

## Letters to the Editors

### An Upper Limit for the Effective Capture Cross Section of $\text{Po}^{210}$ for Thermal Neutrons

Measurements were carried out to determine the thermal-neutron-capture cross sections of 138-day  $\text{Po}^{210}$  for the formation of both isomers of the capture product,  $\text{Po}^{211}$ . Although large quantities of  $\text{Po}^{210}$  have been widely used for many years and although it would be formed in large quantities by radiative capture in any reactor using bismuth or its compounds as a reactor coolant or fuel carrier, no measurement of its cross section seems to have been made.

The 0.52-sec isomer of  $\text{Po}^{211}$  emits primarily 7.43-Mev alpha particles, and the 25-sec isomer largely 7.14-Mev alpha particles, with several more energetic alphas in low abundance. These alpha groups are all sufficiently energetic to be readily distinguishable from the 5.30-Mev alphas of  $\text{Po}^{210}$ . The irradiations were carried out in the pneumatic tube facility of the Oak Ridge Research Reactor, where a maximum thermal flux of about  $7 \times 10^{13}$  neutrons  $\text{cm}^{-2}$   $\text{sec}^{-1}$  is available and where the ratio of thermal flux to resonance flux per  $\ln E$  interval is about 25. The alpha particles were measured with a silicon semiconductor radiation detector of the surface barrier type (capable of 1% energy resolution for  $\text{Po}^{210}$  alphas) and counted through a filter sufficiently thick to just absorb the  $\text{Po}^{210}$  alphas. The energy calibration was made with a radium source using  $\text{Po}^{214}$  (Ra C') of 7.68 Mev and  $\text{Po}^{218}$  (Ra A) of 6.00 Mev alphas. Transit time from the face of the reactor to the detector was about 2 sec; no chemical separations were required.

Using sources of  $\text{Po}^{210}$  ranging in intensity up to about 20  $\mu\text{C}$  and counted in geometry configurations as high as  $\sim 30\%$ , no evidence was found for the production of either of the isomers of  $\text{Po}^{211}$  (or for that matter of any alpha emitting product of energy in excess of  $\sim 6.0$  Mev). On the basis of a capacity to detect activity levels of the product  $\text{Po}^{211}$  equal to that of the background, upper limits were set for the activation cross sections of  $\text{Po}^{210}$ . The thermal cross sections were thus shown to be  $< 30$  mb for the formation of the 0.52-sec isomer and  $< 0.5$  mb for the 25-sec isomer.

A low cross section for  $\text{Po}^{210}$  is not unexpected, since it contains a closed shell of 126 neutrons and one pair of protons beyond the closed shell of 82. The isotones of  ${}_{84}\text{Po}_{126}^{210}$ , namely  ${}_{82}\text{Pb}_{126}^{208}$  and  ${}_{83}\text{Bi}_{126}^{209}$  have similarly low thermal cross sections: 0.6 and 34 mb, respectively (1). It is also of interest to note that radiative capture with thermal neutrons is exoergic by 4.5 Mev for the 0.52-sec and 3.2 Mev for the 25-sec isomers of  $\text{Po}^{211}$ . These low energies for radiative capture (in contrast to the more usual  $\sim 6$  Mev in the heavy mass region) again reflect the enhanced stability of the closed shell nuclide. This enhanced stability of the closed shell tends to lead to wider level spacings and, consequently, to lower thermal cross sections.

### REFERENCES

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J. HALPERIN  
J. H. OLIVER

*Chemistry Division  
Oak Ridge National Laboratory\*  
Oak Ridge, Tennessee  
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### Radiation Damage to Freon-11, $\text{CCl}_3\text{F}$

Numerous inquiries concerning Freon-11 ( $\text{CCl}_3\text{F}$ ) for use as a refrigerant in radiation fields, have prompted the authors to publish experimental results previously reported (1) in a laboratory memorandum.

The radiation stability of some organic compounds containing a single halogen had been summarized in a review by Tolbert and Lemmon (2). Davidge (3) had given some data for the production of fluorine induced by reactor irradiations of  $\text{CF}_4$  at about  $70^\circ\text{C}$ . Data for the effect of temperature on G values of a multihalogenated organic compound ( $\text{CCl}_3\text{Br}$ ) were reported by Firestone and Willard (4). However, no specific information was available for compounds of this type containing fluorine.

Freon-11 was used as a refrigerant in an early ORNL reactor installation, the Homogeneous Reactor Experiment (1952). No data were obtained on its stability. However, the Freon was subjected to relatively low radiation dosage. When the same refrigeration system was considered for use in the Homogeneous Reactor Test (1955), to be subjected to radiation fields 100-fold greater than those previously encountered, the question of radiation damage to Freon-11 and possibly subsequent corrosion in a mixed metal system was considered. A mass spectrographic cracking pattern of technical grade Freon-11, performed in this Laboratory (5), showed no parent ion ( $\text{CCl}_3\text{F}^+$ ). The relative abundances of the most prevalent ions were  $\text{CCl}_2\text{F}^+$  (100),  $\text{Cl}^+$  (15),  $\text{CClF}^+$  (10.6) and  $\text{CCl}^+$  (7.0).<sup>1</sup> This indicated

\* Operated by Union Carbide Corporation for the U. S. Atomic Energy Commission.

