World Academy of Science, Engineering and Technology
International Journal of Physical and Mathematical Sciences
Vol:5, No:4, 2011

Survey Gamma Radiation Measurements in Commercially-used Natural Tiling Rocks in Iran

A.Abbasi, F.Mirekhtiary

***Abstract*—**The gamma radiation in samples of a variety of natural tiling rocks (granites) produced and imported in Iran use in the building industry was measured, employing high-resolution Gamma-ray spectroscopy. The rock samples were pulverized, sealed in 0.5 liter plastic Marinelli beakers, and measured in the laboratory with an accumulating time between 50000 and 80000 second each. From the measured Gamma-ray spectra, activity concentrations were determined for ^{232}Th (range from 6.5 to 172.2 Bq kg^{-1}), ^{238}U (from 7.5 to 178.1 Bq kg^{-1}),^{226}Ra(from 3.8 to 94.2 Bq kg^{-1}) ^{40}K (from 556.9 to 1539.2 Bq kg^{-1}). From the 29 samples measured in this study, "Nehbndan (Berjand)" appears to present the highest concentrations for ^{232}Th,"Big Red Flower (China) "for ^{238}U , " Khoram dareh" for ^{226} Ra and " Peranshahr" for ^{40}K , respectively.

***Keywords*—**activity concentration, natural radioactivity, tiling rocks (granites)

I. Introduction

THE natural environmental radiation mainly depends on geological and geographical conditions. Higher radiation levels are associated with igneous rocks, such as granite, and lower levels with sedimentary rocks [1].Granites are the most abundant plutonic rocks of mountain belts and continental shield areas. They occur in great batholiths that may occupy thousands of square kilometers and are usually closely associated with quartz monzonite, granodiorite, diorite, and gabbro. Typical granites are chemically composed by 75 % silica, 12 % aluminium, less than 5 % potassium oxide, less than 5 % soda, as well as by lime, iron, magnesia, and titania in smaller quantities. However, because of the large quantities of granites that occur in nature, geologists believe now that most of the granites have been formed either by melting, partial melting, or metamorphism of deeply buried shale and sandstone [2]. In terms of natural radioactivity, granites exhibit an enhanced elemental concentration of uranium (U) and thorium (Th) compared to the very low abundance of these elements observed in the mantle and the crust of the Earth [3].

A.Abbasi is with the Physics Department, Faculty of Arts and Sciences, Eastern Mediterranean University, Famagusta T.R. North Cyprus via Mersin 10, Turkey; (Tel: +90 542 876 99 23 ; e-mail: akbar.abbasi@emu.edu.tr)

F.Mirekhtiary is with the Physics Department, Faculty of Arts and Sciences, Eastern Mediterranean University, Famagusta T.R. North Cyprus via Mersin 10, Turkey; (Tel: +90 542 886 19 11 ; e-mail:fmirekhtiary@gmail.com).

Geologists provide an explanation of this behaviour in the course of partial melting and fractional crystallisation of magma, which enables U and Th to be concentrated in the liquid phase and become incorporated into the more silica-rich products. For that reason, igneous rocks of granitic composition are strongly enriched in U and Th (on an average 5 ppm of U and 15 ppm of Th), compared to rocks of basaltic or ultramafic composition (< 1 ppm of U) [4]-[5].

In this paper, the results from gamma radiation measurements in samples of a variety of natural tiling rocks imported in Iran and sold under the commercial name of "granites" are presented. These results are of general interest since such rocks are globally used as building and ornamental materials. The measurements have been carried out in the Nuclear Analysis Laboratory of the Department nuclear Science and Technology Institute labs, using a high-resolution γ − ray spectrometry system.

II. Experimental Method

A. Gamma-ray Detection System

A stand-alone high-resolution spectrometry system is used for the measurement of the energy spectrum of the emitted gamma rays in the energy range between 50 keV and 3000 keV. The system consists of a high-purity germanium (HPGe) detector (coaxial cylinder of 72 mm in diameter and 120 mm in length) with an efficiency of 80 %, relative to a 3″×3″ NaI(Tl) scintillator. The spectrometry system is linked with a Multi-Channel Buffer (MCB) which is a PC-based plug-in PCI card consisting of an 16k Analogue-to-Digital Converter (ADC). An advanced Multi-Channel Analyser (MCA) emulation software (Gamma 2000) enables data acquisition, storage, display and online analysis of the acquired gamma spectra [6].The detector is surrounded by a graded-Z cylindrical shield consisting of lead, iron, and aluminum with thickness of 8 cm, 1.4 cm, and 1 cm, respectively, which provides an efficient suppression of the background gamma radiation present at the laboratory site.The energy-dependent detection photopeak efficiency has been determined using a calibrated mixed source gamma reference source, sealed in a standard Marinelli beaker. The energy resolution (FWHM) achieved in the measurements is 1.2 keV at the 1.33 MeV reference transition of ^{60}Co. Depending on the peak background and the counting time of each measured spectrum, the calculated Minimum Detectable Activity (MDA) varies within the range of 0.02 - 0.2 Bq kg^{-1} for all of radionuclide ^{232}Th , ^{238}U and ^{226}Ra and within the range of 0.05 - 0.5 Bq kg^{-1} for ^{40}K.

