*Chemistry*

*Analytical Chemistry fields*

Okayama University *Year* 2006

# A membraneless gas diffusion unit: Design and its application to determination of ethanol in liquors by spectrophotometric flow injection

N. Choengchan[*] T. Mantim[†] P. Wilairat[‡]
P. K. Dasgupta[**] Shoji Motomizu[††] D. Nacapricha[‡‡]

[*]Flow Innovation-Research for Science and Technology Laboratories, Department of Chemistry, Faculty of Science, Mahidol University

[†]Flow Innovation-Research for Science and Technology Laboratories, Department of Chemistry, Faculty of Science, Mahidol University

[‡]Flow Innovation-Research for Science and Technology Laboratories, Department of Chemistry, Faculty of Science, Mahidol University

[**]Department of Chemistry and Biochemistry, Texas Tech University

[††]Department of Chemistry, Faculty of Science, Okayama University, motomizu@cc.okayama-u.ac.jp

[‡‡]Flow Innovation-Research for Science and Technology Laboratories, Department of Chemistry, Faculty of Science, Mahidol University

# A membraneless gas diffusion unit: design and its application to determination of ethanol in liquors by spectrophotometric flow injection

**N. Choengchan[a,b*], T. Mantim[a], P. Wilairat[a], P. K. Dasgupta[c], S. Motomizu[b], and D. Nacapricha[a]**

[a]*Flow Innovation-Research for Science and Technology Laboratory, Department of Chemistry, Faculty of Science, Mahidol University, Rama 6 Road, Bangkok, 10400 Thailand*

[b] *Department of Chemistry, Faculty of Science, Okayama University, 3-1-1 Tsushimanaka, Okayama, 700-8530 Japan*

[c] *Department of Chemistry and Biochemistry, Texas Tech University, Lubbock, TX 79409-1061 USA*

## Abstract

This work presents new design of a gas diffusion unit, called 'membraneless gas diffusion (MGD) unit', which, unlike a conventional gas diffusion (GD) unit, allows selective detection of volatile compounds to be made without the need of a hydrophobic membrane. A flow injection method was developed employing the MGD unit to determine ethanol in alcoholic drinks based on the reduction of dichromate by ethanol vapor. Results clearly demonstrated that the MGD unit was suitable for determination of ethanol in beer, wine and distilled liquors. Detection limit (3S/N) of MGD unit was lower than the GD unit (GD: 0.68%, v/v; MGD: 0.27%, v/v). The MGD design makes

the system more sensitive as mass transfer is more efficient than that of GD and thus, MGD can perfectly replace membrane-based designs.

*Keywords:* Membraneless gas diffusion, Flow injection, Ethanol determination, Liquor, Spectrophotometry.

*****Corresponding author. Tel.: +66-2-201-5127; Fax: +66-2-354-7151.

*E-mail address: nchoengchan@gmail.com (N. Choengchan).*

## 1. Introduction

For most analytical procedures, at least a separation step is required for sample preparation prior to measurement. Separation or sample clean up can be performed by various means, such as liquid-liquid or liquid-solid extractions. For volatile or semi-volatile compounds, separation of these species from the matrix are conveniently performed in flow injection (FI) format [1]. Previous reports for on-line separation with subsequent detection of the volatile compounds are mostly carried out using two types of membrane-based apparatuses. The apparatuses are known as gas diffusion (GD) and analytical pervaporation (PV) units. Inside a single unit, a hydrophobic membrane is fitted for, normally, a passive transfer of a gaseous compound from one side (donor stream) to the other side of membrane (acceptor stream). In this way, selective detection of partially diffused volatile is accomplished [2]. Examples of the use of GD in flow analysis are determination of ammonia [3,4] and halogens [5-7].

PV is also a membrane-based kit for on-line separation of volatile species from donor stream to acceptor stream and is perfectly compatible with flow-based measurement. Although, the principle of PV is similar to GD, a constant volume of air

gap is typically maintained between the level of donor solution and the membrane [8]. PV does provide some advantages over GD, when it is applied to liquid suspension samples, because membrane dose not contact directly with the samples. This prolongs the lifetime of the membrane.

Although GD and PV are very useful for selective analysis of volatile compounds, they might not be so cost-effective, especially for the cases, which membrane has to be changed frequently. Also, use of the membrane can somehow reduce the sensitivity of analysis.

In this work, design and development of new gas diffusion unit is presented. The current unit does not rely on use of the hydrophobic membrane and thus the unit is named 'membraneless gas diffusion unit' (MGD unit).

