

This document was prepared in conjunction with work accomplished under Contract No. DE-AC09-96SR18500 with the U. S. Department of Energy.

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, nor any of their contractors, subcontractors or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or any third party's use or the results of such use of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof or its contractors or subcontractors. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

**SELECT THERMAL AND PHYSICAL PROPERTY DATA
FOR LEAD CARBONATE (U)**

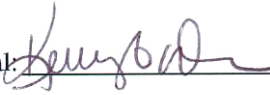
W. L. Daugherty

Savannah River National Laboratory
Materials Science & Technology

Publication Date: December 2005

DOES NOT CONTAIN
UNCLASSIFIED CONTROLLED
NUCLEAR INFORMATION

ADC &

Reviewing Official: 

Date: 1-3-06

Westinghouse Savannah River Company
Savannah River Site
Aiken, SC 29808

This document was prepared in connection with work done under Contract No. DE-AC09-96SR18500 with the U. S. Department of Energy

DISCLAIMER

This report was prepared for the United States Department of Energy under Contract No. DE-AC09-96SR18500 and is an account of work performed under that contract. Neither the United States Department of Energy, nor WSRC, nor any of their employees makes any warranty, expressed or implied, or assumes any legal liability or responsibility for accuracy, completeness, or usefulness, of any information, apparatus, or product or process disclosed herein or represents that its use will not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, name, manufacturer or otherwise does not necessarily constitute or imply endorsement, recommendation, or favoring of same by Westinghouse Savannah River Company or by the United States Government or any agency thereof. The views and opinions of the authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

Select Thermal and Physical Property Data for Lead Carbonate (U)

APPROVALS:

W. L. Daugherty
Author W. L. Daugherty Date 29 Dec 2005

K. A. Dunn
SRNL Pu Surveillance Program Lead K. A. Dunn Date Jan. 3, 2006

N. C. Iyer
Manager, Materials Science & Technology N. C. Iyer Date Jan 4, 2006

B. M. Loftin
Savannah River Packaging Technology B. M. Loftin Date January 3, 2006

K. H. Subramanian
Technical Review K. H. Subramanian Date January 9/2006

Revision Log

Document No. WSRC-TR-2005-00573

Rev. No. 0

Document Title Select Thermal and Physical Property Data for Lead Carbonate (U)

<u>Rev. #</u>	<u>Page #</u>	<u>Description of Revision</u>	<u>Date</u>
0	all	Original document	12/29/2005

Select Thermal and Physical Property Data for Lead Carbonate (U)

Introduction

The 9975 shipping package has been used for shipping and storing radioactive materials. Field surveillance activities of 9975 shipping packages following storage in the K Area Material Storage (KAMS) facility have identified that a lead carbonate layer forms on the shield of these packages after a period of time. Analysis of this corrosion product has identified the presence of lead carbonate (PbCO_3) and/or basic lead carbonate ($2\text{PbCO}_3 \cdot \text{Pb(OH)}_2$). In order to update calculations of the performance of the package in service, Materials Science & Technology was requested to compile thermal and physical properties of lead carbonate. This report transmits the information which was located.

The requested properties include thermal conductivity, specific heat capacity, density and emissivity. Several of the identified properties were found in the literature, and others were not. For the properties that were not located for lead carbonate, the same properties were located for other compounds which might behave similar to lead carbonate. Some general guidance as to the applicability of these data to lead carbonate is provided in lieu of actual lead carbonate properties. Where data were provided for both lead carbonate and basic lead carbonate, the values for each are reported. However, in most cases basic lead carbonate was not included in the references.

Data

Several references were located which contain the identified properties for lead carbonate in particular, or for a number of compounds in general. Thermal conductivity data for lead carbonate was not found in any of the listed references. In this case, thermal conductivity data for a variety of other compounds is identified, and used to provide a potential range for lead carbonate. While specific heat capacity data for lead carbonate was identified, a comparison to other compounds was made prior to locating that data. This comparison is included for completeness. Data for density and emissivity of lead carbonate were located, and are reported. Should additional information be required, further searches might focus on information from the paint/pigment industry.

Thermal Conductivity

Thermal conductivity data for lead carbonate was not found. However, Reference 1 compiles thermal conductivity data for a number of other compounds. Data for several compounds which might be similar to lead carbonate are listed in Table 1.

Table 1. Thermal conductivity for compounds which might behave similar to lead carbonate. Data are for 300K unless otherwise noted.

