

## THE EFFECT OF TEMPERATURE AND CARRIER GAS ON THE LOSS RATE OF TRITIUM FROM RADIOACTIVE FOILS\*

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### INTRODUCTION

In this and other laboratories commonly employing ionization detection devices the source of radiation selected is often the relatively stable hydrogen isotope, tritium, a pure beta emitter. The tritium is usually available as the titanium tritide, with either copper or stainless steel backing. Detailed descriptions of the foils proper have been given by VANDERSCHMIDT *et al.*<sup>1</sup> and WILSON<sup>2</sup>. The tritium betas with a mean energy of 5 keV are not sufficiently energetic to penetrate even a thin layer of restraining material. Bremsstrahlung from these weak betas should be negligible. For these reasons, a source contained within a detection cell should not constitute a hazard due to surface activity at the detector body.

The hazards involved with initial loading and subsequent modification of any ionization device should be of little concern to the average operator since by law this procedure is to be carried out only by qualified, licensed personnel. It has been shown<sup>2</sup> that the action of sample components upon the source will be minor in nature owing to their presence in such minute amounts. The most noticeable action would be from components having a corrosive effect upon the metal backing of the foil. A more detailed treatment of possible hazards arising from use of radioactive foils has been given by TAYLOR<sup>3</sup>.

The National Aeronautics and Space Administration contemplate the use of gas chromatography employing ionization detectors in some of their interplanetary probes. Quite often it is necessary to operate these devices under varying conditions. For this reason, it would be desirable to know the effect of temperature and carrier gas upon the stability of these foils since tritium is a very attractive source in the laboratory development program of most of these devices. If losses should occur anomalous results would most likely be encountered without frequent calibration. Further, extreme losses might lead to hazardous conditions unless proper ventilation was provided, due to enrichment of effluent vapor from the system in tritium gas. This paper reports on such a study.

### EXPERIMENTAL

The radioactive foil used in these studies was obtained from U.S. Radium Corporation. The metal backing was of copper. The data were obtained from the experimental

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arrangement shown in Fig. 1. A known flow of clean, dry carrier gas was passed into a glass chamber containing the radioactive foil. The glass chamber was stationed in a furnace capable of elevating the temperature to 300°. The effluent gas was led through a short section of narrow diameter tubing into a large volume ionization chamber and from here to a fume hood.

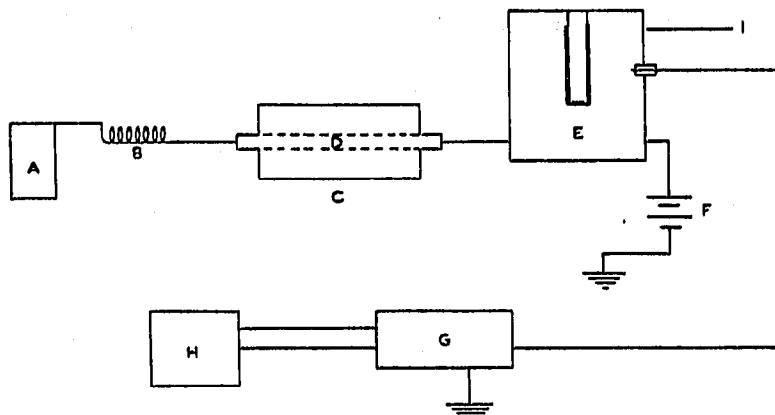


Fig. 1. Experimental arrangement: (A) carrier gas supply; (B) 5 Å molecular sieve drying column and capillary restrictor; (C) furnace; (D) glass chamber containing the radioactive foil; (E) ionization chamber; (F) polarizing potential; (G) electrometer; (H) recorder; (I) effluent gas to fume hood.

Fig. 2 illustrates the actual ion chamber used in the experimental operations. The collection electrode is a 3 cm diameter brass tube isolated from the body of the chamber by a teflon supporting rod. The chamber body is a cylindrical brass tube 13.5 cm in diameter. Gas enters the chamber through a gas-tight connection in the lower section and exits through a similar connection in the upper section. The total volume was measured and found to be approximately 2 l. The necessary electrical connections are made through the body of the chamber using gas-tight teflon insulators to minimize electrical leakage. A polarizing potential is developed between the detector body and the collection electrode. The electrical signal generated was fed into a high gain electrometer and subsequently recorded graphically by means of a potentiometric recorder. Details of the considerations to be made in measurements of this type as well as construction details of ionization chambers in general have been given by OVERMAN AND CLARK<sup>4</sup> and Nuclear of Chicago<sup>5</sup>.

