Development of a High-Accuracy Capacitance-Type Densimeter for Slush Hydrogen*

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Slush hydrogen is a mixture of solid hydrogen particles and liquid hydrogen, and is being considered as a spaceplane fuel or as a means of transport with hydrogen used as a source of clean energy. In this paper we describe the fabrication of three varieties of capacitance-type densimeters using combinations of a flat plate and two cylinders for the electrodes, and present the subsequent density measurement of slush hydrogen obtained using the freeze-thaw method. The density values measured for slush hydrogen with a solid weight % of up to 30% were generally found to correspond to within $\pm 0.5\%$ of the density values estimated from the latent heat of evaporation of liquid hydrogen. Since the flat-plate/cylinder electrode-type densimeter allowed easy penetration of solid hydrogen particles within the gap between the electrodes, highly accurate density measurements were obtained compared to those simultaneously measured using densimeters with parallel flat plate electrodes. The practical applicability of the new type of slush hydrogen densimeter was thus confirmed.

Key Words: Cryogenics, Liquid Fuel, Solidification, Densimeter, Liquid Hydrogen, Slush Hydrogen

1. Introduction

Since liquid hydrogen has a high specific impulse as fuel for rockets, it is being used in the first- and second-stage engines of the H-II rocket in current use and for the H-II A now under development, although it suffers from the disadvantage of requiring a large fuel tank due to the low density of liquid hydrogen (about 1/14 of that of water). Slush hydrogen is being considered as a spaceplane fuel to decrease vehicle weight during the transition period from one-time-use rockets to completely reusable space shuttles (spaceplanes) in the coming age of mass space transportation.

Slush hydrogen is a mixture of solid hydrogen particles and liquid hydrogen, and can be transported through a pipeline as smoothly as a normal fluid up to a solid weight ratio (solid fraction) of 50%⁽¹⁾.

Compared with liquid hydrogen, the density of slush hydrogen with a solid fraction of 50% increases by about 16%, and its cryogenic capacity by about 18%.

If slush hydrogen is used as a spaceplane fuel, an increase in cryogenic sources for cooling the engines and aerodynamic heating of the fuselage can be expected, along with a lightweight vehicle design facilitated by the decreased fuel tank volume. Thus, development of slush hydrogen technology has become an important issue for the realization of a spaceplane for which disposable boosters (as in conventional multistage rocket propulsion) are not permitted.

Furthermore, in the World Energy Network System (WE-NET) project currently being pursued as part of Ministry of International Trade and Industry's New Sunshine Project, the use of slush hydrogen is being considered as a future fuel of clean energy, due to the potential for improved transport efficiency resulting from the decrease in boil-off gas when hydrogen is carried by ship.

In the case of liquid hydrogen, the fuel load for the rocket can be determined by measuring its volume or level. However, a densimeter with a simple

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configuration and high accuracy is required to allow suitable loading of slush hydrogen, which is characterized by uneven distribution of solid particles.

Since the 1970s, methods for measuring beta- and gamma-ray decay or propagation of ultrasonic waves (including capacitance-type densimetry) have been studied for use in densimeters for slush hydrogen⁽²⁾. However, the beta- and gamma-ray methods cannot generally be used when handling or loading radioactive substances, and the ultrasonic wave method has not yet been practically applied because the effect of solid particles still remains unclear. For the capacitance-type densimeter, as described subsequently, an electrode configuration allowing a large capacitance based on the change in the solid fraction of slush hydrogen is desirable because the specific dielectric constant between liquid and solid hydrogen is small.

Sensors measuring change of capacitance have already been practically applied in the cryogenic field for use in densimeters for Liquid Natural Gas (LNG), level gauges for LNG tanks, and level gauges for liquid hydrogen and oxygen used as fuels for rockets. In the case of densimeters for LNG, for example, the double cylinder type has generally been employed. This is because the substance to be measured can easily penetrate into the gap between the electrodes, and measurement accuracy can therefore be increased by increasing the electrode area and narrowing the distance between the electrodes.

However, for the capacitance-type densimeter used in cases where solid particles coexist in liquid, as seen in slush hydrogen, thorough investigations remain to be performed concerning the high-accuracy electrode configuration, taking into consideration the behavior of solid particles and the required compact design of the densimeter.

