Bibliography of a Promising Tritium Breeding Material – $Pb_{83}Li_{17}$

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UWFDM-994

FUSION TECHNOLOGY INSTITUTE

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I. Introduction

The United States began producing tritium in a heavy water reactor at Savannah River Laboratory (SRL) for thermonuclear weapons in 1952. This activity continued up to 1988 when all reactors at SRL were shut down. At that time it was expected that a new production reactor would be built to provide tritium for the nation's stockpile of nuclear weapons. The New Production Reactor (NPR) Office was established in 1988 but in 1991 President Bush announced a significant reduction in the U.S. stockpile of nuclear weapons. The NPR was canceled in 1991-92 but several methods of producing the tritium needed for the stockpile continued to be examined.

Almost immediately after the SRL reactors were shut down, scientists at LANL proposed the use of an accelerator to manufacture neutrons that could be used to make tritium (the original idea to produce tritium with an accelerator was made in the 1950's).(1) The basic idea is that a high energy, ≈ 1 GeV, beam of protons would be impacted on a high Z target (e.g., W, Pb, etc.) that would produce copious amounts of spallation neutrons. These neutrons would then be slowed down in a surrounding blanket and absorbed by selected isotopes (i.e., ^{3}He, ^{6}Li, etc.) that could produce tritium.

The concept of Accelerator Produced Tritium (APT) has been greatly expanded and refined since 1989. Since the exact amount of tritium needed for the U.S. stockpile is classified, the desired production levels are usually stated in terms of the production rate when the SRL reactors were shut down, i.e., 3/8, 1/4, 3/16, etc. of the 1988 rate. A brief chronology of the APT highlights at LANL is given below (after G. P. Lawrence, LANL).

1989-90	Design study for full-goal linac; reviewed by ERAB 1.6 GeV, 250 mA protons
1992	1/4 Goal design presented to JASON Group 0.8 GeV, 150 mA protons
1992-94	DOE funded study of 3/8 Goal design; Topical Report 1 GeV, 200 mA protons
1994-95	3/16 Goal design upgradable; reviewed by LLNL and JASON Group 1 GeV, 100 mA protons
1995	3/16 Goal design, upgradable to 3/8 Goal 1 GeV, 100 mA (200 mA with funnel)

One particularly clever scheme for producing tritium was proposed by scientists at LANL and involved the use of a W target which was surrounded by a moderating material such as heavy water. The breeding blanket contained ^{3}He gas which would intercept the thermalized neutrons to produce the tritium by the following reaction:

$$^3\mathrm{He} + n \rightarrow {}^3\mathrm{H} + {}^1\mathrm{H}\ .$$

Another method of producing T_2 was studied by scientists at Brookhaven National Laboratory (BNL). This involved the use of the LiAl alloy that had been used in the SRL reactors for over 35 years. The tritium producing isotope in the breeding blanket of that design was ^6Li via the following reaction:

$$^6\mathrm{Li} + \mathrm{n} \rightarrow {}^3\mathrm{H} + {}^4\mathrm{He} .$$

More recently,(2,3) an even more attractive design has been proposed which uses the low melting point eutectic alloy $Pb_{83}Li_{17}$ (see the phase diagram(4) in Figure I-1). The suggestion to use this alloy comes from earlier work at the University of Wisconsin in 1980(5) related to the design of tritium breeding blankets for fusion reactors. Although the first Pb-Li alloy suggested, in 1978,(6) for the tritium breeding blankets was the $Pb_{38}Li_{62}$, a high melting alloy, it was quickly discovered that the lower melting alloy, $Pb_{83}Li_{17}$ was much more attractive. Particular attributes of $Pb_{83}Li_{17}$ include:

- Low parasitic neutron absorption in the Pb
- Very high efficiency in converting neutrons into tritium
- Low vapor pressure at working temperatures
- Low tritium solubility in the alloy
- Good compatibility with inexpensive alloy steels
- Good high temperature stability
- Low long lived radioactivity.

Some of the drawbacks of this alloy as it pertains to making tritium include:

- Production of ^{210}Po which presents a hazard in an accident
- High density and high static loads on pipes and pumps.

A more complete description of the advantages and disadvantages of this alloy system for the APT concept is given elsewhere.(2) The purpose of this report is to collect as much information as is possible in a short time, from the key references on the $Pb_{83}Li_{17}$ alloy published over the past 15-20 years. We have attempted to collect and categorize those open literature papers, conference proceedings, and reports in a way that they can be easily retrieved and used.

II. Overview of the Bibliography

II-A. Method of Cataloging References

The commercial bibliography program, End Note Plus™, version 2.0.1, has been used and the keyword file includes the following 7 descriptors:

- Tritium
- Safety
- Corrosion
- Chemistry
- Reactor
- Country
- Experimental.

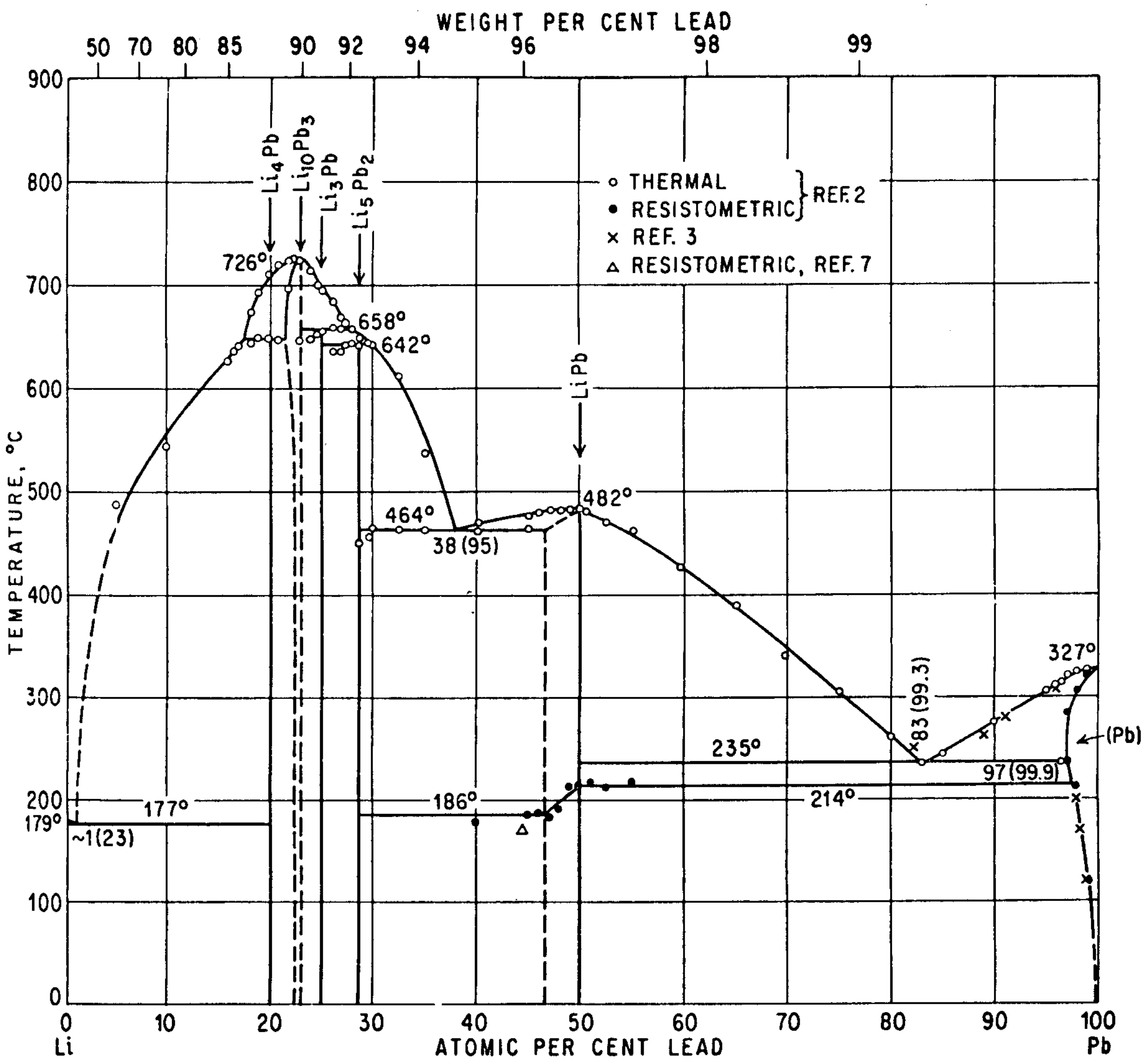


Figure I-1. Phase diagram of the Pb-Li system.(4)

The types of information included in each of the 7 keywords is summarized below.

