

DISSOLUTION OF URANIUM OXIDE MATERIALS IN SIMULATED LUNG FLUID

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Depleted uranium (DU) oxide aerosols prepared in the laboratory and collected in the field were tested to characterize their dissolution in simulated lung fluid and to determine how dissolution is affected by aerosol preparation. DU, a by-product of the uranium fuel cycle, has been selected by the US military for use in several types of munitions. During development, manufacture, testing, and use of these munitions, opportunities exist for inhalation exposure to various (usually oxide) aerosol forms of DU. The hazard potential associated with such exposures is closely related to the chemical form, the size of the DU aerosol material, and its dissolution properties. Five DU sample materials produced by exposing uranium alloy penetrators to certain controlled oxidation atmospheres¹ were studied (oxidation temperatures ranged from 500 to 900°C). In addition, two DU sample materials collected in the field were provided by the US Air Force. All sample materials were generated as aerosols using a Wright dust feed. The "respirable fraction" of these aerosols, according to the British Medical Research Council criteria, was separated and collected using a size-selective horizontal elutriator.

Bulk and respirable fraction samples analyzed by x-ray diffraction indicated the uranium crystalline material was α - UO_2 and α - U_3O_8 . Some amorphous material was detected in the respirable fraction of one of the field sample materials. This analysis also showed a significant difference between the composition of bulk and respirable fraction sample materials. The UO_2 content was greater in the bulk samples.

Respirable fraction samples of each study material were subjected to *in vitro* dissolution analysis. A gross-gamma technique^{2,3} was used to determine the sample mass content of uranium before dissolution. Individual samples were then sandwiched between 0.1- μm -pore-size Nucleopore filters. The sandwich was placed in a one-sided radial flow dissolution chamber. A simulant of lung serum at a temperature of $37^\circ\text{C} \pm 0.5^\circ\text{C}$ and a pH of 7.4 ± 0.1 was pumped past the filter sandwich. Eluate from each system was collected and analyzed for uranium by a delayed-neutron activation technique.

Analysis of the data from the dissolution study indicates that multiple-phase (apparently, three different phases) dissolution behavior occurred in all samples. Long-term dissolution parameters were independently calculated in two ways, one using a weighted nonlinear least-squares analysis³ and the other using a simple averaging technique.^{2,3} Additional details for the calculational technique may be found in the referenced work.^{2,3} Both calculation methods resulted in long-term dissolution half-times (third phase) that ranged from 170 to 490 days. The fraction of material associated with the second dissolution phase varied from 4 to 25% with the dissolution half-times in this phase being <2 days. The initial dissolution rate is extremely rapid (in contrast to the rate during the second phase) and could not be accurately measured.

Particular trends regarding the physiochemical character of uranium oxides described by other investigators were supported by the data generated in this study, which suggest that under some conditions a rapidly dissolving U_3O_8 fraction may be formed concurrent with the production of UO_2 . This fraction may play an important role in determining hazard potential associated with inhalation exposure to certain uranium aerosols.

References

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ADDITIONAL SHORT-TERM PLUTONIUM URINARY EXCRETION DATA FROM THE 1945-1947 PLUTONIUM INJECTION STUDIES

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A recent publication by Rundo¹ on the long-term urinary excretion of plutonium, 10 000 days after intravenous injection of known amounts, has shown that the amount of plutonium excreted per day is significantly higher than predicted by the Langham power function model.² The Langham equation for daily urinary (Y_u) excretion rates, in per cent/day of the injected dose at time t (days) after the intake, follows:

$$Y_u = 0.2t^{-0.74} \quad (1)$$

Complete details on the Langham experiment are given in the Langham report and in a follow-up publication by Durbin.³

A review of the original injection experimental records was made because the published 10 000-day excretion data and observations made at Los Alamos⁴ and at the Oak Ridge National Laboratory⁵ have shown that at long times after occupational exposure, the urinary plutonium excretion deviates from the Langham power function model. Each of the Los Alamos National Laboratory notebooks used to record the analytical data was taken from storage and was studied for details that could influence the published findings. The most interesting discovery from this review was that there were additional urine excretion data for case HP-3. This case was one of the two cases from which Rundo obtained the 10 000-day excretion rate. The reason the data were not used in the original Langham publication is unknown, but remarks included in the notebooks suggest that there were some questions about the analytical methodology and an uncertainty with regard to the collection order. These two remarks may have influenced the exclusion of the data from the Langham report. The other case considered by Rundo was HP-6.

Table VII lists the results for case HP-3, recorded in the Los Alamos notebooks, starting with day 1 through day 23 and for days 321 through 324. Additional urine excretion data² referenced in the Langham² publication from day 1645 (reported as four daily samples showing an average daily urinary excretion of 0.0008% for the injected dose) and the Rundo data at day 9934 are also listed in Table VII. The Los Alamos notebook records did not identify the data from day 1645, but two samples collected on day 1674 are noted in the notebook and are included in Table VII. The recorded values for these two samples are 1.29 and 0.83 counts/min and correspond to 0.0004% and 0.0002% of the injected dose. These values do not relate to the per cent excreted value of 0.0008% reported by Langham. There were no records of spiked control samples analyzed concurrently with the injection study samples, and there is no record of correction factors being applied to the recorded results to correct for chemical losses. Each of the samples through day 324 was analyzed in duplicate. When a serious procedural problem was noted with either of the aliquots, the result was not utilized by Langham in the mathematical treatment of the data to calculate his model.

The data for case HP-6 are given in Table VIII. The samples collected on day 523 and day 1610,