§6. Remove of Tritium from Tritiated Water and Isotope Separation by Microchips

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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:

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

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

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 hydrogen isotopes. Now the widest variety of stable isotopes are mainly separated at electromagnetic separators and gas centrifuges. Flexible highly efficient centrifugal technology is only possible for those elements (about 20) which have gaseous compounds at room temperature. Therefore, these methods cannot meet the separation of hydrogen and calcium isotopes.

At first, we found that calcium isotopes were actually fluctuated by liquid-liquid extraction using dicyclohexano-18-crown-6 and the advantage of our chemical exchange method is verified not only for an ion separation, but also for an isotope separation. We also evaluated each contribution ratio of the field shift effect and the hyperfine splitting shift effect to the mass effect of the calcium isotopes and found the contribution of the field shift effect is small . [3] Recently chromatography experiments using benzo-18-crown-6 resin were also carried out and calcium ion selectivity and isotope effects was observed. [4] However, it was found that the absorption of calcium depends on the concentration of "additional" hydrochloric acid in solution and its observed separation factor is about one order smaller than the direct liquid-liquid extraction. These indications are promising towards the tritium separation by the liquid-gas phase transfer system with the help of a microchannel chemistry.

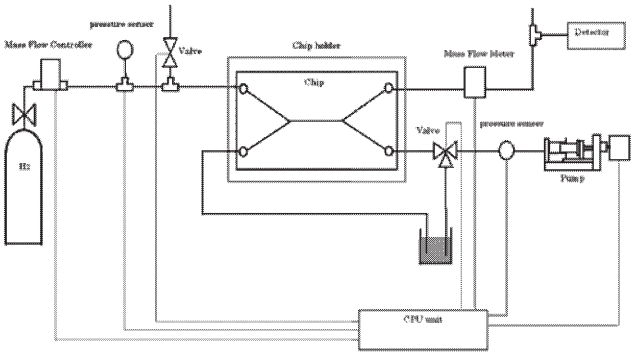

Fig. 1. Flow chart of Experimental Device.

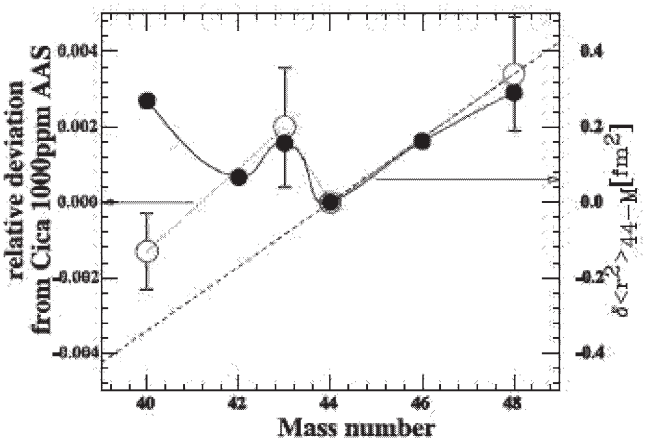

Fig. 2. Observed calcium isotope effects (left axis) and experimental values for the mean square nuclear charge radii (right axis) of the calcium isotopes relative to ^{44}Ca. Filled (line) and open (dotted line) points correspond to nuclear charge radii and our obtained data, respectively. The dashed line is the expected mass effect, which shows a linear relationship with mass number.

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.: arXiv:0710.3840.[nucl-ex]
4) Hayasaka, K et al.: Prog. Nucl. Energy **50** (2008) 510