X-822

UNCLASSIFIED

ORNL MASTER COPY

OAK RIDGE NATIONAL LABORATORY
Operated By
UNION CARBIDE NUCLEAR COMPANY

UCC

POST OFFICE BOX P
OAK RIDGE, TENNESSEE

ORNL
CENTRAL FILES NUMBER
55-9-163

COPY NO. 78

DATE: September 30, 1955

SUBJECT: Radiation Damage To Freon

TO: Distribution

FROM: C. Michelson

Distribution

1. HRP Director's Office, Room 259, Bldg. 9204-1
2. G. M. Adamson
3. R. E. Aven
4. S. E. Beall
5. E. G. Bohlmann
6-7. R. B. Briggs
8. F. R. Bruce
9. R. H. Chapman
10. R. D. Cheverton
11. H. C. Claiborne
12. C. J. Claffey
13. E. L. Compere
14. F. L. Culler
15. H. N. Culver
16. N. W. Curtis
17. W. K. Eister
18. D. E. Ferguson
19. T. B. Fowler
20. W. R. Gall
21. H. E. Goeller
22. C. B. Graham
23. J. C. Griess
24. J. J. Hairston
25. D. C. Hamilton
26. P. N. Haubenreich
27. P. W. Hembree
28. J. W. Hill
29. H. K. Jackson
30. G. H. Jenks
31. R. W. Jurgensen
32. S. K. Kaplan
33. P. R. Kasten
34. A. S. Kitzes
35. J. O. Kolb
36. R. B. Korsmeyer
37. J. E. Kuster
38. J. A. Lane
39. M. C. Lawrence
40. M. I. Lundin
41. R. N. Lyon
42. T. H. Mauney
43. C. Michelson
44. R. G. McGrath
45. J. R. McWherter
46. E. C. Miller
47. R. L. Moore
48. E. O. Nurmi
49. L. F. Parsley
50. M. L. Pickelsimer
51. L. R. Quarles
52. R. C. Robertson
53. J. N. Robinson
54. W. Robinson
55. M. W. Rosenthal
56. W. L. Ross
57. H. C. Savage
58. H. K. Search
59. C. H. Secoy
60. C. L. Segaser
61. I. Spiewak
62. J. A. Swartout
63. W. F. Taylor
64. W. Terry
65. T. H. Thomas
66. M. Tobias
67. D. S. Toomb
68. W. E. Unger
69. R. VanWinkle
70. A. M. Weinberg
71. F. C. Zapp
72. P. C. Zmola
73-74. REED Library
75-77. Laboratory Records
78. ORNL-RC Lab Records

NOTICE

This document contains information of a preliminary nature and was prepared primarily for internal use at the Oak Ridge National Laboratory. It is subject to revision or correction and therefore does not represent a final report.

UNCLASSIFIED

RECOMMENDATIONS

This study predicts the decomposition of Freon-11 by gamma radiation, resulting in the formation of ions which are incompatible with stainless steel. The possibility of a corrosion problem in the HRT cold traps and refrigeration system, and also the possibility of gas binding and heavy oil formation must therefore be considered.

An experimental investigation of the effects of gamma radiation on Freon and an evaluation of the resulting problems are recommended.

RADIATION DAMAGE TO FREON

The present HRT design specifies Freon-11 ($C\ Cl_3\ F$) as a coolant for the cold traps and process line freeze jackets. A question has been raised as to whether Freon will be subject to radiation damage, and, if so, what effect it will have on the refrigeration system.

Radiation damage studies on similar organic materials (APEX-172) indicate that an organic structure such as F-11, $\left[Cl\text{-}\overset{\overset{Cl}{|}}{\underset{\underset{Cl}{|}}{C}}\text{-}F\right]$, can be broken down by ionizing radiations. When this occurs some of the products which form might be corrosive. Of particular concern is the formation of ions, such as F^+, which can attack the stainless steel cold traps. Other products to be expected are light gases which might bind the refrigeration system unless particular provision for venting is made, and possibly heavy oils which could accumulate in the freeze jackets and reduce their efficiency.

The principal sources of ionizing radiation in the cold traps will be background gammas from the reactor cell and decay gammas from fission product gases in the cold traps. Most of the damage to Freon will result from ionization caused by Compton scattering of the harder gamma rays.

A search of available literature has been made. There are no significant references to this problem. Some early investigations with 15-Mev deuterons on F-12 were conducted by the University of Chicago Metallurgical Laboratory (CC-238, CC-465), but no detailed results were reported. In 1952 the Radiation Laboratory at the University of Pittsburg conducted a number of gamma radiation studies on materials of interest to army ordinance. F-12 was included in the study but the only results reported were words to the effect that decomposition did occur, accompanied by some sign of corrosive gas formation (NP-5218).

An interesting experience with radiation damage to Freon was encountered recently in a gamma radiation facility at the Dugway Proving Ground (Army)

near Arco. J. R. McWherter of the Radiation Corrosion Group visited the installation and brought back the following account of their problems.

The gamma facility at Dugway utilizes eight spent MTR fuel elements arranged in a horizontal, cylindrical array to give about 1.5×10^6 r/hr of gammas along the axis. A stainless steel cooling jacket is placed around the periphery of this array to keep the entire facility at below 32°F.

Initially the facility was cooled by a refrigeration system consisting of a Freon primary loop removing heat from a closed secondary loop through an intermediate heat exchanger. The secondary loop contained ethylene glycol which circulated through the freezing jacket. This system became inoperative due to breakdown of the ethylene glycol.

