Routine Method of 77Kr Production

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III. 7 Routine method of ^{77}Kr production

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Among the krypton isotopes, ^{77}Kr has some excellent characteristics. It has a rather short half-life(74.7 min) and a high positron branching ratio(~80 %). It is used for regional cerebral blood flow measurement by positron tomography.[1] The $^{79}Br(p,3n)^{77}Kr$ reaction on natural bromine seems suitable for the routine production of ^{77}Kr.[2] The separation of ^{77}Kr from a target, however, may be not so convenient when a pellet target of NaBr is used as previously reported.[3] ^{77}Kr produced in a matrix of the pellet is recoverd by melting it in a quartz tube. As an alternative method, that of using an aqueous solution of NaBr as the target has been investigated since it was expected that this method gives a lower yield but makes the seperation easier. The present report describes the ^{77}Kr production method suitable for its routine medical use.

The irradiations were carried out with 38 MeV protons by varying the current and the thickness of the target at the cyclotrons of Tohoku University and National Institute of Radiological Sciences. The concentration of NaBr in water was selected as 40 wt % because the solvent water might be decreased by radiation decomposition or evaporation resulting in the deposite of NaBr in the target vessel during the irradiation when a saturated solution(about 48 wt %) was used. In general, when a thin liquid target is irradiated with a high current of charged particles, a cavity is born in a beam passage by local evaporation or evolved gases produced by radiation reactions, and the decrease in the effective current is observed. The target vessel made of titanium was specially designed to suppress the above effect. Its design is shown in fig. 1. The net volume of the target was about 30 ml but that of the beam passage about only 4 ml(30 mm of diameter and 5.0 mm of thickness) so that it could be expected that when a cavity was born in the beam passage the surrounding target filled it swiftly. Figure 2 shows the flow chart of the production system. The gas transfer tube was well-swept with a He current before the irradiation because ^{11}C, ^{13}N and ^{15}O simultaneously produced by the reactions of $^{16}O(p,3p3n)^{11}C$, $^{16}(p,\alpha)^{13}N$ and $^{16}O(p,pn)^{15}O$ might contaminate ^{77}Kr by taking the chemical forms of $^{11}CO_2$, ^{13}NN and ^{15}OO, respectively, if the air was present in the system. During the irradiation, the solenoid valves of EV-2 and EV-3 were opened, and after the irradiation the ^{77}Kr produced was carried from the target under a He current with a controlled flow rate(usually 30 ml/min) and collected in a baloon. The yield and radionuclidic purity were measured with a Ge(li) or pure Ge detector.

The separation of the ^{77}Kr from the aqueous target could be performed simply by sweeping it under a He current through the target vessel. As seen in fig. 3, it was collected in 150 ml of He within 5 min after the irradiation. No contaminant derived from the water was observed on the decay curve of the collected radioactive gas.

The threshold energy of the $^{79}Br(p,3n)^{77}Kr$ reaction was calculated to be 23.0 MeV corresponding to a degraded energy after passing through the target of 6.5 mm thickness. In fig. 4, a decrease in the yield of ^{77}Kr is observed at the thickness of 5 mm though it is constant over that of 6 mm. Figure 5 shows the effect of the proton intensity on the yield. Over the current of 5 μA, the effect of cavitation is remarkable as the yield is reduced to about 80 % of the expected yield with a 10 μA-irradiation. From this result, no definite conclusion can be drawn on the usefulness of the present target vessel design as no comparable data is available.

When natural bromine is used as the target instead of enriched ^{79}Br, the contamination of ^{79}Kr is inevitable owing to the simultaneous reaction of the $^{81}Br(p,3n)^{79}Kr$. It is also produced by the $^{79}Br(p,n)^{79}Kr$ reaction when a thick target is used. Figure 6 shows the radionuclidic purity of ^{77}Kr as a function of the target thickness. At the thickness of 5 mm, the purity was 92 % and this value agrees with that of the literature which is obtained with a thinner target ($34.8 \leqq E_p \leqq 40$ MeV)[2], showing that the ^{79}Kr produced by the $^{79}Br(p,n)^{79}Kr$ reaction has no contribution to the purity. ^{76}Kr produced by the $^{79}Br(p,4n)^{76}Kr$ reaction was not observed on the gamma ray spectrum obtained with a pure Ge detector as shown in fig. 7.

From the results obtained, the optimal target thickness seems to be 5~6 mm on the basis of the yield and purity. When the aqueous NaBr solution of 40 wt% with 5 mm thickness is irradiated by 38 MeV protons with a 5 μA-20 min irradiation, about 20 mCi of ^{77}Kr can be obatined 5 min after the irradiation. This value is thought to be sufficient for medical use. In addition, the target can be used for a successive production. It can be concluded that the method of using the aqueous NaBr solution as the target is very suitable for the routine production of ^{77}Kr.

