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LiPb, A NOVEL MATERIAL FOR FUSION APPLICATIONS

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Abstract

LiPb in various compositions is a unique material for fusion application, combining the breeding material and neutron multiplier. It provides a high tritium breeding ratio, low tritium solubility and, at the Pb rich end, is relatively inert with water. This paper summarizes the neutronic calculations, the tritium chemistry information, the material compatibility problems and the available material properties. The material compatibility problem may be the most severe problem in using LiPb in a fusion environment.

Introduction

Lithium-lead in various compositions has been proposed to fulfill the tritium breeding function in D-T fusion reactors. The phase diagram of LiPb has been well established and is shown in Fig. 1.[1] The form of LiPb can be a solid, as Li_7Pb_2, a liquid, as $Li_{17}Pb_{83}$, or even in a two-phase mixture, as $Li_{62}Pb_{38}$. Most breeding compounds either have a high lithium atom density, or require a neutron multiplier to insure adequate breeding. The uniqueness of LiPb is the combination of breeding material and multiplier into one phase. Thus, lithium inventory can be minimized while the blanket design will be less complicated. It is also possible to use a low melting point LiPb compound to function as a breeder, neutron multiplier, and as a coolant. This will further simplify the blanket design.

The use of LiPb as breeding material results in good tritium breeding, high blanket energy multiplication, low tritium inventory, low blanket temperature, and low lithium inventory. The lead rich region of this alloy is of particular interest due to its inertness with water. Tritium recovery appears to be feasible by either inert gas purging or molten salt extraction. The great difficulty of using LiPb is the very high tritium partial pressure due to the low tritium solubility. This causes considerable difficulty in tritium confinement. The compatibility problem between LiPb and ferrous alloys at temperatures up to 500°C is another potential problem.

This paper reviews the existing literature and recent developments on LiPb. In particular, we will discuss tritium chemistry, neutronic results, compatibility problems, and chemical reactions with water. However, the scarcity of experimental results in physical data, tritium chemistry, and corrosion forces us to make a number of extrapolations from the few available results. Further experiments are urgently needed in these areas.

Neutronics

The tritium breeding and nuclear heating characteristics of the lithium-lead material are discussed in this section. The tritium breeding potential of the lithium-lead material with a graphite reflector was investigated in the NUWMAK study.[2] The NUWMAK blanket, which is made of Ti-alloy, consists of a 0.5 m breeding zone and 0.2 m graphite reflector in the outer region and a 0.2 m breeding zone and 0.4 m tungsten-based hot shield in the inner region close to the toroidal axis. It was concluded from this study that the tritium breeding ratio is adequately bracketed between 1.45 and 1.55 tritons per D-T neutron once the lithium atomic content in the lithium-lead materials (comprised of only natural lithium) is more than 20%. With only 5% lithium atom content in the lithium-lead materials, the tritium breeding ratio achieves about 1.1 tritons per D-T neutron. Recently, an attempt at using a metal reflector to enhance the blanket nuclear heating was made[3,4] and incorporated in a lithium-lead blanket design.[4] This blanket design employed a $Li_{17}Pb_{83}$ eutectic as the tritium breeding and cooling material. Tritium breeding as a function of volume percent structure was studied with a 1 m breeding blanket without a reflector and shield. At 22% structure, tritium breeding was 1.0 triton per D-T neutron and increased to 1.4 tritons per D-T neutron at 0% structure. It was also concluded that a tritium breeding ratio of 1.1 can be obtained if the breeding zone is 0.75 m thick.[4]

In the following we investigate the tritium breeding and nuclear heating for a reference blanket with reflector/shield as a function of lithium-lead material composition, breeding zone thickness and ^6Li enrichment in lithium. The reference blanket is made of ferritic steel, HT-9, and consists of a 3 mm first wall, a tritium breeding zone with variable thickness, and a 0.25 m ferritic steel reflector/shield. The tritium breeding zone is comprised of 10% structure and 90% lithium-lead material, both by volume. All the neutronic calculations were performed using the one-dimensional transport code, ANISN[5], with P_3S_8 approximation, in cylindrical geometry. The nuclear cross section library used is the DLC-37 library[6] with 25 neutron and 21 gamma-ray group structure.

