UCRL-51910

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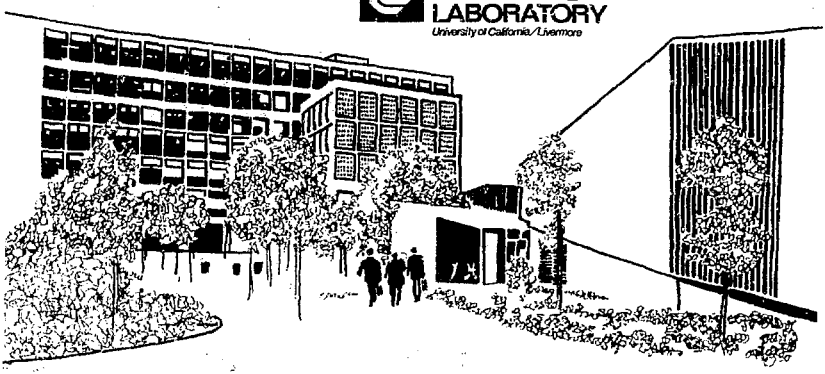
Prepared for U.S. Energy Research & Development Administration under contract No. W-7405-Eng-48



Printed in the United States of America
Available from
National Technical Information Service
U. S. Department of Commerce
5285 Port Royal Road
Springfield, Virginia 22151
Price: Printed Copy $___*; Microfiche $2.25

*Pages	NTIS Selling Price
1-50	$4.00
51-150	$5.45
151-325	$7.60
326-500	$10.60
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Distribution Category
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MS. date: September 19, 1975

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Abstract

The performance of a uranium trap as a means of removing tritium from an inert gas was measured for varying trap conditions, using deuterium (to represent tritium) in argon at room temperature. Performance was expressed as a purification factor, which is the ratio of deuterium concentration at the inlet to that at the outlet of the trap. Purification factors vary inversely with both the ratio of deuterium to uranium already contained in the trap and with the rate of flow of gas through the trap. Varying the inlet deuterium concentration had no apparent effect.

Introduction

Processing and handling of tritium frequently result in waste inert gases contaminated with tritium. These gases come from such operations as flushing of equipment prior to making repairs and recovery of tritium from earlier experiments. The tritium must be scavenged from these waste gases prior to their disposal, and it can be recovered later for reuse.

A uranium getter bed offers a simple means of scavenging tritium from inert gases. Uranium traps have been used for purification and storage of hydrogen for many years, including the separation of ^3He from tritium. Equilibrium partial pressures of hydrogen isotopes over uranium are well known.[1] Uranium metal is readily converted to an active getter powder by simply hydriding and dehydriding it[2]; dehydriding is easily done by heating to about 400°C.

We have studied the use of uranium as a potential getter bed for tritium in a flowing gas stream. Since uranium reacts with all isotopes of hydrogen, we used mixtures of deuterium in argon in these experiments. Preliminary studies by Weed[3] showed that the efficiency of a uranium trap varies inversely with the

flow rate. We have extended his work, and we have also studied the effects of varying the atomic deuterium-to-uranium (D/U) ratio in the trap; i.e., the relative amount of UD_3 already formed on the trap, and also the effects of varying the D_2 concentration in the gas entering the trap.

Equipment and Procedures

The original experimental system has already been described,[3] and the major components are shown in Fig. 1. Deuterium and argon flow rates are measured by mass flowmeters. Flow rates varied from 11 to 44 cm^3(STP)/s (0.07 to 0.27 mole/m^2·s). The test gas mixture was 1 or 2 vol% deuterium in argon. Total pressures ranged from about 119 to 153 kPa, depending on the flow rate.

The uranium trap shown in Fig. 2 was developed by Carlos Colmenares[4] for use in a portable tritium cleanup system. It contains 5.71 moles (~1.36 kg) of ^{238}U powder arranged in three layers, each separated into eight sections, to keep the uranium powder distributed evenly. The trap has a cross-sectional area of 73.8 cm^2, giving a uranium distribution of 18.4 g/cm^2 of flow area. The D/U ratio was assumed to be zero after dehydriding the trap at about 400°C and simultaneously pumping off the outgassing deuterium until the steady-state pressure was <1.3 mPa (1×10^{-5} Torr).

Low pumping speed in the high vacuum system resulted in a high D_2 background during all runs. For this reason, the Varian partial pressure gauge* (hereafter, PPG: a residual gas analyzer) could not detect D_2 at concentrations less than ~250 ppm in the argon stream, although we had expected an order-of-magnitude better performance.

The amount of deuterium introduced into the uranium trap was measured in order to calculate the D/U ratio for each run. This was done by filling a tank of known volume with D_2 gas and monitoring the pressure change for each run.

Measurements of the PPG background and argon-only readings were always made before reading Ar-D_2

*Reference to a company or product name does not imply approval or recommendation of the product by the University of California or the U.S. Energy Research and Development Administration to the exclusion of others that may be suitable.

