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BERKELEY, CALIFORNIA

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ABSTRACT

This report describes a method that can be used to produce milliliter amounts of tritiated water from tritium gas. The report emphasizes safety precautions as part of the method, which involves oxidation of tritium gas over hot copper(II) oxide in a sealed glass system.

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Introduction

Papers reporting methods of synthesizing tritiated water are meagre^{1, 2} and fail to include precautions that are necessary to insure the safe handling of tritium. This report grew from a need of tritiated water which was to be used in following the path of hydrogen from water in photosynthesis in green algae. To meet this need it was necessary to design a method of producing tritiated water of high specific activity (5-20 curies/ml) from sealed ampoules of tritium gas. The method described in this report is so designed that the procedure is workable and yet the possibilities of exposing operating personnel to tritium are at a minimum.

Apparatus

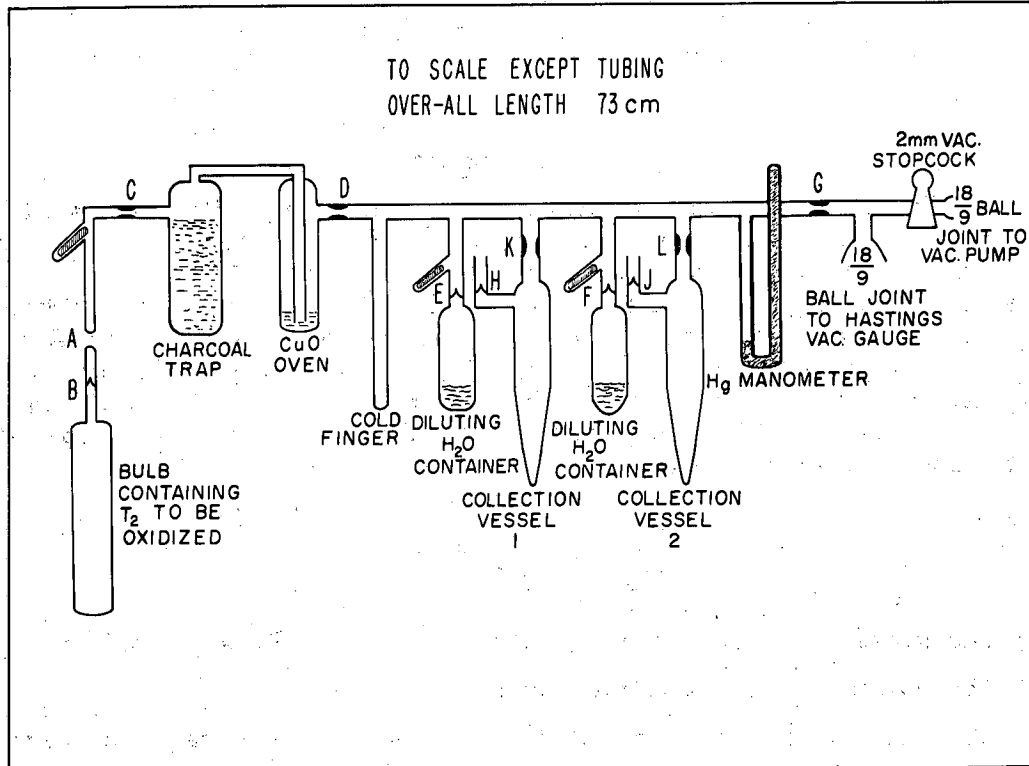
The apparatus consists of:

- (a) the glass system pictured and described in Figs. 1 and 3;
- (b) the vacuum pump and vacuum gauge;
- (c) the resistance oven, controlled by variac and potentiometer;
- (d) the Berkeley gloved box, vented to a manifold (slight negative pressure);
- (e) the vacuum flasks; liquid nitrogen; oxy-methane torch; and heat lamp; and
- (f) the two tritium "sniffers" (portable tritium air monitors).

Procedure

1. Preparation of Diluting H₂O Containers

The diluting H₂O containers are filled before they are attached to the line as follows (see Fig. 2). After an excess of H₂O has been admitted, a water aspirator is fitted on the 14/35 standard-taper joint and the excess is evaporated out by use of a heat lamp. (This assures complete deaeration of the H₂O.) The stopcock is closed and the container is moved to a vacuum line. The H₂O is then frozen in a liquid nitrogen bath and the stopcock opened. When it is clear that there is no leak in the breakoff seal, the container is sealed at M and attached to the tritium line.



MU-14555

Fig. 1. Tritium oxidation line.

Collection vessels are adapted 12-ml. centrifuge tubes graduated in 0.1 ml.

