^2 is the Lorenz Number.

This law has been confirmed for many liquid metals [Giordanengo, 1999]. The contribution of phonons to the thermal conductivity of metals of interest is small and can be neglected in the temperature range of 400-1200 K (127-927°C). Therefore, the approximate prediction of thermal conductivity of liquid metals and alloys is possible by combining the Wiedemann-Franz-Lorenz law and the existing, reliable values of the electrical resistivity.

2.17.1 Lead

The following data on lead thermal conductivity were analysed for the handbook: [Lyon, 1954], [Kutateladze, 1959], [Crean, 1964], Friedland [Friedland, 1966], Smithells [Smithells, 1983, 2004], [Jauch, 1986], [Iida, 1988], [Millis, 1996], [Kyrillov, 2000b], [TECDOC-1289, 2002] and Yamasue.

The results reported by R.N. Lyon [Lyon, 1954] and by S.S. Kutateladze, *et al*. [Kutateladze, 1959] show very different temperature dependencies: the lead thermal conductivity decreases with temperature in the first, while it increases in the second. Smithells Metal Reference Book [Smithells, 1983, 2004] cited [Ho, 1968] and [Davies, 1970] as sources. T. Iida and R.I.L. Guthrie [Iida, 1988] used values taken from [Powell, 1972]. K.C. Millis, *et al*. [Millis, 1996] considered many sources: [Touloukian, 1970], [Hemminger, 1989], [Filippov, 1973], [Banchola, 1973], [Wittenberg, 1973], [Duggin, 1972], [Osipenko, 1970], [Nakamura, 1990] and recommended the linear correlation for the temperature dependence of thermal conductivity of molten metals.

Friedland [Friedland, 1966] used values given in [Lyon, 1954] and in [Kutateladze, 1959]. U. Jauch and B. Shultz [Jauch, 1986] took their data from [Touloukian, 1970], that have been taken into account in [Millis, 1996]. These sources were not included in the database.

Figure 2.17.1 shows the values for lead thermal conductivity taken from the literature cited above. A very large dispersion of data is present from different sources. A decrease of the lead thermal conductivity with temperature reported by Lyon [Lyon, 1954] (also by L.E. Crean, W.E. Parkins [Crean, 1964], and M.J. Duggin [Duggin, 1972]) and an anomaly reported by S.S. Kutateladze, *et al*. [Kutateladze, 1959] can be attributed to oxidation effects.

Figure 2.17.1. Thermal conductivity of molten lead versus temperature
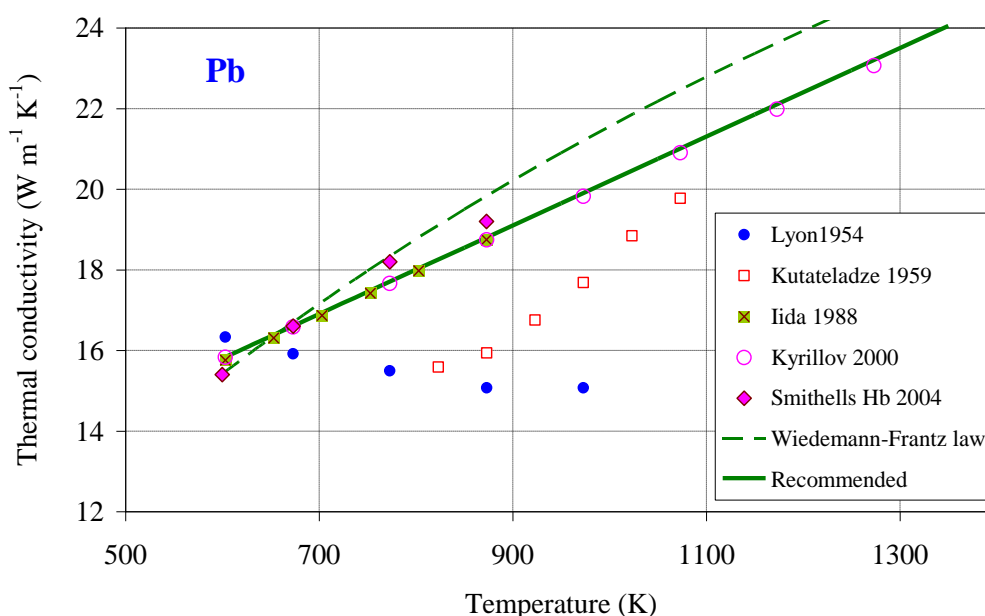
In an effort to find a physically reasonable compromise between the available data and taking into account the Wiedemann-Franz-Lorenz law, a linear correlation was chosen as recommendation for the thermal conductivity of molten lead:

$$\lambda_{Pb}\left[\text{W m}^{-1}\text{ K}^{-1}\right] = 9.2 + 0.011 \cdot T \qquad (2.70)$$

where temperature is in kelvins. This correlation is applicable in the temperature range of T_{melt}-1300 K (1027°C).

2.17.2 Bismuth

The data for bismuth thermal conductivity reported in this handbook were taken from Lyon [Lyon, 1954], Kutatuladze [Kutatuladze, 1959], Crean and Parkins [Crean, 1964], Smithells Metal Reference Book [Smithells, 1983, 2004], Iida and Guthrie [Iida, 1988] and Millis, *et al.* [Millis, *et al.*, 1996].

Smithells [Smithells, 1983] recommended the bismuth thermal conductivity values based on the results of [Samsonov, 1969], [Viswanath, 1972], [Bush, 1963] and [Stull, 1956]. Iida and Guthrie [Iida, 1988] used values taken from Powel [Powel, 1972]. Millis [Millis, 1996] initially reviewed the available data on bismuth thermal conductivity from the compilation of Touloukian [Touloukian, 1970] and then analysed more recent data. He used linear correlations expressing thermal conductivity as a function of temperature. Satisfactory agreement was found between values measured by Filippov [Filippov, 1973] and those derived using the Wiedemann-Franz-Lorenz law and the electric resistivity reported by [Iida, 1988].

Figure 2.17.2 shows the values of thermal conductivity taken from the literature for liquid bismuth, plotted in the range of temperatures of interest.