B. Sample Sreparation and Counting

A total of 29 different kinds of "granites" of those imported in Iran have been collected (Table 1). The measured samples were pulverized, sieved through 0.2 mm mesh, sealed in standard 1000 ml Marinelli beakers, dry-weighed and stored for four weeks before counting in order to allow the reaching of equilibrium between ^{226}Ra and ^{222}Rn and its decay products. However, significant non-equilibrium is uncommon in rocks older than 106 years, and the ^{232}Th series may be considered in equilibrium in most geological environments [7]. Each sample was put into the shielded HPGe detector and measured for an accumulating time 22 hours. Prior to the samples measurement, the environmental gamma background at the laboratory site has been determined with an empty Marinelli beaker under identical measurement conditions (B.G) It has been later subtracted from the measured γ−ray spectra of each sample.The results of the reported ^{232}Th, ^{238}U ,^{226}Ra and ^{40}K activity concentrations obtained for each of the measured samples together with their corresponding total uncertainties are summarized in Table 1. It is noted here that no other radionuclide than naturally occurring were detected in the measured samples and that the small contribution of the environmental γ-ray background at the laboratory site has been subtracted from the spectra of the measured samples.For compare of results uncertainty we used IAEA standard material such as soil-6, soil-375, and soil-327 IAEA-RGK-1, IAEA-RGU-1 and IAEA-RGTh-1 that all obtained results were accordance complete[8].

C. Calculation of Activity Concentrations

Calculations of count rates for each detected photopeak and radiological concentrations (activity per mass unit or specific activity) of detected radionuclides depend on the establishment of secular equilibrium in the samples. Since secular equilibrium was reached between ^{232}Th , ^{238}U and ^{226}Ra and their decay products, the ^{232}Th concentration was determined from the average concentrations of 228Ac ($T_{½}$ = 6.15 h) lines at 338.4 and 911.21 keV with an emission percentage 11.1% and 26.6%, and that of ^{238}U was determined from the average concentrations of the ^{234}Th (63.29 keV) with an emission percentage of 4.471% and for ^{226}Ra was determined by measuring its gamma emitters ^{214}Pb (295.213 and 351.921 keV) with an emission percentage of 19.3% and 37.6% respectively and ^{214}Bi (609.312 keV) with an emission percentage of 46%. The concentration of the ^{40}K was calculated using the gamma line at energy 1460.75 keV of emission percentage 10.7% [8].Thus, an accurate radionuclide concentration of ^{232}Th and ^{238}U was determined, whereas a true measurement of concentration was made. The specific activity (in Bq kg^{-1}), A_{Ei}, of a nuclide i and for a peak at energy E, is given by:

$$A_{E_i} = \frac{N_{E_i}}{\varepsilon_E . t . \gamma_d . M_s} \quad (1)$$

where N_{Ei} is the Net Peak Area of a peak at energy E, ε_E the detection efficiency at energy E, t the counting live-time, γ_d the gamma ray yield per disintegration of the specific nuclide for a transition at energy E, and M_S the mass in kg of the measure sample [9]. The total uncertainty (σ_{tot}) of the calculated activity values (Table1) is composed of the counting statistical (σ_{st}) and weighted systematic errors ($\sigma_{sys,i}$) calculated by the following formula [5].

$$\sigma_{tot} = \sqrt{\sigma_{St}^2 + \frac{1}{3}\sum_i \sigma_{sys'i}^2} \quad (2)$$

The systematic uncertainties considered include: the uncertainty of the source activity (3%), the uncertainty in the efficiency fitting function (1-10%), and uncertainties in the nuclide master library used (1-2%). As can be seen in Table 1.

