

## Temperature dependence of the liquid eutectic lead-lithium alloy density

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**Abstract:** Lead-lithium alloys are of great interest for practice as the advanced materials to be used in new technique, nuclear energetics, and so forth. Therefore, study on the physico-chemical properties of the latter is of major significance. An analysis of the available literature shows that there are a few works, devoted to study of Pb-Li alloys densities. However, temperature dependence of the density  $\rho(T)$ , and its temperature coefficient

$K = d\rho/dT$  for eutectic alloy were obtained by either extrapolation of the density data up to the eutectic alloy's composition, or calculation method. There is a certain discrepancy amounting to as high as 4%, while the allowable error in the density measurements is less than 0.5%. The discrepancy between the results for the temperature coefficients of density amounts to 80%.

In this work we present the experimental data on the temperature dependence of  $Pb_{0.83}Li_{0.17}$  eutectic alloy's density in the temperature range 520K to 643 K. The alloys were prepared using Pb and Li with 99.999% and 99.8% contents of the basic elements, respectively. We use the improved device, which permits to get the results with error less than 0.15%. The results of 115 measurements of density in 520K to 643K temperature range were processed by the least-square method. Density polytherm of  $Pb_{0.83}Li_{0.17}$  eutectic alloy is described by linear equation

$$\rho(T) = 9507.89 - 0.79813(T - 508), \text{ kg/m}^3,$$

where  $T$  is the absolute temperature by K. Measurement error was 0.12% at 95% reliability. Discrepancy in the temperature coefficient data was 1.08%.

Thus, the temperature dependence of the Pb-Li eutectic alloy density was studied by the precise two-capillary method. The obtained results may be recommended as the most reliable reference data.

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Lead-lithium alloys are of great interest for practice as the advanced materials to be used in new technique, nuclear energetics, and so forth<sup>[1-3]</sup>. However, the properties of the alloys with lithium are not studied well enough due to their high chemical activity. Therefore, it is necessary to study in detail the physico-chemical properties of the latter, such as density, surface tension, wettability of the construction materials by them and so on. Especially, it is important to know the properties of the liquid eutectic lead-lithium alloy.

There are no the experimental data on the temperature dependence of the  $Pb_{0.83}Li_{0.17}$  eutectic alloy density

in the literature.

The concentration dependence of the Pb-Li alloy's density was studied in the single work only [4], however, the nearest to eutectic alloy's composition among the investigated alloys is Pb+20% (at. fraction) of Li.

Using the experimental results by [4], the equation for the temperature dependence of the  $Pb_{83}Li_{17}$  liquid alloy density has been obtained by extrapolation method in [5]:

$$\rho(T) = 9519 - 0.71(T - 508), \quad (1)$$

where  $T$  and  $\rho$  are measured by K and  $kg/m^3$ , accordingly;  $T_{eut} = 508$  K.

Approximating the results of [7], an equation for the temperature dependence of Pb-Li eutectic alloy's density was given in [6]:

$$\rho(T) = 9495 - 0.695(T - T_{eut}). \quad (2)$$

The density value of the eutectic alloy by eq. (2) is less by  $24 kg/m^3$  that given by eq. (1). In [8] the equation for the eutectic alloy's density polytherms, obtained according to the additivity law, has been suggested. However, there is a considerable deviation of the concentration dependences of the physico-chemical properties of this alloy from the additivity law, due to the well-known geometrical and electrochemical inequality of the atoms of the Pb-Li alloy components [9]. For example, an appreciable negative deviation from the additivity law for the concentration dependences of density,  $\rho(x)$ , of Pb-Li alloy has been found in [4].

Analysis of the available literature on the subject reveals there are only approximate values of the eutectic alloy's density, and a discrepancy between values of temperature coefficients of density data amounts to 30%. So, the precise measurements of the temperature dependence of the Pb-Li eutectic alloy's density are a highly actual problem.

In Table 1, the most essential results on the temperature dependence of the eutectic Pb-Li alloy's density are given.