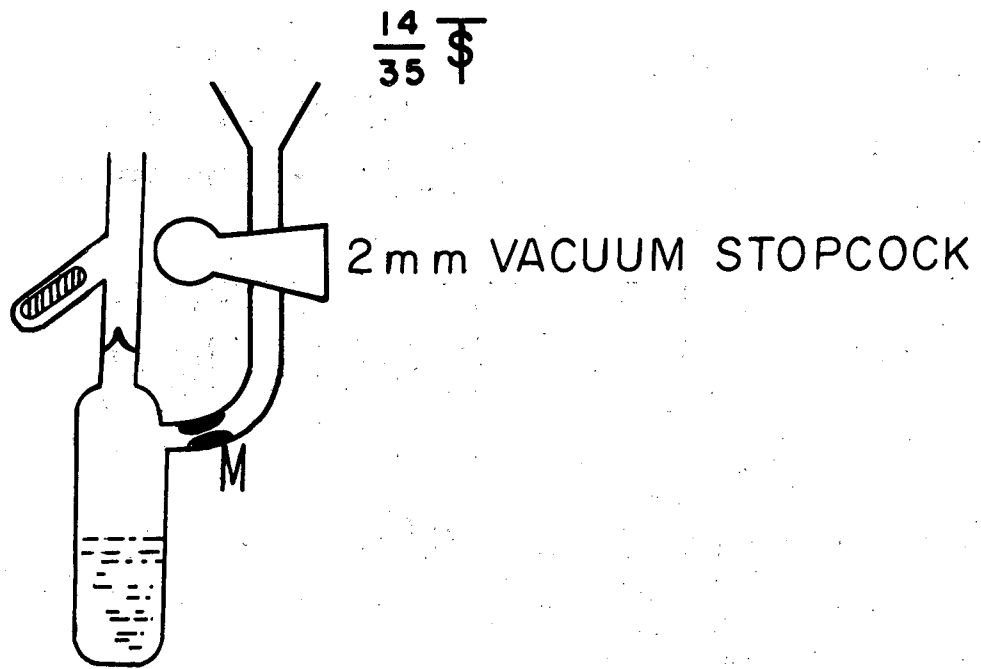
Side arms on collection vessels extend to rear, not to side as shown.

All tubing 10 mm o. d. except manometer which is 6 mm o. d.

All sealoffs at least 2.5 cm long with minimum i. d. 0.5 mm

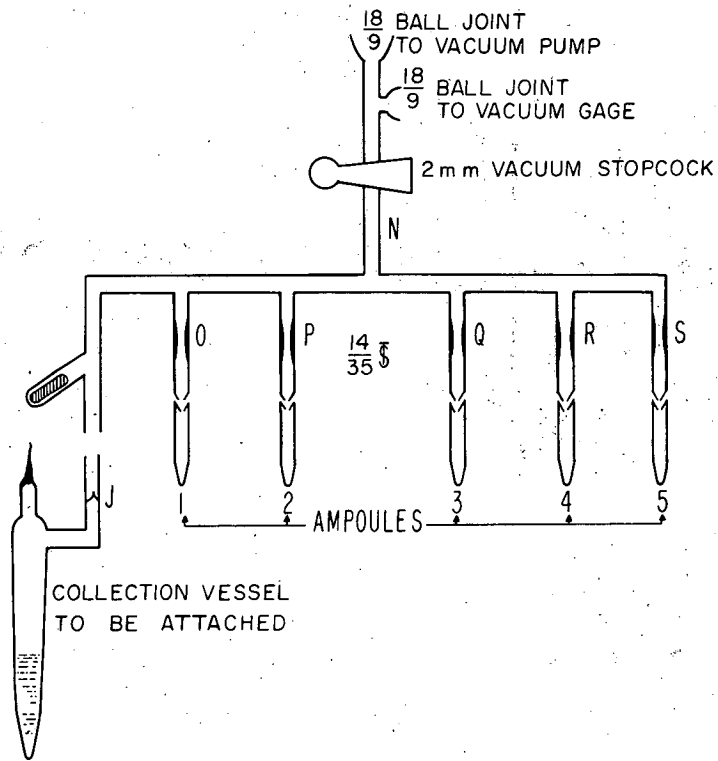
B, E, F, H, J are breakoff glass seals.

All glass pyrex.



MU-14556

Fig. 2. Diluting H₂O container during preparation.



MU-14557

Fig. 3. Glass system for transfer of tritiated water to ampoules.

2. Attachment of T₂ Container and Evacuation of System

The glass system is clamped in a Berkeley gloved box and the bulb containing the tritium to be oxidized is attached to the line at A. (Pressure inside the line is required for this glass-blowing job. Pressure in a tube from the operator's mouth collapses a polyethylene bag inside a 4-liter flask. The bag is attached via tubing to the glass system at the vacuum-gauge ball joint.) The vacuum gauge is replaced, the charcoal is baked out with a heat lamp, and the system is then evacuated to 70 microns pressure (or less).

