In-vivo monitoring of nuclear fuel workers

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Introduction

As well as being a major source of uranium, Canada possesses a sizable uranium processing industry. Several plants located in Southern Ontario fabricate fuel rods for use in the Canadian nuclear power industry and also for export.

Working conditions in these plants are specified by the Atomic Energy Control Board, and monitoring of workers by means of criticality badges and urinalysis is performed by the Radiation Protection Bureau and the Ontario Department of Health.

Urinalysis results frequently show evidence of uranium ingestion, but the relation of this to total body burden is uncertain and a direct, in-vivo method of body burden measurement is required.

Cofield (1) made such measurements using a standard whole body counter in a steel room, and Quastel et al (2) of this Bureau made similar measurements, together with extensive urinalysis and other biological measurements, on 15 uranium workers. This study confirmed that body burden could not be accurately estimated from daily urinary excretion.

Unfortunately, the use of a conventional whole body counter means that the subjects have to travel to Ottawa, a distance of 250 miles from the major uranium processing plants, and while this can be arranged for small sample populations it is obviously impractical for the plants' total exposed work force. It was therefore necessary to move the equipment to the plants, and the present study was undertaken to see whether a portable system would have sufficient sensitivity to be useful.

Theory

The usable radiations from U-Nat. are as follows:

U-238 -	none	U-235 -	185 keV (54%)
Th-234 -	63 keV (3.5%) 93 keV (4%)	Th-231 -	84 keV (10%)

Since the subjects to be monitored had long exposures to uranium it was considered reasonable to assume secular equilibrium of the ingested material, which being of refined reactor grade had a very low content of radium or radium daughters. It was therefore decided to use the low energy radiations

from the thorium daughter, for which a 20 cm diameter x 3 mm thick NaI crystal, already available, would have a high efficiency. The response of this detector to a U-Nat. phantom is shown in fig. 1 as a broad peak extending from around 50 to 90 keV. The detector has negligible sensitivity to the 185 keV gammas from U-235.

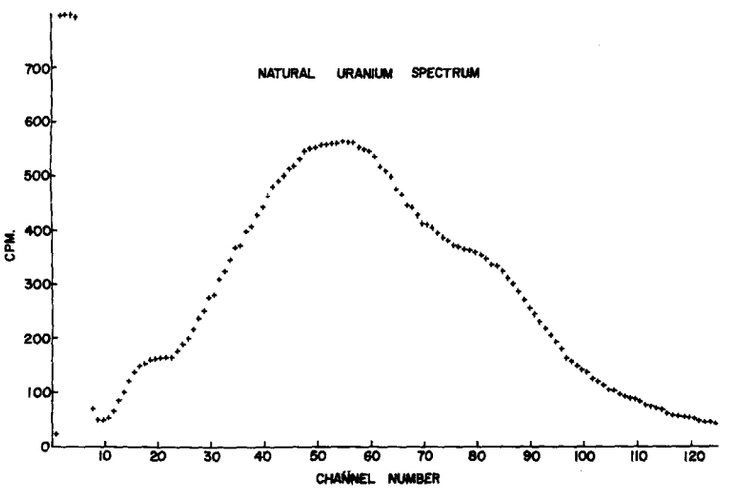

Fig. 1 Response of 20 cm x 3 mm NaI crystal to U-Nat. phantom

The phantom used for calibration purposes was a Remcal (Alderson Research Laboratories Inc.) phantom, with 9.2 g of uranium oxide dust in the lung cavities. The dust was sprayed onto adhesive coated paper tissues which were then made to adhere to the inside of four polythene bags. One bag was introduced into each of the four lung cavities and inflated with air. The basal lobes contained about four times as much uranium as the apical lobes.

The remainder of the phantom was filled with water and the net count rated observed was 3.86 cpm/mg U-Nat., which with a background of 3000 cpm gives an S^2/B ratio of 0.0049.

