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DETECTION SYSTEMS FOR THE LOW-LEVEL RADIOCHEMICAL
ANALYSIS OF IODINE-131, IODINE-129 AND NATURAL
IODINE IN ENVIRONMENTAL SAMPLES (a)

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DETECTION SYSTEMS FOR THE LOW LEVEL RADIOCHEMICAL
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Summary

A procedure based on chemical separation techniques and activation analysis has been developed for the sequential analysis of low levels of ^{131}I , ^{129}I and natural iodine in environmental samples. The iodine is first separated from the samples by oxidation. The separated iodine is then counted by low level, beta-gated gamma-ray spectrometry for the measurement of ^{131}I . The chemical yield for the separation is measured by means of ^{125}I tracer. Activation analysis is used for measurement of the separated natural iodine (^{127}I) and ^{129}I . The natural iodine is estimated from either the ^{126}I or ^{128}I activity produced in the sample; the induced ^{130}I is used to estimate the ^{129}I concentration.

The measurement of ^{130}I at low levels requires specialized counting methods. Interfering activities which may be present in the irradiated sample include ^{125}I , ^{126}I , short-lived fission-product iodine activities produced by activation of uranium impurities, and ^{82}Br from bromine impurities. Since ^{130}I decays with several coincident gamma-rays, multiple gamma-ray coincidence counting techniques can be used to reduce background and discriminate against interfering activities. Several

three-and four-segmented NaI(Tl) detectors have been used for these measurements and compared with other detector systems.

The sensitivities for different coincidence counting modes were compared for several of the detector systems. Detection sensitivities achieved for ^{131}I and ^{129}I are 0.02 dpm and 10^{-6} dpm respectively.

Measurements have been made of iodine radioactivity in the environment at concentrations below the limits established for radiation protection purposes.

Introduction

The measurement of radioactive iodine is useful for tracer studies and detection of potential contamination problems in environmental samples at concentrations much below those routinely used for radiation protection and environmental surveillance. The radioactive iodine isotopes of major interest are fission-produced 8-day ^{131}I and 1.6×10^7 -year ^{129}I .¹⁻¹⁹ Determination of the concentration of natural iodine in the same environmental samples is also required when evaluating the impact of radioactive iodine on the environment and population.^{16,19}

A procedure based on the iodine activation analysis method of Studier and co-workers²⁰ has been developed for the sequential analysis of low levels of ^{131}I , ^{129}I and natural iodine in environmental samples. It consists of the following techniques:

1. Gamma-ray spectrometric analysis of the bulk sample for measurement of gamma radioactive nuclides.

2. Carrier-free separation of iodine from the sample with ^{125}I tracer being used to measure the chemical yield.
3. Measurement of ^{131}I activity by beta-gated gamma-ray spectrometric methods when more sensitive measurements are required than can be obtained by analysis of the bulk sample.
4. Measurement of the ^{125}I tracer to determine the iodine recovery.
5. Thermal-neutron activation of the iodine for determination of ^{129}I and natural iodine by activation analysis. A comparator standard of mixed ^{125}I , ^{127}I and ^{129}I is irradiated with the sample.
6. Rapid separation and purification of the iodine from the irradiated sample.
7. Measurement by gamma-ray spectrometry in the purified iodine of standard ^{125}I tracer, ^{126}I and ^{128}I produced by activation of natural iodine, and ^{130}I produced by activation of ^{129}I . Beta-gated, multiple gamma-ray, coincidence spectrometric methods are used when maximum sensitivity is required.
8. Computer analysis of the counting data and calculation of the concentration of ^{131}I , ^{129}I and natural iodine in the samples.

The sensitivities of the iodine estimates are primarily dependent upon the sensitivity of the radioactive measurement

methods selected. Other factors affecting the sensitivity (some of which can not always be controlled) are the available sample size, the chemical yield, the thermal neutron flux, and the level of interfering radioactivity. This paper describes the gamma-ray spectrometric methods developed for measurement of ^{131}I , natural iodine (through the activation products ^{126}I and ^{128}I) and ^{129}I (through the activation product ^{130}I). These methods are evaluated for their sensitivity and selectivity.

^{131}I Measurement Methods

The measurement of ^{131}I in bulk samples by gamma-ray spectrometry is normally attempted prior to chemical separation of the sample iodine. The sensitivity of such measurements is limited by the fallout and natural radioactivity in the sample. Bulk environmental-sample counting of ^{131}I by gamma-ray spectrometry generally requires at least one d/min. (dpm) for detection.