Figure 2.17.2. Thermal conductivity of molten bismuth versus temperature
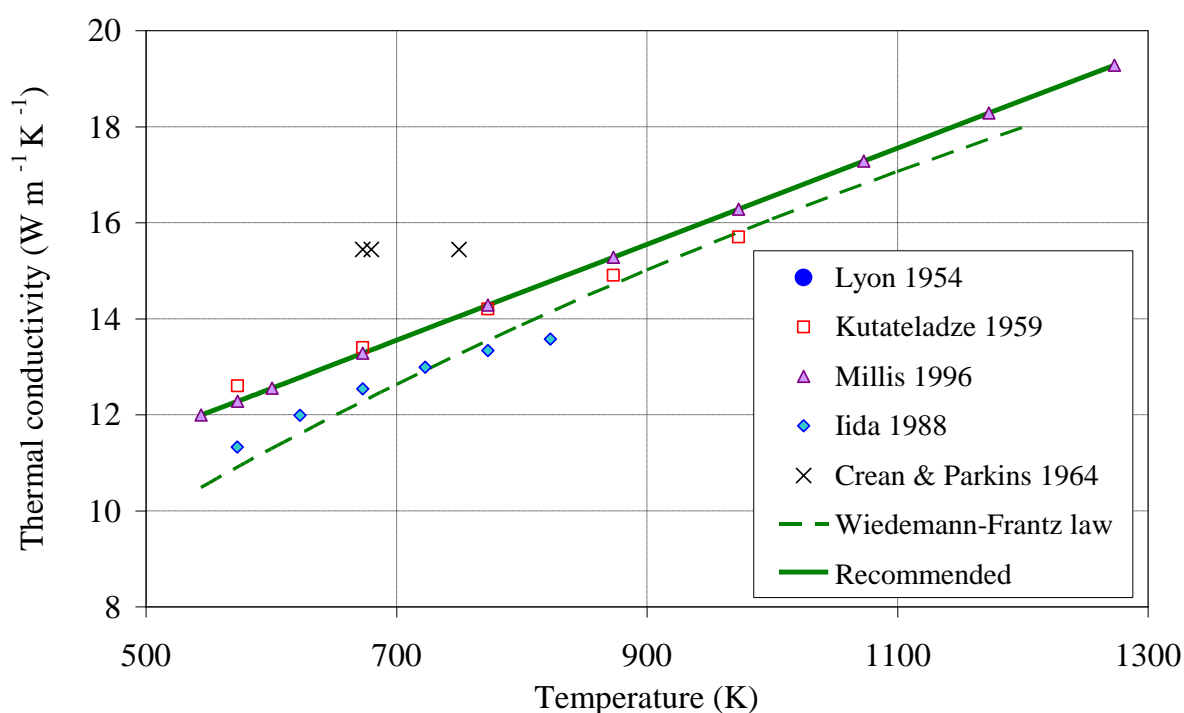
A discrepancy exists between the values taken from [Crean, 1964], that indicate a decrease of the Bi thermal conductivity with temperature up to 650 K (377°C) (not shown) and a constant value at higher temperatures, and the data given by the other sources. This source was not included in the database.

In this version of the handbook, the following empirical correlation recommended by K.C. Millis, *et al*. [Millis, *et al*., 1996] was chosen:

$$\lambda_{Bi}\left[W\,m^{-1}\,K^{-1}\right] = 12 + 1\cdot 10^{-2}\cdot\left(T - T_{melt}\right) = 6.55 + 1\cdot 10^{-2}\cdot T \qquad (2.71)$$

where temperature is in kelvins, and which is applicable in the temperature range from T_{melt} to about 1000 K (727°C).

2.17.3 LBE

Information on thermal conductivity for molten LBE mainly comes from the old handbooks [Lyon, 1954] and [Kutateladze, 1959]. Later handbooks or reviewers reference these data. Only in the handbook of Iida [Iida, 1988] did some values appear that differ from those cited above. In the Russian literature new data were summarised in the handbook of Kirillov [Kirillov, 2000b] without direct references to the sources. Later, the last recommendations were practically reproduced in [TECDOC-1289, 2002]. The data from these four sources for LBE thermal conductivity were included in the database described in Table 2.17.1.

Table 2.17.1. Thermal conductivity database for the melted LBE

Reg. no.	Parameter	Measurement method	Estimated accuracy %	Temperature range, K	Pressure range, Pa	Values	Ref.
17.3.1	Thermal conductivity	Absolute	5% (?)	473-593	~ 10^5		[Lyon, 1954]
	Interpolation function: $\lambda_{LBE}(T) = 3.48 + 0.0129 \cdot T$ W m^{-1} K^{-1}						
	Comments: 44.5 wt.% Pb + 55.5 wt.% Bi. Unknown purity.						
17.3.2	Thermal conductivity	Comparative	?	403-973	~ 10^5 (?)		[Kutateladze, 1959]
	Interpolation function: $\lambda_{LBE}(T) = 6.86 + 0.0102 \cdot T$ W m^{-1} K^{-1}						
	Comments: 44.5 wt.% Pb + 55.5 wt.% Bi. Unknown purity.						
17.3.3	Thermal conductivity	?	?		~ 10^5 (?)		[Iida, 1988]
	Interpolation function: $\lambda_{LBE}(T) = -1.25 + 3.04 \cdot 10^{-2} \cdot T - 1.343 \cdot 10^{-5} \cdot T^2$ W m^{-1} K^{-1}						
	Comments:						
17.3.4	Thermal conductivity	?	?	403-1073	~ 10^5 (?)		[Kyrillov, 2000a]
	Interpolation function: $\lambda_{LBE}(T) = 7.03 + 0.000993 \cdot T$ W m^{-1} K^{-1}						
	Comments: 44.5 wt.% Pb + 55.5 wt.% Bi. Unknown purity.						

The values of the LBE thermal conductivity extracted from the selected sources are presented in Figure 2.17.3 together with the recommended values for lead and bismuth.

