



Reactor design optimization for direct synthesis of hydrogen peroxide

Tomoya Inoue^{a,*}, Yoshikuni Kikutani^b, Satoshi Hamakawa^a, Kazuma Mawatari^b, Fujio Mizukami^a, Takehiko Kitamori^{b,c}

^a Research Center for Compact Chemical Process, National Institute of Advanced Industrial Science and Technology (AIST), Sendai, Miyagi, Japan

^b Micro Chemistry Group, KAST, Kawasaki, Kanagawa, Japan

^c Department of Applied Chemistry, Graduate School of Engineering, The University of Tokyo, Bunkyo-Ku, Tokyo, Japan

ARTICLE INFO

Article history:

Received 15 July 2009

Received in revised form

24 December 2009

Accepted 15 February 2010

Keywords:

Direct synthesis of hydrogen peroxide from hydrogen and oxygen
Glass-fabricated microreactor
Gas–liquid distribution

ABSTRACT

A glass-fabricated microreactor was applied to the direct synthesis of hydrogen peroxide from hydrogen and oxygen. The direct synthesis reaction is a three-phase reaction, with gas (hydrogen and oxygen), liquid (reaction solution) and solid (catalyst) being involved. We designed an advanced microreactor in which an ideal gas–liquid distribution could be accomplished throughout the catalyst packed bed. We also set up a reaction system that enables the reaction operation at more than 2 MPa over 1 week. With these efforts, more than 3 wt% of hydrogen peroxide was successfully produced using the microreactor technology.

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1. Introduction

Hydrogen peroxide has a wide range of application, including oxidation process, bleaching, wastewater treatment, and semiconductor wafer cleaning. So far, it has been mainly produced by the anthraquinone process, in which an organic solution containing modified anthraquinone (called the working solution) is circulated in the redox cycle to produce hydrogen peroxide. However, since the contamination from the partially degraded working solution occurs, a purification process, which consumes a large amount of energy, is required in this process [1,2].

The direct synthesis of hydrogen peroxide from hydrogen and oxygen has been developed as an alternative in past decades. It usually consists of a three-phase reaction, which involves gas (hydrogen and oxygen), liquid (reaction solution to store hydrogen peroxide produced), and solid (catalyst). This process would be environmentally benign as it only produces water as byproduct, and would be merited by its less energy consumption, reducing the post-reaction processes, condensation and purification.

Its development is mainly based on a continuous stirred tank reactor (CSTR), in which both hydrogen and oxygen are fed into the reaction solution suspending a slurry catalyst [3,4]. The main issue of the process development is to promote the productivity under a severe safety procedure, limiting hydrogen content below 4 vol.%

to prevent an explosive reaction. Intensified mixing is preferred to maintain well-dispersed gas bubbles in the reaction solution, but it may cause another challenge for a batch reactor design, as the reaction is conducted at a high pressure up to 10 MPa [5].

Recently, efforts in process miniaturization, namely microreactor technology, have shown an opportunity for conducting chemical processes by preventing potential danger [6–8]. The examples are such as direct fluorination with gas phase fluorine, ozonolysis, and hydrogen oxidation reaction in gas phase [9–11]. Rapid heat dissipation by the factor of 100 prevents the generation of hot spot during reaction, owing to the higher surface to volume ratio of microchannels compared to conventional reactors. In addition, the microstructure of the reactor helps generate a higher interfacial area to promote mass transfer in multiphase reactions.

It is reasonable to regard these characteristics of microreactor technology as suitable for the direct synthesis of hydrogen peroxide from hydrogen and oxygen. In fact, it has been shown that the nature of this technology, such as rapid heat dissipation, suppresses a radical propagation in hydrogen oxygen mixture to conduct the reaction safely [8,12–17]. Furthermore, the author successfully analyzed the reactor performance thorough the efficiency estimation of the mass transfer from gas to liquid. The overall mass transfer coefficient (K_1a_1) for hydrogen has been estimated to be 3.8 s^{-1} , which is much more efficient compared to conventional reactor technologies, in the range from 0.01 to 0.08 s^{-1} [12,13,18,19].

