

AN OVERVIEW OF THE HAZARDS ASSOCIATED WITH DEPLETED URANIUM

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SUMMARY

The use of depleted uranium (DU) in new programs or facilities can introduce new hazards to health and safety specialists, particularly those unfamiliar with basic radiological protection. It is incorrect to assume that these hazards originate exclusively from radiological properties; chemical properties must also be evaluated. This report describes the dangers of external irradiation, internal toxicity, and flammability associated with DU.

During DU handling operations, individuals may be exposed to beta particle and photon radiation fields. Due to the large dose contributed by beta particles, particular attention should be directed to preventing excessive loses to the sensitive basal opithelium of the hands. A good estimate of the contact dose from a slab of DU is 235 mrad/hr. However, the use of heavy leather or rubber gloves can reduce hand doses.

Inhalation of depleted uranium compounds presents the predominant internal toxicity hazard for occupational workers. For soluble compounds, which are rapidly translocated from the lung into the circulatory system, guidelines for allowable intake originate from the DU levels at which toxicity occurs in the kidney. The upper limit for soluble DU aerosols is 200 ug U/m³.

When insoluble DU compounds are deposited in the lung, excessive lung irradiation is the primary concern. Lung doses are dependent on the residence time of DU in the lung. Unfortunately, discrepancies between laboratory and occupationally derived data can complicate the evaluation of an exposure hazard, leading to a need for separate analyses of each situation. An occupational upper limit for insoluble DU in the air is 1 x 10^{+10} µCi/cc.

Since large masses of compact metal are relatively inert, with oxidation usually confined to the outer surfaces, the fire hazards of uranium metal components are typically associated with the pyrophoric nature of small fragments. Nonaqueous agents are preferred for extinguishing uranium fires to avoid uranium hydride production and vigorous steam - uranium interaction.

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AN OVERVIEW OF THE HAZARDS ASSOCIATED WITH DEPLITED URANIUM

INTRODUCTION

In the natural environment, uranium exists as a composition of three primary isotopes, ²³⁸U, ²³⁵U and ²³⁴U. While physically and chemically similar, these isotopes pessess different nuclear properties, and the ratio in which they occur strongly influences havard appraisals. Consequently, comparisons of the hazards of natural uranium with other, artificallyproduced isotopic compositions will depend on the relative abundance of the three isotopes.

A by-product of the 235 U enrichment process, depleted uranium (DU) consists of a larger percentage of 238 U and lesser percentages of 235 U and 234 U than natural uranium. The nonnuclear industry has advantageously used DU because of its high-density heavy metal properties. This has placed large amounts of DU into programs where the specific hazards associated with uranium are not recognized.

An accurate hazard evaluation of uranium compounds depends on their chemical forms and isotopic compositions. Therefore, nonnuclear programs utilizing Du have different hazards from those of the nuclear industry, which uses enriched uranium. This report identifies the general hazards and infjuencing factors associated with DU uses. These hazards are classified as external radiation exposure, internal toxicity, and flammability. To provide a sense of perspective, several comparisons between the hazards from DU and other isotopic uranium compositions are presented.

EXTERNAL RADIATION EXPOSURE

Concern about external radiation exposure arises primarily from the beta particle emissions of thorium-234 and protactinium-234m, the first two daughters of the uranium decay series.⁽¹⁾ Three beta particle spectra are associated with the decay of the two daughters: two spectra from ²³⁴Th with maximum energies ($E_{\rm max}$) of 0.103 and 0.193 MeV, and one spectrum from ^{234m}Pa with an $E_{\rm max}$ of 2.29 MeV.⁽²⁾ A minor beta contribution is made by thorium-231. the daughter of ²³⁵O.^{*} in addition to beta particles, several of the daughters and ²³⁵U also emit photons. However, these photons are either emitted infrequently or are of low energy. Consequently, they make a minor contribution to the total external radiation nazard.