Tritium

1. Diffusion/permeation of tritium (experimental results, mathematical model)
2. Tritium separation/recovery
3. Tritium removal techniques
4. Solubility and diffusivity of hydrogen isotopes
5. Interaction with hydrogen isotopes

Safety

1. Activation characteristics
2. Reaction with water
3. Reliability investigations
4. Sensitivity to atmospheric contamination
5. Chemical kinetics
6. Interaction with hydrogen isotopes
7. Production of bismuth and polonium

Chemistry

1. Impurities
2. Surface analysis
3. Deposition behavior
4. Thermodynamic properties
5. Thermophysical properties
6. Interaction with hydrogen isotopes
7. Production of bismuth and polonium
8. Diffusion/permeation of tritium
9. Tritium separation/recovery
10. Compatibility of structural materials
11. Chemical kinetics

Reactors

1. Major studies of reactors/devices that utilized liquid Li-Pb alloys for breeding/cooling
2. Generic work that may be applied to any reactor

Countries

In this section one will find a tabulation of the publications produced by institutions in various countries. The procedure for determining the country of origin is as follows:

- The country is assumed to be associated with the institution of the first author.
- Countries that are considered are: U.S., Japan, USSR, Germany, U.K., France, and Belgium.
- It is known that the former USSR has a strong program in the use of liquid Pb alloys metals, but unfortunately the publications in the USSR are hard to find and the work on Pb-Li may have been somewhat underestimated.

Experimental

1. Tritium release rates
2. Irradiation of liquid breeder material
3. Extraction of tritium
4. Solubility and diffusivity of hydrogen isotopes
5. Corrosion behavior of different structural steels
6. The reaction with water

II-B. Alphabetical Listing by Author

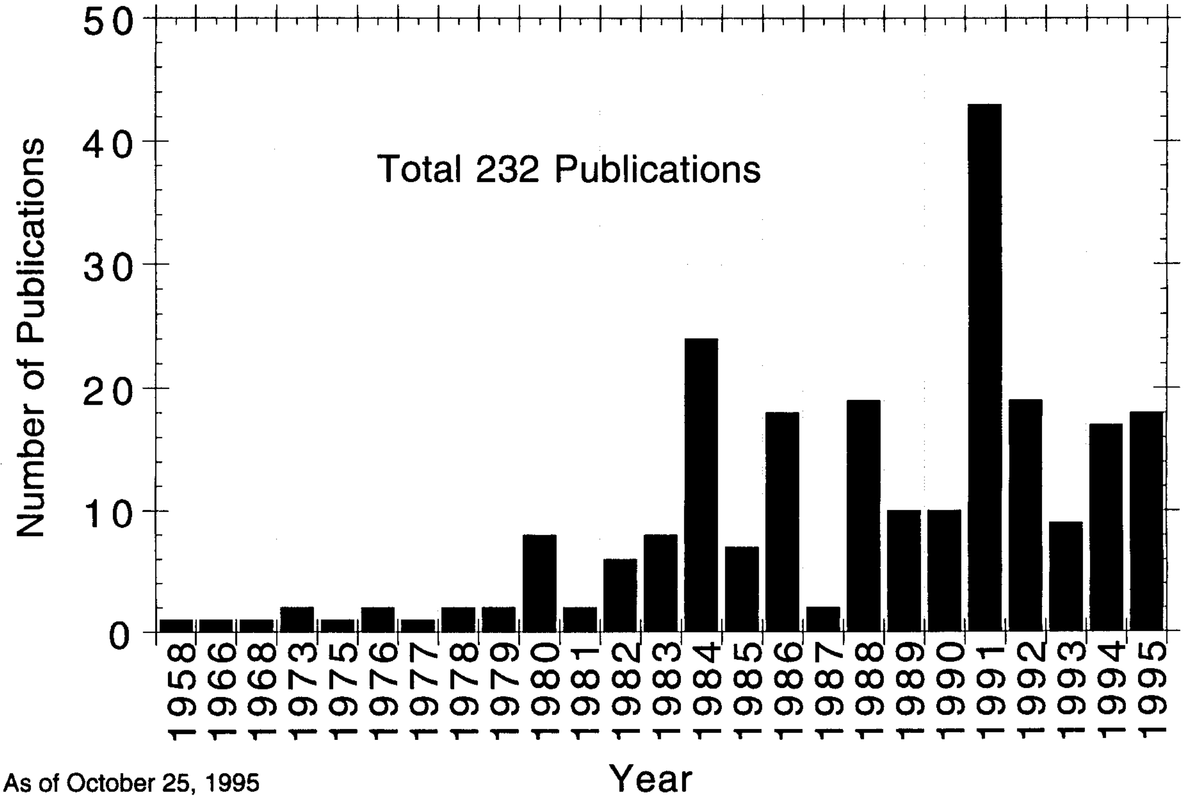
A total of 232 references that refer directly to the use of $Pb_{83}Li_{17}$ over the past 37 years have been cataloged alphabetically by author (see Appendix A). There are 109 different lead authors but approximately half of the articles were published by only 21 individuals and the other half were published by ≈88 authors.

II-C. Chronological Listing

The total list of references from Appendix A has been reformatted chronologically in Appendix B. A graphical display of this information is shown in Figure II-1. The articles appearing before 1980 deal only with some physical properties of the $Pb_{83}Li_{17}$ alloy. Since the 1980 proposal to make tritium in fusion reactors,(5) there have been 219 publications. This amounts to an average of 1 publication every 4 weeks over the past 15 years, or approximately 1.5 publications per month since 1990.

II-D. Listing by Country of Origin

The geographical distribution of publications on the $Pb_{83}Li_{17}$ alloy is given in Appendix C and graphically displayed in Figure II-2. It is interesting to note that while the largest number of papers from a single country come from the United States, the European countries of Germany, Italy, U.K., France, the former USSR, and Belgium account for more than 60% of the total papers. In the authors' opinion, the quality of papers coming from Germany far exceeds those from other countries and in particular, the work at KfK is at the cutting edge of this field.



As of October 25, 1995

Figure II-1. Number of lead-lithium publications.

