In Situ Quantitative Determination of Transuranic Elements in Areas of High-Level Gamma Radiation*

by

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ABSTRACT

A technique is described for passive neutron monitoring of transuranic elements. The method provides quantitative determinations of transuranic element concentrations in a variety of field situations where no other measurement method is possible. The technique can measure concentrations of transuranic oxides as low as 8 nCi/cm^3 and is capable of operating in gamma radiation fields up to megarads per hour. Information on chemical and isotopic composition can also be obtained from the data. Several successful applications of the technique are discussed.

KEYWORDS: Transuranics; neutron monitoring

INTRODUCTION

The in situ detection and measurement of transuranic elements is an extremely difficult problem. These isotopes are primarily alpha-emitting nuclides which emit relatively low energy gamma-rays through very low branching fractions. Transuranic elements also emit characteristic x-radiation in high yield, but this can only be measured in special circumstances. Neutrons are the only characteristic radiations emitted by the transuranic elements which are readily distinguishable in the presence of other radionuclides. These neutrons come from spontaneous fission or from (α,n) reactions on light isotopes such as ^{19}F, ^{17}O, or ^{18}O. For many applications, such as criticality, accountability, or waste management, requiring the quantitative determination of plutonium or other transuranic elements, these neutrons are the only radiations suitable for measurement. For example, the characterization of residual plutonium in process or fabrication hoods undergoing decommissioning is not possible by alpha detection and is only occasionally possible by x-ray measurements. Although gamma detection will frequently work for fairly high quantities of plutonium, shielding by massive machinery may introduce substantial uncertainties. Neutrons are not as severely attenuated by bulk quantities of steel, and remote pockets of plutonium may be detected by neutron measurements.

It may be necessary to measure plutonium concentrations for potential criticality evaluation in trenches, cribs, or storage tanks which have received process wastes. These locations frequently have large quantities of fission products present which would interfere with the detection of plutonium photons, and only neutrons can be unequivocally identified. Similarly, it may be desirable to measure the quantity of transuranic elements released to the environs during an accidental discharge of fission product waste or to determine the transuranic element content of fuel cycle fission product effluents for disposal purposes. The high level gamma radiation present in these cases also restricts the detection of transuranic elements to neutron monitoring.

This communication describes a technique for quantitatively measuring the neutrons emitted by transuranic elements. This technique can evaluate the neutron energy spectrum and thereby infer the chemical and/or isotopic composition of the transuranics in question.

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DISCLAIMER

Finally, this technique can make quantitative measurements of transuranic element concentrations at <10 nCi/g in gamma radiation fields exceeding megarads per hours.

EXPERIMENTAL

There are two basic detection techniques for measuring neutrons emitted by transuranic species. The first of these is the active technique, which incorporates real time measurement of neutrons with ^{3}He, BF_3, fission chamber, or similar detectors and collection and storage of signals with appropriate electronic equipment. This technique is not applicable to operation in high gamma radiation fields, since the electronic detectors will not function properly. In addition, neutron energy evaluation is virtually impossible with this active technique, since the detectors are generally operated in a moderating medium to effectively utilize their enhanced efficiencies at thermal neutron velocities.

The second neutron detection technique, a passive neutron activation method, is the one utilized in this work. The passive technique involves the exposure of various materials to transuranic generated neutrons and subsequent remote determination of the neutron-induced reaction products. This technique is not influenced by high gamma fields and can yield energy related spectral information with appropriate use of threshold reactions.

The following criteria were considered in choosing appropriate materials for passive neutron monitors. The neutron-induced reaction product had to be easily determinable. This was quickly narrowed to radioactive products having distinctive gamma-ray emissions which could be nondestructively determined in high sensitivity gamma-ray spectrometers. The product had to have a relatively high branching fraction through the gamma cascade of interest and a half-life between a few hours and a few days to allow the sample to reach near saturation values within a reasonable exposure period. Time required for transfer from exposure to counting precluded very short half-lives. The material needed to be in a physically manageable and durable chemical form. The material needed to be readily available at reasonable cost; and cross sections for the neutron-induced reactions needed to be as large as possible in the interest of sensitivity.

