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LOWER PLENTY ROAD  
YALLAMBIE VICTORIA 3085  
TELEPHONE: 433 2211

EFFICIENCY CALIBRATION OF LOW ENERGY PHOTON SPECTROMETERS

P A Burns, P N Johnston and J R Moroney

INTRODUCTION

A low energy photon spectrometer (LEPS) with Ge(HP) detector (Ortec Model No. 1013 200 mm<sup>2</sup>, 7 mm thick, 230 eV FWHM at 5.9 keV) is being used in metrological studies of the heavy elements (Burns et al. (1983a,b); Johnston et al. (1983a,b)). For immediate purposes, an efficiency calibration of high accuracy is required for the energy range of 8 to 25 keV.

Two complementary approaches are being employed - one experimental and the other theoretical. Radionuclide calibration sources were used for direct and independent measurement of the efficiency of the spectrometer at eight energies covering the range of interest, Table 1. A computer model was constructed, simulating photon transport between the source and detector, to calculate spectrometer efficiency and to provide insight into the relative importance of the various parameters and processes involved. When fitted to the set of independent measurements, the calculated efficiency should afford a sound basis for combining those results to optimum overall accuracy and for interpolation at energies between them.

EXPERIMENTAL CALIBRATION

The coincidence method (Burns et al. (1983a)) was applied to determine the full energy peak (FEP) efficiency of the LEPS, using the radionuclides <sup>57</sup>Co, <sup>65</sup>Zn, <sup>75</sup>Se, <sup>85</sup>Sr, <sup>88</sup>Y, <sup>99m</sup>Tc, <sup>109</sup>Cd and <sup>113</sup>Sn prepared as calibration sources (Moroney et al. 1983a, b)). The number of K xrays being detected per electron capture (EC) to a known state was used for the first five radionuclides, while the number of K xrays detected per K-shell internal conversion was used for the other three. Some details of the experimental methods are summarised in Table 1., together with the approximate uncertainty expected in each calibration point.

Coincidence gated K xray spectra were obtained for a set of five sources for each radionuclide. Statistical uncertainties were generally less than 0.5% (2σ) for each source. Analysis of the spectra is well advanced but the corrections for interference in the coincidence gate remain to be made.

TABLE 1. CALIBRATION OF LEPS BY DIRECT MEASUREMENT  
USING THE COINCIDENCE METHOD

Nuclide	Calibration energy $K_{\alpha 12}$ xrays	Coincidence gate		Expected order of uncertainty ( $^{\circ}/\cdot$ ) in calibration point ( $2\sigma$ )
		Radiation	Detector*	
$^{57}\text{Co}$	6.4 keV	$\gamma$ , 136 keV	GE(HP)	3
$^{65}\text{Zn}$	8.0 "	$\gamma$ , 1115 "	NaI(Tl)	2
$^{75}\text{Se}$	10.5 "	$\gamma$ , 401 "	GE(HP)	4
$^{85}\text{Sr}$	13.4 "	$\gamma$ , 514 "	NaI(Tl)	1
$^{88}\text{Y}$	14.1 "	$\gamma$ , 1836 "	NaI(Tl)	1
$^{99\text{m}}\text{Tc}$	18.3 "	$\text{Ce}_K$ , 119 "	SiSBD	3
$^{109}\text{Cd}$	22.1 "	$\text{Ce}_K$ , 61 "	SiSBD	2
$^{113}\text{Sn}$	24.2 "	$\text{Ce}_K$ , 363 "	SiSBD	1

\* Ge(HP): hyper-pure germanium. NaI(Tl): Thallium-activated sodium iodide.  
SiSBD: silicon surface barrier detector.

#### COMPUTER MODELLING

A computer code implementing Monte-Carlo methods, was written to model the physical processes, and the source-detector geometry, which determine the FEP efficiency of the LEPS. Several simplifying assumptions were justified, including

- photons are emitted isotropically from the source with negligible interaction within the source
- the materials between source and detector act only as narrow-beam attenuators - specifically, there is no elastic or inelastic scattering from them into the Ge crystal

- the orientation of the axes of the GE crystal can be ignored; that is, the detector can be treated as an amorphous block of Ge
- following photon interaction in the crystal, there is immediate absorption of any secondary electrons and any Ge xrays from above the K shell.

Physical dimensions were taken from the manufacturers specifications, supplemented by radiographs of the detector. The Ge deadlayer on the crystal was determined by a new technique using the As K xrays from  $^{75}\text{Se}$ .

Comparison of the preliminary experimental efficiencies with those from the modelling shows encouraging agreement.

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FLUORESCENCE YIELD AND COSTER-KRONIG TRANSITION PROBABILITY FOR THE  
L<sub>2</sub> SUBSHELL OF Ra, Th, Pa, U, Np and Pu.

P N Johnston, J R Moroney and P A Burns

INTRODUCTION

The investigation being undertaken to make preliminary determination of the Coster-Kronig transition probability ( $f_{23}$ ) and the fluorescence yield ( $\omega_2$ ), of the L<sub>2</sub> subshell of Ra, Th, Pa, U, Np and Pu, is proceeding. The investigation and its methodology were previously outlined (Burns et al. (1983a); Johnston et al. (1983)).

L-SUBSHELL VACANCY DISTRIBUTION

The distributions of primary vacancies were calculated for all but one of the systems under study. The outstanding data are for the delayed 86.5 keV state in <sup>233</sup>Pa following <sup>237</sup>Np decay; before that distribution can be established adequately it is necessary to re-determine the intensities of the three gamma-rays involved in the de-excitation of the 86.5 keV state. Those measurements have not yet been made.

(ALPHA, L XRAY) COINCIDENCE SPECTROSCOPY

L xray spectra, gated by alpha-particle emission, were taken for <sup>230</sup>Th, <sup>234</sup>U, <sup>238</sup>Pu and <sup>244</sup>Cm and are discussed elsewhere (Burns et al. (1983b)). L xray spectra, delayed after alpha-particle emission, were taken for <sup>237</sup>Np and <sup>241</sup>Am. The six sets of spectra are being analysed preparatory to calculation of  $f_{23}$  and  $\omega_2$  on completion of calibration of the spectrometer (Burns et al. (1983c)).

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PREPARATION OF ULTRA-THIN SOURCES AND CALIBRATION STANDARDS FOR  
COINCIDENT STUDIES, AT HIGH RESOLUTION, OF RADIATIONS EMITTED BY  
THE HEAVY ELEMENTS

J R Moroney, P A Burns and P N Johnston

The techniques for source preparation described previously (Moroney et al. (1983)) were used to prepare sources of  $^{230}\text{Th}$ ,  $^{234}\text{U}$ ,  $^{237}\text{Np}$ ,  $^{238}\text{Pu}$ ,  $^{241}\text{Am}$  and  $^{244}\text{Cm}$  for studies in atomic inner-shell physics (Johnston et al. (1983a,b)) and of  $^{57}\text{Co}$ ,  $^{65}\text{Zn}$ ,  $^{75}\text{Se}$ ,  $^{85}\text{Sr}$ ,  $^{88}\text{Y}$ ,  $^{99\text{m}}\text{Tc}$ ,  $^{109}\text{Cd}$ ,  $^{113}\text{Sn}$  and  $^{125}\text{I}$  for spectrometer calibration (Burns et al. (1983a,b)). A set of five sources was made with each radionuclide.

The preferred preparation procedure was electrodeposition, which was applied successfully for all but two of the radionuclides. The exceptions were  $^{99\text{m}}\text{Tc}$  and  $^{125}\text{I}$ , sources of which were prepared by drop evaporation. A fine ring of grease, 7.0 mm inside diameter, defined the source geometry and contained a drop of the solution of the radionuclide; this was spread more-or-less uniformly within the ring by addition of a dispersing agent (Catanac) and then evaporated. Although less satisfactory than if electrodeposited, the two sets of sources proved to be adequate for the purpose.

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MEASUREMENT OF ABSOLUTE EMISSION RATES OF L XRAYs

P A Burns, P N Johnston and J R Moroney

L xray spectra of  $^{230}\text{Th}$ ,  $^{234}\text{U}$ ,  $^{237}\text{Np}$ ,  $^{238}\text{Pu}$ ,  $^{241}\text{Am}$  and  $^{244}\text{Cm}$ , gated by alpha-particle emission, were taken with the experimental apparatus previously described (Burns et al. (1983a)). The measurements for each radionuclide were made on a set of five sources (Moroney et al. (1983a)) with L xray counting statistics of better than 0.2% ( $2\sigma$ ) for each source. Source-detector geometry was closely controlled throughout the measurements - to within 70  $\mu\text{m}$  (Moroney et al. (1983b)) - and source-to-source variation, in the gated L xray spectrum for a radionuclide, was held to less than 1%.

Absolute L xray emission rates will be derived from the sets of spectra when the detector calibration is completed (Burns et al. (1983b)) and appropriate corrections for summing and other coincidence effects are made. It is anticipated that an overall accuracy approaching 1% ( $2\sigma$ ) should be attainable.

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## NON-IONISING ELECTROMAGNETIC RADIATIONS

SPECTRORADIOMETRIC MEASUREMENT OF SOLAR ULTRAVIOLET RADIATION

P H Gies, C R Roy and G Elliott

INTRODUCTION

The aim of the project is to determine accurately the solar ultraviolet (UV) irradiance at the earth's surface over the wavelength range 250 to 400 nm.

The ultraviolet region can be divided into three sections, UV-A (400 to 315 nm), UV-B (315 to 280 nm) and UV-C (280 to 100 nm). Unattenuated solar radiation contains UV-A, -B and -C, but the lower wavelengths are absorbed by oxygen and ozone in the atmosphere, so that wavelengths below about 300 nm are not usually detected at the earth's surface. The exact cutoff, however, will depend upon the season, time of day and atmospheric ozone concentration, and results from the combined effect of the rapid falloff of intensity of the incident solar radiation with decreasing wavelength and the large change of the ultraviolet absorption coefficient of ozone with wavelength.

