Numerical study of helium solubility and helium bubble stability in mercury

Bo Lu [a,*], Arthur E. Ruggles [b], Matthew W. Francis [c]

[a] Institute of Plasma Physics, Chinese Academy of Sciences, PO Box 1126, Hefei, Anhui 230031, China
[b] Department of Nuclear Engineering, University of Tennessee, Knoxville, TN 37996-2300, USA
[c] Oak Ridge National Laboratory, PO Box 2008, MS6172, Oak Ridge, TN 37831-6172, USA

ABSTRACT

Dispersing small helium bubbles in the liquid mercury target of the high-power spallation neutron sources was proposed to add compressibility to the target made of liquid mercury. The pressure rise from proton beam deposition is reduced due to added compressibility, which in turn mitigates cavitation damage to the target boundary. A gas volume fraction of ~0.5% with a nominal bubble diameter of ~30 μm is desired for optimal pressure pulse relaxation at the beam power of >1 MW. Initial gas injection experiments performed in the Oak Ridge National Laboratory encountered difficulty in obtaining the required volume fraction in mercury. Gas dissolution and diffusion in mercury were candidate mechanisms for this behavior. To clarify this, the solubility of helium in mercury is evaluated in this study and compared to the available experimental data. The results indicate that helium has very small solubility in mercury and that the solubility increases with system temperature. Based on the predicted solubility values, bubble size evolution due to mass diffusion is simulated numerically. Mass diffusion induced bubble size evolution does not significantly affect bubble behavior for conditions expected in high power spallation targets.

© 2013 Elsevier Ltd. All rights reserved.

1. Introduction

Spallation neutron source (SNS) is an accelerator-based pulsed neutron source developed in the Oak Ridge National Laboratory (ORNL) (Kustom, 2000; Mason, 2000; Mason et al., 2006). The design parameters of SNS include 1.4 MW proton beam power at 1.0 GeV beam energy on target, operating at 60 Hz repetition rate, with each pulse duration near one microsecond. Mercury is selected as the target material because it provides high neutron yield per spallation. Since mercury is liquid at target operating temperature, it is not susceptible to the mechanical damage and radiation damage experienced by solid targets, such as tungsten. The mercury circulates through the target where beam energy is deposited to remove heat, with the flow following a pattern as shown in Fig. 1. Neutrons are knocked off the target atoms by high-energy protons and are successively moderated and collimated into various beam lines. Beam line stations are positioned outside the shield block for neutron scattering measurements. Neutron scattering is an advanced probing method to study the arrangement, motion, and interaction of atoms in materials (Mason, 2000). Neutron scattering can provide information that is complementary to other probing methods, such as transmission electron microscopy (TEM) and X-ray diffraction (XRD). Currently, SNS provides the strongest pulsed neutron beams around the world.

Repetitive deposition of the high-power proton beam results in periodic pressure waves of high amplitude propagating inside the mercury target. The pressure wave interacts with the target vessel and causes cavitation damage to the material surface. This damage progresses to create holes in the vessel, and limits the service lifetime of the target vessel. One solution for mitigating the cavitation damage to the target vessel boundary is injection of small helium gas bubbles into mercury to add compressibility and attenuate the periodic pressure pulse and reduce the cavitation damage. Helium bubble injection experiments revealed the difficulty in obtaining the desired gas volume fraction, bubble retention time and bubble size distribution. However, experimental data indicated that cavitation damage was reduced by bubble injection induced pressure wave attenuation (Futakawa et al., 2008; Riemer et al., 2010, 2012). The helium–mercury target needs further study to ascertain the feasibility of helium injection to extend target endurance.

The solubility of helium in mercury is important to predicting the behavior of the proposed helium bubbles in the mercury target. The bubble size evolution and retention time will be dependent on the amount of gas that is dissolved in mercury and the gas absorption and desorption as the bubbles move around the target flow

* Corresponding author. Tel.: +86 551 65591309; fax: +86 551 65591310.
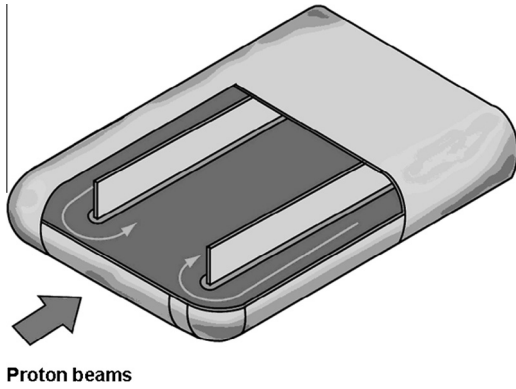*E-mail address:* blu@ipp.ac.cn (B. Lu).

Fig. 1. Cutaway view of SNS target schematic (Riemer et al., 2003).

loop. Gas solubility in liquid metal is also important in fast reactors that use liquid metal as coolant. The production of gas from nuclear fission reactions and its dissolution in the liquid–metal, such as liquid sodium or lead, can significantly affect the flow and heat transfer performance of the reactor (Thormeier, 1970). Theoretical and experimental investigations of inert gas solubility in liquid metals, especially in alkali metals, were performed to understand these effects. Thormeier (1970) first investigated the solubility of helium and argon in liquid sodium experimentally and derived a hard-sphere solubility model that agreed with the experimental results. More recently, Shpil'rain et al. (2000) revisited Thormeier's model and detailed the calculation procedure for inert gas solubility in liquid metal. Based on these models, inert gas solubility in liquid metal (liquid alkali metals, tin, lead and bismuth) have been evaluated with an emphasis on high system temperature, which are relevant to fast reactors using a liquid metal as heat transfer agent (Shpil'rain et al., 2000, 2002a, 2002b, 2007; Skovorod'ko and Mozgovoi, 2010). For inert gas in mercury, results on the solubility of inert gas in mercury were obtained, where very low but rather different solubility values were found (Francis, 2008; Skovorod'ko et al., 2011), indicating more work is needed.