Because of the abundance of beta particles, control of personnel exposure is primarily directed at the prevention of excessive irradiation of extremities, particularly the hands, during uranium handling operations. A commonly accepted value for the surface dose rate from a large metal slab of natural uranium through 7 mg/cm² of tissue-equivalent material is 235 mrads/hr. (1,2) Based on the quarterly dose limit of 18.75 rems to the extremities, direct hand-to-uranium contact would be limited to about 6 hours per week. The interactions of beta particles with matter favor the use of thick gloves to reduce hand-exposures, and significant reductions have been noted. (1,3) Photon exposure to gloved hands continues; however, photons only contribute about 4 mrads/hr of the total dose rate received at close proximities to uranium objects. (3)

At greater distances, whole-body exposure to photons becomes the fundamental consideration. Occupational experience indicates that wholebody exposures due to uranium have not been significant and have been considered a secondary hazard.⁽⁴⁾ However, exposure rate measurements may be justified to determine personnel dosimetry needs at large DU stockpile installations. Shielding and distance can readily reduce exposure rates to acceptable levels. Precautions should be taken to avoid exposure rate increases from the production of Bremsstrahlung (secondary radiation generated by beta particle interactions), when shields or containment devices composed of high atomic number elements are placed close to uranium objects. Maximum exposure rates at 50 cm from an aluminum-shrouded 3.3-kg DU bar are 0.039 mR/hr.⁽⁵⁾

Differences between surface dose rates from depleted and natural uranium are minimal. However, theoretical estimates of the photon contribution from DU are less than those from natural uranium, due to very slight differences in the ²³⁸U content and a large reduction in the ²³⁵U content.

A special exposure hazard exists where DU is melted, purified and cast. During such operations, 234 Th and 234m Pa migrate to form areas of high radioactivity along the surfaces of molds and in cooler areas of furnaces. Workers have received 2 to 3 rads per week to the hunds and possibly to the eyes during vacuum pellet casting. (1)

Freshly purified DU will produce greatly reduced dose rates. As the daughters build up from the subsequent decay of ²³⁸U and ²³⁵U, dose rates will increase until the radioactive decay equilibrium is attained. Therefore, the degree of daughter buildup should be considered when evaluating possible exposure hazards. Because of secular decay equilibrium conditions, the fractional daughter buildup, F, can be estimated by

$F = 1 - e^{-\lambda_1 t}$

where λ_1' (the decay constant of 234 Th) = 0.0288 day⁻¹ and t = time in days after purification.

To summarize, dose rates from DU are mean rable and result primarily from the emission of beta particles. The primary consideration for personnel protection is to prevent excessive hand doses. Exposure rates associated with whole-body irradiation are generally of secondary importance. Overall, external radiation hazards associated with DU utilization are small, and simple but adequate means are available to control any exposure hazards.

INTERNAL HAZARDS

The introduction of uranium compounds into the body is of primary concern in the industrial and occupational environment. Inhalation is the most common intake route, but internal deposition from contaminated wounds and ingestion can also occur. Information on uranium metabolism is available. However, the development of general statements on relative degrees of hazards following intake is complicated by discrepancies between laboratory and occupational date.

Following the inhalation of DU compounds, potential for harm arises from the radiological and toxicological properties of uranium, with special danger from lung irradiation and chemically related kidney damage. The total deposited mass, the compound's solubility in lung fluids, and the site of the lung deposition influence the degree and type of hazard. Other factors which must be considered on an individual basis are exposure history and deviations of metabolic rates from accepted norms.

INHALATION OF SOLUBLE URANIUM COMPOUNDS

Inhaled soluble unanium compounds rapidly enter the circulatory system and are complexed by proteins and bicarbonate ions. (6,7) Clearance from the blood leads to accumulations in the bone and kidney. For DU, the accumulation in the kidney is the crucial factor because kidney damage will occur before DU deposits in the bone or kidney exceed radiation dose limits.

