Helium bubbles inside liquid Pb16Li
A molecular dynamics study

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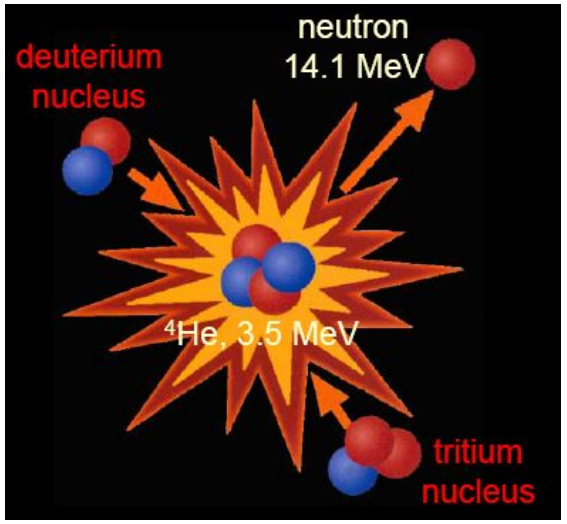
Outline

I Why PbLi?
II Why MD?
III PbLi interatomic potential
IV PbLi+ He
V He bubbles
VI Next steps?

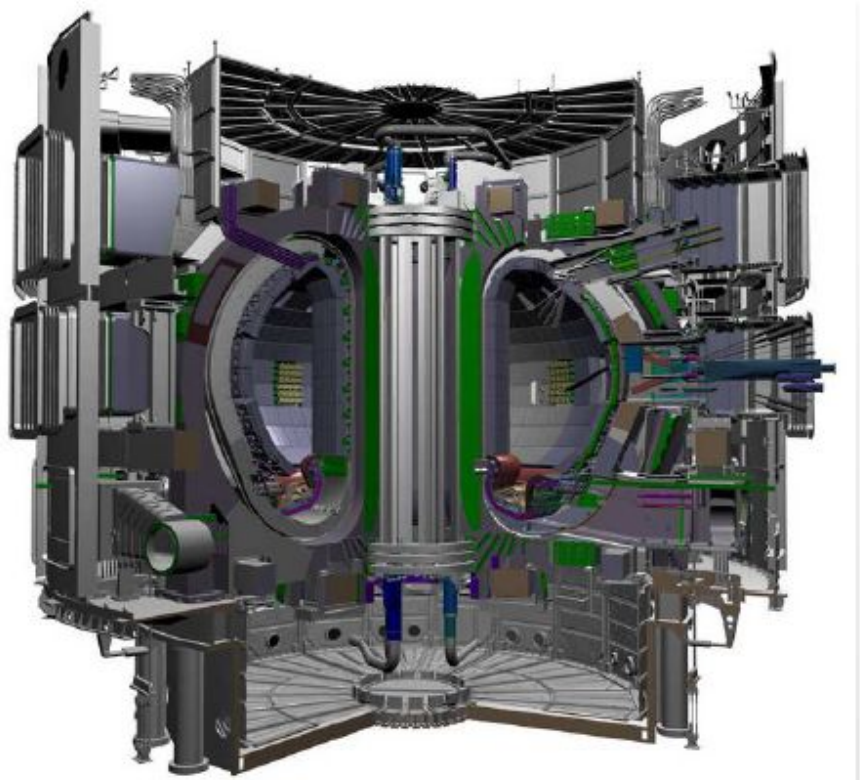
A breeding blanket consists of:

A tritium breeding material (Li-containing alloy)
A neutron multiplier (Be)
A coolant (water, He and/or Pb-Li)
A structural material

$$_3Li^6 + n \rightarrow {}_2He^4 + {}_1T^3$$



ITER



Breeding blanket: the kinetic energy of 14 MeV neutrons is transformed into heat that is transferred to a coolant.

Blanket solutions

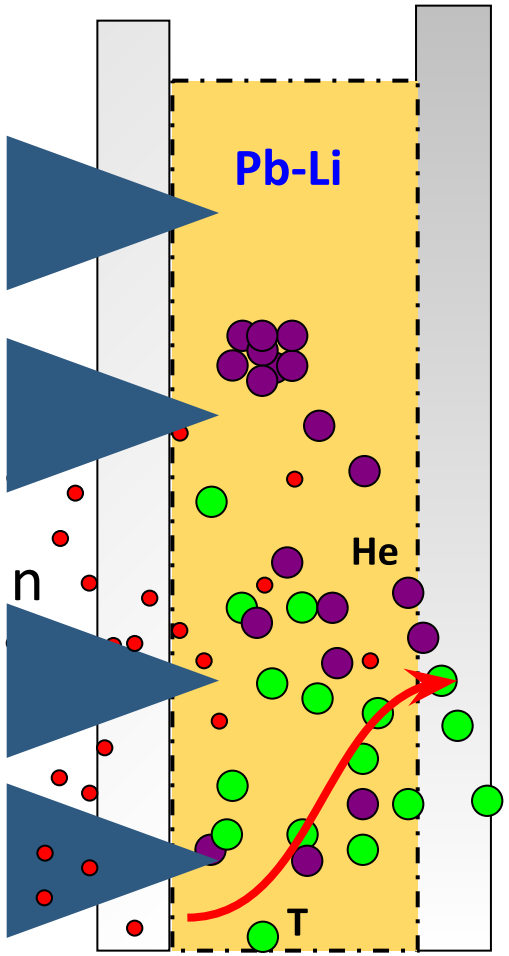
Pb16Li & other possible candidates:

Liquid breeder	**Li**	**Pb16Li**	**Flibe**	**$Li_{20}Sn_{80}$**
Melting point (ºC)	180	235	459	320
Density (g/cm^3) 873K	0.48	8.98	2.0	6.0
Li Density (g/cm^3) 873K	0.48	0.061	0.28	0.09
Breeding property	Good	Fairly good	Neutron multipler required	Neutron multipler required
Chemical stability	Active	Middle	Almost stable	Almost stable
Corrosion	Severe	Middle	HF exist severe	?
Tritium release form	HT, T_2	HT, T_2	HT,T_2 TF	HT, T_2

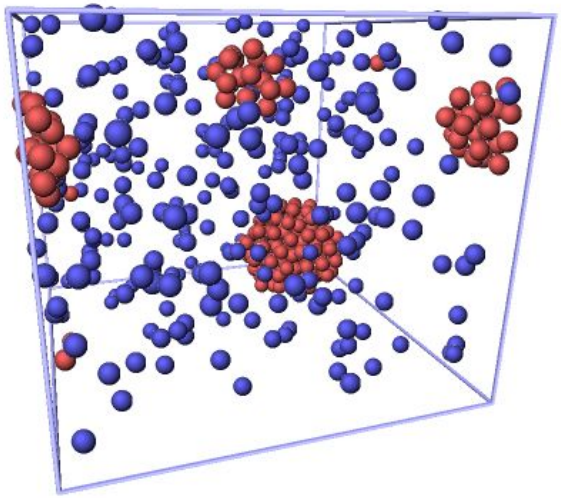
Giancarli L, Chuyanov V et al. 2007 **Test blanket modules in ITER: An overview on proposed designs and required DEMO-relevant materials** J. Nucl. Mater. 367–370, Part B, **1** 1271-1280

$$_3Li^6 + n \rightarrow {}_2He^4 + {}_1T^3 + 4{,}78MeV$$

$$_3Li^7 + n \rightarrow {}_2He^4 + {}_1T^3 + n + 2.47MeV$$



- Bubble nucleation & growth phenomena
- **Tritium is absorbed into He bubbles*??**
- Residence time of eutectic atoms in a material surface. Diffusion inside the structural material, corrosion ?