In the current study, the solubility of helium in mercury is evaluated systematically at various temperatures. The calculation accounts for the effect of temperature on mercury density and viscosity. Based on the solubility predictions, bubble radius evolution due to mass diffusion across the bubble wall is studied using a mass-diffusion dominated bubble evolution model to illustrate the behavior of the injected bubbles in mercury.

2. Determination of helium solubility in mercury

The models developed by Thormeier (1970) and Shpil'rain et al. (2000) for a mixture of inert gas and liquid alkali metal rendered relatively good agreement with the experimental data available for those metals. The model is based on the thermodynamic equilibrium between the solvent and the solute, so it is also appropriate for evaluating the solubility of inert gases in mercury and is briefly reviewed below.

The derivation of the model starts with the Fowler–Guggenheim formula (Fowler and Guggenheim, 1949) for the free energy of a binary solution, which is individual nuclear and physical properties, and system temperature and pressure,

$$F = n_1 \left\{ -\Phi_1 - kT \left[\ln(Z_1 v_1) + 1 - \ln\frac{n_1}{n_1 + n_2} \right] \right\}$$
$$+ n_2 \left\{ -\Phi_2 - kT \left[\ln(Z_2 v_2) + 1 - \ln\frac{n_2}{n_1 + n_2} \right] \right\}$$
$$+ \frac{n_1 n_2}{n_1 + n_2} \Delta\Phi_{12} \tag{1}$$

where n_1 and n_2 are the number of moles of the solvent and the solute, respectively; Φ_1, Φ_2, and $\Delta\Phi_{12}$ are the molar binding energy of the two components and the correlation part, respectively; v_1 and v_2 denote the molar volumes of the two substances; and Z_1 and Z_2 are the state functions for the two components. The solubility is usually expressed as the ratio of the solute's number of moles to the total number of moles of the solution,

$$x_2^{(1)} = \frac{n_2}{n_1 + n_2} \tag{2}$$

Thermodynamic equilibrium requires that the chemical potential of gas in gas phase ($\varphi_2^{(2)}$) is equal to the chemical potential of gas in mercury ($\varphi_2^{(1)}$). For the chemical potential of inert gas in mercury, the partial derivative of free energy (F) over the number of moles of the solute (n_2) in the solution is evaluated, and for the chemical potential of gas in its gas phase, it has an explicit form (Shoor and Gubbins, 1969),

$$\varphi_2^{(1)} = \frac{\partial F}{\partial n_2}, \quad \varphi_2^{(2)} = -kT \left[\ln Z_2 - \ln\left(\frac{p_2^{(2)}}{kT}\right) \right] \tag{3}$$

By equating the two chemical potentials and performing some mathematical manipulations, the model renders the following expression for the solubility,

$$x_2^{(1)} = \frac{v_1 p_2^{(2)}}{RT} \exp\left(\frac{-F_{12}}{RT}\right) \tag{4}$$

where $p_2^{(2)}$ is the cover gas pressure above the solution; F_{12} is the free energy of the gas in the solution, which is a function of material properties, system pressures and temperatures and can be evaluated numerically.

To compute the solubility, accurate nuclear and physical properties of helium and mercury are required. Table 1 lists some properties used for the solubility calculation (Barton, 1991; Skovorod'ko et al., 2011). Additionally, mercury's density, dynamic viscosity, thermal expansion coefficient and isothermal compressibility are expressed as a function of temperature. Representative properties of mercury were plotted as a function of temperature (250–550 K) in Fig. 2 (Grosse, 1966; Mehdipour and Boushehri, 1997).

The solubility was evaluated using these properties and 1 atm coverage gas pressure for temperatures ranging from the melting point to the boiling point of mercury. The results of this evaluation are shown in Fig. 3. The solubility was very small and ranged from $\sim$$1.57 \times 10^{-14}$ at 250 K to $\sim$$1.07 \times 10^{-12}$ at 550 K. Helium solubility in mercury increases with temperature as with other liquid metals. The computed results were well fitted with an exponential function, which is also the case for liquid alkali metals (Shpil'rain et al., 2007). The low solubility is expected as mercury is used as the blocking material in ASTM testing standards for solubility measurement of gas in liquid (ASTM International, 2002), in which zero gas solubility in mercury was assumed.

The calculated results were compared with those values published by Skovorod'ko et al. (2011) using the same atomic radii, polarizability, and effective charge for mercury and helium. The results of the models presented here predicted much lower solubility, but it was found through correspondence that the results in Skovorod'ko et al. (2011) were misrepresented. The corrected results are of the same order of magnitude as in the current work.

Table 1
Nuclear and physical properties of inert gases and mercury.

Substance	Atomic mass (amu)	Atomic diameter (10^{-10} m)	Effective charge	Polarizability (10^{-30} m³)
Helium	4.0026	2.70	1.7	0.205
Mercury	200.59	3.19	8.0	10.34