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PAPER XI

PRACTICAL DOSIMETRY IN URANIUM MINING

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## 1. INTRODUCTION

The need for dosimetry measurements in the uranium extractive industry arises in four ways:

1. to meet legislated obligations under the appropriate State or Commonwealth Territory Radioactive Substances Act and allied uranium mining code,
2. to demonstrate the adequacy and reliability of plant equipment designed to reduce any potentially hazardous operations,
3. to demonstrate the adequacy of administrative control over prescribed procedures, and
4. to make administrative decisions on duty roster etc.

In essence, these needs are met by determining:

- . External irradiation levels (whole body gamma and surface  $\beta/\gamma$  dose and dose-rates).
- . Exposure due to inhalation (U, Ra particulate activity, levels of Rn and associated daughter products).
- . Exposure due to ingestion (U, Ra in drinking water, local food supplies etc.).
- . Surface contamination levels for  $\alpha$  emitting contaminants.

Naturally the accuracy and thus the practicability of each of these determinations depends on the reason for the measurement.

Each type of measurement is now considered in turn, discussing the range in methods of detection and the associated accuracy.

## 2. EXTERNAL RADIATION

### 2.1 Whole Body Gamma Irradiation

#### 2.1.1 Gamma surveys

Ore bodies, primary ore stock piles, stored yellow cake, and tailing dumps are essentially infinite with respect to the mean free path of gamma rays. The gamma spectrum from such systems therefore contains a large percentage of low energy photons and it is essential that the dose meters have a response that is reasonably independent ( $\pm 20\%$ ) of photon energy. In the main, this means that the detector should be an ionisation chamber with an effective cut-off energy of  $\sim 50$  keV. Suitable instruments of the type are readily available.

The frequency with which gamma surveys should be made will probably be covered by the appropriate legislation; it can be expected to range from quarterly (calculated exposure ~ 75% of allowable limit) to yearly (calculated exposure ~ 50% of allowable limit).

A guide to suggested procedures for the measurement of external radiation is given in American National Standards Institute (1960).

### 2.1.2 Personnel dosimetry

Personnel dosimetry of gamma rays in Australian facilities normally involves the use of film badge packs. However the high humidity and temperatures together with the rough handling to be expected in uranium mining, indicate that a more satisfactory solution might be the use of thermoluminescence dosimeters (the quantity of light given off from a heated quantity of activated lithium fluoride is proportional to the dose).

While it is not clear which authorities would be responsible for the personnel dosimetry service in order to meet legal requirements, most State legislation is sufficiently flexible for a private company to be made 'an approved laboratory' for the purpose.

## 2.2 Surface $\beta$ Irradiation

Surface  $\beta$  dose rates from uranium ore and stocked yellow cake are generally not significant and would not be monitored routinely.

The better class of survey instruments used as  $\gamma$  monitors can, by removal of an end cap, provide an estimate of  $\beta + \gamma$  dose rate.

## 3. INHALATION

Radon daughters are normally of primary concern in uranium mines although other radioactive dusts are produced by mining operations. Radon emanates continuously from all ore bodies and needs to be routinely monitored.

### 3.1 Uranium/Radium

The distribution of a given amount of uranium ore in equilibrium with a body of air will involve equal amounts of activity of uranium-238 and its daughters e.g. radium-226. The maximum permissible concentration (mpc) values for both soluble and insoluble forms of uranium and radium vary little, one from the other. Thus air contaminated with uranium ore as a result of mining, grinding or crushing etc., will be equally hazardous with respect to uranium and radium and no attempt need be made to identify these activities separately.

Of course the radon daughter alpha activity must be allowed to decay before the long lived U and Ra activities are estimated from the particulates collected on a filter paper during an air sampling survey. A 24 hour decay period is more than ample for this.

### 3.2 Radon Daughters

The hazard associated with atmospheric levels of radon gas arises from the presence of the daughter products. This is shown in Table 1 where the daughter ratios shown as (1,  $\ell$ , m, n) refer respectively to radon-222, polonium-218 (RaA), lead-214 (RaB) and polonium-214 (RaC<sup>-</sup>). As discussed in a subsequent paper, the ratios of the various daughter products depend markedly on the origin of the radon, the age of the air mass (in terms of the half-life of radon-222), ventilation rate etc. Since the half-lives of the various daughter products are different, the  $\alpha$  activity on a filter paper, obtained during air sampling, is a complex function of initial disequilibrium ratios, duration of sampling and subsequent decay period. These relationships are discussed in the paper by Evans (1969).