References

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3) Houdayer, A. J., Meyer, E. and Yamamoto Y. L., Int. J. Nucl. Med. and Biology, 4 (1977) 83.

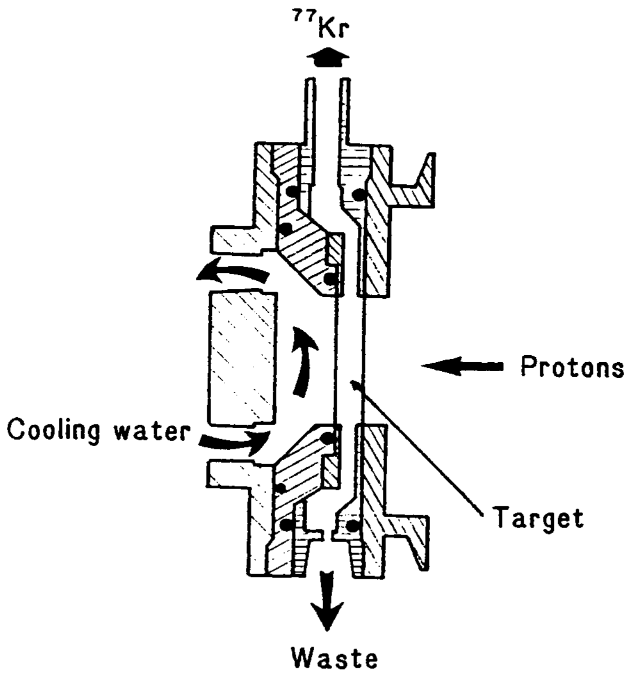

Fig. 1. Sectional side view of the target vessel.

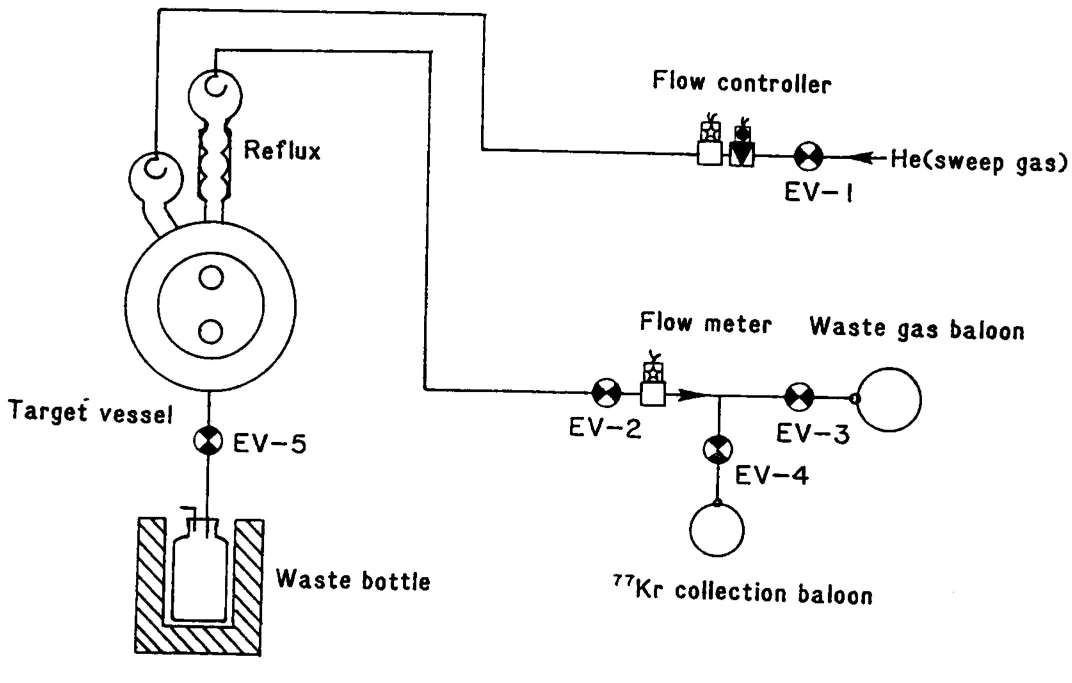

Fig. 2. Flow chart of the ^{77}Kr production system.

