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Solubility of helium in mercury for bubbling technology of the spallation neutron mercury target

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A B S T R A C T

The pitting damage of mercury target container that originates in the pressure wave excited by the proton beam incidence becomes a large problem to reach the high-power neutron source in JSNS and SNS. The lifetime of mercury container is decreased remarkably by the pitting damage. As one of solutions, the pressure wave is mitigated by injecting the helium micro bubbles in mercury. In order to inject the helium micro bubbles into mercury, it is important to understand the characteristic of micro bubbles in mercury. The solubility of mercury–helium system is a key factor to decide bubbling conditions, because the disappearance behavior, i.e. the lifetime of micro bubbles, depends on the solubility. In addition, the bubble generation method is affected by it. Moreover, the experimental data related to the solubility of helium in mercury hardly exist.

In this work, the solubility was obtained experimentally by measuring precisely the pressure drop of the gas that is facing to mercury surface. The pressure drop was attributed to the helium dissolution into mercury. Based on the measured solubility, the lifetime of micro bubbles and the method of the bubble generation is estimated using the solubility data.

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1. Introduction

High-intensity pulsed spallation neutron sources are developed in the world. For the spallation neutron target, a liquid-mercury target system was adapted as taking an advantage of a forced flow for heat removal and high neutron yield. The Japan Spallation Neutron Source (JSNS) has been constructed under the Japan Proton Accelerator Research Complex (J-PARC) project promoted by a joint collaboration between the Japan Atomic Energy Agency (JAEA) and the High Energy Accelerator Research Organization (KEK) [1]. The JSNS aims to produce the world highest intensity of pulsed cold neutron beams and high quality pulse structure for fundamental research in materials and life science [2]. In the JSNS, the high-intensity neutron is generated by high-power proton beam (1 MW, 25 Hz, 3 GeV) injected into the liquid-mercury target.

In developments of the high-power liquid-mercury target, the damage by cavitation erosion was confirmed on the surface of mercury target vessel [3]. This damage occurs as follows. Pressure waves will be generated in the mercury by the abrupt heat deposition at the moment the proton beam injection [4]. After the negative pressure causes cavitation into the mercury, collapse of the cavitation bubbles damages the surface of the vessel wall. This damage reduces the lifetime of the target vessel [5]. Therefore, the mitigation techniques for pressure waves and pitting damage are required to realize the high-power liquid-mercury target. Through our R&D on the mitigation technology, helium micro bubble injection into liquid-mercury becomes one of the prospective candidates from the result of bubble dynamics and experimental observation [6].

Much research is necessary to develop techniques to inject and flow the micro-scale bubbles of the helium in liquid-mercury and methods of generating micro bubbles and the behavior of micro bubbles in mercury, etc. Since fundamental physical constants have not yet been measured in mercury–helium system, it is difficult to predict the characteristics of such bubbles. In particular, the solubility is related to the formation and the life of a micro bubble. The solubility is, therefore, a very important parameter to control and generate helium micro bubbles in mercury. But data related to the measured solubility of mercury–helium hardly exist.

In this paper, we estimate the solubility experimentally and discuss techniques for bubble generating system for mercury targets.

2. Solubility of helium in mercury as application of micro bubble

2.1. Lifetime of micro bubble

Micro bubble research has been done extensively in water–air system. A short lifetime of micro bubble is clarified as one of the features of micro bubble in past research in water–air system. For static state of a single bubble, the lifetime is determined due

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to an expansion or shrinkage itself. It is clear that micro bubbles rapidly disappear in water. This is the effect of micro size based on the scale factor of bubbles, shown in Fig. 1, and is based on the following reasons. In the interface of bubble and liquid, following the Young–Laplace equation can describe a pressure relationship of gas (micro bubble) and liquid:

$$p_g = 2\sigma/R + p_\infty, \tag{1}$$

where p_g is the pressure of gas in bubble, p_∞ is the pressure of liquid, σ is the surface tension of liquid and R is the bubble radius.

