ANRCP -1997-4

Amarillo National Resource Center for Plutonium

A Higher Education Consortium of The Texas A&M University System,
Texas Tech University, and The University of Texas System

RECEIVED
DEC 09 1997
OSTI

TECHNICAL SUMMARY: **FEASIBILITY STUDY OF CONDUCTIVITY** **MONITORING FOR LEAK DETECTION IN** **DOUBLE-WALLED PLUTONIUM CONTAINERS**

November 1997

This report was prepared
with the support of the
U.S. Department of Energy
(DOE), Cooperative
Agreement No.
DE-FC04-95AL85832.

However, any opinions,
findings, conclusions, or
recommendations
expressed herein are
those of the author(s) and
do not necessarily reflect
the views of DOE. This
work was conducted
through the Amarillo
National Resource Center
for Plutonium.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

600 South Tyler • Suite 800 • Amarillo, TX 79101
(806) 376-5533 • Fax: (806) 376-5561
<http://www.pu.org>

df
MASTER

DISCLAIMER

**Portions of this document may be illegible
in electronic image products. Images are
produced from the best available original
document.**

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

ANRCP-1997-4

Technical Summary: Feasibility Study of Conductivity Monitoring for Leak Detection in Double-Walled Plutonium Containers

J.-X. Lu

Department of Nuclear Engineering
Texas A&M University
College Station, Texas 77843

W. H. Marlow

Department of Nuclear Engineering
Texas A&M University
College Station, Texas 77843

November 1997

Published by the
Amarillo National Resource Center for Plutonium

Technical Summary:
Feasibility Study of Conductivity Monitoring
for Leak Detection in Double-Walled Plutonium Containers

J.-X. Lu and W. H. Marlow

Department of Nuclear Engineering

Texas A&M University

College Station, TX 77843

I. INTRODUCTION

Currently, the storage container for the pit from a dismantled warhead is a sealed outer container, or *drum*, within which the pit is suspended. Since the pit itself is a sealed, stainless steel container for the plutonium, the inner plus the outer containers constitute the "double-walled" configuration for plutonium storage. If either inner or outer wall of the container fails, the fill-gas between the pit and drum walls will contain species that will modify the physical properties of that gas. The work summarized here reports the initial feasibility study for an innovative approach for monitoring for leakages both for radioactive materials from the pit and for the intrusion of outside air into the drum by monitoring the electrical conductivity of the fill-gas.

For the gas present in a drum containing a pit, alphas from decays of plutonium are stopped by the primary container wall of the pit itself unless pit leakage occurs. If plutonium leaks from the pit and enters the fill-gas (either noble gas or air) of the outer container, each of the alpha particles due to the decay of plutonium will create about 10^5 electron-ion pairs along its track. If the fill gas is a noble gas, these electrons will diffuse in the gas as free

electrons. On the other hand, if the fill-gas is air, the electrons will quickly form negative ions by attachment to neutral molecules in the gas such as oxygen, then these newly-formed negative ions will diffuse in the gas. In order to avoid recombination between positive and negative species, a weak electric field is applied across the fill-gas. Then, we expect a current pulse due to the drift and diffusion of the above positive and negative ions. We also expect that the numerous gamma decays within the pit give rise to a constant ionization of the fill-gas, creating the normal background conduction current of the container. The striking difference between the constant low background current and the current pulse due to an alpha particle provides a unique means, similar to an ionization chamber, to determine if there is a leak through the inner wall of the container. Conversely outer container leakage permits atmospheric gases to mix with the fill-gas resulting in changes in the steady-state conductivity of the gas.

Specifically, we have two cases depending on the fill-gas. If the container is filled with a noble gas, the electrons produced by the gas ionization of a single alpha particle due to plutonium decay are rapidly transported to the anode for the imposed electric field without forming negative molecular ions. This electronic current is sharply peaked and is followed by much slower and two orders of magnitude weaker ionic peak. Detection of either of these two current peaks against the low level constant background current due to gamma ionization indicates uniquely the presence of plutonium external to the pit. If the outer wall has a leak, ambient air and possibly aerosol particles will enter the container. Then we will not detect the electronic current peak but only an ionic peak since electrons will attach to molecules to form molecular ions. Further, the previously constant background current will also measurably decrease due to ambient gases. If the container is filled with relatively clean dry air, the previous discussion indicates that we can detect only ionic current pulse in the case of inner wall leakage.