<sup>1</sup> The cracking pattern of Freon-22,  $\text{CHClF}_2$ , indicated that the parent ion was less than 0.9% of all the ions observed from  $m/e$  13 through 88 (5).

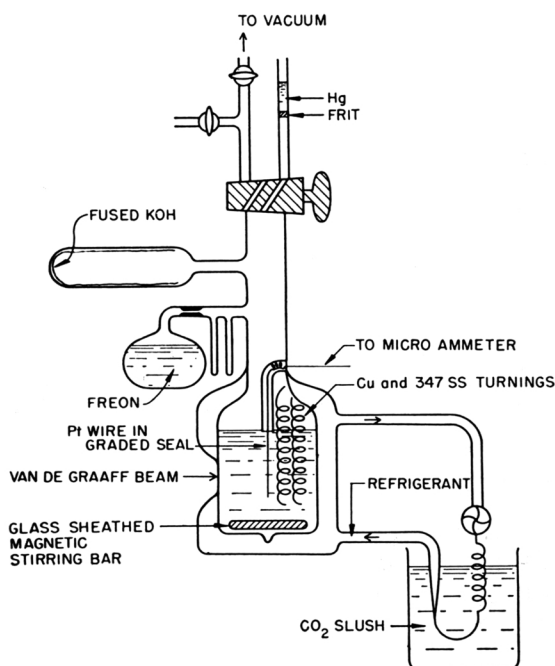


FIG. 1. Irradiation apparatus

that halogen and halogenated ions might present a serious corrosion problem. Hence a radiation experiment was conducted to determine the decomposition products and their corrosive effects under conditions approximating those anticipated for the reactor cell.

The radiation cell (Fig. 1) was constructed of Pyrex. It was cooled to about  $-70^{\circ}\text{C}$  where the vapor pressure of the liquid Freon is about 4 mm, by circulating chilled trichloroethylene around the cell. Degreased copper and stainless steel turnings were included in the cell so that the metals were 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.

The Freon was irradiated for 28 min using an electron beam of  $5\ \mu\text{amp}$  at 1.5 Mev, supplied by a Van de Graaff accelerator. 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 their surfaces; both the wash solution and the metal turnings were analyzed for chloride and fluoride.

The Freon received a radiation exposure of about  $2.6 \times 10^{21}$  ev/ml. A total of 28.3 mg of chlorine (as  $\text{Cl}^-$ ) and 4.1 mg of fluorine (as  $\text{F}^-$ ) formed during the irradiation reacted with the stainless steel and copper turnings, as well as with the fused KOH in the system. Approximately half of the halogen formed was found in the KOH trap. The surfaces of the metal turnings were completely covered with a yellowish-green deposit, indicative of metal halides. Mass spectrometric analysis of the irradiated distillate indicated traces of  $\text{SiF}_4$  probably resulting from reaction of a reactive form of fluorine with the glass wall.

The  $G$  values for the production of chlorine and fluorine atoms formed per 100 ev absorbed are 0.62 and 0.16, respectively. The value for fluorine compares favorably with twice the reported  $G$  value of 0.20 at  $-78^{\circ}\text{C}$  for the production of  $\text{Br}_2$  from solid  $\text{CCl}_3\text{Br}$  (4), especially when the C-F and C-Br bond energies are considered. These values are 105.4 and 65.9 kcal/mole (6), respectively. Extrapolation of our results to room temperature in accordance with the data of Firestone and Willard (4) indicate that the  $G$  value of 0.78 (for total halogen atom formation) would be raised by an order of magnitude.

Our results for fluorine are also in agreement with Davidge's (3) value,  $G(\text{F}) = 0.56$ , for  $\text{CF}_4$  when one corrects for the 1:4 ratio of fluorine in the two compounds.

The results of this experiment would indicate that Freon-11 is not stable nor is it compatible with copper or stainless steel in a radiation field. The authors' opinion is that any of the Freons would decompose in a strong radiation field and would present corrosion problems in ordinary metal systems.

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M. D. SILVERMAN\*  
B. O. HESTON†  
P. S. RUDOLPH

Chemistry Division  
Oak Ridge National Laboratory‡  
Oak Ridge, Tennessee

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### Determinations of the Total Emissivity of Polished and Oxidized Uranium Surfaces§

Calculations of the ignition (1) and burning temperatures (2) of uranium have been carried out as part of an intensive

\* Now with Reactor Chemistry Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee.

† Summer participant, 1956, from the Chemistry Department, University of Oklahoma, Norman, Oklahoma.

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