Tritium Breeding and Nuclear Heating

Figure 2 depicts the tritium breeding ratio as a function of breeding zone thickness for $Li_{17}Pb_{83}$, LiPb and Li blankets. Note that all the lithium-lead materials used here are based on natural lithium unless otherwise mentioned. From this figure we see that LiPb and Li blankets produce about the same amount of tritium per D-T neutron. This is because the LiPb blanket produces more Pb(n,2n) reactions per D-T neutron than the Li blanket produces $^7Li(n,n'\alpha)$ reactions per D-T neutron that tend to compensate the decrease of tritium production due to the lower lithium concentration in the LiPb blanket. The tritium breeding ratio of these blankets reaches 1.1 tritons per D-T neutron as the breeding zone thickness increases to a value of more than 0.4 m. However, for the $Li_{17}Pb_{83}$ blanket, the tritium breeding ratio will never exceed 1.1 unless the breeding zone thickness is allowed to be more than 1 m. The reason is that the $Li_{17}Pb_{83}$ material consists of low atom percent lithium that is not adequate to breed tritium when the breeding zone is not thick enough, although the Pb(n,2n) reaction is capable of achieving 0.65 reactions per D-T neutron when the breeding zone is 0.5 m thick. Figure 3 summarizes the tritium breeding ratio as a function of atomic percent of lithium for 0.2, 0.4 and 0.8 m breeding zone thicknesses.

The total nuclear heating is also shown in Fig. 2 as a function of breeding zone thickness for $Li_{17}Pb_{83}$, LiPb and Li blankets. Because more neutrons per D-T neutron are generated in lithium-lead systemswith high lead content, more neutrons leak into the ferritic steel reflector/shield. Therefore, it produces more nuclear heating in the high lead content lithium-lead blanket as revealed in Fig. 2. Note that for a tritium breeding ratio of 1.1 tritons per D-T neutron, the total nuclear heating is 18.6, 18.7, and 17.8 MeV per D-T neutron for the $Li_{17}Pb_{83}$, LiPb and Li blanket, respectively. The energy multiplication of the $Li_{17}Pb_{83}$ blanket can reach 1.38[4] with a tritium breeding ratio of 1.07.

Effect of ^6Li Enrichment in Lithium

The effect of ^6Li enrichment in lithium on the tritium breeding and total nuclear heating for these lithium-lead blankets is discussed in this subsection. A reference design with 0.4 m breeding zone was chosen for our study because this blanket design produces 1.1 and 1.15 tritons per D-T neutron when the breeding material employed is LiPb and Li, respectively. However, the tritium breeding ratio for this blanket becomes 0.71 tritons per D-T neutron when $Li_{17}Pb_{83}$ is employed. Note that all lithium-lead and lithium materials mentioned above are based on natural lithium.

Figure 4 shows the tritium breeding ratios and total nuclear heating as a function of percent ^6Li in lithium for $Li_{17}Pb_{83}$, LiPb and Li blankets. It is seen from this figure that the tritium breeding ratio increases for all blankets except the lithium blanket as the percent ^6Li in lithium increases. The tritium breeding ratio in the Li blanket reaches a maximum value of 1.2 at about 20% ^6Li in lithium and then decreases as the percent ^6Li increases further. This is due to the fact that the tritium production due to the $^7Li(n,n'\alpha)T$ reaction is drastically reduced if the amount of ^6Li in lithium increases further. Eventually, the overall tritium breeding ratio drops below one when the ^6Li enrichment in lithium exceeds 80%. However, in contrast to the lithium blanket, the tritium breeding in LiPb and $Li_{17}Pb_{83}$ blankets depends mostly on the $^6Li(n,\alpha)T$ reaction which favors the increase of ^6Li content in lithium. The tritium breeding ratio for the $Li_{17}Pb_{83}$ blanket will reach 1.1 at about 40% ^6Li in lithium, and it continues to increase as the percent ^6Li in lithium increases. When the ^6Li content in lithium reaches 100%, it is 1.3 tritons per D-T neutron. The tritium breeding ratio for the LiPb blanket will be 1.4 at 50% ^6Li in lithium and it levels off as the percent ^6Li in lithium increases because of the relatively larger amount of lithium existing in the LiPb material.