We have performed a computational investigation that shows the feasibility of the detection principles discussed above. In particular, we have shown that both the electronic and ionic peak currents are greater than $10^{-12} A$, i.e., 1 pA (picoampere), which is a feasible

lower limit for detection of current. If the fill-gas is a noble gas, the electronic current peak is on the order of a nanoampere and is in general two orders of magnitude larger than the ionic current peak. If the fill-gas is air, the ionic current peak resembles that of noble gas. In terms of ease of detection, the choice of fill-gas is certainly noble gas.

Before we present the detail of the results, we address the question of how much material must be present in the outer container so that the resulting ionization current is detectable on a practical basis. This study shows that the current due to a single alpha ionization is detectable. So the above question is now equivalent to asking what is the practical time scale for which there is one alpha decay to occur, i.e., the radioactivity. We have made simple calculations of the mass of Pu-239 for one α -decay over each of the time scales: 10 hours, 1 hour, 1 minute and 1 second. The plutonium isotope Pu-239 is chosen in our estimations since this isotope has the largest mass-percent in various grades of plutonium (e.g., weapons grade, fuel grade and power grade). The results are summarized in Table I. In our estimations, we made the assumption that the alpha decays are all from the parent Pu-239.

II. THEORY

An alpha particle from the decay of a plutonium isotope has a typical energy around 5 MeV. In air or noble gas, this alpha particle will create about 10^5 electron-ion pairs along its track. The track range in air or noble gas is about 2 cm. Unlike in noble gas, the electrons in air quickly attach to molecules to form negative ions. With a weak applied electric field, electrons or ions drift towards the proper electrodes, thereby giving rise to electric current pulses. Such an applied electric field also suppresses the recombination between positive and negative charged ions in addition to the diffusion effect. If there are no outer wall leaks, we expect either electronic current pulse or ionic current pulse or both, depending on the fill-gas as discussed above.

Completely solving the electron or ion transport in the container with an imposed electric

field is extremely difficult. However, to show the feasibility of the detection principle, a theoretical characterization of the electron or ion transport is sufficient. For this reason, we model the transport processes of electrons and ions due to a single alpha track by considering the transport of the electrons or ions in a homogeneous space with a constant applied electric field. In other words, we neglect the influences of the container walls and internal structure on the transport of the electrons or ions and on the applied electric field. This model description should be fairly accurate for those alpha tracks not very close to the containers' inner and outer walls and to the electrodes. We show that the current pulses due to a single alpha particle ionization is indeed measurable. We also investigate how the current profiles depend on the the applied electric field and the distance between the alpha track and the electrode. This study will also provide the guidance for the design of the prototype system.

The most general equation governing the transport of electrons or ions is

$$\partial_t n(\vec{r}, t) + \nabla \cdot \vec{j} = -\alpha n(\vec{r}, t) + S(\vec{r}, t), \quad (2.1)$$

where $\partial_t \equiv \partial/\partial t$, $n(\vec{r}, t)$ is the electron or ion density, the constant α characterizes the loss of electrons or ions by chemical reactions which is not important in our case, and $S(\vec{r}, t)$ is the source term. In the above, $\vec{j}$ denotes the current density divided by the charge of the electron or ion, which is given by

$$\vec{j} = \vec{v}_d n(\vec{r}, t) - \mathcal{D} \cdot \nabla n(\vec{r}, t), \quad (2.2)$$

where $\vec{v}_d$ is the drift velocity of electron or ion under an applied constant field $\vec{E}$ and is given by $\vec{v}_d = K \vec{E}$ with K the mobility. The diffusion coefficient tensor $\mathcal{D}$ is in general given by a 3×3 matrix as

