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Lead–lithium eutectic material database for nuclear fusion technology

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Abstract

Fully validated material databases are needed for coherent technological developments in any R&D field. For nuclear fusion technology (NFT), within a near-term perspective of qualification and licensing of nuclear components and systems, this goal is both compulsory and urgent. This mandatory requirement applies for the particular case of the Pb–Li eutectic database as fusion reactor material. Pb16Li is today a reference breeder material in diverse fusion R&D programs worldwide. Technical consensus on most part of the material database inputs seems a major technological objective. In this work Pb16Li material database inputs for NFT have been systematically reviewed. Database inputs (bulk, thermal, physical-chemistry properties, and H-isotopes transport) are discussed and extended to base magnetohydrodynamic (MHD) properties, values for non-dimensional parameters and pipe/channel correlations in 2-phases dispersion models. Ongoing efforts to develop the Pb16Li material database as a computing expert system are reported.

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1. Introduction

Validated materials databases are needed for progress of any R&D technology. For fusion reactor materials some efforts of this type are ongoing today within diverse fusion programmes worldwide. NFT is today within the short-term perspective of licensing nuclear components and systems. It is the case of ITER test blanket modules (TBMs). Licensing of TBMs anticipates more demanding licensing issues of future DEMO reactors. Technical consensus for databases on properties demanded by design means a major technical goal particularly important for the case of Pb–Li eutectic as tritium breeding material. The Pb–Li eutectic properties are key licensing aspects concerning on tritium control capabilities and tritium confinement related issues.

This paper proposes a systematic revision of the Pb–Li material database for its use in NFT. The classic entrances, already gathered in previous efforts [1–3] are processed and extended to other properties: magnetic, hydrodynamic properties and non-dimensional correlations, tritium transport and material reactivity properties.

Material properties data ranges are imposed by fusion reactor systems conceptual design. Fusion reactors are both, electromagnetic and nuclear devices. For tenths MW m^{-3} deposed powers and few MW m^{-2} of wall loads, lead–lithium working temperatures window comes determined by the structural materials one restrained by corrosion limits under eutectic flowing conditions (commonly ranging between mm s^{-1} for pure breeding concepts and m s^{-1} for self-coolant ones). Thermal windows typically assumed range between eutectic points, 550 °C, case of ferritic–martensitic steel, or 700 °C, provisionally assumed SiC_f/SiC corrosion limit in flowing lead–lithium. Electromagnetic fields can modify corrosion behavior through

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the modification of velocity fields close to the wall. Maximum values of electric and magnetic field range within 0–1 kV m^{-1} and some Tesla (<15 T). Tritium partial pressure values come determined by tritium residence times of the liquid metal in the breeding zone and could typically range from few Pa up to kPa. Available database inputs and discrepancies are discussed together with the experimental technique used and associated uncertainty in a brief form.

Of particular interest is to configure this database as an expert computer tool. The present approaches in this direction are just anticipated.

2. Pb–Li eutectic standard database

Lead–lithium alloy does not have uses out of fusion technology what has meant a handicap for an extension of database properties and in particular for a systematic and precise validation of the alloy phase diagram.

The summary of standard inputs are given in Table 1. Standard properties include constitutive, bulk, thermal and electric properties. Here, assumed key aspects are discussed.

2.1. Lithium title discrepancies

Lithium determines the eutectic chemical activity and fine variation of Li title can significantly impact key database properties as physical–chemical and solute transport properties. Quality assurance (QA) of lead–lithium eutectic as nuclear material is intimately related with: (i) accurate determination of Li title, (ii) the eutectic production technique guaranteeing its short-scale homogeneity (in order to avoid Li aggregation) and (iii) the analysis technique certifying at 0.1%Li, the eutectic title. Table 2 shows present disagreements. Statistical data analysis in Table 2 place eutectic title at 15.7 at.% Li and at 508 K, as experimentally determined in [9](Table 3).

