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Influence of atomic hydrogen, physical properties, and surface impurity film on droplet ejection from liquid metals interacting with inductively coupled hydrogen plasma

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This paper reports characteristics of droplet ejection from liquid metals interacting with inductively coupled hydrogen plasma. It was observed that the ejection frequency was independent of the flux of atomic hydrogen, whereas it increased with the ion flux. The ejection frequency was dependent on the physical properties of liquid metals, and the transport height of droplets was shortened by the impurity film formation on the liquid metal surface. These results gave us an insight into a method to avoid the transport of droplets to the core plasma, when we apply liquid metals to divertor materials in nuclear fusion reactors.

The design of divertor devices is the most challenging task in the development of nuclear fusion reactors. The energy flux irradiating the divertor material ($\geq$ 10 MW/m^2) is so great that even tungsten is subjected to severe erosion.[1)] The need to replace divertor materials and perform other maintenance in an environment where radioactivation has occurred is also a major burden. An unconventional idea to address the issue is the concept of liquid metal divertors.[2–7)] The use of liquid metals as the divertor materials can improve in the tolerance to high thermal and neutron loads. In addition, if the divertor material is a liquid, it can be introduced into the fusion reactor as a fluid from the outside with keeping the operation. A serious problem that must be considered in the concept of the liquid metal divertor is the production of metal droplets via the interaction with plasma.[8–14)] The transport of metal droplets to the high-temperature core plasma must be avoided strictly since it results in significant power loss. Therefore, the development of a method to avoid the transport of metal droplets to the core plasma is vital for realizing a nuclear fusion reactor with a liquid divertor system.

In a previous work,[15)] we reported the following mechanism for the ejection of droplets from liquid gallium interacting with hydrogen plasma. The droplets are produced by the burst

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of a bubble formed in liquid gallium. The bubble formation is owing to the supersaturated density of dissolved hydrogen. The irradiation of positive ions plays an important role for the supersaturation of the dissolved hydrogen density. This mechanism is supported by the experiment reported by Tamura *et al.*[16] In the present work, we compared the droplet ejections from liquid gallium and liquid tin, which told us the importance of the physical properties on the frequency of the droplet ejection. Another important result was the decrease in the kinetic energy of droplets when the liquid metal surface was covered with an impurity film. In addition, we examined the influence of atomic hydrogen, which had a much higher flux than ion, on the frequency of the droplet ejection.

The experimental apparatus has been described in the previous paper.[15] Briefly, liquid gallium was stored in a quartz container which was installed in a stainless-steel vacuum chamber. We also used liquid tin and a brass container for comparison in the present work. The use of liquid lithium, which would be better as the diverter material because of the small atomic number, was abandoned due to its high chemical reactivity. The temperature of liquid metal was controlled using a heater. The gas in air (nitrogen and oxygen) was dissolved in gallium if it was melted before the installation in the vacuum chamber. Nitrogen and oxygen dissolved in liquid gallium were removed by evacuating the chamber for 10 h at a liquid gallium temperature of 580 K. After the evacuation, we produced inductively coupled hydrogen plasma by applying rf power to a two-turn antenna placed on a quartz plate at the top of the chamber. The positive ion flux toward the liquid metal surface was controlled by the rf power, while the ion energy was controlled by connecting liquid metal to a negatively biased dc power supply. The space above the sample was illuminated by a continuous-wave laser beam at a wavelength of 457 nm. The shape of the laser beam was arranged to be planar using two cylindrical lenses. We observed Mie scattering of the laser light when a droplet passed through the planar laser beam. The scattered laser light was captured using a video camera with an image intensifier. We counted the number of frames with the scattered laser light in the movie and obtained the frequency of the droplet ejection.

In the previous work,[15] we examined the ejection frequency of droplets as a function of the ion flux, and pointed out the importance of the ion irradiation to the droplet ejection. However, the flux of atomic hydrogen was much higher than the ion flux in the experimental condition. In the present work, we examined the influence of the atomic hydrogen flux on the ejection frequency to investigate the influence of the penetration of atomic hydrogen as well as the effect of the simultaneous irradiation of ions and atoms. The atomic hydrogen density was estimated from the difference between the molecular hydrogen densities before and

after the plasma production. The density of molecular hydrogen after the plasma production was estimated based on the absolute optical emission intensity at the Fulcher-α band (the $d^3\Pi_u - a^3\Sigma_g$ transition).[17] The densities of the rovibrational states of $d^3\Pi_u$ were obtained from the optical emission intensities, and they were connected to the rovibrational state densities of the electronic ground state using a collisional-radiative model developed by Sawada and Goto.[18] We substituted the electron density and the electron temperature which were measured using a Lagmuir probe into the model. The rovibrational state densities of the electronic ground state were estimated by the fitting between the experimental and simulation spectra. The optimal fitting was searched by scanning the vibrational and rotational temperatures of the electronic ground state in the model. The atomic hydrogen flux was estimated from the rotational temperature and the decrease in the molecular hydrogen density after the plasma production.