We chose to develop a method for determination of ethanol in alcoholic beverages to demonstrate the potential use of our unit, since there have been quite a number of reports formerly presented use of GD and PV for alcohol analysis [9-13]. Our MGD method for ethanol is based on the reaction between ethanol and dichromate in acidic solution. Reduction of dichromate to chromium (III) with ethanol is spectrophotometrically monitored at 590 nm. The development has been carried out using FI technique for automation.

## 2. Experimental

### *2.1 Reagents and samples*

All chemicals used were of analytical reagent grade, and solutions were prepared using Milli-Q water. Working standard solutions were freshly prepared by appropriate dilution of 99.5 % (v/v) ethanol (Wako Pure Chemical Industries, Japan).

The acceptor stream of the FI system in Fig. 1 (acidic 0.03 mol $l^{-1}$ $K_2Cr_2O_7$), was prepared by dissolving 4.41 g of potassium dichromate crystal (Katayama Chemicals, Japan) in 500 ml of 1.5 mol $l^{-1}$ sulfuric acid.

Eight samples of alcoholic drinks from Thailand and Japan were examined. Ethanol contents in these samples range from 5 to 40 % (v/v). Almost all of the beverages were directly injected into the developed FI system (Fig. 1), except the Thai whiskies. These whiskies were diluted with water (4 times) prior injection.

*2.2 Membraneless gas diffusion unit*

Schematic diagram, which presents construction of the MGD unit, is shown in Fig. 2. The unit is made of Perspex. In practical use, the unit was covered with lid and was held together by using bolts.

*2.3 The MGD-FI system*

The MGD-FI system for determination of ethanol is depicted in Fig. 1. The system was automatically controlled by LabView 7.1™. The peristaltic pump (Cavro, USA) was used with Tygon™ pump tubes (0.79 mm i.d.). A six-port injection valve (SNK, Japan), with a 300-µl injection loop, was employed for injecting standard and sample solutions. A Soma S-3250 spectrophotometer (Japan), equipped with a 10-mm flow-through cell and a FIA monitor/data processing apparatus (F.I.A Instruments, Japan), were respectively utilized for the detection and recording of signals. The manifold in Fig. 1 was constructed by using 0.5 mm i.d. PTFE tubing.

*2.4 GD-FI system for validation*

Another FI system, with GD unit, was employed for validation. The FI manifold was set up similarly to the MGD-FI system drawn in Fig. 1, by replacing the MGD unit with the GD unit. The membrane of the GD unit was a tubular shape made of microporous PTFE (2.0 μm pore size and 20 cm long). Assembly of the unit has been previously described [3,4].

*2.5 Comparison of efficiency with a conventional GD unit*

In this work, we compared the mass transfer of the MGD unit with a GD unit. The GD unit is the same unit that was used for the experiment described in 2.4. The manifold in Fig. 1 was used for this comparison. The GD unit was placed in the FI system at the same place where the MGD unit is depicted. For the tubular GD unit, the effective area (membrane surface) that contributed to the transfer of ethanol was 400 $mm^2$ (2 mm i.d. x 200 mm length). The MGD unit (Fig. 2) had the transfer area of 100 $mm^2$ (2 mm width x 50 mm length).

In this work, we determined the mass transfer efficiencies (GD and MGD) for the transfer of ethanol from donor stream to acceptor stream. We describe here the procedure to evaluate the efficiency of MGD, as example. Similar procedure was used for the GD unit.

The mass transfer efficiency is defined as percentage of the signal obtained from the 'actual diffusion', compared to the signal obtained from 'maximum diffusion'. The signal of 'actual diffusion' was evaluated firstly by constantly propelling a standard ethanol solution into the donor channel ('DS' in Fig. 1). This was done while dichromate solution was pumped into the manifold as normally operated in the ethanol analysis. After the signal reached its plateau, 25 ml of the waste at detector was

collected. This solution was shaken and left to stand for 30 min at 25 °C, for the complete reaction of ethanol with dichromate. This 25-ml aliquot was fed through the 'AS' channel in Fig. 1, while 'DS' channel was continuously fed with water. The obtained constant reading is the signal that represents 'actual diffusion'.

The signal called 'maximum diffusion' is defined as the representative signal for a condition that gives 100% diffusion of ethanol. Experimentally, the signal was obtained by propelling a mixture of ethanol and potassium dichromate to the 'AS' channel in Fig. 1. This mixture was prepared in a way to contain equal amounts all the reactants that would present in the 25-ml aliquot when actual diffusion was examined. The mixture was mixed off-line and was kept standing at 25 °C for 30 min before feeding to the 'AS' channel. At the same time, water was fed as the 'DS' stream. The stable reading of absorbance is referred to as the signal at the 'maximum diffusion'.