Table 2
Eutectic title

Year	Ref.	at.% Li	Uncertainties
1986	[2]	16.76	±0.21
1988	[4]	16.98	
1991	[5]	16.55	
1991	[6]	16.98	±0.25
1992	[7]	15.7	±0.2
1998	[8]	15.8	
2005	[9]	15.8	±0.2
2006	[10]	16.97	±4.99

2.2. Electrical conductivity divergences

Another source of experimental dispersion appears for the eutectic *electrical resistivity* since more than one order of magnitude difference for the two existing correlations [2,17]. *Electrical resistivity* enters in key non-dimensional numbers determining fluid MHD regimes. Additional electric resistivity measurements for a certified eutectic appear as a straightforward need.

3. Pb–Li eutectic extended database

Refinement and evolution of fusion reactor systems design naturally demand extension for materials databases.

Table 1
Summary of standard database inputs

Property	Expression	Range	Ref.	Consensus
Constitutive properties				
Eutectic title (at.% Li)	15.7–16.98	–	[2,4–10]	Rough on 15.7
Melting point (K)	507–510	–	[3,7,11]	Rough on 508
Molec. mass (g/mol)	175.76–173.16	–	[4–10]	As previous
Bulk properties				
Density (g/cm^3)	$\rho = 10.52\ (1–113 \times 10^{-6}\ \mathrm{T})$	T_m – 880 K	[6,10,12]	Large
Dyn. viscosity (Pa s)	$\mu = 1.87 \times 10^{-4} \exp(11640/\mathrm{RT})$	T_m – 625 K	[7]	Large
Surface tension (N/m)	$\sigma = 0.52–0.11 \times 10^{-3}\ \mathrm{T}$	520–1000 K	[13]	Unique entry
Thermal properties				
Specific Heat (J/g K)	$C_p = 0.195–9.116 \times 10^{-6}\ \mathrm{T}$	T_m – 800 K	[7]	Unique entry
Heat of melting (J/g)	$\Delta H_f = 33.9$	T_m	[14,15]	Large
Vapour pressure (Pa)	$P_v = 1.5 \times 10^{10} \exp(-22900/\mathrm{T})$	550–1000 K	[16]	Unique entry
Thermal conductivity (W/K m)	$\lambda = 1.95 \times 10^{-2} + 19.6 \times 10^{-5}\ \mathrm{T}$	T_m – 625 K	[7]	Unique entry
Thermal expansion coeff. (K^{-1})	$\beta = 1.124 \times 10^{-4} + 1.505 \times 10^{-8}\ \mathrm{T}$	T_m – 880 K	[6,10,12]	Theor. approx.
Electric properties				
Electric resistivity (Ω m)	$\rho_{el} = 102.3 \times 10^{-6} + 4.26 \times 10^{-8}\ \mathrm{T}$	T_m – 933 K	[2,7]	None
Characteristic adimensional numbers [6]				
$Pr = \mu C_p/\lambda = 1.4 \times 10^{-3}$ (Prandtl)		$Ha = BL(1/(\rho_{el}\mu))^{0.5} \rightarrow 1.4 \times 10^4$ (Hartmann)		
$Re = \rho u\ L/\mu \rightarrow 6 \times 10^4$(Reynolds)		$N = B^2L/(\rho_{el}\rho u) \rightarrow 3.0 \times 10^4$ (Interact. Number)		
$Rm = \mu_m Lu/\rho_{el} \rightarrow 10^{-6}$ (Magn. Reynolds)		$Ha/Re \rightarrow 2$		
$Gr = g\rho^2\beta\ (\Delta T)L^3/\mu^2 \rightarrow 5.0 \times 10^{12}$ (Grassof)		$Sc = \mu/(\rho D) \rightarrow 71$ (Schmidt Number)		

Table 3
Summary of extended database inputs

Magnetic properties

Lithium	Paramagnetic, $\chi_v > 0$	$\chi_M = 24.5 \pm 0.3$ (10^{-6} cm^3/mol) [19,20]
Lead	Diamagnetic, $\chi_v < 0$	$\chi_\rho = -0.79 \pm 0.01$ (10^{-9} m^3/kg) [21], but paramagnetic in [18]
It seems that magnetic permeability of Pb16Li is almost the permeability of vacuum		

