

Crustal Radioactivity

B K Bhaumik

*Physics lab, Atomic Minerals Directorate for
Exploration and Research, Hyderabad*

Crust is the outermost layer of earth containing only about one percent of the mass of the whole earth. Molten mantle material cools and hardens to form rock largely of coarse crystalline type called granite.

Rock specimens emit gamma radiations due to their natural content of thorium ($^{232}\text{Th}_{90}$ present in 100 % abundance), uranium ($^{238}\text{U}_{92}$ present in 99.3 % and $^{235}\text{U}_{92}$ present in 0.7 % abundance), and potassium (^{40}K present in 0.0118 % abundance). ^{40}K , however, is present in earth's core also and is contributing to core - heating. It is estimated that a column of granite 1 km long with a cross sectional area of 1cm^2 produces 2 calories of heat per year.

In earth crust, there are suitable geological environments, which are primarily meant for the deposit of uranium/thorium minerals. Noteworthy among different environments are (i) unconformity contact representing halt in the process of deposition (e.g. Athabasca basin, Cigar lake, Rabbit lake, Canada; Jabiluka II, Ranger, Naberlek, Australia), (ii) veins (Pribram, Czechoslovakia; Beaverlodge, Canada), (iii) sandstone (Shirley basin, Benavides, USA; Beverley, Australia; Domiasiat, India), (iv) breccia complex (Olympic dam, Australia), (v) surficial (Yellerrie, Australia; Flodell creek, Pryor mountain, USA), (vi) volcanic (Michelin,

Labrador, Canada; Ben Lemond, Australia) and (vii) quartz pebble conglomerate (Elliot lake, Canada; Witwatersrand, South Africa).

Beach sands (coastal stretches of Karnataka, Kerala, Tamil Nadu, Andhra Pradesh, Orissa) are excellent hosts for thorium minerals. However, beach placers are environments originating under conditions of interaction between the geological environments on land and wind-wave action.

Terrestrial gamma radiations (maximum up to 2.62 MeV of energy) on an average can penetrate 30-40 cm of rocks of average density 2.5 gm/cc. Thus depth information of about 30-40 cm is available. This is why radiometric surveys *in situ* are mostly based on detection and measurements of gamma rays. The two other radiations namely, alpha (α) and beta (β) particles have poor capability of penetration and are illustrated in Fig.1

Parent radioactive nuclides obtainable in nature are given in Table 1. Example of naturally occurring well known series of daughter products occur from that of $^{238}\text{U}_{92}$

Wonder in earth crust (at 2000 ma ago)

It is Oklo phenomenon at Gabon, in West African rainforest, where 17 active natural nuclear fission reactors were producing fission with naturally available ^{235}U at 2000 ma earlier. Reaction lasted from 1 to 8×10^5 years. Small areas (few meters in length and breadth but less than a meter in thickness) in clastic sedimentary rocks became the source of these natural reactors, a wonder. Reactor using light water (H_2O) requires an enriched fuel (2 – 3% of ^{235}U isotope). It may therefore be interesting to calculate the percentage of ^{235}U that would have been prevalent in nature in the past at 2000 ma. It turns out to be 3.5%.

^{235}U being 3.5% there remains a chance of fission reaction with light water. Small masses of uranium could initiate nuclear chain reactions in nature subject to certain conditions. At Gabon the natural reactor sites contained 20 to 60% U_3O_8 though it is uncommon at 2000 ma. The neutron capturing nuclei like boron and rare earth elements were low and the neutron moderators like hydrogen in water were present.

and $^{232}\text{Th}_{90}$ which through combined α and β^- decays end up as isotopes