For practical dosimetry, a method must be found for relating the measured  $\alpha$  activity on the filter paper back to an atmospheric concentration of radon daughters (i.e. to working levels (WL's)). Many methods have been reported.

#### 3.2.1 The Kusnetz method

This was one of the earliest methods reported (US Public Health Service 1957) for converting gross  $\alpha$  activity to WL and remains the most widely used.

A 5-10 minute air dust sample is collected at a constant flow rate on a suitable filter (5 $\mu$  pore size membrane paper) and allowed to decay for 40 minutes. The  $\alpha$  activity remaining on the paper over the next 50 minutes is counted and the average decay rate computed.

This value is converted to WL units by means of standard factors of theoretical origin.

In practice the uncertainties in this method are less than expected on theoretical grounds (~  $\pm$  10%) and it is now the most convenient method in use.

In some applications the  $\alpha$  activity is simply measured with a countrate meter after 40 - 90 minute decay. This approach is not recommended since much accuracy is lost simply as a result of counting statistics.

With this method, as with all others that demand a knowledge of volume of air sampled, the rotameter or other flow measuring device needs frequent calibration checks.

### 3.2.2 The Tsivoglou method

In this method (Tsivoglou, Ayer and Holaday 1953) the decay curve is recorded over a 90 minute period from the end of air sampling. Effective half-lives at specified decay times are determined from this curve and a set of simultaneous equations is then solved to give specific concentrations for each of the radon daughters.

Because poor counting statistics normally apply in this method, any particular result is fairly unreliable and it is generally only used in computing average weekly values from 10 or more separate determinations.

### 3.2.3 The least squares regression analysis method

In this technique (Raabe and Wrenn 1969) full use is made of the counting statistics available from sequential counting. The calculation proceeds in three parts; first, the differential equations which describe the collection and the decay during collection of the daughters are solved; then the differential equations which describe the decay of the daughters on the sample after collection is complete are solved; and lastly, a weighted least-squares regression analysis is performed to fit the observations of counts during various counting periods to the general decay equation for the sample.

Computer facilities are needed for this approach but it is the only suitable method for dealing with an atmosphere containing significant and varying amounts of  $^{222}\text{Rn}$  (radon) and  $^{220}\text{Rn}$  (thoron).

### 3.2.4 The two-filter method

Strictly speaking, this is a method for measuring the concentration of radon gas (Thomas and Le Clare 1970). If used for determining WL values, severe over-estimation would result because of the need to assume complete equilibrium between the various daughter products.

However, if run in parallel with one of the above methods it allows an estimate of the degree of disequilibrium (the so-called WL ratio) and thereby provides guidance on how much more control could be effected by increasing ventilation rates.

Briefly, air to be assayed is drawn through two filters in series which are separated by a delay volume to permit radon decay. The first filter removes the radon daughters present in the air, while the radon concentration is calculated from the decay products (formed in the delay volume) caught on the downstream filter.

### 3.2.5 Personnel radon monitors

The methods described above are most useful for determining the radon hazard at particular locations. If the hazard is not easily controlled then some form of staff roster may be necessary and the use of personnel radon monitors becomes mandatory.

In practice, the change of emphasis for personnel dosimetry relates to the method of detecting the  $\alpha$  activity on the filter paper. For station monitors, standard ZnS-photomultiplier scintillation probes are used. For personnel dosimeters, a more convenient method is to use track etch films. In this method suitably prepared 0.0007 cellulose nitrate films are placed close to the filter paper in the air sampling device. Any  $\alpha$  particle of energy between limits that are characteristic of the film material, will, on striking the film, produce bulk damage. This 'latent image' can subsequently be 'developed' with caustic soda solution and then counted optically or with a spark counter.

With this method, a personnel dosimeter, battery, air pump, filter detector, can be built into a 4 inch x 1 inch cylinder weighing approximately 2 ounces. We have had little difficulty perfecting methods for manufacture of the film or with the operation of the spark counter. We have not attempted to build up the air sampler believing that commercial units will be available, at least from the USA.

Dosimeters of this class will be suitable for integrating the WL exposure to miners over periods of a week or more (Auxier et al. 1971).

### 3.3 Unattached RaA

As explained in Mr. Cook's paper, the significance of unattached RaA daughters has been questioned by the ICRP Task Group on Lung Dynamics. Nevertheless, the current ICRP recommendation specifically includes the percentage of unattached RaA in their derivation of mpc levels for radon gas. Unless the Mining Code deliberately supersedes these recommendations, the unattached RaA percentage will need to be measured, because the probably conservative value of 10%, as used by the ICRP, would otherwise have to be assumed.