## 3. Results and discussion

### *3.1 Design of the membraneless unit*

The membraneless gas diffusion unit is illustrated in Fig. 2. The unit contains two parallel channels inside a closed module for which the diffusion of gas takes place between both open channels. These grooves are used as donor and acceptor channels. The unit was designed so that both grooves are separated by the 2 mm-thick barrier (depicted in Fig. 2 a and Fig. 2 b). The height of this barrier is made slightly lesser than the depths of the channels (Fig. 2 b). Thus any volatile or semi-volatile compounds can diffuse across this barrier, from the donor side to the acceptor side. With this configuration, separation and detection of the volatile analyte can be done selectively without use of any porous membrane.

The depth of the channels must be carefully designed. When the channel depth was shallow (3 mm), creeping of solutions across the barrier was observed. This flooding problem was solved by using the unit with deeper channels (6 mm). As we did this, we also increased the volume of the headspace and this lowered the sensitivity. Nevertheless, we noticed that the sensitivity (for 50-mm channel length) was lowered only by 11 %, when the 6-mm channel depth was used, compared with the 3-mm depth. Thus, the unit with 6-mm depth was selected for further experiments.

### *3.2 Optimal conditions of the MGD-FI system*

Some FI parameters were investigated by means of univariate approach. Table 1 shows the range over which the parameters were studied and their optimal values. The influence of these parameters on the analytical signal or sensitivity is discussed in the following sections.

#### *3.2.1 Effect of chemical compositions of the acceptor stream*

The acceptor solution ('AS' in Fig. 1), contained potassium dichromate in sulfuric acid solution. The signal was greater with increasing concentrations of potassium dichromate and sulfuric acid. To compromise between the sensitivity and the chemical expense, the concentrations of 0.03 and 1.5 mol $l^{-1}$, were selected (Table 1) for potassium dichromate and sulfuric acid, respectively.

#### *3.2.2 Flow rate*

Flow rates of the donor and the acceptor stream are one of the most important parameters. Flow rates at the *'inlet'* and the *'outlet'* of donor and acceptor streams must be set equally, to avoid flooding of the MGD channels. This can be done by using the

same set of pump tubes for propelling the solutions into and out from the MGD unit (Fig. 1). Operation at high flow rate could increase the throughput. However, this resulted in the decrease in the sensitivity of the ethanol detection. For this work, the flow rate of 0.3 ml $min^{-1}$ was chosen (Table 1) for all the FI streams in Fig. 1.

*3.2.3 Sample size and sensitivity*

When the injection volume of samples was increased, the sensitivity was improved. However, the sample throughput was decreased with increasing volume of samples. An injection volume of 300 µl was selected as a concession between the sensitivity and sample throughput.

*3.2.4 Influence of channel length for MGD*

Effect of channel length on the sensitivity and the analysis time is shown in Fig. 3. As the channel length was increased from 25 mm to 100 mm, the absorbance reading was increased by 54 %, and little difference in the sensitivity was observed between 50 mm- and 100 mm-channel lengths. As a compromising, between the sensitivity and the throughput, the 50 mm length was chosen.

*3.2.5 Influence of temperature*

The temperature of donor and acceptor streams can affect considerably both the evaporation of ethanol and its diffusion in the MGD unit. Higher temperature increases the vapor pressure of the analyte in the headspace area inside the unit and accelerates the diffusion of the gas.

In this work, the influence of temperature was investigated by placing the MGD unit inside a hot-dried air box (Model GAS DIF, TCI, Japan). The temperature was

varied from room temperature (25 °C) to 50 °C. As expected, the sensitivity was enhanced by increasing the temperature. The average peak height at 50 °C was about 50 % greater than the average height at 25 °C. However, the sensitivity is already sufficient at 25 °C. Therefore, we decided to carry out the analysis at the room temperature, which is the most convenient.

### *3.3 Mass transfer efficiency compared to GD*

Although employment of membrane is very useful and suitable for the designs of GD and PV units, the sensitivity would have been reduced with membrane permeation. In dynamic system, like flow injection, membrane lessen the mass transfer of the gaseous species.

Comparison of the mass transfer efficiency, between the GD-FI and the MGD-FI, is shown in Fig. 4. Although the effective area of donor stream in MGD is 4 times smaller than the GD unit, the mass transfer efficiency of MGD is always greater. This result (Fig. 4) clearly demonstrated that the MGD design is more effective and give a better sensitivity than the GD design. The result also supports that for a dynamic diffusion system like this, membrane can reduce the mass transfer of gas.

