TRITIUM IN $[^{15}\text{O}]$ WATER, ITS IDENTIFICATION AND METHOD OF REMOVAL

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Oxygen-15 labeled water ($[^{15}\text{O}]$ water) is a widely used radiopharmaceutical for PET imaging. The nuclear reaction $^{14}\text{N}(d,n)^{15}\text{O}$, which is frequently employed for the production of $^{15}\text{O}$ for $[^{15}\text{O}]$ water, is known to accompany another reaction $^{14}\text{N}(d,t)^{13}\text{N}$ (1). This leads to the possibility that $[^{15}\text{O}]$ water containing $^3\text{H}$. The presence or absence of $^3\text{H}$ in $[^{15}\text{O}]$ water has never drawn attention of PET chemists or been investigated in detail, probably because its amount, if any, is considered too small to affect the radiation dosimetry. In those countries where radioactive waste is treated differently depending on the half life, however, the amount of $^3\text{H}$ in $[^{15}\text{O}]$ water could be a matter of consequence. In Japan, when the $[^{15}\text{O}]$ water synthesized in some PET centers with deuteron irradiation was examined after it had decayed out according to the proposed new regulation on radioactive waste, activity above background level was detected by liquid scintillation counting (2). The present investigation was undertaken to identify the long-lived radionuclide and its chemical forms in $[^{15}\text{O}]$ water and to develop a method of its removal to facilitate the radioactive waste disposal.

Oxygen-15 was generated by bombarding a nitrogen target with 8.6 MeV (energy at in target) deuterons by $^{14}\text{N}(d,n)^{15}\text{O}$ nuclear reaction using an AVF compact cyclotron (74 cm dia.; CYPRIS 370, Sumitomo Heavy Industries). The target chamber was made of aluminum and magnesium (97:3), and the window foil was made of 600µm thick aluminum. Under the irradiation with a beam current of 15µA, the target gas (N2:O2=99.5:0.5) was continuously supplied to the chamber at a flow rate of 500mL/min. Generated oxygen-15 in nitrogen and oxygen gas (99.5:0.5) together with 5mL/min of hydrogen gas was passed over a heated (150°C) palladium black catalyst column for one minute, and produced $[^{15}\text{O}]$ water vapor was bubbled and trapped in 10 mL of sterile 0.9% saline.

![Diagram of oxygen-15 production process](image-url)
Comparison of energy spectrum in multichannel liquid scintillation counter between decayed \[^{15}\text{O}\]water (10 mL 0.9% saline) and \[^{3}\text{H}\]toluene, as a tritium standard, indicated that the long-lived nuclide in \[^{15}\text{O}\]water was tritium. When the activity in sufficiently decayed \[^{15}\text{O}\]water was repeatedly measured for 12 months, it decreased at a rate equal to that of \[^{3}\text{H}\]toluene. The 8.6 MeV deuteron irradiation with a 15 µA beam for 1 min resulted in the yield of 1.656±0.085 GBq for \[^{15}\text{O}\]water together with 5.708±0.423 Bq (342.5±25.4 dpm) of tritium. The radiation exposure to the human body by the tritium was estimated to be negligible compared to that by \[^{15}\text{O}\].

Table 1. Tritium and \[^{15}\text{O}\] radioactivity in the \[^{15}\text{O}\]water synthesized from the target gas after various treatments.

