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°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 μamp 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 2.6×10^{21} ev/ml. 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. 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 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°C for the production of Br_2 from solid CCl_3Br (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, $G(F) = 0.56$, for 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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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

study of metal combustion. In those studies it was necessary to estimate the heat losses due to radiation, and reliable values for the total emissivities of polished and oxidized uranium surfaces were required. Since no satisfactory values were found in the literature, the experiments described below were carried out.
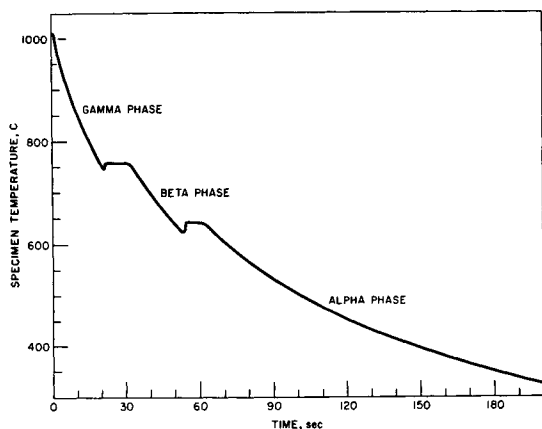

FIG. 1. Typical cooling curve for uranium cube. (Run #13, weight = 10.54 gm, area = 4.16 sq cm.)

The total emissivity measurements were made using nominal 1-cm cubes of high purity uranium metal (approximately 250 ppm total analyzed impurities). A $\frac{1}{16}$-in. hole was drilled in one face to the center of the cube. The cube surfaces were then polished to a 1-μ diamond paste finish. A 10-mil Pt-Pt, 10% Rh thermocouple was spotwelded to the bottom of the hole, and the polished cube was suspended by the insulated thermocouple wires in a Pyrex bulb, which was evacuated to a pressure of about 3×10^{-7} mm Hg.

The cube was heated by an external induction coil to a temperature somewhat below the melting point of 1133°C. The induction power was then turned off suddenly, and thermocouple voltage versus time was recorded on a high speed millivolt recorder. A typical cooling curve is shown in Fig. 1.

It was assumed that all heat loss was by thermal radiation from the cube surface. The rate of heat loss by thermal radiation can be expressed by means of the Stefan-Boltzmann law as follows (symbols are defined in Table I):

$$[\text{Heat loss rate}] = \epsilon \sigma A (T^4 - T_a^4). \tag{1}$$

The rate of sensible heat loss by the sample is

$$[\text{Heat loss rate}] = -WC_p \left(\frac{dT}{dt}\right). \tag{2}$$

TABLE I

LIST OF SYMBOLS

A	Area of radiating surface, sq cm
C_p	Specific heat of metal, cal/(gm) (°K)
ΔH	Heat of phase transformation, cal/gm
T	Temperature of specimen, °K
T_a	Ambient temperature, °K
t	Time, sec
W	Mass of test specimen, gm
α, β, γ	Solid phases of uranium metal
ϵ	Total emissivity
σ	Stefan-Boltzmann constant; 1.356×10^{-12} cal/(sec)(sq cm)(°K^4)

TABLE III

AVERAGE TOTAL EMISSIVITY VALUES FROM TABLE II[a]

Surface condition	γ Phase ($\sim$1000 to 760°C)	β Phase (760 to 643°C)	α Phase (643 to 400°C)
Polished	0.37	0.36	0.33
Oxidized $\sim$700 μg O$_2$/sq cm	0.61	0.61	0.58
Oxidized $\sim$1000 μg O$_2$/sq cm	0.73	0.71	0.68

[a] Values based on phase transformations are not included in these averages.

TABLE II

TOTAL EMISSIVITY VALUES FOR URANIUM AND URANIUM OXIDE

Run no.	Surface condition	γ Phase ($\sim$1000 → 760°C)		γ, β transition	β Phase (760 → 643°C)		β, α transition	α Phase (643 → 400°C)
		Slope method	Two-point method		Slope method	Two-point method		Slope method
6	Polished	0.34	0.33	0.34	0.34	0.33	0.31	0.31
7	Polished	0.36	0.37	0.37	0.36	0.36	0.34	0.33
8	Polished	0.38	0.40	0.39	0.36	0.37	0.36	0.34
9	Polished	0.40	0.40	0.40	0.38	0.38	0.37	0.35
12	Oxidized $\sim$700 μg O$_2$/sq cm	0.60	0.62	0.61	0.60	0.61	0.59	0.59
13	Oxidized $\sim$700 μg O$_2$/sq cm	0.60	0.61	0.62	0.61	0.61	0.61	0.58
10	Oxidized $\sim$1000 μg O$_2$/sq cm	0.74	0.76	0.83	0.69	0.71	0.75	0.68
11	Oxidized $\sim$1000 μg O$_2$/sq cm	0.68	0.73	0.74	0.71	0.72	0.68	0.69

Equating the two above expressions, and solving for total emissivity, ϵ, yields:

$$\epsilon = \frac{(-dT/dt)}{(T^4 - T_a^4)} \frac{WC_p}{A\sigma} \quad (3)$$

This is designated the "slope method" as it depends on a value of the slope of the cooling curve.