Eq. (1) indicates that the pressure of the gas in the micro bubble is higher than liquid pressure always. For example, the pressure difference reaches 0.1 MPa for 50 μm radius in water–air. This differential pressure becomes the driving force, and the gas in micro bubble dissolves into the liquid and disappears in the end. That was experimentally made clear.

The bubble shrinkage speed can be described as follows equation by Takemura [7]

$$\frac{dR}{dt} = \frac{\sigma\rho \cdot \Re T}{R(p_\infty R + \sigma)} \cdot \frac{D}{H}, \tag{2}$$

where ρ is a density of liquid, $\Re$ is gas constant, T is the temperature, D is the diffusion constant, H is the Henry's constant corresponding to solubility x_i.

The shrinkage speed of micro bubble can be estimated with the surface tension, the diffusion constant and the solubility. Although the surface tension of mercury is known, in the mercury–helium system, the diffusion constant and solubility are unknown. When helium micro bubbles are introduced in mercury, they must last a minimum amount of time. In JSNS, it takes about a hundred seconds to circulate mercury around the mercury circuit. If the lifetime of micro bubble is over 100 s, an installation area of micro bubble generator could be placed anywhere in the mercury circulation system. To satisfy that condition, D/H should be over 10^{-18} ($m^2\ s^{-1}$/Pa kg^{-1} mol).

In order to estimate D/H, we carried out to measure the shrinkage speed of a single micro bubble in a mercury–helium system [8]. Since the mercury is an opaque liquid, helium micro bubbles in contact with a transparent wall were observed to estimate the parameter ratio D/H. It was determined that D/H in the mercury–helium system was 7×10^{-20} ($m^2\ s^{-1}$/Pa kg^{-1} mol), and therefore large enough to satisfy our condition.

The relation between the Henry's constant and the solubility can be described as follows:

$$Pi = D \cdot x_i, \tag{3}$$

where Pi is partial pressure of gas. Therefore, $D \cdot x_i$ is 7×10^{-16} ($m^2\ s^{-1}$Pa kg^{-1} mol, 0.1 MPa, 300 K) from Eq. (3) and our measurement results.

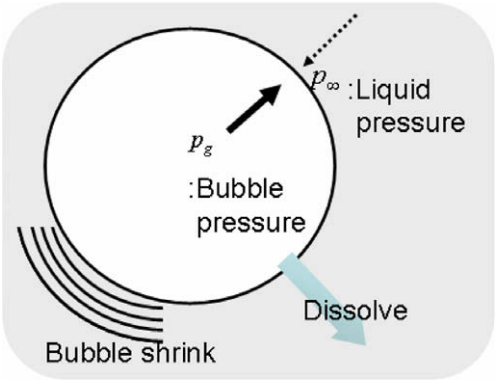

Fig. 1. Shrinkage mechanism of a single micro bubble.

2.2. Generation method of microbubble: pressurization dissolution

In the water–air system, many types of equipment to generate micro bubble have been developed and have been put to practical use. For example, a rotation diffuser type, a porous plate type, a venturi method, a needle pipe and a pressurization dissolution method are micro bubble technology in the water–air system. However, the microbubble methods in a special liquid like mercury are not established yet.

In the above-mentioned, the pressurization dissolution method, shown in Fig. 2, can generate extremely small micro bubbles, 10–40 micro-meter radius in water–air. Furthermore, a size distribution of micro bubbles is also enough small for our purpose. In this method, helium will be dissolved in mercury by pressure compression, and generates the extraction gas as micro bubbles by de-compression. For this method to work, the solubility must be sufficiently high.

We estimated the required void fraction to mitigate the pressure wave in the mercury target. The required void fraction is higher than 0.1% from the calculation result of bubble dynamics [9]. Using the pressurization dissolution method, a simple estimation is performed as follows. In the mercury target of JSNS, de-compression as 0.1 MPa will be reliable. The target temperature is mostly maintained at room temperature. The solubility of mercury–helium system will need as 10^{-6} (mol mercury/mol helium, 0.1 MPa, 300 K), in order to attain the void fraction 0.1% using the pressurization dissolution method. Therefore, a solubility of 10^{-6} (mol mercury/mol helium, 0.1 MPa, 300 K) is required if the pressurization dissolution method is to be feasible.

