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Title: Micro-bubble generation rate and bubble dissolution rate into water by a simple multi-fluid mixer with orifice and porous tube

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Abstract: A multi-fluids mixer by Sadatomi and Kawahara [1] is described as well as its performance as a micro-bubble generator for several trial products. In the experiments, air micro-bubble generation rate at water depths up to 3.6 m and the dissolution rates of oxygen in air and carbon dioxide into tap water at 20 °C were measured. In the analyses, the micro-bubble generation rate data could be well predicted by Sadatomi et al.'s model [2] by choosing suitable energy loss coefficients needed in the model, and the oxygen dissolution rates in tap water could be well correlated with Kawahara et al.'s model [3]. The detail of the multi-fluid mixer and its practical significances together with a result of experiments and analyses are reported in the present paper.

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Dear Editor of Experimental Thermal and Fluid Science,

We want to submit our following paper to Experimental Thermal and Fluid Science.

Title: Micro-bubble generation rate and bubble dissolution rate into water by a simple multi-fluid mixer with orifice and porous tube

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Thank very much for editing of our paper.

Best regards,

Prof. Michio Sadatomi

## Highlights

> The present author's multi-fluids mixer is described together with its performance as a micro-bubble generator. > Micro-bubble generation rate at water depths up to 3.6 m and the dissolution rates of oxygen in air and carbon dioxide in tap water were measured. > The micro-bubble generation rate data could be well predicted by Sadatomi et al.'s model by choosing suitable energy loss coefficients needed in the model, and the dissolution rates in tap water could be well correlated with Kawahara et al.'s model.

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2 Micro-bubble generation rate and bubble dissolution rate into water by a simple multi-fluid  
3 mixer with orifice and porous tube  
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## ABSTRACT

A multi-fluids mixer by Sadatomi and Kawahara [1] is described as well as its performance as a micro-bubble generator for several trial products. In the experiments, air micro-bubble generation rate at water depths up to 3.6 m and the dissolution rates of oxygen in air and carbon dioxide into tap water at 20 °C were measured. In the analyses, the micro-bubble generation rate data could be well predicted by Sadatomi et al.'s model [2] by choosing suitable energy loss coefficients needed in the model, and the oxygen dissolution rates in tap water could be well correlated with Kawahara et al.'s model [3]. The detail of the multi-fluid mixer and its practical significances together with a result of experiments and analyses are reported in the present paper.

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## 1. Introduction

Micro-bubbles are tiny bubbles, less than a few hundred micrometers in diameter, and have several characteristics, such as high dissolubility in water around them. The most famous application of them is enriching oxygen into water in fisheries of oysters, pearl oysters and so on. Ohnari [4-6] reported that the enrichment promotes the oxygen consumption by the oysters etc. and their blood circulation and metabolism, resulting in the speed-up of their growth. Other applications of the micro-bubbles in industries and several micro-bubble generation methods are described in some books, say by Ueyama and Miyamoto [7].

Sadatomi [8, 9] invented a micro-bubble generator (MBG for short) with a spherical body in a flowing liquid tube and with a lot of drilled small holes on the tube for gas suction, in which micro-bubbles could be generated by supplying liquid alone because gas was automatically sucked by a negative pressure arisen behind the body. After that, Sadatomi et al. [2] proposed a model which can predict well the air micro-bubble generation rate by the MBG placed at any water depth, and Kawahara et al. [3] proposed a model which can predict well the dissolution rate of oxygen in air micro-bubbles into water and seawater. However, the MBG has two defects of (a) the difficulty of fixing the spherical body especially in smaller generator and (b) the troublesome drilling of a lot of small holes.

Recently, in order to overcome the above defects, Sadatomi and Kawahara [1] invented a new device with an orifice and a porous pipe instead of the spherical body and the small drilled holes. The new device is called a multi-fluids mixer in our laboratory because of multifunctional, which can generate (a) micro-bubbles by supplying liquid and sucking gas, (b) mists (i.e., tiny liquid droplets) by supplying gas and sucking liquid, and emulsion of immiscible liquids by supplying one of the liquids.

In the present paper, the structure of the multi-fluids mixer and its performance as a MBG are described for several trial products. In the experiments, three kinds of test were conducted: (a) hydraulic performance test of the present MBG, (b) bubble diameter measurement and (c) micro-bubbles dissolution performance test. In (a), air micro-bubble generation rate was measured at water depths up to 3.6 m by changing water supply rate to the MBG systematically. In (b) and (c), bubble diameter and the dissolution rate of oxygen into water through air micro-bubbles in 1.2 m deep water tank at 20 °C and at atmospheric pressure were measured by changing both water supply rate and air suction rate systematically. In (c), the dissolution rate of carbon dioxide into water through carbon dioxide micro-bubbles was also measured. In the analyses, Sadatomi et al.'s model [2] is tested against the present micro-bubble generation rate data by choosing suitable energy loss coefficients needed in the model and Kawahara et al.'s model [3] is tested against the present dissolution rate data of oxygen in air bubbles. A result of such experiments and analyses together with the detail of the multi-fluid mixer and its practical significances are reported in the present paper.

## Nomenclature

$A_H$  total area of gas suction hole in porous pipe ( $m^2$ )  
 $C$  concentration ( $kg\ m^{-3}$ )

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3	$D_L$ liquid-phase molecular diffusion coefficient ( $\text{m}^2 \text{s}^{-1}$ )
4	$d$ diameter of MBG pipe (m)
5	$d_{BM}$ mean bubble diameter (m)
6	$d_{BS}$ Sauter mean bubble diameter (m)
7	$d_G$ diameter of gas suction hole in porous pipe (m)
8	$d_o$ orifice diameter (m)
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10	$E_A$ ratio of oxygen dissolved into water to that supplied (dimensionless)
11	$E_O$ Eotvos number (dimensionless)
12	$f_C$ correction factor (dimensionless)
13	$H$ water depth (m)
14	$h_G$ thickness of porous pipe (m)
15	$h_p$ height of bubble diameter measurement (m)
16	$K_{La}$ volumetric mass transfer coefficient ( $\text{s}^{-1}$ )
17	$L_L$ water power (W)
18	$l$ length of porous pipe (m)
19	
20	$N_C$ oxygen mass transfer rate ( $\text{kg s}^{-1}$ )
21	$N$ number (dimensionless)
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23	$p$ gauge pressure (Pa)
24	$Q$ volume flow rate ( $\text{m}^3 \text{s}^{-1}$ )
25	$T_L$ water temperature ( $^{\circ}\text{C}$ )
26	
27	$t$ time (s)
28	$u_B$ bubble rise velocity ( $\text{m s}^{-1}$ )
29	$V$ volume ( $\text{m}^3$ )
30	$v$ mean velocity ( $\text{m s}^{-1}$ )
31	$W_{O_2}$ mass flow rate of oxygen supplied to water ( $\text{kg s}^{-1}$ )
32	

### ***Greek symbols***

35	$\beta$ area ratio of orifice to MBG pipe (dimensionless)
36	$\rho$ density ( $\text{kg m}^{-3}$ )
37	
38	$\zeta$ energy loss coefficient (dimensionless)
39	

### ***Subscripts***

41	$E$ section far downstream from the exit
42	$G$ gas
43	$H$ homogeneous
44	$L$ liquid
45	$S$ saturation
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47	1 inlet section of MBG
48	2 contraction section of MBG
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## **2. Experiment**

### ***2.1. Micro-bubble generator***

Fig. 1 shows the orifice type MBG [1] newly developed for the present experiment. The generator has an orifice in a flowing water tube. When pressurized water is introduced into the generator, the water velocity through the orifice becomes several times of that at the generator exit, thus from the energy conservation principle the

1  
2 pressure at a little downstream of the orifice becomes negative. With the aid of the  
3 negative pressure, air is automatically sucked through a porous pipe embedded in the  
4 pipe, and the air sucked is broken into a huge number of micro-bubbles by a high shear  
5 water flow with strong turbulence. Thus the generator can discharge a water jet with  
6 micro-bubbles from the exit.  
7

8 Table 1 lists the specification of the orifice type MBG tested. The first three, named  
9 LP-8.8 to LP-14.6, are large types with the same stainless steel punched porous pipe  
10 except for the orifice diameter,  $d_o$ , being 8.8, 12.5 and 14.6 mm. The diameter of the  
11 circular tube was 22.0 mm; the area ratio of the orifice to the tube,  $\beta$ , was changed from  
12 0.16 to 0.44 in order to study the effects of the area ratio; the length and the thickness of  
13 the porous pipe were  $l = 8$  mm and  $h_G = 0.15$  mm; the diameter of each punched holes  
14 was  $d_G = 300$   $\mu\text{m}$ , and the total area of the holes was  $A_H = 152.7$   $\text{mm}^2$ . The last three,  
15 SF-4.0 to LF-12.5, were geometrically similar to LP-12.5, but in order to study the  
16 effects of the MBG size the size of SF-4.0 and MF- 8.4 were around 1/3 and 2/3 of  
17 LF-12.5. In addition, SF-4.0 to LF-12.5 had the polyolefin (polypropylene  
18 polyethylene) fiber porous pipe with the porosity of  $d_G = 25$   $\mu\text{m}$ . The thickness and the  
19 length of the fiber porous were  $h_G = 1.5$  mm and  $l = 3$  to 8 mm depending upon the pipe  
20 size.  
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## 25 2.2. Test apparatus and measurement systems