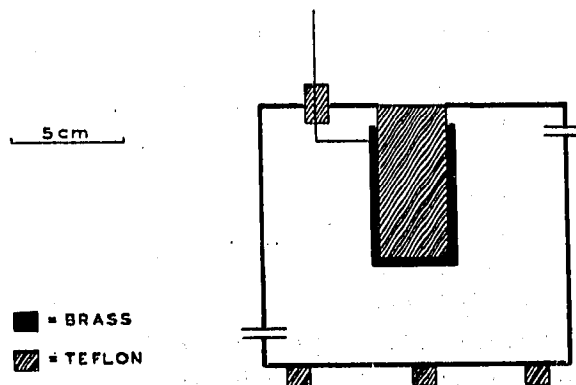


Fig. 2. Ionization chamber. Shaded sections of brass; lined sections of teflon.

A voltage *versus* current relationship was determined to establish the proper operating conditions for the system. Fig. 3 demonstrates that the chamber is operating as a simple ionization chamber at 300 V and that all the ions formed are being collected before there is a chance for recombination to occur. A measured volumetric flow of carrier gas, 100 ml/min, was allowed to pass through the system for several hours until equilibrium was reached. The signal generated indicated a normal back-

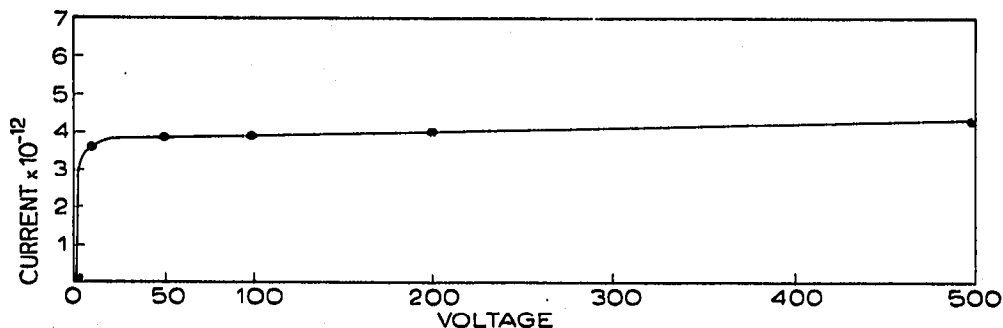


Fig. 3. Voltage *versus* current for the ionization chamber.

ground. The furnace temperature was adjusted to the desired level and several hours were required to reach a steady state equilibrium whereby the amount of tritium gas entering the chamber was equivalent to the amount of tritium gas being expelled. The signal thus obtained indicated the concentration of radioactive species within the chamber.

Tritium betas have a maximum range of about 1 cm in air at S.T.P. The dimensions of the ionization chamber were so arranged that the distance between electrodes far exceeds this value. This would imply that the total energy of the tritium betas should be dissipated in the gas phase. The number of ion pairs formed per beta particle should, to a very close approximation, be equal to the average energy of the tritium beta divided by the specific ionization energy of the gas under consideration. Values for specific ionization energies have been presented by SHARPE<sup>6</sup>. The average value is around 30 eV. Since 1 microcurie ( $\mu\text{C}$ ) is equal to  $3.700 \cdot 10^4$  disintegrations per second, the following relationship may be written:

$$C = \frac{IS(6.281 \cdot 10^{18})}{(5 \cdot 10^3)(3.700 \cdot 10^4)}$$

where:

$C$  = concentration of radioactive species ( $\mu\text{C}$ ),

$I$  = observed current (A),

$S$  = specific ionization energy of the carrier gas (eV).

The rate of loss from the tritium foil is now described by:

$$R = \frac{CF}{V}$$

where:

$R$  = rate of tritium loss ( $\mu\text{C}/\text{sec}$ ),

$C$  = concentration of radioactive species ( $\mu\text{C}$ ),

$F$  = volumetric flow of carrier gas ( $\text{cm}^3/\text{sec}$ ),

$V$  = volume of ionization chamber ( $\text{cm}^3$ ).

A single radioactive foil was used in all the experiments herein reported. Its activity was measured before and after each investigation in terms of the current it provided in a small volume ionization chamber under a standard set of conditions. Allowances were made in the calculations for any change in observed activity.