Table 1 Literature data on the temperature dependence of the eutectic Pb-Li alloy's density

Authors	Temperature dependence of the density	Notes
	$\rho(T) = \rho_m - \frac{d\rho}{dT}(T - T_{eut}), \text{kg/m}^3$	
Jauch U. , Haase G. , Karcher V. , Schulz B. [13], 1983	$\rho_{al} = 10.60(1 - 122 \times 10^{-6} T), \text{g/cm}^3$ $\rho_{liq} = 10.45(1 - 161 \times 10^{-6} T), \text{g/cm}^3$ $\rho(T) = 9595.3 - 1.682(T - T_{eut}), \text{kg/m}^3$	Method of immovable drop. 508 < T < 625, Measurements error is 5 %
Calculations by Coen V. [6] by [7], 1985	$\rho(T) \times 10^{-3} = 9495 - 0.695(t - 235^\circ\text{C})$ $\rho(T)^{11} = 9495 - 0.695(T - T_{eut}), \text{kg/m}^3$	Approximate equation
Calculations by G. Kuhlbornsch and F. Reiter [5] by experimental results of [4], 1984	$\rho(T) \times 10^{-3} = 9519 - 0.710(t - 235^\circ\text{C})$ $\rho(T)^{11} = 9519 - 0.710(T - T_{eut}), \text{kg/m}^3$	Approximate equation
Kalinin G. M. , Sidorenko A. V. , Uchitnov A. G. et al. [14], 1991	$\rho_{al} = 10.69(1 - 80 \times 10^{-6} T), \text{g/cm}^3$ $\rho_{liq} = 10.47(1 - 110 \times 10^{-6} T), \text{g/cm}^3$ $\rho(T)^{11} = 9884.9 - 1.152(T - T_{eut}), \text{kg/m}^3$	Gamma-method. Measurements error is 1%

Authors	Temperature dependence of the density		Notes
	$\rho(T) = \rho_m - \frac{d\rho}{dT}(T - T_m), \text{kg/m}^3$		
Kirillov P. L. , Deniskina N. B. Calculations by the additivity law [8], 2000	$\rho(T) \times 10^{-3} = 9482 - 1.145(t - 235^\circ\text{C}), \text{kg/cm}^3$ $\rho(T)^{1)} = 9482 - 1.145(T - T_m), \text{kg/m}^3$		Calculation by the additivity law
Kirillov P. L. , Deniskina N. B. [8], 2000	$\rho_{\text{LiP}}(T) = 9449 - 0.849(T - T_m), \text{kg/m}^3$		Analysis of the literature data

Note: 1) equations derived by us using the data given in the above mentioned works

In this work we present the experimental results on the temperature dependence of the  $\text{Pb}_{83}\text{Li}_{17}$  eutectic alloy's density, as it is the most interesting alloy for practice<sup>[10]</sup>. The measurements were performed in the 520K to 643 K temperature range in steps of 3 to 4 degrees. The eutectic alloy was prepared under  $10^{-4}$  Pa vacuum condition using the high purity components, such as 99.999% (mass fraction) of Pb and 99.8% (mass fraction) of Li. The temperature measurements error was less than 0.03%.

In the present work we used a completely soldered two-capillary vacuum pycnometer, graduated using a high purity Hg with density equal to  $\rho_{\text{Hg}} = (13.545884 \pm 13 \times 10^{-6}) \text{ g/cm}^3$ <sup>[11]</sup>.

The density measurements error did not exceed 0.15%. Description of the device, the measuring cell and the technique of preparing the samples were given in<sup>[12]</sup>. We obtained more than 80 density values of the liquid  $\text{Pb}_{83}\text{Li}_{17}$  eutectic alloy. In Table 2, we present a part of the experimental results, obtained in the studied temperature range.

The experimental data on the  $\rho(T)$  was processed by the least-square method and it was found that they are well described by linear equation:

$$\rho(T) = 9507.89 - 0.79813(T - 508), \quad (3)$$

where  $\rho$  and  $T$  are expressed by  $\text{kg/m}^3$  and K, respectively.

Table 2 Temperature dependence of the  $\text{Pb}_{83}\text{Li}_{17}$  eutectic alloy's density

No.	T / K	$\rho / (\text{kg} \cdot \text{m}^{-3})$	No.	T / K	$\rho / (\text{kg} \cdot \text{m}^{-3})$	No.	T / K	$\rho / (\text{kg} \cdot \text{m}^{-3})$
1	520.08	9501.32	11	584.88	9446.71	21	622.69	9416.36
2	526.02	9495.25	12	591.05	9441.71	22	625.00	9414.64
3	533.80	9490.08	13	594.91	9441.64	23	627.32	9412.68
4	540.03	9485.10	14	598.77	9435.56	24	629.63	9411.01
5	546.62	9478.81	15	602.62	9432.21	25	631.17	9409.84
6	552.64	9473.62	16	605.71	9430.08	26	632.72	9408.68
7	558.06	9471.62	17	608.80	9426.75	27	635.03	9406.59
8	564.00	9462.34	18	611.88	9425.05	28	638.12	9404.18
9	572.40	9458.42	19	615.74	9422.10	29	639.66	9402.98
10	580.24	9449.98	20	620.37	9418.26	30	641.98	9401.11

