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## EARTH MATERIALS FOR LOW-BACKGROUND RADIATION SHIELDING

Master of Sciences  
(Engineering Sciences)

Harold Tollenberg

Geological Engineering

Bldg 72

UC Radiation Laboratory

Berkeley

(Chairman)

Thesis Committee

### ABSTRACT

A search was conducted for materials for construction of a low-background counting room enclosure at the Lawrence Radiation Laboratory, University of California, Berkeley. Gamma-ray spectrographic analyses of sources of concrete aggregates in Central California indicate that ultramafic rocks are best suited for use as low-background aggregates.

It is expected that the background level to be achieved inside the enclosure will be 1/100 that presently existing at the site. To achieve this the counting room enclosure must not only serve as a shield but also have inherently low radioactivity. Material with a gamma-ray spectrometer counting rate  $\leq 0.004$  counts/minute/gram would be adequate for this purpose.

$^{238}\text{U}$ ,  $^{232}\text{Th}$ , their decay products, and  $^{40}\text{K}$  are the principal contributors to the gamma-ray activity of most earth materials. The U, Th, and K concentrations vary with the acidity of igneous rocks, being practically nil in the ultramafic rocks and highest in the igneous rocks. In carbonate rocks U is the principal radioactive contaminant and is most abundant when carbonaceous material is present. In sediments U and Th are concentrated with the heavy minerals; zircon, apatite, cassiterite, sphene, and rutile.

Samples of possible sources of low-background aggregate were collected and subjected to gamma-ray spectrographic analysis on a 100-channel analyzer. Data obtained for each sample enabled computation of total counting rate in counts per minute per gram, and the individual contributions (in counts per minute) of U, Th, and K to the total gamma-ray spectrum.

Standard portland cement and concrete-aggregate material obtained locally were found to be too radioactive to be suitable for low-background material. Local sources of sand also contained too much radioactivity for use in low-background shielding.

ABSTRACT (Cont.)

H. Wollenberg

Contamination of low-background rocks is derived primarily from adjoining higher background rock and dikes intruding the deposits.  $Co^{137}$  fallout is a source of surface contamination on exposed material.

Possible sources of low-background material which were examined included serpentized ultramafic rocks in the Coast Ranges and Sierra Nevada foothills, the Calera, Calaveras, and Gabilan limestones and dolomites, vein and placer quartz of the Sierran Foothills, scapolite deposits near Dutch Flat, California and Battle Mountain, Nevada, talc from the vicinity of Keeler, California, boron minerals from the Kramer area, California, and diatomite from Napa County.

Recommended materials are listed in order of their feasibility:

1. Freshly broken, serpentized ultramafic rock from the Sonora area,
2. Dolomitic limestone from Salinas,
3. Pure quartz from the Sierra foothills.

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## THE PROBLEM

Present-day facilities for counting very low levels of gamma-ray radioactivity or moderately radioactive materials in a nearly "zero" background environment usually consist of small enclosures of lead blocks or fairly large steel enclosed rooms. The Health Physics Department of the Lawrence Radiation Laboratory, Berkeley, California is presently studying the applicability and availability of natural earth materials for use in large low-background counting enclosures. The purpose of the enclosure is essentially two-fold. Primarily it must possess adequate thickness and density so that gamma rays are attenuated and fast neutrons thermalized and captured. Therefore, material with fast neutron attenuation characteristics should be present. Characteristics of a neutron shield and capture of thermal neutrons by boron are discussed more fully later in this report. Almost of equal importance is the criterion that the material composing the enclosure must itself be of adequately low radioactive background.

The present natural gamma-ray background at the site of the proposed enclosure to be constructed at Berkeley measures about 170 counts per second (0.00815 mr/hr) on a portable scintillation counter utilizing a sodium iodide crystal. It is hoped that a background level of 1/100 of this level can be achieved inside the proposed enclosure.