Compound	Thermal Conductivity (w/cm-K)
MgCO ₃	0.0465
CaCO ₃ (marble powder)	0.005 - 0.006
PbO + SiO ₂	0.0053
SiO ₂ crystalline	0.06 – 0.12
SiO ₂ fused	0.01 – 0.02
PbTiO ₃	~0.04, ~0.03 (~580K)
PbZrO ₃	0.0136 (345K), 0.0150 (530K)
KNO ₃	0.0090 (353K), 0.0045 (573K)
AgNO ₃	0.00447 (469K)
NaNO ₃	0.00565 (582K)

The following observations are drawn from the thermal conductivity data.

- Two other carbonate compounds have values ranging from 0.005 to 0.046 w/cm-K.
- Three nitrate compounds have values from 0.004 to 0.009 w/cm-K
- The range among the three nitrate compounds is relatively small, and similar to the value for CaCO₃.
- Since the values for SiO₂ are much higher than those for PbO+SiO₂, the value for just PbO may be less than 0.005 w/cm-K
- Values for 2 other oxides are 0.014 and 0.04 w/cm-K
- Over the temperature ranges given in the reference (not included in Table 1), values for the carbonates vary more with temperature (data for ~300K and less) than values for the lead oxides.

There are no obvious trends among these compounds that suggest a likely thermal conductivity value for PbCO₃. It may be that the primary factor influencing thermal conductivity is the form of the material; a porous layer of any of these compounds will be a relatively effective thermal insulator. Reference 1 in turn lists the sources from which this data was compiled. It may be possible to track down these source references and determine which compounds were tested in a form most like the PbCO₃ film that forms on a 9975 lead shield. Otherwise, the limiting value listed above (0.004 or 0.046 w/cm-K, each value might be limiting for different scenarios) might be an appropriate approximation.

Specific Heat Capacity

Specific heat capacity data for lead carbonate were found in Reference 2, and are summarized in Table 2.

Table 2. Specific heat capacity for lead carbonate. Data from Reference 2 are in cal/mol-K. These values are also shown converted to cal/g-K.

T (K)	PbCO ₃ cal/mol-K	PbCO ₃ cal/g-K	PbO-PbCO ₃ cal/mol-K	PbO-PbCO ₃ cal/g-K
298	20.917	0.0783	31.875	0.0650
300	20.970	0.0785	31.940	0.0651
400	23.830	0.0892	35.440	0.0723
500	26.690	0.0999	38.940	0.0794
600	29.550	0.111	42.440	0.0865
700	32.410	0.121	45.940	0.0937
800	35.270	0.132		

Conversion from mol to g: 267.1 g/mol PbCO₃, 490.41 g/mol PbO-PbCO₃

It is possible that the carbonate layer might form along with an oxide layer, or on top of an oxide layer. If this is considered credible, the data for PbO-PbCO₃ might be considered. Otherwise the PbCO₃ data would be appropriate.

Prior to locating the above data, the specific heat capacity data for similar compounds were examined. These data are in Table 3. It is noted that the specific heat capacity generally varies with the molecular weight. A consistent trend was observed between the weight of the metal atom and the specific heat capacity of the carbonate compound for those compounds with a single metal atom. The data for these compounds at 300K are plotted in Figure 1. A curve fit for these data identified by Excel spreadsheet is extrapolated to the atomic weight of lead. It is noted from Figure 1 that this gives very close agreement to the value reported in Reference 2. This relationship is shown primarily to suggest possible approaches in the use of the thermal conductivity data above.

Table 3. Specific heat capacity (C_p) for several carbonate compounds. Units are cal/g-K

Compound	At. Wt. of metal	C _p at 100K	C _p at 300K	C _p at 600K
PbCO ₃	207.2			
BaCO ₃	137.3	0.0608	0.104	0.1368
Ag ₂ CO ₃	107.9	0.0614	0.097	
SrCO ₃	87.6	~0.07	~0.13	
MnCO ₃	54.9		~0.17	~0.22
CaCO ₃	40.1	~0.09	0.2	~0.19
Li ₂ CO ₃	6.9			0.5