$$\mathcal{D} = \begin{pmatrix} D_T & 0 & 0 \\ 0 & D_T & 0 \\ 0 & 0 & D_L \end{pmatrix}, \quad (2.3)$$

where D_T denotes the transverse diffusion coefficient that describes the rate of diffusion in the direction perpendicular to the applied electric field $\vec{E}$ and D_L is the longitudinal diffusion

coefficient. Using Eq. (2.2) and choosing $\vec{E}$ along positive z-axis, we can rewrite Eq. (2.1) as

$$\partial_t n(\vec{r}, t) = D_T(\partial_x^2 + \partial_y^2)n(\vec{r}, t) + D_L\partial_z^2 n(\vec{r}, t) - v_d\partial_z n(\vec{r}, t) - \alpha n(\vec{r}, t) + S(\vec{r}, t). \quad (2.4)$$

If we make the following transformation

$$n(\vec{r}, t) = e^{-\alpha t + \frac{v_d}{2D_L}(z - v_d t/2)} \tilde{n}(\vec{r}, t), \quad (2.5)$$

the above equation can be simplified as

$$\partial_t \tilde{n}(\vec{r}, t) = D_T(\partial_x^2 + \partial_y^2)\tilde{n}(\vec{r}, t) + D_L\partial_z^2 \tilde{n}(\vec{r}, t) + \tilde{S}(\vec{r}, t), \quad (2.6)$$

where $\tilde{S}(\vec{r}, t) = e^{\alpha t - \frac{v_d}{2D_L}(z - v_d t/2)} S(\vec{r}, t)$.

Eq. (2.6) can be solved using Green-function methods and combining with Eq. (2.5), we have the general solution

$$n(\vec{r}, t) = \frac{1}{(4\pi)^{3/2} D_T D_L^{1/2}} \int d^3 \vec{r}' \int_{-\infty}^t dt' \frac{S(\vec{r}', t') e^{-\alpha(t-t')}}{(t-t')^{3/2}} e^{-\frac{(x-x')^2 + (y-y')^2}{4D_T(t-t')}} e^{-\frac{[(z-z') - v_d(t-t')/2]^2}{4D_L(t-t')}}}. \quad (2.7)$$

Given any source $S(\vec{r}, t)$, the electron or ion density at any space point and at any time can be determined either analytically or numerically from the above equation. For the present case, i.e., the ionization in a fill-gas due to a single alpha particle from the decay of plutonium, the short range nature of the alpha particle permits us to represent the source $S(\vec{r}, t)$ in terms of delta functions as

$$S(\vec{r}, t) = N_0 \delta(\vec{r} - \vec{r}_0) \delta(t - t_0), \quad (2.8)$$

where $\vec{r}_0$ is the center position vector of the alpha track, t_0 is the time when the single alpha particle is created and N_0 is the number of electrons or ions created by the alpha track and is given by $N_0 = E/W$ with E the energy of the alpha particle and W the W-value of the fill-gas. N_0 is in general on the order of 10^5 . Using Eq. (2.8), the time-dependent electron or ion density distribution can be obtained from Eq. (2.7) as

$$n(\vec{r}, t) = \frac{N_0}{(4\pi)^{3/2} D_T D_L^{1/2}} \frac{e^{-\alpha(t-t_0)}}{(t-t_0)^{3/2}} e^{-\frac{(x-x_0)^2 + (y-y_0)^2}{4D_T(t-t_0)}} e^{-\frac{[(z-z_0) - v_d(t-t_0)/2]^2}{4D_L(t-t_0)}}}. \quad (2.9)$$

Using Eq. (2.2), we have

$$\vec{j}(\vec{r}, t) = \frac{n(\vec{r}, t)}{2(t - t_0)} [\vec{r} - \vec{r}_0 + v_d(t - t_0)\vec{e}_z], \quad (2.10)$$

where $n(\vec{r}, t)$ is given by Eq. (2.9) and $\vec{e}_z$ is the unit vector of z-axis. Then the current through any given surface area is

$$I(r, t) = q \int \vec{j} \cdot d\vec{S}, \quad (2.11)$$

where r denotes the shortest distance between the center of the alpha track and the surface, and q denotes the charge carried by an electron or ion.