III. Results and Discussion

Activity concentrations of ^{232}Th ranged from 6.5 to 172.2 Bq kg^{-1} ,of ^{238}U from 7.5 to 178.1 Bq kg^{-1}, of ^{226}Ra from 3.8 to 94.2 Bq kg^{-1} and of ^{40}K from 556.9 to 1539.2 Bq kg^{-1}. From the 29 samples measured in this study, "Nehbndan (Berjand)" appears to present the highest concentrations for ^{232}Th,"Big Red Flower (China) "for ^{238}U , " Khoram dareh" for ^{226} Ra and " Peranshahr" for ^{40}K , respectively.Investigated, reaching levels of 172.2 Bq kg^{-1} for ^{232}Th, 178.1 Bq kg^{-1} for ^{238}U, 94.2 Bq kg^{-1} for ^{226}Ra and 1539.2 Bq kg^{-1} for ^{40}K.All measured samples except of four , named "Morvared sabz" , "Gareh dash", "Sangeh lorestan" and "Gal exec India" show concentrations of 40K above the value of 500 Bq kg-1. In addition, 24 samples appear to present concentration of ^{232}Th lesser than 100 Bq kg^{-1}, while only 6 samples exhibit concentration of ^{238}U that surpass the above limit. Analytical results for the activity concentrations of ^{232}Th, ^{238}U, ^{226}Ra and of ^{40}K determined for each of the measured samples together with their total uncertainties are presented in Table 1.The measured activity concentrations of ^{232}Th, ^{238}U , ^{226}Ra and ^{40}K can be converted into total elemental concentrations of Th , U , Ra (in ppm) and of K (in percent), respectively [9].The extracted values for the elemental concentration are: for Th (range from 1.6 to 42.3 ppm), for U (from 0.6 to 14.4 ppm) , for Ra (from 0.1 to 2.6 ppm) and for K (from 1.8 to 85.7%) Fig.1.In average, the lower values correspond to sample No.16 ("Gal excei") and the higher ones to sample No.17 ("Garmez goldorosht"). The results are summarized in Fig. 2 and Fig. 3.

The strong correlation between uranium and radium in granite samples under investigation (correlation coefficient=0.885) which indicate the secular equilibrium between ^{238}U and ^{226}Ra shows in Figure 4. While Figure 5 shows the good correlation between the concentrations of the two radioactive isotope (^{232}Th, ^{238}U) (correlation coefficient = 0.689).

IV. Conclusions

Exploitation of high-resolution γ-ray spectroscopy provides a sensitive experimental tool in studying natural radioactivity and determining elemental concentrations in various rock types. Most of the tiling rock "granite" samples studied in this work reveal high values for the activity and elemental concentrations of Th, U, Ra and K, thus contributing to high absorbed dose rates in air. In general, the extracted values are distinctly higher than the corresponding population-weighted (world-averaged) ones, and, in some cases (e.g. "Nehbndan (Berjand)", "Big Red Flower (China)", "Khoram dareh" etc.), In view of worldwide concern about the radioactivity contents of various construction materials, experimental measurements of the activity concentrations of various granite types(local and foreign) commonly used in iran have been carried out. The measured values of the activities of ^{232}Th, ^{238}U, ^{226}Ra and ^{40}K in the granite samples have been found to lie in the ranges: 6.5–172.2, 7.5-178.1, 3.8 – 94.2 and 556.9–1539.2 $Bqkg^{-1}$, respectively. Environmental monitoring should be carried out for granites where people might be exposed to radioactivity. It emphasizes the importance of accurate information concerning commercial names and origins of these granites, because simple mistakes on that can produce serious economical and social consequences in the stone market sector.

Acknowledgements

This work is conducted with assist from the boulder confederacy of Iran. We would also like to thank the Iran's Association of Manufacturers, which provided us with the original granite materials from which the samples are prepared and gave us information about their origin.

References

[1] Florou, H., Kritidis, P., 1992. Gamma radiation measurements and dose rate in the costal areas of a volcanic island, Aegean Sea, Greece. Radiation Protection Dosimetry 45 (1/4), 277-279.

[2] Snelling, A., Wood morappe, J., 1998. Rapid Rocks Granites… they didn't need millions of years of cooling. Creation Ex Nihilo 21(1), 37-39.

[3] Luigi Bruzzi, Maurizio Baroni, Renato Mele and Elisabetta Nanni.,1997. " Proposal for a method of certification of natural radioactivity in building materials". Journal of Radiol.Vol.17,No.2 , P.85-94.

[4] Thomas, J., Hulka, J., Salava, J., (1993). New houses with high radiation exposure levels. Proceedings of the International Conference on High Levels of Natural Radiation, Ramsar, 1990.

[5] Hamby, D. M., Tynybekov, A. K., 2002. Uranium, thorium and potassium in soils along the shore of lake Issyk-Kyol in the Kyrghyz Republic. Environmental Monitoring and Assessment 73, 101-108.

[6] EG&G ORTEC, 1999. GammaVision-32: Gamma-Ray Spectrum Analysis and MCA Emulator. Software User's Manual (V5.1). EG&G ORTEC.