3. Sorption of T₂ on Charcoal; Pump-Out of Helium

If the T₂ is not freshly prepared, enough He³ may be present to interfere with the vacuum distillations. If there is, the stopcock is closed, the charcoal is frozen in a liquid nitrogen bath, and the inside break-off seal B is opened. When the pressure drops to a constant level, the stopcock is opened and the He³ is pumped out for about 30 seconds. Since preliminary studies with hydrogen indicated that about 0.5% of the tritium is lost during this pumpout of the line, the vacuum pump exhaust is led into the manifold, which draws air through the gloved box. The seals at C and G are cut through, the end pieces are set aside, and the "zero" pressure reading is noted on the manometer.

4. Oxidation of T₂; Dilution and Collection of Tritiated Water

The oven is turned on to 400°C and the charcoal is warmed and baked out by a heat lamp. Concentrated tritiated water collects in the cold finger, which is chilled in a liquid nitrogen bath. When the pressure drops to "zero," seal D is cut through, and the charcoal trap and CuO bulb are set aside. Break-off seal E is opened and the H₂O in container No. 1 is evaporated by heat lamp and collected in the cold finger and in collection vessel No. 1, which have been cooled in liquid nitrogen baths. The cold finger is warmed and the diluted tritiated water is collected in collection vessel No. 1. The frozen diluted tritiated water is melted and mixed, and an aliquot is evaporated and collected in collection vessel No. 2, which is chilled in a liquid nitrogen bath. Seal K is cut through, seal F is opened, and the contents of the diluting-H₂O container No. 2 are evaporated by heat lamp and collected in collection vessel No. 2. Seal L is cut through. The collection vessels containing different concentrations of tritiated water are ready to be prepared for storage.

5. Storage of Collection Vessels and Disposal of Glassware

There is a possibility that bursting pressures of H₂ may build up in the sealed bulbs from self-decomposition of the tritiated water they contain.³ Therefore they are placed in cylindrical ice cream cartons and mechanically sealed in cans for storage. The remaining glass parts of the line are enclosed in a plaster of Paris cast, removed from the gloved box, and concreted in barrels for sea burial.

6. Transfer of the Tritiated Water to Ampoules for Use

The collection vessel is attached to a vacuum line (see Fig. 3) set up in a gloved box. The stopcock is opened and the line is evacuated and sealed at N. Break-off seal T is opened and the tritiated water is vacuum-distilled into ampoules for use in the immediate future. Seals O, P, Q, R, ... are cut through as desired amounts fill each ampoule. The ampoules are sealed in cans during transit to the gloved box where they are to be used.

DISCUSSION

Tritiated water produced by this method contains unknown trace amounts of Cu, Hg, and Fe from the oxidant, manometer, and break-off rods respectively. The manometer is not essential but with it in the line one can follow the pressure rises and falls during the T₂ sorption, the T₂ oxidation, and the vacuum distillations.

A fairly good initial vacuum is necessary in the line to insure that during evaporations the tritiated water will condense in the desired chilled bulb and not in the connecting tubing, which is at room temperature. The entire system should be checked for vacuum leaks before it is put in the gloved box.

The procedure from Step 2 through Step 4 takes about 6 hours, the greater part of which is for the vacuum distillations, which require about 5 minutes per milliliter evaporated. The oxidation of the T₂ is complete in about 45 minutes.

One hundred curies of tritiated water in concentrations from 5 to 20 curies per milliliter have been produced by this method. It would also be equally convenient to produce other concentrations. An unknown loss of activity occurs in the sorption of tritium on the charcoal, on the CuO, and on the inside of the glass system. This loss could be reduced by following the T₂ oxidation with H₂ flushes from another bulb attached to the line.

If the T₂ is freshly prepared and is free of He³, the line can be sealed at G before the T₂ bulb is opened, and the charcoal bulb can be omitted from the line. The tritium is then enclosed in sealed glass at all times during the operation.

The pressure of gases from decomposition of the tritiated water in one of our collection vessels containing 60 curies in 3.4 ml was measured 3½ months after being sealed. The volume of the collection vessel above the water was 15 ml and the pressure in this volume was 33 ± 14 mm Hg at S. T. P. This pressure was due to gases which are not condensed at liquid nitrogen temperatures. As this pressure can be completely accounted for by He³ build-up during 3½ months of storage the equilibrium pressure of hydrogen over our tritiated water as a result of the self-decomposition-recombination reaction is low.

As a double precaution two portable tritium "sniffers" were used during the operation. One sampled air near the sliding door of the Berkeley gloved box; the other, which was placed several feet away, sampled air that the room air currents carried past the gloved box.

SUMMARY

This report describes a method for producing milliliter amounts of tritiated water from tritium gas by oxidation over hot CuO in a sealed glass system. The method is somewhat slow, and trace amounts of certain impurities may be present in the product, but there is a minimum of exposure of operating personnel to tritium.

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