There are two basic approaches for the measurement of unattached RaA. In the first (Chamberlain and Dyson 1956) the windpipe is simulated by a tube 15 cm long, 5.7 cm circumference, with a smooth paper insert. Air is drawn through the tube at 16 l/min for about 5 minutes; the paper is removed and

the deposited  $\alpha$  activity determined. It is assumed that only RaA is unattached and that plate-out effectively removes any unattached RaA that is present.

For the second method, much care is taken to design a diffusion battery for the complete removal of unattached products and the RaA component is determined by spectroscopically comparing two parallel sampling systems, one of which is preceded by the diffusion battery (Duggan and Howell 1969).

The first method will invariably produce the lower result but the question as to which is more correct depends solely on the rationalisation behind the recommendation on the significance of unattached RaA.

#### 4. INGESTION HAZARD

The determination of any ingestion hazard would normally be the responsibility of the appropriate Health Department but the techniques involved would be no different to those that the mining companies would need in order to demonstrate compliance with criteria for effluent release. The determination of U, Cu, Pu, As, etc. involves well developed chemical methods and only the determination of radium-226 will be discussed here.

##### 4.1 Radium-226 Determination

Radium-226 is determined by the emanation method. This procedure is capable of measuring with high accuracy quantities of radium-226 as low as  $2.10^{-14}$  g; it discriminates against other radium isotopes, as well as all other radioactive elements which may be present.

Radium-226 is determined through its daughter, the noble gas radon-222. The daughter is purged from a solution of its parent with an inactive gas and collected on activated charcoal at  $-80^{\circ}\text{C}$ . The charcoal is then heated and the radon transferred to a zinc sulphide coated counting vessel where after 3 hours during which radioactive equilibrium between radon and its daughters has been established it is counted by scintillation techniques.

##### 4.1.1 Preparation of samples

All samples are taken into solution by various means or an acid extraction is carried out on them; they are then made up to 200-400 ml with distilled water, placed into a special flask, the residual radon is purged out with an inactive gas and the flask sealed for 1-30 days to allow the daughter radon-222 to establish equilibrium with its parent radium-226. The methods used to bring the samples into solution for radium-226 analysis by the emanation technique vary with the nature of the sample.

Water samples: Water samples are collected in polythene bottles and acidified with 4 ml concentrated  $\text{HNO}_3/\ell$ . The samples are then filtered through a  $0.47 \mu$  filter and a separate analysis is carried out on the water and the suspended sediment. The suspended sediment is taken into solution by a nitric acid/perchloric acid digestion.

Organic samples: These samples are first dried, ashed at  $450^{\circ}\text{C}$ , and then the ash is digested with concentrated nitric acid/perchloric acid, evaporated to near dryness and taken up in distilled water.

Soils and sediments: These samples are dried, ashed at  $450^{\circ}\text{C}$  and then digested for one hour in 2M  $\text{HNO}_3$  and the resulting solution used for radium-226 analysis.

#### 5. SURFACE CONTAMINATION

The question of surface contamination was fully discussed in an earlier paper. To reiterate, no radiological hazard from surface contamination is expected to exist but there is certainly a nuisance value with respect to counting laboratories etc. In practice, the greatest source of yellow cake contamination will be at the drum packing end. Suffice to say that manual stirring of uncovered drums which was acceptable in the past would no longer be tolerated.

#### 6. SUMMARY

While there are many dosimetry measurements that must be made in the uranium extractive industry, all involve well established techniques and employ commercially available instrumentation.

The instruments themselves, though 'ruggedised', are still essentially laboratory equipment and must be treated accordingly. In planning a mining operation, it is essential to provide adequate laboratory space that is both airconditioned and free of airborne contamination, as well as adequate staff so that all equipment can be calibrated and standardised on a routine basis.

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TABLE 1

INHALATION HAZARD OF NON-EQUILIBRIUM

RADON-DAUGHTER MIXTURES

Equilibrium Ratios	% of Equilibrium Dose
1, 1, 1, 1.	100
1, 1, 0.90, 0.80	87.5
1, 0.90, 0.80, 0.70	77.4
1, 0.85, 0.65, 0.45	59.8
1, 0.75, 0.50, 0.30	45.3
1, 0.75, 0.40, 0.15	34.3
1, 0.50, 0.25, 0.10	21.9
1, 0.35, 0.15, 0.05	13.4
1, 0.25, 0.10, 0.05	9.6
1, 0.15, 0.08, 0.03	6.9