The biological effectiveness of ultraviolet radiation varies by approximately four orders of magnitude within the UV-B region. Hence there is a need for accurate measurement of the spectral distribution of solar ultraviolet radiation.

Measurements of the spectral distribution of Australian daylight have been reported by Dixon<sup>(1)</sup> for a limited time span while Paltridge and Barton<sup>(2)</sup> have reported results from a network of erythemal meters but without providing information on the spectral distribution. The measurement period needs to be extended to a year or more so that data on the daily and seasonal variations of solar ultraviolet radiation can then be correlated with atmospheric ozone concentrations and compared with northern hemisphere results and model predictions.

METHOD

Measurements of both direct and scattered solar ultraviolet irradiance at ground level were taken using a micro-computer controlled Optronics 740A spectroradiometer, primarily to test its suitability as a measuring instrument. Atmospheric conditions during the measurement period varied from zero cloud cover to almost total cover. Interpretation of the data collected during days of non-zero cloud cover was made difficult by the variations in

light levels that resulted. Until this light level variation can be monitored in some way, it has been decided to avoid this problem by taking solar spectral measurements only on clear days. The spectroradiometer is calibrated immediately before and after solar spectral measurements against a tungsten halogen standard lamp traceable to the U.S. National Bureau of Standards.

## RESULTS

The initial phase of this programme has been completed, and analysis of these results has given some insight into future requirements. A typical solar scan is shown in Fig. 1. The calculated noise equivalent spectral irradiance is also shown for comparison purposes. The discontinuities at 275 nm are caused by a filter change at that wavelength.

Evaluation of the results reveals that while the data obtained in the UV-A region is in agreement with previously published results, spectral irradiances at the lower wavelength end of the UV-B region are affected by the relatively high levels of stray light inherent in the present monochromator. Because this problem is most severe wherever there is a rapid change in spectral irradiance, the critical region in solar spectral distributions is therefore around 300 nm, where the spectral irradiance changes by at least five orders of magnitude over a wavelength interval of a few nanometres. This rapid change is illustrated by the "true spectrum"<sup>(3)</sup> in Fig. 1. The problem can be largely overcome by the incorporation of a double grating monochromator in the measurement system. Fig. 1 shows a calculated spectral irradiance curve obtainable with a double grating monochromator.

## CONCLUSION

If the project is to continue, then changes to the present measurement system and procedures will be necessary. Both the accuracy and the frequency of measurement needs to be improved. The reduction of stray light levels in the UV-B region required to improve the accuracy of the measurements can be achieved with the substitution of a double grating monochromator, with its inherently lower stray light levels.

A data collection system dedicated to the measurement of solar ultraviolet radiation will be permanently established. The minimum desirable measurement frequency is two or three times a week, weather permitting. Maintaining this measurement frequency with the present portable equipment is logistically difficult, in addition to being detrimental to the equipment. A permanent, dedicated system will enable measurements to be made daily.

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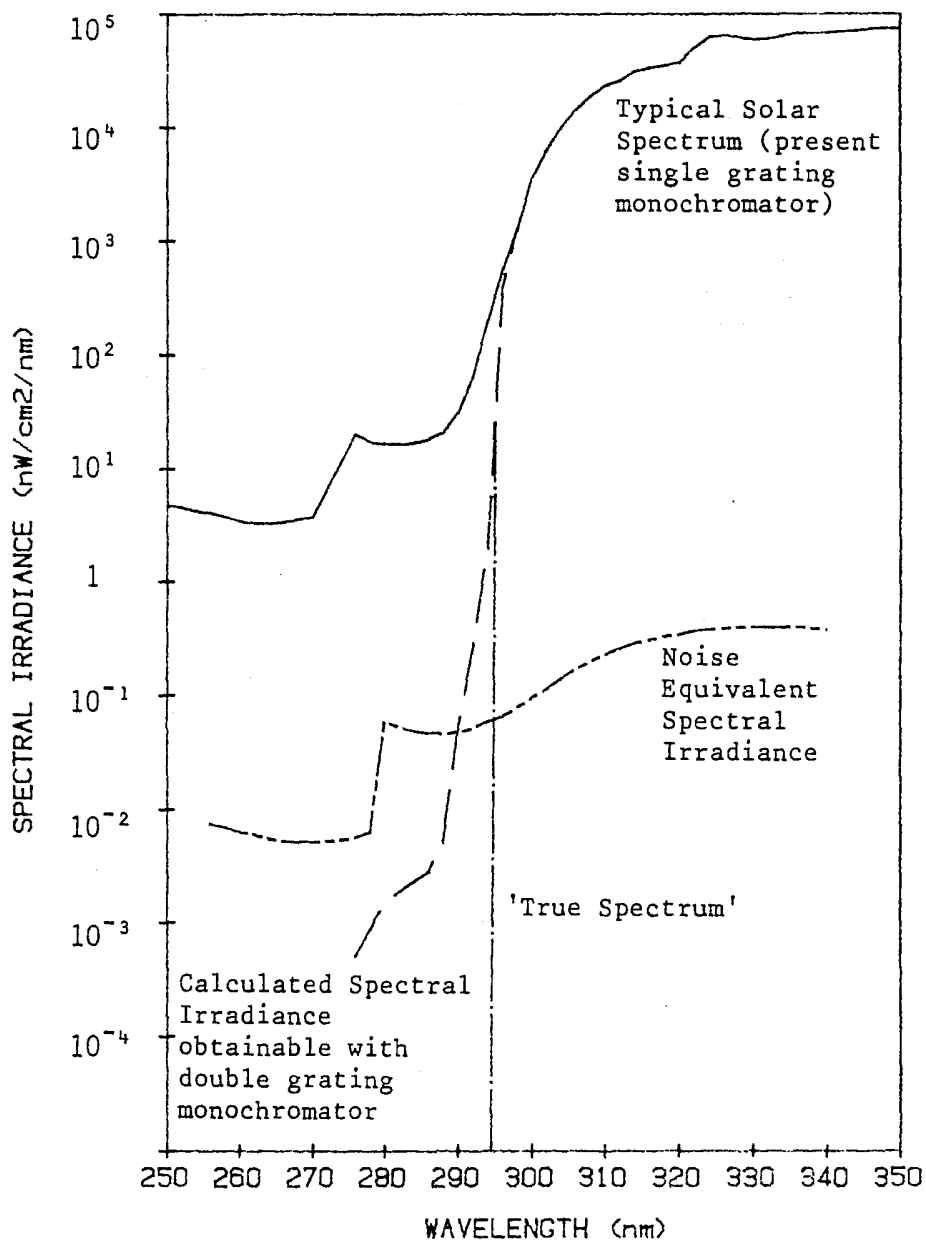


Figure 1. Solar spectral irradiance versus wavelength.

ULTRAVIOLET LASER INDUCED REACTIONS IN ORGANIC  
AND INORGANIC SYSTEMS

C R Roy, V Delpizzo, D W Tomlinson and P H Gies

INTRODUCTION

The primary aim of the project is to study the fluorescence from short-lived intermediates following the absorption of a pulse(s) of ultraviolet (UV) radiation. However steady-state experiments are planned in which final products will be analysed using standard techniques (e.g. chromatography, spectrophotometry etc.).

1. DETECTION SYSTEM

A computerised detection and data acquisition system is to be used for both steady-state and pulse experiments. This system, in its present state, is shown schematically in Figure 1.

Current status:

- (i) steady-state - the system is complete with only final testing of the software, to read the status and wavelength of the monochromator, yet to be done.
- (ii) pulse - for pulse experiments it was necessary to construct a memory board for the transient digitizer. A STD Bus is now being developed to read data from the digitizer memory (10 bit) and to convert it to 7 bit ASCII co-ordinates for subsequent display and/or plotting.

Some further development work is required on the selection and wiring of photomultiplier tubes (PMTs) for fast (nanosecond) detection. A new base circuit with a hard-wired PMT has been completed and is being tested.

2. LASER SOURCE

An MSNW Exci-lite XL-401 laser was recently acquired. This laser should produce an ultraviolet output of 5-10 mJ per pulse at repetition rates up to 100 pulses per second. The laser is designed to operate on the rare gas halide excimers and on many other laser gases.

The laser operates successfully as a N<sub>2</sub> laser (337 nm) with mixtures of N<sub>2</sub> and He. However in tests with F<sub>2</sub>/Kr/He, KrF (249 nm) pulses are obtained for only several minutes (instead of 2-3 hours). The problem is one of either gas

purity or of incomplete fluorine passivation of the internal surfaces of the laser. This problem should be solvable in the short term.

Initial photochemistry studies will be on previously studied systems where it is known that fluorescence is detectable with normal techniques. This will permit optimisation of the detection and data acquisition system.

The laser, to be useful must be capable of generating of the order of  $10^{-5}$  M of transient in the monitored volume. In general it will be necessary to store and average many waveforms. This technique still needs to be developed.