Chemical toxicity in the kidney results from decreased solubility of the DU compounds in the urine due to the reabsorption of complexing bicarbonate ions and increased acidity. ⁽⁸⁾ The free urinary uranium affects the tubule cells by interfering with normal membrane respiration. ⁽⁹⁾ An early indication of uranium intoxication is the presence of catalase in the urine. ⁽¹⁰⁾ Renal damage has traditionally been indicated by the presence of casts, red blood cells, and albumin in the urine. ⁽¹¹⁾ Tissue damage is usually localized in the proximal convoluted tubule, with total repair possible 14 days after a mild intoxication. ⁽¹²⁾

According to Alexander, (13) the nephrotoxic limit of 3 mJ U/H of kidney tissue established by the International Commission on Kadiological Protection (ICRP), can be related to a single deposition of 2.7 mg of uranium into the blood. The validity of this value assumes an effective half life of 15 days. Total blood uranium estimates are commonly assumed to be twice the uranium content in 1 L of urine from a 24-hour sample. Albuminuria has been associated with a urinary excretion of 2 mg/L for acute exposure: for prolonged or chronic exposures, albuminuria may occur with smaller urinary uranium levels. (15) Alexander (13) has thoroughly discussed the interpretation of urinalysis data as a function of sampling frequency to obtain hazard estimates.

To relate working place atmospheric levels of uranium to the development of nephrotoxic conditions, several concentration guides have been established. Most guides, like those listed below, do not specifically identify DU, but the values for natural uranium or 238 U can be applied because the toxic limit for DU is uniquely dependent on the total mass of uranium deposited in the body. Ford⁽¹⁶⁾ has stated that the kidney toxicity limit is applicable for all 235 U enrichments up to 5%. The most common concentration guides are as follows:

- The current Threshold Limit Value (TLV) for soluble uranium is 200 µg/m³, which replaced the conservative 50 µg/m³ because of the lack of harmful evidence associated with the use of the older value. (17)
- The maximum permissible concentration in air (MPC.) for natural uranium based on a 40-hour work week is $7 \times 10^{-110} \, \mu \text{Ci/cm}^3$ (18) This is a mathematically derived limit based on principles of biological modeling. To convert the MPC_a to mass units, the specific activity of natural uranium (0.33 $\mu \text{Ci/g}$, based on the special curie for natural uranium 7.57 $\times 10^{10}$ alpha dis/sec) must be used. The resulting mass MPC_a is 210 $\mu \text{g/m}^3$. The use of the special curie has been a source of confusion and has been eliminated. Based on the usual curie definition, the specific activity of natural uranium is 0.676 $\mu \text{Ci/g}$, which leads to an MPC_a of 1.4 $\times 10^{-10} \, \mu \text{Ci/cm}^3$ or 1 $\times 10^{-10} \, \mu \text{Ci/cm}^3$

when rounded off. An adapted MPC_a in activity units for DU is 7.6 x 10^{-11} µCi/cm³. This value is based on the permissible level of 210 µg/m³ and a specific activity of 0.36 µCi/g for DU.

Legal limits specific for DU have been listed in Appendix B of 10 CFR 20. $^{(19)}$ For occupationally exposed individuals, the allowable concentration is 200 $_{\rm Vg/m}^3$.

INHALATION OF INSOLUBLE URANIUM COMPOUNDS

The inhalation of insoluble uranium compounds presents radiological health implications due to the extended pulmonary region clearance times associated with lung deposition of insoluble particulates and due to alpha particle emission from the uranium isotopes. Of principle concern is the development of latent effects associated with excessive irradiation of the lung. Because the effects are radiation-induced, the hazards of inhaling DU are similar to those of inhaling the other insoluble alpha-emitting substances. However, it has been suggested that the chemical effects of uranium may reinforce the development of latent tissue damage in cases involving large uranium lung burdens. (20)

Leach et al.⁽²¹⁾ have used monkeys and dogs in an extensive investigation into the biological effects of UO₂ when inhaled under chronic conditions simulating a 5-year occupational exposure. Organ burdens of monkeys and dogs were analyzed during the 5-year exposure period and up to 6 1/2 years post-exposure. The inhaled aerosol was characterized as 1 μ mass median diameter (MMD) with a concentration of 5 mg/m³.

The lungs and associated tracheobronchial lymph nodes (TLN) contained over 90% of the deposited uranium. Maximum equilibrium lung levels were attained after 6 months of exposure, and were 200 μ g U/g and 3600 μ g U/g for the dog and monkey respectively. Translocation to the tracheobronchial lymph nodes was represented by a slow, 4-year increase in TLN uranium. Maximum levels were 50,000 to 70,000 μ g U/g in dog and monkey. Based on an allowable dose to the lungs of 15 rem/yr, the maximum equilibrium concentration of uranium in the lung is 25 μ g U/g of tissue.⁽²²⁾ At the end of the total exposure period, dose rates were about 60 rads/week

to the TLN and 2 to 3 rads/week to the lung. Pathological changes due to radiation damage were infrequently observed during the 5 years; however, the results of the postexposure survival study indicated latent radiation effects.

