

Radiation Cleanup of Vacuum Systems

(Radiation-Induced Outgassing)

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Abstract

A stainless steel-304 vacuum system has been designed and constructed to study radiation-induced outgassing when this material is exposed to cobalt-60 gamma radiation. The system is pumped with an ion pump and sorption roughing pump. No foreign materials have been introduced except for copper seals at the flanges. An analytical model has been developed which predicts the outgassing from SS-304 to be 8.27×10^{-12} torr-liters/(cm²)(sec) per megarad/hr. Extrapolation of existing data for aluminum suggests a lower value of 1.0×10^{-12} torr-liters/(cm²)(sec) per megarad/hr. Experiments to determine the value are currently in progress.

Introduction

The long range goal of the Fusion Division of the Energy Research and Development Administration (ERDA) is the successful generation of power through the fusion of light nuclei. One of the prominent problems in fusion is the influence of radiation from a plasma on the first wall (plasma cladding) and related structural materials. The evaluation and resolution of these factors may well dictate the criteria which must be satisfied in order to maintain a plasma "burn" which will not be quenched by impurities.⁽¹⁾ The vacuum vessel that contains the plasma should be at UHV (about 10^{-7} Pa) for start up.⁽²⁾ A pressure of this same magnitude must be maintained and contamination from heavy (high atomic number) ions must be prevented since the radiation losses are proportional to the square of the atomic number of the ion in the plasma.

It has been shown in two experiments by a group at the National Bureau of Standards that the outgassing effect is due primarily to electron and gamma flux and not the neutron flux.⁽³⁾ These important data allow experiments to be performed with gamma radiation alone. In the present project a stainless steel-304 system is being irradiated with cobalt-60 gamma rays and the attendant outgassing measured. The ultimate goal is to obtain a yield fraction Δ for SS-304 under high energy radiation:

$$\begin{aligned}\Delta &= \frac{\text{gas atoms released per unit area per unit time}}{\text{energy absorbed per unit time per unit mass}} \\ &= \frac{\text{torr-liters}/(\text{cm}^2)(\text{sec})}{\text{megarad/hr.}}\end{aligned}$$

These data will be important in the design of vacuum pumping systems for the future fusion reactors.⁽⁴⁾

Objectives of the Current Program

The principal objectives of the current research program are:

- (1) The design, construction, and procurement of the equipment necessary to determine the yield fraction Δ for SS-304,
- (2) The development of an analytical model of radiation-induced outgassing,
- (3) The experimental determination of Δ , and
- (4) Comparison of the magnitude of gamma-induced outgassing with ordinary thermal-induced outgassing.

Objection (4) has broader implications. If radiation induced outgassing is comparable to thermal induced outgassing, then radiation could be used for systems where baking is not possible. This could be especially useful when the system has an unusual geometry which prevents adequate baking methods.

To date, objectives (1) and (2) have been met. The program is essentially on schedule and it is anticipated that objectives (3) and (4) will be met by the conclusion of the current program period.

Design of the Vacuum System

In the design of the vacuum system, several criteria were deemed to be of high importance:

- (1) The system should be constructed entirely of SS-304, even in the regions not exposed to gamma radiation.
- (2) The system should have accurate means of determining the outgassing rate, not only in total, but by gas species.

- (3) The system should be "clean" with respect to the possible introduction of foreign species.
- (4) The surface area exposed to the gamma radiation should be as large as possible compared to the surface area of the remainder of the system.
- (5) The entire system should be capable of thermal bakeout to 300 C, with control of the bakeout temperature.
- (6) The system must be capable of being inserted under high vacuum conditions into the cobalt-60 source located in a water pool.

With these principal criteria, the system that resulted is sketched in Figure 1 and 2. The components are detailed by manufacturer in Table I. This system meets the previously-stated criteria as follows:

- (1) All piping, valves, tees, etc., are constructed of SS-304. The only foreign metal introduced is high purity copper used for the gaskets at the flanges in the system. Copper is required because it "flows" to make the high-vacuum seal, whereas stainless steel does not.
- (2) As shown in Figure 2, a mass spectrometer is included in the system. By using a quadrupole mass spectrometer, not only can the gas species be identified, but changes, if any occur, can be monitored. It will be important to note the possible generation of gaseous species from inside the metal--species trapped during the formation