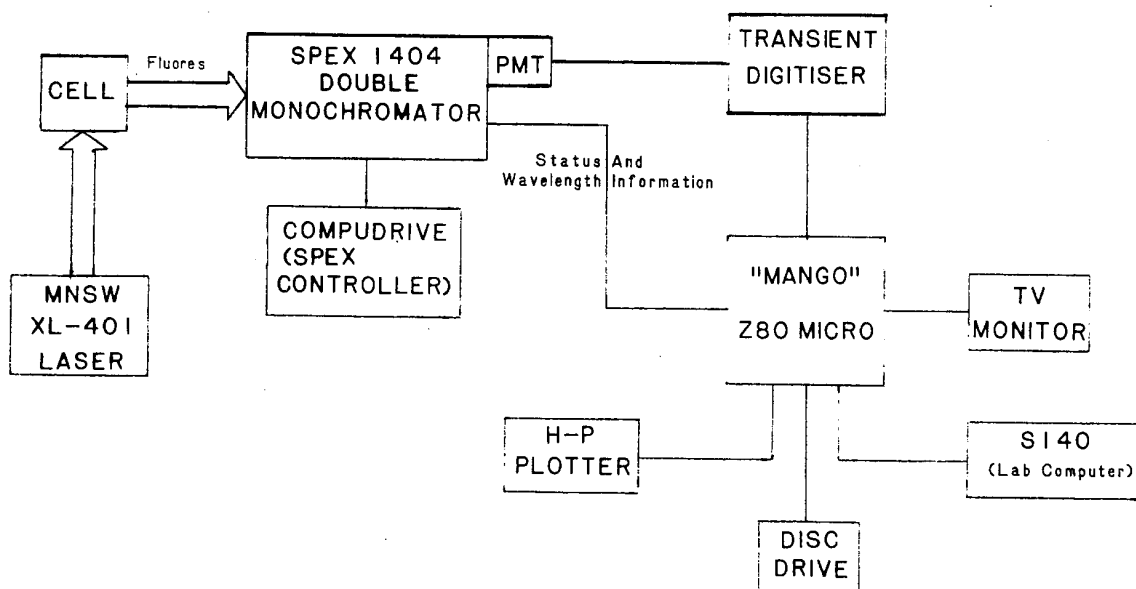


Figure 1. Schematic of Fluorescence Detection and Storage System.

THE PHOTOBIO SYNTHESIS OF VITAMIN D3 IN THALASSAEMIC CHILDREN

Colin R Roy, Peter H Gies

John A Eisman, Department of Medicine,  
University of Melbourne, Repatriation General Hospital

Rae N Mathews, Physician-in-Charge,  
Thalassaemic Clinic, Royal Children's Hospital

INTRODUCTION

The thalassaemias are a group of diseases characterised by the deficient production of one or more of the globin (protein) chains of the haemoglobin molecule. The globin chains are of four different types ( $\alpha$ ,  $\beta$ ,  $\delta$ , and  $\gamma$ ) and aggregate in groups of four (tetramers)  $\alpha_2\beta_2$ ,  $\alpha_2\gamma_2$  and  $\alpha_2\delta_2$ . In thalassaemia there is a genetically determined deficiency in the production of either the alpha or beta chain of the normal adult haemoglobin ( $\alpha_2\beta_2$ ). The resulting tetramers take up oxygen normally but release it very poorly and hence cannot substitute for normal haemoglobin.

Management of thalassaemia (major) is with blood transfusion, usually every 4-6 weeks. However, with each bottle of blood transfused 200 mg of iron is added to the body which cannot be excreted. In order to minimise iron deposition in tissues a chelating agent, desferrioxamine, is given by daily intramuscular injection.

A common clinical problem in children with thalassaemia is osteoporosis with liability to fractures. A possible cause of osteoporosis in a growing child is chronic calcium and phosphate deficiency. In this regard the abnormally low circulating levels of 25-hydroxyvitamin D (25-OH D) which have been found in thalassaemic children, particularly in winter (1,2), indicate that abnormal vitamin D metabolism is an important factor.

Since skin pigmentation is a factor in the photobiosynthesis of vitamin D3 in man it is possible that the increased skin pigmentation induced by iron overload in thalassaemics may be an etiologic factor in the low 25-OH D levels. The photobiosynthesis and 25-hydroxylation of vitamin D3 in thalassaemic children with varying degrees of pigmentation will be compared with that in their normal siblings.

## EXPERIMENTAL DETAILS

### (a) Clinical-Based Studies

Using the technique of 'in vivo' exposure to ultraviolet radiation it will be possible to compare the vitamin D<sub>3</sub> synthetic ability of thalassaemic children with that of their sex-matched siblings. By appropriate choice of control siblings it should also be possible to have a reasonable age-match of the affected and control siblings. The thalassaemic and normal siblings will be exposed to a controlled amount of ultraviolet radiation; plasma vitamin D<sub>3</sub> and 25-OH D levels will be measured before and at 12, 24, 48, 72 and 168 hours following exposure.

It is planned to deliver the radiation from a commercial sunbed with the normal UV-A tubes replaced with UV-B (280-315 nm) tubes. The spectral irradiance will be determined with a calibrated Optronics 740A spectroradiometer.

Another important section of the proposed study depends upon the rapid lightening of skin pigmentation seen in young thalassaemic children shortly after starting the desferrioxamine programme. The vitamin D and 25-OH D responses of these children to UV radiation will be examined immediately prior to and at 6-8 weeks after initiation of chelation therapy.

One requirement of the subjects is that they avoid prolonged exposure to sunlight for about six weeks prior to and during the course of study. It is hoped that arrangements can be made to commence the clinical aspect of the study during autumn 1984.

### (b) Analytical Details

Established techniques for the accurate determination of vitamin D and its metabolites from a single lipid extract of human plasma or serum requires preparative chromatography for separation and initial purification of vitamin D and its metabolites before final assay by high-performance liquid chromatography with UV detection. Adams et al. (3, 4) have described a rapid batch-elution preparative chromatographic technique using disposable silica cartridges to achieve the separation and initial purification. This technique has been reproduced in our laboratory studies and is now being modified to suit, more exactly, the needs of the present project.

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SHIELDING OF 'SEWING MACHINE' DIELECTRIC WELDERS

M J Bangay and K H Joyner

The use of radio frequency (RF) dielectric welders in the plastics industry has produced types of welders that are difficult to shield. A survey of unshielded welders has revealed that operators are exposed to excessive RF electromagnetic fields.

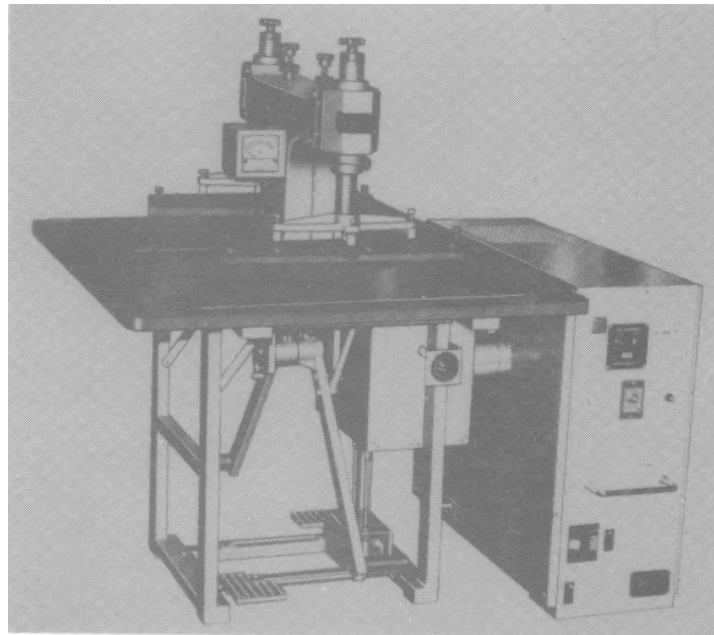


Figure 1. "Sewing Machine" Dielectric Welder (Bar Welder).

A particular type of welder in this category is the sewing machine or bar welder (fig. 1). This welder is used for welding long seams of PVC sheet as used in pool liners, water beds and tarpaulins.

The shielding of more conventional RF welding units is achieved by completely enclosing the weld die and object being welded in a continuous earth shield. Because of the large areas of PVC being processed with the bar welder this technique cannot be used.

A three kilowatt RF bar welder has been loaned to the Laboratory so that effective shielding measures can be developed. However there are certain constraints faced by trying to shield this kind of welder. They are as follows:-

1. The operator must have a clear view of the surfaces being welded (enabling proper setting up).
2. The shield must allow sufficient free area on the work table so that the operator can position the PVC sheet correctly during a weld sequence.
3. The shield must allow sufficient clearance around its edge so that large PVC sheets can be accommodated.

The object of shielding the "sewing machine" dielectric welder is to reduce the RF radiation to below that recommended by the Draft Australian Standard.<sup>2</sup> At the same time the shield should not be expensive, limit the speed of operation or the size of material being welded.

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LASER RADIOMETRY - A STUDY OF THE MECHANISM OF  
PYROELECTRIC SIGNAL GENERATION IN POLYVINYLIDENE FLUORIDE

W A Cornelius

INTRODUCTION

An accurate model of the pyroelectric response mechanism in the material polyvinylidene fluoride (PVDF) is required for accurate laser radiometry where PVDF detectors are employed. Accordingly, an analog computer previously designed and constructed at A.R.L. (1) (2) was used for further investigations of pyroelectricity in PVDF.

RESULTS AND DISCUSSION

The theoretical pyroelectric frequency response and waveform derived using the analog computer to generate the primary pyroelectric signal component [see refs. (1), (2)] were found to be significantly different from the frequency response and waveform obtained from a real detector. The possibility that these anomalies may be accounted for by a large inhomogeneity in polarisation magnitude through the thickness of a PVDF detector was investigated by constructing a detector that could be illuminated on either face of a poled PVDF layer.

Each side of a PVDF film was provided with a vacuum deposited gold/palladium electrode. The detector was completed by employing water as a transparent heat sink on each side of the coated PVDF layer. The water on each side of the PVDF was enclosed within a perspex window containment vessel. Silicon grease was used to complete the seal.

The response waveforms obtained by illuminating alternate sides of the PVDF/water detector were found to be identical. These waveforms were not significantly different from the waveforms obtained from the detectors described in a previous report (2).

The results obtained with the PVDF/water detector led to the following conclusions:

1. The pyroelectric signal from PVDF is not solely derived from a rapid relaxation process where the signal is proportional to the time derivative of the temperature.

2. Either;
- (a) the PVDF is uniformly poled, or
  - (b) the PVDF is not uniformly poled but the poling distribution is centrally symmetric.

Alternative 2(b) was subsequently ruled out by further modelling with the analog computer. It was also shown by further experiment that vacuum deposition of the gold/palladium layer does not significantly depolarise the PVDF nor does the gold/palladium layer act as a significant heat reflector.

