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ABSTRACT

This paper presents a method for the rapid characterization of gamma-ray-emitting radioisotopes in large samples of earth-like materials: concrete shielding blocks in this case. Active regions are identified with a sensitive radiation-survey instrument and then examined in detail with a high-efficiency lead-shielded Ge spectrometer. Naturally-occurring gamma-ray emissions from the decays of uranium, thorium, and potassium are used to calibrate the spectrometer. A simple relationship exists between the observed counting rate in a characteristic gamma ray and the activity in the block. This method, taking only tens of minutes per sample at the nano-Curie/gram sensitivity level, replaces much of the expensive coring and laboratory analysis methods needed otherwise.

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INTRODUCTION:

Following many years of productive work, the SuperHILAC and Bevalac accelerators at Lawrence Berkeley National Laboratory were closed, leaving thousands of concrete shielding blocks available for reuse or disposal. The process history of these blocks as shielding precludes free release pending radiological characterization.

For the first half of its nearly 40 year lifespan, the Bevalac accelerator ran beams of protons at energies as high at 6.2 GeV. (Subsequent heavy-ion beams contributed small amounts of additional activation.) Interactions of these primary-beam protons on targets and machine components generated secondary particles, predominantly neutrons with long mean-free-paths, that activated parts of the shielding. As detailed in References 1 and 2, the dominant process by which accelerator shielding blocks are activated involves capture by thermal neutrons. The isotopes ^{46}Sc, ^{59}Fe, ^{60}Co, ^{65}Zn, ^{134}Cs, ^{152}Eu and ^{154}Eu are produced this way. To a lesser extent, fast neutron activation accounts for the production of ^{22}Na and ^{54}Mn. As discussed in References 1 and 2, thermal-neutron activation may increase slightly with depth, reaching a maximum several cm into the concrete. Fast neutron-activation, on the other hand, is at a maximum at the surface. Activities fall off approximately exponentially after the maximum with a half-thickness on the order of 10 cm. The very few blocks at 0° to GeV beams could have very different distributions. Since most of the high-intensity light-ion runs at the Bevalac occurred prior to 1971, only the longest lived products remain, namely ^{60}Co and ^{152}Eu.

A large number of Bevalac shielding blocks have been examined with a 2.54 cm diameter x 2.54 cm thick NaI survey instrument and found to exhibit surface activities over a range from several μR/hr above background to over 1000 μR/hr above background. Subsequent to this survey, several examples of blocks were examined with a high-efficiency lead-shielded Ge spectrometer to identify and quantify the activity. This spectrometer can easily distinguish natural activity (U, Th, and ^{40}K) from added activity (mainly ^{22}Na, ^{60}Co, and ^{152}Eu). The natural activity may be used to calibrate the detector for the quantification of added activities. Following calibration, this technique makes most coring and laboratory analysis unnecessary.

THEORY:

As shown in Figure 1, the detector views a volume of the sample defined by the solid angle of the lead collimator and a depth determined by the gamma-ray attenuation in the sample. Each unit volume contributes gamma rays that may be detected in a lead-shielded detector located next to the slab. For an activity of A_V Disintegrations/minute/unit volume, and a fraction (BR) of disintegrations that produce a particular characteristic gamma ray, A_V x BR gamma rays at the energy E of the characteristic gamma ray are emitted per unit volume. The observed count rate in the detector in counts/minute (CPM) is:

$$A_V \times BR \times \sum_{i=1}^{\infty} \mu(Z,E,\rho,r_i) \times e(E,\Omega) \times \Delta V_i = CPM$$

where $\mu(Z,E,\rho,r_i)$ is the gamma-ray absorption in a thickness r_i, (Z is the effective atomic number, E is the gamma-ray energy, and ρ is the density.) $e(E,\Omega)$ is the detector efficiency. (Dependent on gamma-ray energy E and detector solid angle Ω, and the sum extends over all elements ΔV_i. Note that $e(E,\Omega)$ depends on Ω, and not on the distance between the sample and the detector face.