#### RESULTS AND DISCUSSION

The gases studied were  $H_2$ , He,  $N_2$ , air, Ar + 5%  $CH_4$  and Ar saturated with water vapor. The temperature ranged from ambient to  $290^\circ$ . Fig. 4 shows the ion current observed *versus* temperature for the above gases. As might be expected, the current rises very rapidly above  $200^\circ$  indicating an appreciable loss of tritium from the radio-

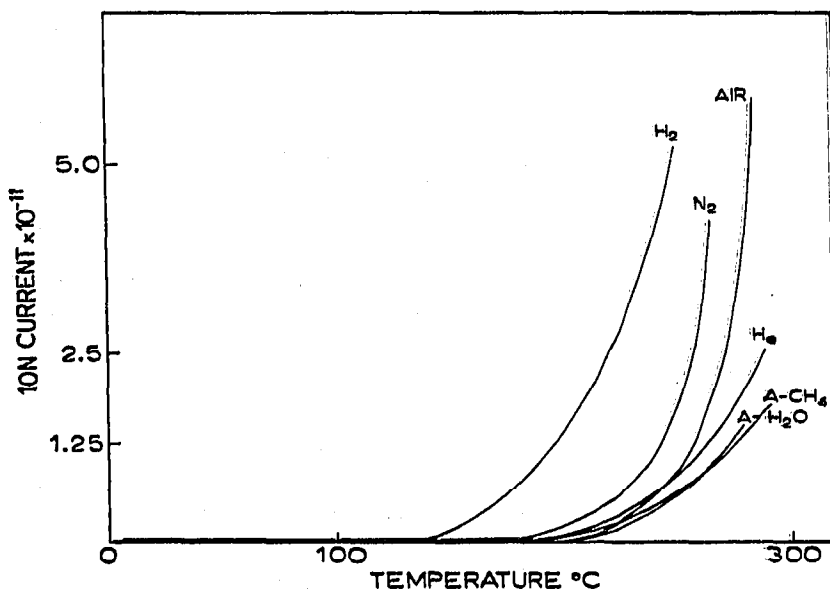


Fig. 4. Ion current *versus* temperature for various carrier gases, before correction for changes in source strength.

active foil. The exception noted was with hydrogen. In this case, noticeable losses occur above  $140^\circ$ . This observation could be accounted for if an exchange occurred between the hydrogen gas and the tritium contained on the foil as titanium tritide. Such an observation would appear to be reasonable since similar observations could not be made with other gases.

Table I lists the calculated loss rate of tritium in various carrier gases with respect to temperature. The loss rate  $R$  is given in units of both  $\mu C/\text{sec}$  and the more practical unit of  $\text{mC}/\text{day}$ .  $R$  is seen to range in value from  $10^{-2}$  to  $6 \text{ mC}/\text{day}$  depending upon the gas used and operating temperature. These values are in close agreement with those given by WILSON<sup>2</sup>. The concentration  $C$ , of tritium gas in air, assuming a 1000 cu.ft. room with a turnover rate of 1 volume per hour is presented. The assumed enclosure and ventilation conditions are worse than those normally encountered. Nevertheless, it should be noted that the maximum permissible concentration in air for tritium gas is not exceeded.

Table II gives the calculated half life of an average tritium foil in various gases at the indicated temperatures. This half life is based solely on the loss rate and does not

