

Radiation Physics Note 47

Some Observations on the Dependence of Surface Contamination
of Uranium on Temperature and Humidity

A. J. Elwyn, B. Arnold, and J. Luoma

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Introduction

It seems likely that a large amount of depleted uranium in the form of metal plates will be brought to Fermilab in connection with the construction of a liquid argon-uranium calorimeter. These plates will be uncoated and, since uranium is very active chemically, will oxidize rapidly on reaction with the environment forming a black powder that poses a potentially serious radioactive contamination problem.

To plan for the on-site storage and use of uranium in a location that minimizes this hazard, we have investigated the dependence of the rate of corrosion on some controllable environmental parameters, temperature and humidity. It should be emphasized that the present observations provide only qualitative information on surface oxidation rates. Quantitative data that aims at the establishment of the basic factors affecting the growth of oxidation layers requires precise determination of the thickness of the scale or of the weight of the sample.¹ Such research while of intrinsic interest requires a more complete laboratory setup and commitment, and in any event transcends the present practical needs.

The Experiment

Small rectangular plates of depleted uranium (2"x3"x0.125" thick), initially cleaned by wire brushing and dipping in a 50% nitric acid - water solution,² were arranged three plates each in three wide-mouth glass jars under different conditions of temperature and humidity as indicated in Table 1, and placed in the vented hood in the Chemistry Lab at Site 21. The 100% relative humidity environment for jars 2 and 3 was provided by enclosing small water-filled beakers within the jars and covering them tightly. These jars were opened periodically to replenish the oxygen. The higher temperature in jar 3 was obtained by use of a thermostatically controlled hot plate.

The corrosiveness of the environments in the three jars was monitored at intervals of 10-14 days by wiping the oxidized surface of the plates and counting the beta radioactivity of the contaminated oxide coating at the shielded proportional counter of the automatic sample changer. At each monitoring period one-half of one-side of one of the plates in each jar was wiped. The experiment was continued until both halves of each side of each plate had been sampled (mid-April to mid-August, 1984).³

Results

The surface wipe results in counts per minute of beta activity⁴ for the plates in each of the jars are shown as a function of time in Fig. 1. Although an effort was made to perform the surface wipes in the same way at each

monitoring interval differences occurred. These differences as well as effects due to non-uniformities on the plate surfaces within the same jar probably account in large part for the observed fluctuations in data.

Even so the general trend of the results for each jar is reasonably clear. The solid line on each plot in Fig. 1 represents a linear fit to the data points. The slopes of these lines, which give the daily increase in surface radioactivity, are shown in Col. 4 of Table 1; the range of values in parentheses reflect the fluctuations in the data. The numbers in Col. 4 normalized to a standard wiping area of 100 cm^2 and converted to activity by use of the known⁵ detection efficiency of the sample changer for depleted uranium are shown in Col. 5. It might be noted that at Fermilab whenever surface contamination levels exceed $0.5 \text{ nCi}/100 \text{ cm}^2$ wipe for general $\beta\gamma$ emitters, or $0.05 \text{ nCi}/100 \text{ cm}^2$ wipe for α -emitters a contamination zone must be established and protective measures instituted.⁵

The observations in the present study suggest that at temperatures near 70° an increase in the relative humidity by about a factor of two increases the daily rate of surface contamination by factors from 4 to 20. More dramatically, a rise of 40% in temperature shows an increase in contamination rates by factors of 100-200 at least under conditions of high humidity.⁶

Because of the rather qualitative nature of this experiment the present results reflect the general trend of the rate of oxidation of uranium in air, not quantitative time and temperature dependences. More detailed and accurate measurements¹ (mostly done at very high temperatures) show that below 150°C the

oxidation of uranium in dry air or oxygen actually follows a parabolic rather than linear dependence on time, while in moist air or water vapor the oxidation may proceed linearly with time although little data exists. For uranium as for most metals a very rapid (probably exponential) dependence on temperature is apparently observed.

In conclusion, the observations summarized above suggest that depleted uranium plates, for assembly into a calorimeter, should be stored and used in a location for which both the temperature and humidity can be controlled. In particular, the rate of surface contamination can be minimized at temperatures of 70-75°F, and values of the relative humidity of up to 60%.

References

1. O. Kubaschewski and B. E. Hopkins, Oxidation of Metals and Alloys (Academic Press, Inc., New York, N.Y., 2nd Edition, 1962); M. G. Fontana and N. D. Greene, Corrosion Engineering (McGraw-Hill Book Co., New York, N.Y., 1978).
2. This was done at the Uranium Shop facilities at Argonne National Laboratory.
3. Unfortunately one of the plates in Jar 3 started to crumble after falling and remaining in water for an extended period. These data are not included in the present results.
4. The wipes counted for α -radioactivity had cpm values of about a factor of 10 lower.
5. Radiation Guide (Fermi National Accelerator Laboratory, Batavia, IL., 4th Edition, April 1983).
6. In retrospect it would have been of interest to include a fourth jar containing uranium plates maintained at $\sim 100^{\circ}\text{F}$, but at ambient relative humidity.

Table 1: Experimental Conditions and Results

Jar Number	Temperature (°F)	Relative Humidity (%)	Slope of straight line on Fig. 1 (cpm/day)	Activity per 100 cm ² wipe (nCi/100 cm ² wipe-day)
1	69-73	35-60	0.03 (0-.3)	0.0005 (0-.005)
2	69-73	100	0.63 (0.5-1.4)	0.01 (.008-.023)
3	90-104	100	114 (74-128)	1.9 (1.2-2.1)

Fig 1: Surface $\beta\gamma$ -Wipe Results