### PROPOSED STRUCTURE

The proposed low-background counting room (or "cave" as such enclosures are called) will have inside rectangular dimensions of 21' x 10' x 10'. The four walls and roof will have a thickness of 5'. A reinforced concrete slab has already been poured for the floor, but due to a high radioactive background in this slab it must be covered by an adequate thickness of low-background shielding material, preferably material similar to that composing the walls and roof. Steel or aluminum reinforcing may be used in the walls and roof provided that the metal is kept at least 3" away from the interior of the enclosure. Neglecting the steel, the total volume of low-radioactive-background material necessary for such a rectangular enclosure amounts to approximately 310 cu yd. The decision as to the type of construction, e.g. block or pour, has not yet been reached. Specifications call for the material in place to have a specific gravity of not less than 2.0.

## THEORETICAL CONSIDERATIONS

### I. Radioactivity of Shielding Material

Considering a spherical idealized shield, Mevill (1951) has stated that the number,  $n$ , of gamma rays/cm<sup>2</sup>/sec from the shield is roughly equal to  $q/\mu'$ , where  $q$  is the number of gamma rays from the source/cm<sup>2</sup>/sec and  $\mu'$  is the gamma-ray absorption coefficient in cm<sup>2</sup>/gm. Therefore, the more effective the shield material the smaller is  $n$ . Within an energy range of from 0.05 to 1.46 million electron volts (MeV) a value of  $\mu'$  of about 0.03 cm<sup>2</sup>/gm can be used for materials with low and medium atomic number.

(2). For high Z materials such as lead, a  $\mu'$  value of 0.07 cm<sup>2</sup>/gm is used. A suitable shield material should have an activity of  $10^{-14}$  curies/cm<sup>3</sup> of lead. Therefore, using the conversion factor that 1 curie =  $3.7 \times 10^{10}$  disintegrations per second, a material with a gamma-ray efficiency of 0.01 and a background of 100 counts/minute would require a shield of lead 10 cm thick to be adequate for low-background shielding.

### II. Radioactive Elements

Natural radioactivity in the earth's crust varies markedly with rock types. The principal radioactive elements which affect the background radiation in rocks are potassium-40, uranium, and thorium and their associated decay products.

#### A. Potassium

The potassium isotope of mass 40 and an isotopic abundance of 0.0117 percent is radioactive. Eleven percent of potassium-40 decays to argon-40



with emission of a gamma ray with an energy of 1.46 Mev. The long half-life of  $K^{40}$ ,  $1.3 \times 10^9$  years, plus the presence of some potassium in almost all of the earth's crustal material makes this element one of primary concern when evaluating the radioactivity of earth materials. The ratio of  $K^{40}$  to  $K^{39}$ , 1/8500, is considered to be fairly constant throughout the earth's crust. Eighty-nine percent of  $K^{40}$  decays to  $Ca^{40}$  with emission of a 1.3 Mev  $\beta$  particle. This  $\beta$  has no influence on a counting device with a minimally-shielded crystal.

The principal potassium-bearing minerals are the feldspar orthoclase, the micas muscovite, and biotite. Hornblende and plagioclase may contain up to 1% potassium. The clay mineral, illite, also has potassium as a principal constituent. Though not a principal constituent of many clay minerals, montmorillonite, potassium may be incorporated by cation exchange with aluminum.

In igneous rocks the concentration of potassium varies roughly with the abundance of silica, potassium being more prevalent in the acidic igneous rocks such as granite than in the ultramafics (peridotite, dunite, serpentine). Rankama and Sahama (1950) show that due to the large ionic radius of potassium (1.33A), and its twelve-fold coordination with respect to oxygen, the element is excluded from the early-formed crystallites of magmatic differentiation and becomes enriched in residual melts and solutions. Potash feldspars are therefore characteristic of the late crystallites, the igneous rocks, most abundant in silica.

Abrams (1954) gives the following concentrations of potassium for various igneous rock types:

In granite the range is from 2% to 6% K.

In basalts potassium concentrations vary with individual flows (potassium being sensitive to fractionation in a basaltic magma). A variation between 0.65% and 1.4% potassium was found by Abrams et al. (1952) in Columbia River basalt. Daly (1933) measured 0.37% potassium in oceanic basalt and 0.65% in plateau basalt.

Peridotite, pyroxenite, and dunite and their serpentinized forms have the lowest potassium concentrations of the igneous rocks, about 10 ppm.