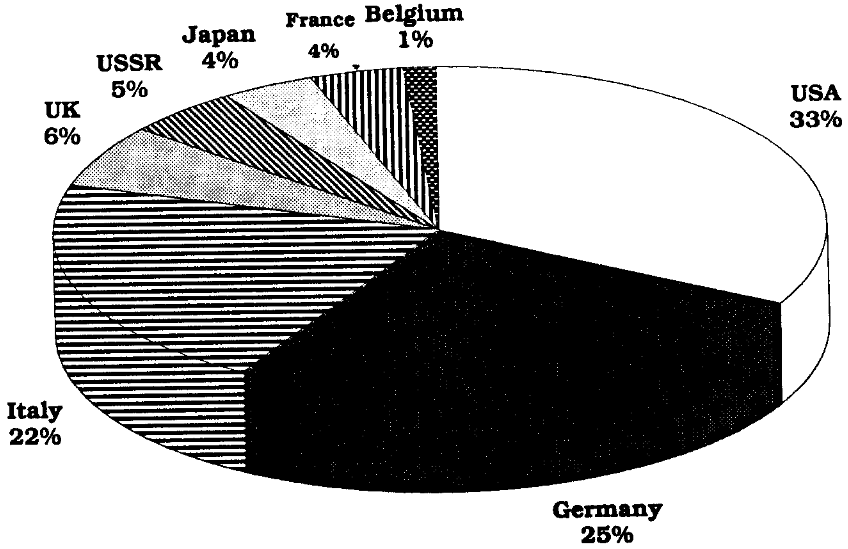


Figure II-2. The U.S., Germany, and Italy account for 80% of the literature published on Pb_{83}-Li_{17} alloys.

III. Thermophysical Properties of $Pb_{83}Li_{17}$ Alloys

A series of graphs (Figures III-1 through III-7) displays some of the key thermo-physical properties of $Pb_{83}Li_{17}$ alloys taken from references 94, 104, 114, 126, and 190 of Appendix A.

According to Bhatti and Shah(7), the correlation for the heat transfer coefficient (Figure III-7) of liquid metals which have fully developed turbulent flow in a smooth tube with uniform heat flux at the wall is,

$$Nu_d = \frac{hd}{k} = 0.625\ (Re_d\ Pr)^{0.4}\ .$$

All properties are evaluated at the bulk temperature. The above equation is valid for:

$$10^2 < Re_d\ Pr < 10^4 \text{ and for } \frac{L}{d} > 60$$

or,

$$Nu_d = \frac{hd}{k} = 4.82 + 0.0185\ (Re_d\ Pr)^{0.827}\ .$$

The above equation is valid for $10^2 < Re_d\ Pr < 10^4$ and for

$$3{,}600 < Re_d < 905{,}000\ .$$

The heat transfer coefficients in fully developed turbulent flow of liquid metals in a smooth tubes with constant wall temperature

$$Nu_d = \frac{hd}{k} = 5.0 + 0.025\ (Re_d\ Pr)^{0.8}\ .$$

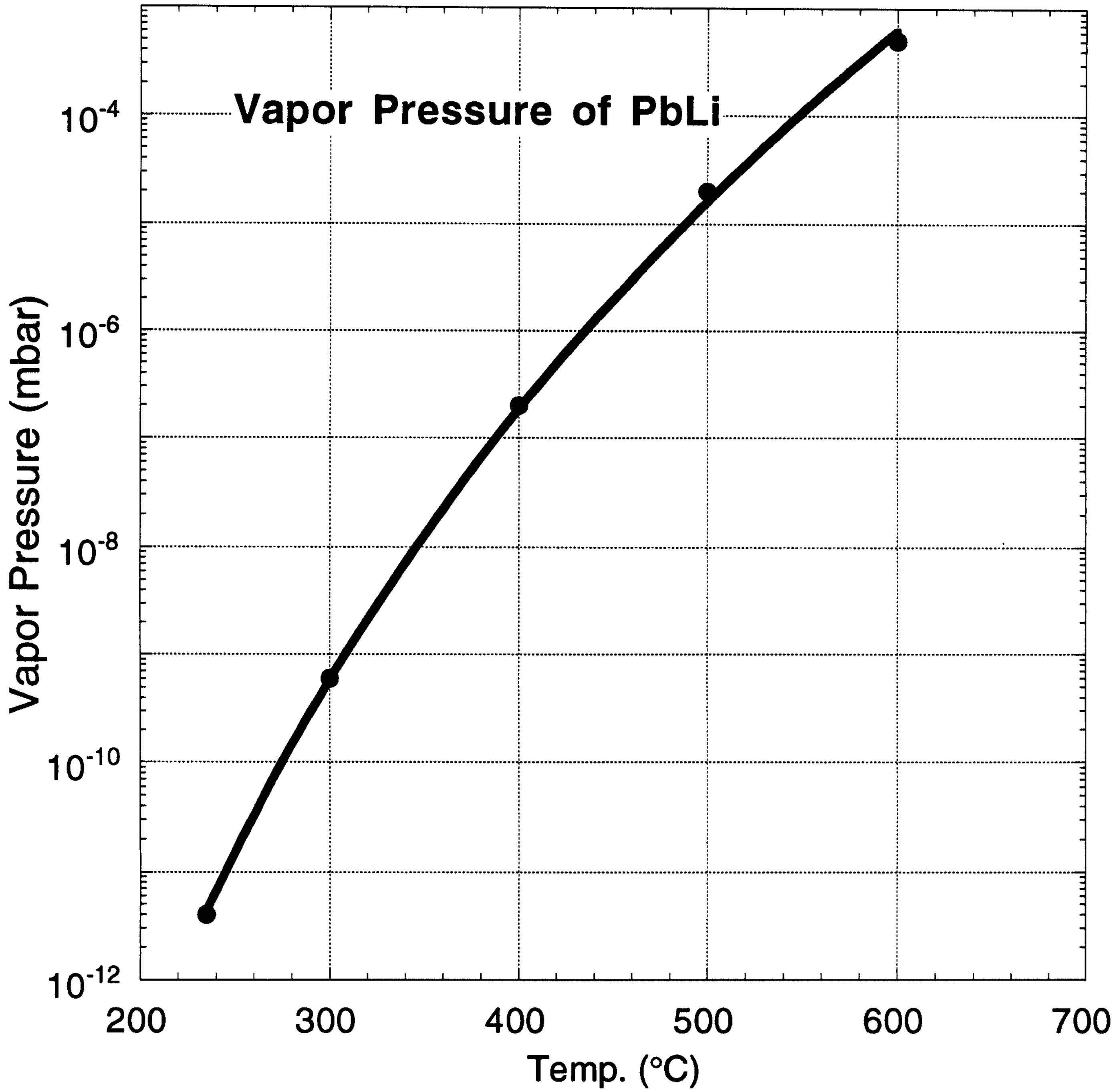
Again, all properties are evaluated at the bulk temperature. The equation is valid for $Re_d\ Pr > 100$ and for $\frac{L}{d} > 60$, where

$$(Re_d\ Pr) = \frac{ud}{a}\ ,\ \text{ and } a = \frac{k}{rc}\ .$$

For the sake of relevancy, the heat transfer coefficient for the $Pb_{83}Li_{17}$ alloy is given at a temperature of 400°C.