As far as the total nuclear heating is concerned, it decreases for all blankets as the percent ^6Li in lithium increases. This is no surprise because more neutrons are absorbed by ^6Li that produces less energy than if those neutrons are parasitically absorbed in the reflector/shield. Note that at 40% ^6Li in lithium, the total nuclear heating for the $Li_{17}Pb_{83}$ blanket is 18.5 MeV per D-T neutron while it is about 20 MeV per D-T neutron at 7.4% ^6Li in lithium.

The Chemistry of $Li_{17}Pb_{83}$

Although the phase diagram has been known for 50 years and the density as a function of LiPb composition measured,[7] there is a somewhat limited data base for this material. Sieverts constants have been reported for various

LiPb compositions,[8] but these data must be viewed as preliminary. A number of measurements of the activity of lithium in the LiPb system have been made, and the results are reasonably consistent.[8,9,10,11] The reactivity of lithium-lead materials, including $Li_{17}Pb_{83}$, has been studied qualitatively by heating samples and dropping them into water.[12] In addition to studies of lithium-lead alloys, some additional useful information can be obtained from data from lithium and lead. There is an extensive data base for lithium[13,14] and at least one method (molten salt extraction) has been demonstrated to extract tritium from liquid lithium at concentrations of one wppm or less.[15] The data base for Pb in the context of fusion applications is much more limited, and a report of Sieverts constants[16] has been refuted.[17] It now appears that the solubility of hydrogen in lead is negligibly small, less than 0.02 appm.[17] In addition there is currently underway at Argonne National Laboratory an investigation of a ternary LiPbBi system. The melting point of $LiPb_4Bi_5$ was measured to be 140°C,[18] 40° less than that of lithium, and nearly 100° lower than that of $Li_{17}Pb_{83}$.

The activity of lithium in the LiPb system has been measured by Knudsen effusion mass spectrometry at 750 K and reported to continuously decrease from 4.0×10^{-3} to 2×10^{-5} as the lithium composition drops from 61 atom per cent to five atom per cent.[8,9,10,11] The activity of lithium in $Li_{17}Pb_{83}$ can be represented in the following form as a function of temperature.[19]

$$\ln a_{Li} = - 6960/T + 0.0245 .$$

Thus, the lithium activity in $Li_{17}Pb_{83}$ can be estimated to be about 1×10^{-4} at 750 K. This is an extremely low value, four orders of magnitude lower than that of pure lithium, and it is reasonable to expect that the chemical reactivity of lithium and related safety concerns will be markedly reduced. The chemistry in many respects may be expected to be more similar to that of lead than of lithium. However, the thermodynamic stability of lithium compounds with nonmetallic elements (e.g., H, O, N, etc.) is very large and despite the low activity, lithium effects can be significant. The activity of lead can be calculated from the activity of lithium by using the Gibbs-Duhem equation.[9] A typical curve of activity coefficient of LiPb is reproduced from reference 9 and shown in Fig. 5. Since $P = (rx)P_o = aP_o$ in which

P_n is partial pressure of the material

r is the activity coefficient

x is the mole fraction

a is the activity

P_o is the pressure of the pure material,

the partial pressure of lithium and lead can be calculated from the information of activity. Furthermore, on the lead rich end of the phase diagram, the vapor phase is dominated by lead and, as a first approximation, the vapor pressure of LiPb is the same as that of lead.

The Sieverts constants for deuterium in LiPb alloys have been measured at two temperatures, 1040 K and 950 K.[8] It is extremely difficult to obtain precise measurements for this system because of the low dissolution of hydrogen isotopes and therefore the results must be regarded as very preliminary. The only data on Sieverts constants of deuterium in $Li_{17}Pb_{83}$ in units of $atm^{1/2}$ per atom fraction was about 200 $atm^{1/2}$ at 1040 K. These values are extremely high, about two orders of magnitude higher than those for pure lithium. Extrapolation to lower temperatures is not possible, but the solubility will remain low.