An experimental result is shown in Table I, which compares the ejection frequencies of droplets from liquid gallium at atomic hydrogen fluxes of 1.7×10^{17} and 2.4×10^{18} $cm^{-2}s^{-1}$. These atomic hydrogen fluxes were obtained at pressures of 30 and 50 mTorr, respectively, whereas the ion flux, which was measured as the current passing through liquid gallium, was adjusted at the same value of $(6 - 7)\times10^{14}$ $cm^{-2}s^{-1}$ by controlling the rf power. The liquid gallium temperature of 500 K and the ion energy of 13-15 eV were the same at the two irradiation conditions. As shown in Table I, the ejection frequency of droplets was not affected by the atomic hydrogen flux even though it was three orders of magnitude higher than the ion flux. It has been confirmed that the process that results in the supersaturated density of dissolved hydrogen is the forced transport of positive ions accelerated in the sheath electric field, and atomic hydrogen cannot break the equilibrium determined by the Henrry's law even if hydrogen atoms with a much higher flux are irradiated simultaneously with ions.

Figure 1(a) shows the frequency of the droplet ejection from liquid tin at a temperature of 530 K as a function of the ion flux. We gave up experiments at higher temperatures because of the limitation in the heating system. As shown in the figure, the ejection frequency increased with the ion flux. Figure 1(b) shows the incubation period. The definition of the incubation period is the delay time between the initiation of plasma irradiation and the first ejection of a droplet. The incubation period decreased with the ion flux, as shown in Fig. 1(b). These trends were the same as those observed in the previous experiment using liquid gallium,[15] and they support the scenario that the dissolution of hydrogen beyond the saturation density, which is caused by the forced transport of ions, results in the formation and the burst of a bubble. However, the absolute values of the ejection frequency and the incubation period observed

using liquid tin were different from those observed using liquid gallium. As reported in the previous paper,[15] the ejection frequency from liquid gallium at a temperature of 490 K was 0.05 s^{-1} at an ion flux of 2×10^{14} $cm^{-2}s^{-1}$, which was much lower than that shown in Fig. 1. The incubation period at an ion flux of 2×10^{14} $cm^{-2}s^{-1}$ was 35 s in the case of liquid gallium, which was much longer than that shown in Fig. 1.

The differences in the ejection frequencies and the incubation periods are understood qualitatively by considering the physical properties of liquid gallium and liquid tin summarized in Table II. According to Mazayev *et al.*[19] and Ou *et al.*,[20] the dissolution limits of hydrogen in liquid gallium and liquid tin are 2×10^{16} and 5×10^{15} cm^{-3}, respectively. These dissolution limits have been examined when the samples are irradiated with hydrogen plasmas. Bubbles are formed more quickly in liquid tin with a lower dissolution limit, resulting in the higher ejection frequency of droplets. The surface tension and the viscosity listed in Table II are estimated based on a paper by Gancarz.[21] The lower surface tension of liquid tin than liquid gallium means that the burst of a bubble occurs at a lower pressure inside the bubble, which also results in the higher ejection frequency. The higher viscosity of liquid tin may be the most important difference from liquid gallium. As reported in the previous paper,[15] the incubation period observed in liquid gallium was explained by assuming the uniform distribution of dissolved hydrogen. The uniform distribution may be owing to the stir of liquid gallium due to the convection. The convection is less efficient in liquid tin with a higher viscosity. Hence, the density of dissolved hydrogen is expected to be higher in the region near the liquid tin surface, resulting in the more frequent formation and the burst of bubbles. Based on the differences in the dissolution limits and the incubation periods, it is estimated roughly that the hydrogen-dissolved layer in liquid tin is 2-3 times thinner than that in liquid gallium.

