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HEALTH PHYSICS ASPECTS OF HANDLING URANIUM-233 FEED MATERIAL

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1. INTRODUCTION

In the past year or so there has been considerable thought given to the use of natural thorium, which is primarily the isotope Th-232, in proposed nuclear reactors. It follows that the uranium feed material bred by this process will be separated and used as nuclear fuel. This "feed material" contains primarily U-233, with smaller quantities of the isotopes U-232, U-234, U-235, and U-238. The processes of separation, purification, and fabrication of the feed material require much closer health physics supervision than do the various enriched or naturally occurring mixtures of isotopes U-234, U-235, and U-238 found in natural uranium.

Two distinct radiation problems exist in the purification and fabrication of the feed material. These are the control of the alpha and gamma radiation. The isotope U-233 presents only a simple alpha control problem. However, the feed material encountered over the past three years has contained from 20 to 80 ppm of the isotope U-232[1]. It is the daughters of U-232 which constitute the gamma radiation problem. In the absence of either the gamma or alpha consideration, the remaining activity could be dealt with in a routine manner.

2. ALPHA PROBLEM AND PERMISSIBLE CONCENTRATIONS

Although the half life of U-232 is some 2×10^3 shorter than that of U-233, the relatively low concentration of the U-232 does not appreciably change the specific activity of the feed material. With no U-232 the specific activity of the uranium isotopes in the feed material[2] would be about 9.4×10^{-3} curie per gram. With 80 ppm of U-232 the activity is raised to about 10.2×10^{-3} curie per gram. Because of this high specific activity as compared with the other uranium mixtures encountered, at the Los Alamos Scientific Laboratory (LASL) it was decided that all work of chemical purification, metallic casting, and fabrication would be done in dryboxes.

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DISCLAIMER

In this connection the maximum permissible body burden, q, and the maximum permissible air concentration, MPC(air), should be considered. The Internal Dose Subcommittee (IDS) of the International Commission on Radiological Protection (ICRP) has prepared, for presentation at this Conference, a revised and extended table of permissible concentrations of radioisotopes in air, water, and for various critical organs of the body. At the time of this writing only the "Tables for 98 Elements, which Contain the Preliminary Biological Constants, Effective Energies, q and MPC Values" were available. The tables were based on an exponential elimination function and were for soluble materials. With the permission of the IDS of the ICRP, the data to follow were taken from the "Preliminary" tables, and should be revised according to the data presented to this Conference by the ICRP. For U-233, which accounts for most of the activity of the feed material, the q and MPC(air) were 0.133 μc and 6.1×10^{-11} $\mu c/ml$ (136 $d/m\text{-}M^3$) of air, respectively. These are based on the bone as the first critical organ, a biological half-life (after fixation) of 300 days, and the fact that only 0.0825 of that quantity inhaled actually becomes fixed in the bone.

The MPC(air) of 6.1×10^{-11} $\mu c/ml$ of air is for continuous exposure and should be modified for occupational exposure. Assuming a 40 hour work week, a 50 week work year, and that one-half of the air breathed each 24 hours of the day is breathed during the 8 hour work period, the MPC(air) for occupational exposure becomes 1.8×10^{-10} $\mu c/ml$ (390 $d/m\text{-}M^3$) of air. During the years 1956 and 1957, in those areas at LASL where the uranium feed material is chemically purified, the average air count in the rooms containing the dry box chains has been 4 $d/m\text{-}M^3$. This is substantially below the permissible value.

Protective measures for occupational exposure consist of daily air sampling, use of respirators, and urine assays. It is the practice at LASL to require the wearing of respirators when the average level of U-233 in the air exceeds the permissible occupational level. The level of activity is not routinely determined until the day after the sample of air is collected. However, because of experience it has been possible to adopt satisfactory policies that assure a minimum of inhalation of U-233. High air counts are expected whenever any of the following jobs are done: changing drybox gloves, performing other maintenance on the dryboxes, decontaminating a spill of radioactive material, and performing air transfers into or out of the dryboxes.