Figure 2.17.3. Thermal conductivity of molten lead, bismuth and LBE versus temperature
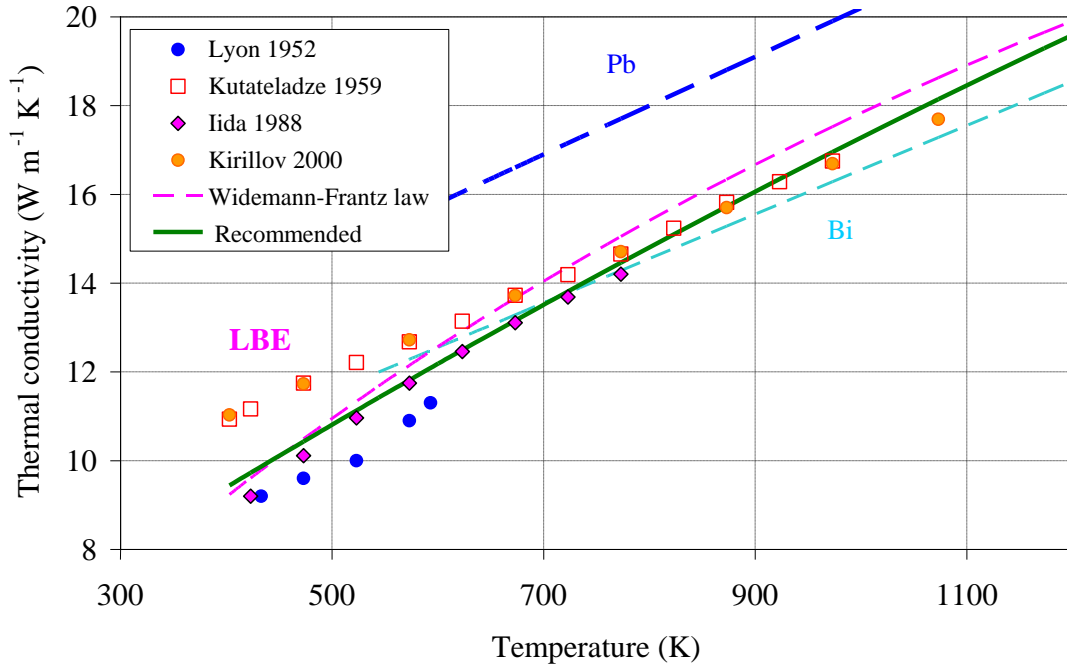
A non-negligible difference exists between the old Western and Russian recommendations, the latter giving 15-20% higher values. The recommendation of Iida [Iida, 1988] provides intermediate values. The data from the handbook of Kyrillov [Kyrillov, 2000a] probably reproduce the old results of Kutateladze [Kutateladze, 1959].

Similar to viscosity and electric resistivity, the results on thermal conductivity are strongly affected by the purity of LBE. Thermal conductivity is not an additive parameter and therefore the Vegard-Kopp law can not be used to estimate it. Contrarily, the Wiedemann-Franz law, which is valid for most metals, can be used.

Substituting in Eq. (2.69) the above recommended correlation for the LBE electrical resistivity, one can obtain the following correlation for the thermal conductivity of molten LBE:

$$\lambda \left[\text{W m}^{-1}\,\text{K}^{-1} \right] \approx \lambda_e = \frac{2.45 \cdot T}{86.334 + 0.0511 \cdot T} \tag{2.72}$$

where temperature is in kelvins.

Eq. (2.72) gives values close to the recommendations of Iida [Iida, 1988] at lower temperatures: 400-550 K (127-277°C), and close to the recommendations of [TECDOC-1289, 2002] at higher temperatures: 600-1100 K (327-827°C).

Fitting the data of Lyon [Lyon, 1954], Kutateladze [Kutateladze, 1959] and Iida [Iida, 1988] with a parabolic function yields the following recommended correlation:

$$\lambda \left[\text{W m}^{-1}\,\text{K}^{-1} \right] = 3.61 + 1.517 \cdot 10^{-2} \cdot T - 1.741 \cdot 10^{-6} \cdot T^2 \tag{2.73}$$

where temperature is in kelvins.

The difference between the selected data and the recommended Eq. (2.73) is plotted in Figure 2.17.4.

**Figure 2.17.4. Deviation of the available data set on
thermal conductivity of molten LBE from Eq. (2.73)**
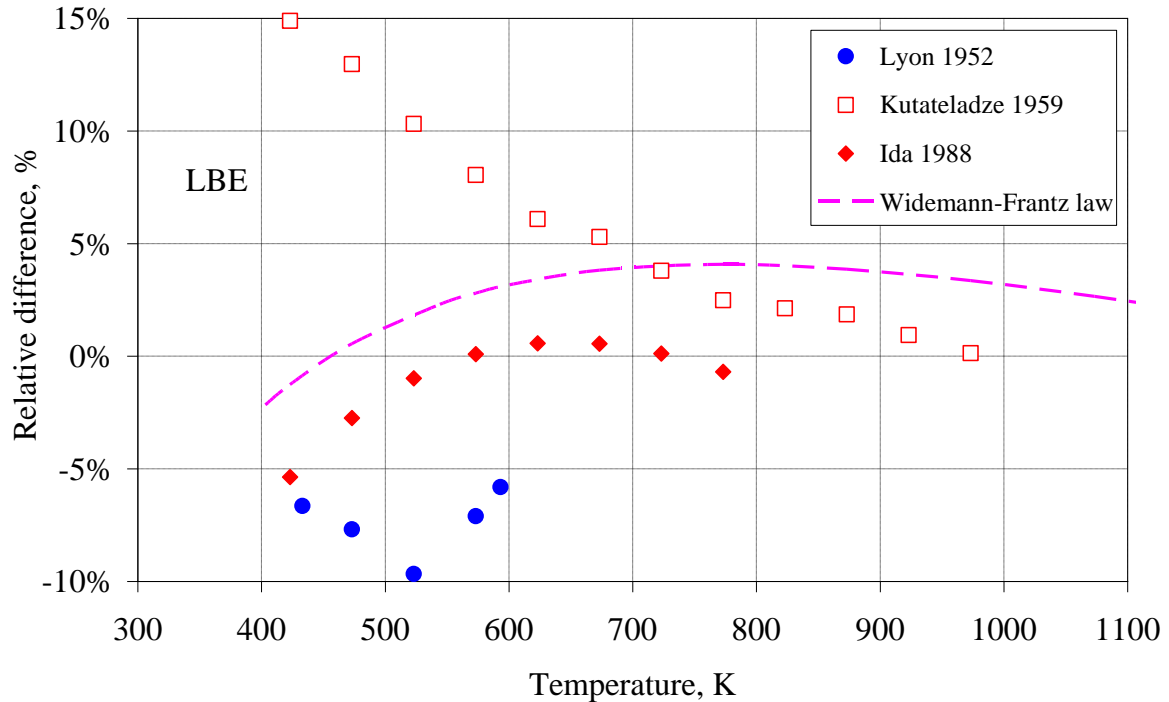
The maximum deviation is 10-15% at low temperatures. The Wiedemann-Frantz law can be used for an estimate of the LBE thermal conductivity if its electrical resistivity is known.