The potassium content of sedimentary rocks depends largely upon the relative amounts of the feldspars, micas, and clay minerals which partially comprise the mineral-aggregate sediments. A sandstone derived from a close granitic source would contain an appreciable amount of feldspar and therefore exhibit a potassium content roughly equal to that of its source granite. A pure quartz sandstone derived from a quartzitic source, or a sandstone at a great enough distance from its granitic source so that the feldspars have been removed during the transport, would contain a relatively low potassium concentration.

Shales or argillaceous sediments with an abundance of mica and clay minerals contain appreciable potassium. Limestones are generally low in alkalis though the presence of the authigenic feldspars and some argillaceous material filling cracks in the limestone (hydroxides developing from detrital clays during diagenesis of the limestone) may increase the percentage of potassium over that of pure limestone. The potassium contents of the broad groups of sedimentary rocks are listed:

Shales and argillaceous sediments	3%
Sandstones	1%+
Limestones	tenths of 1%

## B. Uranium and Thorium

Uranium-238 (half-life  $4.49 \times 10^9$  years) and thorium-232 (half-life  $1.39 \times 10^{10}$  years) and their decay products are also major contributors of radioactivity in the earth's crust. The higher energy gamma rays of the uranium and thorium decay series are of greatest concern in the problems of low-background shielding. Due to the presence of lower energy gamma rays of uranium and thorium the overall uranium and thorium contribution to the total gamma-ray spectrum is appreciably higher than the contribution of  $K^{40}$ . However, the predominance of the lower energy uranium and thorium gammas tends to minimize the importance of uranium and thorium with respect to potassium in the low-background shielding problem.

### 1) Uranium and Thorium in Igneous Rocks

As with potassium, the uranium and thorium contents in igneous rocks vary with the percentage of silica. Along with being potassium rich, the granites are usually uranium and thorium rich while the ultramafics are quite lean in all three elements. Keevil (1944) gives the probable values for the thorium-uranium ratio in igneous rock as 3.0 to 3.5. Evans and Williams (1935) show an almost linear relationship of  $K_2O$  with uranium concentration in the Lassen Peak region volcanics. J. A. S. Adams (1954) attributes the nearly linear relationship to the fact that both uranium and potassium were systematically concentrated in the liquid phase of the magma as crystal fractionation proceeded. Thus uranium and

potassium oxide were, to a degree, excluded from the common minerals formed during crystallization of the Lesser magmas. Uranium and potassium oxide do not substitute readily for other common ions due to the different space requirements of potassium and uranium. Their ionic radii and coordination numbers with respect to oxygen do not permit their ready substitution with other common ions. This would suffice as a partial explanation of the higher uranium and potassium concentrations in the more acidic igneous rocks.

Larsen et al. (1956) from their study of the uranium concentrations of three Mesozoic batholiths of the western United States show that where fractional crystallization is assumed to have been the major factor in magmatic differentiation uranium is enriched in the youngest rocks, these being high in  $\text{SiO}_2$  and  $\text{K}_2\text{O}$  and low in  $\text{CaO}$  and  $\text{MgO}$ . A maximum enrichment of greater than 20 ppm is found in differentiates very poor in  $\text{CaO}$ . From chemical analysis of samples from the Southern California batholith Larsen et al. show that the uranium content of the common rock-forming minerals increases with total uranium in the bulk rock as the percentage of silica increases in the rock.

Davis and Hess (1949) have studied the concentration of radium in ultramafic rocks. Average values for igneous rocks range from  $0.01 \times 10^{-12}$  gm/gm of radium in ultramafics to  $1.0 \times 10^{-12}$  gm/gm in felsic rocks. The absence of uranium (parent element for the radium) in ultramafics is attributed to its strong concentration in residual liquids during magmatic differentiation with uranium being apparently excluded from the crystal structures of the early-formed minerals, olivine, pyroxene, spinel, and

## COMMERCIAL CONCRETE AND AGGREGATES

It was evident soon after commencement of this project that standard concrete materials obtained locally were too radioactive to be suitable for low-background material. A principal source of aggregate in the San Francisco Bay Area is the alluvial gravel beds in the Livermore Valley near Pleasanton. These gravels, composed of pebbles of red, white, and green cherts, glaucophane schist, and gabbro, are derived principally from the Franciscan formation. Samples of these various rock types collected from the stockpile of the Pacific Cement and Aggregate Company near Pleasanton all indicated too great a radioactivity for our use. Thus, the main commercial source of aggregate was ruled out. Table 1 recapitulates the sampling and gamma-ray spectrographic evaluation of some commercial aggregate sources in the San Francisco Bay Area.