*J. Fradera *et al*. Journal of Nuclear Materials 417 (2011) 739–742

Tritium (& He) behaviour

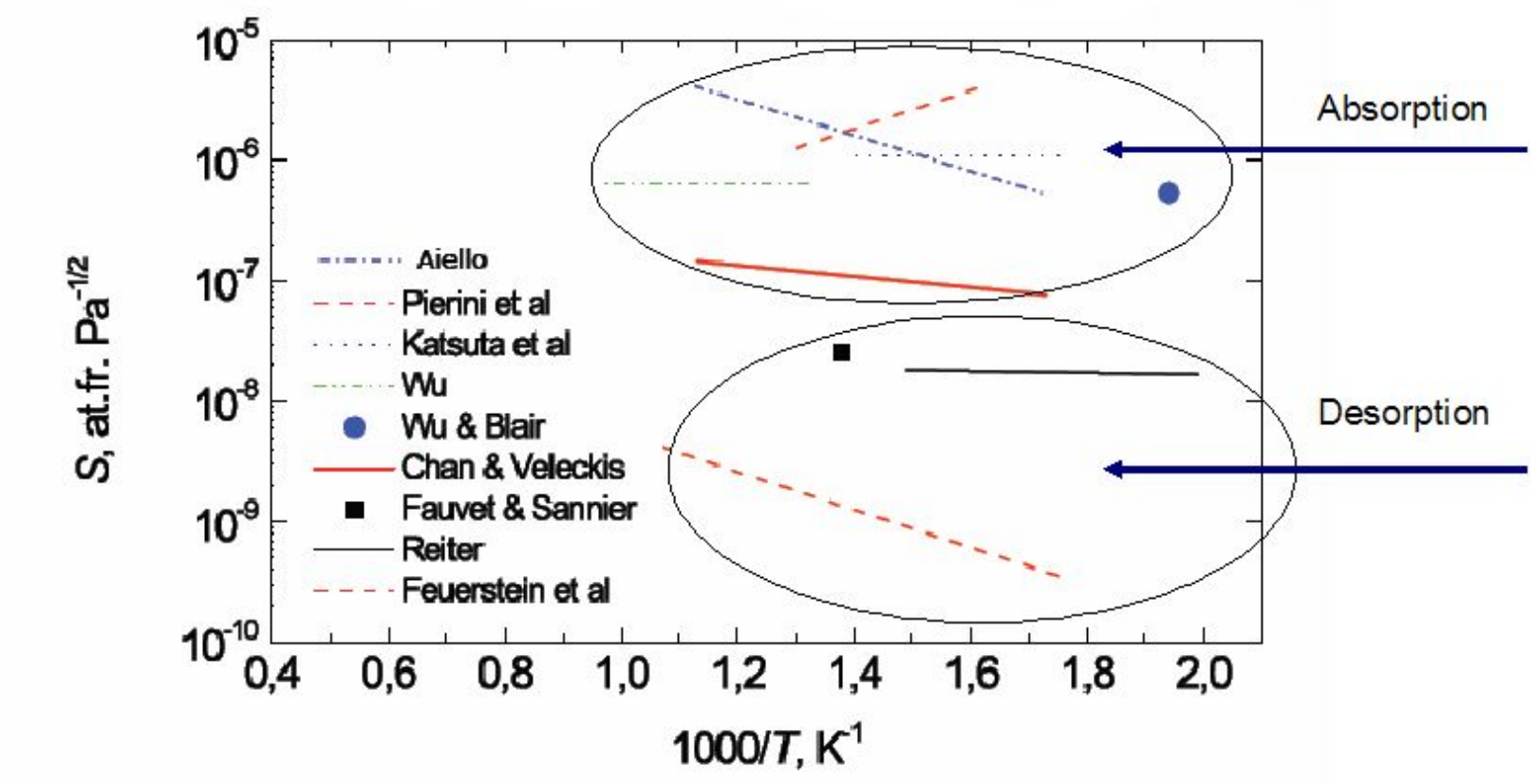
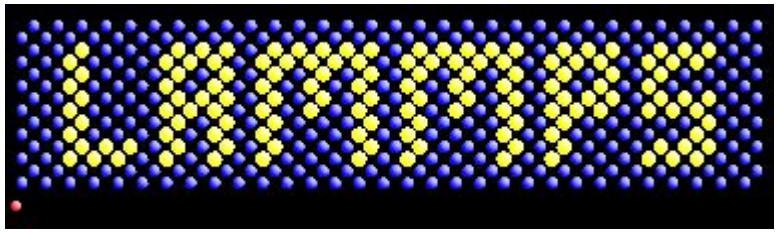
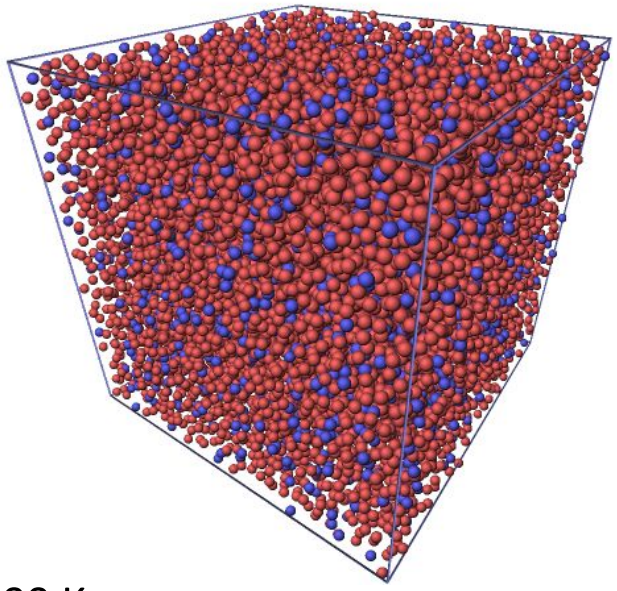


Fig. 1: Solubility database is inadequate for design. Scatter reflects experimental approaches and measurement techniques applied. Knowledge of dynamic transport properties (diffusion, mass transfer, interface processes) is limited [1].

[1] RICAPITO I. Liquid Metal Blankets for Fusion Reactors, Fusion Summer School, 10 Sept (2010)

Why MD





T = 1000 K
N = 100,000 atoms
Eutectic LiPb (16% Li)

MD simulations cost:

- Interatomic Potential ▫ EAM “cheap”
- Number of atoms ▫ 10^6 “expensive”
- Real Time ▫ 10 ns “expensive”

Price per core hour for the year 2020 is CZK 1,36 (excluding VAT) **(0,053 e)**

[1] S. Plimpton, Fast Parallel algorithms for short-range molecular-dynamics, J. Comput. Phys. 117 (1995) 1.
[2] http://lammps.sandia.gov – LAMMPS — Large-scale Atomic/Molecular Massively Parallel Simulator.

Pure metals

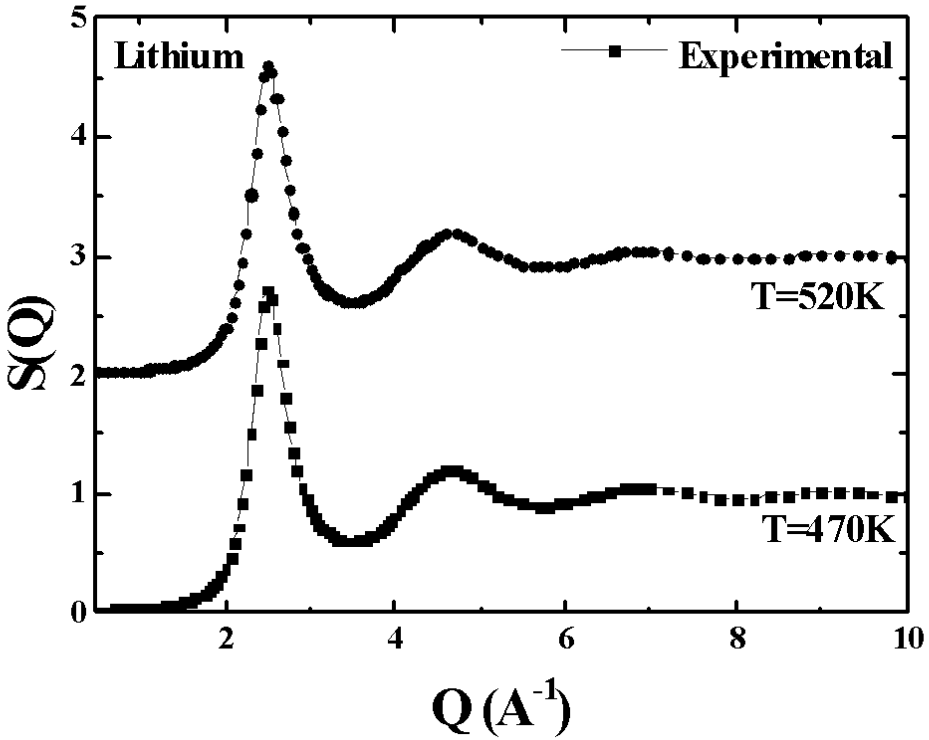


Fig2. Lithium S(Q) experimental (black)