26 Three kinds of test were conducted: (a) hydraulic performance test, (b) bubble  
27 diameter measurement test and (c) micro-bubbles dissolution performance test. Fig. 2  
28 shows the present test apparatus and measurement systems. Two water tanks were  
29 used as the test water tank: a small transparent acrylic resin water tank with 2.0 m in  
30 height and 0.30 m in diameter, and a large opaque poly-vinyl-chloride water tank with  
31 4.0 m in height and 0.489 m in diameter. Water was circulated with a centrifugal  
32 pump from the bottom of the tank to the MBG in the tank via a flow control valve and a  
33 calibrated magnetic flow meter for the measurement of water volume flow rate,  $Q_L$ .  
34 The air suction rate into the MBG,  $Q_G$ , was measured with a calibrated mass flow meter.  
35 The uncertainties in the measurements of  $Q_L$  and  $Q_G$  are about 1 % and about 3 %,  
36 respectively.  
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## 41 2.3. Hydraulic performance test

42 In order to familiarize the present MBG to various application fields, the generation  
43 rate of the micro-bubbles has to be predictable, depending upon the water depth of  
44 MBG placed and water supply rate to the MBG. In addition, a pumping power to  
45 supply water to the MBG must be predictable. So, the hydraulic performance test was  
46 conducted to obtain experimental data necessary to validate the performance prediction  
47 model [2]. In the test, in order to study the effects of water depth of the MBG in the  
48 water tank,  $H$ ,  $H$  was changed as 0.4, 0.8, 1.2 m in the small water tank and 2.4 and 3.6  
49 m in the large water tank, and the needle valve in the air suction line was full opened.  
50 The water supply rate to the MBG,  $Q_L$ , was varied up to 106 l/min depending upon both  
51 the MBG size and the MBG depth from the water surface, while the air suction rate,  $Q_G$ ,  
52 was changed up to 21 l/min depending upon  $Q_L$ . In addition to the measurements of  
53  $Q_L$  and  $Q_G$  mentioned in 2.2, the air pressure,  $p_G$ , and the water pressure,  $p_L$ , at the MBG  
54 inlet were also measured with two different pressure transducers calibrated within the  
55 uncertainties of 50 Pa. As shown in Fig. 2, the output signals from the above  
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2 mentioned flow rate and pressure sensors were fed to a personal computer via an A/D  
3 converter to determine the respective time-averaged values. The water power needed  
4 to generate micro-bubbles by the MBG,  $L_L$ , was calculated by substituting the measured  
5  $p_L$  and  $Q_L$ , and the mean water velocity at the MBG inlet,  $v_{L1}$ , into Eq. (1).  
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$$L_L = (p_L + \rho_L v_{L1}^2 / 2) Q_L \quad (1)$$

#### 10 2.4. Bubble diameter measurement

11 The bubble diameter is known to affect the rising velocity and the dissolution  
12 performance of the bubble. So, filling water to  $H = 1.2$  m in the small transparent  
13 tanked, we measured bubble diameter with a high-speed video camera and an image  
14 processing system. Bubble images against a back flash light at  $h_p = 0.6$  m were taken  
15 with the video camera at a shutter speed of 1/30,000 s. 100- and 30-power microscope  
16 lenses, corresponding to 2.32 mm and 7.73 mm in frame height, were attached to the  
17 camera for measuring smaller and larger bubbles than 0.1 mm. The minimum  
18 measurable bubble diameter was 0.01 mm, thus the uncertainty of the bubble diameter  
19 measurement is 0.01 mm. The number of pictures taken was so determined that the  
20 total frame area captured becomes identical between the smaller and the larger frames.  
21 In the bubble diameter measurement, although most of the bubbles were spherical the  
22 major and the minor axes of the bubble on pictures were measured for distorted bubbles  
23 larger than 1.5 mm, and averaged value of the two axes was taken as the bubble  
24 diameter. In order to obtain reliable bubble diameter distribution, more than 350  
25 bubble diameters were measured, and the mean and the Sauter mean bubble diameters  
26 were calculated by  
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$$d_{BM} = \frac{\sum n_i d_{Bi}}{\sum n_i}, \quad d_{BS} = 6 \times \frac{\sum (n_i \pi d_{Bi}^3 / 6)}{\sum n_i \pi d_{Bi}^2} \quad (2)$$

32 Here,  $n_i$  is the number of bubbles classified in a bubble diameter range of  $d_{Bi}$ .  
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#### 39 2.5. Micro-bubble dissolution performance test

40 Enrichment of oxygen into water through air micro-bubbles is very important in  
41 fisheries, sewage treatment system etc. So, the micro-bubble dissolution test was  
42 conducted with the small water tank by filling 0.13 m<sup>3</sup> tap water, corresponding to  $H =$   
43 1.2 m. The test liquid was tap water, while the test gas was air from atmosphere or the  
44 carbon dioxide from a CO<sub>2</sub> cylinder. The water supply rate to the MBG,  $Q_L$ , was  
45 limited to 72 l/min, and air or carbon dioxide suction rate to 4 l/min. The water  
46 temperature,  $T_L$ , was kept at  $20 \pm 0.1$  °C using a cooling system in the experiment,  
47 because it affects bubble dissolution characteristics. Salinity effects were not studied  
48 in the present study because the study will be done as a future work.  
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51 Most tests were conducted with air as the test gas. Firstly, nitrogen gas was blown  
52 into water in order to reduce the oxygen dissolved in water,  $DO$ , to about 4 mg/l  
53 because water from our laboratory source always showed a high  $DO$  value at the  
54 beginning and the time to reduce it to zero is too long. Secondary, air was blown as  
55 micro-bubbles through MBG at assigned flow rates of air and water, and the time  
56 variation of oxygen concentration in water,  $C$ , was measured with a DO meter  
57 (OE-270AA and MM-60, DKK-TOA Co.) within the uncertainties of 0.02 mg/l. The  
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2 volumetric mass transfer coefficient,  $K_La$ , was determined from the time variation data  
3 by fitting Akita and Yoshida's equation [10]:  
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$$5 \quad dC/dt = K_La(C_S - C) \quad (3)$$

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8 Here,  $C$  and  $C_S$  are the concentration of dissolved oxygen at a time  $t$  and at saturation,  
9 **respectively**. In addition, in order to confirm a well-mixed condition, we measured  $C$   
10 at the bottom, the middle and the top of the test tank, and confirmed  $C$  values at the  
11 three positions to be similar within the accuracy of DO measurement. **As a result,  $K_La$**   
12 **could be determined within the uncertainty of about 10 %**.  
13

14 Oxygen absorption efficiency,  $E_A$ , the ratio of the oxygen dissolved in water to that  
15 supplied to, is calculated from:  
16

$$17 \quad E_A = \frac{N_C}{W_{O_2}} \times 100 \% \quad (4)$$

18  
19 Here,  $W_{O_2}$  is the mass flow rate of oxygen supplied to water as air bubbles, being 23%  
20 of air mass flow rate supplied.  $N_C$  is the oxygen mass transfer rate given by  
21

$$22 \quad N_C = K_LaV_L C_S \quad (5)$$

23  
24 Here,  $V_L$  is the volume of water in the test tank.  
25

26  
27 Similar measurements were also conducted for carbon dioxide,  $CO_2$ , micro-bubbles  
28 instead of air micro-bubbles. **The time variation in  $CO_2$  concentration in water was**  
29 **measured with a  $CO_2$  meter (Ti-9004, Toko Kagaku Co.) within the uncertainty of 2.2**  
30 **mg/l**.  $K_La$  was determined by substituting the time variation data on the concentration  
31 of carbon dioxide into Eq. (3). **As a result,  $K_La$  could be determined within the**  
32 **uncertainty of about 10 %**. Furthermore,  $E_A$  was determined from Eqs. (4) and (5),  
33 **though  $W_{O_2}$  and  $C_S$  must be replaced by the values for carbon dioxide**. In addition,  
34  $K_La$  so determine for carbon dioxide micro-bubbles was compared with that for air  
35 micro-bubbles.  
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### 41 **3. Results and discussion**

#### 42 **3.1. Hydraulic performance**

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44 Figs. 3 (a) to (c) show typical results of hydraulic performance tests for the large  
45 size, punched porous type MBG of LP-8.8, LP-12.5 and LP-14.6, each different in  
46 orifice diameter but the other sizes were the same. **The MBG depth in the water tank**  
47 **in Fig. 2 was set at  $H = 0.4$  to 3.6 m**, and the needle valve in the air suction line was full  
48 opened. Unfortunately, however, LP-8.8 did not work at  $H = 2.4$  and 3.6 m **because**  
49 **the water pressure required at the MGB inlet was higher than that given by the present**  
50 **water circulation pump**.  
51

52  
53 Fig. 3 (a) shows the data of air suction rate,  $Q_G$ , against the water supply rate,  $Q_L$ .  
54 The data points are labeled with different symbols according to both the MGB type and  
55 the water depth,  $H$ . In addition, data point in each MBG type at  $H = 0.4$  m is  
56 connected each other with line segments, to facilitate the understanding of the trend of  
57 data. With increasing of  $Q_L$  at a fixed  $H$ , the water gauge pressure in the air suction  
58 section downstream of the orifice decreases from positive to negative. After that,  $Q_G$   
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2 increases steeply with increasing of  $Q_L$ . In addition,  $Q_G$  increases with decreasing of  $H$   
3 because the water pressure at the MBG exit decreases and thus the water pressure in the  
4 air suction section decreases. Among these three types, LP-8.8 gives the largest  $Q_G$  at  
5 a fixed  $Q_L$  because the water pressure in the air suction section becomes the lowest at a  
6 fixed  $Q_L$ .