H-transport properties

H-isotope	Expression	Ref.	Range	Method
Diffusion coefficient D (m^2/s)				
T	$2.62 \times 10^{-3} \exp(-6630/RT)$	[31]	573–773 K	GE-NI
H	1.5×10^{-9}	[29]	723 K, 10^3–10^4 Pa	GE-ID
H,D,T	$4.03 \times 10^{-8} \exp(-19\,500/RT)$	[5]	508–700 K, 10^3–10^5 Pa	GE-ID, extr.
T	$2.50 \times 10^{-7} \exp(-27\,000/RT)$	[22]	573–973 K, 3×10^3 Pa	GE-NI
Sieverts constant k_s (at.frac/Pa$^{1/2}$)				
D	6.33×10^{-7}	[27]	850–1040 K, 10^{-1}–10^0 Pa	HA-p
H	$4.7 \times 10^{-7} \cdot \exp(-9000/RT)$	[28]	573–773 K, up to 10^4 Pa	HA + TD
H	1.08×10^{-6}	[25]	573–723 K, 10^4–10^5 Pa	HA
H	2.7×10^{-8}	[29]	723 K, 10^3–10^4 Pa	GE-ID
H	$8.98 \times 10^{-7} \exp(-6100/RT)$	[26]	508–1040 K, 10^4–10^6 Pa	HA-p
H	$2.44 \times 10^{-8} \exp(-1350/RT)$	[5]	508–700 K, 10^3–10^5 Pa	GE-ID
D	$2.36 \times 10^{-8} \exp(-1350/RT)$	[5]	508–700 K, 10^3–10^5 Pa	GE-ID
T	$2.32 \times 10^{-8} \exp(-1350/RT)$	[5]	508–700 K, 10^3–10^5 Pa	Extr.
H	$4.66 \times 10^{-6} \exp(-13\,399/RT)$	[30]	600–900 K, 10^3–10^5 Pa	HA-p
T	$2.61 \times 10^{-6} \exp(-1274/RT)$		508–975 K	QRCSM
Recombination coefficient k_r (m^4/at s)				
T	$9.51 \times 10^{-26} \exp(-29\,717/RT)$	[32]	573–700 K, 3×10^3 Pa	Recomb.
T	$1.01 \times 10^{-25} \exp(-29\,350/RT)$	[32]	673–700 K, 10^3 Pa	Mass transp.

He-transport properties

Solubility	Semi-empirical approx. based on thermodynamic liquid model [33].	Li	8×10^{-13} at.frac./Pa
	High dependency on lithium activity	Pb	10^{-15} at.frac./Pa
		Pb–15.7Li	3×10^{-14} at.frac./Pa
Diffusion	Qualitatively: D(eut.-He)/D(Li–He) = D(eut.-H)/D(Li–H) [33]		D(eut.-He)/D(eut.-H) ~ 30

Two-phase dispersion models and correlations [34]

Dispersion coef. (l)	$\Sigma_l = u_{g,0} d (1 + 6.5 Fr^{0.8})/(13 Fr)$	Dispersion coef. (g)	$\Sigma_g = 0.2\ u_{g,0} d^2$
Gas hold-up	$\varepsilon_g/(1 - \varepsilon_g)^4 = 0.2\ Bn^{1/8}\ Ga^{1/12} Fr$	Mass transf. coef.	$ah_l = (D_{T,l}/d^2) 0.6 Sc^{0.5} Bn^{0.62} Ga^{0.31} \varepsilon_g^{1.1}$
Mean bubble diam.	$d_b = d 26 Bn^{-0.5} Ga^{-0.12} Fr^{-0.12}$	Froude number	$Fr = u_{g,0}/(gd)^{0.5}$
Bond number	$Bn = g d^2 \rho_l/\sigma_l$	Galilei number	$Ga = \rho g d^3/\mu_l$

$u_{g,0}$ inlet gas velocity; d extractor diameter

He bubble behaviour in blankets (ref. HCLL) [33]