There is a further point relating to the hydride chemistry of LiPb alloys. Lithium is an exothermic ionic hydride former, whereas lead is a covalent hydride former, and it is expected to be endothermic.[19,20] The solubility of hydrogen in lithium therefore decreases with increasing temperature, whereas the solubility of hydrogen in lead is expected to increase with increasing temperature. In the limited data discussed above for LiPb alloys with up to 90% lead, the dissolution decreases with increasing temperature. Therefore, the hydrogen is determined chiefly by the lithium chemistry. However, as the lithium fraction decreases to zero in the LiPb system, the character of hydration may change from exothermic to endothermic, i.e., the hydrogen pressure isotherms will cross.

Tritium Recovery

As discussed above, the Sieverts constants are very high for dissolution of hydrogen in $Li_{17}Pb_{83}$, about two orders of magnitude higher than those for lithium. This tends to make tritium recovery easier and it is likely that very low inventories of tritium in the blanket can be attained. However, tritium pressure above $Li_{17}Pb_{83}$ will tend to be very high and it will be necessary to obtain very low tritium levels in the liquid so that permeation losses will not be excessive. Therefore, a very efficient tritium recovery method is required.

The technical feasibility of molten salt extraction of tritium from liquid lithium has been demonstrated and tritium levels of less than one wppm appear achievable.[15] This process is proposed for the $Li_{17}Pb_{83}$ system. The solubility of Pb in the reference LiCl-LiF-LiBr salt was measured to be about 10 wppm[15] and therefore mutual solubilities are expected to be acceptably small. In contrast to the case for Li,

the density of $Li_{17}Pb_{83}$ (9.4 g/cm^3)[7] is much greater than that of the salt (2.6 g/cm^3). Therefore, the extraction process is simpler and in the event of alloy carry-over to the salt, the alloy will not interfere with the electrolysis step.[21]

To a first order of approximation (i.e., considering only thermodynamics), a molten salt extraction system of a size appropriate for a liquid lithium system would result in about the same tritium overpressure if used in a $Li_{17}Pb_{83}$ system. Therefore, tritium inventories will be less than those in liquid lithium. However, mass-transfer and kinetic limitations will mean that T_2 pressures above $Li_{17}Pb_{83}$ will be higher. Therefore, tritium permeation will be a significant concern. Experimental measurement of the Sieverts constants and more detailed analysis of the mass transfer limitations are required to assess the severity of this problem.

Reactivity of $Li_{17}Pb_{83}$

As is noted above, the activity of lithium in $Li_{17}Pb_{83}$ is very low, about 10^{-4}. Therefore, the reactivity with air and water should be much less than that of pure lithium or lithium-rich alloys. In a series of experiments at Argonne National Laboratory,[12] the reactivity of lithium and lithium-lead alloys with water was tested. Samples of various compositions were heated to about 500°C and dropped into water at about 90°C. It was found that the lithium-rich compositions $Li_{62}Pb_{38}$ and Li_7Pb_2 and liquid lithium itself had vigorous reactions with water. However, $Li_{17}Pb_{83}$ showed very little evidence of chemical reaction. A steel container dropped simultaneously into water showed boiling of water to about the same degree as the sample due to heat capacity effects. Little evidence of chemical reaction or hydrogen evolution was observed. As the result of this experiment and the very low lithium activity, $Li_{17}Pb_{83}$ is considered to be relatively nonreactive and it appears to be attractive from the standpoint of safety.

Material Compatibility

There is great concern about the compatibility between LiPb and structural materials at elevated temperatures. Since lead corrodes the iron based alloys more severely than the alkali metals, experience with liquid lead is the best indication of corrosion by liquid LiPb. The corrosion of lead is primarily due to straightforward solution by lead. In commenting upon such corrosion, it is essential to be specific not only with respect to temperature, but also to temperature gradient, coolant velocity and coolant impurity level. At 1000°C, there is rapid intergranular attack of various steels in one report,[20] but only a slight attack on 304 SS[21] in another report.