### *3.4 Analytical performances and advantages*

In Table 2, the features of the MGD-FI system are shown and compared with features of a GD-FI system. With use of the MGD, we were able to detect the signal of 0.5 % (v/v) ethanol. The MGD unit gave better sensitivity and lower limit of detection than the GD unit. This is due to the more effectiveness in the mass transfer of this new unit. Reproducibility of the MGD-FI is comparable with the traditional GD-FI. For this application, the sample throughput of the MGD-FI is slightly lower than the GD-FI.

There is a major advantage of the MGD-FI method over the GD-FI. MGD does not require any membrane. Thus the MGD unit does not suffer from exposure of the hydrophobic membrane to ethanol, like in GD unit. Direct contact of the membrane with ethanol causes deterioration of the membrane due to the wetting effect.

### *3.5 Interference study*

Effect of foreign species was investigated. The examined species were inorganic mono- and divalent cations [14] and anion [15], which could be found in wine: $Li^+$ (500 mg $l^{-1}$), $Na^+$ (500 mg $l^{-1}$), $NH_4^+$ (250 mg $l^{-1}$), $Mg^{2+}$(400 mg $l^{-1}$), $Ca^{2+}$(400 mg $l^{-1}$) and $SO_3^{2-}$ (0.01 mol $l^{-1}$). Ammonia (0.5 mol $l^{-1}$) was also studied because it can be produced during brewing fermentation. Results showed that signal alteration for all species was less than 3 %. This suggests that the developed method is free from interferents in alcoholic drinks.

### *3.6 Application to liquor samples*

The developed system was applied in determination of ethanol. Results of eight alcoholic drinks are shown in Table 3. Statistical analysis (paired-*t* test [16]) showed that the data from MGD-FI are not significantly different to the data from the GD-FI ($t_{stat}$ = 1.43, $t_{critical}$ = 1.89 at 95 % confidence). The measured values also agree well with the labeled values.

The recovery test (Table 4) for the same set of samples shows that recovery ranged from 92.5 % to 109 %. These results therefore approve the validity of the MGD for quantitative analysis of ethanol in beverages.

## 4. Conclusions

We presented an innovative unit design for separation and collection of volatile compounds. The unit is a membraneless apparatus. The effectiveness of this new unit is remarkable and can be used to replace those membrane-based apparatus like GD or PV units. The developed unit provides a greater mass transfer than the GD unit. Having no membrane, MGD unit is a lower cost apparatus than GD unit. Problem from malfunction of membrane, causing by reagents, is no longer exist. Also it is easier to construct a MGD unit, as compared to flat or tubular type of GD unit.

This work demonstrates the validity of the MGD unit for quantitative analysis of ethanol. Use of the conventional GD unit often results in poor durability of the membrane causing mainly by direct contact with ethanol. Deployment of this new design provides the analysis with a better robustness.

## Acknowledgements

This work was supported by research grants given to DN (The Thailand Research Fund (TRF) and The Postgraduate Education and Research Program in Chemistry (PERCH)) and to NC (Lecturer Development Program for the Shortest Area , The Postgraduate Education and Research Program in Chemistry (PERCH) and The Royal Golden Jubilee, Ph. D. program of the TRF).

The authors would also like to thank Mr. Motonari Kobayashi, a senior technician of Okayama University, who had made several membraneless gas diffusion units.

## References

[1] J. Ruzicka, E.H. Hansen, Flow Injection Analysis, 2nd edn., Wiley, New York, 1988.

[2] G. E. Pacey and B. Karlberg, Flow Injectional Analysis: A Practical Guide, Elsevier Science Publisher, New York, 1989.

[3] K. Higuchi, S. Motomizu, A. Inoue and T. Tsuboi, Bunseki Kagaku, 48 (1999) 253.

[4] T. Tsuboi, Y. Hirano, M. Oshima and S. Motomizu, J. Flow Injection Anal., 17 (2000) 77.

[5] S. Motomizu and T. Yoden, Anal. Chim. Acta, 261 (1992) 461.

[6] D. Nacapricha, K. Uraisin, N. Ratanawimarnwong and K. Grudpan, Anal. Bioanal. Chem., 378 (2004) 816.

[7] N. Ratanawimarnwong, N. Amornthammarong, N. Choengchan, P. Chaisuwan, M. Amatatongchai, P. Wilairat, I.D. McKelvie and D. Nacapricha, Talanta, 65 (2005) 756.

