§9. Development of μTAS System for Remove of Tritium and Isotope Separation

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Tritium is produced by the side reaction of $^{18}O(p,t)^{16}O$ during the production of PET-FDG([^{18}F]-2-fluoro-2-deoxy-D-glucose). Several processes for the tritium separation have been developed, e.g., distillation, chemical exchange, catalytic exchange, and electrolysis. [1] The chemical exchange utilizing microchannel chip is totally a brand-new technique and had the advantage of fast and high conversion phase-transfer synthesis exploiting the liquid-gas(liquid) interface formed in a microchannel chip. [2] Simple introduction of two phases into the microchannel provided a stable liquid-gas(liquid) interface, and the large specific interfacial area and short molecular diffusion distances had a higher conversion than those of any macroscale reaction with strong stirring. Microreactor system is superior to normal batch systems not only for a high conversion close to 100 %, but also for a fast reaction time less than a few seconds.

The liquid-gas two phases-transfer occurs according to the following exchange reaction:

$$^{1}H^{3}H(gas) + {}^{1}H_2{}^{18}O(liquid) \Leftrightarrow$$
$$^{1}H_2(gas) + {}^{1}H^{3}H^{18}O(liquid), \quad (1)$$

where we can treat HT gas rather than severely retrained HTO liquid making use of molecular tritium and isotope exchange reaction.

First, we tested D_2O liquid in eq. (2) instead of HTO liquid in eq. (1) for ease of handling and measuring of its concentration in the phase-transfer reaction.

$$^{1}H^{2}H(gas) + {}^{1}H^{2}H^{16}O(liquid) \Leftrightarrow$$
$$^{1}H_2(gas) + {}^{2}H^{2}H^{16}O(liquid), \quad (2)$$

The experiment system of a microreactor is illustrated in Fig. 1. The microreactor has depths of 90 μm and 22 μm, widths of 194 μm and 64 μm, for gas phase and liquid phase, respectively, and a liquid-gas contact length of 20 mm. The reaction was performed by introducing a D_2O liquid and a H_2 gas through the two-inlets of the microreactor under continuous flow conditions at ambient temperature. The concentration of D_2O was measured by the density meter (Anon Paar DMA 5000M, which is the most accurate density meter on the market). The accuracy is 0.000005 g/cm^3. Next, we examined the effect of the flow rate on liquid phase for the mean residence time of 0.2 or 4 sec (9.65 μl/min for 0.2 sec and 0.57 μl/min for 4 sec). In both cases, the reactions proceeded smoothly to afford the phase-transfer of deuterium. As comparison, the flow rate reduction to about 6 % is effective to increase the reaction yield of about 1 % to 3 %, which are summarized in Table. I. It is noted that the current flow rate will require a hundred minutes to a day to process one ml amount and we are planning to utilize microreactor with patterned surfaces featuring the hydrophilic and hydrophobic and fabrication of a piling-up for the next step.

Thus, the application of recent advances in microchip technology to our liquid-gas extraction by utilizing circulated liquid-gas multi phase flow in a microchannel chip without any stirring will be quite attractive and can be also applied for chemical separation of the other isotopes, such as calcium. [3]

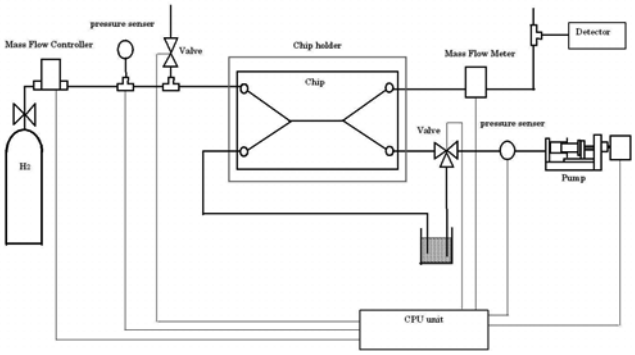

Fig. 1. Flow chart of Experimental Device. Microreactor contains deep (90 μm depth and 194 μm width) and shallow (22 μm depth and 64 μm width) channel areas for gas phase and liquid phase, respectively.

H_2(gas) flow rate (cc/min)	2.3	2.3
$D_2{}^{16}O$ flow rate (μl/min)	9.65	0.57
$D_2{}^{16}O$ density prior (g/cm^3)	1.105276	1.105276
post (g/cm^3)	1.104303	1.101427
$D_2{}^{16}O$ concentration prior (%)	99.844	99.844
post (%)	98.937	96.254

Table. I. Reaction yield and the effect of the flow-rate on liquid phase for the liquid(D_2O)-gas(H_2) two phase transfer reaction.

1) Ogata, Y. et al.: J. Radioanal. Nucl. Chem. **255** (2003) 539
2) Hisamoto, H. et al.: Chem Commun. (2001) 2662
3) Hazama, R. et al.: Proc. of 6th Rencontres du Vietnam, Gioi Publishers (2007) 383: arXiv:0710.3840.[nucl-ex]