It became apparent that the pyroelectric response of PVDF could be readily explained by assuming that a pyroelectric signal component results from long relaxation time processes occurring within the PVDF. These processes may be identified with  $\alpha_c$  dielectric relaxation phenomena as described by Broadhurst and Davis (3) and/or temperature dependent changes in crystallinity as postulated by Kepler and Anderson (4). In either case, it is assumed that the mean relaxation time for these processes is very much longer than the thermal cycle period arising from chopped optical input and that the resulting pyroelectric signal component due to long relaxation time processes ( $I_2$ ) will be proportional to the instantaneous temperature variation ( $\Delta T$ ) of the PVDF from the mean.

The analog computer circuitry was modified to conform to this theory (i.e. analog temperature was summed from the PVDF analog transmission line). Figure 1 shows the frequency response curves predicted by the analog computer for uniform polarization distribution in comparison to the frequency response of an actual 16  $\mu\text{m}$  PVDF detector. These data were obtained by measuring the r.m.s. a.c. voltage from the signal preamplifier. The input stimulus was square wave for all data shown. Curve 1 shows the analog computer prediction for the primary pyroelectric effect alone: i.e.  $I_1 \propto \frac{dT}{dt}$  where  $I_1$  is the fast relaxation pyroelectric current and  $\frac{dT}{dt}$  is the instantaneous time derivative of the temperature  $T$ . Curve 2 shows the analog computer result for the long relaxation pyroelectric effect alone: (i.e.  $I_2 \propto \Delta T$ ). Curve 3 shows the sum of the two curves 1 and 2 (i.e.  $I_1 + I_2$ ). Curve 4 is the frequency response measured in an actual detector. The vertical response scale in figure 1 is linear. Error bars are not shown for these data. The maximum random error in the data occurs for curve 4 and arises chiefly from room temperature thermal noise and some piezoelectric signal due to acoustic noise. The noise error in curve 4 is estimated not to exceed  $\pm 3\%$  of the values shown.

It is not strictly valid to add curves 1 and 2 to obtain an accurate frequency response prediction (curve 3) because the signals should be added in their correct phase relationship. However, it is estimated that the resultant error will be small for frequencies in excess of 100 Hz.

The results obtained are in very good agreement with the previously stated hypothesis. Individual phase correlated waveforms (not shown) obtained at specific frequencies also support the conclusion that the additional pyroelectric signal from PVDF arises from a process with long relaxation time. It is also concluded from these results that the decay time for the long relaxation process is in excess of 5 millisecond. There is some evidence in the waveforms obtained to indicate that the fast relaxation process has a decay time of about 100 nanosecond. The latter observation is in agreement with the work of Kepler and Anderson (4) who used a 200 ps pulsed laser to investigate the pyroelectric response of PVDF.

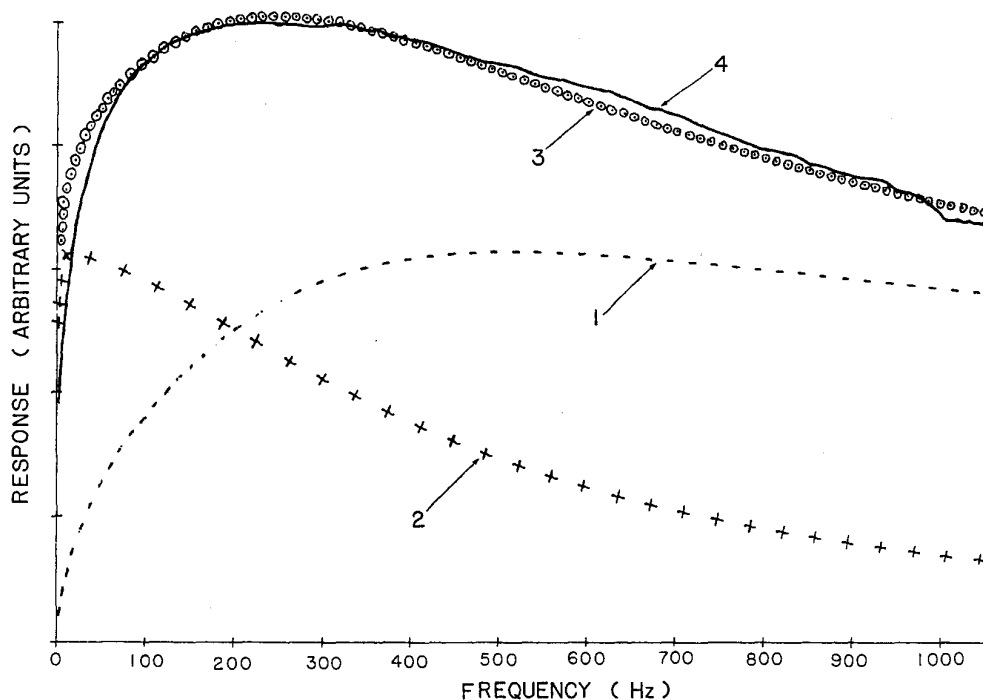


Figure 1. Predicted and measured frequency response curves for 16 $\mu$ m PVDF detector - (1) predicted primary pyroelectric effect, (2) predicted long relaxation pyroelectric effect, (3) sum of (1) plus (2) and (4) measured response in actual detector.

## CONCLUSION

These investigations led to a modified analog model of pyroelectricity in PVDF that accounts for all of the observed response characteristics of PVDF detectors within the frequency range of interest (20 Hz to 1 kHz).

The pyroelectric response of PVDF has been shown to consist of two major components; (i) a fast relaxation signal with a decay time of 100 nanosecond or less and (ii) a signal arising from relaxation processes with a decay time in excess of 5 millisecond.

All experimental evidence obtained indicates that the polarisation distribution through the thickness of a PVDF layer (16  $\mu\text{m}$  thick) is uniform.

In conclusion; it would appear that PVDF is eminently well suited to the purpose of laser radiometry because the component of signal arising from long relaxation time processes will help to reduce signal phase shift and waveform inequivalence between the calibration signal from the nickel heater layer and the optical signal deposited in the gold black [see ref. (2) for construction details of detector].

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ENVIRONMENTAL RADIOCHEMISTRY AND RADIATION MONITORING

THE SPECIATION OF RADIONUCLIDES IN SEDIMENTS AND SOILS

G A Williams and M B Cooper

A knowledge of the concentration levels of naturally-occurring radionuclides in the environment surrounding uranium ore deposits is of importance in any assessment of the environmental impact of uranium mining and milling operations. However, the total metal concentration is a poor criterion for assessing the biological activity, or the potential for environmental transport, of a radionuclide bound up in sediment or soil, as not all physico-chemical forms of a particular metal will have the same properties. Therefore, a study of the bio-availability of radionuclides in soils and sediments must attempt to determine not just total concentration levels, but also to which soil or sediment component a particular radionuclide is bound.

The observation of significant levels of radioactivity in the environment due to a particular radionuclide is, in general, associated with only ultra-trace (e.g. pg/g) quantities of the radionuclide in terms of mass. It is, therefore, impossible to isolate and study the chemical compounds and complexes of the radionuclides by conventional chemical and spectroscopic techniques. Highly desirable though this would be, we must be content with inferences drawn from a series of observations of the extractive behaviour of the relevant chemical forms of the radionuclides. The aim of this work is to conclusively relate particular behaviour patterns with specific chemical species.

Our previous sequential extraction study<sup>1</sup> of speciation in sediments and soils from the vicinity of uranium ore deposits has been extended in the present work<sup>2</sup> by use of a series of extractions using various organic solvents. Of all the organic solvents used, only acetic acid was found to remove large amounts of the radionuclides  $^{210}\text{Pb}$  (ca. 30-70%) and  $^{226}\text{Ra}$  (ca. 10-55%) from the soil and sediment samples studied. The failure of the other organic solvents, and in particular DMF which is generally an excellent solvent for most metal complexes of organic ligands, to extract appreciable amounts of  $^{210}\text{Pb}$  and  $^{226}\text{Ra}$  activity suggests that these radionuclides are not present as discrete molecules composed of metal atoms and organic ligands.

Our results show that two broad types of radionuclide species exist for both  $^{210}\text{Pb}$  and  $^{226}\text{Ra}$ , those soluble in boiling acetic acid (which will include many inorganic complexes) and those insoluble (such as polymers or the extended arrays of mineral structures). The solubility in acetic acid of large percentages of these radionuclides suggests that even under mildly acidic conditions likely to be encountered in the environment, significant amounts of  $^{210}\text{Pb}$  and  $^{226}\text{Ra}$  may be re-dissolved and hence available for further transportation.

In one of the sediments studied, both the thorium and uranium series coexist. For this sediment, the percentages of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  removed at each step throughout the various extractions were essentially identical. This suggests that both isotopes, originating from different decay chains, have been transported in the environment in similar chemical and physical forms, and as expected have reacted similarly with the chemical species present in the sediment.

The very different speciation behaviour of  $^{210}\text{Pb}$  and  $^{226}\text{Ra}$  in the soil and sediment samples indicates that  $^{210}\text{Pb}$  (half-life 22.3 y) originates by adsorption of water-transported  $^{210}\text{Pb}$ , and not by ingrowth from its long-lived precursor,  $^{226}\text{Ra}$  (half-life 1600 y).

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ADSORPTION OF NATURAL RADIONUCLIDES UPON COLLOIDAL PARTICLES

M B Cooper and B J Ralph  
(in collaboration with Dr F Greiser,  
Department of Physical Chemistry,  
University of Melbourne).

INTRODUCTION

Natural waters contain suspended solids ranging in particle size from colloidal to macroscopic. One of the important mechanisms, for the transport of trace metals and radioactive elements in surface and ground waters will be, therefore, adsorption of these species upon particulates (1-3).