In a previous paper⁽³⁾ the authors reported the fabrication of a capacitance-type densimeter using parallel flat plates, and demonstrated the density measurements in slush hydrogen along with the characteristics of the densimeter as observed by changing the solid weight %. The results suggested that measurement accuracy was affected by electrode configuration and electrode-to-electrode distance, as well as by the behavior of solid particles in slush hydrogen.

In this study, considering the thermophysical properties specific to slush hydrogen in which solid particles coexist in liquid at cryogenic temperatures, the problem of the penetration of solid hydrogen particles into the gap between the electrodes has been resolved. Density measurement tests of slush hydrogen have also been conducted with a prototype densimeter, fabricated using a combination of a flat plate

and two cylinders, providing for a simple configuration and high accuracy.

In this paper we describe the density measurement tests in slush hydrogen with a solid weight % (solid fraction) of up to about 30% using three different types of flat-plate- and cylinder-type densimeters featuring different cylinder diameters and different electrode-to-electrode distances, along with a parallel flat-plate-type densimeter. As a result, the characteristics of different types of densimeters with different electrode-to-electrode distances were clarified, and a slush hydrogen densimeter having a high accuracy of within $\pm 0.5\%$ in terms of conversion to density was developed and presented.

Symbols

- C : capacitance, F
- C_0 : sensitivity of capacitance, F
- C_d : reactive capacitance, F
- ΔL : latent heat of evaporation of liquid hydrogen, J
- Q : quantity of heat, J
- ΔQ : total heat leakage, J
- S : area of flat plate
- V : volume, m^3
- ΔV : liquid decrement, m^3
- a : radius of cylindrical electrode
- d : distance between electrodes
- f : solid volume ratio in slush hydrogen ($0 < f < 1$)
- h : enthalpy, J/kg
- l : length of cylindrical electrode
- q : heat leakage into vessel of slush hydrogen, W
- t : time of slush hydrogen production, s
- x : solid fraction of slush hydrogen, wt. %
- ϵ_0 : dielectric constant in vacuum 8.854×10^{-12} , F/m
- ϵ : specific dielectric constant
- ρ : density, kg/m^3

Subscripts

- g : gas
- l : liquid
- s : solid
- sl : slush (liquid + solid)
- t : triple point

2. Densimeter Design for Slush Hydrogen

2.1 Thermophysical properties of slush hydrogen

The liquid hydrogen used for the experiment was para-hydrogen, with a para-hydrogen concentration of not less than 95%. The major thermophysical properties of para-hydrogen are listed in Table 1.

The thermophysical properties were obtained

Table 1 Thermophysical properties of para-hydrogen

	Normal boiling point		Triple point		
	Vapor	Liquid	Vapor	Liquid	Solid
Temperature (K)	20.268		13.803		
Pressure (Torr)	760 (101325 Pa)		52.82 (7042 Pa)		
Density (kg/m ³)	1.338	70.79	0.1256	77.02	86.50
Enthalpy (kJ/kg)	189.3	-256.2	140.3	-308.9	-367.2
Dielectric constant	1.0040	1.230	1.0004	1.252	1.286

from the values published by National Institute of Standards and Technology (formerly National Bureau of Standards) of the USA⁽⁴⁾. Since the change in the specific dielectric constant at the triple point of para-hydrogen ($P_t=52.82$ Torr, $T_t=13.803$ K) represents an increase of only 2.7% in the shift from liquid ($\epsilon_l=1.252$) to solid ($\epsilon_s=1.286$), designing a densimeter configuration which can show a large change of capacitance and allow ease of penetration of solid hydrogen particles into the gap between the electrodes is desirable.

2.2 Working principle of the capacitance-type densimeter

Since capacitance in the capacitance-type densimeter is proportional to the dielectric constant of the material placed in the electrode gap, the relationship between capacitance C and the specific dielectric constant ϵ is expressed by

$$C = C_0\epsilon + C_a \quad (1)$$

C_0 is a constant determined by the size and configuration of the electrode, and corresponds to the sensitivity of the capacitance to the specific dielectric constant. C_a is reactive capacitance generated by the non-electrode structures of the densimeter, a value specific to each densimeter and a constant, regardless of change in the specific dielectric constant.