At temperatures around 600°C, however, the alloys are far more corrosion resistant as the following table[22] shows:

Alloy	Max T°C	ΔT°C	Exposure hr	Calculated Corrosion Rate mils/yr
ASTM A106	593	111	5064	12
Croloy 2-1/4	593	111	5156	10
410 SS	655	167	1346	15

For comparison, corrosion of 316 SS by lithium at 600°C is estimated to be 23 mils/yr.[23] If the temperature is reduced to 400°C, 304 SS showed no attack at all after 500 h.[24] On the basis of these data, we concluded that at temperatures below 500°C, dissolution attack by lead is reasonably slow even for stainless steels in uninhibited lead.

Since nickel and nickel base alloys are much more soluble than iron, it is not surprising that the austenitic stainless steels with their higher nickel content are somewhat more easily corroded. Chromium also has a higher solubility than iron, but lower than nickel. At 600°C the solubility of iron was reported[26] as only 2.3 x 10^{-4} wt% while for nickel at 635°C[7] the solubility was 0.85 wt%. For this reason we would favor the use of stainless irons (ferritic or martensitic) rather than the higher nickel austenitic alloys.

Finally, it is noted that little effort has yet been extended to further reduce the corrosion rate by inhibiting techniques. Addition of trace amounts of Zr and Mg have been shown[28,29] to provide complete protection of a 2-1/4 croloy alloy over three years with temperatures up to 550°C in spite of strong thermal gradients in the test loop.

It can be concluded that there are reasonable grounds for optimism for the use of steels as structural materials in liquid lead (and hence PbLi) up to 500°C. This initial conclusion, however, should not be used as an argument to neglect corrosion studies for these systems. The data we have used, while they are encouraging, are far from definitive and we heartily endorse the proposal for simple coupon tests to validate the alloys proposed for first wall materials in liquid lead.

Physical Thermal Data

There is a general lack of thermal physical data for LiPb. Of the material properties important to fusion applications, the only ones available are density and thermal expansion coefficients.

1. Density and thermal expansion coefficients of LiPb at liquid temperature[7] as shown in Fig. 6.

2. Density of solid lithium.[1]

3. Electrical conductivity for solid Li, atomic percent < 60%[1], as shown in Fig. 7. The resistivity of liquid LiPb was reported[31] and shown in Fig. 8.

4. Melting temperature as shown in Fig. 1.

The material properties such as thermal conductivity, specific heat, viscosity, surface tension, latent heat of melting, etc. are all not available.

The material data available, their status and their sources are summarized in Table 1.

Conclusions

There is considerable interest in the fusion community in using LiPb in different compositions as the breeding material for a D-T fusion reactor. This paper summarizes the available material data, as well as some of the up-to-date work neutronics. There is a general lack of material properties data such as thermal conductivity and specific heat. The available information on tritium chemistry, especially Sieverts constants, is very limited and its accuracy is in doubt. However, the high tritium breeding potential, the low tritium solubility, the low reactivity with water, and the high energy multiplication ratio make LiPb a primary candidate for breeding material. Further work, to measure the required material properties, is definitely needed at an early date.

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3. Preparation of manuscript by G. Herrington, University of Wisconsin.
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Table 1 Summary of Available Data for LiPb

Material Property	Status	Reference No.
Phase diagram	Firmly established	1
Sieverts constant	1 set of measurements, accuracy questionable	8
Lithium activity	4 sets of measurements, consistent with each other	8,9,10,11
Reactivity	Qualitative study	12
Density and thermal expansion coefficient	Information available in solid and liquid	1,7
Electrical conductivity	Limited information	1,31
Material compatibility	No data for LiPb, some results for Pb	20 to 29
Vapor pressure	Calculable from activity	8,9,10,11
Neutronic data	Calculable from Li and Pb	2,4

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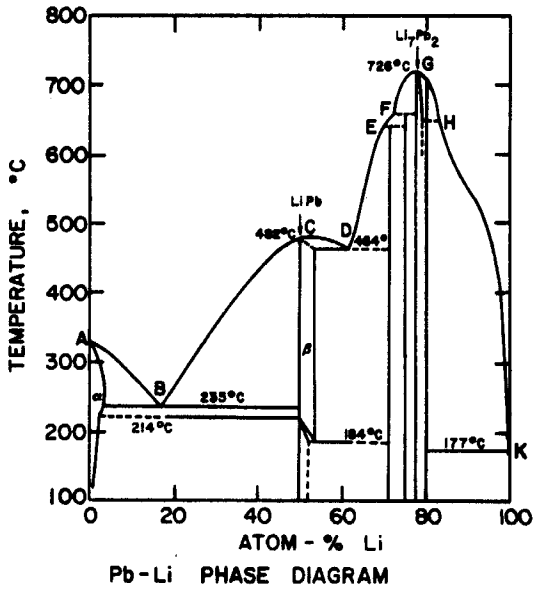