2.3. Estimation of solubility of helium in mercury

As mentioned above, the data of the solubility in a mercury–helium system almost does not exist experimentally. However, the solubility can be estimated from our data of shrinkage speed which we measured, $D \cdot x_i$ is 7×10^{-16} ($m^2\ s^{-1}$Pa kg^{-1} mol, 0.1 MPa, 300 K). Here, the diffusion constant is estimated theoretically and the solubility will be presumed. The most classic formula to estimate the diffusion constant is the Einstein–Stokes formulation [10]

$$D = \frac{k_B T}{4\pi\mu_B R_A}, \tag{4}$$

where k_B is the Boltzmann constant, μ_B is the viscosity and R_A is the atomic radius.

It was assumed that the surface of the solute particle was complete slip condition. It is proper at a molecule and atom level to assume the slip condition. Eq. (4) adjusts and derives the Stokes rule

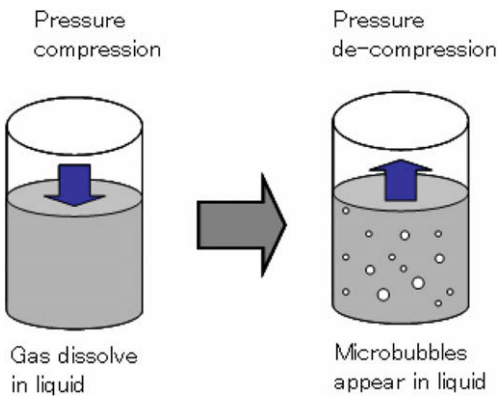

Fig. 2. Microbubble generation with pressurization dissolution.

to the brown particle. Pastermak checked this formation to the self-diffusion and mutual diffusion in liquid metal, which gives the almost right value [11]. To calculate the diffusion constant by Eq. (4), the following value is used; μ_B of mercury is 1.5×10^{-3} Pa-s; R_A is 1.51×10^{-10} m (mercury) and 1.40×10^{-10} m (helium). Then D is calculated to be 5×10^{-9} m^2/s. Here, the Van Del Waals radius is used as an atomic radius of helium. That is taken into account as the particle which behaves as Brown movement. As a calculation result, x_i can be estimated as 1.4×10^{-7} (mol helium/mol mercury, 0.1 MPa, 300 K). From this result, the solubility of mercury–helium is not enough for the pressurization dissolution method.

3. Experimental procedure to measure

In chapter 2, the presumption of solubility from the experimental data and the theoretical formation was done. In this chapter, an experimental measurement of the solubility was described. The predicted solubility is the order of 10^{-7}. It is difficult to observe such a small volume change of helium into mercury solution. Therefore we adapted the method to measure the pressure change instead. The solution volume of helium corresponds to the pressure drop in the helium space. A schematic and photograph of the experimental device are shown in Fig. 3.

A half liter 316SS chamber with polished wall was filled with pure mercury. There are no rubber and resin seals to keep tight for helium gas. In this experimental device, it is possible to measure solubility as low as the order of 1×10^{-8}. The mercury was filled after degassing the chamber by vacuum pumping for 1 day. After degassing, the mercury volume in chamber is calculated by weight measurement with the precision of 1 g. After that, the small evacuated volume above the liquid-mercury is replaced with pressurized helium, 0.1–0.4 MPa. The gas volume is tuned carefully by weight measurement of mercury, because the volume of helium is sensitive to the pressure drop by helium solution into the mercury liquid. The chamber is surrounded by band-heater to control temperature of liquid. During measurement, the temperature is kept by feedback control with thermocouple in mercury. Since the measurement setup is well done, data of pressure and temperature were saved by every 1 min.