Figure 2 is a picture which shows the trajectories of droplets ejected from liquid gallium. This picture was captured by arranging the planar laser beam to be perpendicular to the liquid gallium surface. The ejections of droplets occurred at random positions on the liquid gallium surface, and a long time was necessary to capture droplets with trajectories along the planar laser beam. Considering the fact that the trajectory of a single droplet was captured clearly by laser Mie scattering, we estimate that the size of the droplet is greater than 0.1 mm. As shown in Fig. 2, droplets were ejected in directions nearly perpendicular to the liquid surface. Note that the direction of the ejection is nearly parallel to the liquid surface if the Kelvin-Helmholtz instability is the mechanism of the droplet ejection.[22] The nearly perpendicular ejection of droplets suggests that it is a phenomenon known as “jet drops” at the burst of a bubble.[23] The driving force that throws the droplet in the “jet drops” model is the fluid dynamic motion of

liquid gallium at the surface. Droplets reached a height of over 5 cm above the liquid surface, as shown in Fig. 2. In the previous experiment, we placed the planar laser beam, which was parallel to the liquid gallium surface, at a distance of 3cm from the liquid gallium surface to detect all droplets and to examine the ejection frequency. If we assume 0.1 mm for the diameter of a droplet, its kinetic energy at the ejection is estimated to be on the order of 10^{-9} J if it reaches at a height of over 5 cm above the liquid surface. Since this energy is much higher than the electrical energy acquired by the droplet in the sheath, the trajectory of the droplet is not affected by the sheath electric field.

When we stored liquid gallium in a brass container, it was observed that the surface of liquid gallium was covered with a thin film. The thickness of the film was unknown, but it was thin enough to keep the fluidity of liquid gallium. An analysis by x-ray fluorescence spectroscopy told us that the film was a compound or an alloy of Ga, Zn, and Cu. We observed the erosion of the brass container after a long experiment, indicating that liquid gallium reacted with brass to form the thin film. When the liquid gallium surface was covered with the thin film, we detected no droplets if the planar laser beam, which was parallel to the liquid surface, was placed at a distance of 3 cm from the surface. However, we detected droplets when the distance between the planar laser beam and the surface was shortened to 1 cm. This indicates that a thin film which covers the liquid gallium surface reduces the kinetic energy of droplets by approximately one-third, resulting in the shortened transport height.

We compared the ejection frequencies of droplets from liquid gallium surfaces with and without the thin film. The experiment was carried out by utilizing an accidental situation where a part of liquid gallium surface was covered with the thin film. The ion flux was 7.1×10^{14} $cm^{-2}s^{-1}$ and the ion energy was 15 eV. As shown in Table III, the ejection frequency from the area with the film was higher than that from the area without the film. Note that the dissolved hydrogen density is determined by the balance between the incoming ion flux and the release of hydrogen gas from liquid gallium. The experimental result shown in Table III suggests that ions with kinetic energies can pass through the film but the film may interrupt the release of hydrogen, resulting in a higher dissolved hydrogen density in the region below the film.

The experimental result with a film on the liquid gallium surface gives us an important insight into the development of a method to avoid the transport of metal droplets to the high-temperature core plasma. Even a thin film that does not prevent the fluidity of liquid metal can reduce the kinetic energy of droplets. The thin films did not block the dissolution of hydrogen due to the ion irradiation, and the ejection frequency of droplets became higher if the surface of liquid gallium was covered with the Ga-Zn-Cu film. However, the transport height

was shortened by the film on the liquid gallium surface. It is considered that the shortened transport height is obtained by the effective increase in the viscosity according to the "jet drops" model.[23] If the transport length of droplets is shorter than the distance between the divertor material and the outermost magnetic surface, the ejection of droplets is not a serious problem when applying liquid metals to divertor materials. The development of a method to cover the liquid metal surface with a thin film may be a key for the implementation of the liquid metal divertor.

In summary, we have reported the influence of atomic hydrogen, the physical properties, and the surface impurity film on the droplet ejection from liquid metals interacting with hydrogen plasma. The ejection frequency of droplets was not affected by the atomic hydrogen flux, indicating that the ion irradiation is only the process that can induce the supersaturated density of dissolved hydrogen in liquid metals. The present experimental results give us two insights into the development of a method to avoid the transport of metal droplets to high-temperature core plasma. One is the importance of the physical properties of the liquid metal. The ejection frequency of droplets is reduced by employing a liquid metal with a high surface tension, a low viscosity, and a high dissolution limit. The other is the usefulness of impurity films on the liquid metal surface. It is not effective for reducing the droplet ejection frequency, but it can shorten the transport length of droplets. The development of a method to cover the liquid metal surface with a thin film is important to realize liquid metal divertor systems for nuclear fusion reactors.

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Table I. Ejection frequencies of droplets from liquid gallium at different fluxes of atomic hydrogen.