Thermal diffusivity is defined as follows:

$$a_p = \frac{\lambda}{\rho \cdot c_p} \tag{2.74}$$

So, it can be calculated using data for thermal conductivity, density and specific heat. Fitting the obtained data with a linear function yields the following correlation for LBE which can be recommended for practical use:

$$a_{p\,\mathrm{LBE}}\left[\mathrm{m\,s^{-2}}\right] = \left(1.408 + 0.0112 \cdot T\right) \cdot 10^{-6} \tag{2.75}$$

where temperature is in kelvins.

The calculated thermal diffusivities of lead, bismuth and LBE are plotted as a function of temperature in Figure 2.17.5.

Figure 2.17.5. Thermal diffusivity of molten lead, bismuth and LBE
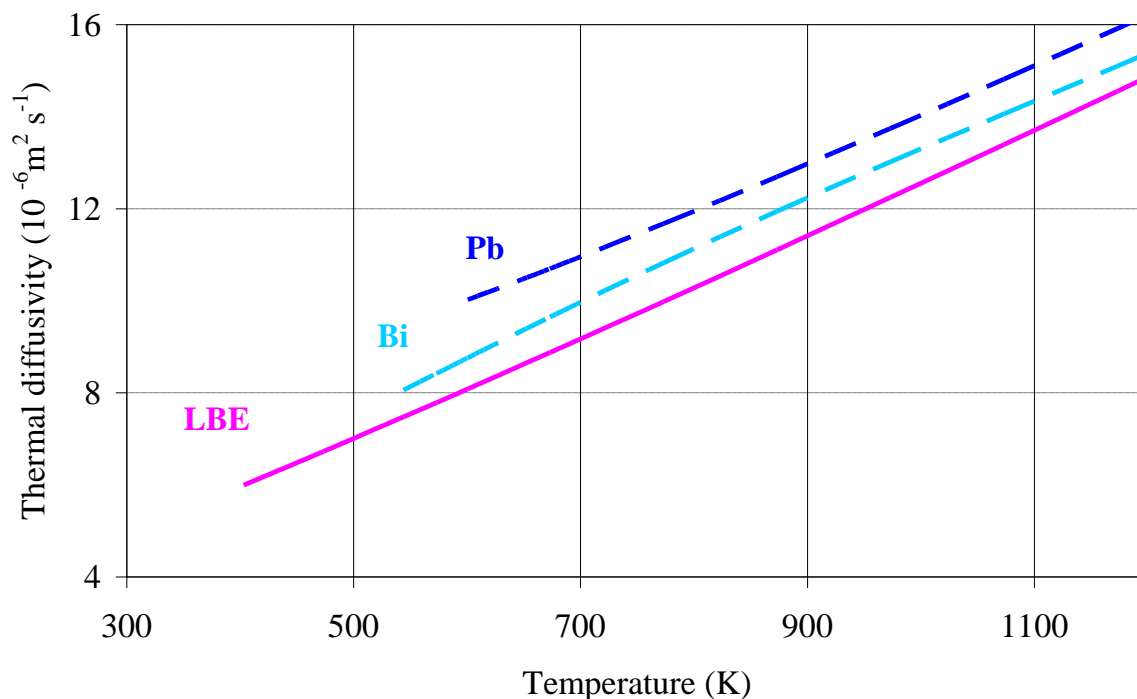
2.18 Conclusions

The data on thermophysical properties of molten lead, bismuth and LBE are generally characterised by significant uncertainty (the exception is the melting point). In general, the reliability of data is satisfactory and good agreement among the different sources of experimental data is often observed. However, a large uncertainty still exists on heat capacity and thermal conductivity – different sources give even different temperature dependence of these parameters. A significant uncertainty also exists in the boiling temperature. The critical temperatures, the critical densities and the critical pressures of Pb, Bi and LBE are not well defined, and this hinders the development of equations of states for these coolants.

On the basis of the review of the data and recommendations available in the literature and of the recent compilations performed in ENEA and SCK•CEN for thermophysical properties of molten lead, bismuth and LBE, correlations are proposed that can serve as temporary recommendations for engineering estimations and design calculations.

The recommended correlations for main thermophysical properties of molten lead, bismuth and LBE are summarised in Tables 2.18.1 to 2.18.3.

Table 2.18.1. Summary of the recommended correlations for main thermophysical properties of molten lead ($p \sim 0.1$ MPa)

Property, parameter	SI unit	Correlation	Temperature range (K)	Estimated error ±
Melting temperature	K	$T_{melt} = 600.6$	n/a	0.1
Latent heat of melting	kJ kg^{-1}	$Q_{melt} = 23.8$	n/a	0.7
Boiling temperature	K	$T_{boil} = 2016$	n/a	10
Latent heat of boiling	kJ kg^{-1}	$Q_{boil} = 858.2$	n/a	1.9
Saturated vapour pressure	Pa	$P_s = 6.189 \times 10^9 \cdot \exp(-22216/T)$	610-2016	15%
Surface tension	N m^{-1}	$\sigma = 0.519 - 1.13 \times 10^{-4} \cdot T$	601-1200	5%
Density	kg m^{-3}	$\rho = 11367 - 1.1944 \cdot T$	601-1900	0.7%
Sound velocity	m s^{-1}	$u_{sound} = 1951.75 - 0.3423 \cdot T + 7.635 \times 10^{-5} \cdot T^2$	601-2000	0.05% ?
Bulk modulus	Pa	$B_s = (42.15 - 1.652 \times 10^{-2} \cdot T + 3.273 \times 10^{-6} \cdot T^2) \times 10^9$	601-2000	–
Isobaric specific heat	J kg^{-1} K^{-1}	$c_p = 175.1 - 4.961 \times 10^{-2} \cdot T + 1.985 \times 10^{-5} \cdot T^2 - 2.099 \times 10^{-9} \cdot T^3 - 1.524 \times 10^6 \cdot T^{-2}$	601-1300	7% ?
Dynamic viscosity	Pa s	$\eta = 4.55 \times 10^{-4} \cdot \exp(1069/T)$	601-1470	4%
Electric resistivity	Ω m	$r = (66.6 + 0.0479 \cdot T) \times 10^{-8}$	601-1300	1%
Thermal conductivity	W m^{-1} K^{-1}	$\lambda = 9.2 + 0.011 \cdot T$	601-1300	10% ?

