ICANS XIX,
19th meeting on Collaboration of Advanced Neutron Sources
March 8 – 12, 2010
Grindelwald, Switzerland

LEAD GOLD EUTECTIC, FIRST STEPS TOWARDS THE QUALIFICATION OF A NOVEL TARGET MATERIAL FOR ESS

KNUD THOMSEN
Paul Scherrer Institut, 5232 Villigen PSI, Switzerland

and

K. CONDER[1], Y. DAI[1], R. FRISON[1], D. KISELEV[1], M. MEDARDE[1],
R. MOORMANN[2], E. PLATACIS[3], E. POMJAKUSHINA[1], S. TÖRÖK[4], L. ZANINI[1],
P. ZIMMERMANN[1]

[1]*PSI, 5232 Switzerland*

[2]*FZ Jülich, 52425 Germany*

[3]*IPUL, 2169 Latvia*

[4]*KFKI, 1525 Hungary*

ABSTRACT

Motivated by substantial problems related to mercury, a so far rather unknown heavy liquid metal, Lead Gold Eutectic (LGE), has been proposed as target material in the European Spallation Source ESS. LGE appears to combine the advantages of mercury and Lead Bismuth Eutectic (LBE) while avoiding their specific pitfalls. In order to arrive at a secure qualification of LGE in relatively short time, diverse efforts have been started. Their first main objective is to identify possible show stoppers. After some initial investigations, a quick and promising route to gain experience and confidence in the new material has been proposed: where applicable, tests involving LGE will be accompanied by basically identical tests using LBE. By performing many different investigations in parallel, a most direct comparison of the behavior and effects on structural materials of the two liquid metals will be possible. Thus it will be attempted to transfer much of the characterisation and qualification of the rather well known LBE to the novel material LGE.

1. Motivation

1.1. Mercury is a most problematic target material

The ESS reference design as devised 2003 uses mercury as heavy liquid metal target material [1]. In the meantime, some boundary conditions have changed and several factors nowadays make mercury a most unlikely choice [2]:

- The consequences of the chemical toxicity of mercury have been reassessed and found to be much higher than previously assumed.
- There is now an international move to ban all avoidable use of mercury.
- Sweden has issued a law to ban all use of mercury in Sweden.
- The baseline design foresees purportedly unavoidable very high temperatures for the mercury in the target.

- With the selection of the site at Lund amidst a densily populated area very strict effective limits and constraints for accident cases apply.

The Swedish law demands the careful assessment of alternatives before any use of mercury could be considered. As demonstrated by the successful design, licensing and operation of the MEGAPIE liquid metal target in PSI's SINQ facility, lead bismuth eutectic offers a viable alternative to mercury [3].

1.2. Lead Bismuth Eutectic LBE is problematic, too

Lead Bismuth Eutectic is rather similar to mercury in terms of neutron production, the major purpose of a spallation neutron source, and it offers two intrinsic advantages compared to mercury:

- The chemical toxicity of LBE is significantly lower than the one of mercury.
- The volatility is very much lower for LBE than for mercury; the boiling points are around 1700 °C and 357 °C, respectively.

The at first sight beneficial comparison of LBE with mercury has to be amended; LBE has two unfavourable features:

- Comparably large amounts of alpha-emitting polonium are produced from bismuth during irradiation [4].
- After solidification the volume increases by about 3 % caused by a phase transition in the solid phase [5].

Given the above shortly listed features it is clear that a more favourable liquid metal target material ought to combine the advantageous characteristics, i.e. low chemical toxicity, low volatility and limited phase changes while solid.

2. Lead Gold Eutectic LGE

From a literature survey the system lead-gold appears to offer an alternative to the hitherto considered liquid metals as target material in a pulsed spallation source. The little known eutectic contains 16 % gold and promises the following attributes:

- Accessible melting point at around 215 °C.
- High boiling point around 1700 °C.
- High density of about 12.5 g/cm^3.
- Little polonium production (due to Bi impurities in the lead).
- Limited mercury production during irradiation.
- Limited expansion due to phase changes in the solid phase.
- From a decommissioning and disposal point of view LGE looks best among the “bad” target materials available.

Figure 1 reproduces a phase diagram of the binary system lead – gold as found in the scarce literature on this material [6,7].

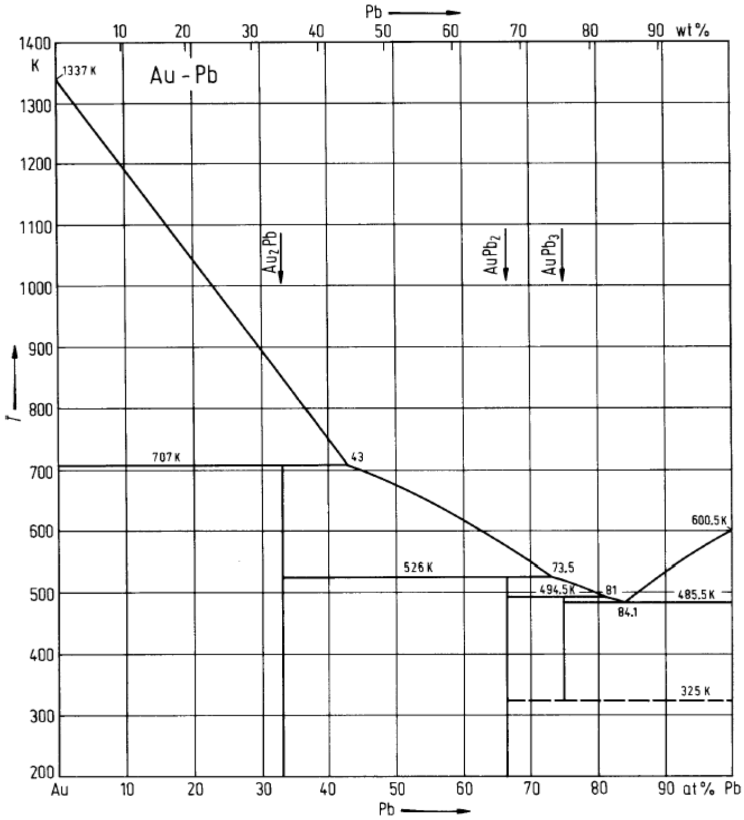

Figure 1. Phase Diagram of Lead – Gold [6,7].