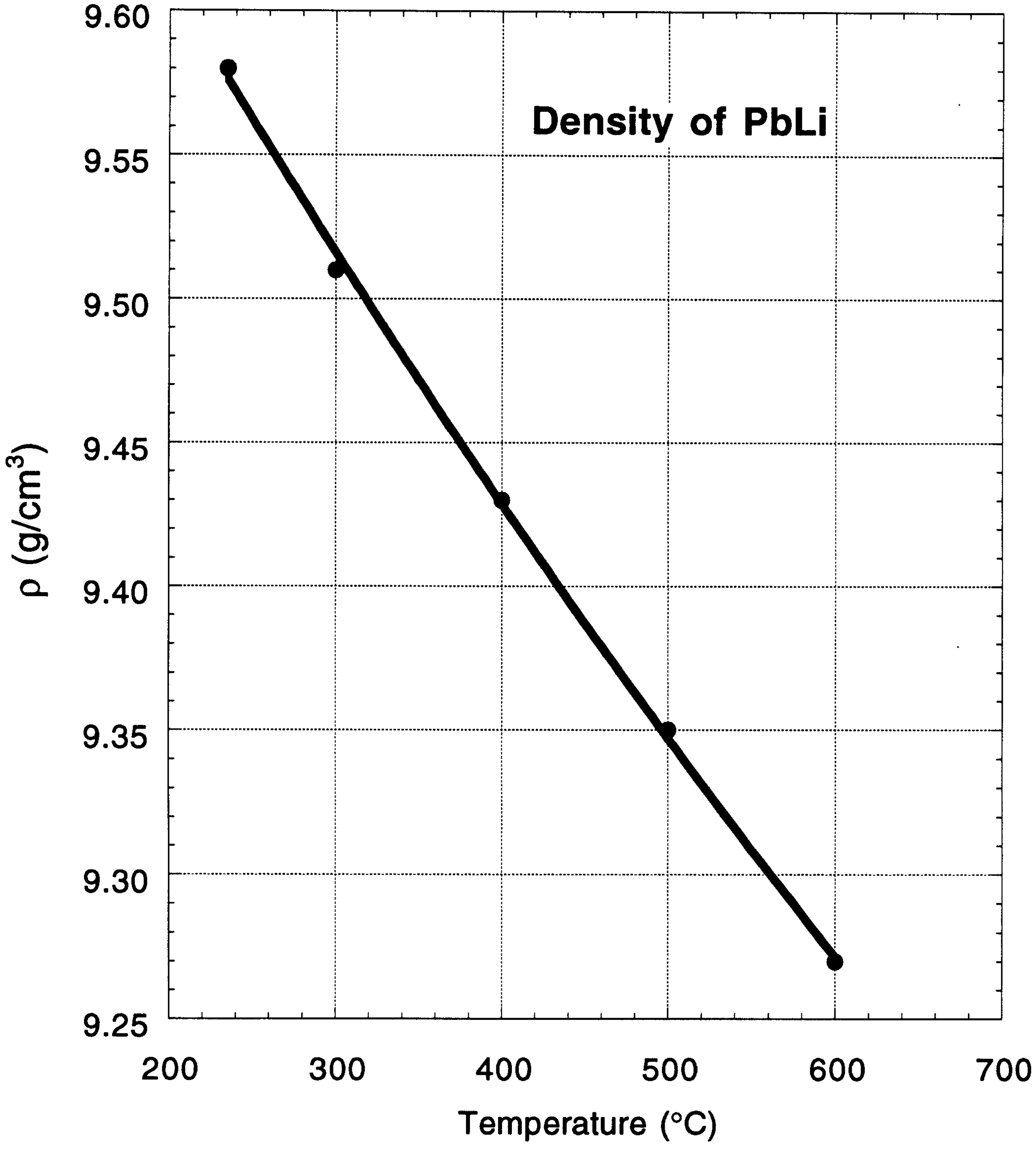
IV. Identification of Material to Contain $Pb_{83}Li_{17}$

The typical structural or piping material proposed to contain the $Pb_{83}Li_{17}$ alloy is steel. Both the austenitic and ferritic types of this steel have been considered. However, the corrosion resistance of the ferritic steels is better. Figure IV-1(8) shows some experimental data on type 316L and 1.4914 ferritic (MANET) steel over the temperature range of ≈400 to 600°C. At 400°C, the corrosion rate of the austenitic steel is ≈30 micron/year while that of the ferritic is <10 micron/year.



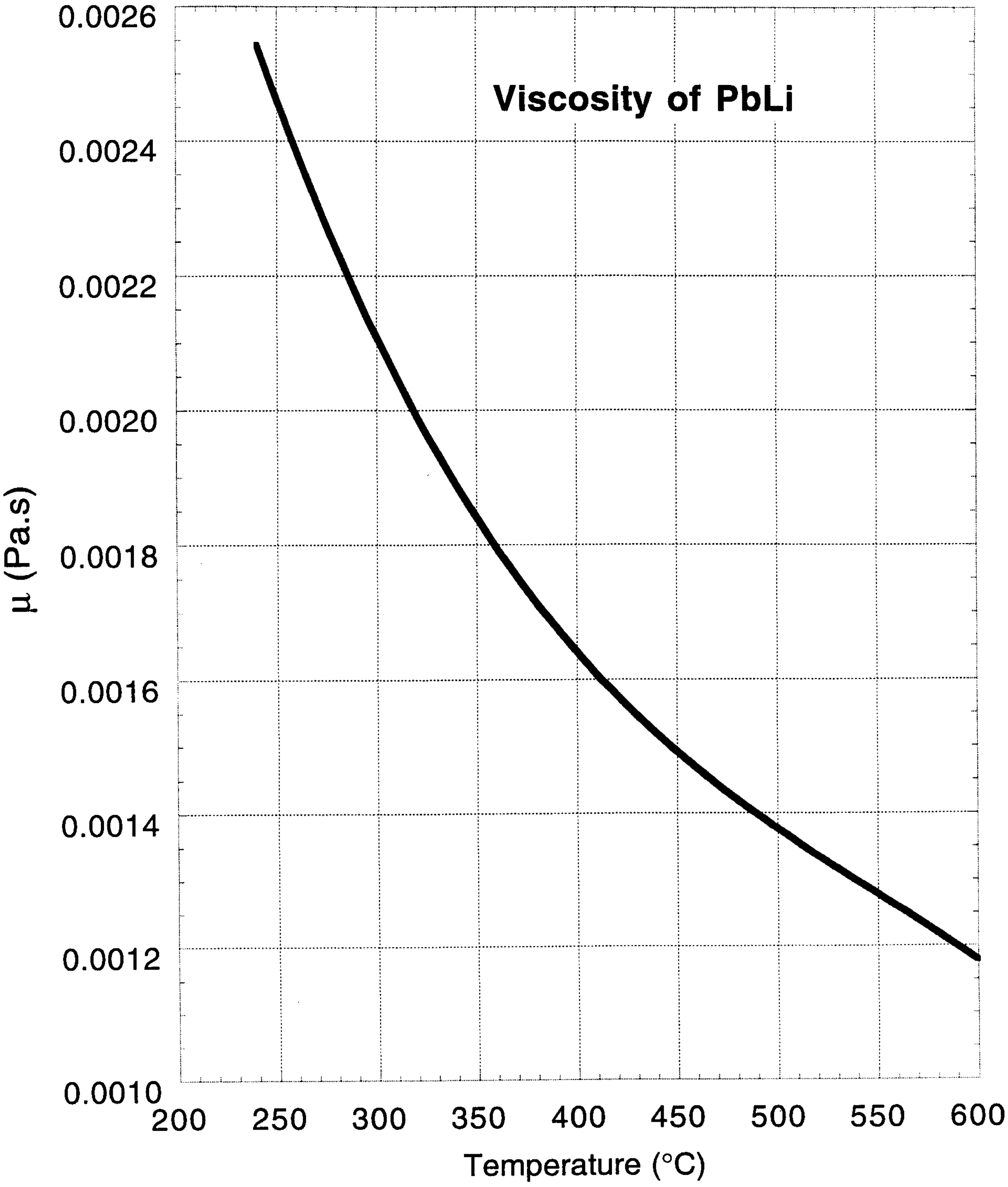
$$P = 1.4508 \times 10^{-59}\ T^{20.025}$$

Figure III-1. Vapor pressure of PbLi.