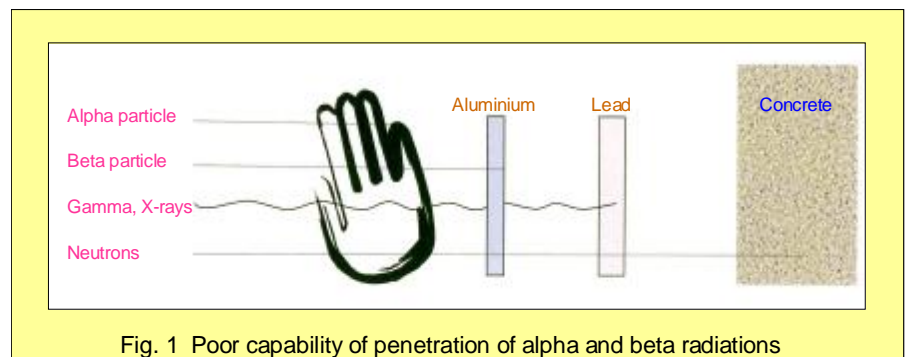


Fig. 1 Poor capability of penetration of alpha and beta radiations

Table 1.0. Parent radioactive nuclides which are found in nature

Nuclide		Atom (percent abundance)	Half-period (year)	Radiation observed	
Z	A				
19	K	40	0.0119	1.2×10^9	β^- , EC, γ
23	V	50	0.24	?	?
37	Rb	87	27.85	6×10^{10}	β^-
49	In	115	95.77	6×10^{14}	β^-
52	Te	130	34.49	$\sim 10^{21}$	Growth of ${}_{54}\text{Xe}^{130}$
57	La	138	0.089	$\sim 2 \times 10^{11}$	β^- , EC
60	Nd	144	23.9	$\sim 1.5 \times 10^{15}$	α
62	Sm	147	15.07	1.4×10^{11}	α
71	Lu	176	2.6	7.5×10^{10}	β^- , γ
75	Re	187	62.93	4×10^{12}	β^-
90	Th	232	100	1.39×10^{10}	α
92	U	235	0.715	7.13×10^8	α
92	U	238	99.28	4.49×10^9	α

of lead. Radon and thoron are intermediate radioactive gaseous daughter products, which may emanate through the fractures of rock. Other than Th – series, U – series and ${}^{40}\text{K}$, there are a few rare earth elements like La, Sm, Lu also available naturally in pegmatite (mineral constituent more or less same as granite like quartz, feldspar, mica etc. but larger crystal sizes) derived from fertile granite source having some special characteristics such as variable $\text{K}_2\text{O}/\text{Na}_2\text{O}$ values, higher Rb, low Sr etc.

Evolved terrain

In rocks containing Rb – Sr system, the growth of daughter ${}^{87}\text{Sr}$ due to decay of parent ${}^{87}\text{Rb}$ can be expressed as: $({}^{87}\text{Sr}/{}^{86}\text{Sr})_{\text{present}} = ({}^{87}\text{Sr}/{}^{86}\text{Sr})_i + ({}^{87}\text{Rb}/{}^{86}\text{Sr})_{\text{present}} (e^{\lambda t} - 1)$; where $({}^{87}\text{Sr})_i$ is the initial ${}^{87}\text{Sr}$ present at the time of formation and λ is the decay constant. The isotope ${}^{86}\text{Sr}$ is non-radiogenic in character. Division of both sides by ${}^{86}\text{Sr}$ to form ratios is useful as these quantities can be measured more precisely by mass spec-

trimeter than measuring the only isotopes.

The equation resembles $y_j = mx_j + c$ where $c = ({}^{87}\text{Sr}/{}^{86}\text{Sr})_i$ and $m = e^{\lambda t} - 1$.

The age estimate is an important factor in concluding whether the rock can be given basement status or a later intrusion into some Super group.

The evolved terrains may have significant potential for enrichment of uranium. Examples are Lambapur in Andhra Pradesh and Bodal in Madhya Pradesh where it appears that uranium minerals are associated with evolved older granite rocks and younger basic intrusive respectively.

Series equilibrium in crustal environments

If the parent has a relatively long half life, then after a long period of time the amount of any given daughter becomes constant, the rate of production from its parent equaling its rate of decay. The series is then said to be in a state of secular equilibrium. The length of time required for secu-

lar equilibrium to become established is dependent on the half-life of the longest-lived daughter in the decay chain below the parent.

The uranium decay series is subdivided into five separate groups named as (i) uranium group (${}^{238}\text{U}_{92} \rightarrow {}^{234}\text{U}_{92}$); (ii) thorium isotope (${}^{230}\text{Th}_{90}$); (iii) radium isotope (${}^{226}\text{Ra}_{88}$); (iv) radon group (${}^{222}\text{Rn}_{86} \rightarrow {}^{210}\text{Pb}_{82}$), and (v) lead group (${}^{210}\text{Bi}_{83} \rightarrow {}^{206}\text{Pb}_{82}$). The elements within each group tend to remain in equilibrium with the parent of the group, although the parent of the group may not itself be in equilibrium with the parent of the decay series ${}^{238}\text{U}_{92}$.

For freshly purified Th, the time required for the attainment of equilibrium is about 50 years for the series to reach in equilibrium with its daughters. This is approximately 7 times the half life (6.7 year) of ${}^{228}\text{Ra}_{88}$ – the daughter having the highest half-life, leaving the parent (Th). This daughter controls for the period of attainment of equilibrium in the series. For geologic specimens it is an instant. So for all practical purposes Th series remains in equilibrium in crustal environment.

For U series, the daughter isotope ${}^{234}\text{U}_{92}$ has the highest half-life (2.5×10^5 year). Seven times of it i.e., 1.75 million year is the time required for fresh U to reach in equilibrium. But in earth's crust the situation is different. The isotope ${}^{234}\text{U}_{92}$ may move preferentially over its parent. It may happen due to following reasons :

As ${}^{234}\text{U}_{92}$ forms via α emission, it may get relocated into crystal defects and micro cracks due to recoil and becomes available to oxidation. Since the recoil energy is 100 keV (approximately), the recoil nucleus may move by a distance of 10^{-7} to 10^{-6} cm, one or two orders larger than the size of

the crystal lattice.