Using the formula of Altshuler and Pasternak (3), the minimum significant measured activity is 12.4 mg and the minimum detectable true activity is 24.8 mg, these figures being for a single 10 min. count. The crystal is also sensitive to scattered radiation from Cs-137 and K-40 to the extent of 8 cpm/nCi Cs-137 and 0.5 cpm/g K. For a normal 70 kg adult with a body burden of 5 nCi Cs-137 and containing 180 g K this would amount to a count rate of 130 cpm. Since the thin crystal is not sensitive to the primary radiation from either Cs-137 or K-40 it is not possible to measure each subject's content of these isotopes directly. The count rate from them has instead been considered part of the subject background, which is discussed further below.

Description of apparatus

The detector is a 3 mm thick by 20 cm diameter NaI (Tl activated) crystal coupled to 3 low-noise PM tubes. Each PM tube has its own H.V. supply, but the signals are collected by a common cable and fed into a single preamplifier and thence to the amplifier which is an integral part of the

Technical Measurement Corporation pulse height analyser. The date is recorded by either a TMC parallel printer or a Tally paper tape punch.

The settings of the H.V. Supplies are adjusted by placing a small uranium source on the crystal axis at a distance of about 40 cms. and switching on one H.V. supply at a time, and adjusting them so that the three spectra overlap as precisely as possible.

The detector is housed in a stainless steel cylinder which is lined with 1/8" lead and supported on a counterbalanced stand. See fig. 2.

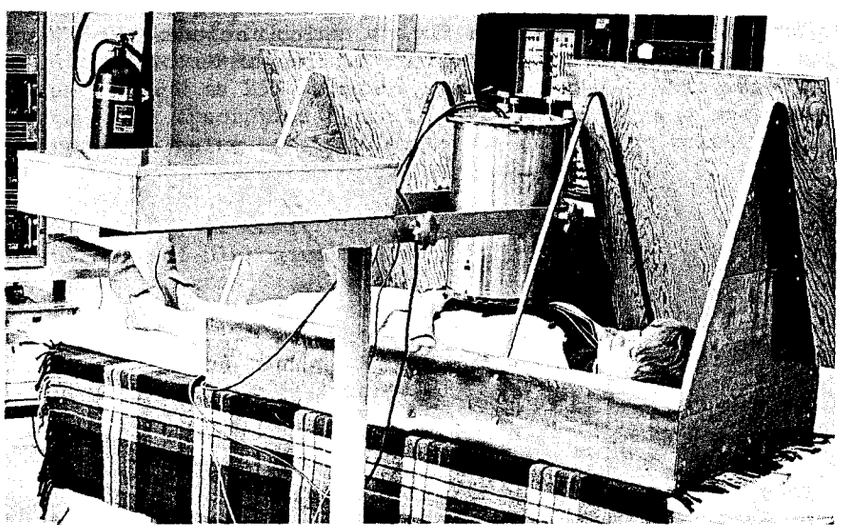

Fig. 2 Detector, counterbalanced stand and lead tent with near-side panels removed.

The shielding is based on a design by Eisenbud et al (4) and consists of 1/8" lead sheet fastened to plywood and placed over a tent shaped framework. A sheet of 1/8" lead is placed under the mattress on which the subject lies.

The effect of this shielding is to reduce the subject background in the 30-100 keV region from 9,500 cpm to 3,000 cpm.

This equipment is portable, the total weight being about 600 lbs. Two men can load it into a station wagon in about 30 mins. and on arrival at the counting location, the system can be unloaded and assembled in about an hour. A room about 10 ft square is sufficient to house the equipment and the counting bed, located away from any active area. So far no unduly high background rates have been encountered, despite the fact that the plants visited process large quantities of radioactive material.

Subject Background

Preliminary work in the laboratory indicated that the background count

on a subject inside the tent varied considerably, depending on his size and shape. This variation was much more than would be expected just from differences in Cs-137 and K-40 content. Therefore forty Radiation Protection Bureau staff members were studied and their backgrounds were measured together with their height, weight and "chest thickness", i.e. the front to back measurement of chest thickness made at inspiration. The latter measurement was chosen as the simplest one to give some index of the bulk of tissue under the detector.