[8] M.D. Luque De Castro and I. Papaefstathiou, Trends Anal. Chem., 17 (1998) 41.

[9] J. Gonzalez-Rodriguez, P. Perez-Juan and M.D. Luque de Castro, Talanta, 59 (2003) 691.

[10] T.R.L.C. Paixao, D. Corbo and M. Bertotti, Anal. Chim. Acta, 472 (2002) 123.

[11] A.O.S.S. Rangel and I.V. Toth, Anal. Chim. Acta, 416 (2000) 205.

[12] E. Mataix and M.D. Luque de Castro, Talanta, 51 (2000) 489.

[13] I.L. De Mattos and E.A.G. Zagatto, Anal. Sci., 15 (1999) 63.

[14] O. Zerbinati, F. Balduzzi, and V Dell'Oro, J. Chromatogr. A, 881 (2000) 645.

[15] T. Yao, M. Satomura and T. Nakahara, Talanta, 41 (1994) 2113.

[16] J.N. Miller and J.C. Miller, Statistics and Chemometrics for Analytical Chemistry, 4th edn, Pearson Education, Essex, 1993.

**Figures' caption**

**Fig. 1** The FI system for spectrophotometric determination of ethanol. DS: Donor stream (water), AS: Acceptor stream (0.03 mol $l^{-1}$ $K_2Cr_2O_7$ in 1.5 mol $l^{-1}$ $H_2SO_4$), MGD: Membraneless gas diffusion unit (50 mm-channel length is optimum), IV: Injection valve, P: Peristaltic pump and W: Waste.

**Fig. 2** Schematic diagram of the membraneless gas diffusion unit: (a) top view and (b) side view.

**Fig. 3** The influence of MGD's channel length on the sensitivity and the analysis time. The experiment was carried out by repetitive injections of 10 % (v/v) ethanol into the system in Fig. 1.

**Fig. 4** Mass transfer efficiencies of MGD and GD.

## List of Tables

### Table 1

Optimal conditions of the MGD-FI system for determination of ethanol.

| Parameters | Range examined | Optimal value |
|---|---|---|
| Sulfuric acid concentration (mol $l^{-1}$) | 0.5 - 2.5 | 1.5 |
| Dichromate concentration (mol $l^{-1}$) | 0.01-0.05 | 0.03 |
| Flow rate (ml $min^{-1}$) | 0.2-0.4 | 0.3 |
| Injection volume (µl) | 100-500 | 300 |
| Channel length (mm) | 25-100 | 50 |
| Temperature (°C) | 25[a], 40 and 50 | 25 [a] |

[a] room temperature

**Table 2**

Analytical characteristics of the MGD-FI, compared with the GD-FI, for the ethanol determination.

| **Analytical characteristics** | **MGD-FI** | **GD-FI** |
|---|---|---|
| Working range (% (v/v)) | 0.5 to 30 | 1 to 30 |
| Calibration equation | $Abs_{590\ nm} = 9.7 \times 10^{-3}$[% (v/v) ethanol] + 0.0014, $r^2 = 0.999$ | $Abs_{590\ nm} = 4.9 \times 10^{-3}$[% (v/v) ethanol] + 0.0019, $r^2 = 0.994$ |
| Detection limit (% (v/v): 3S/N) | 0.27 | 0.68 |
| Reproducibility (RSD, n=10 injections of 1 % (v/v)) | 0.50 | 0.55 |
| Throughput (injections $h^{-1}$) | 16 | 18 |

**Table 3**

Ethanol contents in beverages determined by the MGD-FI and GD-FI method compared with the labeled values.

| **Samples** | **%(v/v) of ethanol, n = 3** | | |
|---|---|---|---|
| | Labeled | MGD-FI | GD-FI |
| Beer | 5 | 4.12 ± 0.04 | 3.77 ± 0.08 |
| Japanese sake 1 | 10 | 10.4 ± 0.16 | 11.1 ± 0.04 |
| Japanese sake 2 | 14 | 15.6 ± 0.20 | 15.9 ± 0.11 |
| Red wine | 14 | 15.7 ± 0.09 | 16.0 ± 0.14 |
| White wine | 14 | 14.4 ± 0.07 | 15.9 ± 0.15 |
| Thai whisky 1 | 35 | 35.0 ± 0.34 | 35.5 ± 0.28 |
| Thai whisky 2 | 35 | 32.6 ± 0.46 | 36.5 ± 0.43 |
| Thai whisky 3 | 40 | 42.2 ± 0.75 | 41.2 ± 0.59 |