Assuming that helium is an ideal gas. Then solubility of helium gas into mercury can be written:

$$x_{\mathrm{He}} \approx \frac{\Delta P V_{\mathrm{He}}/RT}{\rho_{\mathrm{Hg}} V_{\mathrm{Hg}}/M_{\mathrm{Hg}}}, \tag{5}$$

where M_{Hg} is the atomic mass for mercury, ΔP is the pressure change of helium, V_{He} and V_{Hg} are volume of helium and mercury, respectively.

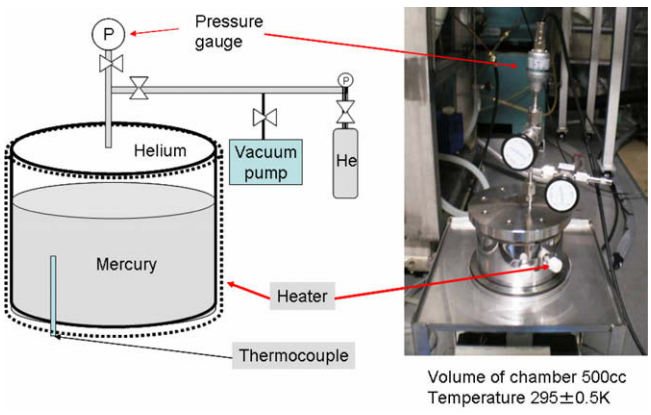

Fig. 3. Setup to measure the solubility in mercury–helium system.

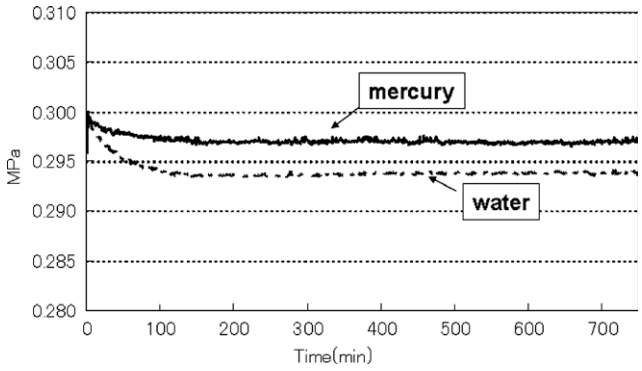

Fig. 4. Pressure as a time function in solubility measurement for mercury–helium and water–helium system.

Before starting to measure the solubility, we carried to observe the leakage for helium as background for pressure change. For 2 weeks, no pressure change was detected. Therefore a lower limit can be placed on the Henry's constant. The smallest observable pressure change with measurement system is 0.5 kPa. This smallest change corresponds to solubility which will be 1.0×10^{-8} (mol Helium/mol Mercury at 0.1 MPa, 300 K).

The pressure trend as a function of time can be shown in Fig. 4. Two trend curves are plotted in Fig. 4. In order to verify the validity of measurement method, the experiment which used water–helium was carried out. The solubility and diffusion constant of the water–helium had been measured experimentally, 7.1×10^{-6} (mol helium/mol water, 0.1 MPa) and 9.2×10^{-9}m^2/s, respectively [12].

The solubility of water–helium obtained from actual measurement is 6.9×10^{-6} (mol helium/mol water, 0.1 MPa). This is very good agreement with the reference data. From this agreement, the solubility estimate by the present method was thought to be reliable. The solubility of mercury–helium system was measured at four different pressure conditions. The results are listed in Table 1. Fig. 5 show that solubility indicates the linear tendency to helium pressure. The solubility of mercury–helium obtained from this trend is 3.8×10^{-7} (mol helium/mol mercury, 0.1 MPa) as a result of this experiment.

4. Discussions

The solubility, which is obtained from the measurement of micro bubble lifetime, is 1.4×10^{-7} (mol helium/mol mercury, 0.1 MPa and 300 K). On the other hand, the solubility, which is appraised with pressure drop measurements, is 3.8×10^{-7} (mol helium/mol mercury, 0.1 MPa). The difference is approximately two times. This difference can be explained as follows. In lifetime measurement, the micro bubbles, which attach to the surface of wall as shown in Fig. 6, are observed in mercury experiment.