Yet, the concentration of hydrogen peroxide achieved was merely 0.2 wt% and an increased productivity is required for indus-

* Corresponding author. Tel.: +81 22 237 3096; fax: +81 22 237 5226.
E-mail address: inoue-tomoya@aist.go.jp (T. Inoue).

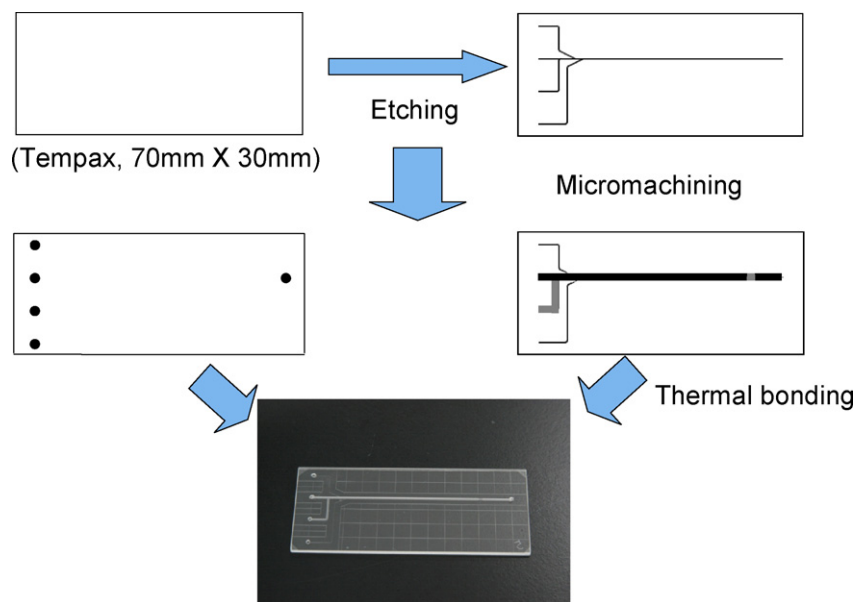


Fig. 1. Fabrication procedure of the microreactor.

trial use. It is suggested that there is more room to promote the productivity by validating a proper gas–liquid distribution during the reaction, as some experimental results indicated the maldistribution problem [12,13].

Based on these considerations, we intend to design a brand-new microreactor by glass fabrication. As glass is transparent, the flow regime inside the reactor can be easily monitored during the reaction. Also the fine chemical processes have proved the applicability of glass as an ideal microreactor material, Lonza's c-SSP process being a good example [20].

2. Experimental

2.1. Reactor fabrication

Fig. 1 shows the fabrication scheme of the microreactor used for reaction. The total reactor design was fabricated on the double side polished Tempax glass sheets (70 mm × 30 mm, 0.7 mm thickness, Nippon Sheet Glass Co.,) by chemical isotropic etching using HF solution. Lattice structures were also etched, which helped the adhesion of each glass sheet so that gas bubbles could escape through the channels during the thermal bonding process. The etched structure was controlled so that the line width is ca. 0.05 mm and the line depth is 0.02 mm. The etched glass sheets were further fabricated by a numerically controlled (NC) drilling system, by which a catalyst packed bed, gas/liquid inlet holes and liquid inlet channels were formed. Finally, two glass sheets were thermally bonded to form the microreactor. It should be noted that we chose to use chemical etching to fabricate thin channels with 0.1 mm width or less, applying mechanical machining to carving out larger channels with more than 0.1 mm width.

The overview of the reactor and its local structure is shown in Fig. 2. The gas delivery channels consist of thin channels fabricated by etching. The liquid delivery channel and catalyst packed bed are fabricated having the width of ca. 0.6 mm. The lattice structures, which were used for bubble-escape to make sure the close contact of each layer during the thermal bonding of the glass sheets, were designed carefully so as not to interfere with the reactor channels. Once the microreactor was prepared, we observed the gas–liquid flow regime by packing an aqueous slurry suspension of silica from the inlet for “catalyst loading”, shown in Fig. 2(a) and

(b). The packed particles were maintained in the bed by the dam structure at the end of the packed bed shown in Fig. 2(c).