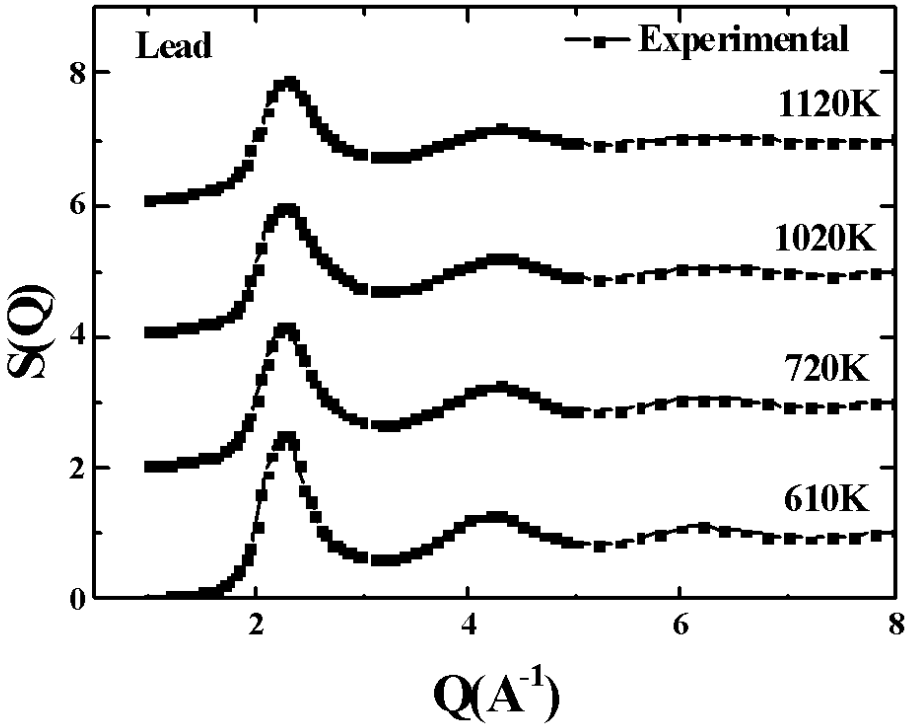


Fig. 2. Lead S(Q) experimental (black)

Pure metals

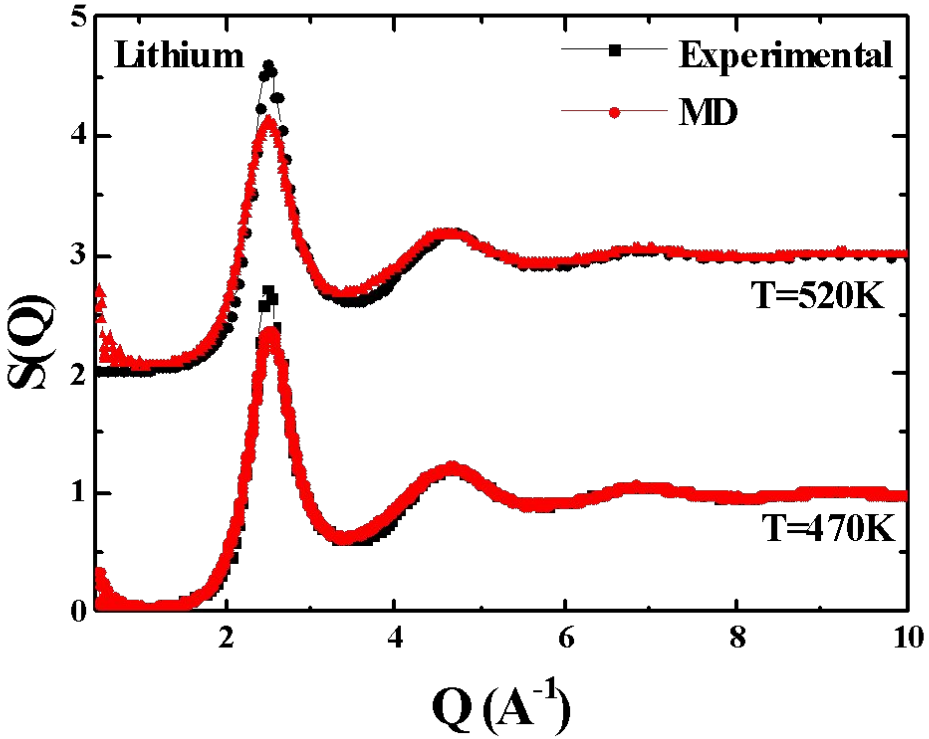


Fig.2. Lithium S(Q) calculated (red) & experimental results (black).

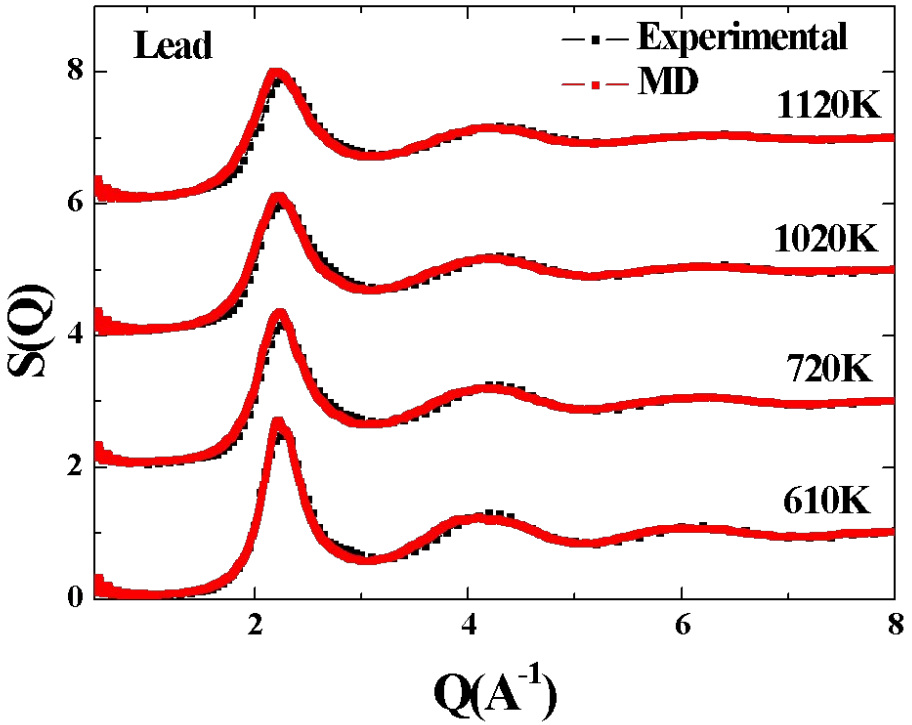


Fig. 2. Lead S(Q) calculated (red) & experimental (black)