7  
8 Fig. 3 (b) shows the data of gauge pressure at the MGB water inlet,  $p_L$ , against the  
9 water supply rate,  $Q_L$ .  $p_L$  in each MBG is almost proportional to  $Q_L^2$ , and  $p_L$  increases  
10 with decreasing of the orifice diameter, because the pressure drop through the MBG  
11 increases with decreasing of the orifice diameter at a fixed  $Q_L$ .

12  
13 Fig. 3 (c) shows  $Q_G$  data against the water power,  $L_L$ , calculated from Eq. (1). The  
14 line tangent to the data through the origin is steeper the more efficient because the ratio  
15 of micro-bubble generation rate to the power required becomes high. Therefore,  
16 LP-14.6 and LP-12.5 is superior to LP-8.8 at  $H < 1.2$  m, and the maximum attainable  
17  $Q_G$  was higher in LP-12.5 than LP-14.6. In addition, the mean and the Sauter mean  
18 bubble diameters,  $d_{BM}$  and  $d_{BS}$ , measured for LP-12.5 under several  $Q_L$  and  $Q_G$   
19 conditions were 1/3 to 1/2 of those for LP-14.6. Thus, LP-12.5 is superior to LP-14.6  
20 at  $H < 1.2$  m. At  $H > 2.4$  m, on the other side, LP-14.6 is superior to LP-12.5.  
21 However, our previous study [11] demonstrated that for the use of MBG in deep water  
22 the supply of pressurized air to the MBG is more efficient than the suction of air by the  
23 increment of water supply rate. This means that the superiority in deep water of  $H >$   
24  $2.4$  m is not important. Thus, LP-12.5, being the diameter ratio of orifice to MGB pipe  
25 to be 0.57, is recommended as a whole.

26  
27 Figs. 4 (a) and (b) show typical results of hydraulic performance tests for the fiber  
28 porous type MBG of LF-12.5, MF-8.4 and SF-4.0, each different in size but the  
29 proportion of orifice diameter to pipe diameter, etc. was the same as that for LP-12.5.  
30 Thus, the size effects of the MBG can be studied by comparing these three.

31  
32 Fig. 4 (a) shows  $Q_G$  data against  $Q_L$ . The trend of  $Q_G$  against  $Q_L$  and  $H$  is similar  
33 to Fig. 3 (a). It is mysterious that the maximum  $Q_G$  for LF-12.5 and MF-8.4 is nearly  
34 the same and limited to 15 l/min at  $H = 0.4$  m. The reason is probably due to a partial  
35 water immersion in fiber porous especially for LF-12.5.

36  
37 Fig. 4 (b) compares  $Q_G$  data against the water power,  $L_L$  at  $H = 1.2$  m. In order to  
38 know the effects of the number of the MBG placed in the same water depth, the data for  
39 double uses of SF-4.0 and MF-8.4 are also plotted and connected with broken line  
40 segments. It is seen that the double use of SF-4.0 is efficient if the demand of  $Q_G$  is  
41 less than 9 l/min at  $H = 1.2$  m, and the double use of MF-8.4 is efficient if the demand  
42 of  $Q_G$  is between 9 l/min and ca. 17 l/min.

43  
44 Fig. 5 compares  $Q_G$  data between LP-12.5 and LF-12.5 (thickened symbols), being  
45 different in porous material. Though the diameter of air suction hole for LP-12.5 is  
46 300  $\mu\text{m}$  and that for LF-12.5 is 25  $\mu\text{m}$ , the difference of  $Q_G$  between them is small,  
47 because the number of the hole is much more for LF-12.5 than LP-12.5.

### 51 52 3.2. Bubble diameter

53  
54 Figs.6 (a) and (b) show typical bubble diameter distribution data for LP-12.5 and  
55 LF-12.5, being different in porous material, at  $Q_L = 67$  and 66 l/min and at  $H = 1.2$  m.  
56 In each figure, the data at  $Q_G = 1.0$  l/min and 4.0 l/min are compared each other, and the  
57 mean and the Sauter mean bubble diameter data, and the total number of bubbles  
58 measured were also shown. In the bubble diameter measurement test and bubble  
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1  
2 dissolution test, in order to control the air suction rate,  $Q_G$ , at a prescribed value, the  
3 needle valve in air line in Fig. 2 was partially closed. The diameter of each bubble was  
4 classified from 0.01 – 0.05 to 3.0 – 4.0 mm. Though the bubbles smaller than 0.01  
5 mm could not be measured in the present experiment, such an extremely smaller bubble  
6 may exist. Excluding smaller bubbles than 0.01 mm, more than 70 % bubbles are  
7 smaller than 0.1 mm. At  $Q_G = 1.0$  l/min, the mean bubble diameter and the Sauter  
8 mean bubble diameter are respectively around 0.12 mm and 0.63 mm, and larger  
9 bubbles than 2.0 mm did not exist, independent of porous materials. At  $Q_G = 4.0$  l/min,  
10 however, 3.0 – 4.0 mm bubbles existed, and caused the enlargement of the Sauter mean  
11 bubble diameter. Similar trend of data was obtained for other MBG types. Thus, in  
12 order to generate micro-bubbles alone,  $Q_G$  must be lower than 1.0 l/min in the present  
13 MBG types. However, even at  $Q_G = 4.0$  l/min, the present MBG are usable as a  
14 micro-bubble generator because most bubbles are smaller than 0.1 mm.

15  
16 Fig. 6 (c) shows the effects of water supply rate,  $Q_L$ , on the Sauter mean bubble  
17 diameter data at  $Q_G = 1.0$  l/min and 4.0 l/min. As anticipated from Fig.3 (a) and Fig. 4  
18 (a),  $Q_L$  for each MBG type could not be changed in a wide range at  $H = 1.2$  m. So, the  
19 mean water velocity at orifice,  $v_{L2} (= 4 Q_L / (\pi d_o^2))$ , is taken as the abscissa. With  
20 increasing of  $v_{L2}$ , the Sauter mean bubble diameter,  $d_{BS}$ , decreases, and over about  $v_{L2} =$   
21 9 m/s it becomes a similar value independent of  $v_{L2}$  and MBG type but dependent on  $Q_G$ .  
22 Thus, in order to generate micro-bubbles alone by the present MBG,  $v_{L2}$  should be  
23 higher than 10 m/s.  
24  
25  
26  
27  
28

### 29 3.3. Bubble dissolution performance

30 Figs. 7 (a) and (b) show typical variation of oxygen and carbon dioxide  
31 concentrations in tap water after bubbling of air and carbon dioxide, respectively.  
32

33 In the air bubbling case, in order to reduce the initial oxygen concentration to about  
34 4 mg/l, the pre-bubbling of nitrogen gas was conducted at first. Then, air bubbling was  
35 conducted, and the concentration goes up gradually to the saturation value of  $C_S = 8.84$   
36 mg/l at 20 °C, and the arrival time to the saturation is about 10 minute at  $Q_G = 4.0$  l/min  
37 and about 35 minute at  $Q_G = 1.0$  l/min. By fitting these data to Eq. (3), we obtained  
38 the volumetric mass transfer coefficient,  $K_{La}$ . In addition, we obtained the oxygen  
39 absorption efficiency,  $E_A$ , by substituting  $K_{La}$  data etc. into Eqs. (4) and (5).  
40

41 In carbon dioxide bubbling case, pre-bubbling of any gas was not needed since the  
42 initial  $CO_2$  concentration is near to zero. Since the saturation concentration of carbon  
43 dioxide is  $C_S = 1724$  mg/l, being much higher than that of oxygen, the increase of the  
44 concentration is very slow, and the arrival time to the saturation value could not be  
45 detected. In addition,  $K_{La}$  for carbon dioxide case is very low and about one-seventh  
46 to one-eleventh of that for oxygen case.  
47

48 Figs. 8 (a) and (b) show  $K_{La}$  data against  $Q_G$  and  $L_L$ , respectively. Data are labeled  
49 according to the MGB type, water supply rate to the MGB and dissolved gas (open  
50 symbol for oxygen, darkened symbol for carbon dioxide). As is noticed from Fig. 8  
51 (a),  $K_{La}$  for the oxygen case (open symbols) increases linearly with  $Q_G$  irrespective of  
52 the orifice diameter, the porous pipe, the MBG size and the water flow rate,  $Q_L$ . A  
53 similar trend is seen for the carbon dioxide case (darkened symbols). In Fig. 8 (b), the  
54 maximum value of  $K_{La}$  data at a fixed  $Q_L$  in each MBG is plotted against the water  
55 power,  $L_L$ , with the same symbol as seen in Fig. 8 (a). Since the ratio of  $K_{La}$  to  $L_L$  is  
56 considered to be one of the indices of efficiency, the line tangent to the data through the  
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origin is steeper the more efficient. Thus, LP-12.5 and LP-14.6 types with punched porous at a higher liquid supply rate of  $Q_L = 67$  and  $72$  l/min are superior to others included LF-type with fiber porous. The main reason of this is that the maximum  $Q_G$  is larger in these LP-types. In addition, the present data are compared with those obtained by Terasaka et al. [12] for four kinds of MBG: spiral liquid flow type (Nanoplanet Co., Ltd.) [5], ejector type (Aura Tech Co., Ltd.), Venturi type (Watanabe et al. [13]) and pressurized dissolution type (Shigenkaiatsu Co., Ltd.). Of these four types, spiral liquid flow type showed best performance in  $L_L < 240$  W [12], so the range of data for spiral liquid flow type is drawn on Fig. 8 (b). From the comparison in Fig. 8 (b), LP-12.5 and LP-14.6 types in the present MBG are superior to spiral liquid flow type and the other types tested by Terasaka et al. [12].