Table 2.18.2. Summary of the recommended correlations for main thermophysical properties of molten bismuth ($p \sim 0.1$ MPa)

Property, parameter	SI unit	Correlation	Temperature range (K)	Estimated error ±
Melting temperature	K	$T_{melt} = 544.4$	n/a	0.3
Latent heat of melting	kJ kg^{-1}	$Q_{melt} = 52.6$	n/a	1.7
Boiling temperature	K	$T_{boil} = 1806$	n/a	51
Latent heat of boiling	kJ kg^{-1}	$Q_{boil} = 857$	n/a	20
Saturated vapour pressure	Pa	$p_s = 2.4723 \cdot 10^{10} \cdot \exp(-22858/T)$	600-1806	70%
Surface tension	N m^{-1}	$\sigma = 0.4255 - 8.0 \times 10^{-5} \cdot T$	545-1300	5%
Density	kg m^{-3}	$\rho = 10726 - 1.2208 \cdot T$	545-1300	0.4%
Sound velocity	m s^{-1}	$u_{sound} = 2111.3 - 0.7971 \cdot T$	545-603	0.1% ?
Bulk modulus	Pa	$B_s = (44.67 - 0.0299 \cdot T) \times 10^9$	545-603	–
Isobaric specific heat	J kg^{-1} K^{-1}	$c_p = 118.2 - 5.934 \times 10^{-3} \cdot T + 71.83 \times 10^5 \cdot T^{-2}$	545-1300	7% ?
Dynamic viscosity	Pa s	$\eta = 4.458 \times 10^{-4} \cdot \exp(775.8/T)$	545-1300	5%
Electric resistivity	Ω m	$r = (97.1 + 0.05534 \cdot T) \times 10^{-8}$	545-1420	0.8%
Thermal conductivity	W m^{-1} K^{-1}	$\lambda = 6.55 + 0.01 \cdot T$	545-1000	10% ?

Table 2.18.3. Summary of the recommended correlations for main thermophysical properties of molten LBE ($p \sim 0.1$ MPa)

Property, parameter	SI unit	Correlation	Temperature range (K)	Estimated error ±
Melting temperature	K	$T_{melt} = 397.7$	n/a	0.6
Latent heat of melting	kJ kg^{-1}	$Q_{melt} = 38.6$	n/a	0.2
Boiling temperature	K	$T_{boil} = 1943$	n/a	10
Latent heat of boiling	kJ kg^{-1}	$Q_{boil} = 854$	n/a	2.0
Saturated vapour pressure	Pa	$p_s = 11.1 \times 10^9 \cdot \exp(-22552/T)$	508-1943	50%
Surface tension	N m^{-1}	$\sigma = (437.1 - 0.066 \cdot T) \times 10^{-3}$	423-1400	5.0%
Density	kg m^{-3}	$\rho = 11096 - 1.3236 \cdot T$	403-1300	0.8%
Sound velocity	m s^{-1}	$u_{sound} = 1773 + 0.1049 \cdot T - 2.873 \times 10^{-4} \cdot T^2$	403-1300	–
Bulk modulus	Pa	$B_s = (35.18 - 1.541 \times 10^{-3} \cdot T - 9.191 \times 10^{-6} \cdot T^2) \cdot 10^9$	430-605	0.05%
Isobaric specific heat	J kg^{-1} K^{-1}	$c_p = 159 - 2.72 \times 10^{-2} \cdot T + 7.12 \times 10^{-6} \cdot T^2$	430-605	7% ?
Dynamic viscosity	Pa s	$\eta = 4.94 \times 10^{-4} \cdot \exp(754.1/T)$	400-1100	5%
Electric resistivity	Ω m	$r = (86.334 + 0.0511 \cdot T) \cdot 10^{-8}$	403-1100	6%
Thermal conductivity	W m^{-1} K^{-1}	$\lambda = 3.61 + 1.517 \times 10^{-2} \cdot T - 1.741 \times 10^{-6} \cdot T^2$	403-1100	5% ?

REFERENCES

Ageron, P., *et al.* (1959), *Technologie des réacteurs nucléaires*, Tome I – Matériaux, Edition EYROLLES, Paris.

Agostini, P., *et al.* (2004), *J. Nucl. Mater.*, 335, 275.

Alchagirov, B.B., T.M. Shamparov, G.A. Mozgovoi (2003), *High Temperature*, 41(2), 210, translated from *Teplofizika Vysokikh Temperatur*, Vol. 41, No. 2, pp. 247-253.

Allen, B.C. (1972), *Liquid Metals*, S.Z. Beer, Dekker.

A. Azad (2005), *J. Nucl. Mater.*, 341, 45.

Banchila, S.N., L.P. Filippov (1973), *High Temp.*, 11, 602 (USSR).

Baker, H. (Ed.) (1992), *ASM Handbook, Vol. 3 Alloy Phase Diagrams*, ASM International, Metals Park, Ohio, USA.

Barin, I., *et al.* (1989), *Thermochemical Data of Pure Substances*, Vol. 2, VCH.

Beyer, R.T., E.M. Ring (1972), *Liquid Metals*, S.Z. Beer, Dekker.

Bonilla, C.F. (1964), *Heat Transfer*, Chapter 2 in "Reactor Handbook", 2nd Ed. (rev., enl.), Vol. IV, Engineering, S. McLain and J.H. Martens (Eds.), Interscience Publishers, New York, USA, p. 46.

J.L. Bretonnet (1988), *Conductivité électrique des métaux liquides*, Techniques de l'Ingénieur, Materiaux métal-liques, n° M153, Form M69, 1.

Bush, G., J. Tieche (1963), *Phys. Condens. Matter*, 1, 78.

Cahill, J.A., A.D. Kirshenboum (1963), *J. Inorg. Nucl. Chem.*, 25, 501.

Cahill, J.A., G.M. Krieg, A.V. Grosse (1968), *J. Chem. Eng. Data*, 13, 504.

Cailletet, L., E. Matthias (1986), *Comp. Rend. Acad. Sc.* (Paris), 102, 1202.

Cevolani, S. (1998), *Review of the Liquid Lead-bismuth Alloy Physical Properties*, ENEA Report DT.SBD.00004, 18 p.

Chawla, T.C., *et al.* (1981), *Nucl. Eng. Des.*, 67, 57.

Chase, M.W., *et al.* (Eds.) (1978), "Thermochemical Tables", JANAF, *J. Phys. Chem. Ref. Data*, 7, 793.

Chase, M.W., *et al.* (Eds.) (1982), "Thermochemical Tables", JANAF, *J. Phys. Chem. Ref. Data*, 11, 695.

Chase, M.W., *et al.* (Eds.) (1985), "Thermochemical Tables", JANAF, *J. Phys. Chem. Ref. Data*, 14, Suppl. No. 1, Vol. 2.

Cheynet, B. (1989), "Thermodynamic Properties of Inorganic Materials", Vol. 2, Elsevier.