2.2. Reaction system set-up

Fig. 3 shows the reaction system set-up using the microreactor. We tested two kinds of catalyst, Pd/C (5 wt% Pd, N.E. Chemcat Co., lot no. 21A-0709549), and Pd/Al₂O₃ (5 wt% Pd, Nippon Engelhard Co., lot no. 97) so far. Both catalysts were suspended in deionized water, and were loaded with the silica-packing procedure mentioned before. The catalyst packed microreactor was set to a jig to connect gas and liquid dispensing tubing. We used both mechanical coupling and adhesives to interface these tubings and the inlet/outlet holes of the microreactor, as shown in Fig. 3(a) and (b). A swagelok ferrule was attached mechanically at the end of each 1/16 PEEK tubing that was then adhered to the reactor using a silicone adhesive. The interconnected area was also mechanically compressed to increase robustness so that the reaction at 2.0–2.5 MPa could be conducted. Fig. 3(c) shows the interconnected microreactor set to the reaction system. Each of hydrogen (deu-

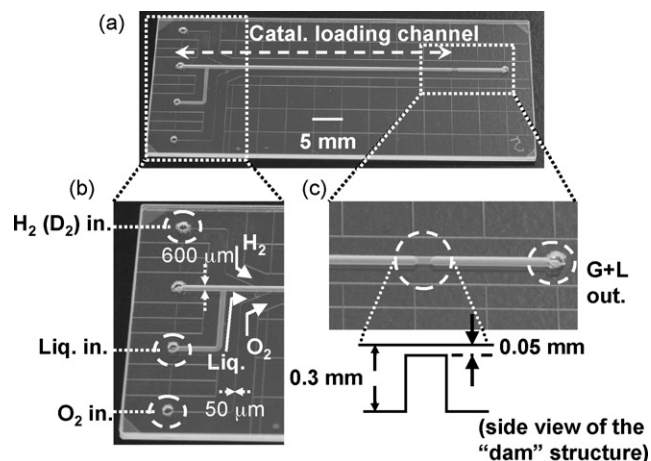


Fig. 2. Design of the glass-fabricated microreactor: (a) whole structure, (b) magnified picture around the inlet, and (c) magnified picture of “dam” structure to retain catalyst particles.

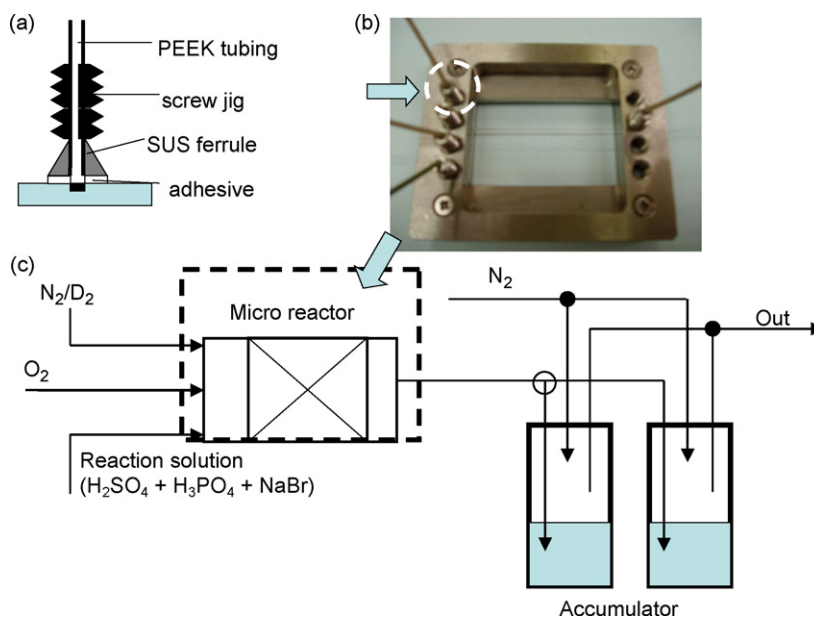


Fig. 3. Reaction set-up for the glass-fabricated microreactor: (a) interconnecting scheme between the reactor and tubing, (b) a picture of microreactor set in the jig, and (c) reaction set-up scheme with gas and liquid inlet, and accumulators at the outlet.