Therefore, if the values of C_0 and C_a are known, the volume ratio of the solid to the liquid within the electrodes can be calculated from specific dielectric constants (known; see Table 1) of solid and liquid hydrogen at the triple point by measuring the output (capacitance C) from the densimeter placed in slush hydrogen.

If the ratio of volume occupied by solid hydrogen in slush hydrogen is represented by f , the following equation can be formulated between the specific dielectric constant ϵ of slush hydrogen obtained from Eq.(1).

$$\epsilon = (1-f)\epsilon_l + f\epsilon_s \quad (2)$$

When f is solved,

$$f = \frac{\epsilon - \epsilon_l}{\epsilon_s - \epsilon_l} \quad (3)$$

Since the values of ϵ_l and ϵ_s are known and ϵ is the value measured as described above, the value of f and the density ρ_{sl} of slush hydrogen can be calculated

Fig. 1 Two different types of densimeter

from the densities (known) of solid and liquid hydrogen, using the following equation.

$$\rho_{sl} = (1-f)\rho_l + f\rho_s \quad (4)$$

2.3 Study of the electrode configuration and fabrication of densimeters

Prototype flat-plate-/cylinder-type densimeters were fabricated, with a cylinder positioned on either side of the flat plate in order to improve sensitivity. A parallel flat-plate-type densimeter was also fabricated with a pair of flat plates arranged in parallel. Figure 1 shows the schematics of both the types. In both cases, square flat-plate electrodes measuring 50 mm $\times$ 50 mm were used.

The capacitance of the parallel flat-plate-type densimeter is approximated by the following equation, where d is the distance between the conductor flat plates and S is the flat plate area.

$$C = \frac{S\epsilon_0}{d}\epsilon \quad (5)$$

The sensitivity of the densimeter calculated from Eq.(5) is shown by the bold broken line in Fig. 2.

The capacitance of the flat-plate-/cylinder-type densimeter is approximated by the following equation when cylindrical conductors with a radius of a are symmetrically placed on both sides of the flat plate at distance d .

$$C = \frac{4\pi l\epsilon_0}{\ln\{(d + \sqrt{d^2 - a^2})/a\}}\epsilon \quad (6)$$

The sensitivity of the densimeter calculated from Eq.(6) is shown by the solid line in Fig. 2. Types I to III in the figure indicate calculated results for the cylinder diameters ($2a$) of 3, 5, and 8.5 mm, respec-

Fig. 2 Sensitivity of densimeter

tively. From Fig. 2, it is seen that when the electrode distance (d) is constant for the flat-plate-/cylinder-type densimeter, sensitivity increases with increasing cylinder diameter. However, if the cylinder diameter becomes too large, its configuration would qualitatively reject the penetration of solid hydrogen between the flat plate and the cylinders. Compared with the flat-plate-/cylinder-type densimeter, the parallel flat-plate-type densimeter provides greater sensitivity than Type I when the distance (d) between the flat plates is 5 mm. Its sensitivity is nearly equal to that of Type II, provided that solid hydrogen is given free access to the center of the electrode. Although it is predicted that solid hydrogen can penetrate relatively easily into the gap between the electrodes with a 10 mm distance between the flat plates, its sensitivity is nearly equal to that of Type I but inferior to that of Types II & III. Thus, the flat-plate-/cylinder-type densimeter is found to have excellent features with respect to slush hydrogen.

The parallel flat-plate-type densimeter was fabricated by selecting 10 mm as the electrode-to-electrode distance (d) to facilitate ease of penetration of solid hydrogen particles, based on the results of Ref. (3). The design sensitivity calculated from Eq. (5) is 2.21 pF, indicated by $\triangle$ in Fig. 2.

Three different types of flat-plate-/cylinder-type densimeters, Types I ~ III, were fabricated by selecting 3, 5, and 8.5 mm as the cylinder diameters ($2a$) and 4.5, 7.5, and 12.75 mm, respectively, as the distance (d) from the cylinder center to the flat plate. The sensitivities of the three densimeters calculated from Eq. (6) are indicated by $\circ$ in Fig. 2. The main objective here was to understand the effects of the electrode-to-electrode distance (d) and the cylinder diameter ($2a$) on densimetric accuracy by selecting the same design sensitivity of 3.16 pF for all three densimeters.