Fig. 9 compares  $E_A$  data mainly for the oxygen case. The effects of MBG type on  $E_A$  are relatively small.  $E_A$  for all cases are almost 25 to 30 %, but decrease a little as  $Q_G$  increases. The difference in  $E_A$  between the oxygen case and the carbon dioxide case is small because in Eq. (5)  $K_{La}$  is smaller but  $C_S$  is larger for carbon dioxide case than for oxygen case.

#### 4. Test of prediction model

##### 4.1. Hydraulic performance

Sadatomi et al. [2] proposed a model which can predict well the micro-bubble generation rate for the spherical body-type MBG (Sadatomi et al. [8, 9]) placed at any water depth. In the model, the following energy equations are simultaneously solved.

Liquid inlet  $\leftrightarrow$  Confined point:

$$p_L + \frac{\rho_L v_{L1}^2}{2} = p_2 + \frac{\rho_L v_{L2}^2}{2} + \zeta_1 \frac{\rho_L v_{L1}^2}{2} \quad (6)$$

Confined point  $\leftrightarrow$  Point far from exit:

$$p_2 + \frac{\rho_L v_{L2}^2}{2} = p_3 + \zeta_2 \frac{\rho_H v_H^2}{2} \quad (7)$$

Gas inlet  $\leftrightarrow$  Confined point:

$$p_2 + \frac{\rho_G v_{G2}^2}{2} = p_G - \zeta_3 \frac{\rho_G v_{G2}^2}{2} \quad (8)$$

Here,  $p_L$ ,  $p_2$ ,  $p_3$  and  $p_G$  are the static pressures, respectively at the liquid inlet, the confined point, the point far from the exit and the gas inlet.  $v_{L1}$ ,  $v_{L2}$  and  $v_{G2}$  are the mean velocities at the liquid inlet, the confined point and the gas inlet.  $\rho_L$  and  $\rho_G$  are the densities of the liquid and the gas.  $\zeta_1$ ,  $\zeta_2$  and  $\zeta_3$  are the energy loss coefficients which must be determined from experimental data as described more detail in Ref. [2], and their values for the present MBG are listed in Table 2.  $\rho_H$  and  $v_H$  are the density and the velocity of the homogeneous mixture of the micro-bubbles and water, defined as:

$$\rho_H = \frac{\rho_G Q_G + \rho_L Q_L}{Q_G + Q_L}, \quad v_H = \frac{Q_G + Q_L}{\pi D^2 / 4} \quad (9)$$

The calculated results by the above model are compared with the present data on the present MBG with the orifice and the porous pipe. Figs. 10 (a) and (b) are the examples of such a comparison for the micro-bubble generation rate,  $Q_G$ , and the gauge pressure at the MBG liquid inlet,  $p_L$ . The agreement between the calculated curves and the experimental data points is good. Thus, if we want to generate micro-bubble at an arbitrary flow rate and at any water depth, we can evaluate the water supply rate to the MBG and the gauge pressure at the MBG inlet by the model, thus we can select the optimum pump for supplying water to the MBG.

#### 4.2. Bubble dissolution into water

Kawahara et al. [3] proposed a correlation of correction factor,  $f_C$ , in the following Nedeltchev et al.'s  $K_{La}$  correlation [14]:

$$K_L \alpha = f_C \sqrt{\frac{4D_L u_B}{\pi d_{BS}}} \frac{6Q_G H}{V d_{BS} u_B} \quad (10)$$

Here,  $D_L$  is the liquid-phase diffusion coefficient, and  $u_B$  the bubble velocity,  $d_{BS}$  the Sauter mean bubble diameter,  $H$  the depth of the liquid phase in the bubble column, and  $V$  the total volume of the gas and the liquid in the bubble column.

Nedeltchev et al. [14] proposed an  $f_C$  correlation for milli-bubbles of  $1.7 < E_O < 7$  as:

$$f_C = 0.185 E_O^{0.737} \quad (11)$$

Here,  $E_O$  is the Eotvos number defined by,

$$E_O = \frac{g d_{BS}^2 (\rho_L - \rho_G)}{\sigma} \quad (12)$$

Kawahara et al. [3] obtained an  $f_C$  correlation for micro-bubbles of  $0.006 < E_O < 0.55$  in  $Q_G \leq 4$  l/min in tap water and salt water generated by the spherical body-type MBG as:

$$f_C = 2.36 E_O^{0.735}. \quad (13)$$

Since micro-bubble diameter in tap water was 5 to 10 times larger than that 3 wt % salt water [3], the range of  $E_O$  for the present micro-bubbles in tap water was  $0.08 < E_O < 0.77$  and biased to higher  $E_O$  side than that in [3]. Since  $f_C$  by Eq. (13) is 40 % higher at maximum for the present micro-bubbles, so for the present micro-bubbles we modified  $f_C$  further by

$$f_C = 1.69 E_O^{0.72}. \quad (14)$$

Fig. 11 compares  $K_{La}$  between the calculations by Eqs. (10), (12) and (14) and the

1  
2  
3 present experimental data in  $Q_G \leq 4$  l/min for the 14 combinations of the MBG types  
4 and  $Q_L$ , where the MBG was placed at  $H = 1.2$  m in water depth. In the calculation,  
5 the present experimental data of  $d_{BS}$  and  $u_B$  were given as the input data. Irrespective  
6 of the orifice size, the MBG size and the porous type, the data are well predicted by the  
7 calculations within  $\pm 20$  %.  
8  
9

## 10 5. Concluding remarks

11  
12 The generation and the dissolution of micro-bubbles in tap water were studied for a  
13 newly developed MBG with orifice and porous pipe. From the experiments of the six  
14 kinds of trial products with different orifice diameter, different porous pipe and different  
15 MBG size together with the analyses of the data, the followings have been clarified:  
16  
17

- 18 (1) As the diameter ratio of orifice to pipe in the present MGB, about 0.57,  
19 corresponding to LP-12.5, SF-4.0, MF-8.4 and LF-12.5 types, is recommended  
20 since the ratio of micro-bubble generation rate to power consumption rate was  
21 higher at  $H < 1.2$  m and the maximum attainable bubble generation rate was higher.  
22
- 23 (2) Mean bubble diameter and Sauter mean bubble diameter for the present MBG was  
24 around 0.12 mm and 0.63 mm at  $Q_G = 1.0$  l/min if the mean water velocity through  
25 the orifice is higher than about 10 m/s. No appreciable effects on bubble diameter  
26 are seen between the 300  $\mu\text{m}$  punching porous and 25  $\mu\text{m}$  fiber porous.  
27
- 28 (3) The volumetric mass transfer coefficient,  $K_{La}$ , increased linearly with  $Q_G$ ,  
29 independent of  $Q_L$  and MBG type, and is higher for the punching porous type than  
30 the fiber porous type.  $K_{La}$  for the carbon dioxide was very low, about one-seventh  
31 to one-eleventh of that for oxygen.  
32
- 33 (4) For the ratio of  $K_{La}$  to  $L_L$ , one of the indices of efficiency, the present LP-12.5 and  
34 LP-14.6 types are superior to four kinds of MBG tested by Terasaka et al. [12]  
35 including spiral liquid flow type MBG.  
36
- 37 (5) The ratio of oxygen dissolved in water to that supplied,  $E_A$ , was almost 25 to 30 %,   
38 roughly independent of  $Q_G$  and MBG type in  $Q_G < 10$  l/min.  
39
- 40 (6) The micro-bubble generation rate could be well predicted by the Sadatomi et al.'s  
41 model [2] by giving the experimentally determined energy loss coefficients,  $\zeta_1$ ,  $\zeta_2$   
42 and  $\zeta_3$  as input data.  
43
- 44 (7) The air micro-bubble dissolution rate in tap water could be well predicted by the  
45 Kawahara et al.'s model [3] by lowering the correction factor by about 40 %.

46 All conclusions mentioned above are effective within the present experimental range.  
47 So, in order to validate the present conclusions to other cases, further studies are needed.  
48 In addition, it is very important to construct a model applicable to the dissolution of  
49 carbon dioxide in water and sea water from a global warming protection point of view.  
50

## 51 Acknowledgements

52  
53 The authors would like to express their heartfelt gratitude to Mr. K. Ariyoshi, a  
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56 supports from JSPS KAKENHI (21560181), Harada Memorial Foundation and Big-Bio  
57 Co. are also appreciated.  
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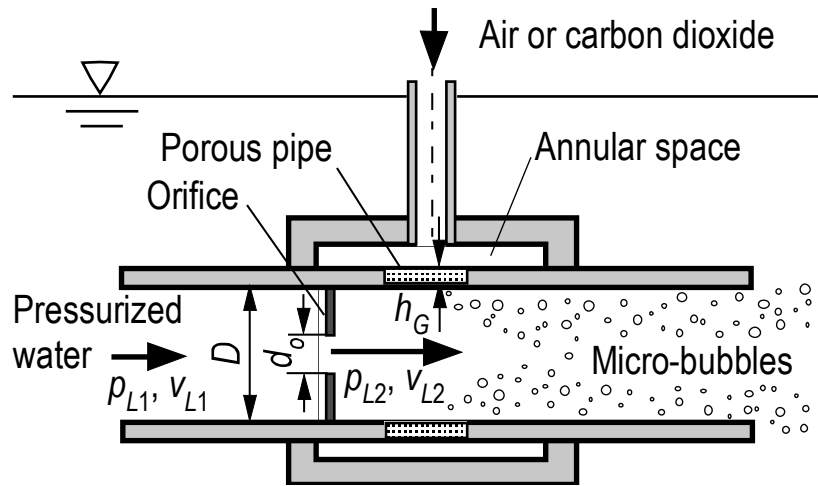
## References

