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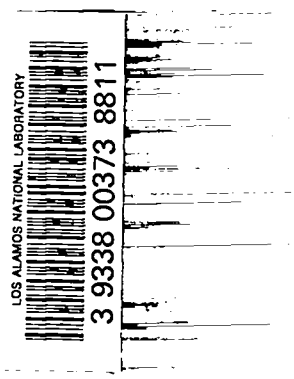
Fission Fragment Removal of Uranium from a Fissioning Surface: II



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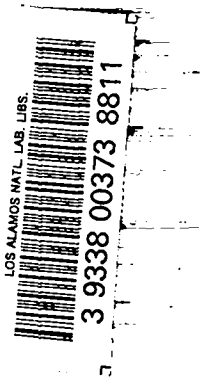

ABSTRACT

Equipment was designed and built for reactor irradiation of U metal in a controlled and measured vacuum with collection of the U atoms ejected on a surrounding Pt foil. At intervals, the U metal source was withdrawn and kept in vacuo while the Pt foil with the collected U became the high-voltage electrode of a fission counting chamber. Accurate measurements of the relative amounts of U collected on the Pt were made at intervals by fission counting techniques. The relative weight of U collected as a function of the neutron exposure of the U metal source was found to conform to an equation developed in Los Alamos Scientific Laboratory Report LA-3352-MS, which stated

$$M' = M_o \left(1 - e^{-\lambda' (\phi t)}\right) + c' (\phi t),$$

where M' is micrograms of U ejected, M_o is micrograms of U originally present on the source surface in UO_2 crystallites of a certain size, λ' is a removal constant for UO_2 crystallites of a given size, ϕt is the integrated thermal neutron flux seen by the source, and c' is a constant.

Exposure of the U metal source to moist He, after the rate of increase of M' had become linear with ϕt, resulted in an acceleration of the U deposition on the Pt with a gradual return toward the rate obtaining before treatment. It is evident from this that the surface condition of the U metal source is an important factor in the ejection of the U atoms from a fissioning surface.

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INTRODUCTION

Capsule experiments in which measurements have been made of the average number of U atoms ejected per fission fragment penetrating the surface of a metallic U or UO_2 emitter are subject to some uncertainties. The principal difficulty is that there is evidence the surface condition of the U emitter is an important factor in determining the basic ejection model.[1] In static capsules, initially sealed at 10^{-4} torr or less, outgassing can radically change the capsule atmosphere during irradiation. Also, in order to measure the U ejected, the emitters must be exposed to air when the collectors are removed from the capsule for analyses. In both cases, changes in the surface of the emitters can occur which may influence the U ejection rate.

To minimize these effects and learn more about the mode of U ejection, an experiment was designed and carried out in which the U emitter was kept in a controlled and measured vacuum during irradiation and determination of the U ejected.

EXPERIMENTAL

To provide a facility for irradiation of a metallic U source and determination of the U ejected from the source, the combination irradiator and fission counter shown in Fig. 1 was built. This chamber, when inserted adjacent to the core of the Water Boiler Reactor at the Los Alamos Scientific Laboratory, furnished a facility in which a U source could be irradiated in a neutron flux of $\sim 5 \times 10^{11}$ at full reactor power of 25 kW and at a pressure of $< 10^{-4}$ torr, with the ejected U atoms being collected on the Pt foil surrounding it. After the