In the postexposure survival study.⁽²¹⁾ monkeys and dogs were sacrificed and examined during the following 6 1/2-year period. Integrated alpha doses for the maximum survival period were 660 rads to the lung and 16,000 rads to the TLN for the dogs, and 1280 rads to the lung and 24.800rads to the TLN for the monkeys. Pathological examinations of dog lungs revealed pulmonary neoplasms of the adenoma and carcinoma variety. Fibrotic and necrotic TLN were also reported. Damage to monkey lung and particularly to TLN was limited to severe fibresis. The investigators commented that the development of neoplasms from natural uranium was unique to this study.

Permissible concentration guides have been developed for insoluble uranium aerosols. Based on a specific activity of 0.36 μ Ci/g, the MPC a listed in ICRP Publication 2 is 1 x 10⁻¹⁰ μ Ci/cm³ for natural uranium and 2380. An acceptable MPC a for DU would also be 1 x 10⁻¹⁰ μ Ci/cm³ because the detrimental effects are dependent on the activity of the deposited material. When orginally developed, the MPC expressed in mass units was 180 μ g/m³. However, conversion of the values just presented to mass units will not equal 180 μ g/m³ due to rounding error effects. Mass concentrations based on the allowable 1 x 10⁻¹⁰ μ Ci/cm³ for natural uranium, DU, and ²³⁸U are 143 μ g/m³, 278 μ g/m³ and 299 μ g/m³ respectively. The ILV for insoluble aerosols is 200 μ g/m³.

Derivations of MPC are based on a maximum amount of radionuclide allowed in an organ under conditions of equilibrium. A critical element is the effective residence time or effective half life. (For long-lived uranium, estimates of the biological half life are used.) Consequently. the accuracy of the estimate determines the level of conservatism associated with MPC. If the estimated value exceeds the true value, conservatism will be established. Unfortunately, many estimates of the lung half life of insoluble uranium compounds have complicated sufety evaluations.

Insoluble aerosols associated with operations involving DU metal are usually UO₅ and $U_{2}O_{2}$. (3) While both are insoluble in water, solubility in

lung fluids has been reported (23,24) and might be attributed to the presence of complexing ions and proteinaceous material in lung fluid. (6)Based on several case histories of occupational exposure to uranium dust, Quigley et al. (23) estine is a biological half life of 30 to 60 days. Spoor (2°) recommended chest half times of 35 days for UO₃, 100 days for UO₂ and 360 days for U₃O₈. In contrast, Leach et al. (20) reported extreme insolubility of UO₂ in animals. Lung half times of 420-660 days and TLN half times of 1500-1950 days were found in monkeys. Comparable lung half times were calculated for dogs but TLN half times ranged from 600 to 780 days. The investigators suggested that extensive TLN damage in monkeys explained the half time differences between the monkeys and dogs.

The ICRP has incorporated a 120-day half time estimate for derivation of the insoluble MPC_a,⁽²²⁾ whereas the Tosk Groups on Lung Dynamics suggested a 150-day half life for UO₂ and a 120-day half life for U₃O₈.⁽²⁵⁾ Alexander ⁽¹³⁾ classified both UO₂ and U₃O₈ as class W compounds (relatively insoluble), following the classification system presented by the Task Group on Lung Dynamics. The half times assumed by Alexander were 50 days for both compounds.

Alexander⁽¹³⁾ compared the model prediction (with a 50-day half life) to human autopsy data to estimate the conservatism associated with a 50-day half life. He found the model could overestimate the lung burden by a factor of 5. When UO_2 or U_3O_8 behaved like class Y compounds (insoluble), a half life of 500 days was used and the model overestimated lung burdens by a factor of 70; however, because comparison data was very limited, the large factor was considered acceptable.