Evaluating the current through a general cross-section and for a general applied electric field can only be done numerically. For the present study, we have chosen the constant electric field along the z-axis. So the drift current due to electrons or ions is either along negative z-axis or positive z-axis. Hence, we should choose the cross-section for the integration in Eq. (2.11) perpendicular to the z-axis, i.e., in the x-y plane. For the purpose of the present study, we can further simplify the calculations. The reason for applying an external electric field is two-fold. One is to suppress the recombination losses, especially the so-called *columnar* recombination loss. The other is to give rise to the drift current. If the applied field is not too strong in the sense that the electric energy gain during a mean free path of electron or ion in the fill-gas is less than the thermal energy kT , we can then take $D_L = D_T = D$. Recall that the relation $\vec{v}_d = K\vec{E}$ requires also the electric field be weak. The actual criterion for a weak field is

$$\frac{E}{p_{293}} (\text{V/cm torr}) \ll 0.26 \left(\frac{m}{m + M} \right)^{1/2} Q_D (\text{\AA}^2), \quad (2.12)$$

where E/p_{293} has unit volts per centimeter per torr with p_{293} the pressure at $T = 293$ Kelvin (i.e., at 20°C), m is the electronic or ionic mass and M is the fill-gas molecular mass. Q_D is the electron or ion-molecule collision cross-section in $\text{\AA}^2$ and it is typically on the order of $10 \text{ \AA}^2$. But in general the ion-molecule cross-section is larger than the electron-molecule cross-section. Taking $Q_D = 10$, we have for electron ($M \approx 1000m$)

$$\frac{E}{p_{293}} \ll 0.1 \text{ V/cm torr}, \quad (2.13)$$

while for ion ($m \approx M$)

$$\frac{E}{p_{293}} \ll 2.8 \text{ V/cm torr}. \quad (2.14)$$

The above clearly shows what may be a weak electric field for an ion may be a very strong field for an electron in the same fill-gas.

With all the above and if we take the cross-section of electrodes as a disk with radius ρ_0 , the electronic or ionic current through the cross-section in x-y plane and located at $z = z_0$ can be obtained from Eqs. (2.9), (2.10) and (2.11) as

$$I(z_0, t) = \frac{qN_0}{2t(4\pi Dt)^{1/2}} (z_0 + v_d t) e^{-\frac{(z_0 - v_d t)^2}{4Dt}} (1 - e^{-\rho_0^2/4Dt}), \quad (2.15)$$

where we have set both $\alpha = 0$ and $t_0 = 0$, and $q = -e$ for electrons or negative ions and $q = e$ for positive ions with $e = 1.60 \times 10^{-19} \text{C}$. The z_0 is now the shortest distance between the center of the alpha track and the cross-section chosen.

III. RESULTS

We have applied Eq. (2.15) to study the electronic and ionic current pulses due to a single alpha track in the fill-gas of helium. Specifically, we have studied the current profiles due to electrons for each of $z_0 = 10, 15$ and 20 cm with $E/p_{293} = 0.01, 0.05$ and 0.1 V/cm torr, respectively. For a given $E/p_{293} = 0.05$ V/cm torr, we have also studied the profiles for $z_0 = 10, 15$ and 20 cm. Similar current profiles due to He^+ have also been studied, but now with $E/p_{293} = 0.05, 0.1, 0.15$ and 0.2 V/cm torr. In our study, $p_{293} = 760$ torr is always chosen. The data from Table II have been used. For He^+ in helium gas, the mobility is $10.4 \text{ cm}^2/\text{V sec}$ at $T = 293$ Kelvin, which gives the diffusion coefficient $D = 0.269 \text{ cm}^2/\text{sec}$ from $D/K = kT/e$. The drift velocity $v_d = KE$ is $395, 790, 1.19 \times 10^3$ and 1.58×10^3 cm/sec for $E/p_{293} = 0.05, 0.1, 0.15$ and 0.2 V/cm torr, respectively. The typical energy of alpha particles from the decay of plutonium isotopes is around 5 MeV , we choose it as $E = 5$

MeV. The W-value (the mean energy expended in a gas per ion pair formed) of helium gas is $W = 42.7$ eV. So the number of ion pairs is $N_0 = E/W = 1.2 \times 10^5$.