	2a (mm)	d (mm)	Min.Gap=d-a(mm)
Type I	3.0	4.5	3.0
Type II	5.0	7.5	5.0
Type III	8.5	12.75	8.5

Fig. 3 Flat-plate- and cylinder-type densimeter (three types)

Fig. 4 Flat-plate- and cylinder-type densimeter (type II)

Figure 3 shows the configuration and dimensions of the flat-plate-/cylinder-type densimeters. For reference, the minimum electrode-to-electrode distances ($d-a$) from the cylinder circumference to the flat plate were, respectively, 3, 5, and 8.5 mm for Types I ~ III.

Figure 4 is a photograph showing an external view of the Type II flat-plate-/cylinder-type densimeter.

Oxygen-free copper was used as the material for both the flat plate and the cylinder. Bakelite material was used as the support material for the flat plate and the cylinders, due to insulation protection considerations.

Fig. 5 Test apparatus

3. Test Apparatus and Method

3.1 Test apparatus

Figure 5 shows the schematics of the test apparatus used. The cryostat used to produce slush hydrogen is composed of a double glass Dewar vessel. Its outer side is a vessel containing LN_2 to prevent heat penetration from the outside atmosphere, and its inner side is an LH_2 vessel for production of slush hydrogen. The two vessels use double vacuum-jacketed structures fabricated of pyrex glass, and the internal surface of the vacuum chamber is deposited with silver to prevent heat penetration due to heat radiation from external sources. A slitted observation window without silver deposition was provided to enable observation of slush hydrogen production conditions and measurement of the liquid level.

The freeze-thaw (intermittent evacuation) method was employed for production of slush hydrogen. Solid hydrogen produced over the liquid hydrogen surface during evacuation of the liquid hydrogen was broken to fine pieces and a stirrer (propeller) to homogenize the density distribution during densimetry was also provided at two points (upper and lower) in the Dewar vessel. The stirrer is driven by an electrical motor placed at the top of the Dewar vessel. Other equipment includes a vacuum pump with a discharge rate of 10 000 l/min used to evacuate the liquid hydrogen, and a pressure gauge, a controller and a solenoid valve to periodically repeat evacuation of the vacuum line. The temperature of slush hydrogen was measured using a germanium semiconductor temperature sensor with a measurement accuracy of within ± 0.01 K. An LCR meter was used for the measurement of capacitance in the densimeter.

3.2 Densimeter calibration method in liquid hydrogen

Steady state heat penetration into liquid hydrogen was assessed by measuring the boil-off gas rate from

liquid hydrogen using a gas meter after temperature stabilization at each point, following the filling of the LH_2 cryostat with LH_2 for production of slush hydrogen at atmospheric pressure.

This heat penetration leakage value was used during the density measurement of the slush hydrogen produced. The measured result of heat penetration obtained from a series of test runs was approximately 3 W.

Subsequently, the vacuum pump was operated to evacuate the liquid hydrogen cryostat from atmospheric pressure to the triple point pressure of 53 Torr. The densimeters are calibrated taking into account the fact that the specific dielectric constant of liquid hydrogen changes during the evacuation process. Calibration with high accuracy cannot be performed at room temperature due to changes in the electrode dimensions and the electrode-to-electrode distance during the shift from room temperature to cryogenic temperature.

In the calibration procedure, the specific dielectric constant is obtained from the temperature and pressure of liquid hydrogen while controlling the pressure at a constant value using the vacuum pump inlet valve at a total of seven points, including the normal boiling point, the evacuation process point, and the triple point. Then, the values of C_0 and C_d in Eq. (1) are calculated using the least-squares method.

The densimeters were calibrated for each of the test runs, considering possible changes in the electrode-to-electrode distance.

