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LITERATURE SEARCH ON LEAD-BISMUTH ALLOYS

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LITERATURE SEARCH ON LEAD-BISMUTH ALLOYS

By M. E. Lee

INTRODUCTION

The use of a lead-bismuth alloy as a coolant in the removal of heat from power-generating nuclear reactors merits careful consideration as does the possibility that its ternary alloy with uranium or plutonium might be of use in a reactor of the circulating-liquid-fuel type. Information collected from the literature covering phase-equilibrium studies, physical-property data, and reactivity of this alloy toward other substances is presented in this report.

PHASE-EQUILIBRIUM DATA

The phase diagram of the lead-bismuth system given in the Metals Handbook for 1948 shows a eutectic at 55.5 wt % bismuth and 124°C (255°F) and a peritectic at 32.2 wt % bismuth and 184°C (363°F). This diagram is given in Fig. 1. In the diagram developed on the basis of thermal, microscopic, and x-ray studies,¹ the solid solution contains 2 per cent Pb; the eutectic is found at 124°C (255°F) and contains 43 per cent Pb; and the beta phase exists at 182°C (363°F). The nature of the beta phase could not be determined, but it was thought to represent a solid solution of Bi in the hexagonal modification of lead not known in the free state. This phase is shown to exist in slowly cooled alloys containing 65 to 70 per cent lead. The region of its existence is increased slightly with the rise in temperature up to the eutectic point. The maximum concentration of bismuth in lead amounts to 20 per cent, decreasing to 13.5 per cent as the temperature rises to 170°C (338°F).

The results of conduction experiments carried out by Herold² at 123°C (253°F) and 0°C (32°F) indicated that the limit of solubility of Bi in Pb was between 17.5 and 18.5 per cent bismuth, whereas the solubility of lead in bismuth was 1.6 per cent lead. The hardness curve indicated that the mixed crystal limit on the lead side was at about 17 to 18 per cent bismuth. The two limits for the eutectic were 40 per cent bismuth and 0.2 to 0.5 per cent lead. One investigator³ found that the solubility of bismuth in lead decreases from 14 per cent at 188°C (370°F) to 13 per cent at 6°C (42°F), and that of lead in bismuth is about 0.5 per cent at 125°C (257°F) (eutectic temperature), remaining practically constant down to 6°C (42°F).

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PHYSICAL-PROPERTY DATA

An alloy consisting of 56.5 per cent bismuth and 43.5 per cent lead is reported⁴ to have a specific gravity of 10.3, a melting point of 124°C (255°F), and an average neutron-capture cross section of 0.18 barn. Richter⁵ stated that the heat capacity of the alloys in this series is greater than that of the components and rises to a maximum 6 per cent high for the compound PbBi. However, near the lead end of the series the heat capacity is less than that of the components. In this series the specific volume decreases.

Data on the surface tension of the Pb-Bi alloys show only relatively small deviations from the law of mixtures.⁶ Measurements made on Pb-Bi alloys showed that their viscosities are less than the values calculated for the rule of mixtures. No maxima or minima existed in the eutectic region.⁷ It has been determined⁸ that the temperature coefficient of viscosity for these alloys is small and at the higher bismuth concentrations appears to be even lower than that of pure bismuth.

It has been reported that the lead-bismuth eutectic contracts slightly on cooling. Richter⁹ observed an amorphous phase in the eutectic. The properties of several alloys near the eutectic point have been studied in view of the possibility of using such a material as a pile coolant. One such alloy containing 58 wt % lead melts at 124.9°C (257°F) and boils at 1510°C (2750°F).¹⁰ A similar alloy has a volumetric heat capacity¹¹ of 0.35 cal/cc/°C. Measurements¹² have been made of the thermal expansion of Pb-Bi alloys at temperatures between 0 and -195°C (32 and -318°F), and the curve of thermal-expansion coefficient vs. composition has a maximum at 33 per cent Bi, the solid-solubility limit of bismuth in lead.

Table 1 is a summary of pertinent coolant properties of the Pb-Bi alloy containing 55 wt % bismuth.

SOLUBILITY OF VARIOUS MATERIALS IN THE ALLOY

Tests were carried out to determine the solubility of aluminum, stainless steel (18-8), 18 per cent chromium steel, and mild steel in a eutectic Bi-Pb (54.9 per cent Bi) solder at temperatures of 300 to 500°C (572 to 932°F).^{13,14} Test samples were freed of oxide, fluxed, and placed in the molten solder bath at the testing temperature. The solder was contained in a clay-graphite crucible and was covered with a blanket of charcoal. At the end of the specified time the samples were removed from the bath and allowed to cool in air. The samples were sectioned for metallographic examination, and the bath was cooled, sectioned, and analyzed. The solder had attacked the Al intergranularly, tearing off large pieces of the alloy. The sample of mild steel tested for 1 hr at 1000°C (1832°F) showed a fairly even attack with a few large pits. The attack on the 18 per cent Cr steel and the 18-8 stainless steel was intergranular. All the materials tested were wet by the solder at the temperatures and conditions of the test.