A series of empirically chosen expressions combining the three anthropomorphic parameters were analysed using a least squares method, for their correlation with the observed count rate. The expressions were of the type $X = (W/H) \log C$ or $X = \exp (W/H)$ etc. and the correlation coefficients were all quite similar at around 0.84. The highest coefficient, 0.851, was obtained with the expression $X = (W/H)\sqrt{C}$ which yielded an equation for Y, the subject background; $Y = K (3517.4 + 259.46X)$ where K = correction factor to allow for differences in ambient background between our laboratory and other counting locations. A plot of Y vs X is shown in fig. 3, which includes the 95% confidence limits for a single estimate of Y given a value of X.

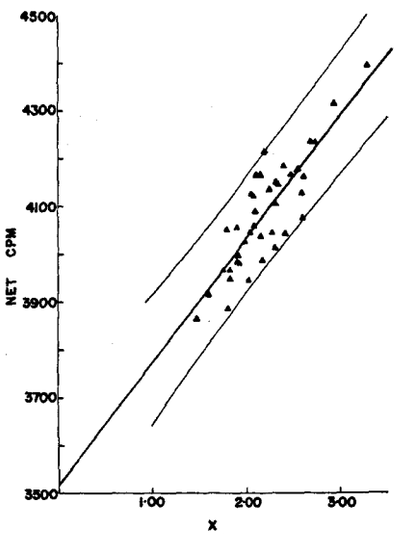

Fig. 3 Plot of observed subject background count-rate vs X where X = weight/height x$\sqrt{\text{chest thickness}}$. (The 95% confidence limits are shown)

In terms of cpm these confidence limits represent ± 120 cpm on a typical subject background of 3000 cpm (K being significantly less than one at the plant where the bulk of the subjects were counted), whereas counting error for 10 min. count = 17.3 cpm. This error is more or less equal to the count rate expected from 30 mg U-Nat., the maximum permissible body burden. This means that only burdens in excess of the maximum permissible can be confidently detected. In an attempt to improve this situation, a more detailed multiple regression test was run on the data. This analysis was carried to the point where an equation containing six terms was derived, but the correlation coefficient associated with this equation was 0.8631, which was not significantly greater than with the much simpler equation originally chosen.

Other corrections

In dealing with radiations at 100 keV or below, tissue absorption must be taken into account. On the assumption that the uranium is located in the lungs or pulmonary lymph nodes, it is necessary to make some estimate of the thickness of tissue overlying these organs. Deane (5) has measured this thickness ultrasonically and correlated it with the weight/height ratio of each subject. His formula is as follows:

$$T \text{ (thickness (mm))} = 0.071 + 0.512 \frac{W \text{ (weight (kg))}}{H \text{ (height (m))}}$$

Ramsden et al (6) did a similar study, and obtained a different formula, which included a smeasurement of the chest circumference (C)

$$T \text{ (cm)} = 15.3 \frac{W \text{ (kg)}}{H \text{ (cm)}} - 0.01\ C \text{ (cm)} - 3.55$$

We were not able to obtain an ultrasonic device and make actual measurements, and so T was calculated both ways and the mean taken, for each subject. As there were differences between the thicknesses obtained with the two formulae, in some cases as much as 25%, a mean value was taken. This was used to read off the appropriate absorption correction from fig. 4, the error associated with this procedure being estimated as ± 4%, due to the relatively small slope of fig. 4.

Newton et al (7) in their work on the measurement of plutonium in the lungs, took into consideration the self-absorption of the lung tissue itself, but for the present work, in view of the absorption curve in fig. 4 this has been ignored.

Collection of data

In the fall of 1971 a brief visit was made to a uranium processing plant in Southern Ontario. This was the first trip with the portable equipment and the main object was to see whether it could be transported and set up easily, and would be stable in operation. The time from arrival at the plant to counting the calibration standard was about 1½ hours, and frequent counting of the calibration standard indicated that the response of the system was very stable.

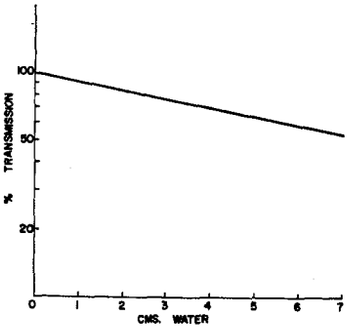

Fig. 4 Relative transmission of U-Nat. radiations in 60-90 keV region in water.