Pb-Li Interatomic potential

ELSEVIER

Journal of Nuclear Materials
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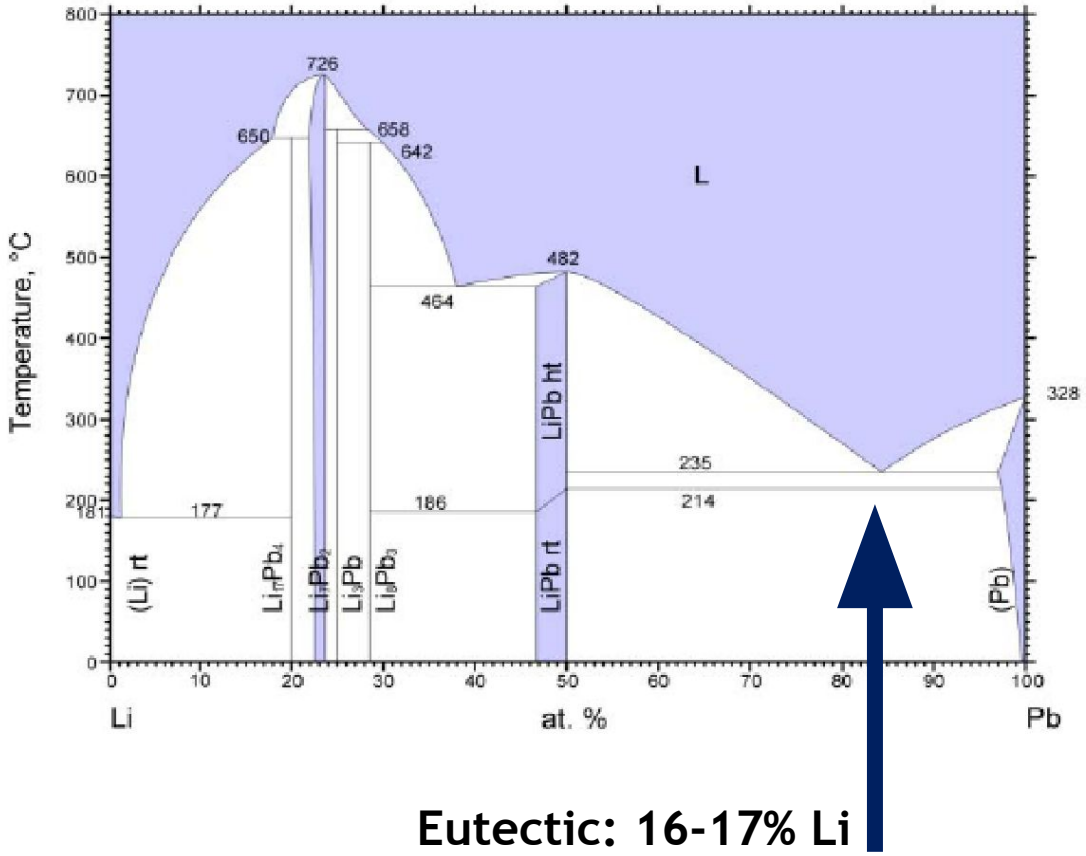
Interatomic potential for the compound-forming Li–Pb liquid alloy

Alberto Fraile [a], Santiago Cuesta-López [b], Alfredo Caro [c], Daniel Schwen [c], J. Manuel Perlado [a]

Pb-Li Interatomic potential



Eutectic: 16-17% Li

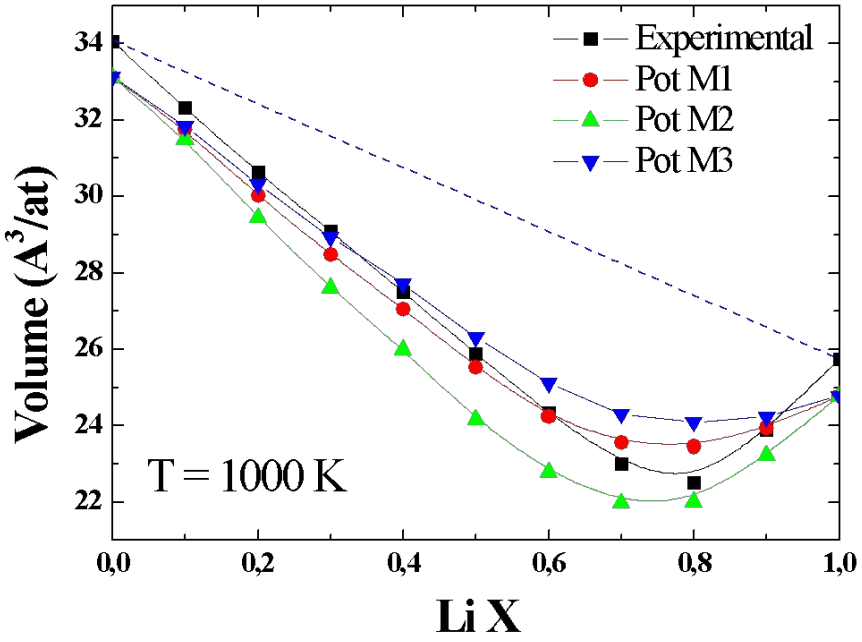


Fig.3 Volume of LiPb **liquid** alloys (T=1000 K). Experimental data (black squares). MD results calculated with 3 different potentials.

Fraile A, Cuesta-López S, Caro A, Schwen D, Perlado J M. 2014. J. Nucl. Mater. 448 103

Pb-Li + He

The potential function for the L-J 12-6 potential for atoms *i* and *j* is written as:

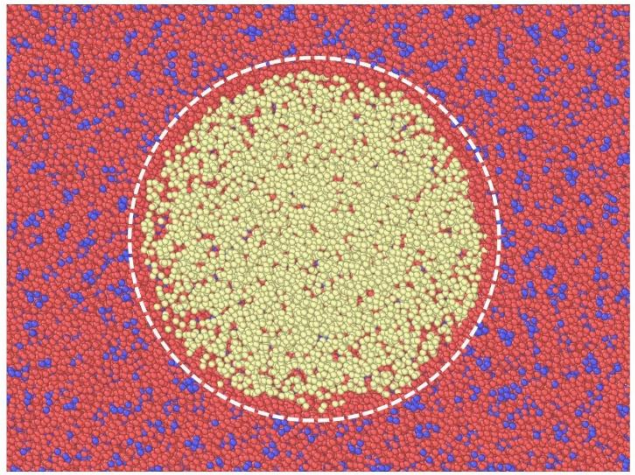
$$\phi_{ij} = \varepsilon_{ij}\left[\left(\frac{\sigma_{ij}}{r}\right)^{12} + \left(\frac{\sigma_{ij}}{r}\right)^{6}\right]$$

where r_{ij} is the distance between the *i*th and the *j*th particle.

For He-Pb we could not find a L-J potential, but knowing the L-J for Pb-Pb [44], and with Lorentz-Berthelot rules [45]

$$\sigma_{ij} = \frac{\sigma_{ii}+\sigma_{jj}}{2} \qquad \varepsilon_{ij} = \sqrt{\varepsilon_{ii}\varepsilon_{jj}}$$

the He-Pb values were calculated.



	ε (eV/atom)	σ (Å)	σ_c (Å)	Ref
He-He	0.00088	2.556	8.0	[42]
He-Pb	0.01057	3.0667	8.0	[44,45]
He-Li	0.000141	5.3565	8.0	[43]

Table 1. L-J parameters for the He-He and He-Metal interactions. σ_c is the cutoff distance.

[42] Aziz R A, et al. 1979 J. Chem. Phys. **70**, 4330.
[43] Dehmer P and Wharton L 1972. The Journal of Chemical Physics **57**, 4821.
[44] Heinz H, at al 2008. J. Phys. Chem. C **112**, 17281-17290
[45] Lorentz H A 1881. Annalen der Physik. **248** (1)