Figure 1

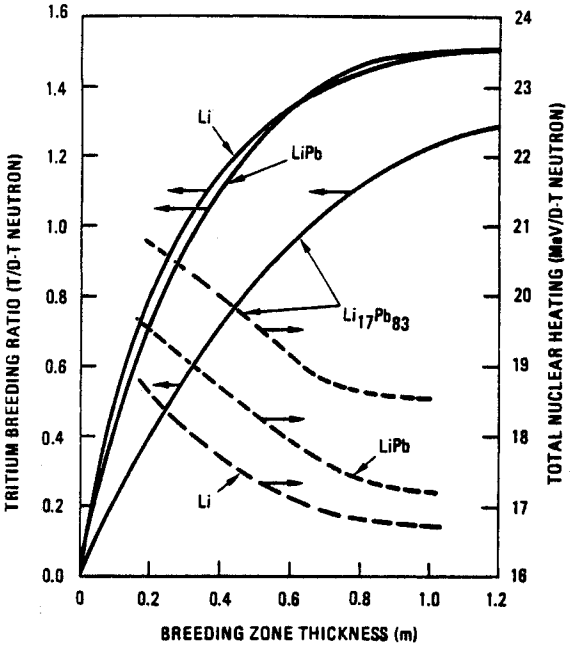


Figure 2 Tritium breeding ratios and total nuclear heatings as a function of breeding zone thickness.

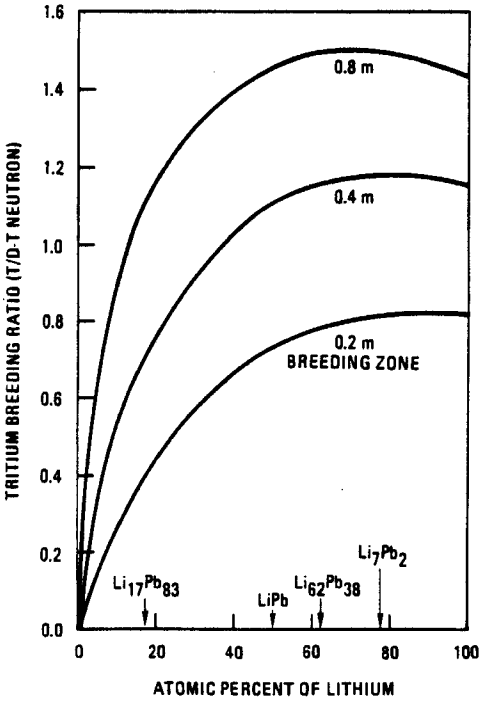


Figure 3 Tritium breeding ratios as a function of atomic percent of lithium in lithium-lead materials.

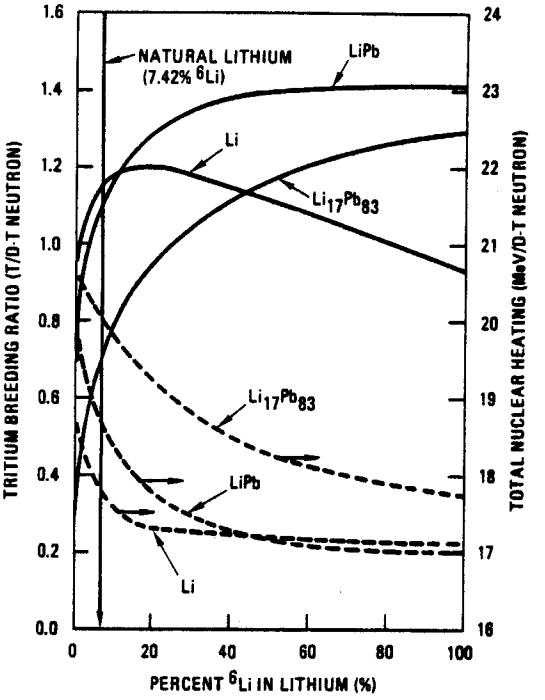


Figure 4 Tritium breeding ratios and total nuclear heatings as a function of percent ^6Li in lithium for lithium-lead blankets.