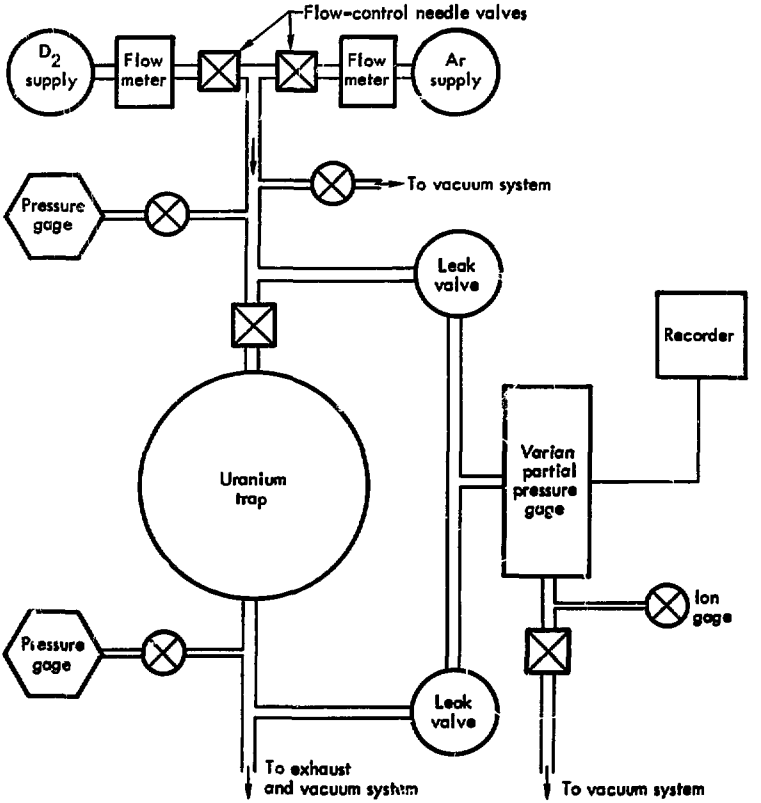

Fig. 1. Schematic diagram of uranium trap system for removing hydrogen isotopes from inert gas.

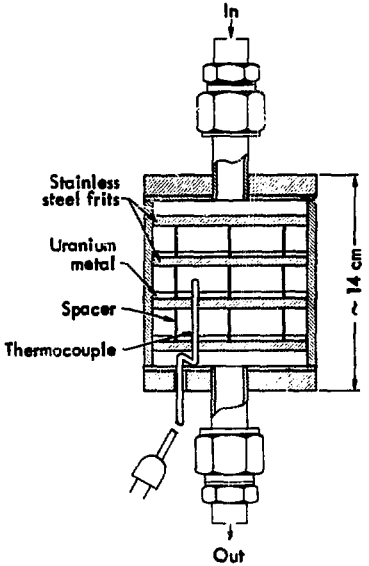

Fig. 2. Cross section of the uranium trap. Total uranium = 1.36 kg, cross section for gas flow = 74 cm^2.

mixtures. The outlet mixture was sampled before the inlet mixture to insure the highest sensitivity at the lower D_2 concentrations, although PPG data was used only for the trap outlet mixtures. Inadequate mixing of the D_2 and Ar gave erroneous readings at the trap inlet sampling valve; inlet D_2 concentrations were therefore derived from flowmeter data.

The D_2 concentrations determined by our PPG for known gas mixtures did not agree with corresponding results from a reliable mass spectrometer. Our PPG was subsequently calibrated using prepared gas mixtures containing 1.0 or 5.9 vol% D_2 in Ar. We still saw run-to-run variations of up to ±20% in the analyses of these mixtures. Similar variations would have a corresponding effect on our data.

Results and Discussion

The uranium trap efficiency is expressed as a purification factor, P.F.:

$$\text{P.F.} = \frac{\text{Concentration of inlet } D_2}{\text{Concentration of outlet } D_2}.$$

The inlet D_2 concentration was calculated from flowmeter data, the outlet D_2 concentration from PPG data.

The atomic D/U ratio was the most significant experimental variable affecting the purification factor. The purification factor varies inversely with D/U ratio as shown in Figs. 3 and 4. We would expect that

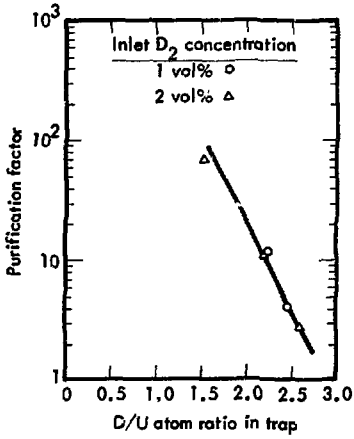

Fig. 3. Performance of the uranium trap as a function of D/U ratio and inlet D_2 concentration. Mass flow rate = 0.27 mole/m^2·s.

at a D/U ratio of 3, the trap would be fully loaded with deuterium and no purification would occur, giving a purification factor of 1. The line in Figs. 3 and 4 extrapolate to D/U ratios quite close to 3 for a purification factor of 1. The D/U ratios are sufficiently close to 3 that the deviation could be due entirely to experimental errors.