Samples of portland cement produced locally also exhibited a background which precluded their use. It then became apparent that we should search farther afield geologically and geographically for sources of both cement and aggregate. In this respect requests were sent to the member companies of the Portland Cement Association and other cement companies in the United States and Canada for samples of their lowest alkali cements, chemical analyses, and any geological information pertinent to the source of the cement raw materials. Response of the cement industry was excellent. The process of evaluating the radioactivity of portland cement samples is now in progress. Results will be published at a later time.

The purpose of this report is to explain the factors leading to the choice of aggregate material for low-background radiation shielding.

TABLE 1. GAMMA-RAY SPECTROGRAPHIC EVALUATION, COMMERCIAL SOURCES OF AGGREGATES

RUN NO.	DESCRIPTION	COUNTING RATE counts/min./170	CONTAMINANTS PRESENT	% K	CONTRIBUTION TO TOTAL GAMMA-RAY SPECTRUM IN COUNTS/MINUTE		
					K	U	Th
855	Red chert, PCA Quarry	0.144	K, U, Th	0.16	22	124	18
848	White chert, PCA Quarry	0.240	K, U, Th	0.16	22	88	61
854	Green chert, PCA Quarry	0.182	K, U, Th	0.30	40	61	55
847	Gabbro, PCA Quarry	0.070	K, U, Th	0.11	15	28	5
663	"Basalito", Hapa	1.00	K, U, Th	1.78	205	183	375
703	"Pre-Alx" (Folton granite mixed with Franciscan gravel and sand)	0.376	K, U, Th	0.53	93	154	178
669	Elliot Pea Gravel (Franciscan)	0.336	K, U, Th	0.57	85	78	159
664	Folton Granite	0.589	K, U, Th	2.63	373	154	72

## RECOMMENDATIONS ON THE SOURCE OF AGGREGATE MATERIAL

Recommended sources of low-background material are listed in order of suitability for crushed aggregate

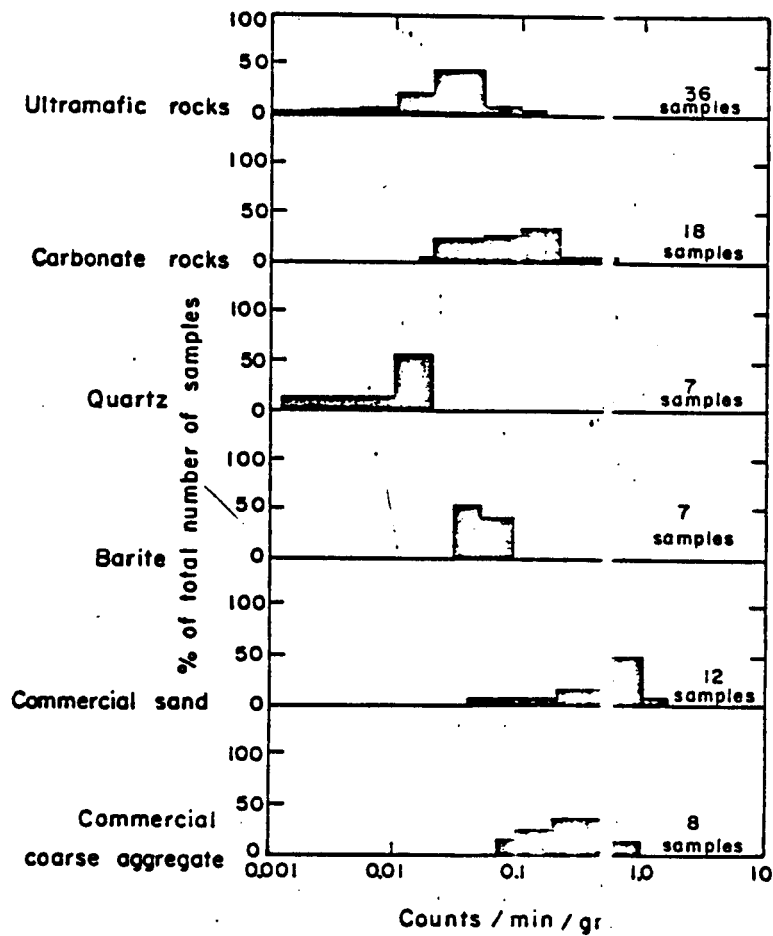
1. Ultramafic rocks of the Sonora area.
2. Dolomitic limestone from the Salinas area.
3. Pure quartz.