**Table 4**

Analytical recovery of the MGD-FI system for determination of ethanol in liquors.

| Sample | %(v/v) of ethanol , n = 3 | | | Recovery (%) |
|---|---|---|---|---|
| | Original | Added | Found | |
| Beer | 4.10 ± 0.04 | 10 | 13.7 ± 0.11 | 96.0 ± 1.1 |
| Japanese sake 1 | 10.4 ± 0.16 | 10 | 21.3 ± 0.09 | 109 ± 0.9 |
| Japanese sake 2 | 15.6 ± 0.20 | 10 | 24.8 ± 0.07 | 92.5 ± 0.7 |
| Red wine | 15.7 ± 0.09 | 10 | 25.3 ± 0.04 | 96.3 ± 0.4 |
| White wine | 14.4 ± 0.07 | 10 | 25.0 ± 0.07 | 106 ± 0.7 |
| Thai whiskeys 1 | 8.80[a] ± 0.09 | 10 | 18.6 ± 0.07 | 98.8 ± 0.7 |
| Thai whiskeys 2 | 8.20[a] ± 0.11 | 10 | 18.9 ± 0.07 | 107 ± 0.7 |
| Thai whiskeys 3 | 10.5[a] ± 0.19 | 10 | 21.1 ± 0.04 | 106 ± 0.4 |

[a] The content found in the diluted samples (4 times).

**Figures' caption**

**Fig. 1.** FI system for spectrophotometric determination of ethanol. DS, donor stream (water); AS, acceptor stream (0.03 mol L−1 $K_2Cr_2O_7$ in 1.5 mol $L^{-1}$ $H_2SO_4$); MGD, membraneless gas diffusion unit (50 mm-channel length is optimum); IV, injection valve; P, peristaltic pump and W, waste.

**Fig. 2.** Schematic diagram of the membraneless gas diffusion unit. (a) Top view; (b) Side view.

**Fig. 3**. Influence of MGD channel length on sensitivity and throughput. Experimentwas carried out by repetitive injections of 10% (v/v) ethanol into the system depicted in Fig. 1.

**Fig. 4.** Mass transfer efficiencies of MGD and GD.

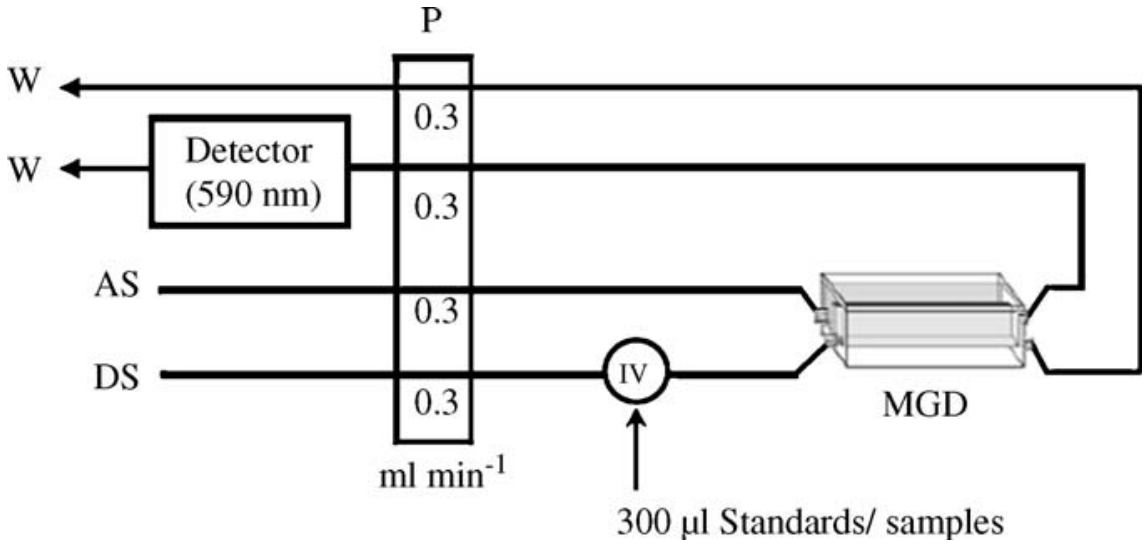
P
W
W
Detector
(590 nm)
0.3
0.3
0.3
0.3
AS
DS
IV
MGD
ml min-1
300 µl Standards/ samples