When such operations are contemplated, it is the practice to require the wearing of respirators in the area while the work is going on as well as for a minimum of one hour afterwards. It is due to this preventative use of respirators that thus far no person working with the feed material has submitted an excessive urine sample.

The only unexpected over-permissible air concentration encountered occurred in February 1957, when a mild explosion took place in one of the drybox lines during nonworking hours. The high count at that time was 1400 $d/m\text{-}M^3$ for an 8 hour period. Although the incident was not discovered until after the area was entered, the persons who entered the room briefly, without benefit of respirators, did not show excessive amounts of U-233 in their urine assays.

Normally, urine samples are required monthly of persons working the feed material line. It can be shown that with no further exposure the initial rate of excretion of a fixed permissible body burden in the bone would be about 455 d/m-l in urine. With a continuing exposure the level to be expected is more nebulous. Since only 8.25% of that taken into the body by inhalation reaches, and is fixed, in the first critical organ (bone), the remaining 91.75% must be eliminated. Actually, urinary excretion of the uranyl carbonate complex is the main method of excretion from the body, and 85% of an acute dose is eliminated by this process in the first 24 hours.[3]

For a working number, LASL has selected the level of 50 d/m-l of U-233 in urine as indicative of a significant exposure. On the occasion of a sample exceeding 50 d/m-l concentration, the person is resampled immediately. If the level has dropped below 50 d/m-l, nothing further is done. However, if the level remains high, the situation is investigated, and the cause is eliminated.

During 1956 and 1957 the average urine counts for these persons working exclusively in the uranium feed material chemical processing area has been about 13 d/m-l. The highest sample encountered thus far was about 75 d/m-l, but this level was not maintained.

LASL is in the process of changing from a solvent extraction method of recovery of U-233 from urine to an ion exchange method. The latter method is expected to give a better clean-up of the thorium and thorium daughters, and should have about 90% recovery of the uranium.

3. GAMMA PROBLEM

As mentioned in Section 1, the gamma problem in handling the feed material is not due to the U-233 isotope and its daughters, but rather to the U-232 isotope and its daughters. While contamination to the extent of 20 to 80 ppm does not seem very significant, a comparison with natural thorium will illustrate the fallacy of this argument. U-232 has a radioactive half life of 74 years, and its immediate daughter, Th-228, has a half life of 1.9 years. Th-228 is also the third daughter of the most prevalent isotope of natural thorium (Th-232). It may be shown that at equilibrium (i.e., the condition when the least quantities of both natural thorium and uranium feed material would be required), it would require 3750 times as much natural thorium as it would feed material with 20 ppm of U-232 to give the same rate of formation of Th-228. Since the gamma radiation problem in both cases is from the daughters of the Th-228, it is obvious that, by comparison with uranium feed material, natural thorium presents practically no external radiation problem in handling. In the case of the natural thorium the much larger bulk would effectively shield and dilute the source of gamma radiation.

In practice, the gamma problem in handling the feed material is one of growth of activity. The U-233 and its daughters and the U-232 itself present no problem. However, the gamma activity of the daughters of U-232 is another story. All of the daughters after Th-228 have half lives of less than four

days. As a result of the 1.9 year half life of the Th-228, there will be an exponential build-up of activity which maximizes at about 10 years, and which then decays with a 74 year half life.

The calculations presented in Table I illustrate the growth of gamma activity and indicate that the predominance of activity comes from the U-232 and its daughters. The dose rates are given in roentgens per hour at 1 cm from a 0.635 cm thick, 5.08 cm diameter disk of feed material (20 ppm of U-232) at various ages. Self-absorption of the gammas is included in these calculations, and it was assumed that no Th-228 was present initially. For feed material with a different concentration of U-232 the figures given may be multiplicatively adjusted by the ratio of the concentrations of U-232 in the feed material under consideration to that used in this calculation.

TABLE I.