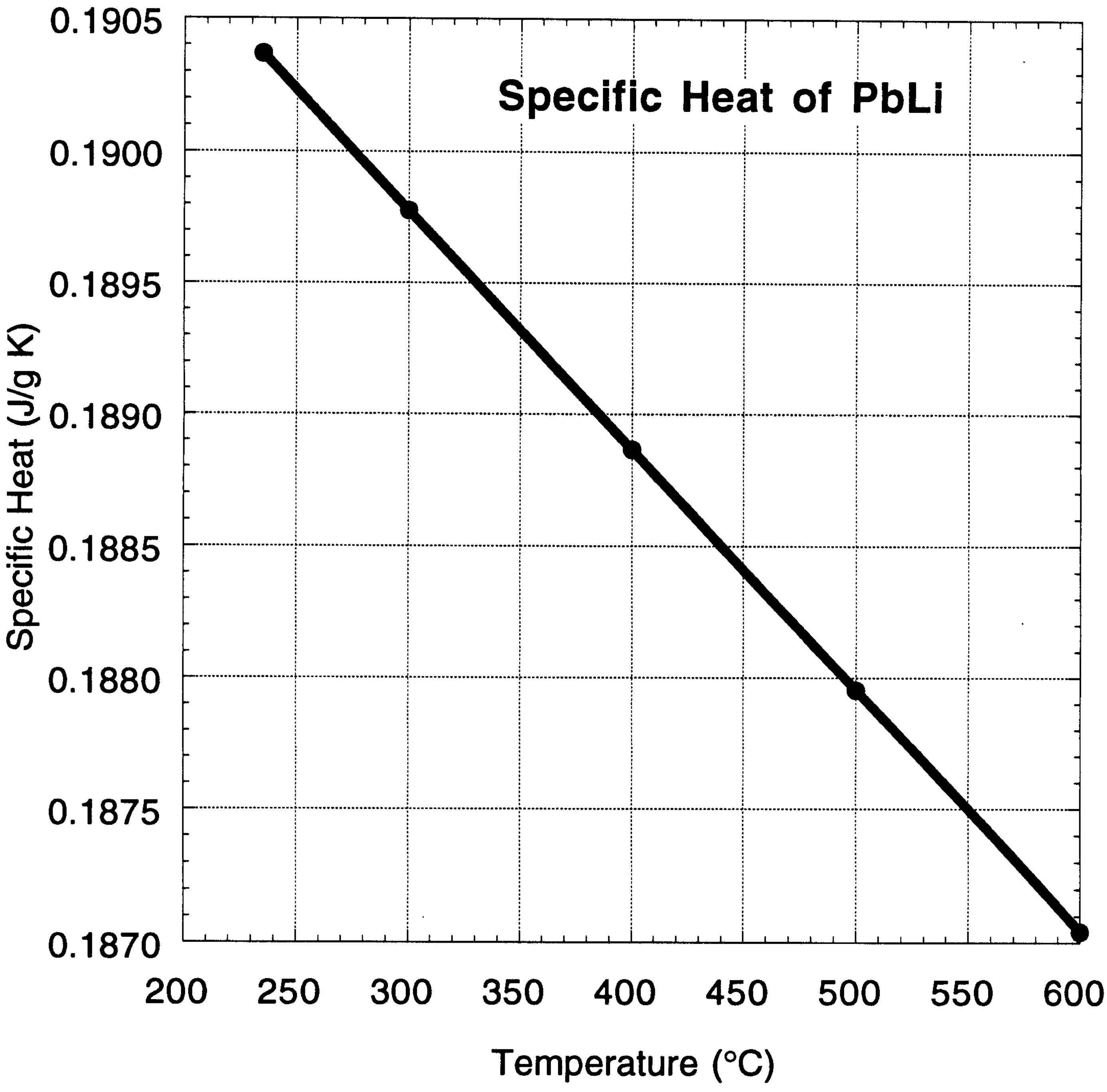
$$\rho = 9.8153 - 1.0876 \times 10^{-3}\ T + 3.0244 \times 10^{-7}\ T^2$$

Figure III-2. Density of PbLi.



$$\mu = 0.0061091 - 2.2574 \times 10^{-5}\ T + 3.766 \times 10^{-8}\ T^2 - 2.2887 \times 10^{-11}\ T^3$$

Figure III-3. Viscosity of PbLi.



$$c = 0.19251 - 9.116 \times 10^{-6}\ T$$

Figure III-4. Specific heat of PbLi.