Converting to disintegrations/gram: $A\,\rho = A_v$

$$A \times BR \times \sum_{i=1}^{\infty} \mu(Z,E,\rho,r_i) \times e(E,\Omega) \times \rho \times \Delta V_i = CPM$$

$$A \times BR \times \varepsilon = CPM$$

where ε is, essentially, the absorption- and efficiency-weighted mass of the sample examined and is determined by comparing the CPM observed with the activity determined by measuring the activity of a core sample in the laboratory.

The activity A is determined by:

$$A = \frac{CPM}{\varepsilon \times BR} \quad \text{in units of disintegrations/minute/g}$$

$$\text{or } A = \frac{CPM}{2.22 \times \varepsilon \times BR} \quad \text{in units of pCi/g}$$

PROCEDURES:

Part I: Determination of intrinsic activity by laboratory analysis

The intrinsic activity of the bulk concrete is determined by gamma-ray analysis of kg-sized samples obtained by drilling 2.5 cm diameter holes into the concrete. Typically 6 holes 15 cm deep were used to obtain enough material. The hammer-drill used pulverized the material into a fine powder which was collected in a plastic bag. The contents were transferred to a Marinelli beaker and counted for ≈1 day on a calibrated 30% p-type germanium spectrometer at the LBNL Low Background Facility. Concentrations of uranium, thorium, and potassium were determined for various samples of normal- and high-density concrete.

Part II: Detector Calibration

Prior to drilling, a lead-shielded 80% Ge spectrometer was used to measure activity in-situ at the same sites as the core samples were taken. (This procedure, shown in Figure 2, will be described more fully in the next section.) The detector response ε is determined by comparing the observed count-rate (CPM) from characteristic gamma-rays from U, Th, and K observed in-situ with the U, Th, and K activities (A) determined via laboratory analysis as described above.

$$\varepsilon = \frac{CPM}{2.22 \times A \times BR}$$

Bevalac shielding blocks are made of two different kinds of concrete having different physical and radiological properties. Normal-density concrete, specific gravity ≈2.4, and high-density concrete, typically containing iron-ore aggregates of specific gravity ≈3.5 were used in different parts of the shielding. Normal-density concrete contains the natural decay products from U, Th, and 40K at levels ≈10 times higher than high-density concrete.

Part III: In-situ measurements

The spectrometer used consisted of an ORTEC 80% p-type germanium detector mounted in a low-background cryostat and powered by an ORTEC Nomad™ system. The 8192 channel analog-to-digital converter and internal memory accumulated and stored data for read-out via an attached lap-top PC. Resolution was 1.9 keV @ 1333 keV. The spectrometer shielded with 5-10 cm of lead (5 cm in the early measurements and 10 cm in later measurements) and mounted on a cart as shown in Figure 2. The cart is positioned next to the block for measurements. Note that the distance between the spectrometer and the block is not critical as long as the field of view from the detector sees only the sample.

Characteristic gamma rays from naturally-occurring radionuclides over the energy interval 238-2614 keV were used to generate detector response curves as shown in Figure 3. The response to characteristic gamma-rays from added activities were determined by interpolation. The curve from Figure 3 was used to generate a table relating the observed CPM in a characteristic line to activity. This table was used to determine added activities.

Most of our observations involved counting times on the order of 1 day, with the exception of background counts (with an additional 5-10 cm lead shielding over the front face of the detector) which lasted several days. With these counting times, we were able to measure activities <0.01 pCi/g for ^{60}Co, <0.1 pCi/g ^{152}Eu, and <0.2 pCi/g ^{22}Na. For activities in the nCi/g range, counting times of only a few minutes are required. Figure 3 shows a spectrum from a sample containing ≈10 pCi/g of ^{60}Co, adjusted to represent a 5 minute counting time.