- [1] M. Sadatomi, A. Kawahara, Fluids mixer and fluids mixing method, Japanese Patent, JP. 2008-173631, 2008.
- [2] M., Sadatomi, A. Kawahara, F. Matsuyama, T. Kimura, An advanced micro-bubble generator and its application to a newly developed bubble-jet type air-lift pump, *Multiphase Science and Technology* 19 (2007) 323–342.
- [3] A. Kawahara, M. Sadatomi, F. Matsuyama, H. Matsuura, M. Tominaga, M. Noguchi, Prediction of micro-bubble dissolution characteristics in water and seawater, *Experimental Thermal and Fluid Science* 33 (2009) 883–894.
- [4] H. Ohnari, Waste water purification in wide water area by use of micro-bubble techniques (in Japanese), *Japanese Journal of Multiphase Flow* 11 (1997) 263–266.
- [5] H. Ohnari, T. Saga, K. Watanabe, K. Maeda, High functional characteristics of micro-bubbles and water purification (in Japanese), *Resources Processing* 46 (1999) 238–244.
- [6] H. Ohnari, Fisheries experiments of cultivated shells using micro-bubbles technique (in Japanese), *Journal of the Heat Transfer Society of Japan* 40 (2001) 2–7.
- [7] T. Ueyama, M. Miyamoto, *World of micro-bubbles – Cooperative action of water and gas* (in Japanese), Kougyoutyousakai, Tokyo, 2006.
- [8] M. Sadatomi, A. Kawahara, K. Kano, A. Ohtomo, Performance of a new micro-bubble generator with a spherical body in a flowing water tube, *Experimental Thermal and Fluid Science* 29 (2005) 615–623.
- [9] M. Sadatomi, Micro-bubble generator, Japanese Patent 4069211, 2008.
- [10] K. Akita, F. Yoshida, Gas holdup and volumetric mass transfer coefficient in bubble columns, *Industrial and Engineering Chemistry, Process Design and Development* 12 (1973) 76–80.
- [11] M. Sadatomi, A. Kawahara, T. Goto, Experiment and performance prediction of a bubble-jet-type air-lift-pump for dredging sediments, *Proceedings of 7th Int. Conf. on Heat Transfer, Fluid Mechanics and Thermodynamics (July 2010), held in Antalya, Turkey, 6 pages in CD-ROM.*
- [12] K. Terasaka, A. Hirabayashi, T. Nishino, S. Fujioka, D. Kobayashi, Development of microbubble aerator for waste water treatment using aerobic activated sludge, *Chemical Engineering Science* 66 (2011) 3172–3179.
- [13] Watanabe, K., Fujiwara, A., Takagi, S., Matsumoto Y., The experimental study of micro-bubble generator by Venturi tube (in Japanese), *Proceedings of the Annual Meeting of the Japanese Society for Multiphase Flow (August 2004)* 185–186.
- [14] S. Nedeltchev, U. Jordan, A. Schumpe, A new correction factor for theoretical prediction of mass transfer coefficients in bubble columns, *Journal of Chemical Engineering of Japan* 39 (2006) 1237–1242.

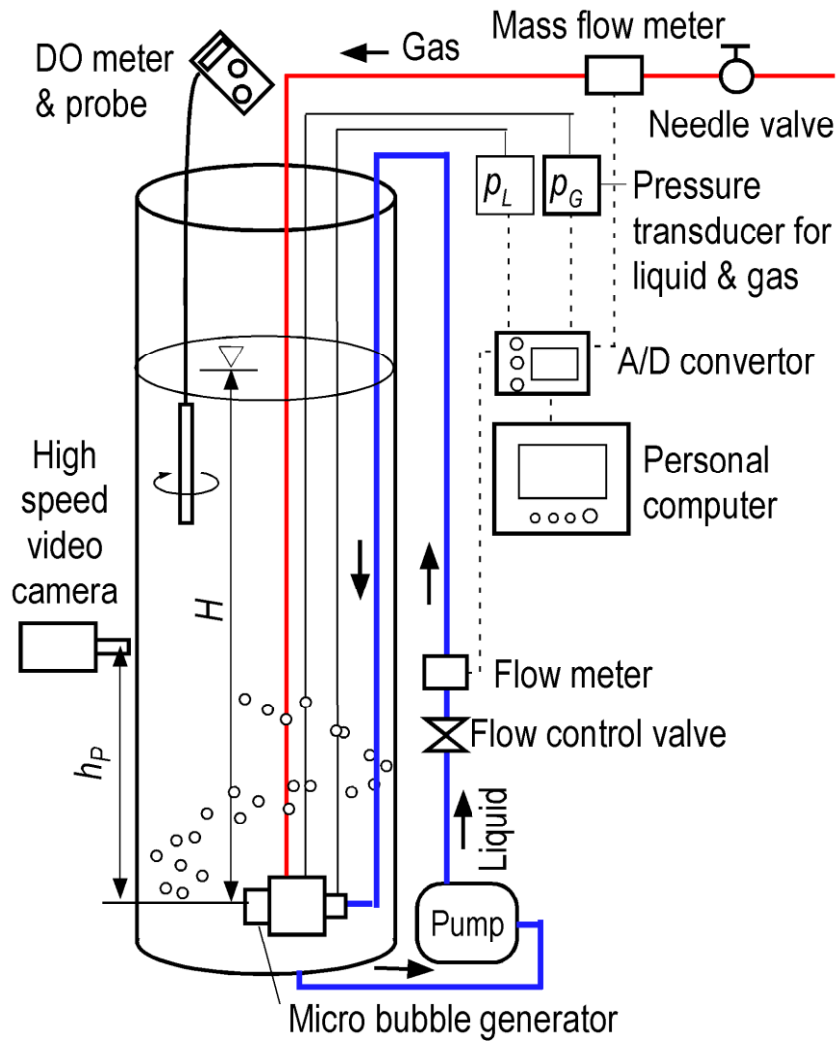


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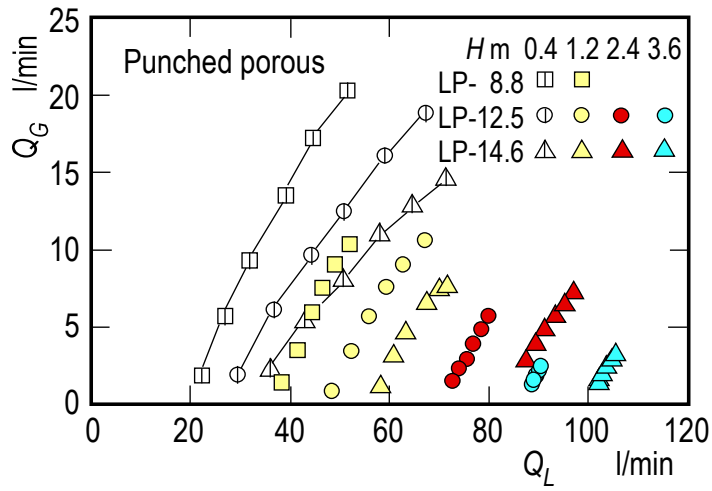
- Fig. 1.** Micro-bubble generator by Sadatomi & Kawahara [1].
- Fig. 2.** Experimental apparatus.
- Fig. 3.** Effects of orifice diameter on hydraulic performance for LP type MBG: (a)  $Q_G$  vs.  $Q_L$ , (b)  $p_L$  vs.  $Q_L$ , (c)  $Q_G$  vs.  $L_L$ .
- Fig. 4.** Effects of MBG size on hydraulic performance for LF type MBG (a)  $Q_G$  vs.  $Q_L$ , (b)  $Q_G$  vs.  $L_L$ .
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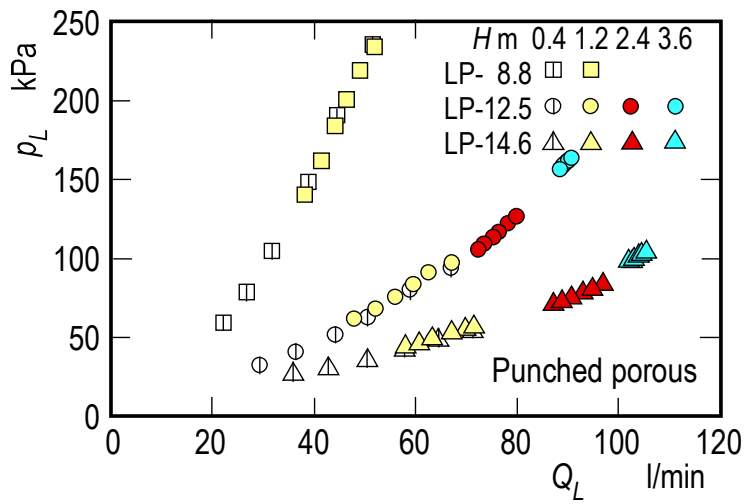
**Fig. 1.**