3.3 Densimetry of slush hydrogen

Solid hydrogen begins forming when liquid hydrogen reaches the triple point. At this point of time, the solid produced over the liquid surface is precipitated in the liquid by periodically operating the vacuum inlet valve (solenoid valve), and the slush hydrogen produced is stirred, to obtain homogenized distribution of solid hydrogen particles. Visual observation showed that the majority of solid particles were about 1 mm in size but some of them were larger flakes with sizes of several millimeters. Measurements using the densimeter were conducted during the slush hydrogen production process. Simultaneously, the liquid level drop was measured from the triple point solidification start point to the density measurement point to calculate the volume decrement (amount of boil off) of liquid hydrogen. The relationship between the level drop and the reduced volume in the LH_2 cryostat was pre-calibrated using pure water. The mean density of slush hydrogen in the cryostat was calculated from the volume decrement of liquid hydrogen. The assumption for this calculation is that the cryogenic energy left over by subtracting heat penetration into

the cryostat from the latent heat of evaporation of LH_2 at the triple point is used for solidification of LH_2 at the triple point. Heat penetration into slush hydrogen generated during stirring was not taken into consideration because the viscosity coefficient of LH_2 (about 3/100 of water at room temperature and atmospheric pressure) and the peripheral velocity of the propeller are negligible. However, heat penetration from the propeller support was measured as a part of heat penetration into the cryostat. The calculation procedure is detailed in section 3.4. The read error for the liquid level drop was a maximum of ± 0.5 mm and the estimated calculation error of the mean density of slush hydrogen in this case was within $\pm 0.03\%$.

An LCR meter was used for the capacitance measurement of the densimeters. Although the dielectric constant of non-polar molecules, such as hydrogen, show no dependency on the frequency of applied voltage, measurement accuracy was confirmed and the frequency was selected by varying the measured frequency using liquid hydrogen at the normal boiling point. The measurements of capacitance carried out by varying the frequencies to 100 kHz, 500 kHz, and 1 MHz with the Type I densimeter immersed in liquid hydrogen verified that the higher frequency reduced the variance in the measured result. It was thus confirmed that the frequency of 1 MHz showed a variance within $\pm 0.01\%$, and it was selected as the frequency of the applied voltage during capacitance measurement.

3.4 Calculation method of slush hydrogen density (solid fraction)

If V_l is used to denote the volume of triple point liquid hydrogen at the onset of slush hydrogen production, and if V_{sl} is used to denote the volume of slush hydrogen after its production, the apparent liquid volume decrement after slush hydrogen production is $V_l - V_{sl}$. However, because liquid and solid components with different densities coexist in slush hydrogen, the actual liquid decrement ΔV is

$$\Delta V = (V_l - V_{sl}) - \frac{\rho_s - \rho_l}{\rho_l} \cdot V_s, \quad (7)$$

where V_s is the volume of solid hydrogen in slush hydrogen.

The heat quantity Q_l contained by the triple point liquid hydrogen at the onset of slush hydrogen production is

$$Q_l = \rho_l \cdot V_l \cdot h_l. \quad (8)$$

The latent heat of evaporation of liquid hydrogen, ΔL , is

$$\Delta L = \rho_l \cdot \Delta V \cdot (h_g - h_l). \quad (9)$$

The total heat penetration into the liquid hydrogen cryostat, ΔQ , is

$$\Delta Q = q \cdot t. \quad (10)$$

The heat quantity contained by the slush hydrogen produced, Q_{sl} , is

$$Q_{sl} = \rho_s \cdot V_s \cdot h_s + \rho_l \cdot h_l (V_{sl} - V_s). \quad (11)$$

From the balance of the heat quantities contained before and after slush hydrogen production,

$$Q_{sl} = Q_l + \Delta Q - \Delta L. \quad (12)$$

Solving for V_s ,

$$V_s = \frac{\rho_l \cdot (V_l - V_{sl}) \cdot (h_l - h_g) + q \cdot t}{\rho_s \cdot (h_s - h_g) - \rho_l \cdot (h_l - h_g)}. \quad (13)$$

The solid mass in the produced slush hydrogen is $\rho_s \cdot V_s$, the liquid mass in the produced slush hydrogen $\rho_l (V_{sl} - V_s)$, and the solid fraction x (wt%) of slush hydrogen is given by

$$x = \frac{\rho_s \cdot V_s}{\rho_s \cdot V_s + \rho_l \cdot (V_{sl} - V_s)} \times 100. \quad (14)$$