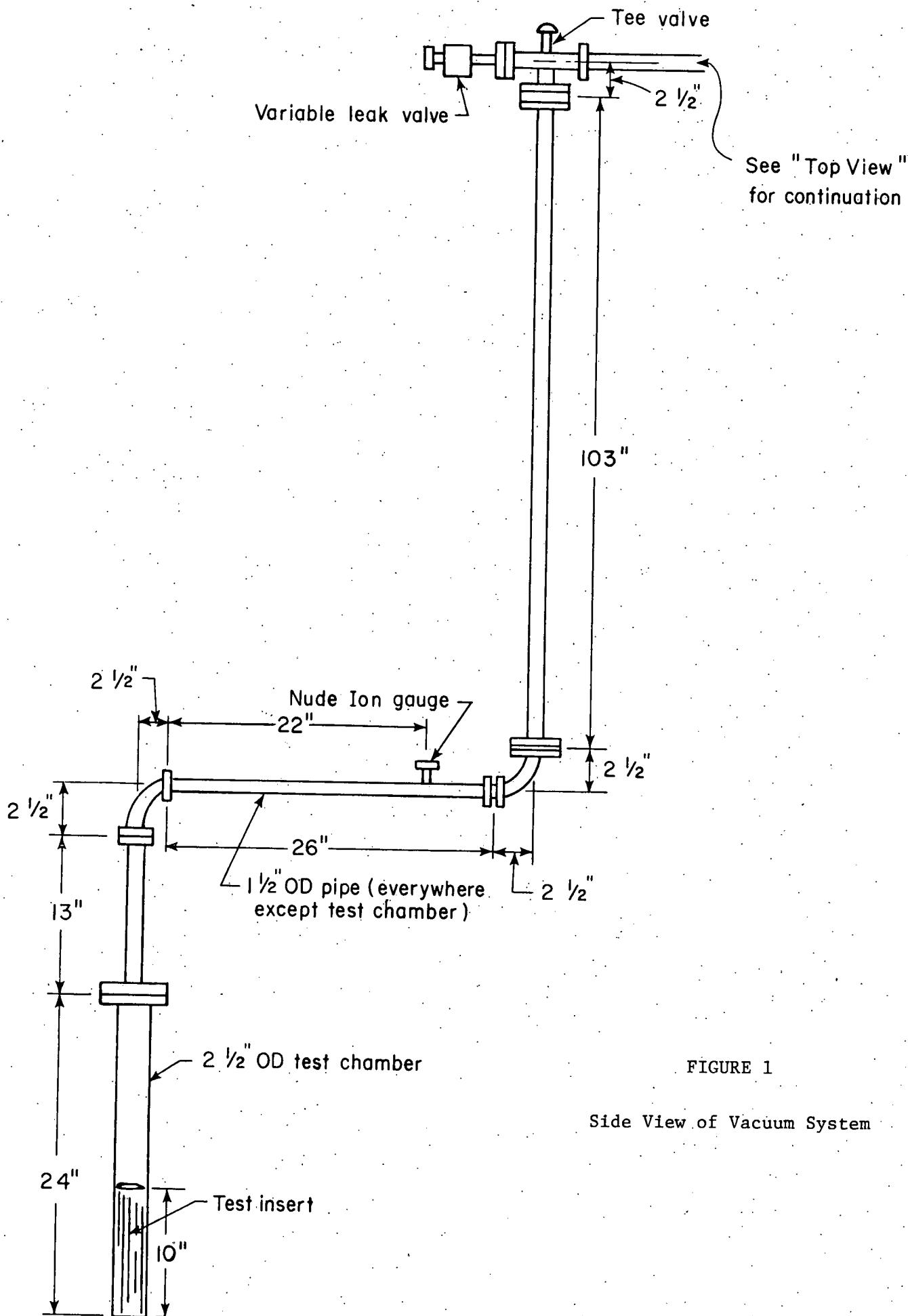


FIGURE 1

Side View of Vacuum System

Top View

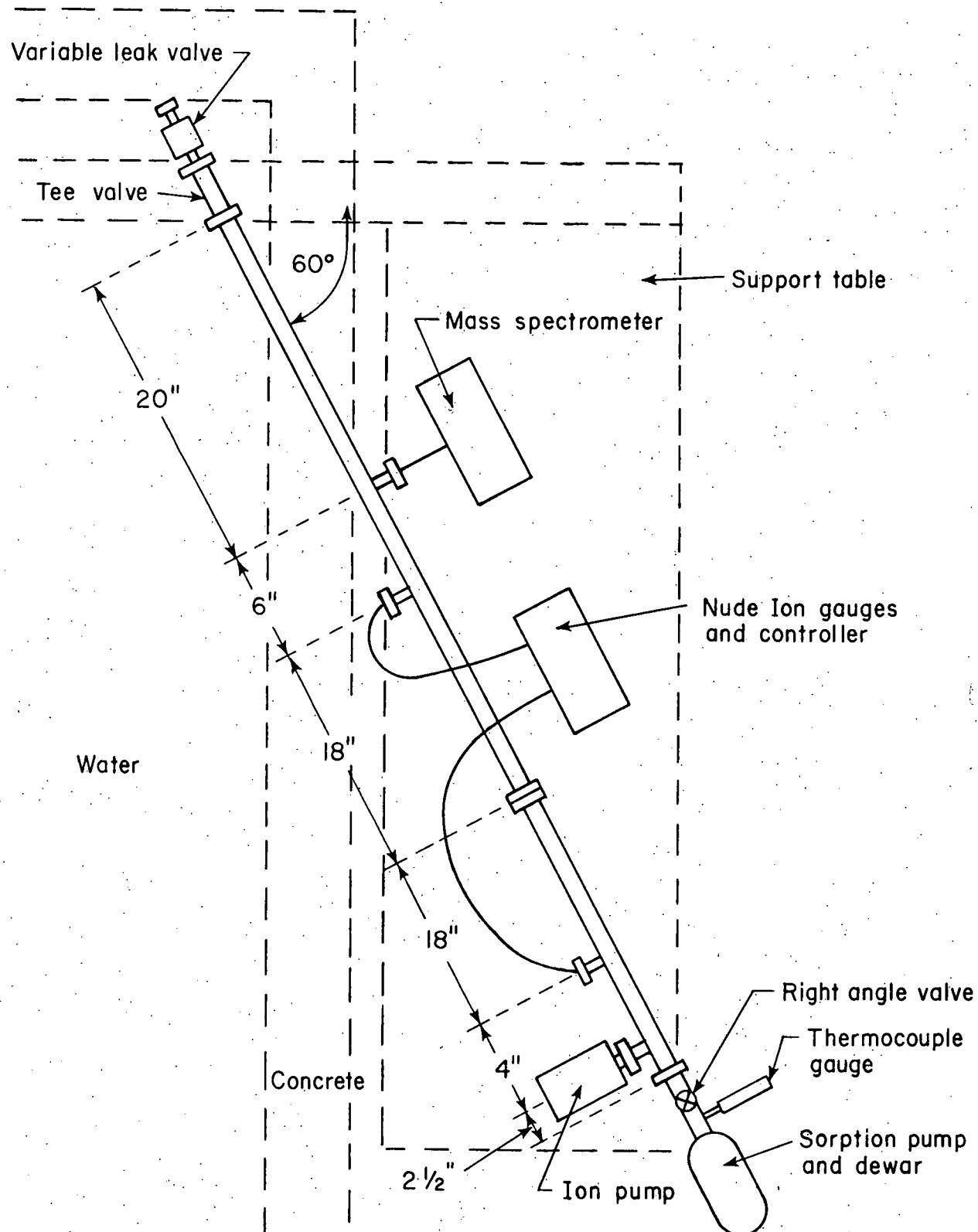


FIGURE 2

Top View of Vacuum System

of the metal. (6) If any high atomic number atoms are evolved, this could quench the fusion reaction in a fusion reactor. The gas evolution rate can be determined in several ways. First, as seen in Figure 2, two nude ion gauges are separated by 36 inches. By measuring the pressure difference between these gauges, from the calculated conductance, the gas flow can be determined. Second, it will be noted that there is a flanged joint between the two gauges. This permits the insertion of an orifice of accurately known conductance into the system. The flow can then be determined by the pressure reading.

- (3) Use of an ion pump with a sorption roughing pump eliminates possible oil or mercury contamination from forepumps and "standard" diffusion pumps.
- (4) As seen from Figure 1, the bottom 10 inches of the system, which is the region exposed to the intense gamma radiation, contains a test insert. It consists of 95 pieces of 1/8-in. SS-304 seamless tubing, each 10-in. long, held in a square array by SS-304 wire mesh. No foreign materials, such as would be contained in a weld, are introduced into the system. Further, the tubes are bevelled at the bottom and so as to approximate point contact with the test chamber, to avoid "infinite leaks" that result from close-tolerance area contact. The test insert and walls of the test

