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July 8, 1948

SAFE HANDLING OF PLUTONIUM PRODUCTS

Introduction

The writer, and the organizations for which he has been privileged to work in Health Physics, have established that it is reasonable to maintain a safety factor up to 10 beyond recognized health safety standards (1). Thus while the national standard for permissible exposure to external radiation has been 100 mr per day, the Hanford operation has been conducted with an average exposure of about 5 mr per day. This has never been regarded as wasteful of money or time; it has been recognized as a contributing cause of the complete avoidance of radiation injury at Hanford Works, and of the desired high morale and efficiency of the operating force.

When a similar philosophy is applied to more nebulous hazards, such as the handling of plutonium products, there is clearly an obligation to operate with working limits even further below the tentatively established standards. This has been expressed as follows:

> "Conservatism is essential when dealing with incompletely evaluated hazards. If X µg is the tolerable deposition of a toxic element deduced from early animal experimentation, and it later turns out that X/10 µg is the limit, then the health-physicist who has a number of colleagues with a body content of this element between X/10 and X µg occupies an infelicitous position."(1)

The whole history of radiation protection has been one of steady reduction of permissible exposure. The only limit which is supposed to stand critical

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investigation is that for general body exposure to penetrating radiation. The National Committee on Radiation Protection is currently engaged in formulating recommendations for the further reduction of this limit. It is highly probable that the hastily formulated limits applicable to plutonium toxicity will be continuously reduced as knowledge advances. Two such changes have already been made. The initial value for permissible deposition of plutonium in the skeleton was 50 μg. This was quickly changed to 5 μg, and then to 1 μg. The writer maintains that there is little or no definitive evidence that such a deposition is harmless to the individual in the long term picture. It is a bench mark built on shifting sands, and a firm foundation may only be found well below this mark. Therefore, the present legitimate exposure to plutonium is that minimum resulting when all feasible operating precautions are applied. If this limit is more than one-tenth of the current formal permissible exposure there is reason to question the safety of operation. If it falls below this, there is probable satisfaction, but still no absolute assurance of safety, and certainly no just accusation of waste of money on excessively conservative operation.

Let this general philosophy be applied to the known specific hazards of plutonium manipulation.

1. Critical Mass

Current manipulations at Hanford Works are such that this hazard is essentially controlled by basic plant design. It is anticipated that physical control in the 234-5 operation will be straightforward.

2. Neutron Emission

This may arise from subcritical accumulations of the material, or from nuclear reactions as when plutonium and fluorine are in intimate contact. Current methods of monitoring neutron exposure of personnel are inadequately developed. Moreover, recent radiobiological studies have introduced two uncertainties into the permissible exposures --

a. Slow Neutrons

The work of Henshaw et al at Oak Ridge has shown that the biological effectiveness ratio of slow neutrons to gamma rays increases as the overall time of the exposures is extended. For long term work, the slow neutron hazard may be as much as five times greater than is currently supposed.

b. Fast Neutrons

Fast neutrons of energy less than 2 Mev are certainly more damaging for equal tissue ionization than the more energetic cyclotron neutrons on which the effectiveness ratio 1 rep = 4 rem was established. It is expected that a ratio 1 rep = 10 rem will henceforth be applicable.

3. Deposition of Plutonium in the Body

The principal sites of deposition are:

a. skeleton
b. liver

Apparently, deposition studies have been made only with more or less soluble compounds eg. Pu Cl_3, Pu $(NO_3)_4$, Pu^{+4} citrate and $PuO_2(NO_3)_2$. (2). Deposition in both skeleton and liver was found to depend on the chemical form. Data on liver deposition from the Chicago, Berkeley and Los Alamos laboratories is particularly conflicting, and presumably quite sensitive to the biochemical situation. It follows that the intake of particulate oxide contamination as from metal machining may yield a quite different deposition pattern. Specifically, it would not be unlikely to find a much higher liver deposition, making that organ the site of primary interest. Such possibilities must be considered if and when the Hanford program includes these forms of exposure.

There is no recognized technique for estimating the total deposition of plutonium in liver. In the skeleton, the deposition is deduced from the low and variable excretion in urine. The best data on the excretory rate has been prepared by W. H. Langham at Los Alamos. His recent findings are unavailable at this site, but fairly recent personal communications show two well established points on the curve

1. At one month, daily elimination by urine = 0.016%
2. At one year, daily elimination by urine = 0.004%

Less certain is a value 0.1% at one week, and as much as 0.5% in the first day or two. Until proven otherwise, it will be assumed that the excretory rate continues to fall slowly beyond one year, becoming say 0.002% after two years. Assuming these data*, one can compute the net excretion rate after continuous exposure for several years. After two years this will be:

a. 0.0045% per day, if there has been no intake within the few days preceding the urine collection
b. about 0.01% if there has been uniform daily intake up to the last day.

After five years, the rate may be guessed to be about 0.002% per day.**

The measured activity in the urine will be highly sensitive to the intake of the last few days before test. One can take frequent samples and hope to detect intake by the favorable signal of recently deposited fractions, or one can take less frequent samples and gear the program to the assumption that all the deposited plutonium is of long standing, say half the exposed career of the individual. The latter is the more conservative course and is followed at Hanford Works.