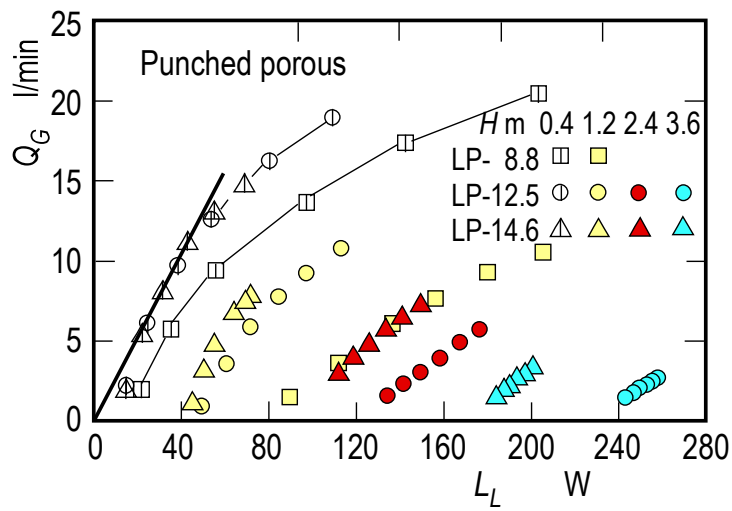
**Fig. 2.**



(a)

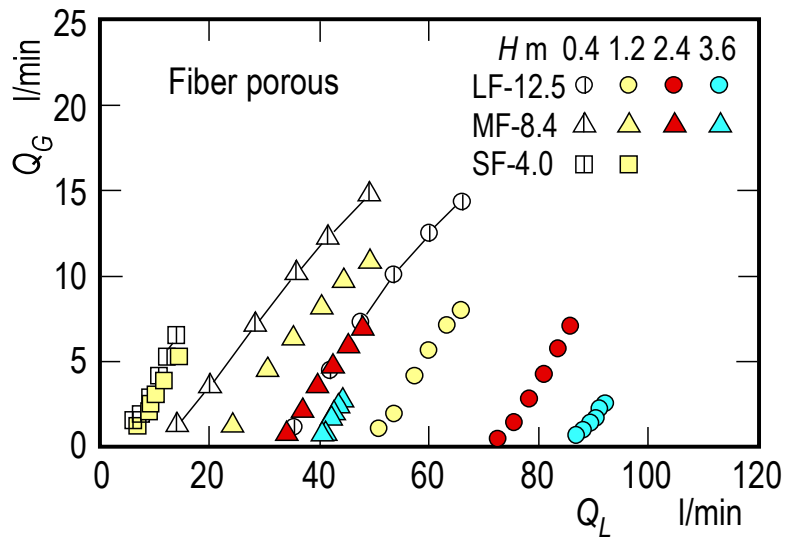


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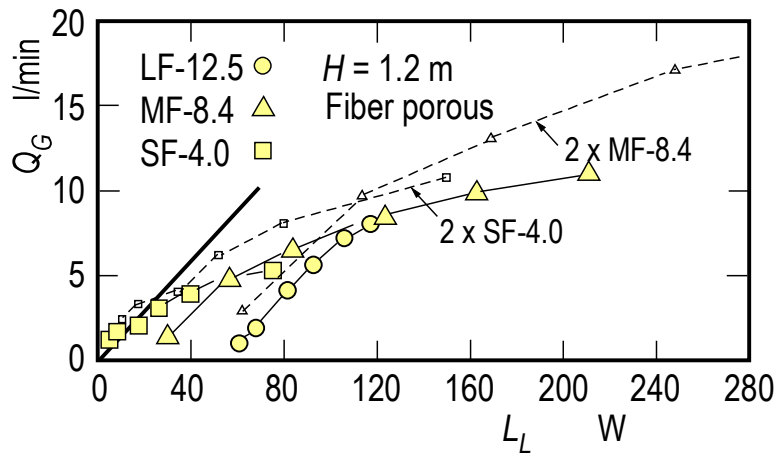


(c)

Fig. 3.



(a)



(b)

**Fig. 4.**

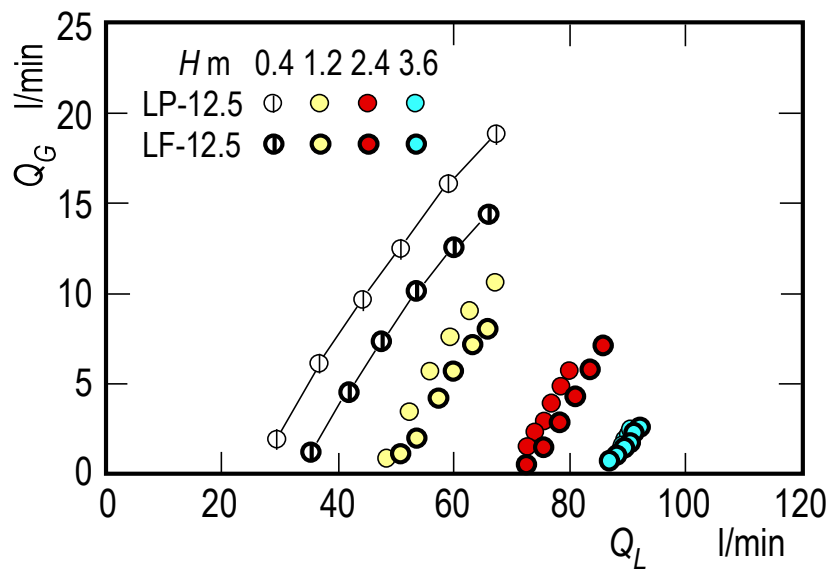
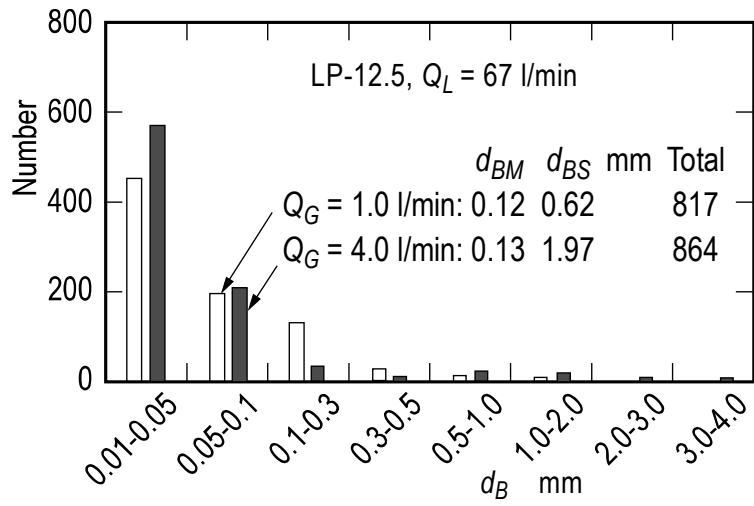
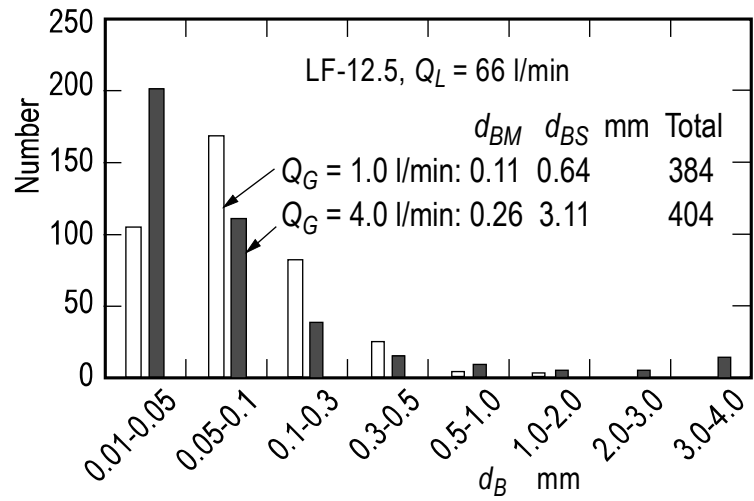


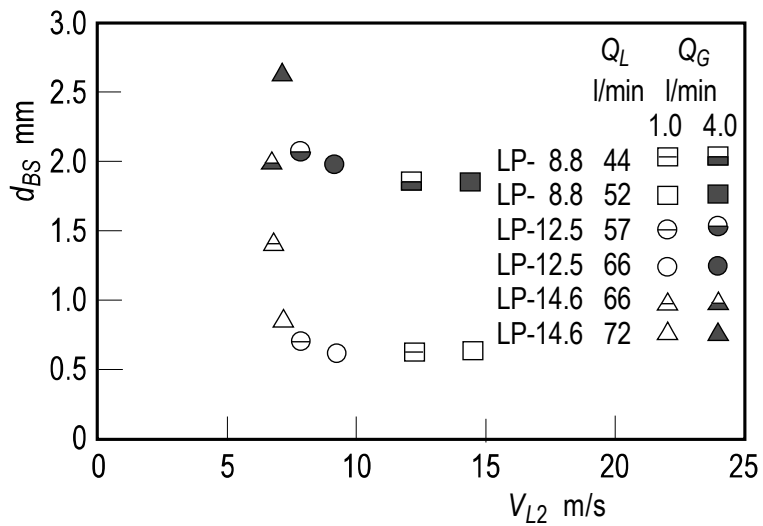
Fig. 5.