A sample being processed for ^{129}I activation analysis can easily be measured for ^{131}I after iodine separation chemistry and before neutron irradiation. Carrier-free ^{125}I tracer is added to the sample; the iodine isotopes are separated by combustion in oxygen and the released iodine is then trapped on activated charcoal. Further purification of the iodine is accomplished by combusting the original charcoal trap in oxygen and

trapping the released iodine on a small (several milligram) activated charcoal trap. The iodine absorbed on this trap is mounted for counting by placing the charcoal between two plastic scintillator discs and heat sealing the discs at the edge. Beta-gamma coincidence spectrometry^{21,22} is then used to measure the ^{131}I in the separated iodine. Corrections are made for self-absorption of beta radiation in the charcoal and iodine contained between the scintillators. Corrections are also made for chemical yield based on ^{125}I recovery measured by an x-ray gamma coincidence system.²²

The measurement of ^{131}I by beta-gated, gamma-ray spectrometry rather than by low-level beta counting allows selective measurement of ^{131}I in the presence of radiochemical impurities. Iodine-131 detection sensitivities as low as 0.02 dpm have been achieved with this method.

Iodine Activation Analysis

Iodine activation analysis requires the transfer of iodine from the charcoal between the plastic scintillators to a quartz irradiation ampoule. This is accomplished by mechanically removing the charcoal from the scintillator and transferring it to a vacuum system. The charcoal is heated to liberate the iodine which is trapped in a quartz tube at liquid nitrogen temperature. The tube is then sealed to make an irradiation ampoule.

Quartz ampoules containing the iodine separated from the samples are irradiated with reactor neutrons for eight to twenty-four hours. Comparator standards containing ^{125}I , ^{127}I and ^{129}I are irradiated with each set of samples.

The neutron capture reactions used for the iodine activation analysis are:

Interfering reactions include:

Following irradiation carrier is added and the iodine samples are further purified by distillation and solvent extraction. The samples are finally precipitated as AgI and mounted on thin plastic scintillators for counting.

The ^{126}I , ^{128}I and ^{130}I activities produced in the sample and comparator standards during irradiation are estimated by gamma-ray spectrometry from several spectra collected over a period of time to obtain half-life information. Low level, beta-gated, multiple gamma-coincidence spectrometric techniques are used when required to measure very small amounts of activity and to discriminate against interfering activities such as those produced by reactions (6) and (7). Post-irradiation chemical processing also limits interference from reaction (7).

Interference from reaction (4) is minimized by use of small activity levels of ^{125}I so that ^{126}I production by reaction (2) predominates. Reaction (5) limits the improved sensitivity that can be obtained by increasing the exposure time and neutron flux. Neutron exposure conditions are selected on the basis of expected natural iodine content of given sample types in order to limit the correction required due to reaction (5) to less than 10%

When the purified iodine sample is available within several hours after the neutron irradiation, ^{128}I measurements can be made on the sample to estimate its natural iodine (^{127}I) content. The sensitivity of this method greatly exceeds normal environmental sample analysis requirements and is dependent primarily on the decay time between irradiation and counting. Well-type 12.7-cm-diameter NaI(Tl) detectors are normally used for these measurements although Ge(Li) detectors can be used with some sacrifice of geometric reproducibility.

When purification of the iodine sample is delayed (usually due to transportation between the irradiation facility and the laboratory) until after the 25-minute ^{128}I activity has decayed, ^{126}I measurements can be used to estimate the natural iodine (^{127}I). These measurements are best made starting about a week after irradiation when the ^{129}I activation product (^{130}I) has also decayed.

The same well-type NaI(Tl) detectors used for ^{128}I measurements are used for ^{126}I measurements. Beta-gated gamma-ray spectrometry is used to reduce background and improve the ^{126}I detection sensitivity when insufficient activity is present for normal gamma-ray spectrometric measurements. The ^{126}I detection limits are 30 dpm and 0.1 dpm by gamma-ray spectrometry and beta-gated gamma-ray spectrometry, respectively. The resulting natural iodine sensitivities depend on the fast (greater than 11 MeV) neutron flux used for the irradiations. For the Washington State University TRIGA reactor operating at the 1 MW level the maximum sensitivity obtained for an 8-hour irradiation is 0.5 micrograms of natural iodine.