Assume the net elimination rate to be 0.004% per day with 1 μg Pu in the skeleton; the 24-hour urine sample will yield 5.6 dis/min. The method of analysis is nominally sensitive to a few parts of a disintegration per minute. Nominal results above 0.65 dis/min (ie: not more

*If the figures are somewhat changed, the general conclusions will not be affected

**We have previously assumed that the excretion rate several years after the intake of new plutonium will be 0.001% per day

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than 0.12 µg Pu deposited) are retested. However, due to poor statistics at these low levels there is a 30% chance of missing a retest if the true activity of the sample just exceeds this limit, but only a 0.1% chance of missing twice this amount. An analysis of 3150 Hanford urine samples has shown an average activity of 0.136 dis/min with a standard deviation of 0.161 dis/min(4) Blank samples gave an average of 0.133 dis/min, with standard deviation 0.140 dis/min. In other words, the Hanford personnel is statistically free from plutonium deposition. Furthermore, no single individual has shown a confirmed emission of more than 0.7 dis/min, or an assumed maximum deposition of 0.125 µg Pu.

This location is not prepared to accept a system which regards 7 counts per minute or 14 dis/min in the 24-hour urine sample as permissible level, for three reasons:

1. According to local views this may imply 2.5 µg Pu deposition now, and 5 µg if continued for 5 years or more.
2. With a formal limit of 1 µg Pu, there is uneasiness about <u>actual</u> depositions above 0.1 µg.
3. Significant deposition has been shown to be unnecessary in the 231 Building operations, without absurd or restrictive precautions. There is no reason why 234-5 Building operations should not be as cleanly conducted.

The foreseeable modes of entry for eventual bone deposition are:

1. ingestion
2. inhalation - transmittal through the lung lining or removal by ciliary action with secondary ingestion
3. implantation, as by a wound
4. absorption through intact or broken skin

Protection thus involves

a. clean surfaces throughout work rooms
b. clean air
c. protection against wounds
d. protection of exposed skin

The proposed design of the 234-5 Building has been predicated on satisfactory management of these phases, as demonstrated in the 231 Building.

4. <u>Inhalation of Plutonium and its Compounds</u>

The inhalation hazard falls partly in the bone deposition hazard as shown above. Separate is the risk of direct irradiation of the lung and associated lymph nodes by more or less permanently fixed deposits. The generally accepted permissible concentration in air is 5×10^{-10} µg Pu/cc. This is the concentration believed to lead to a lung irradiation of 0.01 rep per day if the biological "half life" in the lung is a few months.* Three recent factors favor reduction of this limit:

*J. G. Hamilton reports the "half life" in rat lung as 6 months. See Ref. 2

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1. The biological effectiveness ratio of alpha radiation to gamma radiation was taken as 1 rep = 10 rem almost arbitrarily, because it must be higher than the fast neutron to gamma ratio 1 rep = 4 rem. Since this is now to be changed to 1 rep = 10 rem, the alpha-gamma relation must go to 1 rep = 20 or 25 rem.

2. The permissible weekly exposure is to go to 0.3 rem instead of the previous 0.1 rem per day. (1) and (2) together give a reduction by a factor of about 5.

3. Recent emphasis on particulate contamination points up the possibility of local damage to the lung even when the total deposition is far below that which leads to a tolerable condition for the <u>uniform</u> deposition presupposed in the earlier computations.

The local Health Instrument Division favors a reduction by a factor of at least 50 in the permissible air concentration.

There appears to be no method of measuring the amount of plutonium in the lung. Control hinges on maintenance of clean air, and of clean hands, if one can picture material being "sniffed" off the hands. At Hanford Works, assault masks are worn if the concentration in air is known to exceed 10^{-11} μg Pu/cc. Unfortunately, the readings of air contamination are after-the-fact. The wearing of masks is thus a matter of operating judgement. It is imperative that the design of all work areas be such as to maintain the air-contamination below 10^{-11} μg Pu/cc.

5. <u>Subcutaneous Implantation</u>

In animals, 1 μg Pu below the skin produces degenerative change in one year. A much smaller amount has to be considered ultimately hazardous in man. There is no convenient method of detecting such accidental implants other than by local excision. It is conceivable that a sliver of metal can enter without detection by the individual. Such injuries must be avoided by elimination of possible injury - a significant advance in protection is offered by the remote mechanical type operation. With soluble plutonium compounds there is the added risk of contributing to the skeletal load.

6. <u>Absorption through the skin surfaces</u>

Little is known of the severity of this hazard beyond the early human skin tests at Hanford. Presumably, it is adequately controlled by normal standards of skin cleanliness.

<u>Summary</u>

Present permissible limits of exposure to plutonium both for skeletal deposition and lung deposition are not well-founded, and reasons are advanced why they should be lowered. Other sites of deposition (liver or subcutaneous implant) cannot be dismissed as insignificant.

Methods of measurement of body content either lack precision (urinalysis) or are non-existent (lung).

It follows that conservative operating policy requires the application of the most effective known methods of protection, and continuous efforts to advance the protective art.

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