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UCRL-52801

# TRITIUM RELEASE FROM A NONEVAPORABLE GETTER-PUMP CARTRIDGE EXPOSED TO MOIST AIR AT AMBIENT TEMPERATURE

MASTER

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July 10, 1979

Wurk performed under the auspices of the U.S. Department of Energy by the UCLLL under contract number W-7405-ENG-48.



# MASTER

Distribution Category UC-4



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# TRITIUM RELEASE FROM A NONEVAPORABLE GETTER-PUMP CARTRIDGE EXPOSED TO MOIST AIR AT AMBIENT TEMPERATURE

ABSTRACT

We measured the amount of tritium released when a commercially available getter-pump cartridge was exposed to moist air at ambient temperatures. The cartridge consisted of Zr-Al powder pressed onto an iron substrate, which is the type of cartridge proposed for use in the Tokomak Fusion Test Reactor. While we found that the initial release of tritium was rapid, the total activity released was less than 0.005% of the cartridge loading. Of this amount, at least 80% was released as tritiated water.

## INTRODUCTION

Although the potential problems of tritium technology associated with fusion reactor systems have been anticipated for some time,<sup>1,2</sup> fusion test facilities that use tritium as fuel are now being built; concepts are becoming realities. Handling this highly radioactive hydrogen isotope safely and economically requires knowing how well the proposed components will function during routine operations and knowing the consequences of unexpected failures.

One component proposed for use in the Tokomak Fusion Test Reactor (TFTR) currently under construction at the Princeton Plasma Physics Laboratory is a commercially available getter pump. The pump and cartridges are manufactured by Societa Apparicci Elettrici e Scientifici (SAES), Italy, and distributed in the U.S. by Westinghouse Electric Corp. and SAES Getters/USA.] This pump backs up turbomolecular pumps, which remove residual tritium and deuterium from the torus after each burn, and services neutral-beam injectors.<sup>3</sup> Previous investigations conducted at Lawrence Livermore Laboratory (LLL) determined the pumping speed of tritium, the effects of impurities on pumping speed, and the equilibrium base pressures at loadings anticipated in the TFTR.<sup>4</sup> The present investigation determined the amount of tritium released when a tritium-loaded cartridge was exposed to moist air at ambient temperature. The getter pump consists of a Zr-Al cartridge, a heater, and an outer case with cooling-water coils





(Fig. 1). (An unenclosed pump, consisting of only a cartridge and heater mounted on a flange, is called a nude pump.) The cartridges are made by pressing a

powdered alloy (84% zirconium and 16% aluminum) onto an iron substrate by a proprietary process. (The SAES trade name for the Zr-Al mixture is St 101 alloy.) Folded strips of this material are then arranged in pleated fashion at 4-deg angles to form a circular cartridge with maximum surface area and trapping efficiency (Fig. 1). The cartridge fits around a rod-shaped heating element that is mounted on a Conflat-style flange with electrical

feed-throughs. This heater both activates the cartridge (by heating to 750°C *in vacuo*) and maintains the pump at a desired operating temperature: optimum temperatures depend on the particular application. The heater also regenerates hydrogen pumps. Table 1 lists the manufacturer's performance data for the C-100 cartridge that we investigated and the C-500 cartridge proposed for use with the TFTR.

TABL	Æ	1.	Cartridge	performance.	3
		••			

	Active	Getter	Pun	iping spec	:ds, 1/s	Permane capacit activati	nt sorption y for one on, Torr-1	Reversible						
Cartridge	cm <sup>2</sup>	y	CO	N <sub>2</sub>	H2 <sup>b</sup>	со	N <sub>2</sub>	of H <sub>2</sub>	, Torr-1					
C-100	2600	80	_	_	· _	55	55	135°	780 <sup>0</sup>					
Nude	-	-	300	110	650				-					
Enclosed	-	-	100	50	280			· · -	-					
C-500	13000	385	-	-	-	280	280	670 <sup>c</sup>	- 4000 <sup>d</sup>					
Nude	-	-	750	320	2000	-	- 1	-	_					
Enclosed	····	• ••	500	250	1400	-	18 - T	-	-					

<sup>a</sup>Data from Ref. 6.

<sup>b</sup>Hydrogen pumping speed at 400°C.

<sup>c</sup>Equilibrium pressure:  $3 \times 10^{-6}$  at 400°C.

<sup>d</sup>Equilibrium pressure: 1 × 10<sup>-4</sup> at 400°C.

#### PROCEDURES AND RESULTS

We activated an enclosed SAES C-100 getter pump cartridge used in previous getter-pump evaluations by heating it for 14 h at over 750°C; an activated-uranium trap was used to prevent residual tritium driven off by heating from entering the atmosphere. When the dynamic pressure reached  $5 \times 10^{-4}$  Torr, the uranium trap was isolated and the cartridge was pumped on directly using a mercury diffusion pump until the pressure reached 2.3 ×  $10^{-8}$  Torr. The cartridge was then isolated under vacuum until it was loaded with tritium.