(a)

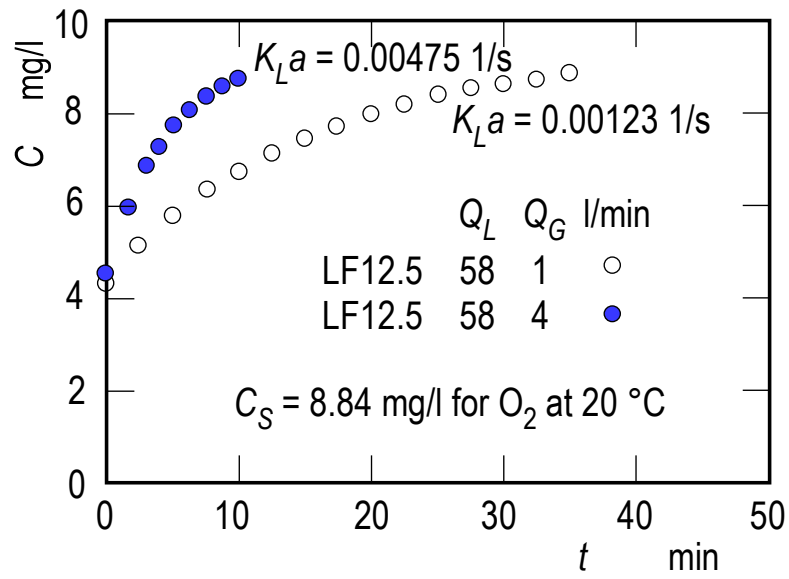


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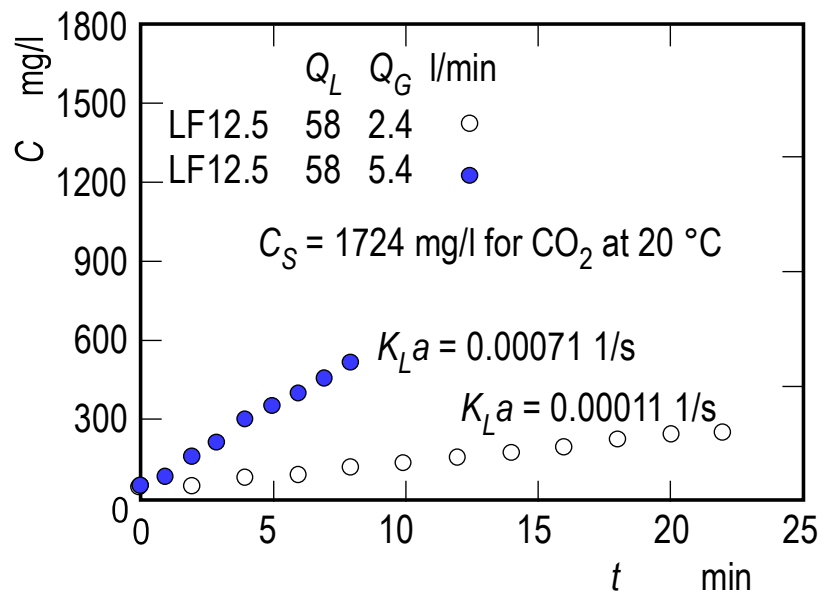


(c)

**Fig. 6.**



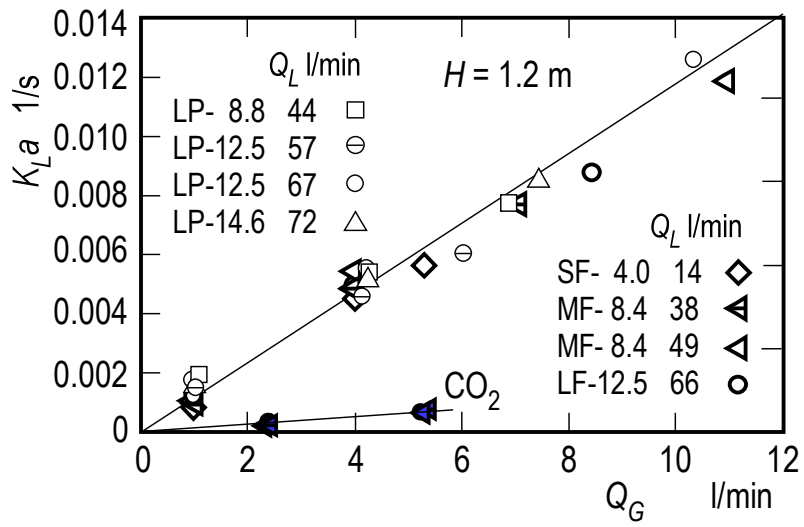
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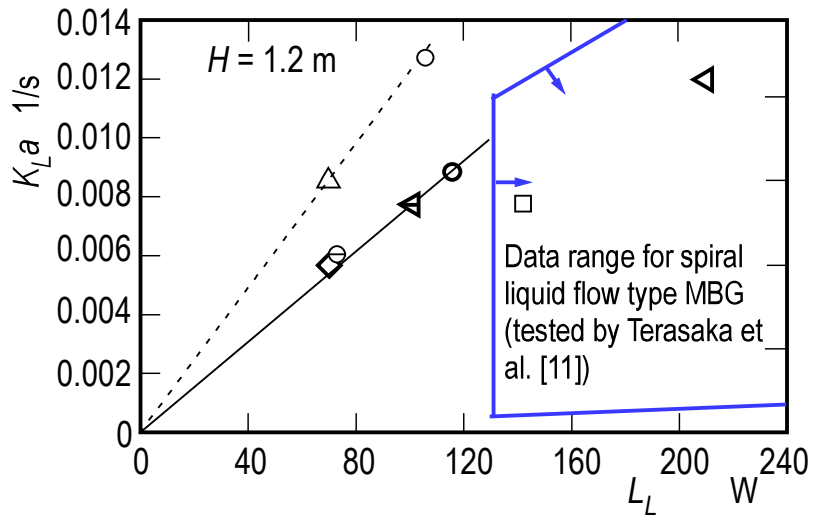
(b)

**Fig. 7.**



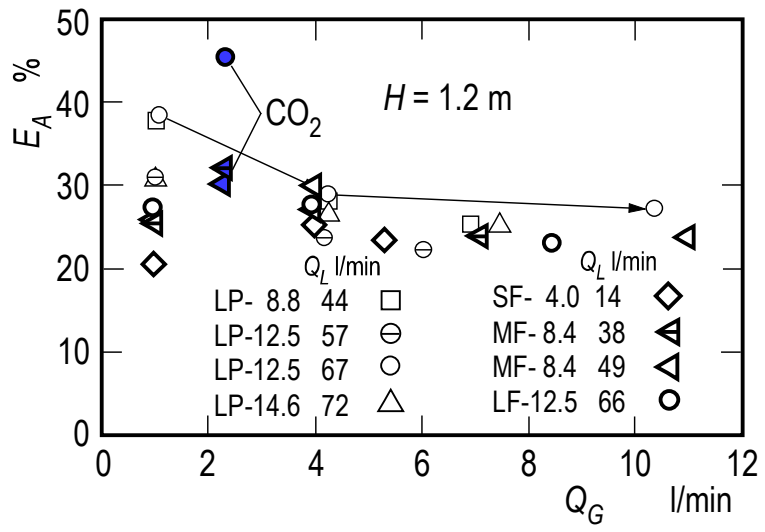


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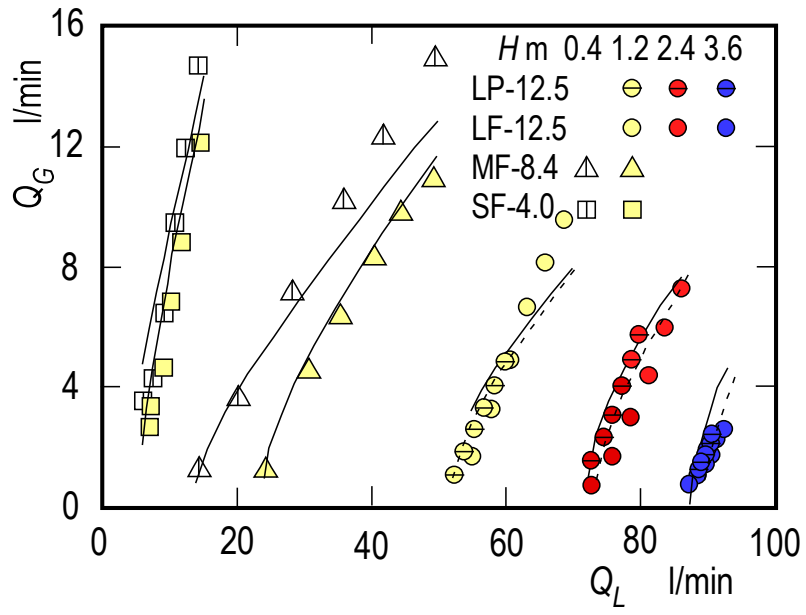


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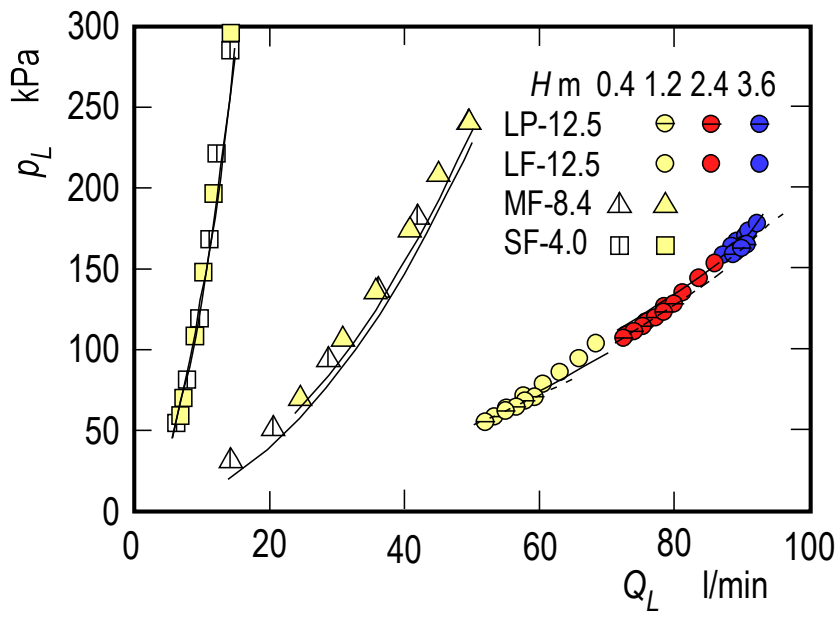
**Fig. 8.**



