Project Sapphire Uranium-Beryllium Dose Rate Analysis

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During a six-week period in the fall of 1994 a team of 31 U. S. government and Y-12 personnel packaged and removed several thousand kilograms of material containing highly enriched uranium from the (former Soviet Union) Republic of Kazakhstan for interim storage at the Y-12 Plant in Oak Ridge, Tennessee. This classified mission, known as PROJECT SAPPHIRE, had been initiated at the request of the Kazakhstan government in order to rid itself of possible security problems. Planning for the mission included assurance of the health and safety of the team members, as well as compliance with all local, I.A.E.A., and U. S. government regulations regarding the handling, packaging, transportation, and storage of radioactive and fissile material. The mission classification restrictions were relaxed following the return of the team and material to the United States.

The material to be removed, in the form of small billets and rods of uranium metal and uranium-beryllium alloy and oxide powder, was sealed by team members on site into two-liter steel cans. Two or three cans each were loaded into more than 400 I.A.E.A. certified fissile material shipping container, and each container was packed into a large steel drum for transport by U. S. Air Force cargo planes to the United States.
RADIATION SOURCES

After criticality safety, a primary item of concern during the mission planning was the amount of radiation dose the team members might receive. Subcritical amounts of uranium of any enrichment give off little penetrating radiation from radioactive decay. But when uranium is mixed with elements such as beryllium, having a large alpha-neutron cross section, a neutron source much larger than that from spontaneous fission can result due to alpha decay in the uranium. This source is increased somewhat due to subcritical multiplication in the highly enriched uranium and the large (n,2n) cross section of the beryllium, leading to a measurable neutron dose rate.

An analysis of material samples provided by the local installation prior to the mission confirmed that all the uranium was enriched to about 89% by weight 235U, 10% 238U and 1% 234U. There were also trace amounts of 232U, 236U, and isotopes of other elements. The total uranium weight in the U-Be alloy varied from less than 10% to more than 30%.

The 234U isotope is increased from its natural concentration during the 235U enrichment process, and it is an active alpha emitter leading to the neutron source in the uranium-beryllium alloy. The two uranium trace isotopes are sometimes introduced during enrichment processes due to contamination from reactor returns. The decay of the 232U isotope, with a 69 year half-life, leads to the emission of a 2.6 MeV gamma ray. A significant gamma ray dose rate can be created with concentrations as low as a few parts per billion (ppb) 232U in the uranium.
DOSE RATE ANALYSIS

Following procedures established at the Y-12 Plant to ensure that all packages containing radioactive material entering or leaving the plant are in compliance with regulations regarding dose rate limits, conservative dose rate calculational analyses were performed prior to the mission. All sources were conservatively increased, and all shielding properties of the package and contents were minimized. The uranium weight in the alloy was taken as 40% and the $^{232}$U concentration was taken as 6 ppb. Dose rates were calculated on the surface of the two-liter storage cans and on the surface and at one meter from the outer shipping drum.

It was assumed in the calculations that the uranium and beryllium were homogeneously mixed. The ORIGEN-S isotope generation, decay, and depletion code was used to generate the neutron and gamma sources. The MCNP and MORSE Monte Carlo radiation transport codes were used to calculate the dose rates resulting from the calculated sources.

Dose rate calculations were also made using nominal source and package data in order to better estimate actual dose rates that might be encountered by the team members. The uranium weight in the alloy was taken as 17%, and the $^{232}$U concentration was reduced to 1 ppb. The maximum and nominal calculated dose rates are compared with those measured on site by Y-12 health physics team members. All calculated and measured dose rates one meter from the drum outer surface were less than 10 μSv/h (1 mR/h). The statistical uncertainties of all calculated values is on the order of 5% or less.
DOSE RATE COMPARISONS

Dose rates in μSv/h
(10 μSv/h = 1 millirem/h)

<table>
<thead>
<tr>
<th></th>
<th>Can surface</th>
<th>Drum surface</th>
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</thead>
<tbody>
<tr>
<td></td>
<td>neutron</td>
<td>gamma ray</td>
</tr>
<tr>
<td>Calculated</td>
<td>511</td>
<td>359</td>
</tr>
<tr>
<td>maximum</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Calculated</td>
<td>74</td>
<td>45</td>
</tr>
<tr>
<td>nominal</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Measured</td>
<td>70</td>
<td>40</td>
</tr>
</tbody>
</table>
NON-DESTRUCTIVE ASSAY

Dose rate calculational analyses were also used to help establish some of the non-destructive assay measurements made on each can of uranium-beryllium alloy before it was sealed and loaded into the shipping container. These assays were part of the criticality safety procedures used to ensure that the mass of the uranium as determined from information supplied by the local installation could be verified. (Other tests were uses to confirm the uranium isotopic concentrations). For the range of uranium weight % in the U-Be alloy (up to 40%), it was shown by calculational analysis that the neutron dose rate increased approximately linearly with the uranium atom fraction in the alloy. It was also found that the density of the alloy followed the same linear curve versus the uranium atom fraction. With zero dose rate at zero atom fraction of uranium, the density of the alloy goes to that of pure beryllium. Some of the alloy rods were measured and weighted on site, and the resulting density and neutron measurements were able to confirm the stated uranium masses to within + or -10%. Much of the uncertainty was due to a similar uncertainty in the measured $^{234}$U weight % used in the neutron source calculation. The neutron response function in the energy ranges of the alpha-neutron production (1-5 MeV) is fairly flat, leading to the predicted dose rate results as measured by a neutron counter on site. Other analyses were used to verify a set of "standard can" measurements made on site, each can with a different and known (measured) mass of uranium in the U-Be alloy. The neutron data for these "standards" served as calibration checks throughout the loading and assay procedures.
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