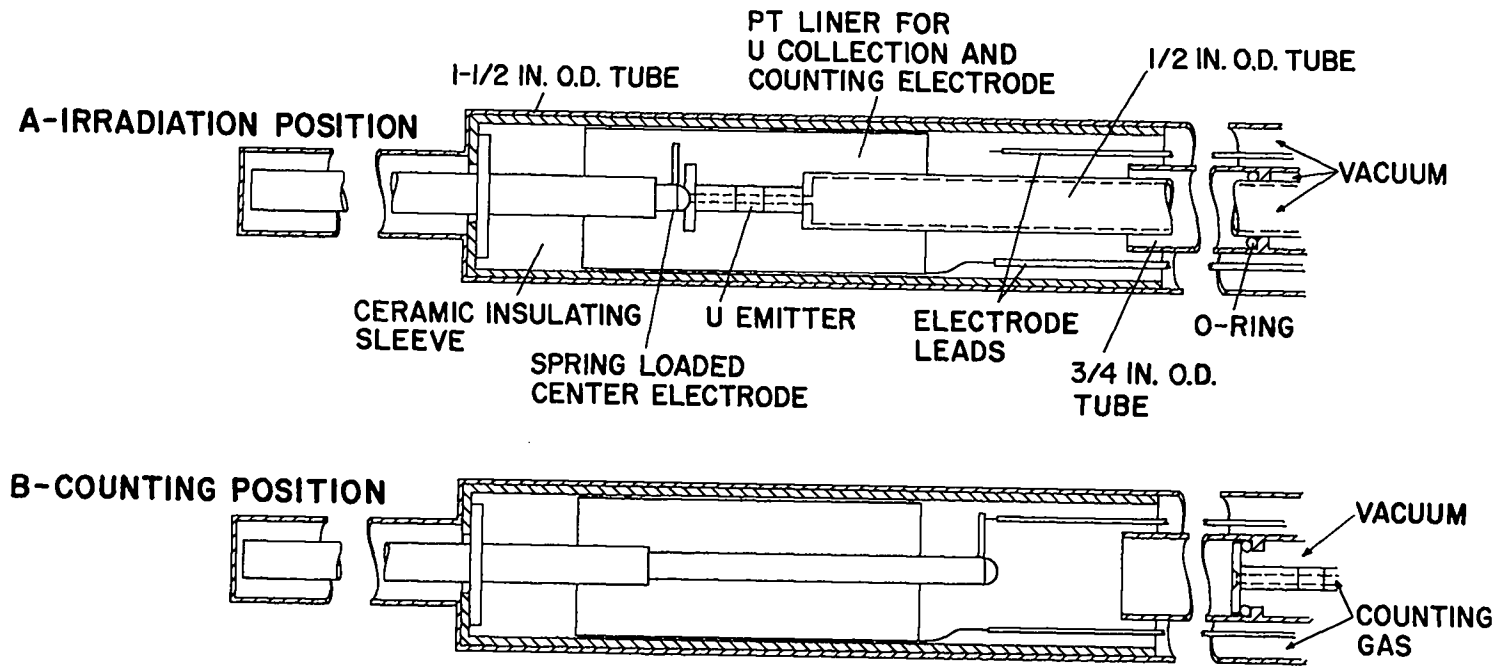

Fig. 1. Combination Irradiator and Fission Counter for Determination of U Ejection from a Fissioning Surface.

desired irradiation period, the source was withdrawn to a low flux position 2 ft away where it was sealed into a 3/4-in. -O. D. tube and kept under vacuum. As the source was withdrawn, a spring-loaded center electrode moved into the position it had occupied, and the irradiator became a chamber for counting the fissions of the U deposited on the Pt foil. The center electrode, as well as the Pt collector foil, were insulated from ground and connected electrically to an external counting circuit. The counting gas mixture, 90% Ar - 10% CH_4, was passed into the chamber through the center 1/2-in. -O. D. tube and removed through the annular space between the 1-1/2-in. -O. D. tube and the 3/4-in. -O. D. tube.

With appropriate valving on an external manifold, the counting chamber could be evacuated after completion of the counting by a liquid N_2 trapped oil diffusion pump. The U emitter could then be pushed back into position and the cycle repeated as often as desired.

The U emitter used in this experiment was a cylinder of 93% enriched oralloy, 0.239-in. O. D. x 0.005-in. wall x 3/16 in. long. The emitter was fitted snugly on a stainless steel pin attached to the 1/2-in. -O. D. tube (Fig. 1), providing a thermally conductive path to a heat sink so that the small amount of fission heat generated would not greatly increase the emitter temperature. Surface preparation of the emitter consisted of pickling in 6N HNO_3 for $\sim$5 min at 90°C followed by washing in de-ionized water and then in ethanol. The surface had a straw-colored appearance, indicating a thin oxide film giving an interference color.