T A B L E 1

Vacuum System for Study of Radiation-Induced Outgassing

<u>Component</u>	<u>Manufacturer</u>	<u>Part Number</u>
1. Nude ion gauges (3)	Granville-Phillips	274-022
2. UHV ion gauge controller (2)	" "	271
3. Variable leak valve	" "	203
4. UHV tee valve (1-1/2")	Ion Equipment Corp.	BVV-153T
5. Ion pump	" "	IP-020
6. Ion pump controller	" "	PS-150
7. UHV right angle valve (1-1/2")	" "	BVV-152
8. Sorption pump	" "	SP-11
9. Dewar	" "	SPD-11
10. Mass spectrometer	Spectrum Scientific	SM-80
11. Thermocouple gauge	Comptech Inc.	TVT-1504
12. Thermocouple control	" "	300-00
13. Thermal bakeout jacket	Briscoe Company	---

chamber have a total surface area of 4600 cm² which is exposed to the intense gamma radiation. The remainder of the entire vacuum system has a surface area of 7500 cm². The S/V (surface-to-volume) ratio of the test insert is 39, and of the entire system including the insert, is 1.6.

- (5) All flanges and valves are bakeable. A custom-tailored bakeout jacket was constructed with variac control of the heaters (broken down into sub-sections for fine control) to enable bakeout of the entire system at various fixed temperature up to 300 C.
- (6) Since the experiments will be performed in the cobalt-60 pool, the vacuum system was designed to be inserted into the pool without breaking the vacuum. The mechanical design of the experiment includes a table which supports the entire vacuum system. The vacuum system is fastened to the table and can be lifted by an overhead winch suspended from an I-beam in the Laboratory. After being properly positioned, the vacuum chamber is lowered into the cobalt-60 region.

Analytical Model of Radiation-Induced Outgassing

In developing an analytical model for radiation-induced outgassing, it is assumed that the electrons produced in the material by the various gamma ray interactions (Compton effect, pair production and photoelectric effect) desorb atoms from the surface. Specifically, the model considers the following events:

- (1) Gamma rays produce electrons of average energy \bar{E} within the material. An isotropic distribution of electrons is assumed.
- (2) The electrons born within a mean range r of the surface travel an average distance \bar{r} to the surface losing energy in the process.
- (3) The electrons with reduced energy \bar{E}_p reach the surface and produce secondary electrons of average energy \bar{E}_s .
- (4) Both the primary and secondary electrons cause electron stimulated desorption of gas molecules from the surface.

From the measured gamma dose rate in the test chamber, the average flux of gamma $\bar{\phi}_\gamma$ can be calculated. The number of electrons produced per cm^3 of material is then

$$N = \Sigma \bar{\phi}_\gamma v \quad (1)$$

where Σ is the macroscopic cross section for electron production and v is the average number of electrons produced per interaction. Of the electrons produced within a mean range r of the surface, one fourth escape from the surface. The flux of primary electrons at the surface is thus

$$\phi_{ep} = \frac{rN}{4} = \frac{r\Sigma \bar{\phi}_\gamma v}{4} \quad (2)$$

These produce Δ_s secondary electrons per primary electron. The primary electrons desorb the gas atoms with a yield Δ (atoms/electron) and the secondary electrons desorb the gas atoms with a yield Δ' . Thus the number of gas atoms desorbed per unit area per unit time is

$$n = \frac{r \sum \bar{\phi} \gamma^v}{4} (\Delta + \Delta_s \Delta') \quad (3)$$

The average distance the electrons produced with energy \bar{E} (within range r from the surface) travel is $\bar{r} = 0.795 r$. The resulting attenuation in energy can be determined from range-energy curves for electrons. There are abundant data on low energy electron stimulated desorption,⁽⁷⁾⁽⁸⁾ but little data are available for electrons of the primary energy range of interest here. A rather wide range is found in the yield depending on the condition of the surface. All available data show that the yield from the surface decreases as a function of irradiation time. Table 2 summarizes some of the available data.⁽⁹⁾⁽¹⁰⁾⁽¹¹⁾

Predictions of Experimental Results

The previously described analytical model was applied to both aluminum and SS-304. The results of the calculations are summarized in Table 3 and compared with available experimental results. The first comparison is of the predicted electron flux at the surface. The value for SS-304 of 2.19×10^{-16} amps/cm² per R/hr is in good agreement with data on gamma-induced currents in a nuclear reactor environment.⁽¹²⁾ However, the value predicted for aluminum is a factor of 6.6 higher than the value measured for reactor gamma radiation. The second comparison is that of the predictions of gamma-induced outgassing for aluminum with the experimental result from the National Bureau of Standards.⁽³⁾ For this comparison, questionable values of $\Delta = \Delta' = 0.1$ were used, with $\Delta_s = 1.0$. The predicted result of 9.35×10^{-12} torr-liters/(cm²)(sec) per megarad/hr is almost a factor of 50 larger than the measured value. The predicted result for SS-304 is thus to be suspected as being too large. If one adjusts the NBS value for aluminum by the ratio of the measured