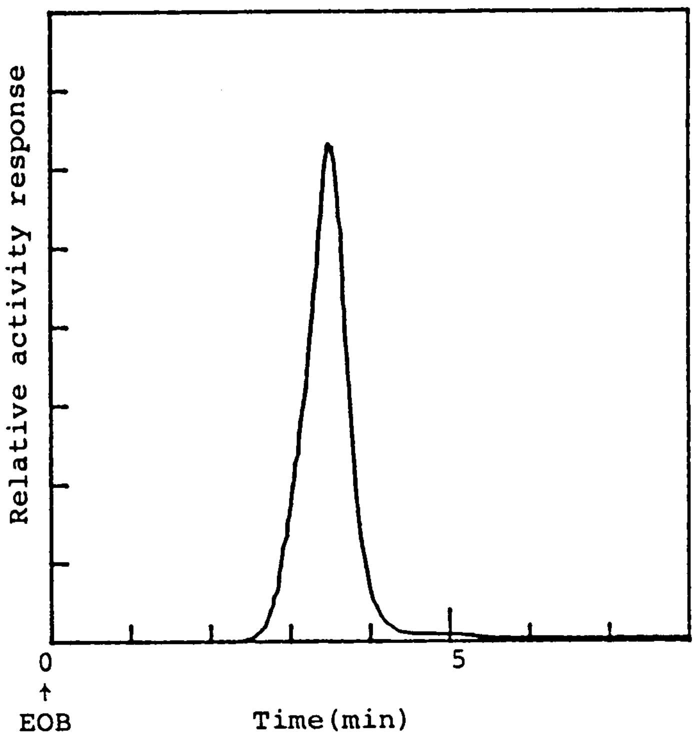

Fig. 3. Typical ^{77}Kr elution curve obtained by monitoring radioactive gas flow in the transfer tube.
sweep gas ; He, 30 ml/min

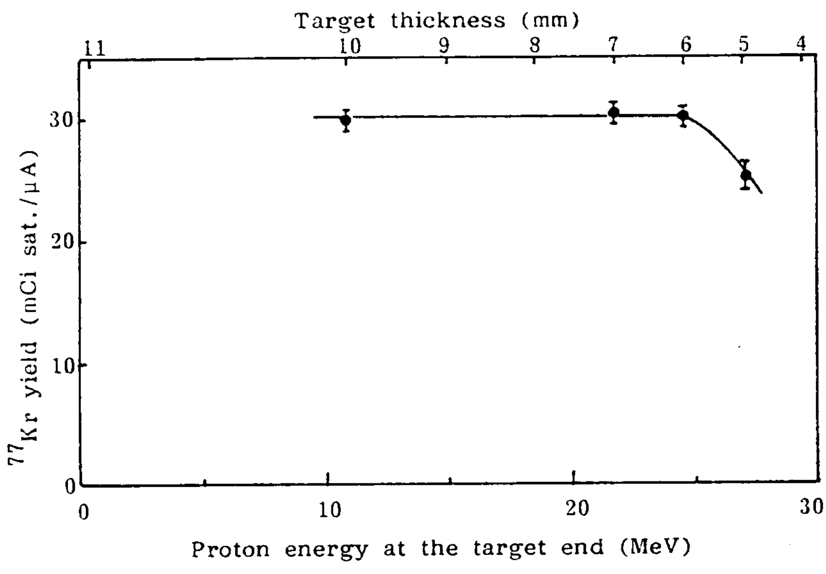

Fig. 4. Correlation between the ^{77}Kr yield and the target thickness.

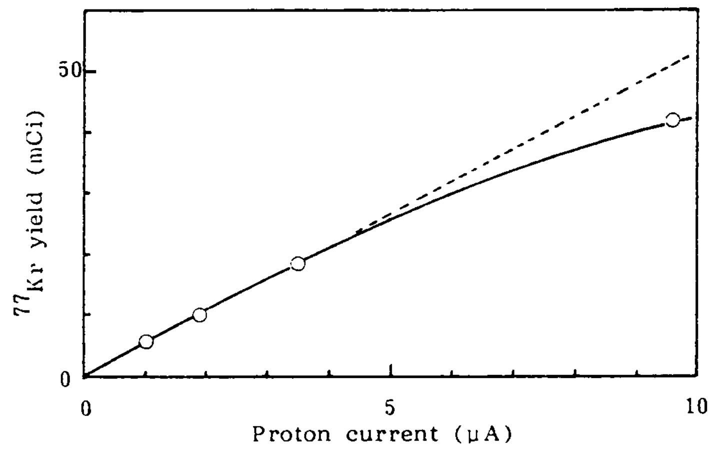

Fig. 5. Effect of the proton intensity on the ^{77}Kr yield.