Since the amount of ^{129}I in environmental samples is much lower than the natural iodine, special detection systems are often required to estimate the ^{130}I produced by ^{129}I activation. Interferences present in the purified iodine sample include ^{128}I and ^{126}I from natural iodine, fission-product iodine from uranium impurities in the irradiated ampoule, and ^{82}Br if adequate decontamination is not achieved in the post-irradiation chemical

processing. The ^{128}I interference can be eliminated by allowing its decay prior to ^{130}I measurements. The fission-product iodine and ^{82}Br interferences are best controlled by before and after irradiation chemistry, but some instrumental discrimination is useful. Iodine-126 interferences can only be resolved instrumentally but this interference can be controlled to some degree by irradiation conditions since the ^{126}I production is dependent on the fast neutron flux. The ^{130}I and ^{126}I decay schemes are shown on Figures 1 and 2. Ge(Li) detectors can be used for resolution of the ^{130}I and ^{126}I activities when sufficient ^{130}I activity is present and the ratio $^{126}\text{I}/^{130}\text{I}$ is not excessive. High $^{126}\text{I}/^{130}\text{I}$ ratios result in obstruction of the ^{130}I photo-peaks by the ^{126}I Compton. Some improvement in the ^{126}I - ^{130}I resolution is possible with a Ge(Li)-NaI(Tl) spectrometer such as shown on Figure 3.²³ The signals from the two Ge(Li) detectors are summed to improve the detection efficiency; separate spectra are recorded in coincidence and anticoincidence with the NaI(Tl) annulus. Due to the high probability of detection of at least one of the ^{130}I gamma-rays by the NaI(Tl) annulus, the ^{130}I activity is recorded mostly in the coincidence spectrum while the ^{126}I activity is primarily recorded in the anticoincidence spectrum as shown on Figure 4. Samples containing large amounts of natural iodine can produce sufficient ^{126}I to obscure the ^{130}I in the coincidence spectrum. The detection limit of the system for ^{130}I is about 2 dpm.

A better way to discriminate against ^{126}I is to count the sample in a well-type NaI(Tl) detector and measure the ^{130}I sum peaks at 1.9 MeV and 2.3 MeV. The maximum energy ^{126}I sum peak occurs at 1.4 MeV. Additional discrimination against ^{126}I and the fission-product iodine isotopes can be achieved by requiring more than two coincident gamma events in the NaI(Tl) detector system.

Two detector systems designed for such measurements are shown on Figures 5 and 6. These systems can also be operated with beta-gating to reduce background^{21,22,23} and achieve additional ^{126}I discrimination. Background spectra measured with the four-segmented detector (Figure 6) are shown on Figure 7. Figure 8 shows the prominence of the ^{130}I sum peaks in the $\gamma\text{-}\gamma\text{-}\gamma$ and $\gamma\text{-}\gamma\text{-}\gamma\text{-}\gamma$ spectra. ^{126}I spectra measured by the same detector are shown in Figure 9 and fission-product ^{135}I spectra are shown in Figure 10. Relative counts are shown in the ordinates of Figures 8-10.

Various detector systems which we have evaluated for ^{130}I measurements are compared in Table I. The efficiencies for ^{126}I , ^{130}I and ^{135}I have been calculated relative to an efficiency of one for the nuclide counted in a large well-type NaI(Tl) detector. Although the ^{130}I counting efficiency is reduced up to a factor of 10 when coincidence counting modes are used, the ^{126}I and ^{135}I efficiencies can be reduced by factors of 1000. The detection limits for ^{130}I have been calculated by means of the GEM computer program, which is a weighted leastsquares method of estimating radionuclides in a mixture using both spectral and decay

information.²⁵ Although the ^{130}I detection efficiency decreases as additional coincidence requirements are imposed on the system, the resulting background reduction and the shift of most of the observed counts to the high-energy region of the spectrum yield improved ^{130}I detection sensitivities. At the same time discrimination against ^{126}I and ^{135}I is improved.

The ^{129}I detection levels achieved for the various counting systems are also listed on Table I. Irradiations in a thermal neutron flux of about 10^{13} n/cm²/sec result in an ^{129}I detection sensitivity of about 10^{-5} dpm of ^{129}I per dpm of ^{130}I for a variety of detector systems. The activation analysis methods when compared with the direct measurement of ^{129}I by low-energy photon spectrometry show improved sensitivities ranging from 10^4 to 10^6 depending on the ^{130}I counting system used. These high sensitivities are required to measure ^{129}I in most environmental samples.