After a thorough examination of the chart of the nuclides and a few preliminary experiments, the number of suitable passive neutron monitors was narrowed to the four metals, magnesium, copper, zinc, and indium. The neutron reactions of interest in these metals are listed in Table I along with the product half-lives, gamma-rays measured, and branching fractions. The copper and indium capture reactions monitor the thermal neutron flux and are used in different situations rather than simultaneously. The copper reaction is used in most cases where time and proximity to laboratory-based large crystal NaI(Tl) multidimensional gamma-ray spectrometers permits. If rapid data acquisition or remote field measurements are required, the indium reaction can be used in conjunction with a portable anticoincidence shielded NaI(Tl) well counter at nearly comparable sensitivity levels.

The zinc reaction has a practical threshold of 2.3 MeV, and the excitation function rises smoothly through a maximum of 280 mb at 10 MeV. The energy of the neutrons produced by (α,n) reactions on oxygen or fluorine is dependent not only on the kinetic energy of the alpha particle but also on the excitation state of the residual nucleus. Therefore, these reactions result in a spectrum of neutron energies with the maximum being 3.22 and 5.76 MeV for plutonium alpha particles on fluorine and oxygen, respectively. The spontaneous fission neutrons have a typical fission neutron energy distribution curve peaking at 1 to 2 MeV and tailing up to ~17 MeV. The zinc monitor reaction is therefore sensitive to both (α,n) and spontaneous fission neutrons.

The magnesium monitor reaction has a practical threshold of 6.2 MeV and is, therefore, not sensitive to (α,n) neutrons but only to the small percentage of high energy spontaneous fission neutrons above this threshold level. The excitation function for this reaction rises smoothly through a maximum of 195 mb at 13 MeV, and although this monitor is not as sensitive as the others, it provides a specific measure of spontaneously fissioning isotopes such as ^{240}Pu.

Table I. Passive Activation Neutron Monitor Reactions

Reaction	Product Half-Life	Gamma-Rays Measured (MeV)	Branching Fraction
$^{63}Cu(n,\gamma)^{64}Cu$	12.8 h	0.511 x 0.511 coincidence	18.5%
$^{115}In(n,\gamma)^{116m}In$	54.0 m	2.391 - 2.529 doublet	86.0%
$^{64}Zn(n,p)^{64}Cu$	12.8 h	0.511 x 0.511 coincidence	18.5%
$^{24}Mg(n,p)^{24}Na$	15.00 h	1.369 x 2.754 coincidence	99.85%

In order to quantitatively determine unknown transuranic element concentrations, isotopes, or chemical composition from a measurement of induced activities in passive metal monitors, it is necessary to calibrate the technique with known quantities of transuranic species under similar environmental conditions. This can be accomplished by constructing mock-up facilities virtually identical to the system being monitored and spiked with known quantities of transuranic elements, by making rough mock-ups with "bugs" of transuranic elements in strategic locations, or by monitoring an analogous field system that has been sampled and analyzed in the laboratory to determine its transuranic element composition. Although each of these techniques has been used successfully in this work, this paper will deal in detail with only one of the more versatile calibrations.

The major demand for this passive activation technique thus far has been for the determination of plutonium in soils, salt cakes, and waste tanks. Plutonium monitoring in soils and sediments is of interest where high level fission product liquid wastes have leaked from storage tanks or where low level process streams have deposited accumulations over the years. Determinations of plutonium concentrations in high level liquid wastes or salt cakes are of interest from a potential criticality standpoint. These field situations all have the common requirements that the monitoring must be done in situ from dry wells and in gamma-ray fields up to megarads per hour. These requirements mandate the use of the passive activation technique.

A calibration facility for the above applications has been constructed from a section of 4-foot (1.22 m) diameter corrugated culvert filled with local aggregate. A 6-inch (15.4 cm) diameter dry well has been installed 11.1 cm off center of the culvert, and five 2½ inch (5.9 cm) diameter source tubes are spaced on 12.7, 22.9, 34.9, 48.3, and 61.0 cm centers from the dry well. Figure 1 shows a top view of this soil calibration facility. Nominal 100 gram sources of plutonium oxides and fluorides of known isotopic composition are placed in the source tubes, and the metal monitors are exposed to the neutron flux in the dry well. Induced activities are determined by gamma-ray spectrometric methods. The plutonium in each source tube is assumed to be uniformly distributed throughout a spherical shell of soil having a mean radius equal to the source tube-dry well separation distance. By normalizing the induced activities from each spherical shell to the plutonium concentration in that shell and summing over all significant shells, the relationship between induced activities and an "infinite" source of uniformly distributed plutonium is determined. This method of calibration has been verified experimentally for gamma radiation. A germanium diode "down well" probe was similarly calibrated using various gamma-ray sources, normalizing the data to each spherical shell, and summing all significant shells.[1] Subsequent calibrations of this diode in a large 4π facility filled with uniformly spiked soil yielded identical efficiency factors. The calibration of neutron-emitting isotopes is completely analogous.