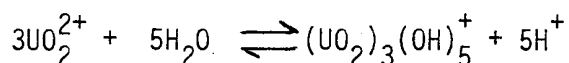
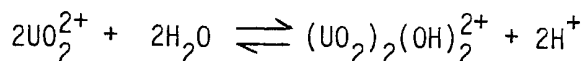
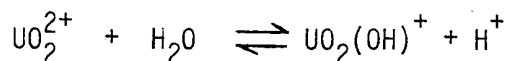
In general, laboratory studies have concentrated upon the adsorption behaviour of metal ions upon metal oxides using model colloid systems such as alumina, silica and amorphous iron oxides (1-5). The surface adsorption of metal ions is strongly pH-dependent. In addition, the presence of hydrolysis products of the metal ion may lead to enhanced adsorption and over certain pH ranges almost complete removal of the metal ion from solution may occur. Three models have been applied to describe the adsorption phenomena (3);

- (a) ion exchange between metal cations in bulk solution and surface protons,
- (b) adsorption of hydrated metal cations on the solid surface and
- (c) the preferential adsorption of certain hydrolysis products upon the oxide surfaces.

It is now well-established that the specific adsorption model (c) is the best explanation for adsorption phenomena involving hydrolyzable metal ions and metal oxides.

The objective of this present study is to investigate the adsorption behaviour of several long-lived radionuclides of the uranium series using colloidal metal oxides as substrates. Knowledge of the solution conditions which give rise to high adsorption densities will be important in predicting the transport mechanisms for radionuclides in surface and ground waters in the environs of uranium mining and milling sites.

The first of the long-lived radionuclides of the natural uranium series to be examined will be uranium ( $^{238}\text{U}$ ) itself. In aqueous solution uranium exists as the uranyl ion ( $\text{UO}_2^{2+}$ ) at low pH and, at higher pH, various monomeric and polymeric hydroxy-species exist,



In addition precipitation of  $\text{UO}_2(\text{OH})_2 \cdot \text{H}_2\text{O}$  will occur at high pH.

### EXPERIMENTAL

Studies of the adsorption of uranium on colloidal silica are made using a 500 mL glass cell which is designed to allow the solution to be de-aerated with nitrogen, temperature and pH to be monitored and the addition of acid to adjust the pH of the solution. Carbon dioxide can have a significant influence upon the nature of the products of the hydrolysis of uranium in solution and therefore it is necessary to de-aerate the solution for 16 hours prior to the commencement of each experimental run. Sodium nitrate at a concentration of 0.001 M is used as the supporting electrolyte. The silica has a known surface area of  $62.75 \text{ m}^2/\text{g}$  and its concentration in solution is 0.01 g/l for each adsorption run. Temperature of the cell is maintained at  $25 \pm 1^\circ\text{C}$ . After each adjustment of the pH the solution is equilibrated for 30 min. and 1-5 mL samples of solution are extracted by syringe. Before the solution is analysed for uranium content any colloid or precipitate is removed by filtration through a  $0.1 \mu\text{m}$  membrane filter.

Three techniques have been evaluated for analysis of the solution for uranium,

- (a) polarography in which both the U(VI) - U(V) polarographic wave and the nitrate catalytic wave were measured,
- (b) U.V. spectrophotometry
- (c) radioactive tracer method using  $^{232}\text{U}$ .

### RESULTS

To date, adsorption curves (% adsorption versus solution pH) have been obtained with initial uranium concentrations of  $10^{-6}\text{M}$ ,  $10^{-5}\text{M}$  and  $10^{-4}\text{M}$ .

With the same conditions except for the absence of silica colloid precipitation curves have been obtained to provide the baseline for the removal of uranium from solution. Typical adsorption curves are shown in Figure 1 for uranium solution of concentration  $10^{-4}$  M in the presence and absence of silica colloid. Curve B, with no silica present, demonstrates the precipitation of uranium hydroxide with increasing pH. In the presence of silica colloid the pH range during which there is significant decrease in soluble uranium moves lower (curve A). This behaviour suggests that adsorption of a hydrolyzed uranium species on the colloid is the dominant mechanism for removal of uranium from solution.

Of the techniques investigated for the assay of uranium, polarography has proved to be unsuitable and results were irreproducible. Measurements based upon the nitrate catalytic wave offer a sensitive method for uranium assay, however the behaviour of the wave is non-linear with uranium concentration. Both U.V. spectrophotometry and the radioactive tracer approach are suitable for this type of work. The former method suffers from one disadvantage due to its lack of sensitivity and reliable analysis has not been possible when the concentration of the uranium solution is less than  $10^{-5}$  M. Experiments performed to date have established that the use of  $^{232}\text{U}$  tracer to determine the percentage of uranium present in solution at a particular pH is the best approach for two reasons. Adsorption data can be obtained with uranium concentrations as low as  $10^{-6}$  M and both the percentage of uranium remaining in solution and that which is bound to the colloidal phase can be determined by alpha counting.

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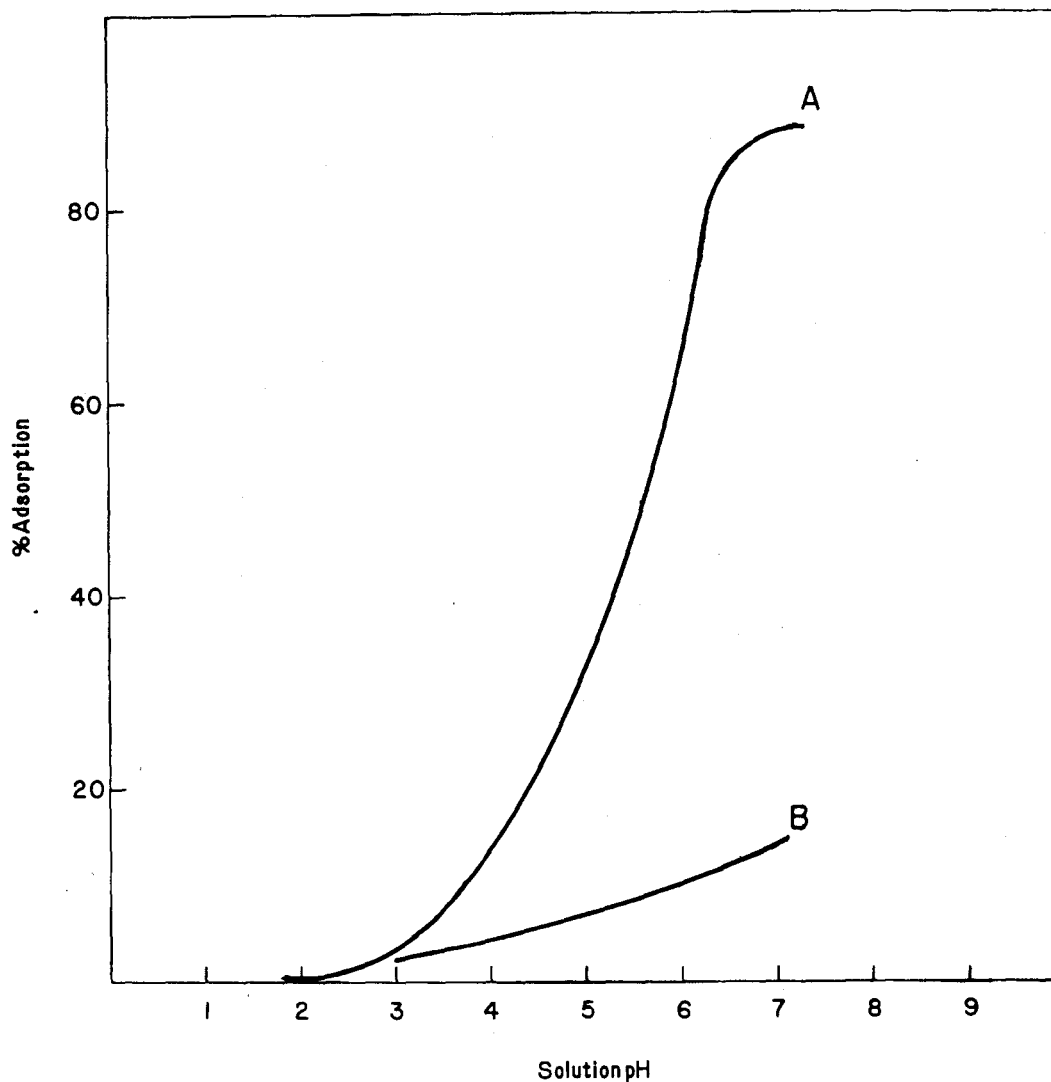


Figure 1 Adsorption curves for  $10^{-4}$  M uranium solution in the presence (A) and absence (B) of silica colloid.

AN ASSESSMENT OF RADIOACTIVITY IN THE ENVIRONS OF A  
TITANIUM DIOXIDE PLANT AT BUNBURY, WESTERN AUSTRALIA

M B Cooper and G A Williams

A previous report<sup>1</sup> describes radioactivity levels at various stages throughout the Laporte Australia Limited titanium dioxide plant at Bunbury in Western Australia. The principal radionuclides detected in the ilmenite feed were  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$  and  $^{228}\text{Th}$ , and it was found that approximately 14 MBq of activity due to  $^{226}\text{Ra}$  and 67 MBq of activity due to each of  $^{228}\text{Ra}$  and  $^{228}\text{Th}$  enter the plant daily. Essentially all of this radioactivity leaves the plant in the form of acidic effluent, which is pumped to disposal lagoons and allowed to neutralise and drain away.

The present study<sup>2</sup> concentrates on the environmental effects of radioactivity both from the effluent and from areas where heavy mineralisation of beach sands occurs naturally. Samples of sediment, algae, water and seafood (crabs and mussels) were collected from Leschenault Inlet (Figure 1) adjacent to the effluent disposal site, and from other areas along the coast including Perth (Swan River Estuary), Mandurah (Peel Inlet) and Busselton (Indian Ocean). Samples of town drinking water from Perth, Bunbury and Capel were also studied.