terium) and oxygen were delivered with a mass flow controller (Brooks Instruments 5850E), while the reaction solution was delivered by a HPLC pump (Nippon Seimitsu Kagaku, NP-KX-1000). The reaction pressure was maintained at the outlet, using accumulators and a pressure controller (Brooks Instruments 5866).

We used PEEK tubings (Upchurch Scientific, 1/16 in.) around the interconnected area as can be seen in Fig. 3(a) and (b). PEEK tubings were also used around HPLC pump while SUS 1/8 in. tubings were used around mass flow controllers and pressure controllers. We used Upchurch valves or swagelok valves occasionally, which is not shown in Fig. 3.

We used deuterium in order to estimate the hydrogen conversion and the selectivity to hydrogen peroxide, as hydrogen peroxide and water, another product of this reaction, were dissolved into the reaction solution [13,21]. The reaction effluent was collected in the accumulators. The sample was taken off from the accumulator every 2 h and the amount of hydrogen peroxide and the deuterium conversion were estimated. Hydrogen peroxide was estimated by permanganate titration, while deuterium conversion was estimated using FTIR-ATR to distinguish deuterized product. The reactant solution was prepared by mixing diluted sulfuric acid (0.025 M), phosphoric acid (0.005 M), and sodium bromide (5.1×10^{-4} M) solution [22].

The reaction performances were calculated as follows; note that the denominator of the right side of Eq. (1) is the total amount of D_2O in the sample, as rapid H-D exchange in the sample solution makes the amount of D_2O represent deuterium conversion [13,21].

$$\text{Selectivity (\%)} = \frac{\text{amount of } H_2O_2 \text{ in the sample (mol)}}{\text{amount of } D_2O \text{ in the sample (mol)}} \times 100 \quad (1)$$

$$\text{Yield (\%)} = \frac{\text{amount of } H_2O_2 \text{ in the sample (mol)}}{\text{amount of } D_2 \text{ introduced (mol)}} \times 100 \quad (2)$$

3. Results and discussions

3.1. Direct synthesis of hydrogen peroxide

We could successfully achieve the hydrogen peroxide concentration more than 3 wt% as a preliminary result, a substantial leap from 0.2 wt% in the previous results, as shown in Fig. 4. It should be

noted that we achieve such high concentration with rather lower pressure; some results are at 1 MPa, whereas more than 2 MPa was applied in the previous studies.

The reaction time is also different between the glass-fabricated microreactor studies (filled plot; \blacklozenge , \blacksquare , \blacktriangle) and the Si-fabricated microreactor studies (white plot; \diamond). The packaging scheme contributed to conducting the reaction operation for long time. We have constructed the reaction system that endures up to 2.5 MPa. Its stable interconnecting between fluid dispensing tubings and the reactor helps achieve the condition. The durability of the system has been validated by a weeklong, continuous operation of the reactor system, with start-ups and shutdowns repeated. In Fig. 4, the reaction was conducted continuously for Pd/C catalyst, while Pd/ Al_2O_3 catalyst experienced a start-up and shutdown for each experimental condition without modifying the packaging scheme.