Figure 2 shows the system used to load the getter-pump cartridge and analyze the residual gas after loading. The calibrated manifold was filled with 96.69% tritium to the pressure that would give a cartridge loading of 750 Ci: all apparatus was at ambient temperature. Within 30 min after loading, we analyzed the residual gas in the manifold with a UTI-100 C residual-gas analyzer; it showed that the

gas was essentially Helium-3, the product of  $T_2$  decay. The getter pump was then evacuated to 1.4 × 10<sup>-5</sup> Torr and isolated.

Before removing the getter cartridge from the loading system, the activity level of the gas over the cartridge was measured using an ion chamber. The manifold, lines, and pump were filled with dry argon (1-2 ppm (v) water) to 760 Torr and the argon circulated through the system and ion chamber using a metal bellows pump. The ion chamber readings (corrected for the argon carrier gas) indicated that about 9 mCi of tritium was in the gas in the getter pump when it was isolated. However, because the system and ion chamber were severely contaminated from previous work, we believe this reading was erroneous. After the ion chamber readings were made, the argon pressure was increased to 790 Torr, the getter pump valve was closed, and the pump was removed from the





manifold and transferred to the inert-atmosphere box for disastembly and removal of the cartridge.

The activity in the inert-atmosphere box was measured using two ion chambers separated by a molecular sieve bed. The total activity (tritium and tritiated water) was measured in the first chamber: the gas sample was then passed through the molecular sieve bed to remove water vapor, and the activity of the tritium gas was measured in the second chamber.

A previous experience prompted us to determine whether the activity measured in an inertatmosphere box after several days was released from the cartridges or from the other pump components stored in the box. We removed and isolated the getter cartridge in a clean container. We then

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reassembled the pump body and stored all contaminated parts inside it so that a separate activity measurement could be made later (see Fig. 3). During these manipulations, about 3 mCi of tritium activity was released in the inert-atmosphere box, about 80% of which was tritium gas.

Another question we hoped to answer was whether the Zr-Al alloy would release tritium at a higher rate than predicted on the basis of dissociation pressure alone. We removed the scaled cartridge container from the inert-atmosphere box, and, using the same tritium monitoring system as before, measured the activity released over a 5-h period as the cartridge container was purged with dry argon. As in the experiment above, most of the activity released was attributable to tritium gas. The total activity measured during this period was  $\pm$  500  $\mu$ Ci, which corresponds to  $\pm$  0.5  $\mu$ Ci/Ci on the cartridge over 8 h. After this measurement, the getter cartridge was isolated under dry argon.

We then measured the level of tritium released when the cartridge was exposed to moist air. The cartridge container was placed in a maintenance enclosure equipped with a tritium monitoring system, pressure sensors, moisture monitors, and a tritiumremoval system (Fig. 4). A diaphragm pump and flowmeter were attached to the cartridge container, and moist air (6000-7000 ppm (v) water) was pumped at about 91/min through the cartridge container, which had a free volume of nearly 1 litre. The activity level in the enclosure increased to 5.77 mCi/m<sup>3</sup> in 16 min. After 20 min, the activity level began to drop because of the removal of tritiated water by the molecular sieve hed in the monitoring system. After 66 min, the getter cartridge was isolated under an atmosphere of moist air, and the tritium-removal system of the test enclosure was turned on. The total activity released during this 66min exposure was 30.4 mCi,

After we began this experiment, we discovered that the second ion chamber had a bias that prevented accurate measurement of tritium released as  $T_2$ . Because of this, we operated the ion chambers in parallel rather than in series, and, by comparing the two readings, we estimated that at least 80% of the tritium released was in the form of tritiated water.

After the cartridge had been stored under moist air for about 24 h, it was again exposed to moist air flowing at about 9 1/min. To prevent removal of tritiated water by the molecular sieve bed in the monitoring system, the bed was bypassed during



FIG. 3. Tritium activity of getter pump body and all contaminated parts measured after storage under argon for one week; total tritium release was about 9,8 mCi, about 80% of which was tritium gas.

this exposure. The activity levels measured in the enclosure were ten times lower than those observed on the previous day. (These readings included the activity released during the storage period.) After 12 min, the measured tritium release rate was about 5.5  $\mu$ Ci/min. Because of the large enclosure volume (5.46 m<sup>3</sup>), small variations in ion chamber readings resulted in large fluctuations in the apparent tritium release rates, e.g., the rate varied between 1.6 and 11  $\mu$ Ci/min during the latter part of the exposure. After 33 min, the exposure was terminated and the cartridge was again stored under an atmosphere of moist air. A total of 2.5 mCi of tritium was released during this exposure.

Because the maintenance enclosure was needed for other experiments and because the tritium release rates were so low, we removed the cartridge in its container and attempted to measure the release rates using other monitoring systems. Unfortunately, because of a combination of factors, the results of these measurements were not entirely satisfactory.