Activities due to  $^{238}\text{U}$ ,  $^{226}\text{Ra}$  and  $^{210}\text{Pb}$  (uranium series), and to  $^{228}\text{Ra}$  and  $^{228}\text{Th}$  (thorium series) in sediments from parts of Leschenault Estuary (notably at the centre of the pipeline, and at sampling sites 3, 4 and 7 in Figure 1 close to the effluent disposal area) were elevated by comparison with sediment from elsewhere. The exception was a sample of organic sediment from Mandurah (Peel Inlet, sampling Station 4) which showed activities similar to the highest found in Leschenault Estuary (typically ca. 30-40 mBq/g for  $^{226}\text{Ra}$  and 60-90 mBq/g for  $^{228}\text{Ra}$  and  $^{228}\text{Th}$ ). Several algae samples from Leschenault Inlet had slightly higher activities than samples from elsewhere, and all water samples (including town drinking water) had only very low activities due to  $^{226}\text{Ra}$  (less than 20 mBq/l) and  $^{228}\text{Ra}$  (less than 70 mBq/l) and no detectable  $^{228}\text{Th}$  activity.

Mussel samples from Leschenault Inlet and from the other areas showed low activities due to all radionuclides in both shell and flesh. Crabs from Leschenault Inlet showed elevated levels of  $^{228}\text{Ra}$  in particular in their

shells (up to 18 mBq/g), but levels of activity in the flesh were much lower (a maximum of 4 mBq/g due to  $^{228}\text{Ra}$ ). Levels of  $^{228}\text{Th}$  in both shell and flesh were lower than those of  $^{228}\text{Ra}$  by a factor of four.

Detailed tabulations of activities, and discussion of the results, are presented in the Report<sup>2</sup>.

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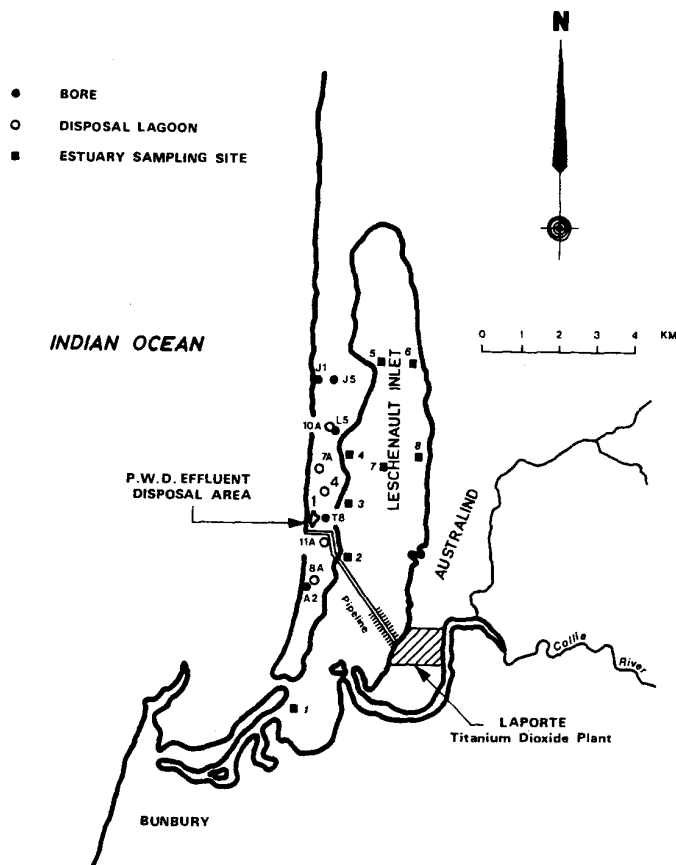


Figure 1. Map of the Bunbury area showing the locations of the Laporte titanium dioxide plant, effluent disposal area, and sampling sites in Leschenault Estuary.

LIQUID SCINTILLATION COUNTING OF ALPHA-EMITTING  
NUCLIDES OF THE URANIUM SERIES

M B Cooper and M J Wilks

INTRODUCTION

The analysis of environmental materials using conventional methods for the determination of naturally occurring alpha emitting radionuclides of the uranium series is associated with drawbacks which include complexity of sample preparation, problems of self absorption and long counting times due to the low efficiency associated with high resolution alpha counting. A comprehensive discussion of the advantages and disadvantages of the various methods employed in the determination of alpha emitting nuclides is provided by McDowell (6). In recent years the potential of liquid scintillation counting of alpha particles has been recognised and has proved useful in the assay of individual actinides (1, 2, 3, 4, 5). The use of a scintillant/extractant solution to isolate the radionuclide of interest simplifies sample separation. Liquid scintillation counting eliminates the problem of sample self absorption whilst providing a system which is almost 100% efficient. Unfortunately, due to the inherent poor resolution, alpha liquid scintillation counting cannot be used to determine individual isotopes unless they are separated by energies of more than 300-500 keV.

In many cases, especially those involving routine monitoring of mining and milling situations, it is advantageous to know the contribution of each of the hazardous radioelements present in a particular sample. Our approach to the development of an analytical scheme to determine the contribution of individual radioelements has involved efforts to sequentially separate radionuclides of the uranium series from a matrix using established chemical procedures followed by radioactive measurement of each nuclide using liquid scintillation counting.

EXPERIMENTAL

The present investigation was confined to environmental samples associated with uranium mining and milling. In these circumstances it is expected that the radionuclides of the uranium series will exceed those of the thorium series to such an extent that the contribution from the latter radionuclides may be neglected when assaying total activity.

$^{229}\text{Th}$  and  $^{232}\text{U}$  were used as tracers to follow the separation steps including the modifications necessary to provide a scintillant/extractant solution of the separated element. It was found that the method due to Sill (7) provided the most acceptable scheme. Yields obtained using the modifications proved similar to those reported by Sill and by Percival and Martin (8). It should be noted that the use of scintillant/extractant solutions can be adapted to any of the recognised separation methods e.g. the Health and Safety Laboratory, New York (9). The method adopted proved satisfactory using the above tracers with a variety of sample matrices. Work is in progress using samples of widely varying origin and composition obtained from the Ranger Uranium Mill to assess the reproducibility of the method.

A key factor in the successful application of liquid scintillation to the determination of alpha-emitting radionuclides has been the use of liquid - liquid extractants to transfer the nuclides of interest from the aqueous phase to the water immiscible scintillator (10). In this way quenching effects are reproducible from sample to sample in terms of pulse height response and peak location. The scintillant mixture which proved most successful comprises 200 g/l naphthalene, 4 g/l (2-(4'-biphenyl)-6-phenylbenzoxazole) PBB0 in toluene. Trioctyl phosphine oxide (TOPO) has proved a suitable extractant for thorium, uranium and polonium. The amount (0.8 g) necessary to effect complete transfer of an element is insufficient to cause extensive quenching. Thus an extractant/scintillator comprising the scintillant mixture previously mentioned combined with 8% TOPO is now being used exclusively in the present study. Elements of interest are being transferred to the organic phase using 2 x 5 ml of extractant/scintillant solution and combined with 10 ml scintillant prior to counting.

Pulse shape discrimination takes advantage of the slower decay of the light pulse produced by alpha particles and allows separation of the beta/gamma components from alpha events. This provides a sensitive method of measurement due to removal of the beta/gamma continuum from the pulse height spectrum and to the reduction of background by two orders of magnitude (11). McDowell (6) has listed the requirements for optimum pulse shape discrimination; these include small volume (1.5 -2.0 ml), complete removal of oxygen and absence of colour quenching. Studies have shown that adequate separation is possible using 10-20 ml samples provided no more than 0.8 g of TOPO is present, the sample is oxygen purged and colourless. Variations in sample preparation, and the concentrations of extractant or scintillant cause fluctuations not only in the separation of alpha from beta/gamma events but also in the alpha count rate when pulse shape discrimination is used.

When pulse height analysis is used quenching produces a decrease in pulse height, however the integrated count under the alpha peak remains unchanged (6). Reduction of PBB0 by half or the doubling of the concentration of TOPO does not affect the count rate when this method of analysis is used. Resolution using 10-20 ml samples is much the same for both methods.

To date investigations have centred mainly upon assessment using pulse height analysis although some preliminary studies have been carried out using the more complex counting array required for pulse shape analysis.

### DISCUSSION

Immediately after uranium separations the spectrum is due solely to three alphas;  $^{238}\text{U}$  (100%),  $^{234}\text{U}$  (100%) and  $^{235}\text{U}$  (4.5%). However if the separated sample is left standing for some time the beta contribution will increase due to the in growth of  $^{234}\text{Th}$  (24 days). While betas due to  $^{234}\text{Th}$  are of low energy and offers little interference to the alpha peak  $^{234}\text{Th}$  is always in equilibrium with  $^{234\text{m}}\text{Pa}$  (1.7 min.) which produces a high energy beta which underlies the alpha peaks thus increasing the background. There is a limiting concentration of uranium which can be extracted. This limit results from the colouration of the scintillator by a yellow uranium complex which causes serious quenching. Because of this colour quenching samples containing large amounts of uranium should be diluted and an aliquot of the resulting solution used to determine uranium content.

$^{234}\text{Th}$  and  $^{230}\text{Th}$  are readily extracted from 3M  $\text{HNO}_3$  with 8% TOPO in toluene with an overall chemical recovery in excess of 95%. However in samples containing uranium and its daughters the thorium fraction will contain both  $^{230}\text{Th}$  and  $^{234}\text{Th}$  and while the method may be used to determine  $^{230}\text{Th}$  the sensitivity of analysis will decrease as the ratio of  $^{234}\text{Th}/\text{Pa}$  to  $^{230}\text{Th}$  increases. The limit of detection will depend upon the amount of  $^{234}\text{Th}$  present and therefore the length of time  $^{234}\text{Th}$  has been separated from its parent.

For example this difficulty arises in the determination of  $^{230}\text{Th}$  in samples such as yellowcake product where the amount of  $^{234}\text{Th}/\text{Pa}$  is greater than the amount of  $^{230}\text{Th}$ . Here pulse shape analysis would be the method of choice as it would separate alpha from beta/gamma components. However pulse height discrimination will be quite sensitive for assessing thorium content of old tailings where uranium has long been separated from its daughters.

A procedure incorporating the use of liquid scintillation counting of Radium-226 is well established. As the details of this method are the subject of an earlier report (11) they will not be discussed at this time.