Table 1 shows the selected examples of the reaction including the selectivity, the yield vs. the hydrogen fed into the reactor, and the productivity of hydrogen peroxide per reactor. As a general trend, a higher hydrogen conversion leads to a lower selectivity to hydrogen peroxide, indicating that water is formed by the sub-

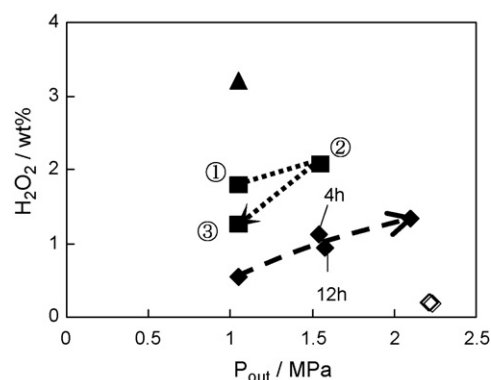


Fig. 4. Glass-fabricated microreactor performance for hydrogen peroxide synthesis. $T = 296$ K; $P = 1.05$ – 2.0 MPa (outlet); catalyst: Pd/C (\blacklozenge) or Pd/ Al_2O_3 (\blacksquare and \blacktriangle); liquid: 0.0075 – 0.0085 mL min^{-1} ; gas: 5 sccm; $D_2/O_2 = 2/3$. Pd/C catalyst was loaded into 0.3 mm-depth channel, while both 0.3 mm-depth (\blacksquare) and 0.9 mm-depth (\blacktriangle) channel was tested for Pd/ Al_2O_3 catalyst. (\diamond) Microreactor performance in the reference [13].

Table 1
Direct synthesis of hydrogen peroxide by the microreactor.

Catalyst	Channel depth (mm)	Reaction pressure (MPa)	H ₂ O ₂				
			wt%	Selectivity (%)	Yield (%)	Productivity	
					(×10 ⁻⁴ mol h ⁻¹) (mol g-cat ⁻¹ h ⁻¹)		
Pd/C	0.3	1.05	0.6	86.5	1.4	0.70	0.069
		1.54 ^a	1.1	70.3	2.7	1.32	0.13
		1.58 ^b	0.9	70.8	2.3	1.14	0.11
		2.10	1.3	59.4	3.4	1.71	0.17
Pd/Al ₂ O ₃	0.3	1.05 ⁽¹⁾	1.8	63.5	5.6	2.78	0.058
		1.55 ⁽²⁾	2.1	62.8	5.4	2.72	0.057
	0.9	1.05 ⁽³⁾	1.3	85.4	3.3	1.64	0.034
		1.05	3.2	29.1	9.9	4.96	0.032

Reaction temperature: 293 K; gas feed rate: 5 mL min⁻¹ (293 K, 0.1 MPa, D₂/O₂ = 2/3); reaction solution feed rate: 0.0075–0.0085 mL min⁻¹.

^a Reaction data were taken from 2 to 4 h after setting the pressure.

^b Reaction data between 4 and 12 h.

sequent reaction of hydrogen peroxide and hydrogen as hydrogen conversion rises. The kinetic analysis of this system is ongoing and will be published later.

3.2. Catalyst

Predominantly, the catalytic reaction on the packed bed is essential in order to attain the reasonable productivity of hydrogen peroxide, as catalyst plays a prominent role in converting a low concentration of reactants (dissolved hydrogen and oxygen) into an adequate amount of product. Here, we focused on Pd/C and Pd/Al₂O₃ taking into account the previous study by the author [13]. Pd/C was useful because of its high selectivity shown in the previous study, while Pd/Al₂O₃ was selected given the commercial availability.

When we used Pd/C, we came across three issues that could hinder a continuous and stable reaction operation. The first was the pressure drop in the catalyst bed during the reaction condition. Although the length of catalyst bed was ca. 40 mm for both Pd/C and Pd/Al₂O₃, the pressure drop reached more than 0.2 MPa for Pd/C while it remained less than 0.1 MPa for Pd/Al₂O₃. The second was the low packed density of Pd/C (0.17 g cm⁻³, 0.8 g cm⁻³ for Pd/Al₂O₃) that limited the amount of catalyst in the reactor.

The third was the oxidation of support material that would erode the catalyst during the process. In the previous study, the support oxidation was not observed because of the short reaction time (ca. 2–3 h), and a longer operation was hampered by the leakage from the reactor–tubing interconnection. In this present study, we developed a packaging scheme that allowed more than 1-week operation and by which also made the catalyst stability issue more visible; 2–3 mm of the catalyst packed bed was eroded through the oxidation of the support during the consecutive operation for 48 h. The catalyst performance deterioration is also noticeable in Fig. 4 (◆), as the H₂O₂ concentration slightly decreased in 12 h operation compared with 4 h.