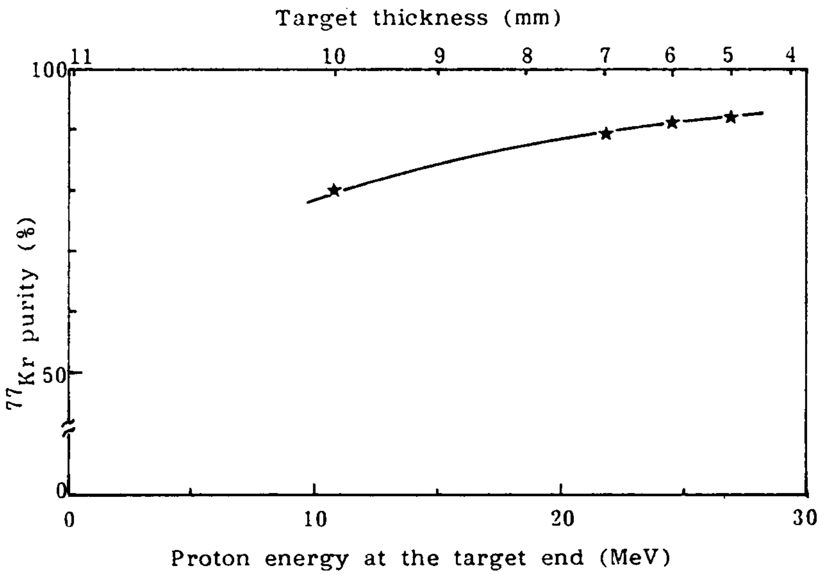

Fig. 6. Correlation between the ^{77}Kr purity and the target thickness.
irradiation : 1 μA, 20 min

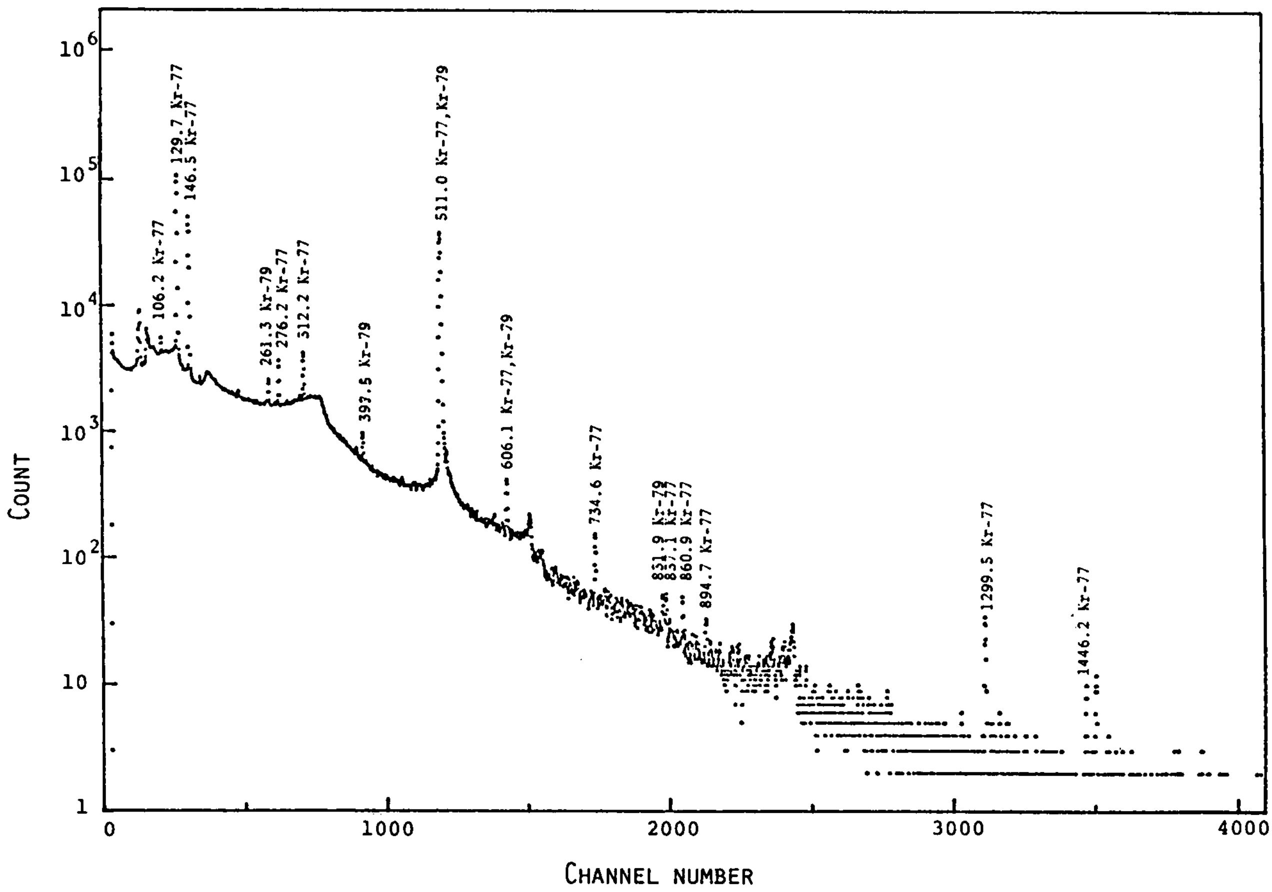

Fig. 7. Gamma ray spectrum of the collected radioactive gas obtained with a pure Ge detector 5 min after the irradiation.