RESULTS

For the sake of brevity, only a single calibration experiment in this facility will be discussed in detail. However, the results of all other calibrations performed to date are given. At this time, only calibrations in dry soil (1.1% moisture content) have been completed. However, it is planned to repeat these experiments at various soil moisture levels up to saturation values. Figure 2 shows the thermal neutron flux as measured in the dry well by copper monitors for a 98.02 gram plutonium oxide source having the isotopic composition of 0.588% ^{238}Pu, 72.659% ^{239}Pu, 19.373% ^{240}Pu, 5.427% ^{241}Pu, 1.907% ^{242}Pu, and 0.866%

[1] H. L. Nielson, N. A. Wogman and R. L. Brodzinski, Nucl. Instrm. Methods 143, 385 (1977).

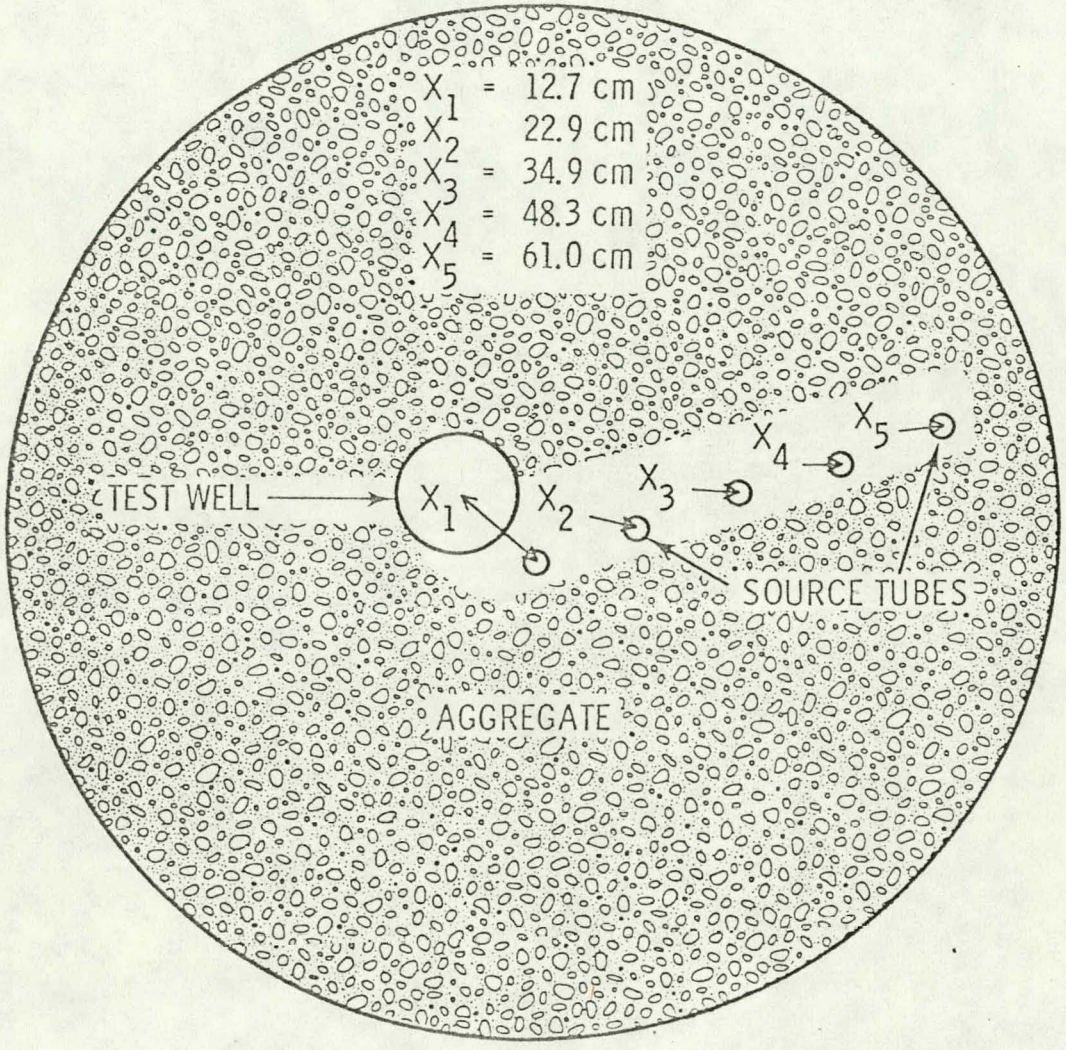

FIGURE 1. TOP VIEW OF SOIL CALIBRATION FACILITY FOR PASSIVE TRANSURANIC NEUTRON MONITORING SYSTEM

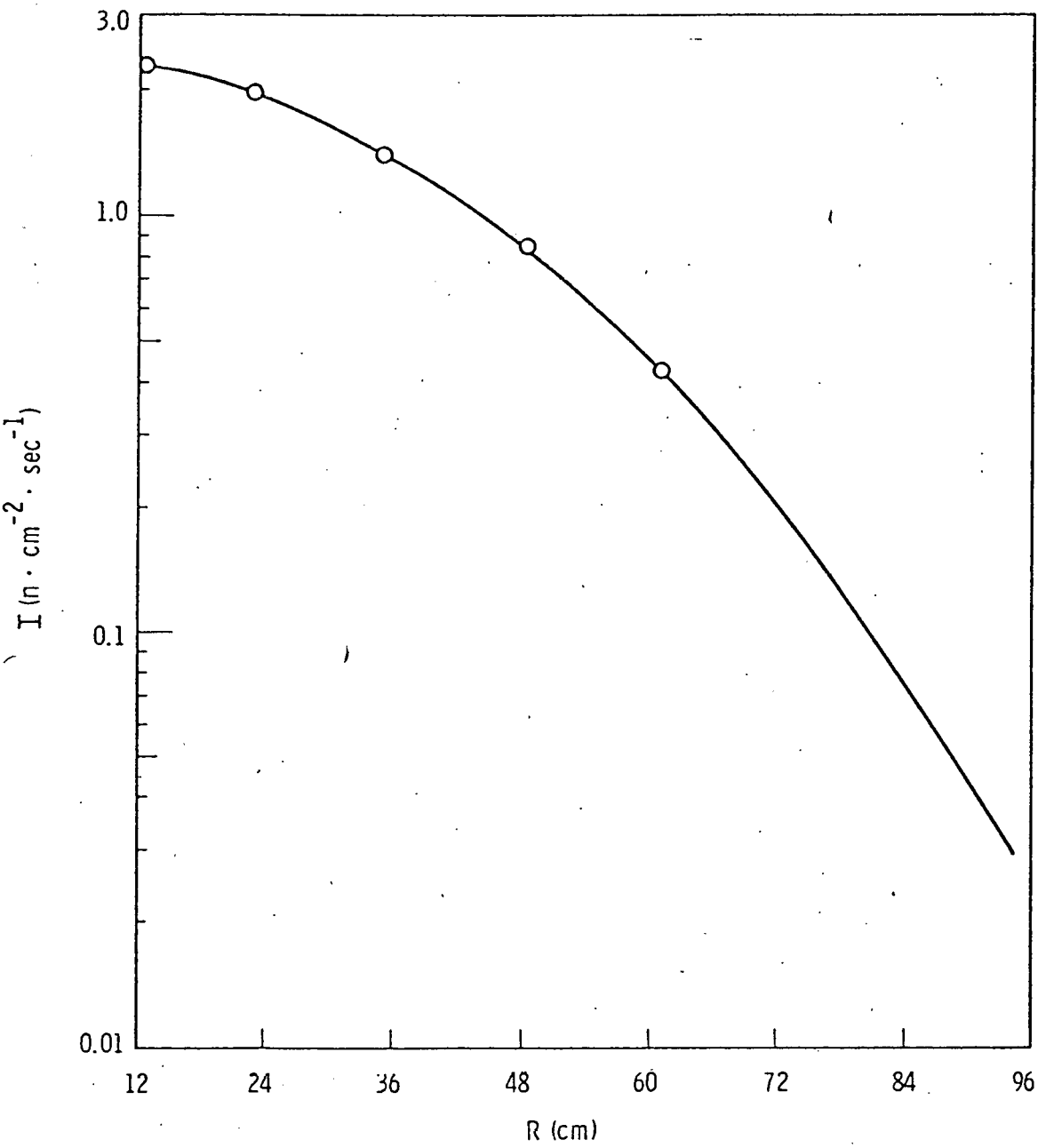

FIGURE 2. THERMAL NEUTRON FLUX FROM A PLUTONIUM OXIDE SOURCE MEASURED THROUGH VARIOUS THICKNESSES OF DRY SOIL

^{241}Am placed in each of the source tubes. In this experiment, a significant contribution to the total induced activity was obtained at the maximum source-monitor separation available, and it was necessary to extrapolate the data to a distance of 112 cm before the contribution became less than 1% of the total. This extrapolation, which amounted to 20% of the total integrated value, was necessary due to the low moderating capacity of the dry soil and hence the long range of the fast neutrons. Extrapolation is not anticipated to be necessary at higher moisture levels where the moderating ability of the soil will be greatly increased. The zinc monitor required a similar extrapolation which amounted to only 1.7% of the total integrated value. No extrapolation was required for the magnesium monitor data because very few collisions are required to reduce the neutron energy below the reaction threshold.