An alternative method is to equate Equations (1) and (2), and obtain the following differential equation:

$$\left(-\frac{dT}{dt}\right) = \frac{\sigma A}{C_p W} \epsilon (T^4 - T_a^4) \quad (4)$$

If it is assumed that the quantity $(\sigma A/CpW)$ ϵ is independent of temperature, and that $T^4 \gg T_a^4$, one can solve the above differential equation, and obtain the following expression for ϵ:

$$\epsilon = \frac{1}{3} \frac{C_p}{\sigma} \frac{W}{A} \left[\frac{1}{t_2 - t_1}\right]\left[\frac{1}{T_2^3} - \frac{1}{T_1^3}\right] \quad (5)$$

This is designated the "two-point method" since it requires time and temperature values from two points on the cooling curve. The resulting value is an average value between T_1 and T_2.

A third method is to calculate emissivity from the time of an arrest in the cooling curve due to a phase transition. This would be calculated by the expression:

$$\epsilon = \frac{\Delta H/\Delta t}{T^4 - T_a^4} \frac{W}{A\sigma} \quad (6)$$

Four runs were made directly with highly polished cubes. The finish appeared somewhat mottled after only slight heating, presumably because of anisotropic growth. Four runs were made in which the polished specimens were deliberately oxidized. Oxidation was achieved by introducing measured quantities of pure oxygen to the Pyrex bulb and heating the specimen in place to about 400°C for approximately ten minutes. Following oxidation, the bulb was evacuated and the run started. In this procedure the oxidized surface suffered no damage from handling.

Tables II and III give the results. The emissivity values given under "slope method" are the average of four or more values calculated by Eq. 3 in the temperature region in question. The "two-point" values were computed between the high and low temperature values in the γ and β phase regions.

Cubes oxidized to 20,000 μg O_2/sq cm resulted in excessive flaking and surfaces covered with loosely clinging flakes and powdery material evidently acting as radiation shields. Therefore, no data is reported for heavily oxidized specimens.

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Note on Spectrum Hardening for Non 1/v Cross Sections

Effective thermal neutron cross sections are well known to depend on the thermal neutron spectrum which is determined by the moderator and neutron absorbers of the medium. While the detailed absorption properties have been accurately measured, determination of the moderator scattering kernel represents a more difficult problem. For an H_2O moderator, considerable success in duplicating measured neutron spectra has been achieved with both a free proton model (1) and with the Nelkin kernel (2, 3). However, both of these models represent somewhat crude approximations. A detailed experimental investigation of the kernel is being carried out (4, 5). In this connection it is necessary to understand for what values of the parameters, including the incident neutron energy, the kernel is most important.

The point of the present note is to present some evidence that for non 1/v absorbers the kernel for incident neutron energies of 0.625 ev and higher energies is significant.

The energy 0.625 ev is a conventionally used value for the upper limit of the thermal group, and the effect of the kernel for energies above this value appears in a solution of the transport equation as a source term. The source terms for the free proton model and the Nelkin model have different energy dependence, so that the sensitivity of the spectrum to the source term may be investigated by using the source for one model with the kernel (for energies below 0.625 ev) for the other model.

For a moderator with $kT_M = 0.0255$ ev, the free proton model has a source term with an energy dependence proportional to erf $[(E/kT_M)^{1/2}]$. The Nelkin model source has an energy dependence given by the same form of function but with a different value of the temperature parameter;

TABLE I

MACROSCOPIC ABSORPTION CROSS SECTIONS (cm⁻¹) FOR DIFFERENT KERNELS AND SOURCE SPECTRA

Composition	Free proton	Diff.	Nelkin kernel free proton source	Diff.	Nelkin kernel Nelkin source
Pure H_2O	0.01913	0.5%	0.01905	0.13%	0.01903
B^{10}, H_2O, 3.15 b	0.1546	3.4%	0.1508	0.9%	0.1494
B^{10}, H_2O, 6.04 b	0.2642	4.8%	0.2556	1.3%	0.2522
Cd, H_2O, 2.67 b	0.1940	0.4%	0.1942	0.5%	0.1933
Cd, H_2O, 5.4 b	0.3712	1.1%	0.3712	1.1%	0.3673
Cd, H_2O, 9.6 b	0.5898	2.3%	0.5868	1.8%	0.5762
Cd, H_2O, 15.4 b	0.8183	3.8%	0.8089	2.6%	0.7882
Sm^{149}, H_2O, 1.97 b	0.1518	1.6%	0.1509	1.0%	0.1494
Sm^{149}, H_2O, 3.67 b	0.2548	2.7%	0.2522	1.7%	0.2481
Sm^{149}, H_2O, 6.20 b	0.3682	4.1%	0.3621	2.4%	0.3536
Sm^{149}, H_2O, 8.71 b	0.4517	5.2%	0.4418	2.9%	0.4293