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SUBJECT: Radiation Damage to Freon

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Radiation damage to Freon-11 (CCl_3F) has been reported at the Army γ-irradiation facility at Dugway Proving Ground[1]. The radiation stability of similar organic compounds has been summarized in recent reviews by Tolbert and Lemmon[2] and by McDonell and Newton[3]. Accordingly, the feasibility of using Freon as a secondary refrigerant for the HRT refrigeration system was questioned and discussed in memos by Michelson[1] and Rudolph[4].

Since Freon was used without difficulty for the HRE, it was originally planned to use essentially the same refrigeration system for the HRT. However, as emphasized by Michelson, the HRE ran for short periods of time and at lower intensities than those contemplated for the HRT. Hence, it was deemed advisable to conduct a radiation experiment on Freon-11 to determine the decomposition products and their amounts under conditions approximating those anticipated for the HRT.

A Van de Graaff accelerator was used for this decomposition study, as it provides a 100-fold more intense radiation source than the 1100 Curie Co-60 source (Chemistry Division) which was not available for prolonged continuous exposures and which was experimentally inadequate.

Experimental

The radiation cell (Figure 1) was constructed of Pyrex. It was cooled to about -70°C where the vapor pressure of Freon is about 4 mm., by circulating chilled trichlorethylene around the cell. Degreased copper and stainless steel turnings were included so that metal was present in both the liquid and gaseous Freon. The trap containing fused KOH was incorporated in the system to serve both as a drying agent and as a reactant for any acid vapors which did not react with the metal. Thirty ml of Freon were distilled directly into the cell since it was found that practically all available stopcock greases were dissolved by the Freon vapors.

The irradiation, using an electron beam of 5 microamps at 1.5 Mv, was completed in three periods of 10, 10, and 8 minutes each. Operational difficulties with the accelerator were responsible for terminating the experiment after 28 minutes exposure. Samples of both the original and distilled Freon, and the irradiated distillate and residue were analyzed. The metal turnings were washed in dilute nitric acid to dissolve the green deposit formed on the surface; both the wash solution and the metal turnings were analyzed for chloride and fluoride.

Results

The Freon received approximately 8% (2.63×10^{21} ev/ml) of the radiation exposure expected by the Freon in the HRT refrigeration system[1]. A total of 28.3 mg of chlorine (as Cl^-) and 4.1 mg of fluorine (as F^-) formed during the irradiation reacted with the stainless steel and copper turnings, as well as with the fused KOH in the system (Table I). Approximately half of the halogen formed was found in the KOH trap. It appears that the exposed surfaces of the metal turnings were completely covered with a layer of metal halide, as further indicated by the fact that all of the metal was covered with a yellowish-green deposit.

In terms of the usual method for expressing irradiation decomposition, the G values for the production of chlorine and fluorine (molecules formed per 100 ev absorbed) are 0.31 and 0.082, respectively. These figures compare favorably with reported G values of 0.26 at -190°C to 7 at 100°C, for the production of bromine from solid and liquid CCl_3Br, respectively[5].

Since Freon in the HRT refrigeration system will be subjected to γ-radiation from -40°C to room temperature, it is probable that the G value of 0.39 (for total halogen formation) will be raised by an order of magnitude.

In view of these results and the literature cited, it is recommended that refrigerants other than Freon be considered for use in the HRT secondary refrigeration system, with special consideration to those which form non-corrosive decomposition products.

Subsequent to the discussion with R. B. Briggs on the design and operation of the cold traps, and a cursory check of the literature[3], either ethanol or methanol would appear to be satisfactory secondary refrigerants for the HRT.

TABLE I

ANALYSES

	Mass Spectrometer		Ionic			
	H_2O	SiF_4	Cl^-	F^-	Cu	347 S.S.
Original Freon	51 ppm	-				
Distilled Freon	131 ppm	-				
Irradiated Distillate	44 ppm	756 ppm				
Irradiated Residue	36 ppm	-				
Dil. HNO_3 Wash			15.8 mg	1.0 mg	42.6 mg	
Fused KOH			12.5 mg	3.1 mg		
Metal Turnings					50 mg	137.4 mg

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REFERENCES

(1) C. Michelson, Radiation Damage to Freon, Memo CF 55-9-163, Sept. 30, 1955.

(2) B. M. Tolbert and R. M. Lemmon, Radiation Decomposition of Pure Organic Compounds, UCRL-2704, August, 1954.