For these calibrations, the monitors were all 15.24 cm diameter discs, 3.175 mm thick for copper and 6.35 mm thick for zinc and magnesium. In practice, the geometry of the monitor can be tailored to specific needs or requirements with no adverse effects. The only necessary consideration, which has been included here, is the amount of self shielding presented by the monitor. An experiment offered in proof incorporated the exposure of a copper disc "facing" a plutonium source in the nearest position and another disc "edge on" to the same source. Both discs yielded identical neutron fluxes which reflects the isotropic nature of the multiply scattered neutrons. Self-shielding corrections are normally not required for zinc and magnesium monitors due to the low macroscopic cross sections for fast neutron interactions.

Thermal neutron flux values are readily calculable from a determination of the quantity of reaction products in the monitor, a knowledge of the cross section, and application of standard radioactive growth and decay equations. The situation is somewhat more complex for the zinc and magnesium reactions, however, since the measured activities are generated by a spectrum of neutron energies at widely varying cross sections. Therefore, it is most useful to determine the average product of the fast neutron flux and the cross section normalized to a unit mass of monitor material. Such calibrations are determined from saturation activity data and are reported as the number of reactions per second per gram.

It was found that the most meaningful method of expressing plutonium concentration in the soil, activity per unit volume, also produced the most consistent relationship between sources of different isotopic compositions. In the second column of Table II, the normalized thermal neutron fluxes as measured by the copper monitors are given for one plutonium fluoride source and three plutonium oxide sources of different isotopic composition. Also shown in the same column are two thermal flux values as measured by the indium monitors for verification of the reliability of the technique. Two epithermal neutron fluxes, obtained by enclosing the monitor discs in cadmium blankets during exposure, are also given. It should be recognized that the thermal/epithermal ratios are related to moderating capacity (moisture level) of the soil and thus can be used to determine the appropriate set of calibration curves in situations where soil moisture levels cannot be determined directly. In the third column, the activity normalized reaction rates for zinc and magnesium monitors are given, and in the last column, the magnesium reaction rates are normalized to the ^{240}Pu concentration since this is the only isotope which contributes to reactions in this monitor.

Agreement between the three plutonium oxide sources is excellent and average values for each monitor are shown in Table II. Similarly, the agreement between the oxide and fluoride source data is consistent with the predicted (α,n) neutron production rates in the two compounds except in the case of the magnesium monitor data which should be independent of chemical composition since only spontaneous fission neutrons contribute to this reaction. As can be seen in the table, this plutonium fluoride source yields approximately a factor of three more high energy neutrons than the oxide sources, a phenomenon which is real and remains unexplained.