These tests were repeated at 500°C (932°F) on the steel samples and at 300°C (572°F) on the aluminum sample. Metallographic examinations of

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the various samples showed no apparent damage to the surface of any of the steel samples at 500°C (932°F) and to the aluminum at 300°C (572°F).

Other investigators¹⁵ found that, when strips of thorium (0.020 to 0.080 in.) were immersed in the Pb-Bi eutectic alloy for periods of 1 hr at 1000°C (1832°F), 900°C (1652°F), 500°C (932°F), and 300°C (572°F), the alloy wet the thorium at all the temperatures; but at 900 to 1000°C (1652 to 1832°F) the thorium was badly eroded or destroyed by the formation of an intermetallic compound. Lead alone caused some oxidation of the thorium, but bismuth caused severe erosion with the formation of a black compound.

Table 1--Summary of Pertinent Coolant Properties
Pb-Bi (55 wt % Bi)

Properties determined	Values obtained	
Atomic density at 405°C N, cm ⁻³ (x 10 ⁻²⁴)	0.0295	
N $\sigma_{a_{th}}$, cm ⁻¹ at 405°C	0.00131	
N σ_s , cm ⁻¹ at 405°C	0.268	
Melting point, °C	126	
Density ρ , g/cc at 405°C	10.18	
Specific heat (liquid) at 405°C		
Mass c, g-cal/g/°C	0.039 (?)	
Volume c ρ , g-cal/cc/°C	0.39 (?)	
Thermal conductivity at 405°C k, cal/sec/cm/°C	0.029	
Heat of fusion, g-cal/g	8.8	
Specific heat (solid) mean value room to melting temp., g-cal/g/°C	0.0298	
Relative heat to melt from room temp., per gram	0.24	
per cc	2.8	
Coolant activity from neutron reactions		
Isotope	45% Pb ²⁰⁸	55% Bi ²⁰⁹
Cross section for reaction		
N σ_a , cm ⁻¹	0.0000031	0.00019
Product decaying	Pb ²⁰⁹	Po ²¹⁰
Type principal radiation	β^-	α
Energy, Mev	0.68	5.3
Half life of decaying product	3.3h	140d

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Beryllium was found to be soluble in the Pb-Bi alloy to between 0.01 to 0.02 per cent at 1000°C (1832°F). Graphite was not soluble, but nickel, manganese, 330 stainless steel, and Hastelloy C showed severe attack with the Pb-Bi alloy.¹⁶

A summary of data on the solubility of other metals in the Pb-Bi eutectic is given in Table 2.¹⁴

DESIGNS PROPOSED

A design for a Pb-Bi-cooled beryllium-moderated reactor was proposed having the following conditions:¹⁶

Heat released in reactor, kw	55,000
Diameter of core	3 ft 2 in.
Height of core	4 ft 9 in.
U ²³⁵ content, kg	14
Maximum metal temperature	950°F (510°C)
Fuel rod	BeU-Be
Coolant entering reactor	600°F (316°C)
Coolant leaving reactor	800°F (427°C)

Another design using a Pb-Bi alloy as coolant calls for a flow velocity of 4.5 ft per second, a pressure drop of 20 psi, and a melting point of 135°F (57°C).¹⁷

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Table 2--Solubility of Various Metals in Eutectic
Bi-Pb Solder at Elevated Temperatures

Material treated	Section of material examined	Temperature*		Area covered, sq in.	Remarks	Chemical analyses, %			
		°C	°F			Al	Fe	Cr	Ni
Aluminum (17ST)	Top	500	932	1.5	As received	0.066			
Aluminum (17ST)	Bottom	500	932	1.5	As received	0.009			
18-8 stainless	Top	500	932	5	Cleaned, fluxed		0.022	Nil	Nil
18-8 stainless	Top	500	932	2	As received		0.003†	<0.002	Nil
18-8 stainless	Bottom	500	932	5	Cleaned, fluxed		0.020	nil	Nil
18-8 stainless	Bottom	500	932	2	As received		0.003	<0.002	Nil
18% Cr steel	Top	500	932	8.5	Cleaned, fluxed		0.010	0.006	
18% Cr steel	Bottom	500	932	8.5	Cleaned, fluxed		0.010	0.008	
Mild steel	Top	500	932	5	Cleaned, fluxed		0.012		
Mild steel	Top	500	932	2	As received		0.004		
Mild steel	Bottom	500	932	5	Cleaned, fluxed		0.008		
Mild steel	Bottom	500	932	2	As received		0.003		
Aluminum (2S)	Top	500	932	3	Cleaned, fluxed	0.067			
Aluminum (2S)	Bottom	500	932	3	Cleaned, fluxed	0.006			
Aluminum (2S)	Average	500	932	3	As received	0.003			
Aluminum (2S)	Average	300	572	15	As received	0.003			
Aluminum (2S)	Average	300	572	15	Cleaned, fluxed	0.004			

*One hour at temperature.

†Pb and Bi used contain 0.003 per cent Fe.

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Fig. 1--Lead-bismuth system.