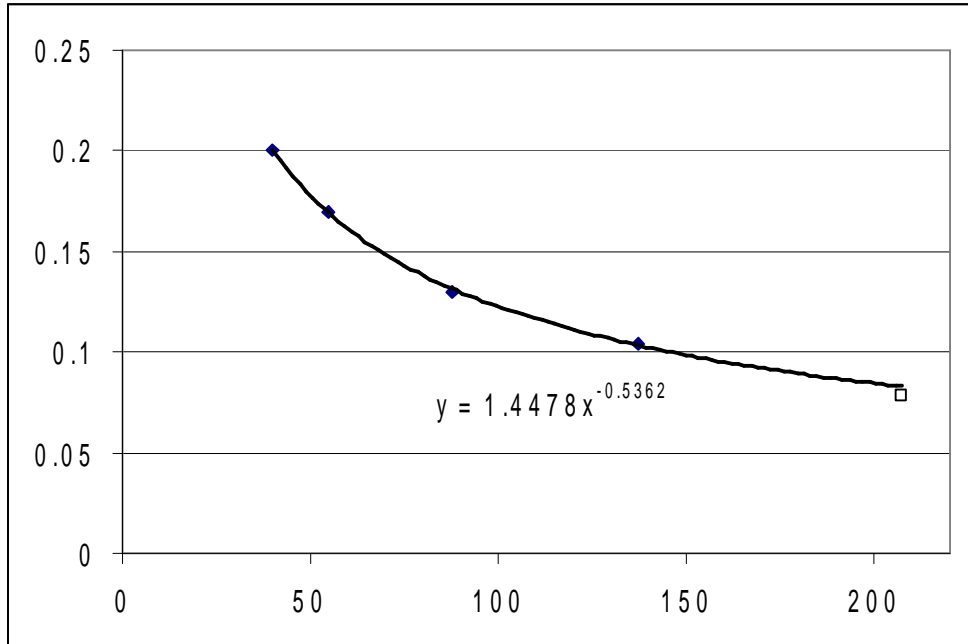


Figure 1. Specific heat capacity of carbonate vs atomic weight of metal (for compounds with a single metal atom). Curve fit by Excel spreadsheet is extrapolated to the atomic weight of lead.

Density

The following data are found in both References 4 and 5.

Lead carbonate, $PbCO_3$, 6.6 specific gravity

Basic lead carbonate, $2PbCO_3 Pb(OH)_2$, 6.14 specific gravity

In contrast, Reference 6 identifies a slightly different value for basic lead carbonate. It gives the following data:

Lead carbonate, $PbCO_3$, 6.6 g/cc density

Basic lead carbonate, $2PbCO_3 Pb(OH)_2$, ~6.5 g/cc density

Emissivity

Reference 7 identifies the following data for lead carbonate emissivity.

Table 4. Lead carbonate emissivity

Temperature (C)	Emissivity (%)
52	89
400	71
2750	8
solar	12

Reference 5 provides the following emissivity data.

For oxidized Pb (metal, not carbonate), emissivity (total) = 0.05 at 200C

For unoxidized Pb (metal, not carbonate), emissivity (total) = 0.63 at 200C

Two additional references for emissivity data were cited, but the references (8, 9) were not located for this review.

Summary

Specific heat capacity, density and emissivity data for lead carbonate were identified and reported. The density of basic lead carbonate was also reported. The thermal conductivity of lead carbonate was not found, but the thermal conductivity of a number of other compounds is summarized. A correlation was observed between the metal atomic weight and specific heat capacity for several carbonate compounds. While this specific relationship does not appear applicable to the thermal conductivity, it might suggest an alternate approach to estimate the thermal conductivity of lead carbonate. However, in the absence of any demonstrated relationship, it is recommended that the maximum or minimum value reported for other compounds (whichever is more limiting) be used for lead carbonate.

References

1. Thermophysical Properties of Matter, Vol. 2, Thermal Conductivity Nonmetallic Solids, Y. S. Touloukian, R. W. Powell, C. Y. Ho and P. G. Klemens, IFI/Plenum, 1970
2. Thermochemical Properties of Inorganic Substances, I. Barin and O. Knacke, Springer-Verlag, 1973
3. Thermophysical Properties of Matter, Vol. 5, Specific Heat Nonmetallic Solids, Y. S. Touloukian, E. H. Buyco, IFI/Plenum, 1970
4. Lead in Modern Industry, Lead Industries Association, 1952
5. CRC Handbook of Chemistry and Physics, 66th Ed, CRC Press, 1985
6. CRC Handbook of Chemistry and Physics, 79th Ed, CRC Press, 1998
7. Gmelins Handbuch der Anorganischen Chemie, Blei, Teil C, Lieferung 2, G. Hantke, Verlag Chemie, 1969, p. 47
8. "Some Optical Properties of White Paint Pigments in the Ultra-Violet Spectrum", Pfund A H, Am Soc Testing Materials Proc, 23, 369-78, 1923
9. "Reflection Factors of Industrial Paints and Pigments", Gardner Henry A, Trans Illum Eng Soc / N Y /, 17, 318-22, 1922

cc: B. M. Loftin, 773-41A
T. K. Houghtaling, 773-41A
J. S. Bellamy, 773-41A
K. A. Dunn, 773-41A
G. T. Chandler, SRNL
K. H. Subramanian, SRNL
J. L. Murphy, 730-4B
K. J. Durrwachter, 705-K
E. L. Bryant, 705-K
S. T. Sweat, 773-52A Document Control