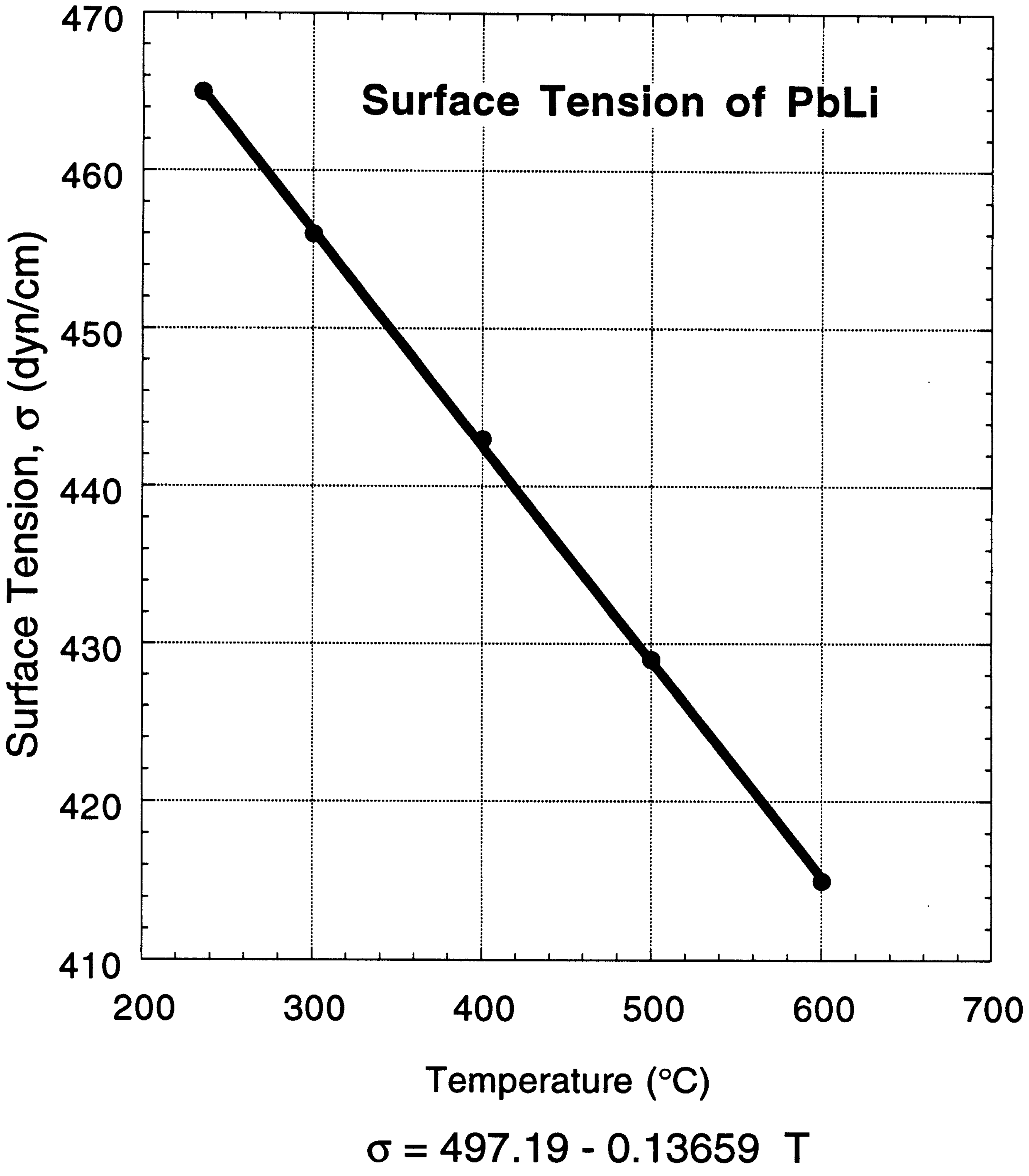
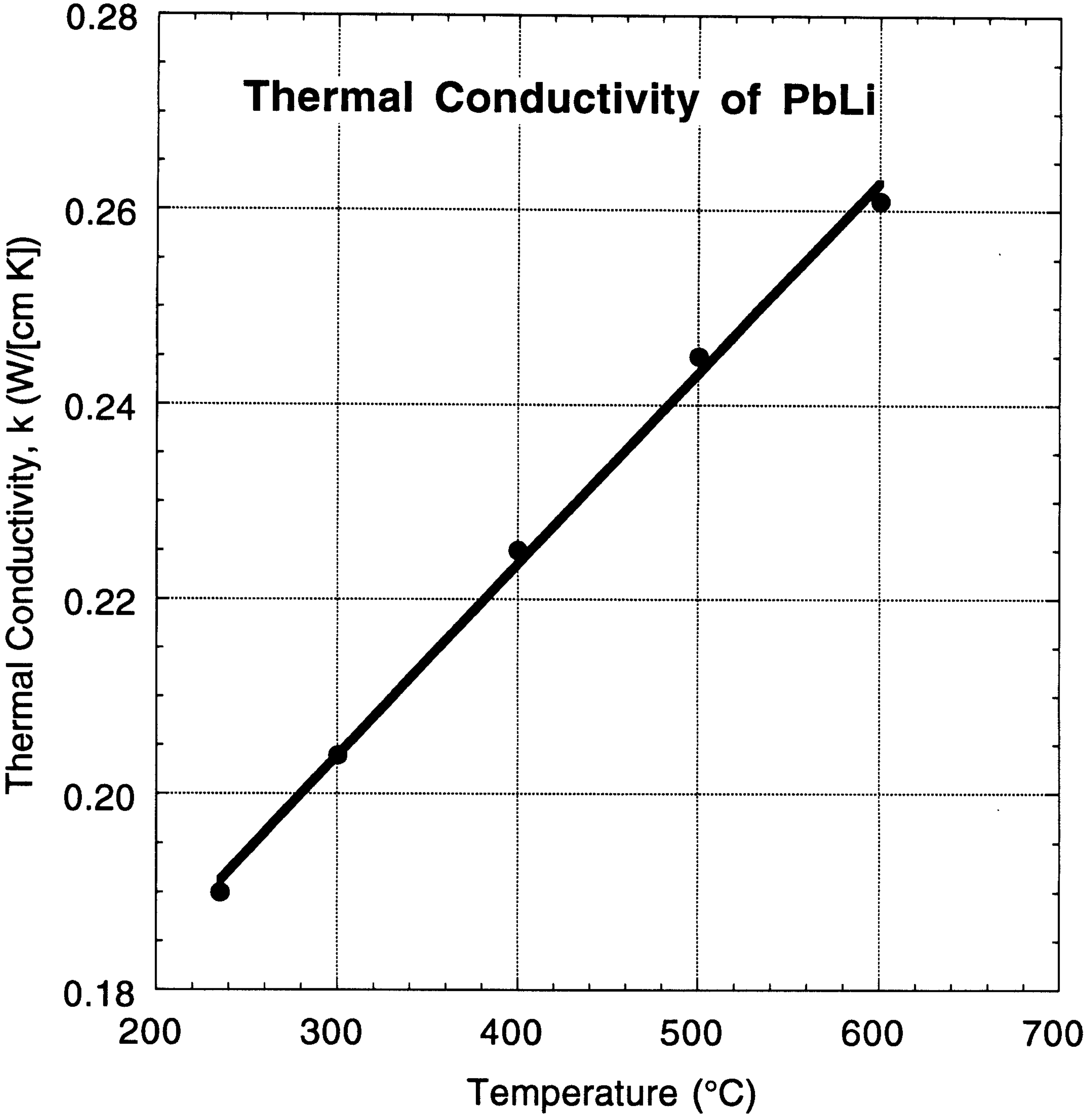
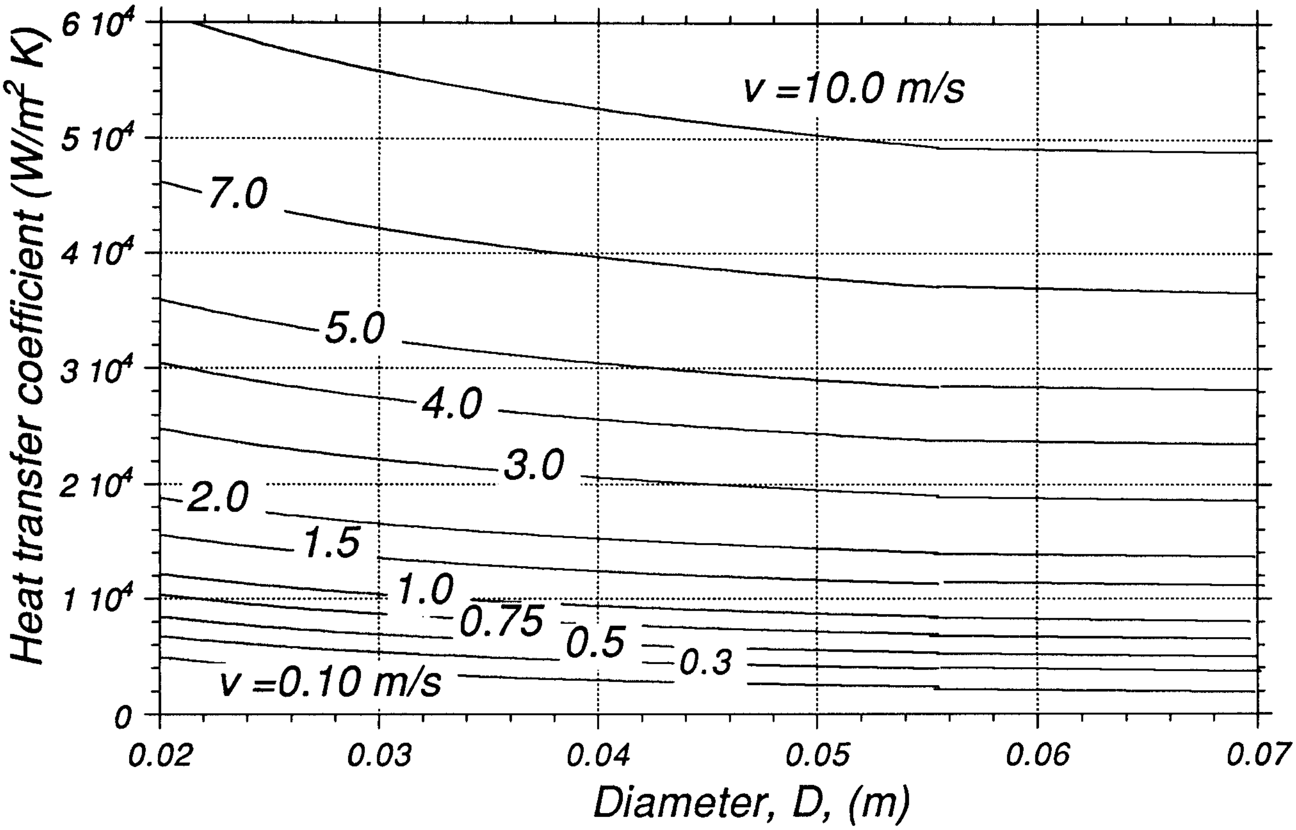


Figure III-5. Surface tension of PbLi.



$$k = 0.1451 + 1.9631 \times 10^{-4}\ T$$

Figure III-6. Thermal conductivity of PbLi.