To test the effect of lower relative-humidity levels on the release rate, we made four runs with





dry air (3.3 ppm (v) water). We used a monitoring system, similar to the one described in Fig. 4, that consisted of two ion chambers separated by a molecular sieve bed with a provision to bypass the sieve bed if desired; the system was open to the atmosphere and vented into a fume hood. Figure 5 shows a typical run; nearly 80% of the activity was attributable to tritium gas. During one of these runs, we also varied the flow rate; although we expected the tritium concentration to increase as the flow rate decreased, Fig. 6 shows that changing the flow rate did not change the measured activity levels. The final moist-air exposure was also made using this open system. After 500 h had elapsed, the tritium release rates were very low and flow rates had to be reduced to extremely low values to see any significant changes in ion chamber readings; unfortunately, it was almost impossible to keep such low flow rates constant. We continued the final exposure for over 770 h. We observed the major change in release rate during the first 100 h; subsequent changes were small and fluctuated with changes in relative humidity (Fig. 7). Figure 8 shows the observed changes in release rate over the entire exposure period of about 1200 h.

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FIG. 5. Ion chamber readings for typical run (No. 8) of cartridge exposed to dry air (3.3 ppm (v) water,); nearly 80% of the activity was attributable to tritium gas. The cartridge had been stored under dry air for about 15 h; the air flow was 740 cm3/min, and the container volume was 930 cm3.



FIG. 6. Ion chamber readings for run No. 9 (cartridge exposed to dry air); the flow rate was varied during this run, but no significant changes in the activity levels were observed. The cartridge had been stored under dry air for about 5.6 h.



FIG. 7. Ion chamber readings for run No. 11 (cartridge exposed to moist air); the major change in release rate occurred during the first 100 h; subsequent changes were small and fluctuated with changes in relative humidity. The air flow was 235 cm<sup>3</sup>/min.



FIG. 8. Change in release rate over the entire exposure period of about 1200 h.

## CONCLUSIONS

Our observations --showed that the initial tritium release from a loaded C-100 getter cartridge exposed to moist air is rapid but that the amount released represents only a small fraction of the cartridge loading. More than 72% of the total tritium released during the exposure was released during the first 4 min; the release measured in 66 min amounted to less than 0.005% of the cartridge loading. As predicted by Singleton and Griffith,<sup>4</sup> the release rate decreased rapidly with time; after more that 1200 h, the total tritium activity released by the cartridge amounted to less than 0.1 Ci (about 0.01% of the cartridge loading).

A comparison of our data with the reported release rate of tritium from titanium tritide sources under dry  $air^{5}$  indicates that the release rate of the Zr-Al alloy is significantly lower. However, there are insufficient data to demonstrate that such a comparison is valid.

#### RECOMMENDATIONS

The C-500 getter pumps proposed for use in the TFTR will contain significantly larger getter elements with approximately five times the active surface area of the C-100 pump cartridges we tested.<sup>6</sup> If we assume equal loading and release proportional to the active surface area, we would expect the initial release areach C-500 pump to be about 0.15 Ci if the getter is exposed to air. If an entire array of 12 pumps were exposed simultaneously, about 2 Ci would be released almost immediately. Because most of the release would be tritiated water and because the biological uptake of tritiated water is much more rapid than that of tritium gas, we recommend that special techniques and handling fixtures be used to prevent or reduce radiation exposures of operating personnel.

### ACKNOWLEDGMENTS

We thank other members of the tritium facility staff for their assistance on this project. Dan Disch and Lyall Miller provided the electronic support required to make the cartridge-loading system

operational. Don Fearon and Karl Johnson constructed a portable monitoring system used for most of our exposure runs, and Augustine Nicolosi furnished the needed monitoring equipment.

#### REFERENCES

- R. G. Hickman, "Tritium Problems in Fusion Reactor Systems," in Proc. Symposium on Tritium Technology Related to Fusion Reactor Systems (Miamisburg, Ohio, 1974), ERDA-50, 1975, pp. 76-86.
- A. Barosi, T. A. Girogi, and L. Rosai, "Characteristics of SORB-AC Non-Evaporable Getter Cartridges and Their Potential Use in Fusion Reactors," in Proc. of Int. Conference on Radiation Effects and Tritium Technology for Fusion Reactors (Gatlinburg, Tenn., 1975), NTIS Conf-750989, IV 203 (1976).
- Tokomak Fusion Test Reactor Facilities, Final Environmental Statement, U.S. Energy Research and Development Administration, ERDA-1544, Princeton Plasma Physics Laboratory, Princeton, N.J. (1975).
- 4. M. F. Singleton and C. M. Griffith, Evaluation of a Nonevaporable Getter Pump for Tritium Handling in the Tokomak Fusion Test Reactor, Lawrence Livermore Laboratory, Livermore, Calif., UCRL-52584 (1978).
- J. A. B. Gibson, The Possible Radiological Hazard from Tritium Sources Absorbed on Titanium, United Kingdom Atomic Energy Authority Research Group, Atomic Energy Research Establishment, Harwell, Berks, Great Britain (1963).
- 6. "Westinghouse Data Sheet B-439," Westinghouse Electric Corp., Horseheads, N.Y. (1978).

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