**Fig. 9.**

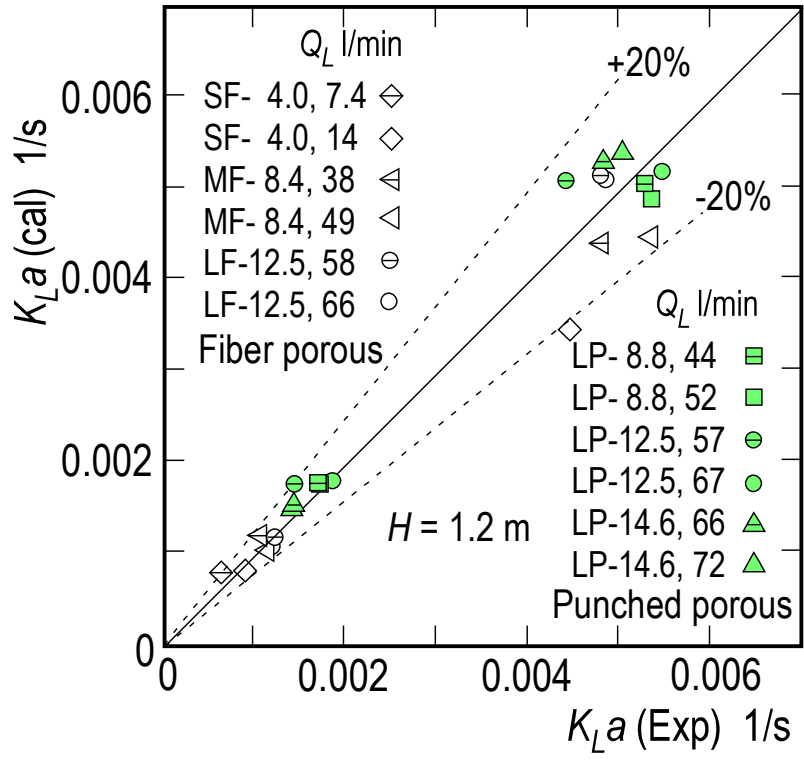


(a)



(b)

**Fig. 10.**



**Fig. 11.**

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**Table 1** Specification of MGB tested.

**Table 2** Energy loss coefficients for water inlet, outlet and porous pipe of MBG.

**Table 1**

MBG type	Porous	$D$ mm	$d_o$ mm	$\beta^2$ -	$l$ mm	$d_G$ $\mu\text{m}$	$h_G$ mm	$A_H$ $\text{mm}^2$
LP-8.8	Punched	22	8.8	0.16	8	300	0.2	152.7
LP-12.5			12.5	0.323				
LP-14.6			14.6	0.44				
SF-4.0	Fiber	7	4.0	0.327	3-8	25	1.5	unknown
MF-8.4		14.7	8.4	0.327				
LF-12.5		22	12.5	0.323				

**Table 2**

Type	LP-8.8	LP-12.5	LP-14.6	LF-12.5	MF-8.4	SF-4.0
$\zeta_1$	55.8	12.2	5.9	14.5	11.7	9.2
$\zeta_2$	30.4	5.0	1.6	4.6	4.6	3.7
$\zeta_3$	1700	1700	1700	4000	4000	4000

Ms. Ref. No.: ETFS-D-11-00485

Title: Micro-bubble generation rate and bubble dissolution rate into water by a simple multi-fluid mixer with orifice and porous tube  
Experimental Thermal and Fluid Science

To Reviewer #1:

Thank very much for giving the authors a lot of good comments. According to the reviewer's comments, the authors revised the manuscript as written in red characters. In addition, the answers to the respective comments are as follows:

1. The number of reference does not coincide with its order of citation in the main body.

We changed the citation number just above Eq. (3) to [10] and that just above Eq. (10) to [14].

2. Measurement uncertainties in flow rates, pressure, bubble diameter, concentration of oxygen/carbon dioxide and mass transfer coefficients much be clearly described in the manuscript.

We added them in sections 2.2 to 2.5.

3. The authors described that averaged value of the two axes was taken as the bubble diameter" in page 4, line 20 -21. This is not correct as the volume-equivalent bubble diameter for ellipsoidal bubbles. How much is the maximum or minimum aspect ratio of a bubble? Why the authors assume spheroid shape to calculate bubble diameter?

We added sentences in 2.4. Most bubbles were spherical as described in the original manuscript. Over about 1.5 mm, however, a little distorted bubbles appeared, but the aspect ratio was within 1.2. Why the authors assume spheroid bubble is that the interfacial area-equivalent bubble diameter is more important than the volume-equivalent bubble diameter for discussing bubble dissolution in water. In addition, the difference in bubble diameter between the interfacial area-equivalent and the volume-equivalent is negligibly quite small for the present bubbles.

4. Measurement method of concentration of carbon dioxide is not described. The method and its uncertainty must be described in the manuscript.

We added them in section 2.5.

5. Although the authors described "LP-8.8 gives the largest QG t a fixed QL because the water pressure



in the air suction section becomes the lowest at a fixed  $QL$ " in page 5, line 48 - 49, LP-8.8 did not work at  $H = 2.4$  and  $3.6$  m as mentioned in page 5, line 36. Why?

We added the reason in section 3.1. As you can see from Fig. 3 (b), the water pressure at the MGB inlet increased with the water depth,  $H$ , and the water flow rate,  $Q_L$ , thus in order to generate micro-bubbles at  $H = 2.4$  and  $3.6$  m much larger water power must be supplied than that given by the present water circulation pump.

6. The authors described "LP-12.5 is superior to others at  $H > 2/4$  in page 5, the last line. However, LP14.6 realizes higher QG at the constant LL than LP12.5 judging from Fig. 3 (c), and therefore, LP14.6 has better performance than LP12.5.

As to Fig. 3 (c) we modified the explanation and added one reference in section 3.1.

7. There is no explanation of experiment using double SF-4.0/MF-8.4. Did the two MBG locate in parallel or tandem?

We added a sentence in section 3.3. Two MBGs are assumed to be placed at the same water depth.

8. In the concentration measurement of oxygen, is there no influence of dissolution of ambient air through the water surface in the tank. How much is the time scale of the dissolution of oxygen through the water surface?

The effect of the oxygen dissolution from the free water surface is negligible, because the interfacial area in the free water surface is extremely smaller than that in the total of micro-bubbles.

9. The description "the increase of the concentration is low" is vague, because the magnitude of the gradient is large due to large difference between  $C$  and  $C_s$ . The authors mentioned that  $K_La$  for carbon dioxide case is unexpectedly small in page 7, line 23. It is recommended to add available literatures on  $K_La$  of  $CO_2$ .

The reviewer misread "slow" as "low", but we modified a little as "very slow". There is no open data on  $K_La$  for  $CO_2$  micro-bubbles, as we know. Why we wrote "unexpectedly small" is that we had had a prejudice  $CO_2$  to be an easily dissolve into water as lemonade and cola.

10. Judging from Fig. 8 (b), LP12.5 with  $QL = 57$  shows lower performance than LP14.6 and LP12.5

with  $Q_L = 67$ , although the authors mentioned that LP-12.5 and LP14.6 type with punched porous are superior to others in page 7. Line 37 - 38.

We added  $Q_L$  values there.

11. In Fig. 6 (a) and (b), there is large difference in the total number of bubbles for  $Q_G = 1.0$  l/min between LP-12.5 and LF-12.5, whereas the difference in bubble size is very small. Why? Does the mass flow rate evaluated from measured bubble size and bubble number coincide with the gas flow rate?

The gas flow rate measured does not coincide with that determined from bubble diameter and bubble number data since the bubble diameter was measured only for bubbles taken by several pictures. In order to obtain reliable bubble diameter distribution in the present measurement, 350 bubbles are sufficient as added in section 2.4. Thus, the number of 817 and 864 in Fig. 6 (a) were too many and time consuming for the measurement.

12. Most of conclusions seem to be just results for the present experimental apparatus and conditions. Applicability of the knowledge to the other MBG or micro-bubble systems is unclear. It is strongly recommended to add discussions on physical background of the results.

We added sentences in chapter 5.

Reviewer #2: The paper describes on generation and dissolving rates of micro-bubbles formed by the authors' prepared bubbler. The paper includes an interesting information for readers of this journal and acceptable after minor revision as pointed out below;

Thank very much for giving the authors good comments. According to the reviewer's comments, the authors revised the manuscript as written in red characters. In addition, the answers to the respective comments are as follows:

- 1) Description on the details of porous materials and measurements of micro-bubbles is insufficient. The authors should to describe more details on those matters.

For porous materials, we added their names in section 2.1. For micro-bubble measurements, we added sentences in 2.4.

- 2) English is necessary to be polished up by native speakers who can understand the scientific contents.

Several sentences are corrected according to the advice.