CALCULATED R/HR DOSE RATES FROM THE TWO PARENT-DAUGHTER CHAINS IN THE URANIUM FEED MATERIAL

	1 Day	4 Weeks	32 Weeks	10 Years
U-233 + Daughters	0.0029	0.0032	0.011	0.15
U-232 + Daughters	0.19	0.37	3.2	14.

Two disks of the LASL uranium feed material (about 20 ppm of U-232), 0.32 cm thick and 5.08 cm in diameter, were procured about two years ago. In order that this material may be handled outside a drybox, one piece is nickel-coated and the other is sealed in a polyvinyl chloride envelope. The growth of the gamma radiation output of each has been followed. The surface dose rates through a 6 mg/cm^2 filter were measured with an extrapolation chamber. By interspersing additional layers of polyvinyl chloride on the plastic-coated piece, it was determined that the dose rate, exclusive of the alpha radiation, of the bare piece of feed material was 1.18 times that measured through the plastic. This factor is believed due primarily to the extra distance from the surface rather than shielding by the plastic. Typical of the growths are the surface dose measurements made on the plastic-encased piece 28, 430, and 540 days after casting. These were 6.7, 58., and 68. rad/hr, respectively. Measurements made with an ionization-type gamma monitoring instrument at shorter intervals during the same period verify the build-up indicated by the extrapolation chamber measurements. For purposes of orientation, the dose rate measured with the surface of the source material about 4. cm from the center of the sensitive chamber of the monitoring instrument was 4.1 r/hr for the plastic-encased piece and 6.0 r/hr for the two pieces stacked. The measure of the two pieces stacked corresponds roughly to the theoretically calculated dose rates. However, the observed rate of build-up is slightly less than the calculated rate. This indicates that some slight amount of Th-228 is apparently present, and it corresponds to that quantity which would be expected to grow in after 30 days.

The primary gammas from the U-232 chain range from about 0.04 to 2.6 Mev. After emerging from feed material 0.32 cm thick or greater, the calculations, mentioned earlier, indicate that about one-half of the total dose rate would be due to the 2.6 Mev gamma of isotope Tl-208.

For gamma rays of this energy spectrum, moderate amounts of lead shielding are not very effective. Some rough measurements made on the disks about 430 days after casting indicate that 0.635 cm of lead shielding reduces the roentgen output to one-half the unshielded value. Each additional 0.635 cm of lead reduces the dose rate by a factor of about 0.7, up to a total thickness of 5.08 cm (measurements with thicker lead shields were not made). This indicates that the gammas emerging after the first 0.635 cm of lead shielding are essentially all 2.6 Mev in energy.

4. LOS ALAMOS EXPERIENCE WITH GAMMA PROBLEM IN CHEMICAL PURIFICATION

From the discussion above, the gamma emission rates are seen to be a function of the degree of thorium clean-up in the previous chemical purification or metal casting, and the period of time since that operation. During 1956 and 1957 batches of the material have been received reading from 0.1 to 3.0 r/hr (readings taken with monitoring instruments in contact with the plastic shipping bottles). Old material, which was reworked, has also shown emission rates in this same range. These emission rates are for about 250 grams of feed material in about 2 liters of solution.

During 1957, in the chemical purification processes which include peroxide precipitations, hydrofluorinations, and reductions to metal, the radiation levels of the material worked have averaged somewhat higher than in 1956. This is reflected in the total gamma exposure received by all persons working the material, and taking into account the quantity of material worked. As measured by film badges, the official record of radiation exposure at LASL, the total gamma exposures received was about 44.0 rem in 1956, while working about 32. kg of material. In 1957 the total gamma exposure was about 18.0 rem while working only about 8. kg of material. Thus, the 1957 exposure rate averaged about 0.9 rem more per kilogram of metal than in 1956.

During 1956 and 1957, a total of nine persons have been involved, to a greater or lesser extent, in the chemical purification of the feed material. In 1956 two persons received about 11.0 rem each, while the others received about 4.0 rem each; only two exposures greater than 0.6 rem in two weeks were experienced. At that time, these exposures were well within the permissible limit of 15 rem per year.