RESULTS

Results of the initial series of irradiations of emitter FFC-2 and countings of U collected are given in Table 1 and plotted in Fig. 2. The neutron

Table 1

Exposure and Counting Data for Initial Irradiation of U Emitter FFC-2*

Point No.	U Collected (fission counts/min) 1st Count	2nd Count	Average	Neutron Exposure (kW-min Reactor Operation)
1	1892	1906	1899	0
2	2058	1848	1953	15
3	3422	3326	3374	165
4	5350	5290	5320	465
5	7164	6846	7005	765
6	8888	8854	8871	1365
7	10758	10596	10677	2465
8	11738	11896	11817	3565
9	13006	13288	13147	5775
10	15904	15570	15737	8685
11	18014	17934	17974	11820
12	20588	20436	20512	15620

*For counting, the Pt collector electrode was charged to -480V, and the reactor operated at a power of 100 W. Gas pressure in the counting chamber was $\sim$10 psig of 90% Ar-10% CH_4.

exposure is given in terms of kW-min of reactor operation, and the amount of U collected on the Pt is expressed as fission counts per minute. Because of the unknown counter efficiency and the difficulty in determining the precise neutron flux at the emitter surface, absolute values of U weight and integrated neutron flux are not used. As a rough guide, 1 kW-min of reactor operation is approximately equivalent to 4.2×10^{11} neutrons/cm^2, and one microgram of U gives

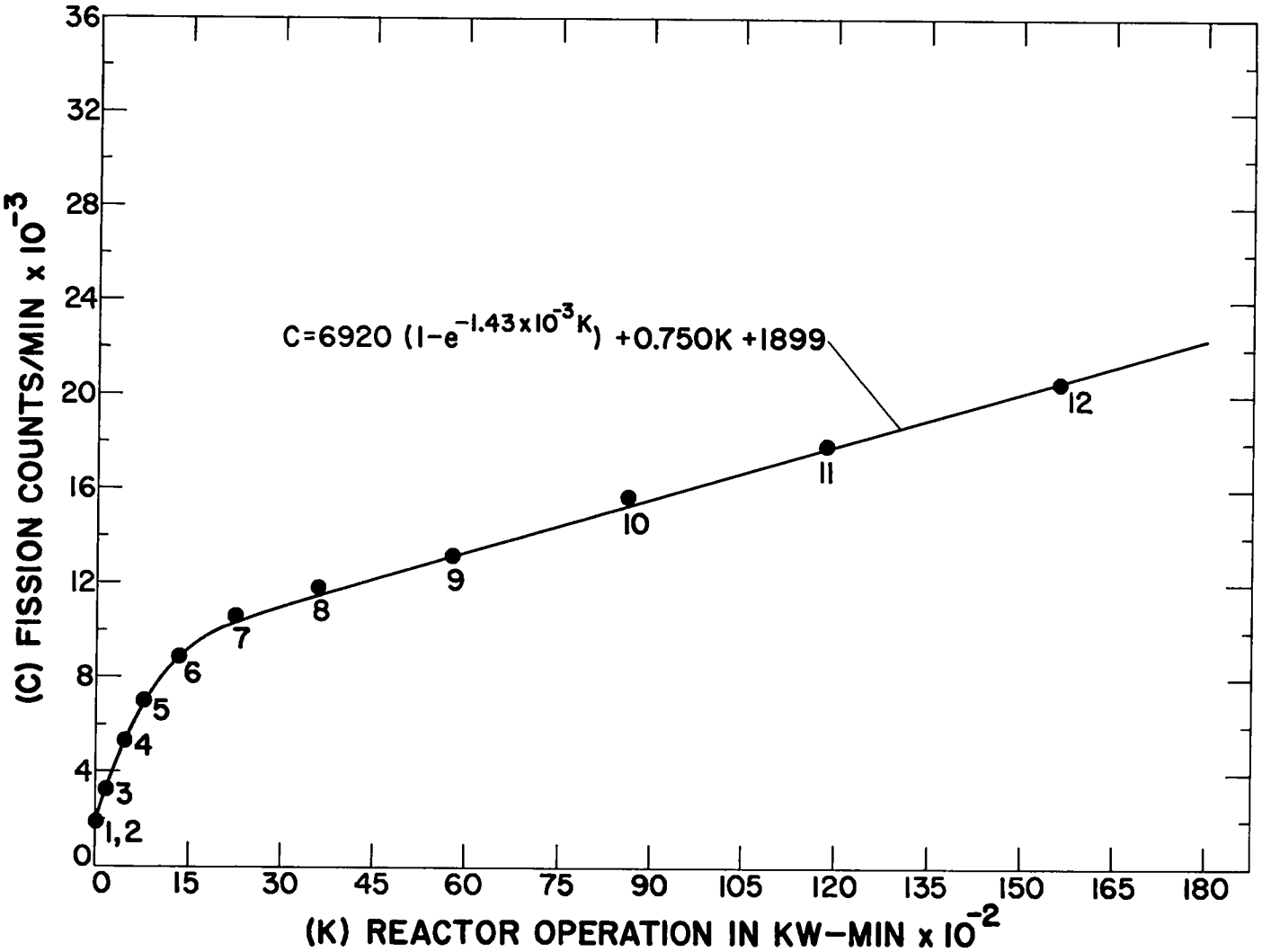

Fig. 2. U Collected in Initial Irradiations of Emitter FFC-2 in Terms of Fission Counts/Min vs Neutron Exposure Expressed in kW-Min of Reactor Operations.