Critical Radius R_c

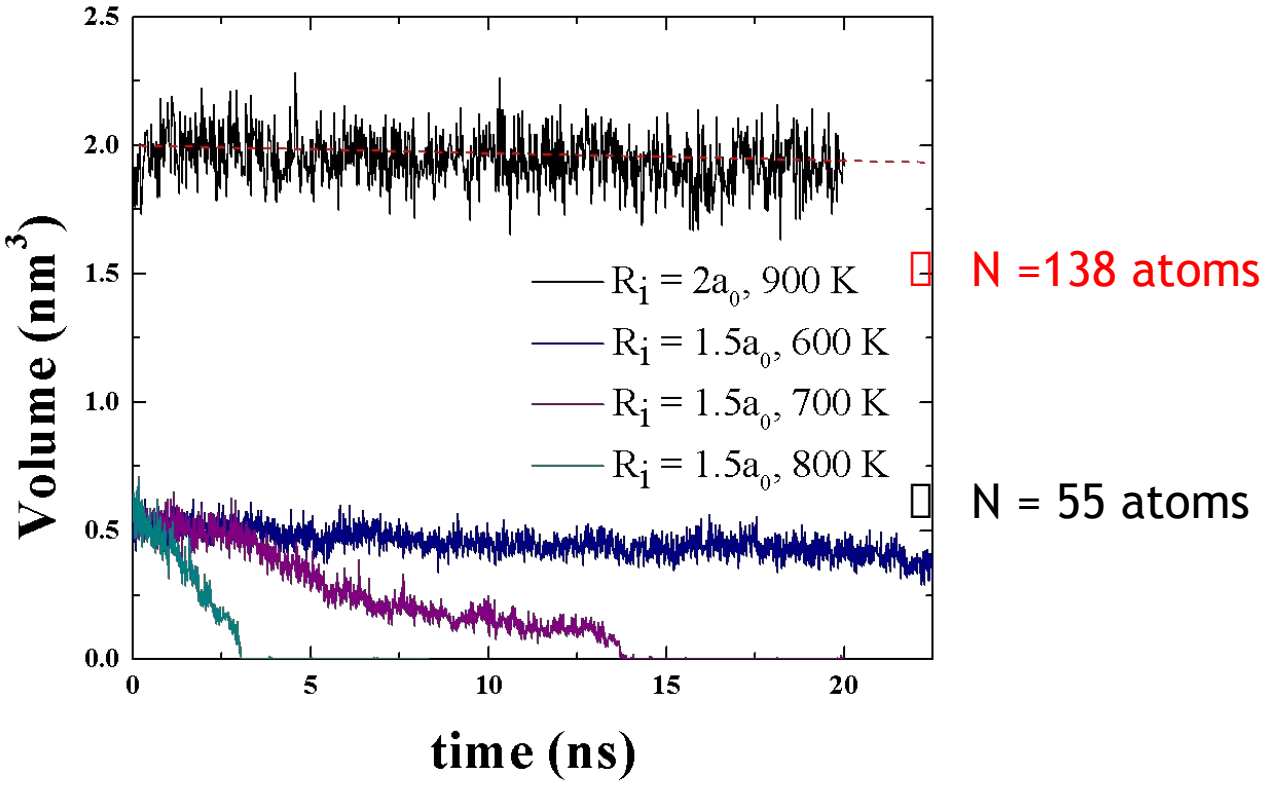


Fig. 4. Time evolution of the volume of bubbles with a different number of atoms (different initial R), for different Ts.

R < R_c ⇨ bubble dissolves

In Ref [48], number of atoms per stable bubble is estimated to be **~10^4.**

Here we show that bubbles with N well below 500 atoms are stable ($R_c \geq$ 2nm).

[48] E. Mas de les Valls *et al* 2008 Lead–lithium eutectic material database for nuclear fusion technology. J. Nucl. Mater **376** 353–357

Radii vs N

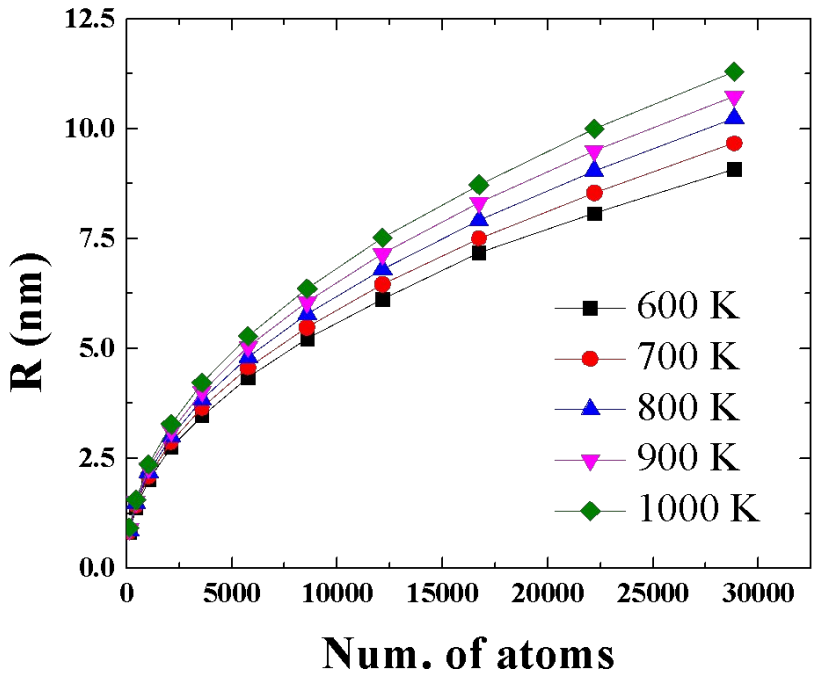
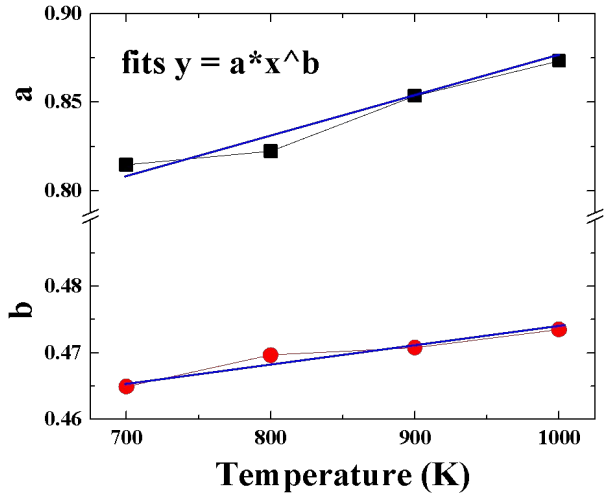


Fig 5. Radii (nm) *vs* number of **He atoms** for five different temperatures, 600, 700, 800, 900 and 1000 K.

Perfect fits are obtained by fitting the curves to a simple **allometric hyperbolic function**

y = a*x^b,



a_1 = 0.66483
a_2 = 2.07151*10^{-4}
b_1 = 0.44688
b_2 = 2.68516*10^{-5}

▫ in the temperature range 700 to 1000 K, R(N, T) in Å :

$R(N,T) = (a_1 + a_2*T)*N^{(b_1+b_2*T)}$

Pressure

the gas pressure P equals the capillarity force
$2\gamma/R$
where γ is the surface energy of the host material and R is the bubble radius, which is known as the **Young-Laplace equation**.

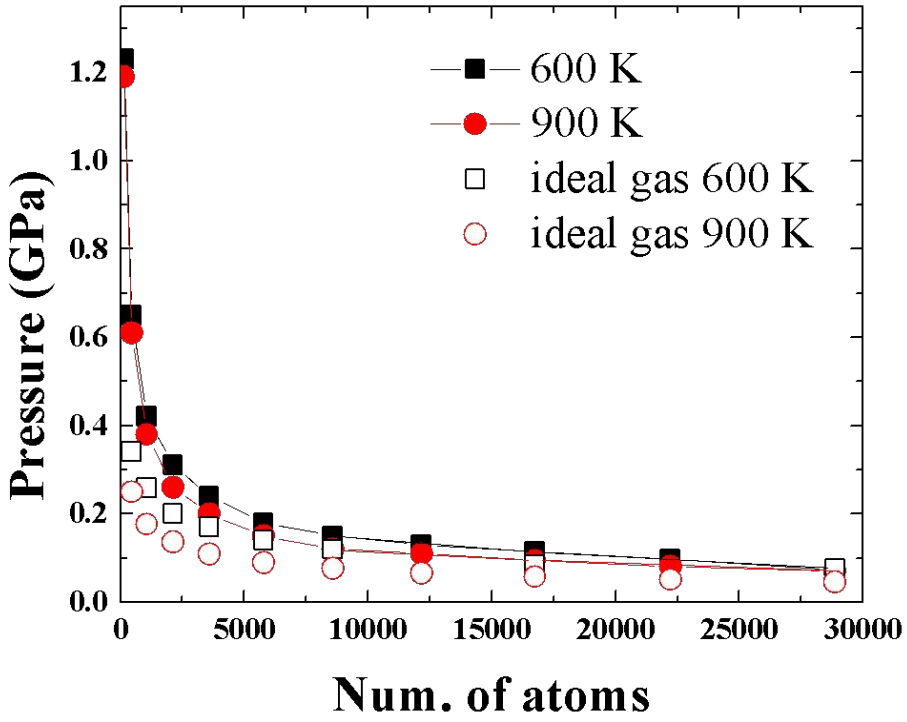


Fig 6. Pressure of He nanobubbles (GPa) *vs* number of atoms for two different temperatures, 600 and 900 K