(3) W. R. McDonell and A. S. Newton, J. Am. Chem. Soc., 76, 4651 (1954).

(4) P. S. Rudolph, Memo to C. Michelson, October 28, 1955.

(5) R. F. Firestone and J. E. Willard, Abstr. Am. Chem. Soc., Paper No. 53, (127th Meeting, Division of Physical and Inorganic Chemistry) April, 1955.

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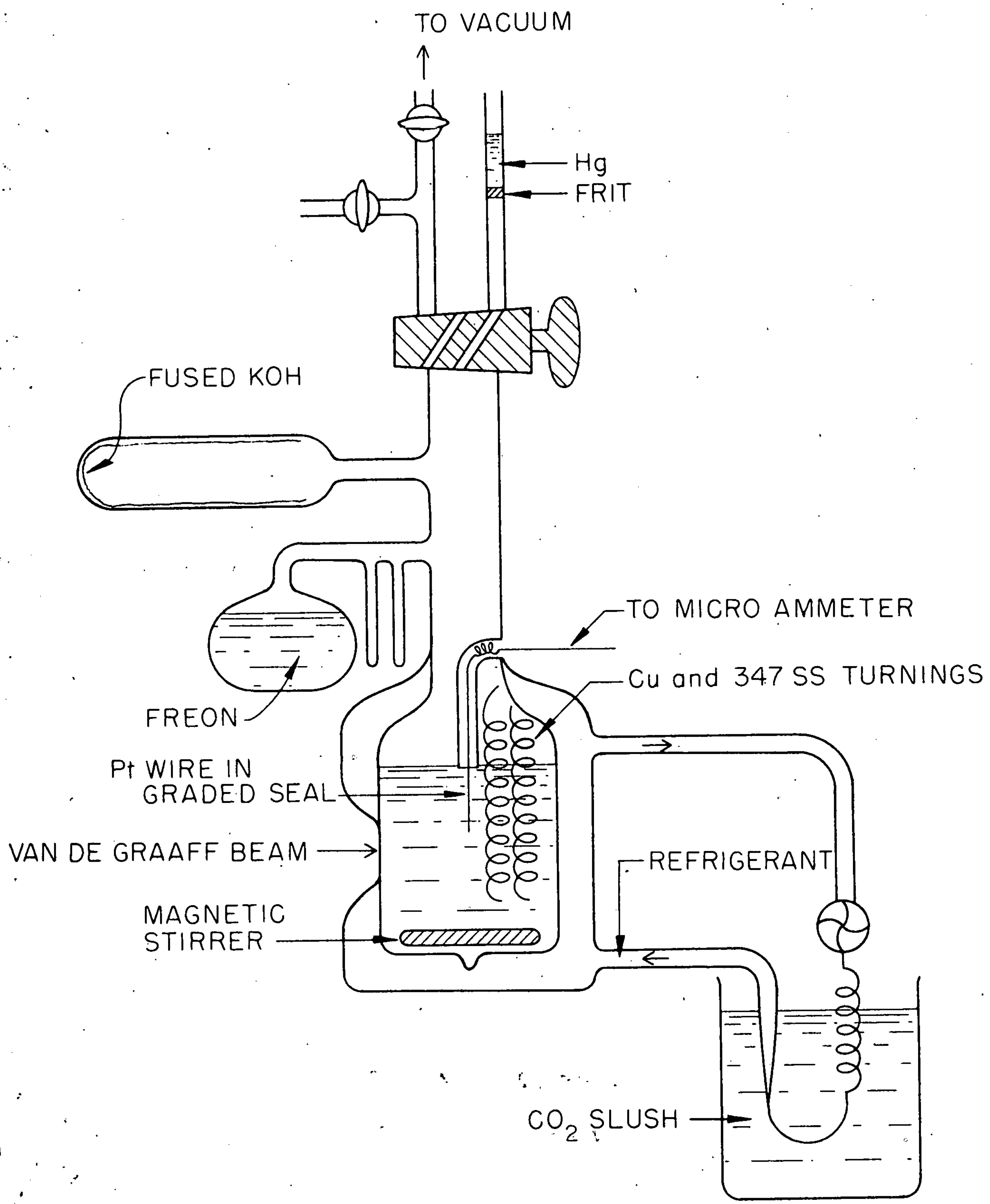

FIGURE 1
IRRADIATION APPARATUS