3. Very First Results

Neutron production being the main point for building a spallation source, as a very first step the possible neutron yield of a LGE target has been quickly assessed [4]. Without any optimisation for the different liquid metals, neutron fluxes have been calculated in the baseline ESS geometry. Whereas some difference is seen for the flux inside the target due to the higher thermal absorption cross section for gold than for bismuth, the interesting fluxes inside the moderators for LGE and LBE are virtually identical, see Figure 2.

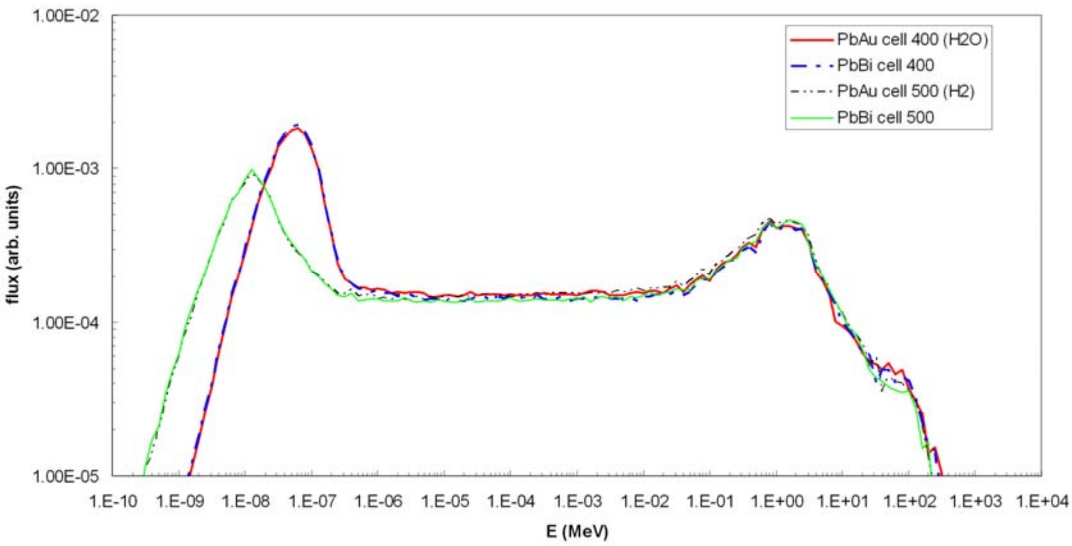

Figure 2. Neutron Fluxes inside two moderators for LGE and LBE, respectively.

During the preparation of LGE on a gram- and also on a kg-scale nothing unexpected was observed, and the measured melting temperatures confirmed the anticipated value.

4. Next Steps

As of the time of the writing of this brief paper, first experiments are underway in an attempt to very quickly identify possible show stoppers. X-Ray diffraction measurements on all samples of LGE available so far aim at quantifying any phase changes and associated volume variations in solid LGE at PSI.

At IPUL, Latvia, a twin loop has been set up where structural materials' specimens are exposed to flowing liquid metals under identical conditions, except the very liquid. By directly comparing corrosion effects in LGE to the ones in LBE, it is attempted to gain a fast cross-reference linking the unknown LGE to the much better understood LBE. This strategy of direct comparison between these two potential target materials will also be followed during more sophisticated tests in case the initial findings comply with our hopeful expectations.

5. Acknowledgements

Stimulating discussions with D. Schumann, J. Neuhausen and S. Heinitz, all PSI, and with O. Lielausis, IPUL, Latvia, are gratefully acknowledged.

6. References

1. ESS Council, The ESS Project Volume III Technical Report, ISBN 3-89336-303-3, May 2002.
2. R. Moormann, M. Medarde, E. Platacis, K. Thomsen, Lead Gold Eutectics as a target material for ESS, Presentation on TCADS-1, Karlsruhe, 15.-17.3.2010, Proc. in press.
3. W. Wagner, F. Gröschel, K. Thomsen, H. Heyck, MEGAPIE at SINQ – The first liquid metal target driven by a megawatt class proton beam, Journal of Nuclear Materials, Volume 377, Issue 1, 30 June 2008, Pages 12-16.
4. F. Carinci and L. Zanini, Neutronic and Nuclear Calculations for the Pb/Au Target Option for ESS, PSI TM-85-09-12, 26 Oct. 2009.
5. H. Glasbrenner, F. Gröschel, H. Grimmer, J. Patorski, M. Rohde, Expansion of solidified lead bismuth eutectic, Journal of Nuclear Materials, Volume 343, Issues 1-3, 1 August 2005, Pages 341-348.
6. Landolt-Börnstein - Group IV Physical Chemistry, Numerical Data and Functional Relationships in Science and Technology, Volume 19B5: Binary Systems. Part 5: Binary Systems Supplement 1, www.springermaterials.com/pdfs/10.1007/978-3-540-45280-5_31.pdf
7. J. Wang, H.S. Liu and Z.P. Jin, Thermodynamic assessment of the Au–Pb system, Calphad 28, Issue 1, 2004, 91-95.