A second explanation points out to the possibility of forming hexavalent ^{234}U atom at the time of its formation as it is stripped of its two electrons at the moment of decay.

The third possibility speaks of a mechanism in which the transformation $^{234}\text{Th}_{90} \rightarrow ^{234}\text{Pa}_{91} \rightarrow ^{234}\text{U}_{92}$ takes a tetravalent Th into pentavalent Pa and finally into hexavalent ^{234}U because each β decay is equivalent to loss of electron and an increase in valence.

Therefore, in such a situation when isotope $^{234}\text{U}_{92}$ moves preferentially over its parent, $^{230}\text{Th}_{90}$ (80,000 year half life) controls for the series equilibrium. About 5,60,000 years is required.

Degree of disequilibrium in terrains

An important aspect of radiometric assay is to ascertain about the degree of disequilibrium in U-series for the uranium present in the sample. Two basic types of disequilibrium can be identified: (i) residual, where uranium is leached out of the system and daughter nuclides remain and (ii) young, where freshly deposited uranium has not reached equilibrium with its daughter products.

A ratio between two concentrations, i.e., uranium (U_3O_8) and radium equivalent part of it [$\text{Ra}(\text{eU}_3\text{O}_8)$] (measurement based on 1.76 MeV) is calculated for each sample from a set of samples collected from a type of rock.

Crustal contribution to background radiation

The cosmic radiation, crustal radioactivity i.e., terrestrial radiation, radiation from building materials (phosphogypsum in cement is the contributor) and radiation from atmo-

spheric radon constitute what is known as background radiation in which we live. Slag containing radioactive material, dumped at a place may also contribute to background radiation.

In India, the cosmic contribution has been estimated to be about 28 mrem per year for cities like Kolkata, Mumbai and Chennai [1 rem is taken to be a quantity of radiation that produces the same biological damage in man as that resulting from the absorption of 1 rad (100 erg of energy absorbed in 1gm of material) of X – rays or gamma rays]. For city like Bangalore situated at an altitude of 921 meter above sea level it is 44 mrem per year.

The terrestrial contribution is a quantity that varies depending upon the concentration of Th, K, and U in

rocks. It can be estimated, for terrain of low topographic relief, using the data shown in Table 2. For example, for abundances of 12 ppm Th, 3 ppm U and 3.4% K in typical common igneous rocks; the exposure rate (mR/hr) at 1m height above the ground is $12 \times 0.31 + 3 \times 0.64 + 3.4 \times 1.53 = 10.84$ mR/hr or 43.36 mrem per year. In order to calculate the same for any other terrain the abundances of radio-elements in various types of rocks are required to be known and is given in Table 3.

The total background radiation can typically range from 150 to 350 mrem per year. In some places, however, it can be much higher. In states of Kerala and Tamilnadu due to monazite [(U, Th, La, Ce)PO₄] in beach sands about 1,40,000 population receive an average gamma dose of 1500 mrem per year. The contribution from radon in this area is approximately another 1500 mrem per year. The figure is comparable to the levels that occur in Brazil, Iran and Sudan with average up to 3800 mrem per year. Above a dose level of 5000 mrem per year, radiation related incidence may start to happen.

Table 2: Exposure rates contributed by thorium, uranium and potassium in the ground at various heights above the ground

z (m)	Exposure rate ($\mu\text{R/hr}$)		
	1 ppm Th	1 ppm U	1% K
1	0.31	0.64	1.53
50	0.18	0.37	0.92
100	0.12	0.24	0.61
150	0.08	0.16	0.43
200	0.06	0.11	0.30

Table 3: Estimated mean thorium, uranium, and potassium concentrations in various rocks (compiled from published data)

Rock Type	Th (ppm)	U (ppm)	K (%)	Th/U	K/U	Th/K
<i>Igneous</i>						
Ultrabasic	0.02	0.007	0.01	2.8	1.4	2.0
Basic	3.4	0.8	1.0	4.3	1.3	3.4
Basic-Intermediate	6.1	1.7	1.9	3.6	1.1	3.2
Intermediate	9.8	3.0	2.4	3.3	0.8	4.1
Intermediate-Acidic	16.0	3.6	3.0	4.4	0.8	5.3
Acidic	21.9	4.1	3.5	5.3	0.9	6.3
<i>Sedimentary</i>						
Evaporite	0.4	0.1	0.1	4.0	1.0	4.0
Carbonate	1.6	1.6	0.3	1.0	0.2	5.9
Sandstone	5.7	1.9	1.2	3.0	0.6	4.8
Shale	11.2	3.7	2.7	3.1	0.7	4.1
<i>Metamorphic</i>						
Amphibolite	2.0	0.9	0.6	2.2	0.7	3.3
Greenstone	3.4	0.8	1.0	4.3	1.3	3.4
Graywacke	6.7	2.1	2.8	3.2	1.3	2.4
Gneiss	10.6	2.3	3.4	4.6	1.5	3.1
Schist	13.5	4.1	2.5	3.3	0.6	5.5