DISCUSSION

This technique has a practical detection limit of $\sim$8 nCi/cm^3 for monitoring transuranic oxides with even lower values measurable in certain circumstances. A word of caution regarding background measurements is advisable here. The normal cosmic-ray neutron flux at sea level is $\sim$0.017 cm^{-2}sec^{-1}, and a piece of copper taken from the laboratory shelf will

Table II. Neutron Reactions in Metal Monitors for Plutonium Sources in Soil Having 1.1% Moisture Content

Source and Monitor	Thermal or Epithermal Flux ($n\ cm^{-2}sec^{-1}/nCi\alpha cm^{-3}$)	Fast Neutron Reactions ($g^{-1}sec^{-1}/nCi\alpha cm^{-3}$)	Fast Neutron Reactions ($g^{-1}sec^{-1}/mg^{240}Pu\ cm^{-3}$)
PuF_4: (8.84% ^{240}Pu)			
Cu	$(2.30 \pm 0.29) \cdot 10^{-3}$		
In	$(2.29 \pm 0.39) \cdot 10^{-3}$		
Cu(Cd)	$(4.15 \pm 0.61) \cdot 10^{-4}$		
In(Cd)	$(4.53 \pm 0.54) \cdot 10^{-4}$		
Zn		$(5.52 \pm 0.65) \cdot 10^{-8}$	
Mg		$(2.29 \pm 0.65) \cdot 10^{-9}$	$(2.11 \pm 0.60) \cdot 10^{-3}$
PuO_2: (5.691% ^{240}Pu)			
Cu	$(4.5 \pm 1.0) \cdot 10^{-5}$		
In	$(3.64 \pm 0.95) \cdot 10^{-5}$		
Zn		$(1.03 \pm 0.22) \cdot 10^{-8}$	
Mg		$(7.3 \pm 4.6) \cdot 10^{-10}$	$(9.8 \pm 6.2) \cdot 10^{-4}$
PuO_2: (16.789% ^{240}Pu)			
Cu	$(6.8 \pm 1.4) \cdot 10^{-5}$		
Zn		$(1.00 \pm 0.21) \cdot 10^{-8}$	
Mg		$(8.8 \pm 4.3) \cdot 10^{-10}$	$(7.6 \pm 3.7) \cdot 10^{-4}$
PuO_2: (19.373% ^{240}Pu)			
Cu	$(6.0 \pm 1.2) \cdot 10^{-5}$		
Zn		$(1.01 \pm 0.22) \cdot 10^{-8}$	
Mg		$(6.7 \pm 2.8) \cdot 10^{-10}$	$(7.6 \pm 3.2) \cdot 10^{-4}$
Weighted Ave. PuO_2			
Cu	$(5.8 \pm 1.5) \cdot 10^{-5}$		
Zn		$(1.01 \pm 0.15) \cdot 10^{-8}$	
Mg		$(7.8 \pm 1.7) \cdot 10^{-10}$	$(8.4 \pm 1.8) \cdot 10^{-4}$

have a measurable intrinsic ^{64}Cu activity. If this same piece of copper is kept 10 meters underground for a day, the ^{64}Cu activity will be substantially reduced. It is therefore necessary when making very low level transuranic element determinations to derive all background corrections based on monitors exposed under similar conditions but in transuranic-element free environments.

In certain instances, this technique can yield information on the chemical composition of the transuranic elements (i.e., metal, oxide, fluoride, etc.) by the copper/zinc, copper/magnesium, or zinc/magnesium ratios. Similarly, it may be possible to determine the isotopic composition of a transuranic source by the magnesium/copper or magnesium/zinc ratios.

This technique has been successfully applied to the quantitative determination of plutonium concentrations in a variety of field situations where other methods of analysis were impossible because of inaccessibility, excessive gamma radiation, or real potential for equipment contamination. Plutonium concentrations have been measured in high level fission product waste storage tanks, in cribs and trenches which have received plutonium-bearing process effluents, and in deep sediments which received accidentally released high level fission product wastes. A plutonium inventory has been performed on a building contaminated during a fire as a prelude to decontamination efforts. Process and fabrication hoods and waste barrels have been examined and inventoried with remarkable accuracy. In these latter cases, an element of directionality was introduced by "backing" the monitors with cadmium-lined moderating collimators. In one waste barrel, a single slug of plutonium was located to with 3 cm, identified as plutonium oxide, and the weight was predicted to within 1%. In perhaps the most spectacular success of the technique to date, a hidden "pocket" of plutonium was found in a recess in a channel iron support beam of a fabrication hood being

decommissioned. The quantity and location of the plutonium was predicted, and the data indicated it to be ∿75% oxide and ∿25% metal. When the hood was breeched, a pile of badly oxidized plutonium metal shavings which had been generated over a period of years by the milling machine in the hood was recovered from where they had fallen through a crack and collected. The quantity recovered was ∿12% greater than predicted and from with 8 cm of the identified location.