Part IV: Comparison with a survey meter

The naturally-occurring radioactivity in concrete can easily provide surface dose rates of several micro-R/hr above background, rates at which a survey meter with a 2.54 cm diameter x 2.54 cm thick NaI crystal can easily measure. Such a meter can be used to survey large areas in short times. For ^{60}Co in high-density concrete, an empirical relationship between dose measured with the survey meter and activity was determined:

Dose rate (μR/hr) = 1.7 * Activity (pCi/g)

For example, 10 pCi/g of ^{60}Co provides approximately 17 μR/hr reading on the survey instrument, an amount easily detectable above the several μR/hr background in the region where we did our measurements. Thus, release criteria of 10 pCi/g for ^{60}Co, for example, can be determined reasonably well with a survey meter. Readings <10 μR/hr, uniform over the surfaces, are probably from naturally-occurring radionuclides alone. Readings above 20 μR/hr, particularly if they are not uniform over the surfaces, almost certainly indicate manmade isotopes. Criteria such as these, combined with judicious application of the detailed characterization described in Part III, can provide inexpensive and sufficient characterization of large samples, such as concrete blocks, for disposition.

CONCLUSIONS:

The combination of survey-meter readings and the detailed characterization techniques described above, provide inexpensive and thorough characterization of concrete blocks for reuse or disposal. Following detector calibration, The characterization technique is simple enough that a technician can be trained to perform in-situ measurements in tens of minutes per sample, including analysis. On-the-spot decisions regarding the suitability of a sample for transportation or disposal can then be made. Spectra are retained as permanent records of the radioisotopes in the block.

REFERENCES:

1) G.C. MOELLER and R.J. DONAHUE, "Induced Radioactivity in Bevatron Concrete Radiation Shielding Blocks," LBL-36072, Lawrence Berkeley National Laboratory, Berkeley, CA. 94720

2) KEN-ICHI KIMURA, TOSHIO ISHIKAWA, MASAHARU KINNO, AKIRA YAMADERA, and TAKASHI NAKAMURA, "Residual Long-lived Radioactivity Distribution in the Inner Concrete Wall of a Cyclotron Vault," Health Physics V67, #6, (1994) 621-631

FIGURE CAPTIONS:

Figure 1: Diagram showing the determination of detector response.

Figure 2: Diagram of the lead-shielded detector used for in-situ measurements.

Figure 3: The detector response ε for normal concrete.

Figure 4: A spectrum showing 5 minutes of counting for a sample with 10 pCi/g concentration of ^{60}Co.