**Fig. 1**

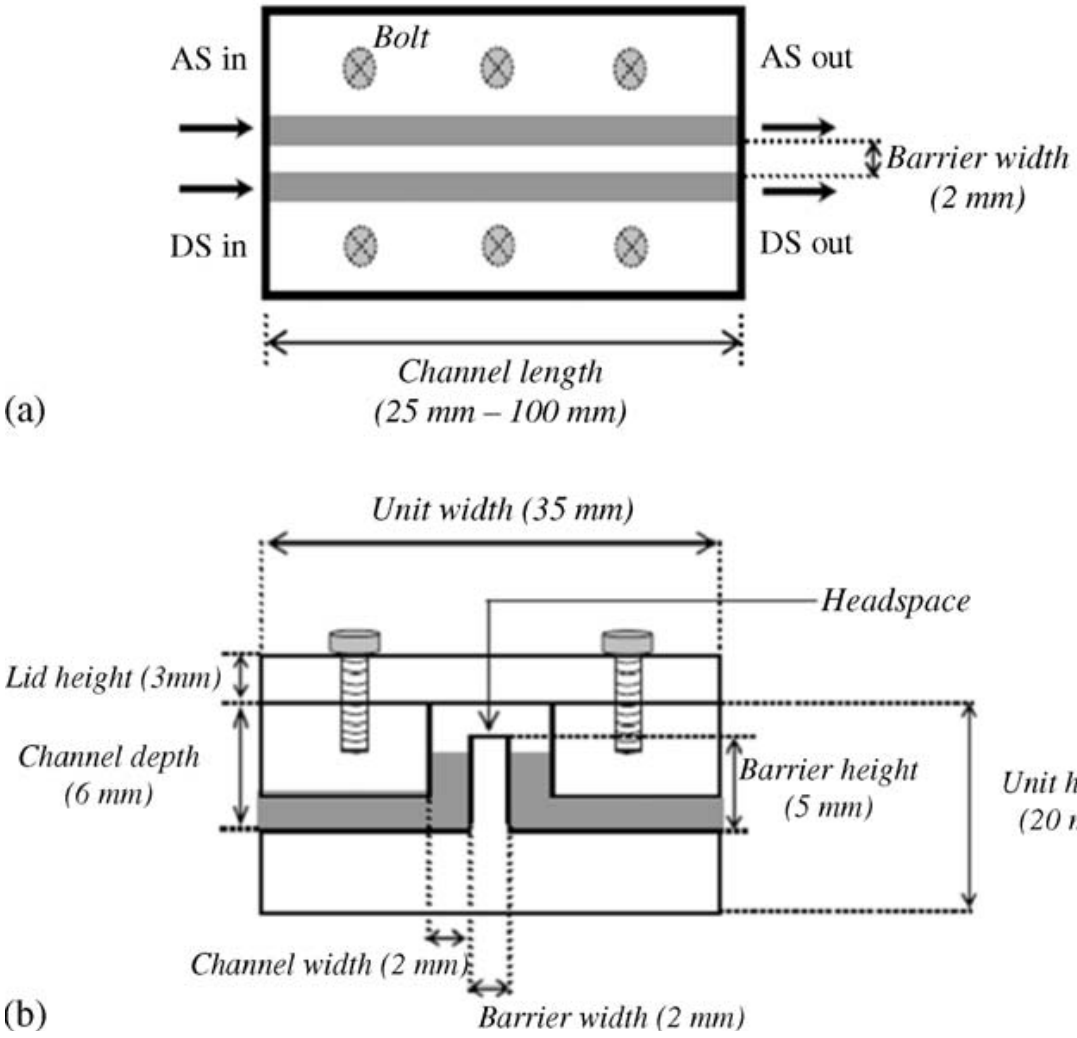
AS in
Bolt
AS out
Barrier width
(2 mm)
DS in
DS out
Channel length
(25 mm – 100 mm)
(a)
Unit width (35 mm)
Headspace
Lid height (3mm)
Channel depth
(6 mm)
Barrier height
(5 mm)
Unit height
(20 mm)
Channel width (2 mm)
Barrier width (2 mm)
(b)