The micro bubble shrinks by dissolving into liquid from the surface of micro bubble by the pressure difference between gas and liquid. Therefore, it is thought that the speed of bubble shrinkage is proportional with the surface area. The shape of micro bubble attaching on the wall is shown in Fig. 6 and depends on a contact angle. The surface area of wall bubble approximately has become a

Table 1
Solubility of mercury–helium at different pressure.

Pressure (MPa)	Solubility (mol He/mol Hg, at each pressure)
0.105	3.89E−07
0.172	7.15E−07
0.228	8.89E−07
0.362	1.53E−06

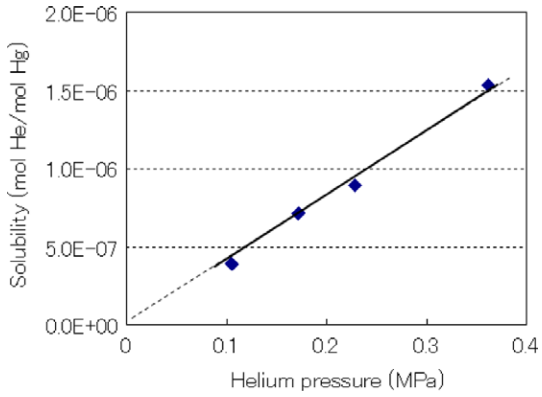

Fig. 5. Solubility of mercury–helium at different pressure.

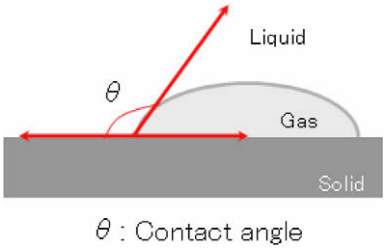

Fig. 6. Shape of micro bubble on wall.

quarter from contact angle, about 130 degrees, of the acrylic to mercury. Assuming the shrinkage speed of micro bubble is proportional to surface area, it is clear that the solubility of wall attaching bubble becomes small due to Eq. (2). Based on interface surface area, 6.8×10^{-7} (mol helium/mol mercury, 0.1 MPa) can be estimated as the value for a bubble attached to the wall. In this case, the solubility is larger than the value obtained from the pressure drop experiment.

Thus, in these two experiments, it is understood that the different results are due to the solubility in the micro bubble form, which is a dynamic influence when the bubble disappears. However, the results are consistent because the solubility indicates on order of 10^{-7} (mol helium/mol mercury, 0.1 MPa).

From experimental results, the solubility of mercury–helium system is less than 10^{-6} (mol helium/mol mercury, 0.1 MPa). For this reason, when the pressurization dissolution method is used as helium bubble generation equipment in mercury, it is calculated that void fraction of micro bubbles is less than 0.1%. Therefore, in order to acquire the mitigation effect of pressure wave as our purpose, it can be concluded that the pressurization dissolution method is inadequate.

However, one-third of the required void fraction can be obtained with the pressurization dissolution method, so the pressurization dissolution method could be useful if it is supplemented by another method.

5. Conclusions

In this paper, the solubility of mercury–helium system was evaluated based on two kinds of experiment results. One is the shrinkage observation of micro bubble, and the other is an observation of the helium pressure drop by dissolution into mercury. The value of solubility obtained from each experiment had twice the difference. However, the difference becomes smaller when the shape effect of micro bubble is taken into consideration. It can be said that it consistent values were obtained from different experiments and evaluation.

It became clear that the required void fraction of 0.1% is not provided using the pressurization dissolution technique from the result of the experimental solubility. Instead, the void fraction would be one order lower than what is required. Therefore, although it can used as an auxiliary method, it is necessary to generate most micro bubbles with other techniques: a rotation diffuser type, a porous plate type, a venturi method or a needle pipe. On this account, future study is necessary for micro bubble generation methods in mercury.

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