A Routine Method for $^{11}$CO Production

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1. Introduction

The development of positron tomography and ultra-compact cyclotrons has been accelerating the studies on automated and efficient productions of radiopharmaceuticals labeled with short-lived positron emitters ($^{11}$C, $^{13}$N, $^{15}$O and $^{18}$F) for medical diagnostic use in hospitals. Of these radioisotopes, $^{11}$C is the most potential for synthesis of various kinds of organic compounds. $^{11}$CO is one of the simplest $^{11}$C-compounds and used mainly for blood flow diagnosis, especially in brain, in combination with $^{13}$NH$_3$ and $^{18}$F-deoxyglucose. It is also useful as a precursor for synthesis of $^{11}$COC$_1$.

In general, $^{11}$CO is produced by reduction of $^{11}$CO$_2$ on zinc heated at 390°C. This method gives a reduction yield of 50~80% depending on the amount of O$_2$ contained in the nitrogen gas target and on the extent of the oxidation of zinc surface. A prolonged use of zinc tends to decrease the yield, and at last to result in the complete oxidation of zinc surface. Therefore it is recommended that fresh zinc should be used on every run.

As an alternative method, $^{11}$CO is produced by reduction of $^{11}$CO$_2$ on heated charcoal at 900°C. The latter method, however, has been used only with the boric oxide target and the He sweep gas containing 1~2% of carrier CO, and no reports have been made on the $^{11}$CO production on charcoal with the N$_2$ gas target containing no carrier CO.

In this note, we present that a convenient method of the $^{11}$CO production on charcoal, instead of zinc, is suitable for routine $^{11}$CO production, along with the optimal reduction conditions.

2. Methods

The irradiation was carried out by using 10 MeV protons from the ultra-compact cyclotron named “Baby Cyclotron”, BC 107 model, made by Japan Steel Works. A current of 4 μA was used with the incident energy of 9.2 MeV after passing through two 30 μm Ti windows.

The radioactive gas production system especially developed for Baby Cyclotron is designed for the automated production of $^{11}$CO, $^{11}$CO$_2$, $^{13}$NN, $^{15}$OO, C$^{15}$O and C$^{15}$OO. The outline of the system is shown in Fig. 1. The conteneous flow method of the target N$_2$ gas was used with a target pressure of 3.5 kg/cm$^2$ and a flow rate of 20~300 ml/min.

Activated charcoal (for chromatography, Wako Pure Chem. Ind. Co.) was used as a reduction reagent in a quartz tube (5.0 mm ID. and 15 cm length). The reduction temperature was varied in the range of 500~1000°C.

The activity of the gas flow after passing through the reduction tube was monitored with the isotope calibrator, and the value was compared with that of the direct gas flow from the
target tube for examining a loss of $^{11}$C in the heated charcoal.

Quantitative analysis was carried out by radio-gas-chromatography on Porapak Q for $^{11}$CO and $^{11}$CO$_2$, and carrier CO was calibrated on Molecular Sieve 5A.

### 3. Results and Discussion

The retention of $^{11}$C by the charcoal was not observed at any reaction temperature. As more than 99% of the total $^{11}$C was found as $^{11}$CO$_2$ in the direct gas flow from the target tube, the ratio of $^{11}$CO per $^{11}$CO$_2$ determined by GC analysis in the gas flow after passing through the charcoal was thought to represent the conversion yield of $^{11}$CO$_2$ to $^{11}$CO:

$$^{11}{\text{CO}}_2 + \text{C} \rightarrow ^{11}\text{CO} + \text{CO}.$$  

Figure 2 shows that the relationships between the $^{11}$CO yield and the flow rate of the target gas at the several reaction temperatures. The results indicate that the reaction temperature is critical for reduction. At 1000°C, $^{11}$CO was obtained in almost 100% of the conversion yield at any flow rate. As the flow rate can be interpreted as a reaction time between $^{11}$CO$_2$ and charcoal, it is expected that an increase of the
reaction time, that is, an increase of the amount of charcoal causes an increase of the yield. Therefore, the reaction temperature of 900°C is practical for ¹¹CO production as previously reported if a sufficient amount of charcoal is used at a flow rate of below 100 ml/min.

The contaminant O₂ in the target gas is also converted to CO. The amount of CO after passing through the charcoal is estimated to be at least twice that of O₂ initially present in the target gas. The results obtained by GC analysis show that the maximum amount of carrier CO was about 200 ppm, whereas it was below 20 ppm in the case of using zinc*. For decreasing the amount of inactive CO, it is recommended that a high pure nitrogen gas should be used as the target and a contamination of O₂ from outside should be carefully prevented. It is also recommended that a sodalime absorber should be still used at the furnace output to remove a trace of unconverted ¹¹CO₂ even at 1000°C⁶.

Charcoal has a much longer life than zinc as a loss of charcoal by oxidation is negligible and it keeps effective for the reduction at a prolonged use. This reduction method on charcoal is much suitable for efficient and routine production of ¹¹CO with ultra-compact cyclotrons in hospitals.

4. Acknowledgement

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References
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* Single inhalation of CO with this concentration (200 ppm) could not give any effect on human. Cf. Lethal concentration for mouse is 200 ppm (8 hours) and threshold limit value of working area is 50 ppm (ACGIH-1978).