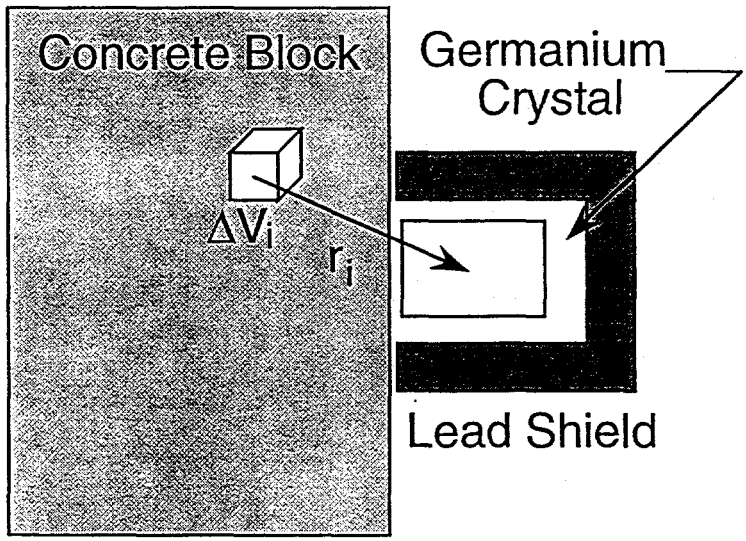

Figure 1

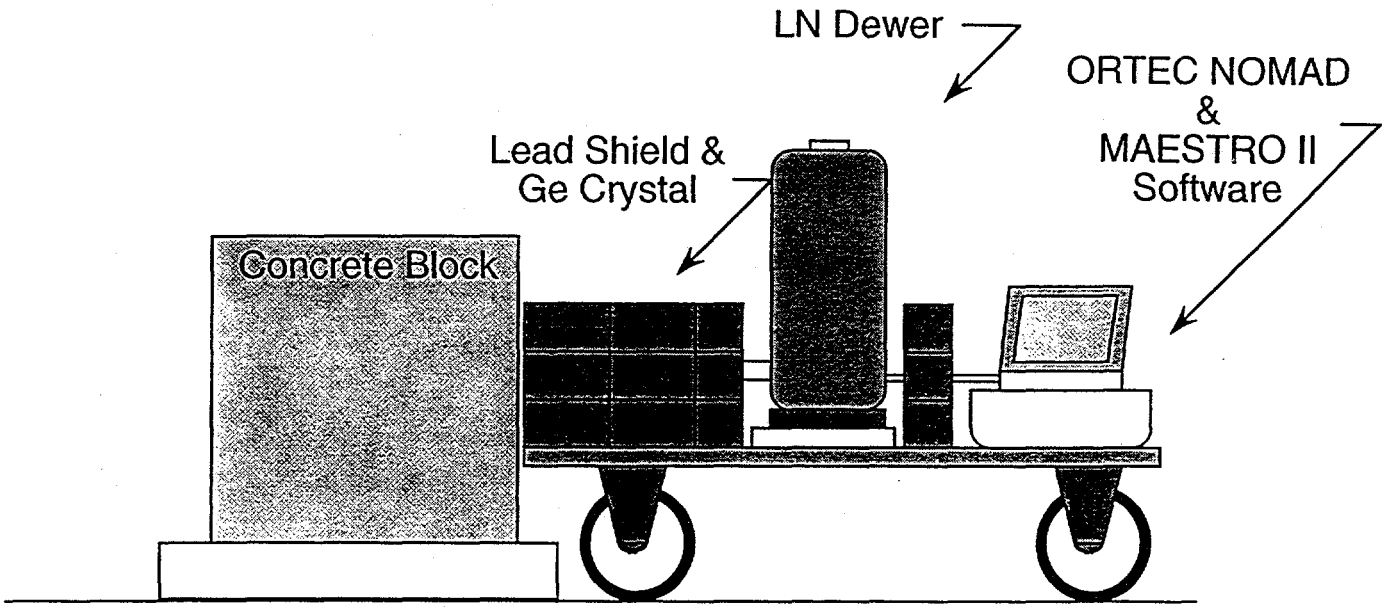
LN Dewer
ORTEC NOMAD
&
MAESTRO II
Software
Lead Shield &
Ge Crystal
Concrete Block

Figure 2

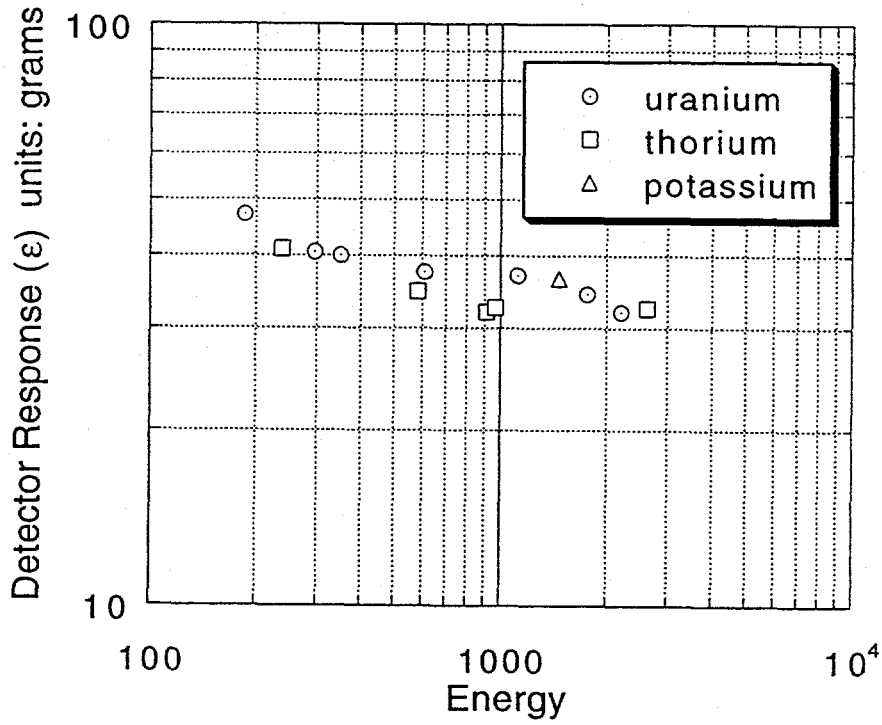
Detector Response (ε) units: grams
100
10
100
1000
10^4
Energy
uranium
thorium
potassium