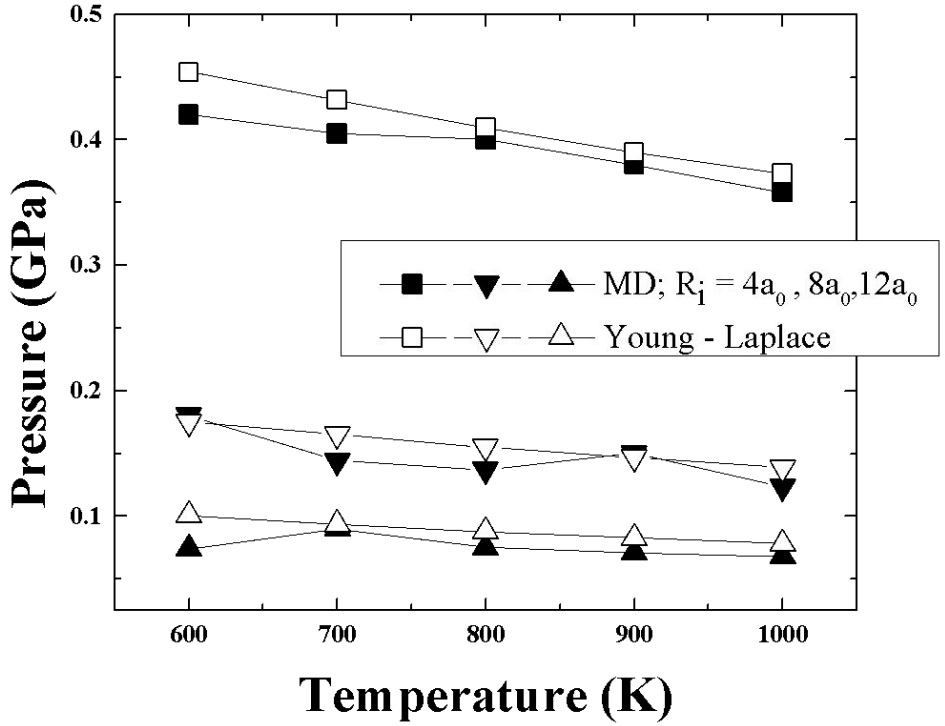


Fig 7. Pressure of He nanobubbles (in GPa) *vs* T for 3 different bubble sizes, (R_i = $4a_0$, $8a_0$ & $12a_0$) according to our MD, and calculated using the Young-Laplace eq.

Caution!

Transport properties are quite potential dependent! **

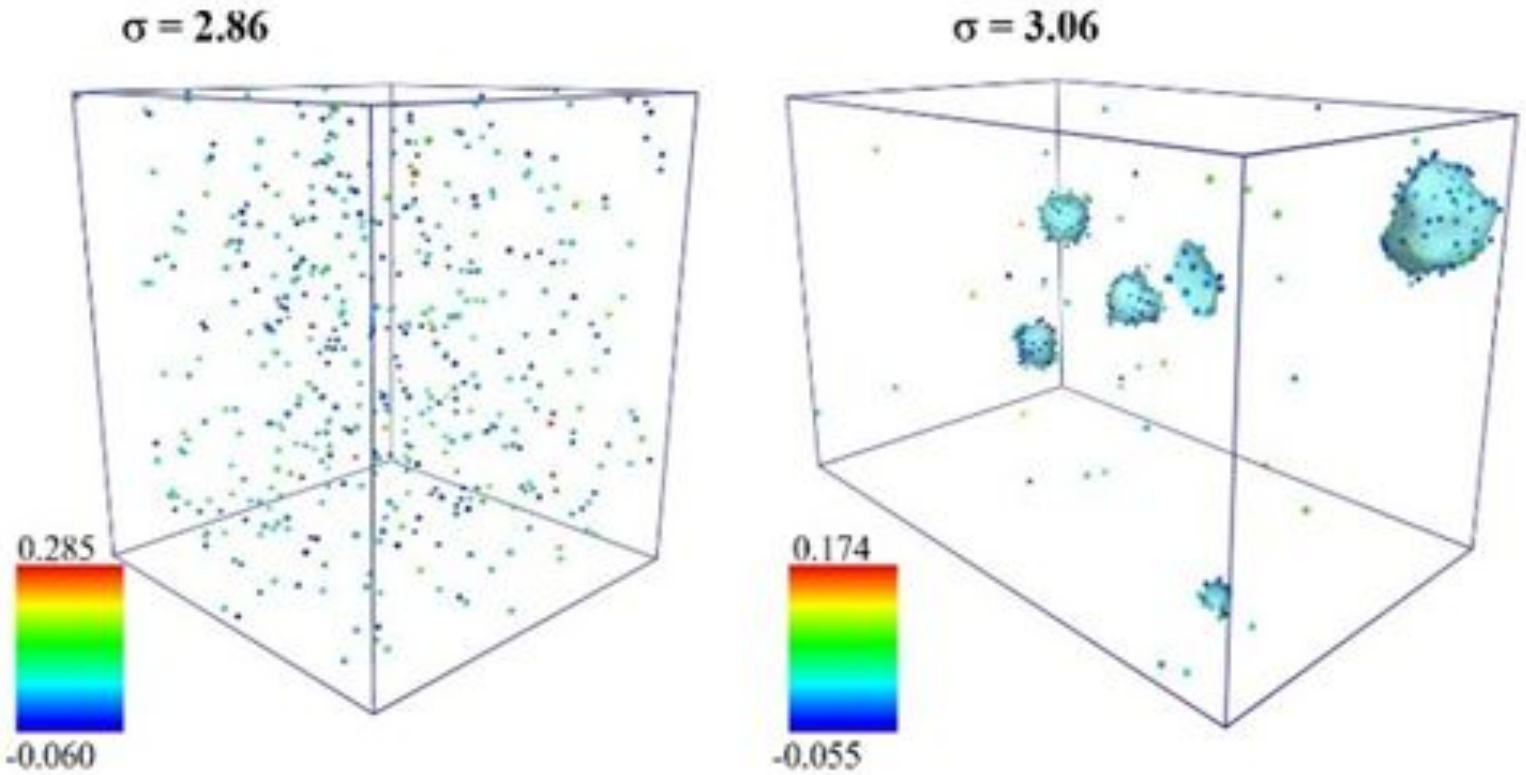


Fig. 8. Two examples of He nucleation; left, He-Pb σ =2.86 Å and right σ = 3.06 Å. T = 900 K. The snapshots correspond in both cases to t =10 ns.

** See Supplementary material in A. Fraile and T. Polcar. Nuclear Fusion 2020.

Summary & Next (?)

for first time…

- Critical radius has been determined (≥500 atoms, R≥2nm)
- Volume *vs* N has been calculated

- $$R(N,T) = (a_1 + a_2*T)*N^{(b_1+b_2*T)}$$

- Pressure *vs* N (or V) has been evaluated
- Caution! Diffusion is highly dependent on the potentials
- Next ? PbLi16 +He+T