Preliminary investigation has shown that the use of the liquid scintillation extraction and counting can provide a suitable means of assessing  $^{210}\text{Po}$  which has been isolated from a matrix using the sequential separation method due to Sill<sup>(7)</sup> and that 8% TOPO/toluene is a satisfactory extractant.

### CONCLUSIONS

While significant improvement in the lower limit of detection may be obtained using pulse shape discrimination in most cases, the less demanding technique of pulse height analysis is adequate to measure at least down to the radiological prescribed maximum of a particular isotope. The combination of chemical separation and liquid scintillation counting can prove useful for many routine monitoring procedures in the assay of specified areas provided one is aware of the limitations associated with a particular sample.

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USE OF SOLID STATE DETECTORS FOR SPECTROMETRY OF CONVERSION ELECTRONS  
EMITTED IN THE DECAY OF LEAD-210

I S Leith

INTRODUCTION

This project, commenced in 1981, was continued along two lines:

- (i) use of a silicon surface barrier detector (SiSBD) of high resolution for measurement in vacuum of conversion electrons emitted in the decay of  $^{210}\text{Pb}$ , and
- (ii) development of L X-ray spectrometry using a thin window NaI(Tl) scintillation detector and its incorporation into a method of measuring  $\text{CE}_L/\text{L}_X$  coincidence electron spectra in air for  $^{210}\text{Pb}$  decay.

EXPERIMENTAL METHOD

A recently-purchased SiSBD of  $100 \text{ mm}^2$  active frontal surface area,  $100 \mu\text{m}$  depletion depth and a rated electronic noise FWHM of 5.7 keV was made available for use in this project. This offered about 2 keV resolution improvement for electrons over the best detector previously available. This detector was set up in a vacuum system and separately in a  $\text{CE}_L/\text{L}_X$  coincidence experiment in air.

A NaI(Tl) scintillation detector of 3 inch diameter and 1/8 inch thick, having a full diameter entrance window of aluminium 0.001 inch thick was set up with a source mount attached to the window surround. To the other side of the source mount was attached a SiSBD, connected to a pre-amplifier by a short cable. During measurements, the whole SiSBD, source holder, pre-amplifier and the front of the scintillation detector was enclosed with aluminium foil, earthed to the signal cable earth connections and to the housing of the scintillation detector. This almost completely eliminated electromagnetic interference, to which the SiSBD's are very sensitive.

The signal output from the scintillation detector was amplified and input to a Nuclear Data model 570 ADC and model 6620 Data Acquisition System for examination of singles X-ray and  $\gamma$ -ray spectra or input to an edge crossover single channel analyser to produce a gating signal for coincidence measurements using a linear gate. This latter signal path was also used to self-gate the scintillation detector singles spectrum in order to set up the Bi L X-ray window precisely.

For the  $CE_L/L_X$  coincidence measurement of electron spectra from  $^{210}\text{Pb}$  decay, the output of the SiSBD was connected to the linear gate signal input. To the gate input was connected the output of the single channel analyser, the window of which was set to allow photons in the energy range of the L X-rays of Bi only to produce an output pulse. The gate was operated in the coincidence mode and its output was fed to the Nuclear Data ADC/DAS.

### HIGH RESOLUTION DETECTOR - RESULTS

The performance of the high resolution SiSBD in vacuum and in air was investigated by measurement of the conversion and Auger electron spectrum of  $^{133}\text{Ba}$  and the conversion electron and  $\beta$ -ray spectrum of  $^{210}\text{Pb}$ . For  $^{133}\text{Ba}$ , in vacuum, the K Auger electrons could be partly resolved from the  $CE_K$  electrons from the 80 keV gamma, showing peak centroids at about 34 and 45 keV. The many conversion electron peaks in this spectrum were used to calibrate the detector/amplifier/ADC combination for energy per channel. In the  $^{210}\text{Pb}$  electron spectrum, shown in Figure 1, the  $CE_L$  and  $CE_M$  peaks are partially, but quantitatively, resolved in vacuum using a thin evaporated source of  $^{210}\text{Pb}$ . The centroid energies of these two peaks correspond closely to the theoretical energies given in Table 1.

Table 1

Theoretical internal conversion electron energies ( $E_{CEX}$ ) for conversion to the subshell  $X$ . Values are given for the L subshells and the three lowest M subshells in  $^{210}\text{Bi}$  for the M1 gamma ray transition energy of 46.52 keV.

Subshell	$E_{CEX}$ (keV)
$L_I$	30.13
$L_{II}$	30.81
$L_{III}$	33.10
$M_I$	42.52
$M_{II}$	42.82
$M_{III}$	43.34

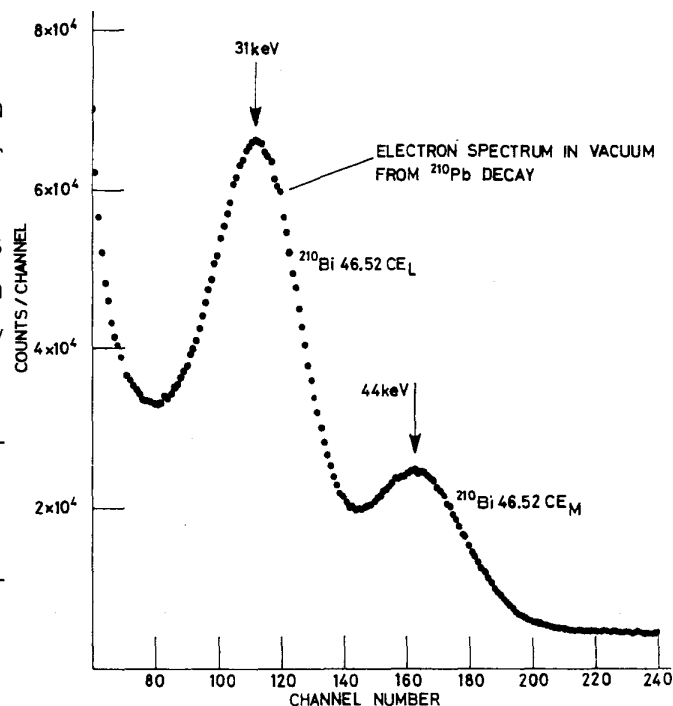


Figure 1. Electron spectrum in vacuum of conversion electrons and beta rays arriving in  $^{210}\text{Pb}$  decay, using a high resolution SiSBD in a measurement system optimised for low noise.

### DEVELOPMENT OF Bi L X-RAY GATING SIGNAL - RESULTS

The photon spectra obtained from the thin-window NaI(Tl) scintillation detector, both singles and self gated, are shown in Figure 2. The peak due to Bi L X-rays at 9.4 to 16.4 keV shows excellent separation from both the 46.5 keV  $\gamma$ -ray and the 3.06 to 4.0 keV Bi M X-rays. The gating signal could be accurately set up to follow detection of L X-rays only, as shown in Figure 2.

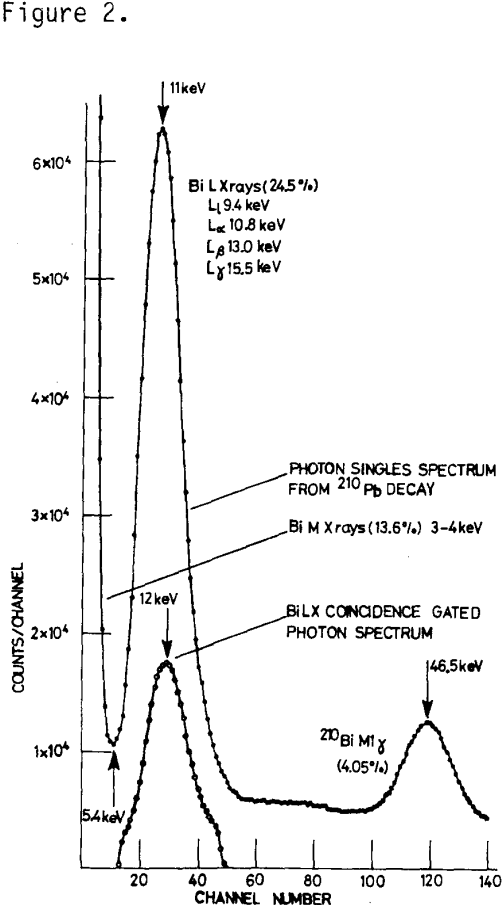


Figure 2. Singles photon spectrum in air of X and gamma rays arising in  $^{210}\text{Pb}$  decay, using a thin-windowed NaI(Tl) detector. Also shown is the same spectrum self-gated in coincidence with the Bi L X-rays in the singles spectrum.

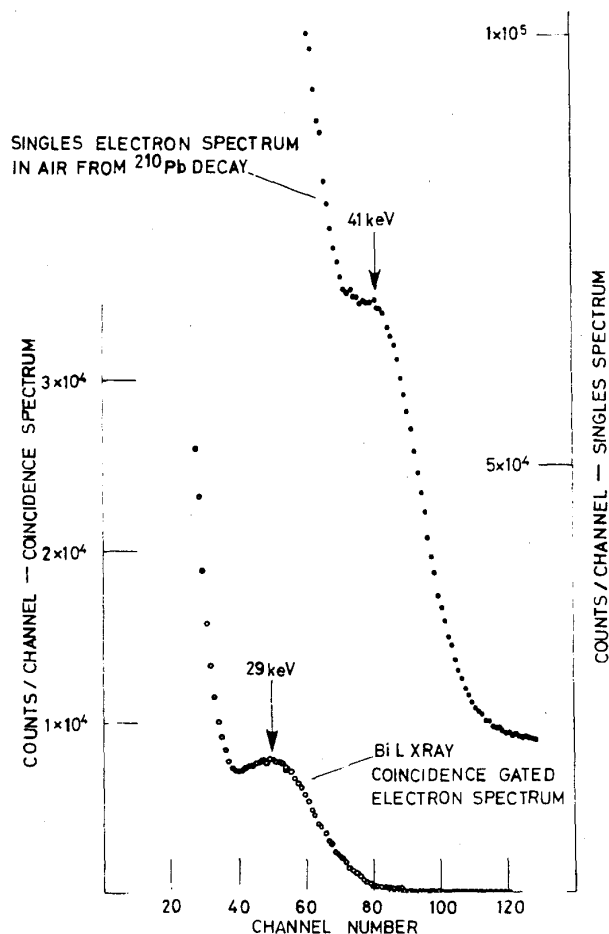


Figure 3. Singles electron spectrum in air of conversion electrons and beta rays arising in  $^{210}\text{Pb}$  decay, using a high resolution SiSBD in a temporary coincidence arrangement. Also shown is the same spectrum gated in coincidence with the Bi L X-ray gate shown in Figure 2.