Figure 3

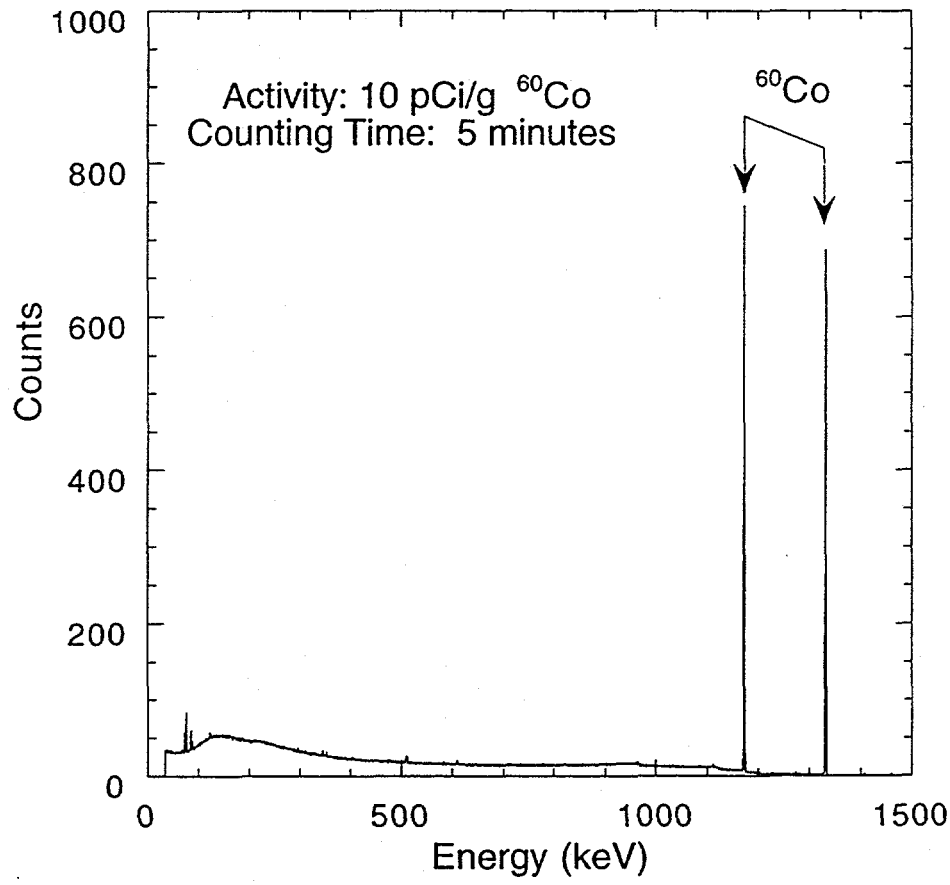
Activity: 10 pCi/g ^{60}Co
Counting Time: 5 minutes
^{60}Co
Counts
1000
800
600
400
200
0
0
500
1000
1500
Energy (keV)

Figure 4