  interaction of T with Li is more complex to model

Thank you for your attention

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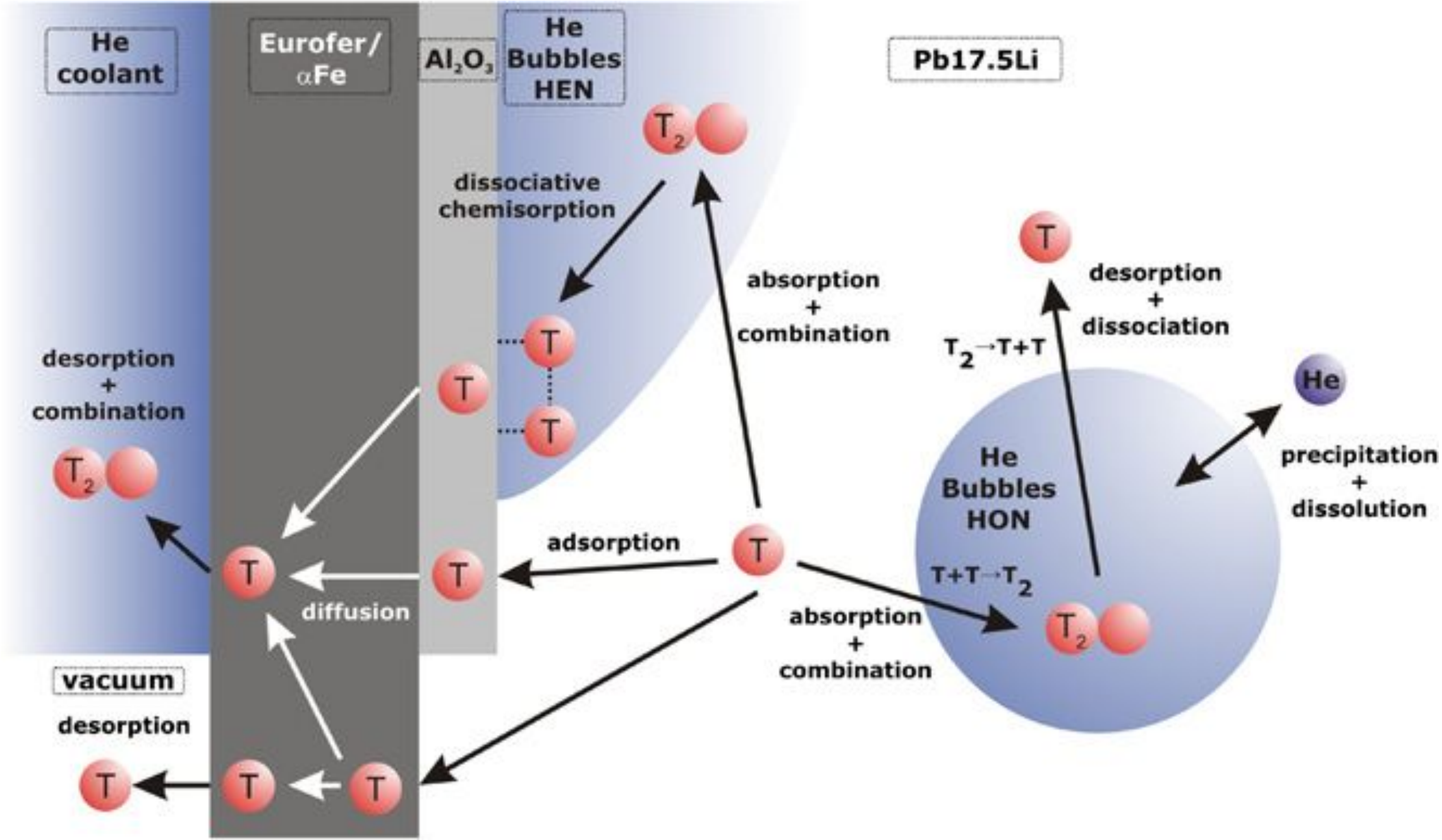
Further reading

[1] Fraile A, Cuesta-López S, Caro. A, Perlado J. M. Journal of Nucl Mater. 440, 1-3, 2013, 98-103
[2] Fraile A, Cuesta-López S, Caro A, Schwen D, Perlado J M. 2014. J. Nucl. Mater. 448 103
[3] A. Fraile and T. Polcar. Volume and pressure of helium bubbles inside liquid Pb16Li. A molecular dynamics study. Nuclear Fusion 2020.
[4] R. Serrano-Lopez et al., "Molten Salts Database for Energy Applications," Chem. Eng. Process., 73, 87 (2013).

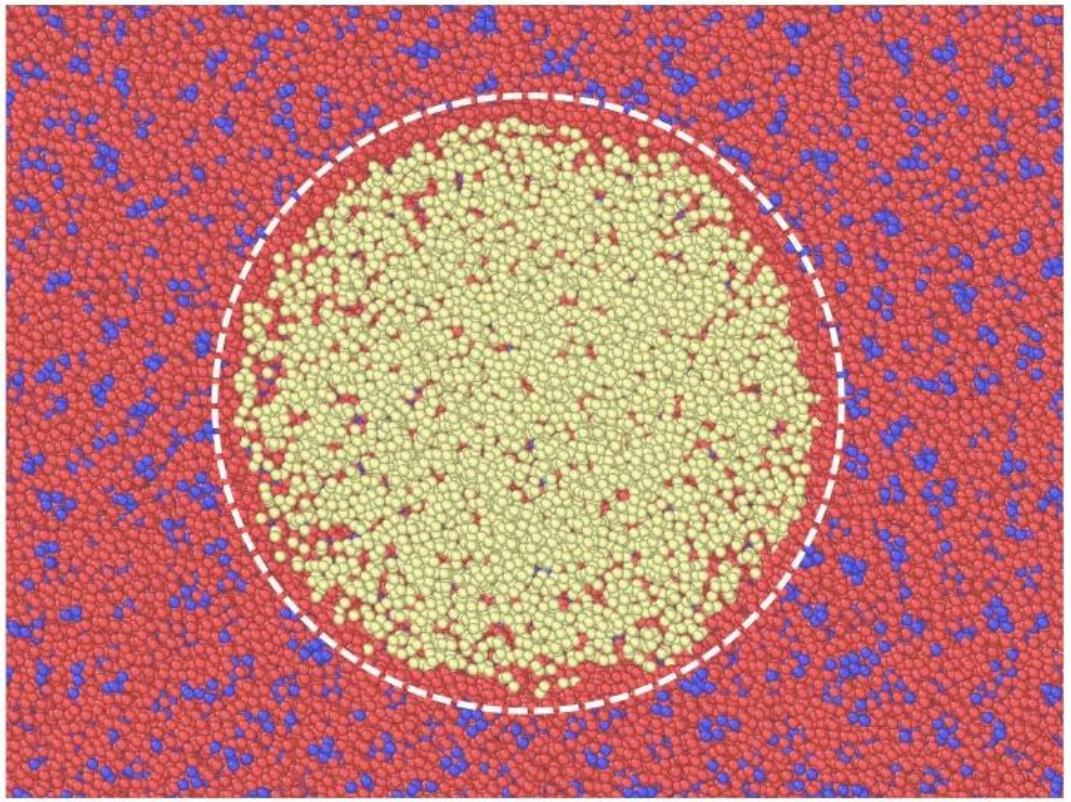


J. Fradera *et al*. Journal of Nuclear Materials 417 (2011) 739–742

Tritium

$$_3Li^6 + n \rightarrow {}_2He^4 + {}_1T^3$$

Due to the expected bonding od T to Li [61-63] and the existence of Pb boundary at bubble-liquid alloy interface, **we may conclude that the concentration of T inside the bubbles will be limited.** Therefore, the effect of T on the volumes and pressures of the He bubbles presented here would be **presumably** negligible.



Structure of a He bubble inside liquid Pb16Li (N=8583 atoms, T=700K, R_f~6nm). Box sliced to show the interior of the bubble. Red atoms Pb, blue atoms Li, yellow ones He. **A two-atom thick shell of Pb atoms around the He bubble is observed.**

[61] Kobayashi M et al 2012 Fusion Science and Technology 62, 1
[62] D. Masuyama, T. Oda, S. Tanaka, S. Fukada 2009. Chem. Phys. Lett. 483 214–218.
[63] N. Jakse and A. Pasturel 2014. Phys Rev B 89, 174302