### CE<sub>L</sub>/L<sub>X</sub> COINCIDENCE SPECTRA - RESULTS

Using the Bi L X-ray gating signal, the electron spectrum obtained from decay of  $^{210}\text{Pb}$  was coincidence gated in air to reveal a broad peak due to the Bi CE<sub>L</sub> merging into the steeply rising  $^{210}\text{Pb}$   $\beta$ -ray (17 keV maximum)

and degraded conversion electron spectrum at low energies typical of such spectra taken in air. This spectrum is shown in Figure 3. Also shown is the smaller electron spectrum in air in the same temporary coincidence arrangement. It is notable that the  $CE_M$  hump at about 41 keV without coincidence gating appears to be completely removed in the coincidence spectrum, as is the 63 keV  $\beta$ -ray of  $^{210}\text{Pb}$  decay and the 1161.5 keV  $\beta$ -ray of  $^{210}\text{Bi}$  decay.

## DISCUSSION

### (a) Conversion Electron Spectrometry

This work has shown that quantitative measurements of the activity of  $^{210}\text{Pb}$  in samples prepared as thin evaporated sources may be made either by use of the small, high resolution SiSBD's for singles spectrometry of the full electron spectrum in vacuum or by performing  $CE_L/L_X$  coincidence in air to permit measurement of the L conversion electrons alone. Neither method permits a high signal-to-noise ratio nor a high efficiency of measurement. Efficiency is limited in both methods by the necessarily thin effective thickness of sources for quantitative measurement of such low energy electrons. Further, the very small area of the high resolution SiSBD's and the geometrical factors for the coincidence technique severely reduce the efficiency in each case.

Resolution of the coincidence technique should be much improved by performing it in vacuum. It may then be practical to use much larger detectors to increase the efficiency. The resolution of SiSBD's may be improved by moderate cooling<sup>(1,2)</sup> or, after careful selection of detectors and field effect transistors, by more severe cooling.<sup>(3)</sup> Cooling of larger SiSBD's may then result in achieving sufficiently high resolution for singles electron spectrometry.

### (b) L X-ray Spectrometry

The singles photon spectrum obtained for  $^{210}\text{Pb}$  decay (Figure 2) shows a clearly resolved Bi L X-ray peak some 6 times in area that of the 46.52 keV gamma ray peak. Examination of compiled data<sup>(4)</sup> for the decay series from  $^{210}\text{Pb}$  to stable lead reveals that the Bi L X-rays arise only in the 46.52  $^{210}\text{Bi}$  transition and that no other L X rays arise in significant intensity. The best available data<sup>(4)</sup> on the intensity of the Bi L X rays is 24.5% vs. 4.05% for the 46.52 keV  $\gamma$  ray, 64% for the 46.52  $CE_L$  and 17%<sup>(4,5)</sup> for the 46.52  $CE_M$ . This leads to numerous considerations

favouring L X-ray spectrometry for quantitative measurement of  $^{210}\text{Pb}$  and other environmental radionuclides with low gamma abundance. These include:

- (i) Quantitative measurement of conversion electrons of energies of the order of 30 keV requires extremely thin sources to reduce self absorption losses and uncertainties. This problem is not so critical for Bi L X rays at about 10 to 16 keV. In this case, greater source thickness can be tolerated, thus greater mass of sample can be used and sensitivity can be increased.
- (ii) The frontal area of the NaI(Tl) detector used in these measurements is approximately  $46\text{ cm}^2$ , some 46 times that of the high resolution SiSBD used for the electron measurements reported here. Thus, in this comparison, X-ray spectrometry permits efficient measurement of samples of much greater area and mass, increasing sensitivity.
- (iii) The efficiency for detection of incident X-rays for data reported here approaches 100%. This is some 6 times the best possible efficiency for detection of the 46.52 keV  $\gamma$  ray following  $^{210}\text{Pb}$  decay. The maximum possible efficiency of detection of the Bi 46.52  $\text{CE}_L$  is some 16 times that for the  $\gamma$  ray. However, in practice, self absorption and absorption in the dead layer of the detector of the  $\text{CE}_L$  severely limit the sensitivity for measurement of  $^{210}\text{Pb}$  by this means. This method will have little advantage over the Bi L X method due to the product of abundance and detection efficiency.
- (iv) L X rays in the case considered here (a purified sample of  $^{210}\text{Pb}$ ) do not suffer interference from X-rays from elements nearby in the periodic classification which will have energies only slightly shifted from those of Bi L X rays. In a mixed sample of natural radioactivity, such interferences may well occur. They may not be separable using NaI(Tl) detectors. A detector of higher resolution (such as a Si or Ge diode) may be needed to do this. In the case of conversion electron spectrometry, the energies depend on the nuclear energy transition in which they arise and are in general more widely dispersed than the X-rays. This may not lessen the apparent advantage of L X-ray spectrometry, if a detector having very high resolution, large area and small frontal dead layer thickness is available.

#### FUTURE WORK

Under the research and development project reported here, it is proposed to explore conversion electron spectrometry in vacuum by the coincidence technique ( $\text{CE}_L/L_\gamma$ ) and by moderate cooling of detector/pre-amplifier

combinations using SiSBD's of somewhat larger area than used to obtain the data reported here, with a view to practical application of these techniques to the measurement of  $^{210}\text{Pb}$  and other radionuclides in environmental samples by conversion electron spectrometry.

Under an associated research and development project, it is proposed to explore L X-ray spectrometry for practical samples using NaI(Tl) detectors, thick silicon detectors and germanium detectors of high resolution having a thin front contact such as the Princeton Gamma Tech hyperpure planar germanium detector purchased some years ago for environmental spectrometry.

It is desirable for these purposes to have good data on relevant L X ray abundances from environmental radionuclides. Such data does not exist at this stage e.g. the  $24.5\%$   $46.52 \text{ CE}_L$  intensity figure quoted here has an associated uncertainty of  $12.25\%$ .<sup>(4)</sup> It may be desirable to set up a measurement of this quantity. Alternatively, it may be possible to apply the method currently employed in this Laboratory for measurement of concentration of radionuclides that emit gamma rays of higher energies. This method involves direct comparison to standards prepared in the same geometry and with the same matrix. In this case neither abundance nor detector efficiency need be known. The method may be applicable at the low energies of the L X-rays from heavy elements.

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ENVIRONMENTAL RADIATION MONITORING AND STUDIES

J.C. Duggleby, P.A. Smith and R.A. Lauder

Radioactive fallout in Australia from nuclear weapons tests in the atmosphere has been monitored on a continuous basis since the 1950's. Since 1974, when the last series of atmospheric nuclear weapons tests of significance to Australia took place, the extent of the monitoring program has been reduced as the levels of long lived fallout have decreased. Between 1975 and 1980, the measurement of the long-lived radionuclides, strontium-90 and caesium-137, had been allowed to assume a lower priority and only sufficient work had been done to confirm that there existed no health hazard to the Australian population from this source. As a result, a considerable quantity of environmental material had been collected but not fully analysed to obtain data of scientific, rather than public health, importance. A major part of the group's effort throughout 1982 was devoted to the radiochemical analysis of this accumulated material. At the end of the year, only a very small number of environmental samples remained to be analysed.

The surveillance program for the early detection of fresh fission products in ground-level air has been maintained. Data from this program are routinely forwarded to the International Reference Centre for Radioactive (a part of the World Health Organisation) for publication in their quarterly reports. No fresh fission products were detected in 1981 but traces of the medium-lived cerium-144 (half-life 284 days), originating from the atmospheric detonation of a nuclear device in China on 16 October 1980, were detected from time to time. Low concentrations of the long-lived fission product caesium-137 (half-life 30 years) continued to be detected in ground-level air, having filtered down from the stratosphere where it had been held since having been injected there from atmospheric detonations during the last 35 years or so; however concentrations were noticeably lower than in previous years.

The Laboratory continued to fulfil its obligations as laid down in the "Guidelines for Environmental Radiation Monitoring During Visits by Nuclear Powered Warships to Australian Ports" and in the Amendment No. 1 published in April 1981. Specified marine samples were collected from regular used berths in Cockburn Sound every quarter and before and after each ship visit. In other ports visited by ships on an occasional basis, similar samples were collected before and after each visit and again three months later. Samples were analysed for the presence of cobalt-60 or any other artificial

gamma-emitting radionuclide known to characterise the radioactive waste likely to be held in a nuclear powered warship. Thermoluminescent dosimeters were provided for placement at selected locations in the vicinity of each visiting ship. The results of the monitoring are published annually by the Department of Home Affairs and the Environment.

All liquid effluent from areas of the Laboratory where unsealed radioactive sources are used is collected in a number of large holding tanks. Before release of the effluent, the activity must be shown, by analysis, to be below the limits specified in the appropriate Victorian Government Regulations (1959). All such samples analysed during the year were found to meet that requirement.

Owing to the high priority placed on completing the analysis of accumulated environmental samples for fallout from nuclear weapons tests, little work was done on the comprehensive study of natural radiation background and the influence of many activities on it.

A study of the technical and administrative problems involved in the regulated disposal of low levels of radioactive waste by the user was begun. The results of the study when completed will be forwarded as a Working Group report to the Commonwealth/State Committee on Radionuclide Waste Management.

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