Cheynet, B., J.D. Dubois, M. Milesi (1996), "Données thermodynamiques des éléments chimiques", *Techniques de L'Ingénieur, Matériaux métalliques*, No. M153, Form M64.

CRC Handbook of Chemistry and Physics (2002), 82nd edition, CRC Press, Boca Raton, Florida, USA.

Crean, L.E., W.E. Parkins (1964), "Liquid Metal Cooled Reactors", Chapter 17 in: *Reactor Handbook*, 2nd Ed. (rev., enl.), Vol. IV, Engineering, S. McLain, J.H. Martens (Eds.), Interscience Publishers, New York, USA.

Cusack, N., J.E. Enderby (1960), *Proc. Phys. Soc.*, 75, 395.

Davies, H.A., J.S.L. Leach (1970), *Phys. Chem. Liquids*, 2, 1.

Dinsdale, A.T. (1991), *CALPAD*, 15, 317.

Douglas, T.B., J.L. Dever (1953), Report NBS-2544, NBS, USA.

Dreyfus, B., A. Lacaze (1971), *Cours de thermodynamique*, Dunod, Paris.

Duggin, M.J. (1972), *J. Phys. F. Met. Phys.*, 2, 433.

Elliott, R.P. (1965), *Constitution of Binary Alloys*, I-Supplement, McGraw-Hill Book Co., Inc., New York, USA.

Eucker, A. (1936), *Metallwirtsch*, 15, 63.

Filippov, L.P. (1973), *Int. J. Heat Mass Transfer*, 16, 865.

Fortov, V.E., A.N. Dremin, A.A. Leont'ev (1975), *High Temp.*, 13, 984.

Friedland, A.J. (1966), "Coolant Properties, Heat Transfer and Fluid Flow of Liquid Metals", in *Fast Reactor Technology: Plant Design*, J.G. Yevick (Ed.), Ch. 2, The M.I.T. Press, Cambridge (MA), USA.

Gasser, J.G., *et al.* (1984), *J. Non-Cryst. Solids*, 61-62, 1237.

Gebhardt, E., M. Becker, H. Tragner (1955), *Z. Metallk.*, 46, 90.

Giordanengo, B., *et al.* (1999), *J. Non-Cryst. Solids*, 250-252, 377.

Giuranno, D., *et al.* (2002), *Intermetallics*, 11, 1313.

Glasbrenner, H., *et al.* (2006), *J. Nucl. Mater.* (forthcoming in 2006).

Glushko, W.P., V.A. Medvedev (Eds.) (1981), *Termicheskie Konstanty Veschestv*, Part III, N, P, As, Sb, Bi (1968); Part IV, C, Si, Ge, Sn, Pb (1970); Part X, Alkaline Metals, *Akad. Nauk*, Moscow, USSR.

Gmelins Handbuch der Anorganischen Chemie (1972), 8, Auflage, Yeil B1, Syst.-Nr.47, Verlag Chemie GmbH, Weinheim.

Gokcen, N.A. (1992), *J. Phase Equilibria*, 13, 21.

Gordon, R.B. (1959), *Acta Met.*, 7, 1.

Gromov, B.F., *et al*. (1999), "The Problems of Technology of the Heavy Liquid Metal Coolants (Lead-bismuth, Lead)", *Proc. Conf. on Heavy Liquid Metal Coolants in Nuclear Technology (HLMC'98)*, 5-9 October 1998, Obninsk, Russian Federation, Obninsk: SSC RF-IPPE, Vol. 1, p. 87.

Gross, P., *et al*. (1948), *Disc. Faraday Soc.*, 4, 206.

Grosse, A.F. (1961), *J. Inorg. Nucl. Chem.*, 22, 23.

Grosse, A.V. (1966), *Revue haute temp. & refrac.*, 3, 115.

Gröschel, G.F., J. Patorski (2005), *PSI Scientific Report 2004*, Volume III, p. 200, ISSN 1423-7326, March 2005.

Gurvich, L.V., I.V. Veyts (1991), *Thermodynamic Properties of Individual Substances*, 4th ed., Vol. 2, Parts 1 and 2, Hemisphere Pub. Corp., New York.

Hansen, M., K. Anderko (1958), *Constitution of Binary Alloys*, 2nd ed., McGraw-Hill Book Co., Inc., New York, USA.

Hemminger, W. (1989), *Int. J. Thermophys.*, 10, 765.

Hiers, G.O. (1948), *Metals Handbook*, Cleveland, Ohio, USA.

Ho, C.Y., R.W. Powell, P.E. Liley (1968), *NSRDS-NBS Report*, 16 Feb. 1968.

Hofmann, W. (1970), *Lead and Lead Alloys*, Springer-Verlag, Berlin, New York.

Holman, J.P. (1968), *Heat Transfer*, MacGraw-Hill, USA.

Howe, H.E. (1961), "Bismuth", in *Rare Metals Handbook*, 2nd ed., C.A. Hampel, Reinhold Publishing Corp.

Hultgren, R., *et al*. (1963), *Selected Values of the Thermodynamic Properties*, J. Wiley, USA.

Hultgren, R., *et al*. (1973), *Selected Values of the Thermodynamic Properties of Binary Alloys*, ASM, Metals Park, Ohio, USA.

Hultgren, R., *et al*. (1974), *Selected Values of the Thermodynamic Properties of the Elements*, J. Wiley, New York, USA.

Iida, T., Z. Morita, S. Takeuchi (1975), *J. Japan Inst. Metals*, 39, 1169.

Iida, T., R.I.L. Guthrie (1988), *The Physical Properties of Liquid Metals*, Clarendon Press, Oxford, UK.

Imbeni, V., *et al.* (1998a), "Stato dell'arte sulle proprietà chimico-fisiche del Pb e Pb-Bi. Parte I. Proprietà di Pb, Bi, Li, Na", in *Studio di un acceleratore superconduttivo di protoni di grande potenza e studio di un sistema sottocritico da esso sostenuto per il bruciamento dei residui radioattivi, Sottotema 2: Prove di corrosione, BoMet*, Accordo di programma ENEA/INFN-MURST, Settore Ambiente, December 1998 (in English).

Imbeni, V., *et al.* (1998b), "Stato dell'arte sulle proprietà chimico-fisiche del Pb e Pb-Bi. Parte II. Proprietà di Pb e Pb-Bi, Na", in *Studio di un acceleratore superconduttivo di protoni di grande potenza e studio di un sistema sottocritico da esso sostenuto per il bruciamento dei residui radioattivi, Sottotema 2: Prove di corrosione, BoMet*, Accordo di programma ENEA/INFN-MURST, Settore Ambiente, December 1998 (Rev. 18.01.99) (in English).