Pd/Al₂O₃ showed a reasonable selectivity to hydrogen peroxide – contrary to the previous studies that had shown a total oxidation activity to water [13]. This discrepancy presumably stemmed from the difference in reduction treatment during catalyst preparation. Pd/Al₂O₃ from Alfa Aesar (the catalyst of the previous study) was reduced at 573 K by gas phase hydrogen, whereas a liquid phase treatment was applied to the N.E. Catalyst. In fact, the reactor performance also deteriorated for Pd/Al₂O₃, as shown in Fig. 4 (■). We conclude that the deterioration was due to the palladium elution in oxidative atmosphere; indeed, in the series of experiment, we have elevated pressure under oxygen flow.

3.3. Reactor optimization

In the optimization of the reaction condition, we examined the effect of the size of catalyst packed bed by changing the width and depth of the channel. The reaction was conducted safely with the width of 0.6 mm, a result consistent with the previous studies [12,13]. A wider channel up to 1 mm has led to a small scale explosion enough to break the catalyst packed bed, which indicates that hydrogen–oxygen gas mixture bubbles had grown so much that the radical propagation could not be prevented (Fig. 5). Therefore, we conclude that the size of 0.6 mm is actually wide enough to propagate flame at 1 MPa in dry condition but the coexistence of water acts as a flame arrest [23]. We examined the depth by 0.3, 0.6, and 0.9 mm to find out that the reaction could be conducted safely for all cases. Due to the limitation of the glass sheet thickness used, we fixed the maximum depth of the catalyst packed bed to be 0.9 mm.

We also examined a gas–liquid multiphase design. Initially, we designed a reactor by the NC machining only, as shown in Fig. 6(a).

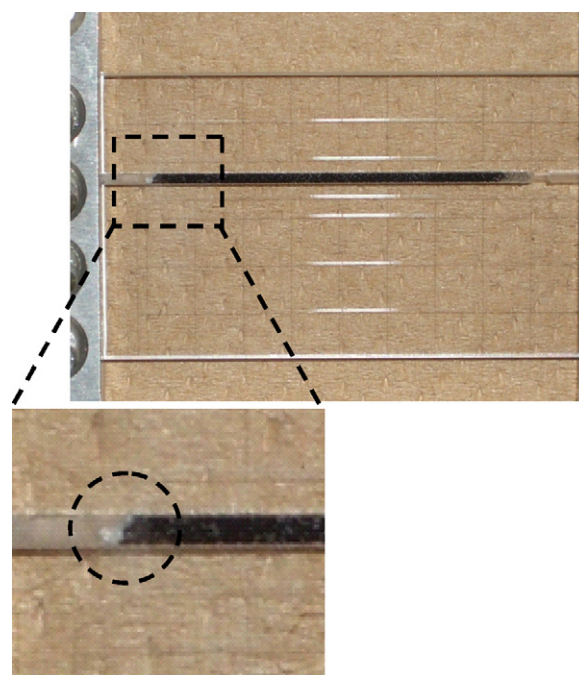


Fig. 5. Microreactor experienced small-scale explosion. Explosion was inevitable with 1 mm width microchannel. White area (in circle, magnified structure) indicates that the wall glass was partially melted by the heat of explosion.