4. Test Results and Discussion

4.1 Densimeter calibration results

As described earlier, each of the densimeters was calibrated using liquid hydrogen (LH_2) before densimetry. Figure 6 shows the time lapse during densimetry as the solid fraction increased after production of slush hydrogen (SLH_2) at the triple point reached by liquid hydrogen at the normal boiling point by evacuation. During evacuation, the temperature

Fig. 6 Slush hydrogen production by freeze-thaw method

Fig. 7 Calibration test results in liquid hydrogen

and pressure of the liquid hydrogen were maintained and calibration was performed at this point. Examples of specific dielectric constants and capacitance figures obtained from calibration testing of Types I ~III and the parallel flat-plate-type densimeters are shown in Fig. 7. Sensitivities (C_0) and reactive capacitances (C_d) of the respective densimeters, calculated from the test results using the least-squares method, are listed in the table contained in the figure. Comparison of the sensitivities of the densimeters, obtained from calibration testing prior to density measurement, is given in Fig. 2.

Although Fig. 2 shows that the measured sensitivity of each densimeter nearly corresponds to the data estimated from calculations, the measured results of Types II and III were found to be lower than the calculated results, probably because Eq. (6) assumes that the flat-plate electrode has infinite dimensions whereas they are finite in the actual electrode. Type I showed a measured sensitivity higher than the calculated results, and this was because the electrode-to-electrode distance during the testing was smaller than its design point.

4.2 Density measurement results

Figure 8 shows density measurement results of slush hydrogen with the Type I ~III densimeters and the parallel flat-plate type placed in the cryostat. The four densimeters were set at different elevations in the cryostat due to limitations of space; Type I was set at an elevation of 164 mm from the bottom, Types II and III at 279 mm from the bottom, and the parallel flat-plate type at 149 mm from the bottom. Although the operation of the solenoid valve was stopped to discontinue the production of solid hydrogen during the measurement test, pressure and temperature were maintained at the triple point.

Fig. 8 Comparison between measured and calculated solid fraction (measurements after stirring)

Since most of the resulting solid particles of hydrogen settled down at or near the cryostat bottom, the stirrer was used for homogenization and then stirring was stopped to measure the capacitance of the densimeters.

Figure 8 shows the comparison of the mean solid fraction in the cryostat, calculated from the liquid volume decrement, with the measured solid fraction. The measured results of Types II and III are basically the same, but the measured values tended to exceed the calculated values in the order of Type I, Type II (Type III), and the parallel flat-plate type. The broken lines in the figure show the differences from the measured values ($\pm 0.5\%$) on the basis of the calculated density of slush hydrogen. The solid fraction here represents the solid weight% where the triple point liquid hydrogen is assumed to be 0% and the triple point solid hydrogen is 100%, such that $\pm 0.5\%$ in density of slush hydrogen with a solid fraction of 50% can be converted to a difference of $\pm 4.3\%$ in the solid fraction. The highest value indicated by Type I may be reasoned as follows: because stirring was stopped immediately before measurement, the effect of gravity caused uneven density distribution in the direction of the cryostat height, thereby generating the highest density near the bottom. The parallel flat-plate-type densimeter shows a smaller measured value than the calculated one, although it was set up closer to the bottom than the Type I densimeter. The reason for this is considered to be that the solid particles of hydrogen could not fully penetrate into the gap between the flat-plate electrodes and since Types II and III were placed at the same height, it is inferred that there is no marked difference due to the different distances between the electrodes.

Furthermore, the production of slush hydrogen commenced from the state of triple point liquid hydrogen, and the liquid level change in the cryostat from the commencement of slush hydrogen production to the level of slush hydrogen with a solid fraction of about 30% was measured in this test series. The liquid level in the cryostat changed from a level of about 590 mm to about 500 mm from the bottom, although this change showed slight variation during each test run.

Figure 9 shows density measurement results using the Types I, II, and parallel flat-plate-type densimeters, set at the same height of 168 mm from the bottom.

The measurement method was identical to the one illustrated in Fig. 8.