Applications

These low-level detection methods have been used by Pacific Northwest Laboratories for a number of environmental iodine studies. The concentration of the iodine isotopes have been measured in underground, surface and atmospheric water samples from the Hanford Project environs.⁸ Thyroid tissue and a variety of other environmental samples from various locations in the United States have been analyzed for natural iodine and ^{129}I .¹⁶ Dose calculations showed that the levels observed were a factor of 100 or more below radiation doses from natural sources. Measurements

of particulate and gaseous atmospheric iodine concentrations have also been made.¹⁷ Iodine-129 radioecology is currently being studied.¹⁸

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TABLE I

PERFORMANCE COMPARISON OF VARIOUS DETECTORS FOR THE MEASUREMENT
OF ^{129}I BY ACTIVATION ANALYSIS AND DIRECT COUNTING

Style	Detector			Coincidence Mode	Relative Efficiency			Detection Limit ^(a)	
	Type	Size (cm)			¹²⁶ I	¹³⁰ I	¹³⁵ I	¹³⁰ I (dpm)	¹²⁹ I (dpm) ^(b)
		Dia.	Ht.						
Well	NaI(Tl)	23	23	γ	1	1	1	10	10.x10 ⁻⁵
Split Well	NaI(Tl)	20	20	γ	1	1	1	11	11 x10 ⁻⁵
Well	NaI(Tl)	13	13	γ	.9	1	1	6	6.x10 ⁻⁵
Split Well	NaI(Tl)	20	20	γ-γ	.1	.7	.2	2	2.x10 ⁻⁵
Figure 5	NaI(Tl)	23	20	γ-γ-γ	.01	.2	.02	1	1.x10 ⁻⁵
Four Half Cylinders	NaI(Tl)	20	20	γ-γ-γ	.02	.4	.04	1.4	1.4x10 ⁻⁵
Figure 6	NaI(Tl)	23	23	γ-γ-γ	.02	.3	.05	1.3	1.3x10 ⁻⁵
Four Half Cylinders	NaI(Tl)	20	20	γ-γ-γ-γ	.001	.1	.002	1.4	1.4x10 ⁻⁵
Figure 6	NaI(Tl)	23	23	γ-γ γ γ	.001	.1	.002	1	1.x10 ⁻⁵
Well	NaI(Tl)	23	23	β-γ	.3	.7	.7	.2	.2x10 ⁻⁵
Split Well	NaI(Tl)	20	20	β-γ	.3	.7	.7	.4	.4x10 ⁻⁵
Two Cylinders	NaI(Tl)	20	20	β-γ	.2	.5	.5	.2	.2x10 ⁻⁵
Well	NaI(Tl)	13	13	β-γ	.3	.7	.7	.1	.1x10 ⁻⁵
Split Well	NaI(Tl)	20	20	β-γ-γ	.02	.5	.1	.2	.2x10 ⁻⁵
Two Cylinders	NaI(Tl)	20	20	β-γ-γ	.02	.3	.1	.2	.2x10 ⁻⁵
Figure 5	NaI(Tl)	23	20	β-γ-γ-γ	.004	.1	.06	.1	.1x10 ⁻⁵
Figure 6	NaI(Tl)	23	23	β-γ-γ-γ	.005	.2	.02	.1	.1x10 ⁻⁵
Figure 6	NaI(Tl)	23	23	β-γ-γ-γ-γ	.0003	.05	.002	.1	.1x10 ⁻⁵
Figure 3	Ge(Li)- NaI(Tl)	28	23	γ-γ	.1	.1		2	2.x10 ⁻⁵
Low Energy	Ge(Li)	2.5	.5	γ	¹²⁹ I Counted Directly				1

(a) $1-\sigma$ limit from GEM computer program⁽²⁵⁾

(b) ^{129}I limit based on thermal neutron flux of 10^{13} n/cm²/sec

Fig. 1. ^{130}I Decay Scheme Showing Multiple Gamma-Ray Transitions.

Fig. 2. ^{126}I Decay Scheme Showing Maximum of Two Gamma-Ray Transitions per Disintegration with Maximum Gamma-Ray Energy Loss of 1.41 MeV

Fig. 3. Diagram of Dual Ge(Li)-Dual NaI(Tl) Gamma-Ray Spectrometer

Fig. 4. Spectra of Activated Iodine Sample Obtained with Ge(Li) Spectrometer

Fig. 5. Diagram of Multiple Coincidence NaI(Tl) Gamma-Ray Spectrometer with Side-Hole Annulus and Scintillation Beta Detector

Fig. 6. Diagram of Multiple Coincidence (NaI(Tl) Gamma-Ray Spectrometer With Plastic Scintillator Annulus and Borated Paraffin and Lead Shield

Fig. 7. Background Spectra in the Four-Segmented NaI(Tl) Detector Showing the Reduced Count Rates Observed in the Multiple Coincidence Modes

Fig. 8. ^{130}I Spectra in the Four Segmented NaI(Tl) Detector Showing the Prominence of the Sum Peaks in the Multiple Coincidence Modes

Fig. 9. ^{126}I Spectra in the Four-Segmented NaI(Tl) Detector Showing the Relative Low Efficiency of the Multiple Coincidence Modes for ^{126}I

Fig. 10. ^{135}I Spectra in the Four-Segmented NaI(Tl) Detector

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