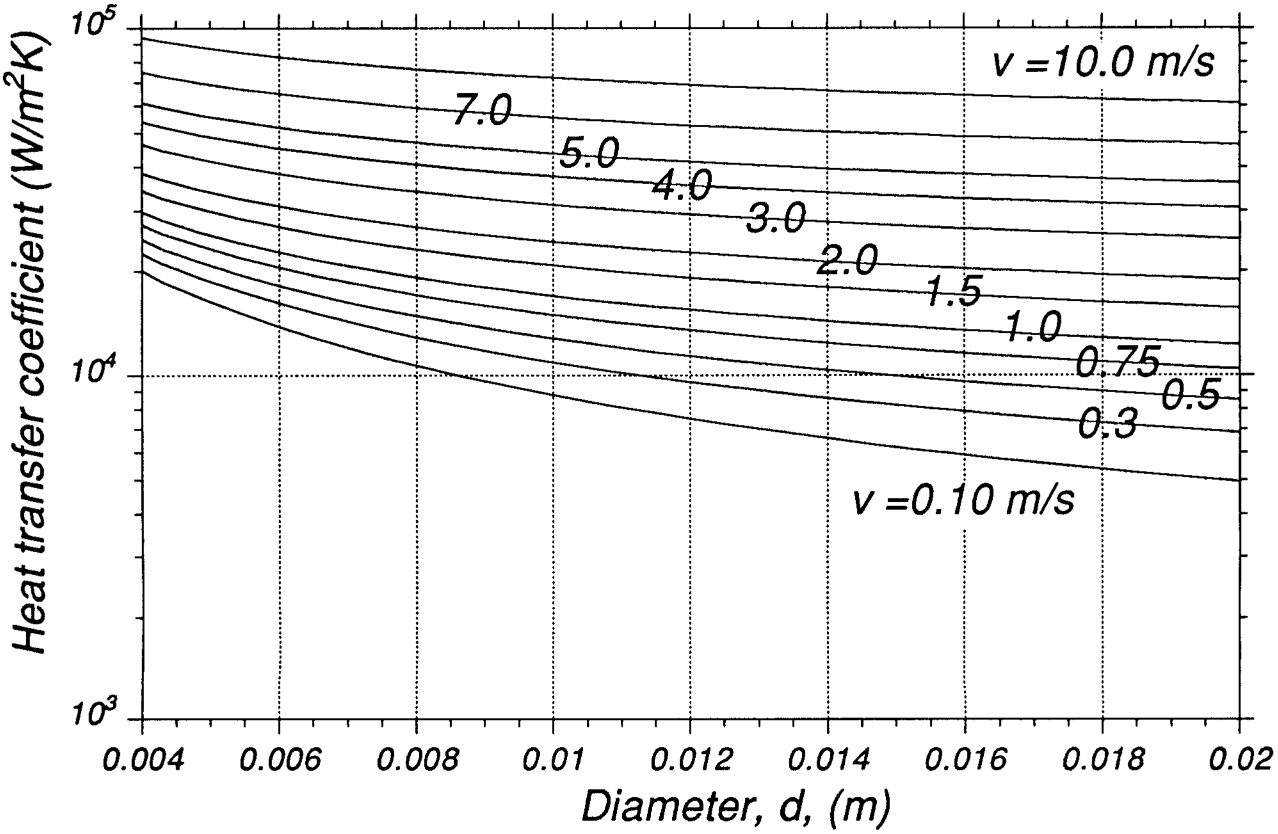


Figure III-7. Heat transfer coefficient (W/m^2 K), for LiPb at 400°C.

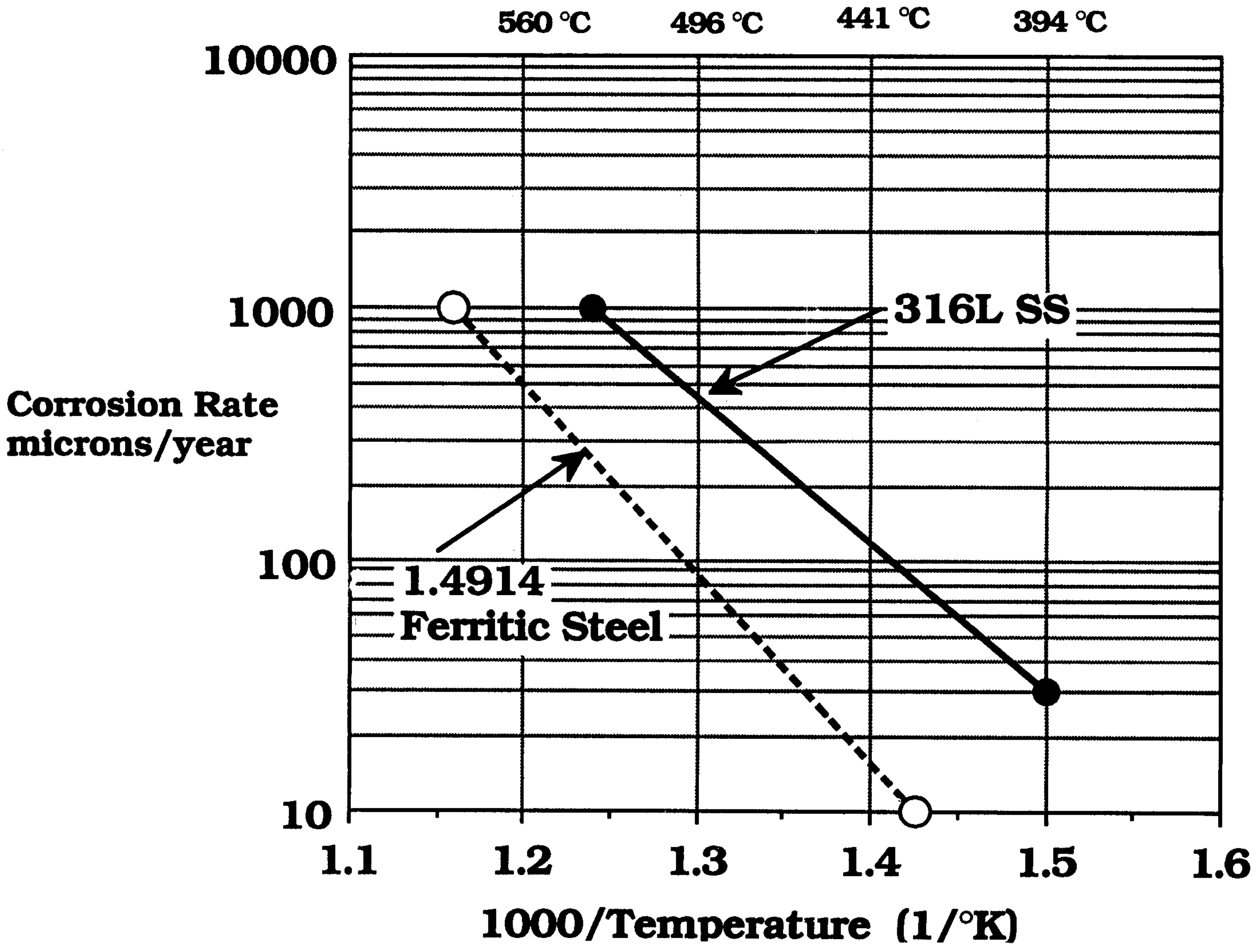


Figure IV-1. The corrosion rates of austenitic and ferritic steels in $Pb_{83}Li_{17}$ are quite low.[(8)]