FLAMMABILITY HAZARDS

Careful attention to potential fire conditions is needed because of the extreme reactivity of metallic uranium and certain uranium compounds. The amount and form of material, as well as its production and u.e., influence the degree of hazard. Also, the extreme variability of uranium's incendiary properties makes predicting potential hazards more difficult. The degree of depletion does not alter flammability because the primary factor is chemical oxidation.

The metallic compound is a commonly used form of DU. As a general rule, massive compact metallic uranium is relatively inert and unable to spontaneously ignite and maintain its burning. Fires have been uncommon during rolling, forging, or storage operations, indicating that external heat sources are incapable of starting a fire. (26) Oxidation of compact metal may result from prolonged heating at temperatures above 350°C,(27) but oxidation of compact metal at low temperatures is usually limited to the surface, forming a permeable layer of UO₂ and U₂O₃.

Isolated instances of spontaneous inginition of compact metal have occurred, generally associated with the presence of residual uranium hydride due to incomplete metal production, the presence of small amounts of moisture, and the presence of contaminating mutals.⁽²⁶⁾ Uranium hydride is a possible by-product of metal oxidation by water and is extremely pyrophoric.⁽³⁾ Uranium hydride and small amounts of moisture have been impricated in surface pyrophoricity of metal during vacuum casting and bomb reduction operations.⁽²⁶⁾ Alternate methods of metal production can reduce compact uranium pyrophoricity by eliminating possible uranium hydride residues.

The pyrophoric behavior of uranium metal sharply increases as object size decreases. The larger surface-to-mass ratio of small fragments permits enough oxidation and retained heat to allow continuous combustion. Small fragments can spontaneously ingnite upon exposure to air at room temperatures, and dispersed powders of metal can be explosive. ⁽³⁾ Consequently, machining operations producing metal chips should be carefully monitored. The liberal use of mineral oil coolants is usually satisfactory for preventing small fragment fires.⁽²⁶⁾ Aqueous coolants are not recommended because water exidation and possible branium hydride formation may allow minor fires which might be prevented by the substitution of mineral oil. Because fires involving a few chips can usually be extinguished with coolant or allowed to burn out, while fires involving many chips can be extremely difficult to extinguish.⁽²⁶⁾ chips should be stored uncompacted, in reduced numbers, and under mineral oil. When using water storage, there is danger of spontaneous combustion with explosive force.⁽²⁶⁾

Once ignited and self sustaining, fires involving compact uranium are slow burning and incandescent, liberating large amounts of heat. In rare cases of compact metal pyrophoricity, ignition occurs after extended periods of heat accumulation from slow oxidation. Inmediate ignition of massive metal is confined to the surface, probably due to moisture-surface interactions forming uranium hydride. ⁽²⁶⁾ In contrast, fragment fires can ignite immediately or slowly, depending on the rate of oxidation and surface area. The intense heat liberated from moist powder oxidation can burn through shouls of iron and steel.

Because of the heat and reactivity of uranium fires, careful selection of extinguishing agents is necessary. Temperatures above 300°C will cause continued oxidation by CO_2 and N_2 .⁽³⁾ A severe steam-metal interaction precludes the use of water as an extinguishing agent; however, uranium fires have been extinguished with water and CO_2 .⁽²⁶⁾ Inert-gas sprinkler systems may be used for small fires if provisions are made to warn personnel of gas release and to maintain sufficient gas flow to prevent reignition due to heat retention and air reentry. Gases should not be used when large amounts of heat are generated because of their poor cooling properties.⁽²⁸⁾ If building integrity is ensured, small amounts of compact uranium can be allowed to burn out because the slow burning permits isolation for controlled incineration.⁽²⁶⁾

*suming a 3% release from a uranium fire and adverse meteorological conditions producing a maximum hazard at a distance of 100 m from the fire, Cook⁽²⁹⁾estimated a safe building inventory of uranium to be 0.5 tonnes.

With such an inventory, the potential release was estimated to be 2 kg in a soluble form. The inventory limit contained a safety factor that accounted for building containment and for explosions releasing the entire cloud. In the latter case, the dispersion height of the cloud would be higher, reducing the level of cloud doses compared to doses resulting from ground releases.

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