Jarzynski, J. (1963), *Proc. Phys. Soc.*, 81, 745.

Jauch, U., B. Schulz (1986), *Estimation of the Thermophysical Properties in the System Li-Pb*, KfK 4144, Karlsruhe.

Joud, J.C., *et al.* (1972), C.*R. Acad. Sc. Paris*, Série C274, 549.

Kaplun, B., *et al.* (1979), "The Viscosity of Lead-bismuth Eutectic Alloy", in *Teplofizicheskie svoystva veshestv i materialov*, Novosibirsk.

Kasama, A., T. Iida, Z. Morita (1976), *J. Japan Inst. Metals*, 40, 1030.

Kazakova, I.V., S.A. Lyamkin, B.M. Lepinskikh (1984), *Russ. J. Phys. Chem.*, 58, 932.

Kažys, R., *et al.* (2002), "Investigation of Ultrasonic Properties of a Liquid Metal Used as a Coolant in Accelerator Driven Reactors", *2002 IEEE International Ultrasonics Symposium Proceedings*, ISSN 1051-0117, Munich, pp. 794-797.

Kelley, K.K. (1960), *Evaluation of High-temperature Heat Capacities of Inorganic Compounds*, US Bur. Mines Bull.

Kelley, K.K. (1935), *Critical Evaluations of Vapour Pressures and Heats of Evaporation of Inorganic Substances*, US Bur. Mines Bull., No. 393.

Keene, B.J. (1993), *Int. Met. Reviews*, 38, 157.

Kirshenbaum, A.D., J.A. Cahill, A.V. Grosse (1961), *J. Inorg. Nucl. Chem.*, 22, 33.

Kittel, C. (1956), *Introduction to Solid State Physics*, Willey, NY.

Kleppa, O.J. (1950), *J. Cem Phys.*, 18, 1331.

Knacke, O., O. Kubaschewski, K. Hesselmann (1991), *Thermodynamic Properties of Inorganic Substances*, Vol. 2, 2nd ed., Springer-Verlag.

Kohlraush, F. (1956), *Praktische Physik*, II, Stuttgart: Teubner.

Konyuchenko, G.V. (1969), *Teplofizika Vysokikh Temperatur*, 10, 309.

Kubaschewski, O., C.B. Alcock (1979), *Metallurgical Thermochemistry*, 5th ed., Pergamon Press, NY.

Kubaschewski, O., C.B. Alcock, P.J. Spencer (1993), *Materials Thermochemistry*, 6th ed. Pergamon Press, NY.

Kutateladze, S.S., *et al.* (1959), "Liquid-metal Heat Transfer Media", *At. Energ.*, Suppl. 2 (transl. Cons. Bur. Inc., New York, 1959).

Kyrillov, P.L., G.P. Bogoslovskaya (2000a), "Heat Transfer in Nuclear Installations", *Energoatomisdat*, Moscow, 2000 (in Russian).

Kyrillov, P.I., N.B. Deniskina (2000b), *Thermophysical Properties of Liquid Metal Coolants. Tables and Correlations*, Review FEI-0291, Obninsk, IPPE, 2000 (in Russian).

Lang, G., *et al.* (1977), *Z.Metallk.*, 68, 113.

Lucas, L.D. (1969), *C.R. Acad. Sci.*, Série C 268, 1081.

Lucas, L.D. (1984a), "Données physico-chimiques des principaux métaux et métalloïdes", *Techniques de l'Ingénieur, Matériaux métalliques*, N° M153, Form M65a.

Lucas, L.D. (1984b), "Viscosité", *Techniques de l'Ingénieur, Matériaux métalliques*, N° M153, Form M66.

Lyon, R.N. (Ed.) (1954), *Liquid Metals Handbook*, 2nd Ed., Report NAVEXOS P-733, Atomic Energy Commission and Dept. of the Navy, Washington, USA, June 1952 (rev. 1954).

Lyon, R.N. (1960), "Liquid Metals", Chapter 49 in: *Reactor Handbook*, 2nd Ed.(rev., enl.), Vol. I, Materials, Sect. F (Coolant Materials), C.R. Tipton, Jr. (Ed.), Interscience Publishers, New York, USA.

Mantell, C.L. (Ed.) (1958), *Engineering Material Handbook*, MacGraw-Hill, USA.

Martynyuk, M.M. (1998), *Russ. J. Phys. Chem.*, 72, 13.

Martynyuk, M.M. (1983), *Russ. J. Phys. Chem.*, 57, 494.

Mera, Y., Y. Kita, A. Adachi (1972), Technical Report, Osaka University, 22, 445.

Michelato, P., *et al.* (2003), "The TRESCO-ADS Project Windowless Interface: Theoretical and Experimental Evaluations", *Proc. Int Workshop on P&T and ADS Development*, Mol, Belgium, 6-8 October 2003.

Miller, R.R. (1951), "Physical Properties of Liquid Metals", in *Liquid Metal Handbook*, NAVEXOS, R.N. Lyon (Ed.), US Gov. Pt. Office, Washington DC, 2nd edition (rev.), ORNL, Tennessee.

Millis, K.C., B.J. Monaghan, B.J. Keene (1996), *Int. Mat. Reviews*, 41, 209.

Morita, K., E.A. Fischer (1998a), *Nucl. Eng. Des.*, 183, 177.

Morita, K., E.A. Fischer, K. Thurnay (1998b), *Nucl. Eng. Des.*, 183, 193.

Morita, K., *et al.* (2004), "Thermophysical Properties of Lead-bismuth Eutectic Alloy for Use in Reactor Safety Analysis", 2nd Meeting of the NEA Nuclear Science Committee, Working Group on LBE Technology of the Working Party on Scientific Issues of the Fuel Cycle (WPFC), Issy-les-Moulineaux, France, 23-24 Sept. 2004.

Morita, *et al.* (2005), "Thermodynamic Properties of Lead-bismuth Eutectic for Use in Reactor Safety Analysis", *Proc. 13th Int. Conf. on Nuclear Engineering (ICONE-13)*, ICONE13-50813, Beijing, China, 16-20 May 2005.

Mustafin, G.M., G.F. Shaikhiev (1983), *Russ. J. Phys. Chem.* 57, 421.

Nakamura, S., T. Hibiya, E. Yamamoto (1990), *J. Appl. Phys.*, 68, 5125.