TABLE I

LOSS RATE OF TRITIUM FOR VARIOUS GASES AT DIFFERENT TEMPERATURES BASED ON A  
ON A 50 mC SOURCE

Gas	Temperature (°C)	R ( $\mu\text{C}/\text{sec}$ )	R (mC/day)	C* ( $\mu\text{C}/\text{cm}^3$ )
He	190	$0.73 \cdot 10^{-3}$	$6.31 \cdot 10^{-2}$	$0.93 \cdot 10^{-7}$
	243	$5.72 \cdot 10^{-3}$	$4.94 \cdot 10^{-1}$	$7.26 \cdot 10^{-7}$
	290	$2.38 \cdot 10^{-2}$	2.05	$3.02 \cdot 10^{-7}$
Ar-5% CH <sub>4</sub>	190	$6.10 \cdot 10^{-4}$	$5.27 \cdot 10^{-2}$	$7.75 \cdot 10^{-8}$
	240	$5.20 \cdot 10^{-3}$	$4.49 \cdot 10^{-1}$	$6.60 \cdot 10^{-7}$
	290	$2.60 \cdot 10^{-2}$	2.24	$3.30 \cdot 10^{-6}$
Ar-H <sub>2</sub> O	240	$6.50 \cdot 10^{-3}$	$5.61 \cdot 10^{-1}$	$8.25 \cdot 10^{-7}$
	280	$2.20 \cdot 10^{-2}$	1.90	$2.79 \cdot 10^{-6}$
N <sub>2</sub>	185	$4.22 \cdot 10^{-4}$	$3.64 \cdot 10^{-2}$	$5.35 \cdot 10^{-8}$
	240	$5.93 \cdot 10^{-3}$	$5.12 \cdot 10^{-1}$	$7.55 \cdot 10^{-7}$
	285	$4.49 \cdot 10^{-2}$	3.87	$5.70 \cdot 10^{-6}$
Air	240	$3.63 \cdot 10^{-3}$	$3.13 \cdot 10^{-1}$	$4.60 \cdot 10^{-7}$
	285	$6.50 \cdot 10^{-2}$	5.61	$8.25 \cdot 10^{-6}$
H <sub>2</sub>	148	$4.57 \cdot 10^{-4}$	$3.94 \cdot 10^{-2}$	$5.80 \cdot 10^{-8}$
	195	$1.05 \cdot 10^{-2}$	$9.00 \cdot 10^{-1}$	$1.33 \cdot 10^{-6}$
	250	$4.62 \cdot 10^{-2}$	3.99	$5.85 \cdot 10^{-6}$
	290	$4.29 \cdot 10^{-2}$	3.70	$5.35 \cdot 10^{-6}$

\* For molecular tritium the occupation maximum permissible concentration in air is  $2 \cdot 10^{-3}$   $\mu\text{C}/\text{cm}^3$ .

TABLE II

ESTIMATED HALF LIFE OF TRITIUM FOIL FOR  
VARIOUS GASES AT THE TEMPERATURES INDICATED

Gas	Temperature (°C)	Half life calculated* (days)
He	190	547
	243	71.2
	290	16.9
Ar-5% CH <sub>4</sub>	190	650
	240	72.6
	290	14.7
Ar-H <sub>2</sub> O	240	61.5
	280	18.2
N <sub>2</sub>	185	960
	240	67.6
	285	8.96
Air	240	109
	285	6.16
H <sub>2</sub>	148	887
	195	38.4
	250	8.64
	290	9.35

\* Half life calculated on basis of simple first order kinetics.

indicate the decrease in activity due to normal radioactive decay. This decrease in activity due to radioactive decay will not be of the same order of magnitude as the loss rate at elevated temperatures and therefore may be disregarded. Calculated half life due to leakage ranges from 960 days at 185° to 6.16 days at 285°, again depending upon the gas present. A slight discrepancy in the data for hydrogen is noted between 250° and 290°. This may be accounted for by considering the rapid decrease in source activity at these elevated temperatures. From the calculated half life values one would predict that the observed saturation current of a detection device should decrease to half of its original value in one half life of the associated radioactive foil. In practice this has not always been found to be true. This apparent contradiction can best be explained by considering the characteristics of each individual source, which can reasonably be expected to vary from one source to another.

#### CONCLUSIONS

The data obtained in this investigation provide a guide for the selection of operating parameters when using tritium foils. Minor variations might occur for a given source, and indeed, variations would be expected to occur between different sources. The general observations, however, should remain valid.

The generally accepted maximum operating temperature for tritium foils is around 225°. The results herein reported indicate that when hydrogen is used as the carrier gas leakage losses become apparent at a much lower temperature than with the other gases tested. For molecular tritium, the occupation maximum permissible concentration in air is  $2 \cdot 10^{-3} \mu\text{C}/\text{cm}^3$  (ref. 2). Thus, if the ventilation conditions for a given site are known, the foregoing guide may be used to determine approximate concentration levels. As suggested by WILSON, it is advisable to lead the outlet of the detection device containing the radioactive foil into an extract or a fume hood if operation at elevated temperature is contemplated.

#### ACKNOWLEDGEMENT

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#### SUMMARY

By means of a simple ionization chamber the loss rate of tritium from tritium impregnated foils was determined. An average 50 mC source was investigated, the loss rate being determined with respect to carrier gas and temperature. The maximum temperature suggested for the use of these foils has been 225° due to health hazards involved. Our results confirm these figures as the maximum allowable concentration in air is not exceeded at this temperature even under the most adverse conditions. From a practical viewpoint, however, it may be desirable to operate somewhat below this temperature in order to obtain a reasonable lifetime of the source in certain carrier gases.

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