It may be concluded from the data by Tables 1 and 2 there is an appreciable divergence between our and the literature results. The average value of the  $\text{Pb}_{83}\text{Li}_{17}$  alloy density at eutectic temperature by literature data is  $(9579.2 \pm 107.3) \text{ kg/m}^3$  with the mean relative error of about 1.12%, while that for the temperature coefficients of density by Table 1 data is about 30%. As it was above mentioned, our results on the  $\text{Pb}_{83}\text{Li}_{17}$  alloy's density are well described by linear equation (3). The eutectic alloy's density value is equal to  $(9511.12 \pm 14.27) \text{ kg/m}^3$  at 508 K with relative error about of 0.15%. The measurements error of the temperature coefficients of density was 1%.

Thus, the temperature dependence of the liquid Pb-Li eutectic alloy's density was studied using the precise two capillary pycnometer method and the high purity samples. In our opinion, the experimental results obtained by us are more precise ones compared with the available literature data, and may be recommended as the reference data.

## References

- [1] Mikhailov V N, Evtikin V A, Lubinski I E, et al. Lithium for Fusion Reactors and Nuclear Power Systems of the XXI Century. M.: Energoatomizdat, 1999, 528.
- [2] Blokhin V A, Ivanovski M N, Kuvshinchikova T A Structure, atomic dynamics, thermodynamics and admixture's state of lead and bismuth melts. (Modern state of the problem). FEI 0290 M; ZNII Atominform. 2000, 76.
- [3] Grjaznov G M, Evtikhin V A, Lubinski I E, et al. Material science of metallic systems for thermonuclear reactors M; Energoatomizdat, 1989.
- [4] Ruppertsberg H, Speicher W. Density and Compressibility of Liquid Li-Pb Alloys. Z Naturforsch, 1976, Bd. 31a. S. 47-52.
- [5] Kuhlbornsch G, Reiter F. Physical properties and chemical reaction behaviour of  $\text{Li}_{17}\text{Pb}_{83}$  related to its use as Fusion reactor blanket material[J]. Nucl Eng Design Fusion, 1984, (1), (2).
- [6] Coen V. Lithium-Lead eutectic as breeding material in Fusion Reactors[J]. Journ of Nuclear Mater, 1985, 133/134: 46-51.
- [7] Holrayd J, Mitchell J T. Liquid Lithium as a Coolant for Tokamak Fusion Reactors[J]. Nucl Eng Design Fusion, 1984, 1(1): 17.
- [8] Kirilov P L, Deniskina N B. Thermophysical properties of liquid metallic heat transfer agents (reference tables and relationships). FEI-0291. M.; ZNII Atominform. 2000, 42.
- [9] Moratchevski A G. Thermodynamics of the metallic and salt melts. M.; Metallurgy, 1987, 240.
- [10] Malang S, Mattas R. Comparison of Lithium and Eutectic Lead - Lithium alloy, two candidate Liquid metal breeder materials for self-cooled blankets[J]. Fusion Engineering and Design, 1955, 28:399-406.
- [11] Fokin L R, Yakovlev A T, et al. The density and thermal expansion coefficient of mercury at the atmospheric pressure and temperatures from 0 to 350°C (Tables of Standard Reference Data). GSSSD 3-77. M.; Izdatel'stvo Standartov, 1987, 8.
- [12] Alchagirov B B, Shamparov T M, Mozgovoï A G. Experimental study of the lead-bismuth eutectic melt's density [J]. TVT. 2003. 41(2): 247-253.
- [13] Jauch U, Haase G, Karcher V, et al. Thermophysical Properties of the System Li-Pb Report KFK-4144. Karlsruhe, FRG, 1983, 82.
- [14] Kalinin G M., Sidorenko A V., Ukhtinov A G, et al. Study of properties of  $\text{Li}_{17}\text{Pb}_{83}$  Eutectic alloy. Preprint ET-91/01. Moscow, USSR. 1991, 92.