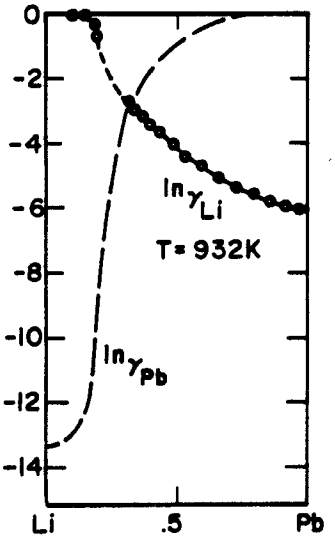
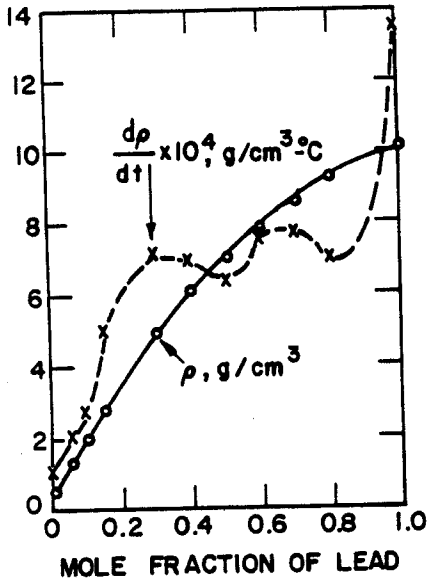


Fig. 5 Activity Coefficients for Li and Pb[9]



DENSITY AND THERMAL EXPANSION COEFFICIENT OF LiPb AT LIQUIDUS TEMPERATURE

Figure 6

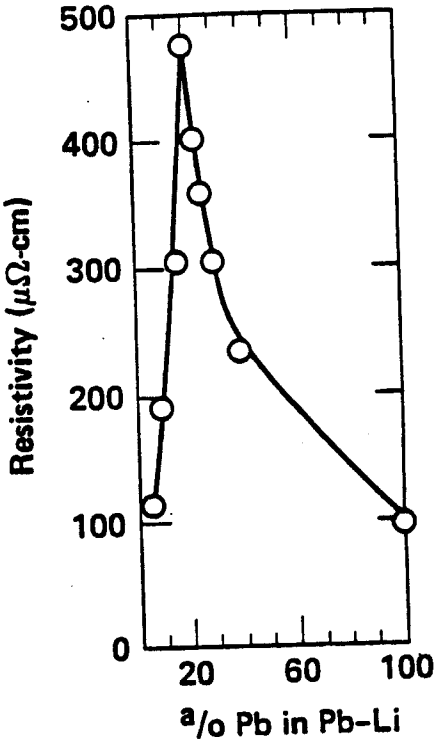
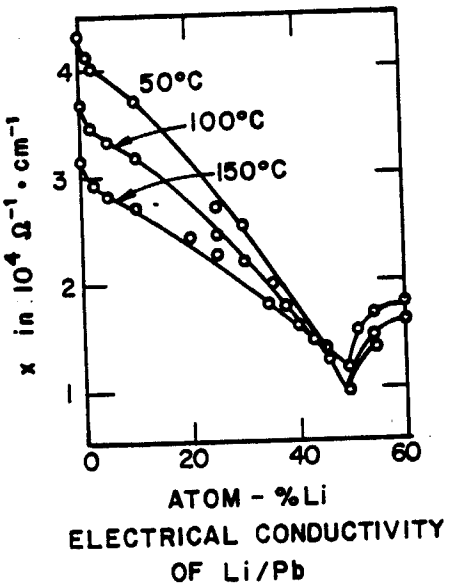


Fig. 8 Electrical Resistance of LiPb at 800°C[31]



ELECTRICAL CONDUCTIVITY OF Li/Pb

Figure 7