about 105,000 counts per minute at a reactor power of 100 W. These estimates are based on U analyses of the Pt collector foil and fission analyses of the U emitter.

When it became evident at the higher exposures that the increase in U collected was linear with reactor operation, the U emitter was exposed to moist He for ~90 hr at room temperature. Irradiation followed by counting then continued. These data are shown in Table 2. Figure 3 is a repetition of the curve of Fig. 2 on a smaller scale with the additional data from Table 2 also plotted. Figure 4 is an expanded portion of Fig. 3, comparing the linear slope before and after the moist He treatment.

Table 2

Exposure and Counting Data for Irradiations of U Emitter FFC-2 after 90 hr Treatment with Moist He*

Point No.	U Collected (fission counts/min) 1st Count	2nd Count	Average	Neutron Exposure (kW-min Reactor Operation)
13	20808	20534	20671	15620
14	22350	22492	22421	16095
15	24258	24164	24211	16705
16	24592	24754	24673	17290
17	25256	25188	25222	17900
18	41810	41816	41813	18860
19	44034	43746	43890	20210
20	44446	43604	44025	21070
21	31706	31654	31680	22545

*For counting, the Pt collector electrode was charged to -480V, and the reactor operated at a power of 100 W. Gas pressure in the counting chamber was ~10 psig of 90% Ar - 10% CH_4.