Pressure (mTorr)	30	50
Atomic hydrogen flux ($cm^{-2}s^{-1}$)	1.7×10^{17}	2.4×10^{18}
Ion flux ($cm^{-2}s^{-1}$)	7.1×10^{14}	6.0×10^{14}
Ejection frequency (s^{-1})	0.026	0.025

Table II. Physical properties of liquid gallium and liquid tin at 520 K. The dissolution limits of hydrogen are the values examined at plasma irradiation conditions.

	Liquid gallium	Liquid tin
Surface tension (mN/m)[21]	709	561
Viscosity (mPa·s)[21]	0.88	2.24
Dissolution limit (cm^{-3})[19,20]	2×10^{16}	5×10^{15}

Table III. Ejection frequencies of droplets from liquid gallium surface covered with and without a thin film consisting of Ga, Zn, and Cu.

Area	Ejection frequency (s^{-1})
Without film	0.087
With film	0.30

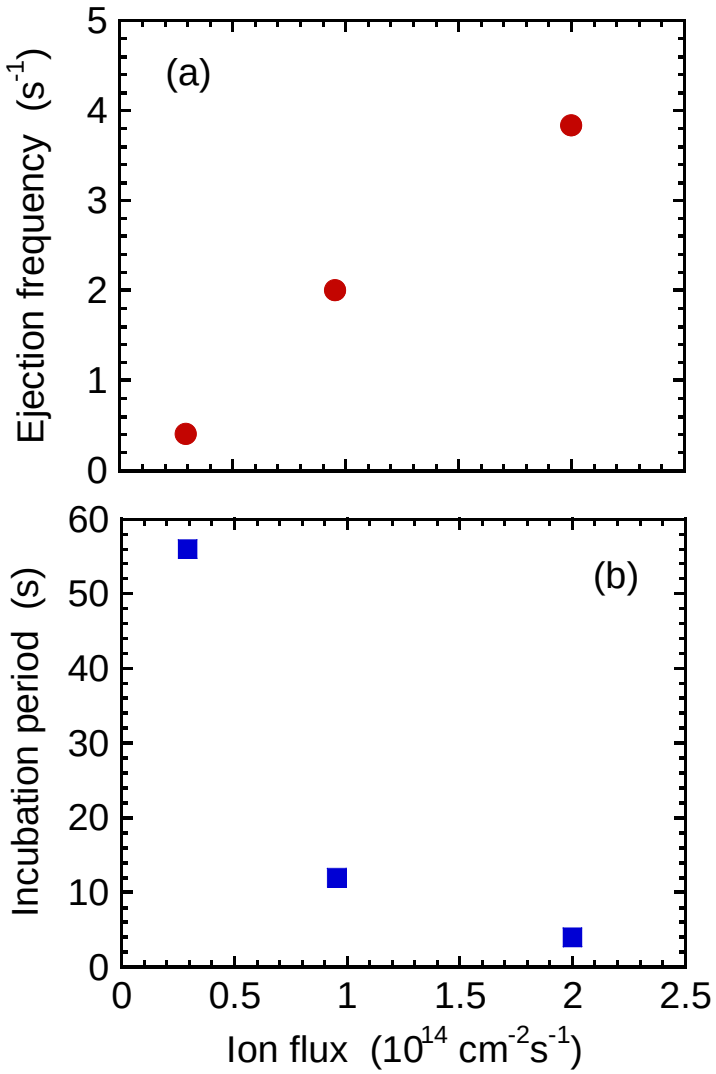

Fig. 1. Frequency of droplet ejection (a) and incubation period (b) as a function of the ion flux. The liquid metal was tin at a temperature of 530 K.

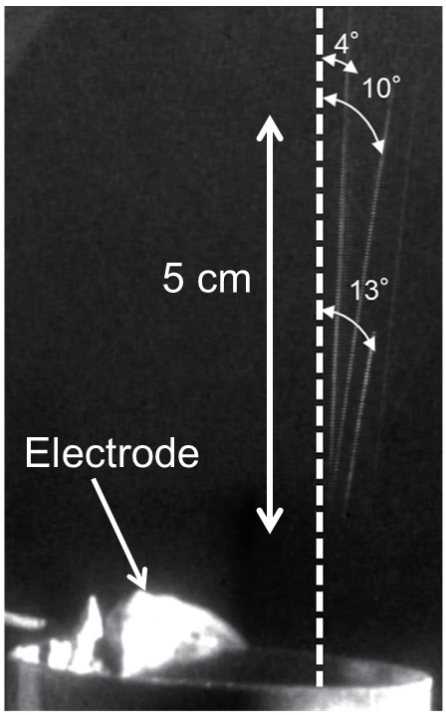

Fig. 2. Trajectories of droplets ejected from liquid gallium without surface impurity film.