Pb-Li Viscosity

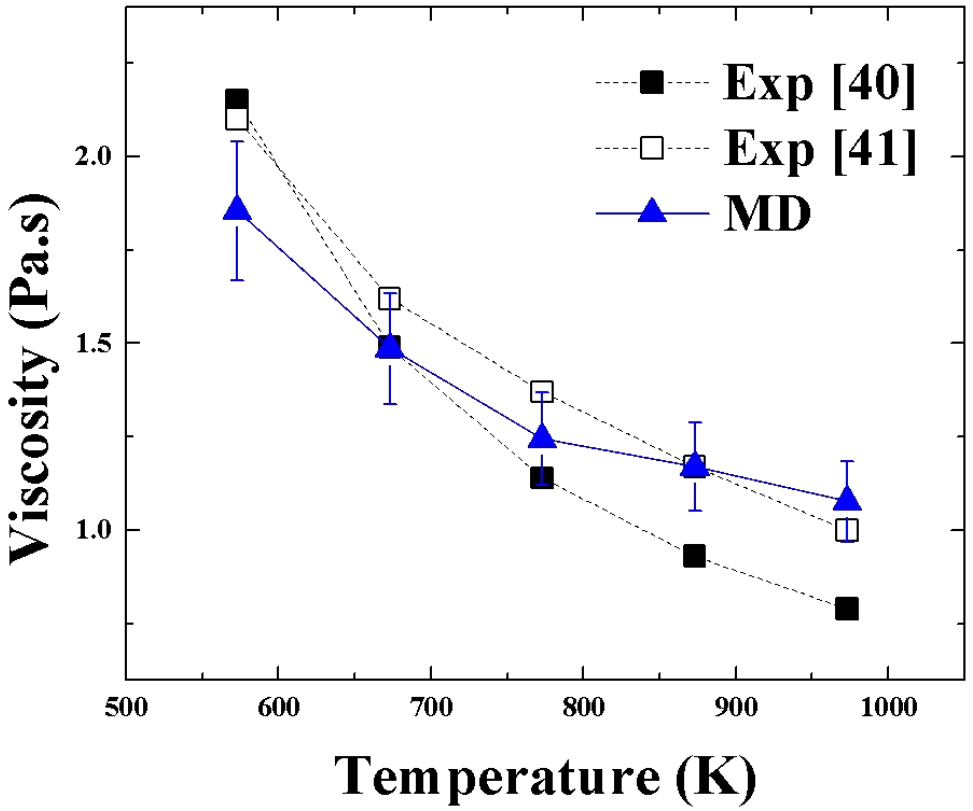


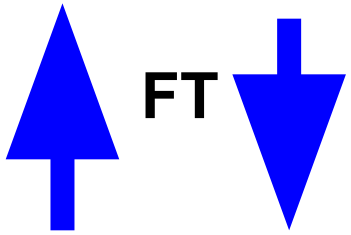
Fig 1. Dynamic viscosity calculated using the EAM potential for liquid Pb16Li eutectic described in our previous work [38]. Experimental values are from [40] and [41].

[38] Fraile A, Cuesta-López S, Caro A, Schwen D, Perlado J M 2014 Interatomic potential for the compound-forming Li-Pb liquid alloy. J. Nucl. Mater. 448 103.
[40] Hubbertsey P, Sample T, Barker M, 1992 J. Nucl. Mater. 191–194 283
[41] Mogahed E A, Kulcinski G L, Bibliography of a Promising Tritium Breeding Material – Pb83Li17, University of Wisconsin, 1995 UWFDM-994.

Testing Li & Pb potentials III

- Structural properties

Pair distribution function



Total structural factor

$$R(r) = \frac{1}{N}\sum_{\nu}\sum_{\mu}\frac{b_\nu b_\mu}{\langle b\rangle^2}\delta(r - r_{\nu\mu})$$

$$\rho(r) = \frac{1}{4\pi r^2}R(r)$$

$$g(r) = \rho(r)/\rho_0 = \frac{R(r)}{4\pi\rho_0 r^2} \qquad \text{thus } g(r) \to 1 \text{ for } r \to \infty$$

$$G(r) = 4\pi r\rho_0(g(r) - 1)$$

$$G(r) = \frac{2}{\pi}\int_0^\infty Q[S(Q) - 1]\sin(Qr)\mathrm{d}Q$$

$$S(Q) = 1 + \frac{1}{Q}\int_0^\infty G(r)\sin(Qr)\mathrm{d}r$$

LiPb MD

CMD
vs
ab initio [13]

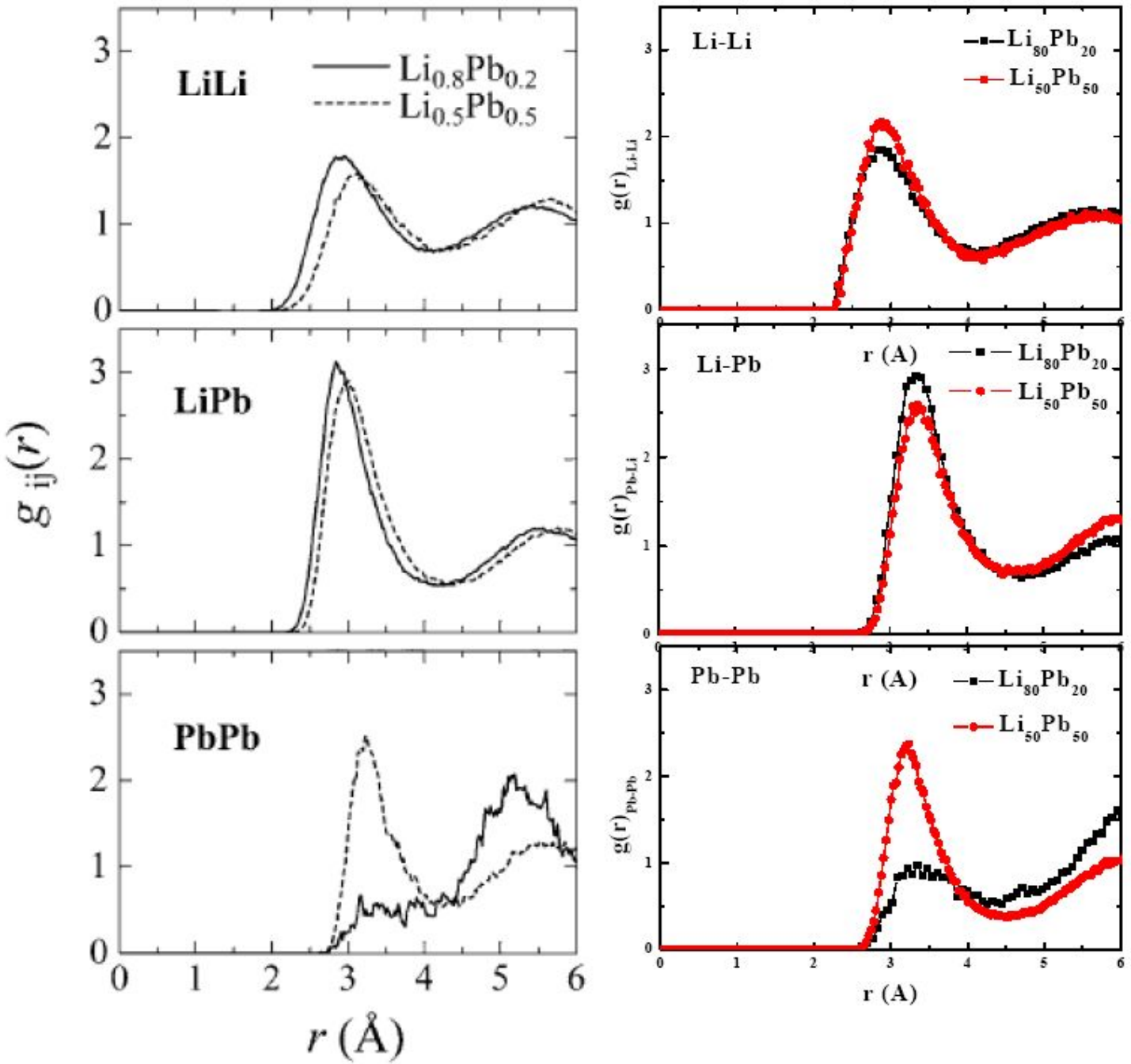


Fig. 5. RDF $g_{LiLi}(r)$, $g_{LiPb}(r)$ and $g_{PbPb}(r)$ for the liquid $Li_{80}Pb_{20}$ (solid line - black) and $Li_{50}Pb_{50}$ (broken line - red) alloys. T= 1075 K and 805 K for the liquid $Li_{80}Pb_{20}$ and $Li_{50}Pb_{50}$ alloys, respectively [1].

[1] Senda et al. The ionic structure and the electronic states of liquid Li-Pb alloys obtained from *ab initio* molecular dynamics simulations. J. Phys.: Condens. Matter 12, 6101 (2000)