In 1957 an attempt was made to keep all operators' exposures below 5.0 rem per year, in accordance with the recommendations of the NCRP and ICRP. By judicious use of personnel rotation, only one man exceeded 5.0 rem, and he received only 5.3 rem. Another received almost 5.0 rem and the others averaged about 1.25 rem. Only one exposure greater than 0.6 rem in two weeks was recorded.

The fact that so few exposures exceeded 0.6 rem in two weeks for this operation during those two years is attributed to the excellent cooperation

of the chemical operators, who have kept their own weekly records of pocket dosimeter exposures and have governed their work accordingly. Wrist film badges, as well as body film badges, were worn by the operating personnel. In neither the case of the body nor the wrist badge was beta exposure significant. It was also observed that for these operators the body exposure, and never the wrist exposure, was the restricting factor.

To date very little shielding has been used. The filtrate bottles have been shielded with 1.27 cm of lead, and an external, portable 1.27 cm lead shield was constructed and used for the first precipitation operations. The exposures stated were accumulated with this meager shielding in position.

In order to reduce the body gamma exposure, it is necessary to separate out the daughters of the U-232, if only for a short while. Some work by J. A. Kircher and others at LASL has indicated cation exchange columns would accomplish the necessary clean-up. A heavily shielded drybox was designed and constructed for this job. It was designed to handle two lots of feed material, which start out at 2 r/hr each. With an operating time of 100 minutes per week, the shielding was designed to allow the operator 0.10 rem/wk. This ion exchange procedure is an addition to the procedures used during 1956 and 1957, and is expected to eliminate one or more of the peroxide precipitation operations.

This box has been in operation only a few weeks. The first few batches of material did not clean up as expected. After a few modifications in procedures it appears that the clean-up will reduce the gamma emission by a factor of 1/3rd to 1/10th the original level. Tenth-scale pilot runs indicate that following the ion exchange step the gamma activity will decrease for a period of 7 to 25 days owing to the removal of the Th-228. Currently it requires about 8 days after the ion exchange to perform the remaining chemistry necessary to produce metallic uranium feed material buttons. It is expected that the use of this ion exchange technique will appreciably reduce the personnel exposure.

5. LOS ALAMOS EXPERIENCE IN METAL FABRICATION

In addition to the chemical processing, some limited experience has been gained in casting and machining the metal. At LASL the casting and machining of this uranium metal is done in the same drybox line in which plutonium feed material (primarily Pu-239) is cast and machined. Since the permissible air concentration for Pu-239 is much less than that for U-233, no additional restrictions were necessary, as far as the alpha problem was concerned. During a seven week period about 14 kg of the uranium metal were fabricated. The average body exposure per week for the five persons who cast, machined, and nickel-coated the metal was 0.7 rem. For these operations the wrist exposure was about the same fraction of permissible exposure as the body exposure. During this period five exposures exceeded 0.3 rem/wk. This case is typical of the problems which have arisen in connection with the metallic fabrication operations. The yearly total exposures received by the persons doing this work are not given. Most of their exposure did not come from these operations, which account for only a small portion of their actual working time.

6. RECOMMENDATIONS

The high exposures experienced at LASL in the metallic fabrication of this uranium feed material is the result of using metal buttons which had been produced over a period of six or more months. If the production of reactor components of this uranium feed material becomes a full time production job, the following would be desirable:

1. A completely remote control line for chemical purification.
2. A shorter delay between chemical purification and metal fabrication.
3. A remote control process for casting and fabrication of metallic components.

If the above cannot be accomplished because of technical or monetary restrictions, a large increase in drybox shielding and in personnel performing the operations, over that discussed in this paper, would be necessary to prevent excessive radiation exposures.

FOOTNOTES

1. It was predicted at the Thorium-U-233 Symposium held at Brookhaven in January 1958 that for maximum economy of reactor operation, the thorium would not be removed until the U-232 concentration would reach 150 to 300 ppm of the U-233 concentration.

2. The combination of isotopes used to calculate the specific activity was U-233, 98.89 wt./%; U-234, 0.88 wt./%; U-235, 0.03 wt./%; and U-238, 0.20 wt./%.

3. Private communication with W. H. Langham, LASL.

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