Types I and II showed a measured solid fraction greater than the calculated value near the solid fraction of 10% (calculated). At solid fractions of 20

Fig. 9 Comparison between measured and calculated solid fraction (measurements after stirring)

Fig. 11 Comparison between measured and calculated solid fraction (measurements during stirring)

Fig. 10 Comparison between measured and calculated solid fraction during slush hydrogen production

~30%, the measured value agreed well with the calculated result within $\pm 0.5\%$.

Since the measured values of Types I and II were nearly the same, the measurement error due to the gap difference between the electrodes is considered to be negligible.

When the solid fraction increased, the parallel flat-plate-type densimeter indicated a measured value smaller than the calculated result. This was probably caused by insufficient penetration of solid particles into the gap between the electrodes.

To facilitate solid particle penetration into the gap between the electrodes of the parallel flat plates, either a decrease of the flat-plate size or an expansion of the gap between the electrodes may be conceivable, but the problem of measurement accuracy would arise due to the corresponding decline in sensitivity.

Figure 10 compares the measured and calculated solid fractions during the time lapsed from the start of slush hydrogen production to the achievement of slush

hydrogen with a solid fraction of about 27% (calculated), using the Type I densimeter. The densimeter was positioned at 168 mm from the bottom.

For Figs. 8 and 9 stirring was stopped during measurements, whereas for Fig. 10 measurement was conducted without stopping stirring. However, stirring was suspended during measurement of the liquid level required for solid fraction calculations to maintain the accuracy of the liquid level measurement. A majority of the measured results show good agreement with the calculated values within $\pm 0.5\%$, and as indicated by Fig. 8, there is variance in density depending on the height of placement of the densimeter in the cryostat, resulting from the effect of gravity during measurement when stirring is stopped.

Figure 11 also shows the results of the density measurement without stopping stirring, as explained for Fig. 10. A total of four test runs were conducted, with the densimeter individually positioned each time. Type I was tested three times, whereas Type II was tested once. All densimeters were positioned at a height of 168 mm from the cryostat bottom.

Most of the measurements agreed with the results calculated from the liquid level decrements within $\pm 0.5\%$, and the maximum difference between the measured and calculated results was within $\pm 0.7\%$.

The cause of variance between the measured and calculated results is probably as follows. Since the freeze-thaw method was used for slush hydrogen production, crushing of solid hydrogen was insufficient. As described earlier, some flakes as large as several millimeters were present, although the majority of solid particles were about 1 mm, leading to various sizes of solid particles being present and distributed within the cryostat or between the electrodes. In addition, the density of slush hydrogen

itself can show local variance, which could also be a possible factor.

5. Conclusions

In order to develop a densimeter for slush hydrogen with high accuracy and simple structure aimed at a compact design, the authors fabricated three different prototype flat-plate/cylinder densimeters, conducted density measurement tests of slush hydrogen having a solid fraction of up to about 30%, and compared their characteristics with the those of a parallel flat-plate-type densimeter simultaneously tested. The following results were obtained.

(1) Densities of slush hydrogen measured with the flat-plate-/cylinder-type densimeters were compared with densities calculated from the latent heat of evaporation of liquid hydrogen and from heat penetration into the cryostat of liquid hydrogen. It was thereby confirmed that most of the measurements agreed with the calculated results within $\pm 0.5\%$. The maximum difference between the measured and calculated results was within $\pm 0.7\%$.

(2) When the measured solid fraction increased (up to 30%), the parallel flat-plate-type densimeter with 10 mm distance between the electrodes showed measured values smaller than the calculated results, and the reason for this could be that the solid particles of slush hydrogen cannot fully penetrate into the gap between the parallel flat plates (50 mm $\times$ 50 mm).

(3) Density measurements using the three

different types of the flat-plate-/cylinder-type densimeters designed with the same sensitivity to capacitance confirmed that there was hardly any influence of the electrode-to-electrode distance on the measured results, and that even in the case of the minimum gap of 3 mm between the electrodes, most of the measured results agreed with the calculated results within $\pm 0.5\%$.

(4) Since the flat-plate-/cylinder-type densimeter has a configuration which can allow easy penetration of solid hydrogen particles into the gap between the electrodes, thereby assuring high-accuracy density measurement results, its practical applicability for slush hydrogen was confirmed.

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