Diameter ~ 20 nm	atoms per stable bubble ~ 10^4	low coalescing probabilities

Reactivity properties

Air and water [14]	ΔHr (LiOH) = −1.1 kJ/cm^3, ΔHr (Li_2O) = − 1.8 kJ/cm^3, no violent reaction is expected			
Gases	H.N	No formation of LiH or Li_3N, considered as no impurities [23]		
	O	Formation of Li_2O, $2.945–4016/T < \log C_0$ (wppm) $< 5.488–6145/T$ [14]		
Metals (solubility)	Ni	$\log_{10} s$(wppm) $= 4.832–981.2/T$ $T \in [520–728]$ K, [36]	Mn	$\log_{10} s$(wppm) $= 6.732–2938/T$ T: [531–783] K, [36]
	Cr	Too low to be measured	Fe	30 wppm [14]
	Bi	Thermodynamic calculations from [37], (Li_3Bi, 723 K) = 2.09×10^{-2} mol%Bi		
Ceramic materials	Theory for Sn–25Li from [24]. Unsatble: Fe_2O_3, NiO, Cr_2O_3, B_2O_3			

The extension of database considered is for: (i) magnetic properties, (ii) solute, both tritium and helium, transport properties and (iii) eutectic reactivity properties. Tritium and helium are both produced by neutron breeding in the eutectic and differently dissolved on it. Two-phase (helium bubbles, 1-phase eutectic) dispersion models represent the appropriate conceptual frame to describe physics of transport involved [33] and to establish extended database entries in terms of values and correlations. Safety analyses of fusion reactor systems refer functionally to tritium confinement. Precise and validated solute (tritium and helium) transport database: *diffusion coefficient*, *solubility constants* and *recombination rate constants* represent primary data with straight impact on component licensing and with a major impact on system sizing.

3.1. Tritium transport properties

Measurement of H-isotopes solubility is a difficult measurement potential full of parasitic effects. Zinkle [3] summarizes available information transport entries, measurement techniques and theoretical assessments. Measurements are visualized in Fig. 1 including extrapolations from other H-isotopes and theoretical approximations.

Such *unacceptable-for-licensing* huge data scattering and discrepancies between hot absorption and gas evolution measurements make independent determinations in connection with the eutectic QA routes urgent and compulsory for NFT. Gas evolution technique, able to check reversibility between absorption and desorption in eutectic samples, results in principle the most convenient technique. The measurements of the diffusion coefficient of hydrogen in the eutectic (Fig. 2) show higher agreement and acceptable dispersion with divergences caused by the experimental device, fitting models, surface effects and control of convection [5].

The surface recombination process depends empirically on the material surface status (cleanliness) and on surface concentrations [32]. Hence, its value depends totally on the experimental conditions and set-up.

3.2. Helium transport properties

Helium production rates in breeder blankets are nearly mol-to-mol linked $n(^{6}\mathrm{Li},^{4}_{2}\mathrm{He})^{3}_{1}\mathrm{H}$ to the compulsory requirement of high *T* self-sufficiency of future deuterium–tritium (DT) fusion reactors. He concentration can modify heat/mass/electrical transfer interfacial exchange coefficients between the liquid metal breeder and the structural material. Thus, the study of the helium concentration profile in the blanket or any other fusion component becomes essential. The two transport values needed are the helium solubility (*Henry's constant*) and diffusion coefficient. The experimental data for Henry's constant in liquid metals is today exotic and scarce. Measurements are not known of such magnitudes for Pb–Li eutectic. A first approximation of helium solubility in the eutectic was proposed in [33] based on values from the thermodynamic liquid hole model in liquid lithium and on cohesion models.

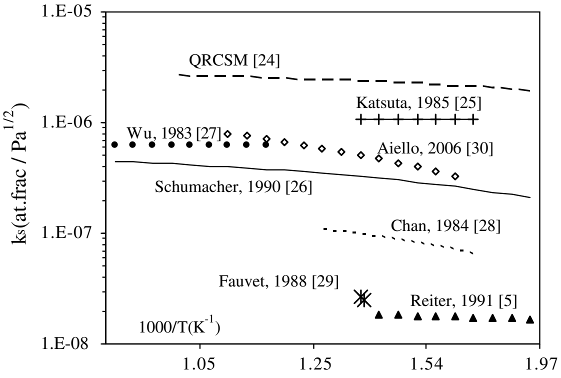

Fig. 1. Values of Sievert's solubility constant. (See above-mentioned references for further information.)