The plant management made a small room 10' x 10' available to us in the administration area of the building and the subjects arrived after a shower and change of clothing. The subjects were 3 workers who had been studied five years earlier by this laboratory (2).

The subjects were counted supine with the 8" detector in contact with the chest surface. The counting time was 10 min.

The results from this preliminary study, while showing some counts above the background of a normal subject, were calculated without the benefit of the subject background equation subsequently developed, and were therefore considered merely an indication that the system could work and that a further field trip to study a larger sample of workers would be justified.

Such a trip was undertaken some time later, and involved a larger nuclear fuel processing plant. This plant contained a large stock-pile of uranium and there were some misgivings as to what the background count would be. It turned out however to be not significantly different from that at the other plant.

A total of twenty-four subjects were counted, with results as shown in table 1, calculated on the assumption that only U-Nat., in equilibrium, was present.

Table 1
Results of Measurements in Nuclear Fuel Workers

Subject #	$X\ (\frac{W}{H}\sqrt{C})$	Estimated background c.p.m.[1]	Observed count rate c.p.m.[2]	Net count rate c.p.m.	Absorption correction*[3]	mg U-Nat.
1	2.47	3056 ± 125	3298 ± 18	242 ± 126	0.78	62 ± 25
2	1.79	2927 ± 121	3401 ± 18	474 ± 122	0.87	109 ± 24
3	2.15	2995 ± 122	3005 ± 17	11 ± 123	0.82	3 ± 24
4	1.93	2953 ± 121	2975 ± 17	22 ± 122	0.85	5 ± 24
5	1.87	2941 ± 120	2981 ± 17	40 ± 121	0.87	9 ± 24
6	2.36	3035 ± 120	3047 ± 17	12 ± 121	0.81	3 ± 24
7	2.42	3046 ± 123	3011 ± 17	-35 ± 124	0.80	-
8	2.09	2985 ± 123	3244 ± 18	259 ± 124	0.83	62 ± 25
9	1.76	2920 ± 120	3090 ± 17	170 ± 121	0.88	38 ± 24
10	1.71	2912 ± 122	3018 ± 17	106 ± 123	0.87	24 ± 24
11	2.17	2999 ± 123	3217 ± 18	218 ± 124	0.82	53 ± 25
12	1.7[illegible]	2919 ± 122	3204 ± 18	285 ± 123	0.88	64 ± 24
13	1.57	3075 ± 121	3041 ± 17	-34 ± 122	0.77	-
14	2.20	3005 ± 123	3291 ± 18	286 ± 124	0.86	66 ± 25
15	1.74	2917 ± 120	2984 ± 17	67 ± 121	0.89	15 ± 24
16	1.87	2941 ± 120	2968 ± 17	29 ± 121	0.87	6 ± 24
17	2.68	3097 ± 123	3326 ± 18	229 ± 124	0.77	59 ± 25
18	2.36	3035 ± 120	3349 ± 18	314 ± 121	0.80	78 ± 24
19	2.97	3151 ± 128	3282 ± 18	131 ± 129	0.75	35 ± 26
20	1.89	2945 ± 120	3211 ± 18	266 ± 121	0.86	62 ± 24
21	2.36	3035 ± 120	3374 ± 18	239 ± 121	0.80	84 ± 24
22	2.07	2980 ± 122	3047 ± 17	67 ± 123	0.83	16 ± 24
23	1.90	2948 ± 120	4908 ± 22	1960 ± 121	0.85	459 ± 24
24	2.07	2983 ± 123	2947 ± 17	-36 ± 124	0.83	-

[1] Errors estimated from fig. 3

[2] Counting error only

[3] Correction factor read from fig. 4 using tissue thickness calculated as described in text.

As will be noted some had very high values, but subsequent discussion with the plant health physicist revealed that these had worked in the UF_6 plant, and during the processing of this material, the thorium does not enter the gaseous phase and is left behind in the "ash". It was therefore assumed that the high readings were due, at least in part, to the inhalation of thorium 234, during the handling of this "ash", and measurements taken on some of the UF_6 workers six months later were down to much lower levels, which tended to confirm this assumption.