Very often the ferritic steel HT-9 has been proposed because of its superior resistance to radiation damage. It is expected that it will also show the same level of liquid metal corrosion resistance as does the MANET steel. Furthermore, the replacement of Mo with W as an alloying element tends to alleviate its long term radioactivity associated with ^{99}Tc ($t_{1/2}$ = 213,000 y). Reductions in the Ni, N, and Nb are also advantageous from the standpoint of reducing long lived radioactivity. Table IV.1 gives a summary of the major alloying components of the steels that are considered prime candidates for containing $Pb_{83}Li_{17}$ in a radiation environment.

V. Design Concepts That Use $Pb_{83}Li_{17}$ Alloys

In the roughly 15 years that the $Pb_{83}Li_{17}$ alloy has been proposed for use in fusion reactors there have been 18 separate reactor designs that have incorporated this breeding/cooling material. The specific designs and their references are included in Table V.1 and the reader is encouraged to look at Appendix G for the complete listing including the generic work done for reactors in general.

VI. Concerns About the Use of $Pb_{83}Li_{17}$ Alloys

As might be expected, there are some aspects about the use of $Pb_{83}Li_{17}$ that require further analysis. Our primary concerns fall into 4 categories:

1. A definitive demonstration of tritium extraction from the liquid alloy with and without impurities from the spallation reactions that occur from 1 GeV protons. (See Reference 2 for further details of the spallation products that may be present).

2. An analysis, and experimental verification, of the containment of tritium in the alloy during its transport through the T_2 and heat extraction systems. This will require the analysis and testing of coating materials for the piping and containment structures.

3. Control of the Hg and Po produced by nuclear reactions in the Pb alloy. In the case of Hg, the experimental testing of the corrosive effects of small amounts of Hg in $Pb_{83}Li_{17}$ should be conducted. If it turns out to be a problem, the methods for extracting the Hg from the liquid Pb alloy should be demonstrated. In the case of Po, methods to prevent the buildup of Bi, the precursor to the ^{210}Po production, need to be demonstrated. This will involve a demonstration of the precipitation of Li_3Bi crystals from the liquid.(2)

4. The high density of Pb-Li (≈10 g/cc) means that there will be considerable mass in any target chamber as well as in the piping. Designs that can safely handle such masses and the momentum associated with circulating ≈1 m^3/s will have to be analyzed.

There are some secondary concerns that should also be experimentally resolved such as: compatibility of $Pb_{83}Li_{17}$ with structural alloys, verification of the neutronic behavior of the $Pb_{83}Li_{17}$ alloy, and establishment of firm price quotes for large batches (≈100 tonne quantities) of the $Pb_{83}Li_{17}$ alloy. While these areas need to be resolved before any final design can be frozen, it is felt that the first 4 primary concern areas listed above should be addressed immediately.

Table IV.1

Key Alloying Elements in Steels That May Be Used to Contain the $Pb_{83}Li_{17}$ Alloy in a Neutron Environment-wt%

Alloy	C	Cr	Ni	Mo	V	Nb	W	Si	Mn	B	N
316 SS	0.8	17	12	2.5	-	0.01	-	1.0	2.0	-	0.06
HT-9	0.2	12	0.5	1.0	0.3	0.11	0.5	0.35	0.55	0.01	0.05
HT-9 (Mod)	0.15	11	0.006	0.0003	0.3	0.0001	2.5	0.20	0.53	0.001	0.001
MANET 1.4914	0.13	10.6	0.87	0.77	0.22	0.16	-	0.37	0.82	0.009	0.02

Table V.1

Specific Fusion Power Reactor Designs That Have Used $Pb_{83}Li_{17}$ Alloys for Breeding and/or Cooling

Design	Year	Main Coordinating Institute or Country	Main Reference in Appendix A
Magnetic Fusion			
Witamir-I	1980	Wisconsin	6
UWTOR-M	1982	Wisconsin	9
TASKA	1982	Wisconsin	8
ITER	1983	U.S., EC, Japan, RF	106
TASKA-M	1983	Wisconsin	10
MARS	1984	LLNL	133
HWL Tokamak	1984	Wisconsin	11
MiniMARS	1986	LLNL	131
Demo	1986	ANL	73
ASRA6C	1987	Wisconsin	25
Titan	1988	UCLA	157
NET	1989	NET Team	160
ATR	1989	ORNL	135
European Demo	1991	NET Team	139
Demo (RF)	1994	Russia	196
SEAFP(E)	1994	Europe	64
Inertial Confinement			
HIBALL	1981	Wisconsin	7
Pulse*Star	1982	LLNL	154
HIBALL-II	1984	Wisconsin	12
LIBRA	1990	Wisconsin	13
LIBRA-SP	1995	Wisconsin	14
$Pb_{62}Li_{38}$-Magnetic Fusion			
NUWMAK	1979	Wisconsin	45

VII. Conclusions

An initial analysis of the literature surrounding the use of $Pb_{83}Li_{17}$ alloys to breed tritium has yielded well over 200 publications since 1980. Essentially all of the reported work has been tailored for the breeding of tritium in fusion devices. While the idea to use this alloy to make tritium originated at the University of Wisconsin, the concept has been pursued vigorously in Europe. At this point, the cumulative work in the United States slightly exceeds that in Europe. However, the recent rate and quality of work done in Europe (mainly in Germany and Italy), greatly exceeds that in the U.S. This observation holds true for both analysis and experiments.

It is a logical extension of the fusion related work to the production of tritium in an accelerator based system. While the physical, chemical, and nuclear properties of this alloy can be used directly, one will have to consider the effect of spallation products on the production, retention, and extraction of tritium. In addition, the effect of spallation products on corrosion will have to be assessed.

At this point in time, we see no "show stoppers" to the successful use of $Pb_{83}Li_{17}$ in the APT program and, in fact, it appears that the advantages of this alloy over the use of the current $W/^{3}He$ approach to breed tritium are compelling.

VIII. Suggested Further Work

This report, compiled in approximately 1 month, represents a significant beginning in the collection of the $Pb_{83}Li_{17}$ literature that has been produced over the past 15 years. However, it is fully expected that there are a few articles in the literature that have been missed. Therefore, a continued effort in literature searches should be conducted over the next few months and a hard copy of every report, conference paper, or journal publication should be collected.

The next logical step is the in-depth analysis of the literature with respect to specific properties which have a strong impact on the production of tritium from a spallation neutron source. In practice, it would be highly desirable to write a monograph on "The Use of Pb-Li Alloys to Breed Tritium". Such a monograph would be of value not only for the APT project, but also for both the Magnetic and Inertial Confinement Fusion Programs. Since it would have to rely on data from around the world, there may even be an international market for such an endeavor.

The form of the monograph would be to collect all the data on the key physical, chemical, and nuclear properties, analyze the data for consistency, and provide recommended values to be used in future design and experimental activities. To our knowledge, there is no such monograph available today.

Acknowledgments

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Appendix C

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Appendix G

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Appendix H

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Appendix I

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