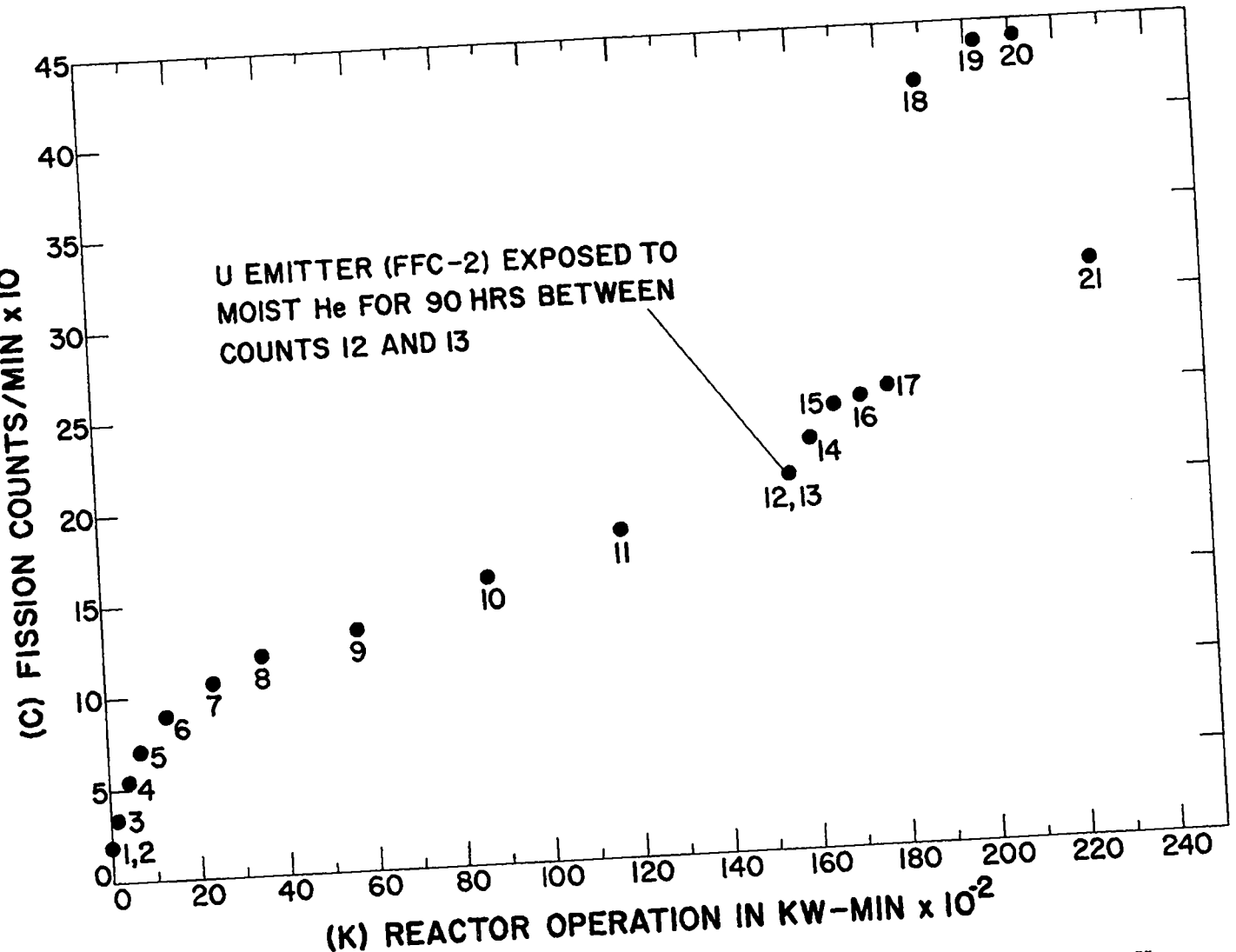

Fig. 3. U Collected from Emitter FFC-2, in Irradiations before and after a Moist He Treatment, in Terms of Fission Counts/Min vs Neutron Exposure Expressed in kW-Min of Reactor Operations.