Changing the inlet D_2 concentration from 1 to 2 vol% had no effect on the purification factor. This is shown in Fig. 3, in which data is given for a mass flow of 0.27 mole/m^2·s with 1 and 2 vol% D_2.

The effect of varying the flow rate is shown in Fig. 4. The inlet gas D_2 concentration was 1 vol%, and mass flow rates were 0.07, 0.10, and 0.27 mole/m^2·s. It is clear that the purification factor varies inversely with flow rate.

Maienschein[5] has developed a model for a gas-phase, mass-transfer-limited process in a packed bed. For our case, this gives

$$\ln\left(\frac{C_o - C_s}{C_i - C_s}\right) = -\frac{A}{V} k_x a Z'$$

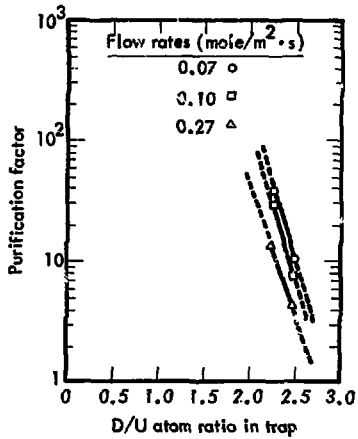

Fig. 4. Performance of the uranium trap as a function of D/U ratio and flow rate. Inlet gas is 1 vol% D_2 in Ar.

where

C_o = D_2 concentration at the trap outlet,

C_i = D_2 concentration at the trap inlet

C_s = D_2 concentration at the uranium surface, taken as the plateau pressure over UD_3,

A = trap cross-sectional area,

V = volumetric flow rate,

k_x = mass transfer coefficient,

a = uranium particle surface area per unit bed volume,

Z' = bed length of active material.

Here, C_s is very small compared to C_i and C_o, and can be ignored.

Evaluating k_x for a given trap at constant temperature and fixed D/U ratio, we find that the model is closely approximated by

$$\ln\left(\frac{C_o}{C_i}\right) = -\left(\frac{P}{G}\right)^{1/2} (K) ,$$

where

P = pressure (kPa),

G = mass flow/unit area (mole/m^2·s),

K = combined constant terms,

or,

$$\left(\frac{G}{P}\right)^{1/2} \log_{10} (P.F.) = K' ,$$

where the purification factor is P.F. = C_i/C_o, and K' represents constant terms describing the bed geometry, temperature, gas properties, etc. Mass flow rates, G, were 0.07 to 0.27 mole/m^2·s. Pressures ranged from 11y to 153 kPa.

A plot of $\left[\left(\frac{G}{P}\right)^{1/2} \log_{10} (P.F.)\right]$ versus D/U ratio is shown in Fig. 5. If our process is like that described by Maienschein's model, we can eliminate flow rate and pressure effects in the plot, to give a single straight line. The plot in Fig. 5 is

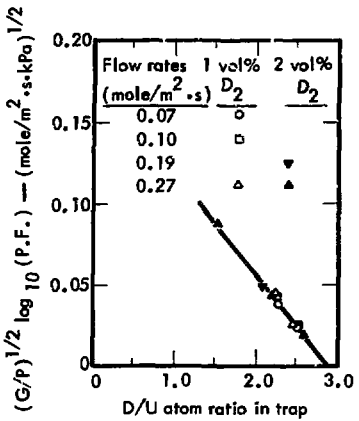

Fig. 5. Purification factor normalized for flow rate and pressure effects, $(G/P)^{1/2} \log_{10} (P.F.)$, plotted as a function of D/U ratio.

in reasonable accord with the model and can be represented by the equation

$$\left[\left(\frac{G}{P}\right)^{1/2} \log_{10} (P.F.)\right] = -0.066(D/U) + 0.19 .$$

These data compare favorably with more recent results,[6] except that our data appear to give a slightly steeper slope to the equation. Our data also indicated the need for better instrumentation, which was achieved in the more recent work.[6]

Conclusions

Expressed as a purification factor, the efficiency of a uranium trap in scavenging deuterium from argon was found to vary inversely both with the flow rate and with the atomic D/U ratio. The inlet deuterium concentration did not appear to affect the purification factor within the range studied, 1.0 to 2.0 volt. The data can be described by an equation of the form

$$\left[\left(\frac{G}{P}\right)^{1/2} \log_{10} (P.F.)\right] = -0.066(D/U) + 0.19 .$$

Acknowledgments

H. C. Weed installed and operated the original experimental equipment for this study; C. A. Colmenares designed the special uranium trap.

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