Nikolskii, N.A., *et al*. (1959), "Thermal and Physical Properties of Molten Metals and Alloys", in *Problems of Heat Transfer*, M.A. Micheev (Ed.), Academy of Sciences of USSR, Moscow, pp. 1-38 (in Russian).

Nosek, M.V., G.V. Sang-Sho-Hsyan, N.M. Semibratova (1967), *Tr. Inst. Khim*, Nauk Akad. Nauk Kaz., SSR 15, 150.

Novacovic, R., *et al.* (2002), *Surface Science*, 515, 377.

Ofte, D., L.J. Wittenberg (1963), *Trans AIME*, 227, 706.

Ofte, D. (1967), *J. Nucl. Mater*., 22, 28.

Ohno, S., S. Tamaki (1975), *J. Phys. Soc. Japan*, 38, 538.

Ohno, S., S. Miyahara, Y. Kurata (2005), *J. Nucl. Sci. and Techn*. 42, 593.

Orlov, Y.I. (1997), "Technology of PbBi and Pb Coolants", *Seminar on the Concept of Lead-cooled Fast Reactor*, Cadarache, France, 22-23 Sept. 1997.

Orlov, Yu., B.F. Gromov, V.A. Gylevskiy (2003), "Bases of Technology of Pb-Bi Coolant", *ATW Workshop*, Washington DC, 16-18 February 2003.

Osipenko, V.P. (1970), *Sov. Phys. J.,* 12, 1570.

Pankratz, L.B. (1982), *Thermodynamic Properties of Elements and Oxides*, US Bur. Mines 682, USA.

Pastor Torres, F.C. (2003), *Surface Tension Measurement of Heavy Liquid Metals Related to Accelerator Driven Systems (ADS)*, Diploma Thesis, FZK (IKET), KALLA, April 2003.

Pottlacher, G., H. Jäger (1990), *Int. J. Thermophysics*, 11, 719.

Pottlacher, G. (1998), personal communication.

Powell, R.L., G.E. Childs (1972), Amer. Inst. of Physics, *Handbook*, McGraw-Hill.

Predel, B., .W. Schwerman (1967), *Z. Metallk*., 58, 553.

Pylchenkov, E.H. (1999), "The Issues of Freezing-defreezing Lead-bismuth Liquid Metal Coolant in Reactor Facilities Circuits", *Proc. Conf. on Heavy Liquid Metal Coolants in Nuclear Technology (HLMC'98)*, 5-9 October 1998, Obninsk, Russian Federation, Obninsk: SSC RF-IPPE, Vol. 1, p. 110.

Ruppersberg, H., W. Speicher (1976), *Z. Naturforschung*, 319, 47.

Samsonov, G.V., A.D. Panasyuk, E.M. Dudnik (1969), *Izv. Vuz. Tsvet. Met.*, 12, 110.

Schulz, B. (1991), *Fusion Eng. Des.*, 14, 199.

Schuurmans, P., *et al.* (2006), *R&D Support for a Windowless Liquid Metal Spallation Target in MYRRHA ADS* (forthcoming in 2006).

Semenchenko, V.K. (1961), *Surface Phenomena in Metals and Alloys*, English edition, Pergamon Press, London.

Smithells, C.J. (1955), *Metals*, Reference Book, 2nd Edition, Vol. I. Interscience Punl. Inc. New York, Butterworths Scientific Publ., London.

Smithells Metals Reference Book (1983), *Thermochemical Data*, Chapter 8 "The Physical Properties of Liquid Metals", E.A. Brandes (Ed.), 6th edition, Butterworths, London, UK.

Smithells Metals Reference Book (8th edition) (2004), W.F. Gale, T.C. Totemeier (Eds.), Elsevier, Amsterdam.

Sobolev, V. (2002), *Database of Thermal Properties for the Melted Lead-bismuth Eutectic*, SCK•CEN report IR-18, Mol, Belgium, May 2002.

Sobolev, V. (2005), *MYRRHA ADS Database: Part I. Thermophysical Properties of the Molten Lead-bismuth Eutectic*, SCK•CEN report BLG-1014, Mol, Belgium, August 2005 (rev. December 2005).

Sobolev, V. (2007), *J. Nucl. Mater.* (forthcoming in 2007).

Steinberg, D.J. (1974), *Met. Trans.*, 5, 1341.

Strauss, S.W., L.E. Richards, B.F. Brown (1960), *Nucl. Sci. Eng.* 7, 442.

Stull, D.R., G.C. Sinke (1956), *Thermodynamic Properties of the Elements*, Am. Chem. Soc., Washington, USA.

Stull, D.R., *et al.* (Eds.) (1971), *Thermochemical Tables*, JANAF, US Dept. of Commerce, Natl. Bur. Stand., US Government Printing Office.

Takeda, Y. (1997), *Volume Change of LBE After Solidification*, PSI TM-97-02-01.

Tamamushi, B., *et al.* (Eds.) (1981), *Iwanami Dictionary of Physics and Chemistry (Iwanami rikagaku jiten)*, 3rd ed., Iwanami Shoten Publishers, Tokyo, Japan.

TECDOC-1289 (2002), *Comparative Assessment of Thermophysical and Thermohydraulic Characteristics of Lead, lead-bismuth and Sodium Coolants for Fast Reactors*, IAEA, Vienna.

Touloukian, Y.S., *et al.* (1970), "Thermal Conductivity of Metallic Elements and Alloys", in *Thermophysical Properties of Matter*, Plenum Press, New York.

Tschirner, H.U. (1969), *Z. Metallk.*, 60, 46.

Tupper, R.B., *et al*. (1991), "Polonium Hazards Associated with Lead-bismuth Used as a Reactor Coolant", *Proc. Int. Conf on Fast Reactors and Related Fuel Cycles*, Kyoto, Japan, 28 Oct.-1 Nov. 1991, Vol. 4, pp. 5.6-1 – 5.6-9.

Viswanath, D.S., B.C. Mathur (1972), *Met. Trans.*, 3, 1769.

von Wartenberg, H., R. Schütte (1933), *Z. Anorg. Chem.*, 211, 222.

Watson, K.M. (1931), *Ind. Eng. Chem.*, 23, 360.

Weeks, J.R. (1971), *Nucl. Eng. Des.*, 15, 363.

Wittenberg, L.J., D. Ofte (1970), *Techniques of Metals Research*, Vol. 4, Interscience.

Wittenberg, L.J. (1973), *Thermochim. Acta*, 7, 13.