Discussion

The results from these field trips indicate that a significant proportion of the workers examined were contaminated with uranium or its daughters. The errors, mainly due to uncertainities in the background count estimate for each subject, were such that only lung burdens greater than 30 mg could be detected with any confidence, and in some cases the material detected could have been mainly 24 day Th-234 rather than U-Nat.

The problem of identification is in fact two-fold, first to identify the degree of enrichment of the uranium, and second, to differentiate thorium from uranium. A recently acquired dual crystal (Phoswich*) detector has been put to use in some preliminary experiments to look into these problems.

The detector is a 12.5 cm diameter, 1 mm thick NaI (Tl activated) crystal backed by a 12.5 cm diameter, 5 cm thick CsI (Tl activated) crystal, and a pulse shape discrimination circuit (Harshaw NC-25) is used to allow only those photons that are completely absorbed in the thin crystal to be recorded. The sensitivity of this detector is 1.25 cpm/mg U-Nat. (50 keV - 110 keV) for an average subject background of 600 cpm. If these figures are corrected to compensate for the smaller area of the 12.5 cm detector compared to the 20 cm detector, an S^2/B ratio of .0067 is obtained. This is better than the figure of .0049 for the 20 cm detector, but could probably be improved further by the use of a thicker front (NaI) crystal, of say, 3 mm.

The natural uranium source gives a spectrum with well-defined peaks at about 60 keV and 90 keV, and the change in relative heights of the peaks with changing enrichment was investigated, on the grounds that the Th-231 from U-235, with its gammas at around 84 keV would affect the 90 keV peak more than that at 60 keV. The peak ratio being defined as $\frac{\text{ch. 103-162}}{\text{ch. 163-215}}$ at a gain of 0.48 keV/ch.

The plot of peak ratio vs enrichment is shown in fig. 5, and there is a marked drop in the 60 keV/90 keV ratio with increasing enrichment up to about 20%. Beyond that the curve levels out but since the material used in the plants under investigation rarely exceeds 20% enrichment, this should not detract from the utility of the curve in estimating the enrichment of the uranium present.

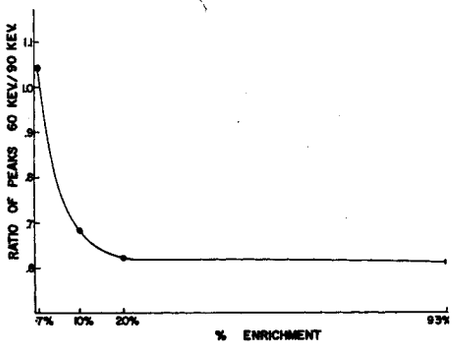

Fig. 5 Ratio of 60 keV/90 keV peaks vs enrichment

The above measurements were made with sources of high activity and so the net count rates were high. With actual subjects the net count rates would normally be much lower, and prone to the errors involved in estimating subject body background discussed already.

(* Harshaw Cehmical Co. Ltd.)

Future work with the dual crystal will include an experiment to determine the relationship between the 60 keV/90 keV peak ratio and tissue thickness, and another to explore any correlation between the body background of a normal subject in the 60 keV-90 keV region and that in the 200 keV and above region, using the 5 cm thick CsI crystal as a detector and counting all the events occurring within it. The range from 200 keV upwards would include contributions from Cs-137 and K-40 but not from the 185 keV line of U-235.

It would also be very useful if the 185 keV line of U-235 could be detected by the CsI crystal in order to distinguish between uranium and separated thorium but early results indicate that the background counts in that region are too high for sufficient sensitivity.

Conclusions

The study shows that given certain conditions, the 20 cm x 3 mm single crystal with portable shielding can detect lung burdens in the region of 30 mg U-Nat. The conditions are that only U-Nat. is present, in equilibrium with its thorium daughters, and that no separated thorium is present.

In the major uranium processing plant studied these conditions hold for a substantial portion of the personnel, and those for which they do not hold can be identified. The system can, therefore, give useful information, and is suitable for routine monitoring within the stated limits.

A dual crystal (Phoswich) system, however, offers considerable promise for refining the method to determine enrichment and to improve the subject background estimates, and hence the sensitivity. The problem of separated Th-234 will require either a much more substantial counter of the shadow-shield type, or a method of repeated counting to detect the decay of the thorium.

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