From the profiles (see attached figures), we have the following: In the case of electronic current, the height of the current peak I_0 is in the range $1 \text{ nA} < I_0 < 10 \text{ nA}$ and the time scale for the peak is from 10^{-4} s to 10^{-5} s , which is given approximately by z_0/v_d , for all the cases studied, i.e., $z_0 = 10, 15$ and 20 cm and $E/p_{293} = 0.01, 0.05$ and 0.1 V/cm torr . In general, the larger the peak current height, the smaller the peak time scale. This corresponds to the smaller z_0 and the larger the E/p_{293} . Similar features also occur for the ionic current profiles studied except that the current peak height I_0 is now in the range $10 \text{ pA} < I_0 < 200 \text{ pA}$ and the peak time scale is around 10^{-2} s .

This study clearly shows that both the electronic and ionic current peaks due to a single alpha particle from the decay of plutonium isotopes are measurable. In terms of ease of detection, the electronic current pulse is the choice, which implies the choice of rare gas as the fill-gas. We have also studied both the electronic and ionic current pulses in Ar as well as the ionic current pulses in air. We found that the electronic current profile in Ar is similar to that in He while all the ionic current profiles are similar no matter whether the fill-gas is rare gas or air. Details of these calculations will be reported in the near future.

TABLES

TABLE I. Minimum Pu-239 Needed For Single α -decay During Each Indicated Time Interval

Time scale	Activity (Bq)	Activity ($\times 10^{-11}$ Ci)	Amount of Pu-239 ($\times 10^{-10}$ gram)
10 hours	2.8×10^{-5}	7.6×10^{-5}	1.2×10^{-4}
1 hour	2.8×10^{-4}	7.6×10^{-4}	1.2×10^{-3}
1 minute	1.7×10^{-2}	4.6×10^{-2}	7.2×10^{-2}
1 second	1.0	2.7	4.3

TABLE II. Data Used In Calculating The Electronic Current Pulse In Fill-gas He

E/p_{293} (V/cm torr)	v_d ($\times 10^5$ cm /sec)	D/K (V)	D (cm^2/sec)
0.010	0.637	0.034	285
0.050	1.840	0.087	421
0.10	2.67	0.154	541

FIGURES

FIG. 1. Comparison of electronic current pulses in the fill-gas of He at $E/p_{293} = 0.05$ V/cm torr for $z_0 = 10, 15$ and 20 cm.

FIG. 2. Comparison of electronic current pulses in the fill-gas of He at $z_0 = 10$ cm for $E/p_{293} = 0.01, 0.05$ and 0.1 V / cm torr.

FIG. 3. Comparison of electronic current pulses in the fill-gas of He at $z_0 = 15$ cm for $E/p_{293} = 0.01, 0.05$ and 0.1 V / cm torr.

FIG. 4. Comparison of electronic current pulses in the fill-gas of He at $z_0 = 20$ cm for $E/p_{293} = 0.01, 0.05$ and 0.1 V / cm torr.

FIG. 5. Comparison of current pulses due to He^+ in the fill-gas of He at $E/p_{293} = 0.1$ V/cm torr for $z_0 = 10, 15$ and 20 cm.

FIG. 6. Comparison of current pulses due to He^+ in the fill-gas of He at $z_0 = 10$ cm for $E/p_{293} = 0.05, 0.1, 0.15$ and 0.2 V/cm torr.

FIG. 7. Comparison of current pulses due to He^+ in the fill-gas of He at $z_0 = 15$ cm for $E/p_{293} = 0.05, 0.1, 0.15$ and 0.2 V/cm torr.

FIG. 8. Comparison of current pulses due to He^+ in the fill-gas of He at $z_0 = 20$ cm for $E/p_{293} = 0.05, 0.1, 0.15$ and 0.2 V/cm torr.

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7

Figure 8