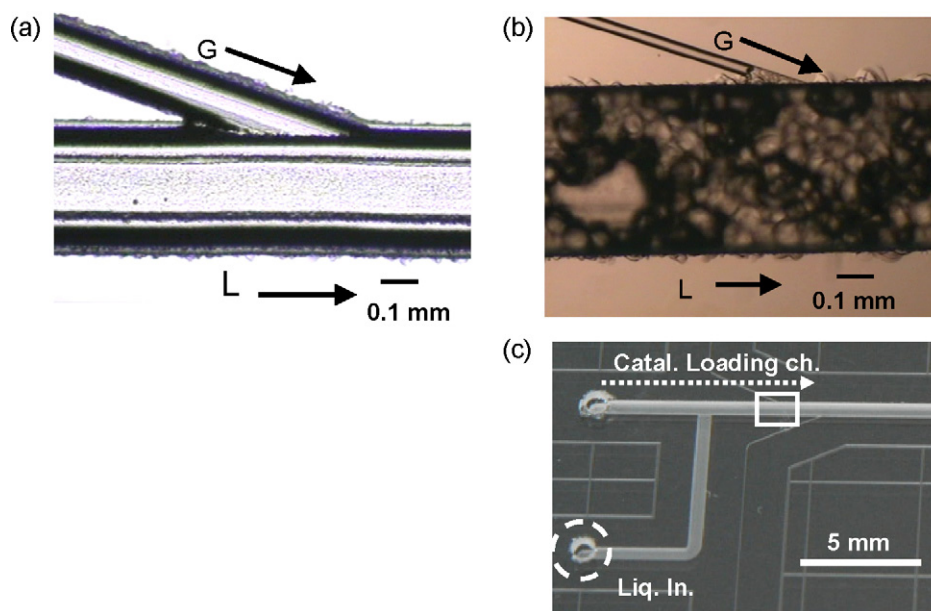


Fig. 6. Magnified picture of gas–liquid mixing area. (a) The microreactor fabricated by NC drilling; channel width for gas inlet is 0.2 mm, and that of liquid dispensing is 0.4 mm. (b) The microreactor fabricated by the combination of chemical etching and NC drilling. The reactor structure is the same shown in Fig. 2, except silica is packed in this figure. The magnified area (b) is corresponding to the rectangle in the reactor, as shown in (c).

However, we failed to achieve a proper gas–liquid distribution. The liquid was likely to “escape” to the gas inlet channel to prevent larger pressure drop at the catalyst packed bed (downstream, not shown), and the gas “blew up” the liquid irregularly. Because of the “cross talk” between the gas and liquid dispensing channels, an irregular pulsing flow was introduced into the catalyst bed occurred. This problem resulted from the difference of pressure drop between gas and liquid, as gas and liquid flows are both pressure-driven. Fortunately, this “cross talk” problem was significantly improved by differentiating the sizes of the fluid dispensing channels (Fig. 6(b)). The liquid did not get into the gas inlet channel as the gas flow push the liquid off the channel, which lead to the controlled gas–liquid flow through the catalyst packed bed.

In order to fabricate the microreactor with proper gas–liquid flow, we applied the combination of chemical etching and NC drilling as mentioned in Section 2.1. Chemical etching was suitable to fabricate microchannels of 0.05 mm width or less. Glass became thin and fragile when we tried to etch channel deeper than 0.2 mm. Mechanical drilling was suitable to fabricate that of sub-millimeter width, but drilling blade was unavailable for fabricating thin channels, width of 0.1 mm or less.

The trickling flow regime was validated by the silica-packed microreactor, the same flow being putatively reproduced during the reaction. The disparity of channel size is crucial for the reactor design, especially for achieving a proper gas–liquid distribution in the reactor. Generally, liquid is 1000 times more viscous compared with gas, which is the main cause of the disparity of pressure drop among the equally fabricated microchannels [24,25]. In order to ensure a proper gas–liquid flow for the reaction, gas phase pressure drop should be countered with increasing a superficial velocity.

4. Conclusions

We have successfully developed a glass-made microreactor suitable for the direct production of hydrogen peroxide from hydrogen and oxygen. The width of the catalyst packed bed channel is a crucial factor to safely conduct the reaction. Gas–liquid distribution is controlled by reactor design, compensating the viscosity difference between gas and liquid. The reactor packaging scheme is

also developed that enables a stable continuous operation up to 1 week.

Reactor performance development as well as numbering-up effort is in progress and will be reported in the near future.

Acknowledgement

This research project was partially supported by the Grant for Industrial Technology Research (Project ID: 07A31002a), NEDO (The New Energy and Industrial Technology Development Organization), Japan.

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