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NEPTUNIUM237 CONTAMINATION PROBLEM, PADUCAH, KENTUCKY, FEBRUARY 4, 1960

SYMBOL : :MDB

Those contacted were Mr. B. Stiller and Mr. Nietsche of the AEC, Paducah Area Office and Dr. Neal Ward and Messrs. Don Levin, Ed Cain and R. G. Brown of Carbide. Mr. Joe Lenhart of OROO came up from Oak Ridge and took part in the discussions.

* Np^{237} seems to be found only in reclaimed feed material provided by Hanford and therefore it is not a problem for the other separations plants.

It is produced by one or both of the following reactions:

(1) ${}_{92}U^{238}$ (N, p.2n) ${}_{93}Np^{237}$

(2) ${}_{92}U^{235}(n,\gamma)\ {}_{92}U^{236}$; ${}_{92}U^{236}(n,\gamma)\ {}_{92}U^{237}$; ${}_{92}U^{237} \xrightarrow[7\ days]{B^- + \gamma} {}_{93}Np^{237}$

This reclaimed U from Hanford now has about 0.05 g of Np/ton of U. The presence of Np was recognized as far back as 1957. At one time during 1958 this feed material had as much as 1 gram/ton but it has been lower lately because Hanford is extracting the Np for other purposes; it would not pay Paducah to try to remove completely this residue and in any case their problem comes from the Np already in the cascade units which now must be taken out, repaired, restored and put back in the systems.

The uranium comes to Paducah as UO_3 which is then reduced to UO_2 and treated with HF to get the green salt UF_4; this is then refluxed with F_2 gas in a hot cyclone type of pipe. The volatile UF_6 so formed goes out the top to be cooled and stored in the solid state in metal "bottles." All contaminants supposedly drop to the bottom of this cyclone pipe and are removed as "ashes," but it appears that Np has sufficiently similar chemical and physical properties to follow along with the UF_6 as NpF_6. The "ashes" show about 15% of Np, the rest entraining with the UF_6.

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There is a slight difference in volatility between the NpF_6 and UF_6, which is enough to result in more NpF_6 than UF_6 remaining behind when the contents of a bottle are fed into the cascades. Thus, the concentration of NpF_6 tends to build up in the "heel" as the bottle is used repeatedly. The fractional retention from a single filling is not known.

The NpF_6 passes into the cascades with the UF_6 but the differential in volatility at operating conditions (optimal for UF_6 enrichment) leads to fine deposits of NpF_6 in the tubes of the barriers and the inside surfaces of the cascade units. It is found more often at the distal end of a system of cascades and in the U^{235} channels. This can't be predicted with certainty since some units will have much NpF_6 and some none. Probably about 60% of the NpF_6 deposits out here.

The problem arises when one of these cascade units is taken out of its operation sequence and opened for replacement of barriers. There is a definite program for such restoration and in some cases they are replacing the old barriers with new ones of improved design. These cascade units are housed in thick stainless steel tanks about 12 feet in diameter and 15 feet tall; they are welded shut and in general much too large to be handled by conventional industrial hygiene measures. The units have to be moved with an overhead travelling crane, special multi-wheel trucks, etc.

The units must be cut open with torches to get at the barrier tubes; the pieces certainly can't be handled gently or contained very readily because they are too massive.

The workers are supposed to wear special MSA nose-mouth face masks but they are not controlled too closely--I watched one man push up his mask and smoke a cigarette using potentially contaminated hands and gloves. They have devised some air-scoopes to fit around the ends of the unit as it is being torched open, but I would judge them to be of limited effectiveness. There may be a filter on the exhaust line for this air collector but it was not obvious; the exhaust simply dumps air outside the building.

Nevertheless, this ventilation was said to be very helpful. Fortunately, NpF_6 does not diffuse very readily, it having been found only within 8 feet of where the cascade unit had been cut free or opened.

According to Handbook 69 calculations (where the 200-year biological half-life is used), the MPC is 8.8d/min/m^3 of air. There are 1526 d/min/µg of Np^{237} so that the MPC equals about 0.0066 µg/m^3 of air.

Also, using the figures in Handbook 69, the maximal body burden would come to 1.3 d/m/24 hr urine sample coming from an 87.3 μg deposit. This, however, is so impossible that they have been using 13 d/m/24 hr urine sample as their standard on the basis that the true body content, after being out of contact with Np for 6 months to a year, would be 10% of the 48 hour lay-off concentration. (I think I am reporting their logic correctly.) Furthermore, the solubility of Np^{++++} is quite different from Np^{++++++} and it is not known which solubility factor should be used in the calculations.

* *

Np^{237} has a $T\frac{1}{2}$ of 2.2 x 10^{6} years and omits α-partic[illegible] 4.77 mev which can't be differentiated from other [illegible] nants by pulse height analysis. Gamma rays are also [illegible] 27.4 day daughter Pa^{233} emits both β- and γ-rays which [illegible].

Recovery of Np from biological samples is poor (80, [illegible] variable) and Dr. Levin reported trouble in making u[illegible] tions at concentrations of 10^{-11} or 10^{-12} where the b[illegible] are supposed to fall, a situation similar to the NBS [illegible] distributed by Dr. Beard.

Np^{239} with its 2.3 day $T\frac{1}{2}$ and γ-rays is useful for some work but [illegible] not be satisfactory for chronic biological experiments.

Np^{237} can now be detected in urine but not consistently and it [illegible] not very reliably. With their present techniques, the average is 0.22 d/m/24 hr urine sample for 75 people. The highest was 11 d/m/[illegible] hr sample. Their spiked blank samples ran 25% to 75% of the expected values.

There are possibly 300 people at Paducah who should be checked out but they hesitate to precede to intensive studies because of the union's use of this as an excuse for hazard pay.

The whole body burden for Np^{237} by Handbook 69 is 6 x 10^{-8} curies and tests with a Np^{237} source on the Y-12 whole body counter put 7 x 10^{-9} curies as the counting limit so that the whole body counter may have some usefulness. They (Dr. Ward and others) were not receptive to the idea of sending 8 or 10 of the men with highest urine counts to Y-12 for counting.

I pointed out that we were planning to initiate biological distribution and radiotoxicological studies of Np^{237} which might have the effect of changing the MPC and burdens, but it would be two years or more before the data would be available. In view of that, I urged both Dr. Ward and Mr. Stiller to improve the industrial hygiene measures surrounding the reworking of the cascade units. I don't have too much faith in masks and the dust particles here are about 0.5μ, the very worst size, biologically speaking.

I also pointed out to Dr. Ward the need to get post mortem samples on any of these potentially contaminated men for correlation of tissue content with urine output, but I am afraid the policy at this plant is to be wary of the unions and any unfavorable public relations.

Dr. Levin seems to be one of the authorities in the field of Np chemistry (others are: Weinstock, ANL; Eugene Lamb, ORNL, George Boyd, ORNL) and is interested enough to want to continue with efforts to improve the bioassay techniques. When he succeeds in this he may be better able to tackle this problem of whole body counting.

The potential situation at Paducah is enough to warrant radio-toxicological studies. In addition, there are the requirements for Np^{237} for various devices and the exposures during separation procedures at the Hanford OPP; perhaps Savannah River also is separating this isotope. I was told that the chemical separation of Np from U is very satisfactory, but the human factor in handling gram amounts should be considered a source of potential exposure.

Thus, it appears that Paducah has a Np problem but we don't have the data to tell them how serious it is. They may get into difficulties with the present Handbook 69 numbers and the problem of the body burden will inevitably come more to the forefront.

cc:
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