T A B L E 2

Surface Atom Desorption

<u>Electron Energy</u>	<u>atoms/electron</u>	<u>Surface Condition</u>	<u>Reference</u>	<u>Remarks</u>
.1 kev	1	dirty	Clausing (9)	Surface was a copper material with diffusion oil contamination. Clean represents a bake out for 20 hrs at 100-145 C.
	5×10^{-2}	clean	"	
	2	dirty	"	
	6×10^{-2}	clean	"	
10 kev	2.5	dirty	"	
	8×10^{-2}	clean	"	
<u>Photon Energy</u>	<u>p atoms/photon</u>			
30 kev	7.5×10^{-4}	non-discharge Cleaned	Brumbach and (10) Kaminsky	Surface was SS-304 but yield is for CO_2 only.
40 kev	6×10^{-4}	"	"	
50 kev	2×10^{-4}	"	"	
50 kev	6×10^{-5}	discharge, clean	"	
0.8 Mev (reactor radiation)	4×10^{-3}	unbaked, initial value	Dobrozemsky (11)	Surface was SS, aluminum and mu metal
0.8 Mev (reactor radiation)	4×10^{-4}		"	After 3.5×10^5 MRad.

T A B L E 3

Comparison of Results from Analytical Model
(Gamma energy = 1.25 Mev)

	Aluminum	SS-304
Average electron energy	.6 Mev	.6 Mev
Maximum range of electron	.76 mm	.25 mm
μ , macroscopic cross-section	$.1479 \text{ cm}^{-1}$	$.3975 \text{ cm}^{-1}$
$\mu\phi$, electrons/cm ³	$8.13 \times 10^9 / \text{cm}^3$	$2.18 \times 10^{10} / \text{cm}^3$
Average range of electrons in material	.60 mm	.198 mm
Average energy of electrons at surface of material	.19 Mev	.18 Mev
I_p , predicted electron flux at surface	$2.50 \times 10^{-16} \frac{\text{amps}}{\text{cm}^2 \text{ R/hr}}$	$2.19 \times 10^{-16} \frac{\text{amps}}{\text{cm}^2 \text{ R/hr}}$
I_p , experimental electron flux for a reactor spectrum of gamma radiation ¹²	$.38 \times 10^{-16} \frac{\text{amps}}{\text{cm}^2 \text{ R/hr}}$	$1-2 \times 10^{-16} \frac{\text{amps}}{\text{cm}^2 \text{ R/hr}}$
Gamma induced outgassing (Model Predictions)	$9.35 \times 10^{-12} \frac{\text{torr-liters}}{\text{cm}^2 \text{ sec M Rad/hr}}$	$8.27 \times 10^{-12} \frac{\text{torr-liters}}{\text{cm}^2 \text{ sec M Rad/hr}}$
Gamma induced outgassing (experimental) ³	$1.94 \times 10^{-13} \frac{\text{torr-liters}}{\text{cm}^2 \text{ sec M Rad/hr}}$	<u>Univ. of Cincinnati Experiment</u> ERDA-E(11-1)-4093

electron currents in a reactor environment, ⁽¹²⁾ the outgassing prediction for SS-304 decreases from 8.27×10^{-12} to 1.0×10^{-12} torr-liters/(cm²)(sec) per megarad/hr. Thus the two predictions of the experimental results for SS-304 are:

$$1.0 \times 10^{-12} \frac{\text{torr-liters}/(\text{cm}^2)(\text{sec})}{\text{Megarad}/\text{hr}} \quad (\text{extrapolation of NBS value})$$

and

$$8.27 \times 10^{-12} \quad " \quad (\text{model prediction})$$

Effort Devoted to the Program by the Principal Investigator

The Principal Investigator, Dr. J. N. Anno, has spent in excess of 17 per cent of his accountable efforts on the project since its inception on September 15, 1976. This rate of effort will continue until the expiration of the project on September 14, 1977.

Acknowledgement

The author wishes to thank Dr. Hatice Cullingford, Division of Magnetic Fusion Energy, U.S. Energy Research and Development Administration for her many positive suggestions and for her encouragement during the course of this work.

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