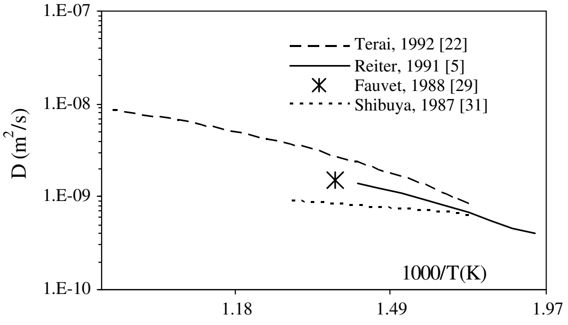

Fig. 2. Values of tritium diffusion coefficient. (See above-mentioned references for further information.)

In LIBRETTO tests [38], bubble nucleation and interfacial nucleation between the liquid metal breeder and the structural material was observed. If this phenomenon is confirmed and helium bubbles are finally formed, a large number of long residence time nucleating nano-bubbles could mean effective tritium sinks in liquid metal breeder channels. This would result in a net reduction of the tritium partial pressures in solution (and hence permeation fluxes) with some kind of additional 'natural' tritium extraction out from the liquid metal channels. Therefore, depending on bubble plume properties in terms of bubble characteristics (size and stability) and then concentration, a plume of bubble would impact on tritium transport schemes in LM channels. Up to now, helium effects and their system design implications have not been considered in system designs. However, 2-phase dispersion models have been studied for quite a long time within the context of tritium recovery systems, i.e. bubble columns [34,35], what can be applied for TBM design. First efforts on determining the bubble diameter and behavior in helium-cooled lithium lead (HCLL) blankets have been advanced in [33].

3.3. Lead–lithium eutectic reactivity

The analysis of reactivity properties of the eutectic is a huge area of research. It is not the aim of this study to report all the reactivity properties. Only the ones of major

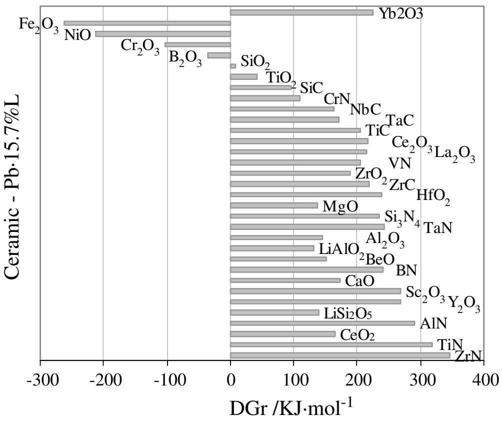

Fig. 3. Energy of reaction of selected ceramic materials in solute (O,N,C) saturated Pb–15.7Li at 773 K.

interest in liquid metal blanket designs are commented. In Fig. 3 the obtained results for ceramic materials stability in the eutectic are exposed.

4. Database as a computing expert system

Technical complexity of data inputs of the lithium–lead eutectic and practical uses for design suggest the need to manage it in a continuous updateable computing form according to the QA database requirements of a nuclear material. Such open tool can be used to fix data quality criteria and to manage data entries, in terms of reproducibility, differences on experimental or theoretical obtaining ways, dispersion, etc. Database interrogation can be useful to provide a given physical magnitude visualization with range constraints with expression of magnitude uncertainties for direct design sensitivity analyses.

5. Conclusions

State of the art of the lead–lithium database has been reported. Database entries needing further research, such as lithium proportion, electromagnetic properties and hydrogen and helium transport properties have been outlined. Extension of database inputs into additional properties in coherence with refinement of design and evolution has been proposed. In this direction, further developments towards the numerical implementation of 2-phase dispersion models for a bubble plume in LM conducts represents parallel coherent efforts.