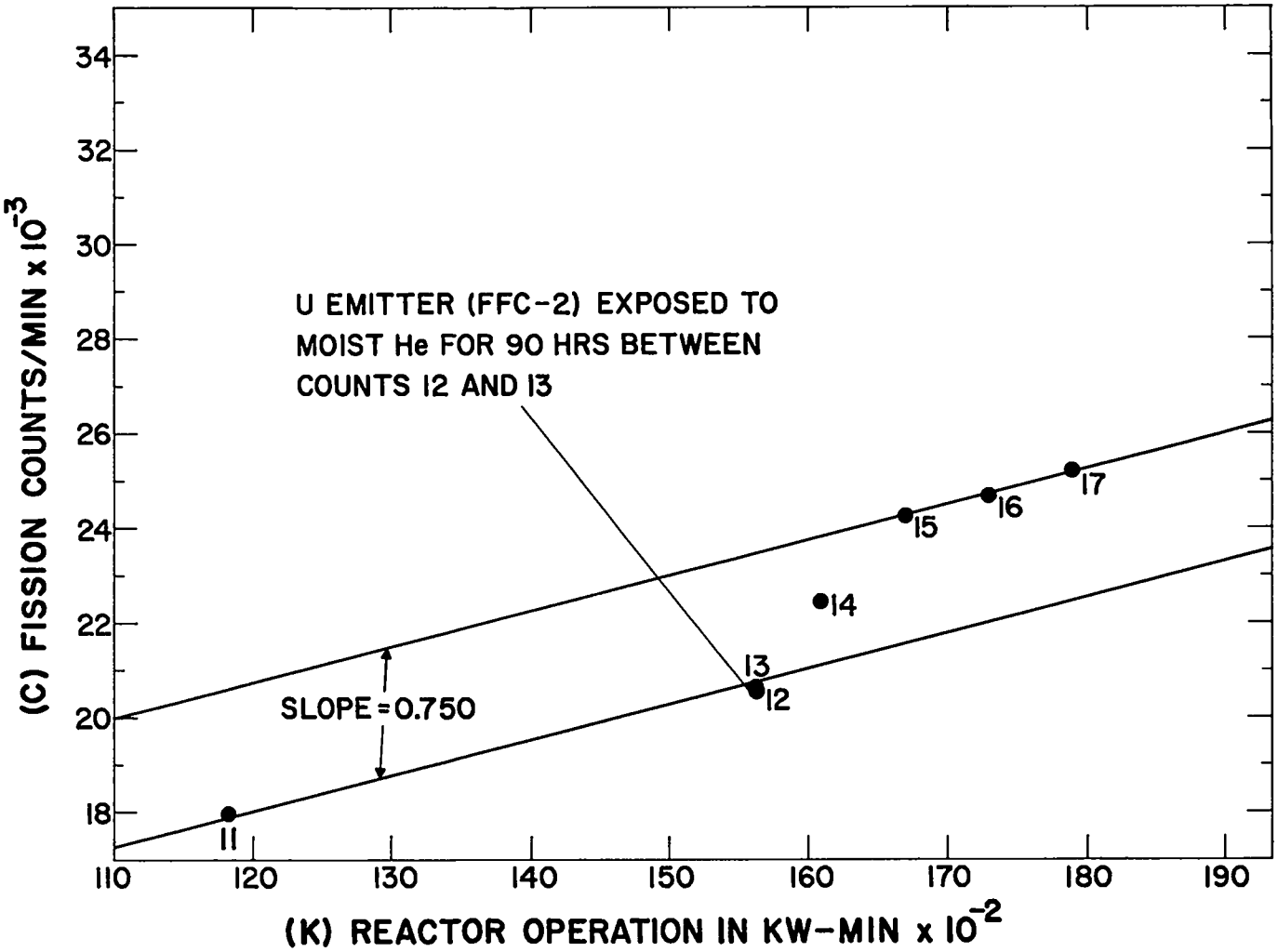

Fig. 4. Data from Fig. 3, immediately before and after Treatment of FFC-2 with Moist He, Showing the Return to a Constant Slope on a Larger Scale.

DISCUSSION

The data gathered in this experiment confirm the results of the capsule irradiations reported in LA-3352-MS. They are also consistent with the hypothesis developed in that Report concerning the modes of U ejection from the emitter surface. Briefly stated, it was suggested that UO_2 crystallites of a critical size on the emitter surface absorbed sufficient fission fragment energy by electronic interaction to be evaporated[2] and that this caused the initially high rate of U ejection. It was further suggested that when crystallites of the critical size range were depleted, an atom current[3] produced by nuclear elastic collisions caused the continuing ejection of U at a much lower rate. Good control of the U emitter environment plus determination of the U ejected in situ gave greater precision and reliability to the results of the present work.

Modifying the equation developed in LA-3352-MS for ejection of U so that mass is expressed as fission counts/min and integrated flux as kW-min, we get

$$C = C_o \left(1 - e^{-\lambda K}\right) + c\,K\,,$$

where C is the count rate of U ejected, C_o is the count rate of U originally present on the emitter surface in UO_2 crystallites of a certain size, λ is a removal constant for UO_2 crystallites of a given size, K is integrated flux in kW-min, and c is a constant.

Using the data of Table 1 to evaluate constants, we find

$$C = 6920 \left(1 - e^{-1.43 \times 10^{-3} K}\right) + 0.750\,K + 1899.$$

This equation represents the solid black line of Fig. 2. The number 1899 is added to adjust for counter background. Using the approximations previously

given for integrated flux as a function of reactor operation and the mass of U collected relative to fission counts/min, calculations show the straight line portion of this curve is equivalent to the order of 6 atoms of U emitted per fission fragment escaping. The initial slope is ~ 12 times that of the straight line portion, or equal to a rate the order of 70 atoms of U emitted per fission fragment escaping. Comparable values reported in LA-3352-MS were respectively 7 and 144 U atoms/fission fragment escaping the surface.

The data in Table 2 and Fig. 3 and 4 show conclusively that oxidation of the U emitter surface by moist He had an effect upon the U ejection rate. Points 13 through 17, taken after the He treatment, show an accelerated ejection rate tending to return to a linear function with the same previous slope. The discontinuity following point 17 is thought to be a result of spallation of the oxide film on the emitter. The fact that the greater share of this increase was subsequently lost at point 21 indicates this material was not firmly attached to the Pt collector foil. It would appear that, in addition to the postulated U removal by evaporation of UO_2 crystallites and an atom current, there exists other possibilities not readily predictable. This would explain the erratic results generally encountered by investigators in this field.

CONCLUSIONS

The results of this experiment show that the surface condition of fissioning U metal has a definite effect on the rate at which U atoms are ejected. Since this surface state was changed by further oxidation in a moist He atmosphere, it is reasonable to believe that the surface condition of interest is some intrinsic characteristic of the oxide film.

While not constituting a proof of U removal by UO_2 crystallite

evaporation in conjunction with an atom current, the data are not inconsistent with the theory and, indeed, lend substance to it.

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