Amplification of these recommendations plus discussion of the feasibility of using barite and boron compounds as additives, and consideration of material to be used for the minus #4 mesh (sand) fraction will follow.

The frequency distribution plots in Figure 5 illustrate the radiation-background ranges of the possible sources of aggregate material. The ultramafic rock samples which exhibit background levels greater than 0.025 counts per minute per gram were collected at the surface and show contamination by Cs-137 fallout. It is safe to assume that fresh ultramafic material would exhibit an appreciably lower background level than the surface samples indicate.

### Ultramafic Rocks:

The overall low radioactive background exhibited by serpentinized rocks appears to hold regardless of the geographic location of the deposits. In individual deposits very little variation in background is observed (with instruments of present sensitivity) except where extraneous material from dikes or overburden has contaminated the ultramafic rocks. Geographically, the most competent ultramafic



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FIGURE 5. Frequency distribution of the occurrence of specific activities of samples.



4. Crushing facilities are cleaned out prior to crushing by, a) thorough washing, b) a preliminary run-through of the ultramafic material.

5. Transportation is in enclosed carriers. An attempt should be made to keep rain water or road dust from coming in contact with the aggregate material.

#### Carbonate Rocks:

There is a marked variation in radioactivity between deposits of carbonate rocks and within individual deposits. Therefore, a stringent quality-control program would be necessary to insure material of suitable low background. The Natividad dolomite quarry near Salinas produces the product with the lowest background of all carbonate rocks examined. However, within this deposit the background level varies by a factor of 10 (from 0.015 to 0.17 counts per minute per gram). Very careful portable counter scanning at the deposit coupled with close sampling and gamma-ray spectrographic evaluation of the dolomite as it is produced would be necessary if a material of sufficiently low background were desired.

#### Barite:

The presence of contaminating wall rock material adjacent to narrow barite veins as well as interlayered shale in bedded deposits necessitates that either careful hand picking or adequate heavy-media separation processes be used if a pure barite aggregate is to be attained. The lack of toughness of barite (a loss of greater than 60% on the L. A. abrasion test) precludes its use as a principal source of aggregate. The curve in Figure 6 gives an approximation of the

density achieved in a serpentine-gypsum-barite mix with varying amounts of barite.

Boron:

If the thermal neutron flux (due to moderation of external fast neutrons) deep within the shield is sufficient to produce gamma-rays close to the interior of the enclosure the addition of boron compounds may be desirable. However, the 5' wall thickness as well as the presence of sufficient hydrogen in the enclosure should suffice for neutron protection. For future enclosures located closer to neutron sources the utilization of boron compounds may be necessary. Henrie (1959) gives various concrete mixes for different neutron and gamma-ray shielding criteria.

Sand:

Since no local sources of sand are adequately low in radioactivity an imported product would be necessary. The Blackhawk silica sand from Ottawa, Illinois is the lowest background sand material tested. However, sand-sized crushed serpentine material can be obtained when the coarse aggregate is prepared. This material will exhibit a background level at least one-quarter that of the lowest silica sand. Therefore, it is recommended that the minus #4 fraction of the crushed serpentine aggregate be utilized as the sand constituent in the final mix. The minus #4 fraction of pure vein or placer quartz could also be used as the sand fraction of a low-background aggregate.

Quartz:

Pure quartz exhibits an adequately low radioactive background. Structurally, it possesses sufficient toughness to serve as a principal aggregate. An acceptable source of quartz should be freshly exposed material, either vein or placer quartz. Some quartz is presently accessible on old waste dumps in the Mother Lode region. The Basalt Rock Company of Nevada has indicated that freshly broken quartz from "Clear Cat", location unknown, is available. This latter source warrants further investigation.