References

[1] M. Kuchle, Technical Report, KfK, 1990.
[2] U. Jauch, V. Karcher, B. Schultz, G. Haase, Technical Report, KfK Report 4144, 1986.
[3] S. Zinkle, Summary of physical properties for lithium, Pb–17Li, and (LiF)nBeF$_2$ coolants, Technical Report, APEX Study Meeting, Sandia National Lab., 27–28 July 1998.
[4] P. Fauvet, J. Sannier, J. Nucl. Mater. 155–157 (1988) 516.
[5] F. Reiter, Fus. Eng. Des. 14 (1991) 207.
[6] B. Schulz, Fus. Eng. Des. 14 (1991) 199.
[7] P. Hubbertsey, T. Sample, M. Barker, J. Nucl. Mater. 191–194 (1992) 283.
[8] H. Feuerstein, L. Hörner, S. Horn, J. Nucl. Mater. 258–263 (1998) 505.
[9] J. Freibergs, J. Klavins, O. Lielausis, A. Mikelsons, J. Zandarts, in: The 15th Riga and 6th Pamir Conference on Fundamental and Applied MHD, Rigas Jurmala, Latvia, 2005, p. 259.
[10] S. Stankus, R. Khairulin, A. Mozgovoi, High Temp. 44 (6) (2006) 829.
[11] P. Hubberstey, in: Proceedings of the First Workshop on Liquid Metal Blanket Experminetal Activities, 16–18 September, CEA Headquarters, Paris, France, 1997.
[12] V. Prokhorenko, A. Kondyr', I. Pazdrii, Investigation of Structural Stability and Physicomechanical Properties of Eutectic in the Solid and Liquid Phases: A Report on Research Work, Lvov Polytechnical Institute, 1990.
[13] R.E. Buxbaum, J. Less Common Met. 97 (1) (1984) 27.
[14] Y. Poitevin, M. Küchle, Pb17Li for EU WCLL Test Blanket Module, Materials Assessment Report ITER Doc. G 74 MA 10 01-07-11 W 0.2.
[15] S. Malang, R.F. Mattas, Submitted to the Third International Symposium on Fusion Nuclear Technology, University of California at Los Angeles, 27 June–1 July 1994.
[16] H. Feuerstein, L. Hrner, S. Horn, J. Nucl. Mater. 258–263 (1998) 505.
[17] S. Malang, M.S. Tillack, Development of Self-cooled Liquid Breeder Blankets, FZKA 5581, November 1995.
[18] F. Sears, M. Zemansky, Física, Ed. Aguilar, 1979.
[19] D. Gugan, Phys. Rev. B 56 (13) (1997) 7759.
[20] N.H. Lu, S.K. Lai, J. Phys.: Condens. Matter 3 (1991) 2175.
[21] C.J. Ford, J. Heighway, E.F.W. Seymour, J. Phys. F: Met. Phys. 2 (1972) 373.
[22] T. Terai, S. Nagai, T. Yoneoka, Y. Takahashi, J. Nucl. Mater. 187 (1992) 247.
[23] P. Hubberstey, J. Nucl. Mater. 247 (1997) 208.
[24] S. Sharafat, N. Ghoniem, S. Zinkle, J. Nucl. Mater. 329–333 (2004) 1429.
[25] H. Katsuta, H. Iwamoto, H. Ohno, J. Nucl. Mater. 133&134 (1985) 167.
[26] R. Schumacher, A. Weiss, Phys. Chem. 94 (1990) 684.
[27] C.H. Wu, J. Nucl. Mater. 114 (1983) 30.
[28] Y.C. Chan, E. Veleckis, J. Nucl. Mater. 122&123 (1984) 935.
[29] P. Fauvet, J. Sannier, J. Nucl. Mater. 155–157 (1988) 516.
[30] A. Aiello, A. Ciampicheti, G. Benamati, Fus. Eng. Des. 81 (2006) 639.
[31] Y. Shibuya, M. Aida, Y. Fujii, M. Okamoto, J. Nucl. Mater. 150 (1987) 286.
[32] A. Pisarev, T. Tanabe, T. Terai, G. Benamati, M. Mullin, J. Nucl. Sci. Technol. 39 (4) (2002) 377.
[33] L.A. Sedano, CIEMAT Internal Report, 2007.
[34] F. Yoshida, Chem. Eng. Technol. 11 (1988) 205.
[35] C. Malara, Fus. Technol. 28 (1995) 693.
[36] M.G. Barker, T. Sample, Fus. Eng. Des. 14 (3-4) (1991) 219.
[37] P. Hubberstey, Thermodynamics of Pb–17Li–Bismuth Interactions, Nuclear Materials 212–15 (1994) 1021.
[38] L.A. Sedano et al., IEA Meeting on Liquid Breeder for Fusion Application ANL, Chicago, US, April 1999.