**Fig. 2**

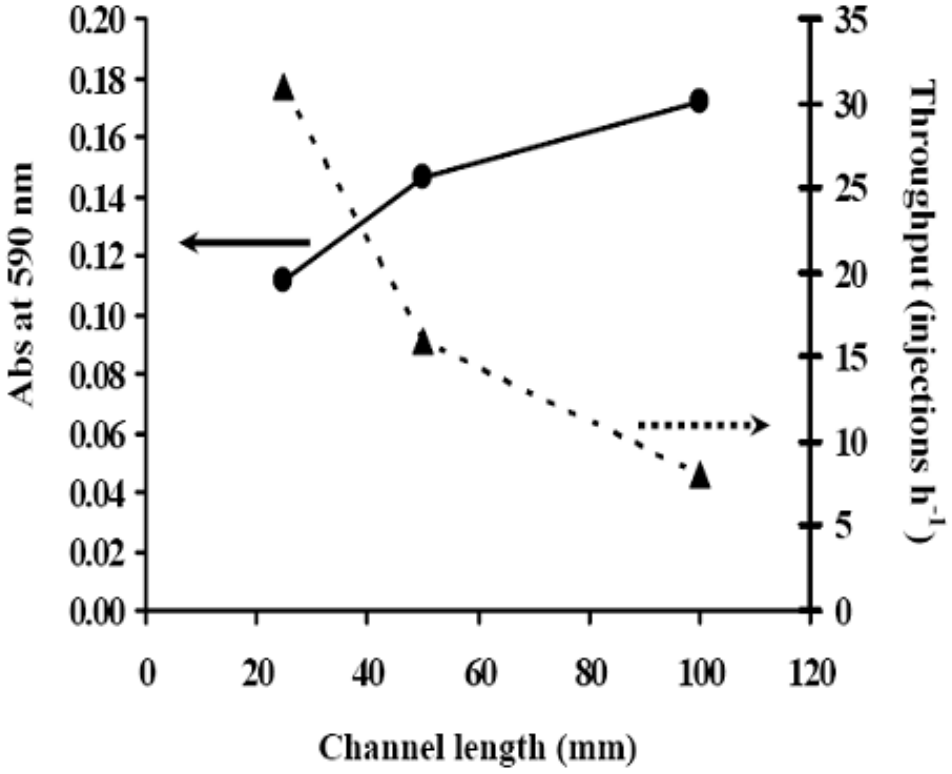
Abs at 590 nm
0.20
0.18
0.16
0.14
0.12
0.10
0.08
0.06
0.04
0.02
0.00
Throughput (injections h-1)
35
30
25
20
15
10
5
0
0
20
40
60
80
100
120
Channel length (mm)

**Fig. 3**

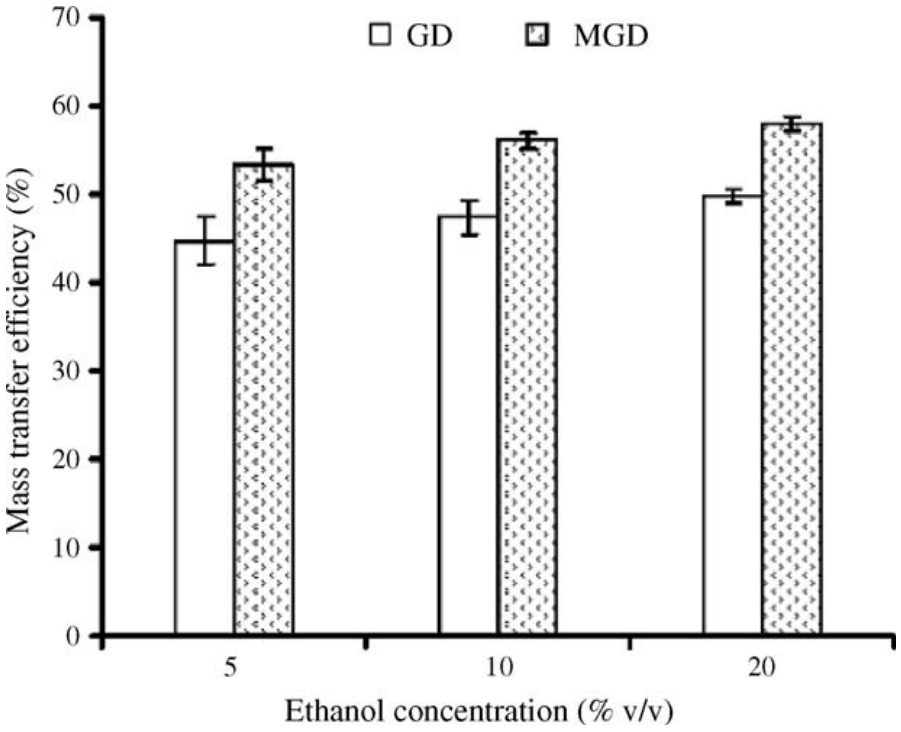
GD
MGD
Mass transfer efficiency (%)
70
60
50
40
30
20
10
0
5
10
20
Ethanol concentration (% v/v)

**Fig. 4**