A second plan was considered using a single-loop Freon refrigeration system connected directly to the jacket. The possibility of radiation damage to Freon was anticipated so a small sample of Freon gas mixed with silver nitrate was placed in the gamma facility for test. A silver chloride precipitate soon formed indicating breakdown of the Freon. However, on the recommendation of a Brookhaven Laboratory consultant it was decided to carry through the plan using Freon. This recommendation was based on the belief that a back reaction would recombine the Freon as rapidly as it was destroyed.

The second system was built using the single Freon loop. After about ten days of operation the cooling began to fall off and the compressor started to overheat. Upon examining the system it was found that oil in the Freon was decomposing, and even the Freon showed signs of decomposion. Also, the soldered joints in the dryer were being destroyed. The copper did not appear to be corroded. The stainless steel jacket was not examined. The recombination of Freon did not occur as expected, perhaps due to the oil picking up free ions before they could recombine. The solder corrosion might be attributed to the use of an acid flux which later combined with water to

attack the joints, or an attack by F^+ ions or even HF that may have been formed.

The system was next rebuilt using a Freon primary loop removing heat from a closed secondary loop of ethyl alcohol. This system has operated satisfactorily for several weeks although there is some evidence of radiation damage.

Samples of the dryer material and oil from the Freon system which failed are available at Y-12. Also available is a sample of the ethyl alcohol which has been exposed for about three weeks in the new system.

The problem of radiation damage to Freon in cold traps was not encountered during operation of the HRE. However, the HRE developed a total of only 580 Mw hr of heat--the equivalent of 580 hrs at full power operation. The HRT is expected to operate several thousand hours at full power and handle considerably larger quantities of radioactive off-gases. Therefore, the total radiation dose received by the Freon will be much larger.

The gamma radiation absorbed by Freon in the HRT cold traps will be about 85 rads/sec when the reactor is operating at 10 Mw. The Freon will contain less than 100 ppm of water. It will reside in the cold traps an average of 14% of its circulating time. According to MDDC-1449, the number of molecules reacting per 100 ev of energy absorbed is about four for most organic liquids. On this basis about 20% of the Freon molecules will undergo reactions in 8000 hrs of operation. The actual type and degree of Freon decomposition, however, remains to be determined experimentally.

CALCULATIONS

Figures are available from CF-55-7-141 on the number of gamma ray photons emitted in the cold traps when the reactor is operating at 10 Mw. Since the pipe jacket covers 56 inches of the 78-inch cold trap it is assumed that 72% of the photons enter the pipe wall under the jacket. It is further assumed that the photons are evenly distributed and are traveling in a radial direction, their only attenuation being due to energy absorption in the pipe walls and Freon. Gamma ray energy absorption coefficients are therefore used. As a further simplification, the pipe walls and annular region in the jacket are treated as parallel planes with the gammas passing through perpendicularly.

It is assumed that all photons from a given emitter are given off with the same (average) energy. All gammas below 0.09 Mev are neglected because of the high cross section of iron for photoelectric encounters. The effects of bremsstrahlung and fast neutrons are neglected. The background gamma radiation is neglected in comparison with the internal sources.

The energy absorption coefficients for C, Cl, and F (Freon) in the 0.5-Mev range are assumed to be similar to C, N, and O--about 0.03 cm^2/g. At 0.17 Mev they are approximately 0.025 cm^2/g. The iron in the walls (0.6 cm thick) exponentially attenuates the photons with an energy absorption cross section of 0.22 cm^{-1} in the 0.5 Mev range and 0.59 cm^{-1} at 0.17 Mev. Those photons which reach the Freon are absorbed according to the equation:

$$E_{abs} = \rho \, \phi_\gamma \, \mu_\gamma \, E_\gamma$$

$$(\text{Mev/cm}^3 \text{ sec}) = (\text{g/cm}^3)(\text{photons/cm}^2 \text{ sec})(\text{cm}^2/\text{g})(\text{Mev/photon})$$

Table I shows the value of flux (photons/cm^2 sec), attenuation (e^{-ux}), and absorption (Mev/cm^3 sec) which occur in the cold traps if the Freon density is 1.6 g/cm^3.

TABLE I

Energy Absorption in Freon

(Mev/cm^3 sec)

Source	Average Energy Mev	Wall Flux $x10^{-10}$	Wall Attenuation	Freon Flux $x10^{-10}$	Freon Absorption $x10^{-9}$
Kr 85	.17	4.5	.70	3.2	0.2
Kr 87	.56	14.7	.87	13.0	3.5
Rb 88	.49	23.3	.87	20.2	4.8
				Total:	8.5 (1)

This amount of energy absorption will cause a certain amount of damage to the Freon as illustrated in Table II.

TABLE II

Radiation Damage to Freon

(8000 hrs circulating time) (1)

Energy absorbed per sec	8.5×10^{15} ev/cm^3 sec
Reaction energy	25 ev/reaction
Reactions occurring per sec	3.4×10^{14} $reactions/cm^3$ sec
Total reactions	1.4×10^{21} $molecules/cm^3$
Original Freon present	7.1×10^{21} $molecules/cm^3$
Percentage reacted	20%

(1) Freon resides in the cold traps an average of 14% of its circulating time (R. C. Robertson).

Carlyle Michelson

C. Michelson

alg