<table>
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<th>[^{3}\text{H}]</th>
<th>[^{15}\text{O}]</th>
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<tbody>
<tr>
<td>Non-treated</td>
<td>5.708±0.423 Bq (100%)</td>
<td>1.656±0.085 GBq (100%)</td>
</tr>
<tr>
<td>Molecular Sieves 4Å</td>
<td>4.195±0.327 Bq (73.5%)</td>
<td>1.414±0.105 GBq (85.4%)</td>
</tr>
<tr>
<td>CaCl(_2)</td>
<td>5.930±0.833Bq (104%)</td>
<td>1.527±0.007 GBq (92.2%)</td>
</tr>
<tr>
<td>Heated Pd, then CaCl(_2)</td>
<td>Not detected (0%)</td>
<td>1.487±0.104 GBq (89.8%)</td>
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When the target gas was passed over a Molecular Sieves 4Å column or a CaCl\(_2\) column (1 cm diam. x 5cm) before being reacted with hydrogen gas in a heated palladium catalyst (1 cm dia. x 1 cm), the tritium radioactivity was reduced only slightly (Table 1). This suggested that most of the tritium was not in the form of H\(_2\)O, NH\(_3\) or HNO\(_3\). When the target gas was passed over a heated palladium catalyst followed by a calcium chloride column and then passed over a heated palladium catalyst together with hydrogen gas (Fig. 1), tritium radioactivity in the \[^{15}\text{O}\]water was completely removed without serious loss on the yield of \[^{15}\text{O}\]water (Table 1). We considered that the tritium labeled compounds in unknown chemical species are completely oxidized, probably to \[^{3}\text{H}\]H\(_2\)O, on the heated palladium catalyst by relative excess O\(_2\) (0.5%) and trapped on the CaCl\(_2\) column. On the other hand, loss of \[^{15}\text{O}\] radioactivity is minimum under this condition, since the amount of hydrogen in the target gas is very low compared to oxygen (Fig. 1). Then the \[^{15}\text{O}\]O\(_2\) containing gas can be passed together with hydrogen gas over a heated palladium catalyst column by the usual procedure and converted to \[^{15}\text{O}\]H\(_2\)O. This provided a practical method of removing the tritium from \[^{15}\text{O}\]water.

Further experiments were performed to identify the exact chemical form of tritium labeled compounds. 1) The tritium labeled compounds were oxidized by O\(_2\) on heated palladium catalyst and trapped on a CaCl\(_2\) column. 2) When the target gas containing tritium labeled compounds was bubbled in water, only ~18% of the total radioactivity was trapped. 3) When the concentration of molecular hydrogen in the target gas increased from 1 to 10%, the tritium radioactivity in \[^{15}\text{O}\]water was decreased by 15.8±3.0%. These observations lead to a speculation of molecular hydrogen being a major chemical form of tritium labeled compounds from the target.

Because tritium was not found in \[^{15}\text{O}\]water produced by \(^{15}\text{N}(p,n)^{16}\text{O}\) nuclear reaction (data not shown), it is speculated that tritium is generated by a reaction via deuteron related nuclear reaction. Possible nuclear reactions that generate tritium include \(^{14}\text{N}(d,t\text{ or }d,n)^{13}\text{N}\) and D(d,p)T. Since the cross-section for D(d,p)T is very small, its contribution to tritium production is estimated to be only 1%. Therefore, tritium is produced mainly by the \(^{14}\text{N}(d,t\text{ or }d,n)^{13}\text{N}\) reaction (2). Decelerating the deuteron energy below the threshold (5.1 MeV) for the \(^{14}\text{N}(d,t\text{ or }d,n)^{13}\text{N}\) reaction may also be effective to prevent the tritium formation.

In conclusion, when \(^{15}\text{O}\) is produced by \(^{14}\text{N}(d,n)^{15}\text{O}\), \(^{3}\text{H}\) is generated by \(^{14}\text{N}(d,t\text{ or }d,n)^{13}\text{N}\) and is present in \[^{15}\text{O}\]water thus synthesized. We have developed a method of removing it to facilitate the radioactive waste disposal.
REFERENCES


DISCUSSION

Syed Qaim - Paper #2 where you talk of tritium production, actually that work got motivated. We published a very detailed paper in Radiochimica Acta in Dec., 1997, where we have looked for all of the products which are formed and the interactions of deuterons on nitrogen. We predicted from Q-value the full excitation function as a measure of the DT reaction given Nitrogen-13 and some other low yield reactions also. So I think it is also very interesting nuclear chemistry which one can do at these low energies also. I was talking to Dr. Hiro and he said he said the reverse girth though the activity measurement and nitrogen 13 measurement and tritium measurement are concordant. But I think it is very interesting.

Syed Qaim - I think this proposal of 1-2 minute irradiation will not work in many cases. With water production yes, but for other products there will not be enough activity. In our laboratory we irradiate for 5 minutes.

T. Ido - I agree. So then I can propose to use the catalyst method.

Syed Qaim - Could it be done centrally for example, so that water is collected and then centrally treated. T. Ido - Yes, That's also where I find some advantage. Even using bheV deuterons, there already may be some tritium due to old tubings. We tried the jet flow and then the collective bubbling through the water. We have some experience with the helium flow at 200 milli liter per minute and then it already is absorbed in tritium, So we have experience. It's a helium flow of 200 mL per minute. In five days any absorbed tritium can be removed. The other way is washing by water, followed by complete drying, otherwise there is some C-150. In that case one has to change the tubing to the new target.