[7] Chiozzi, P., Pasquale, V., Verdoya, M., 2002. Naturally occurring radioactivity at the Alps-Apennines transition. Radiation Measurements 35, 147-154.

[8] Reference sheet IAEA-soil-6 ,1999. soil-375 ,2000 . 327 ,2001. RGU-1 ,1987. RGK-1,1987. RGTh-1,1987 "International Atomic Energy Agency". Analytical Quality Control Services, Wagramer Strasse 5,P.O.Box 100,1400.

[9] Tzortzis, M., Tsertos, H., Christofides, S., Christodoulides, G., 2002. Gamma-raymeasurements of naturally occurring radioactive samples from Cyprus characteristicgeological rocks. Preprint UCY–PHY–02/02 (physics/0212099).

TABLE I
ACTIVITY CONCENTRATIONS OF ^{232}Th, ^{238}U, ^{226} Ra AND ^{40}K of NATURAL TILING ROCKS (GRANITES) IMPORTED AND USED IN IRAN

Sample No.	Commercial name	Origin	Activity (Bq/kg)			
			^{232}Th	^{238}U	^{226}Ra	^{40}K
G-1	Brown india	India	86.0±0.4	11.8±0.6	9.3±0.1	1220.6±0.8
G-2	Chayan sable	Iran	23.4±0.1	28.8±0.6	15.7±0.2	556.9±0.5
G-3	Tekab	Iran	95.1±0.4	69.7±0.3	51.8±0.1	690.9±0.2
G-4	Nehbndan birjand	Iran	172.2±0.5	152.3±0.7	91.6±0.3	1385.4±0.2
G-5	Peranshahr	Iran	62.7±0.2	69.5±0.4	39.5±0.7	1539.2±0.4
G-6	Torbat hydaryeh	Iran	33.6±0.4	40.8±0.2	27.1±0.5	623.9±0.8
G-7	Natanz	Iran	84.8±0.2	113.9±0.4	71.8±0.3	848.1±0.9
G-8	Morvared mashhad	Iran	42.7±0.5	31.8±0.7	18.9±0.1	1374.4±0.5
G-9	Akbatan hamedan	Iran	98.3±0.4	37.4±0.6	24.3±0.7	1319.5±0.8
G-10	Sangeh alamot	Iran	59.8±0.2	80.1±0.7	45.7±0.3	1385.4±0.3
G-11	Garmez yazd	Iran	59.5±0.1	53.5±0.1	41.1±0.7	1077.8±0.2
G-12	Balloch zahedan	Iran	72.7±0.8	57.8±0.4	32.5±0.4	1121.7±0.5
G-13	Morvared sabz	Iran	71.6±0.4	47.2±0.8	28.4±0.7	1451.3±0.9
G-14	Sangeh khoramdareh	Iran	131.6±0.7	149.9±0.5	94.2±0.2	1506.2±0.7
G-15	Garmez golrez	China	86.6±0.2	66.0±0.4	71.4±0.1	1242.6±0.4
G-16	Gal excei	India	6.5±0.3	7.6±0.2	3.8±0.1	146.0±0.4
G-17	Garmez goldorosht	China	160.9±0.7	178.1±0.8	93.4±0.4	1374.4±0.3
G-18	Golzard	China	58.4±0.8	82.2±0.4	55.4±0.5	1244.3±0.5
G-19	Azarbyjan	Iran	43.7±0.5	37.5±0.6	22.1±0.7	1010.4±0.7
G-20	Trasheh sfed	Iran	97.1±0.4	98.1±0.2	50.8±0.2	884.3±0.2
G-21	Khalkhali	Armenia	76.7 ±0.8	87.5±0.3	60.2±0.3	1129.5±0.9
G-22	Trasheh sfed	Iran	132.6±0.7	142.3±0.7	87.5±0.6	1305.7±0.1
G-23	Goldar seyah	Pakistan	87.6±0.2	58.6±0.9	34.2±0.1	741.9±0.6
G-24	Morvared sabz	Iran	38.6±0.4	31.2±0.6	18.4±0.9	259.4±0.5
G-25	Greh dash	Turkish	29.4±0.1	37.1±0.2	14.3±0.4	357.1±0.4
G-26	Yazi dash	Turcoman	67.8 ±0.5	88.5±0.4	45.7±0.7	1179.8±0.7
G-27	Hekmtaneh	Iran	91.6 ±0.2	96.7±0.6	40.9±0.1	992.3±0.2
G-28	Sangeh lorestan	Iran	38.6±0.4	78.4±0.1	32.6±0.8	288.6±0.6
G-29	Alborz	Iran	135.4±0.8	127.5±0.8	67.8±0.6	1239.7±0.4

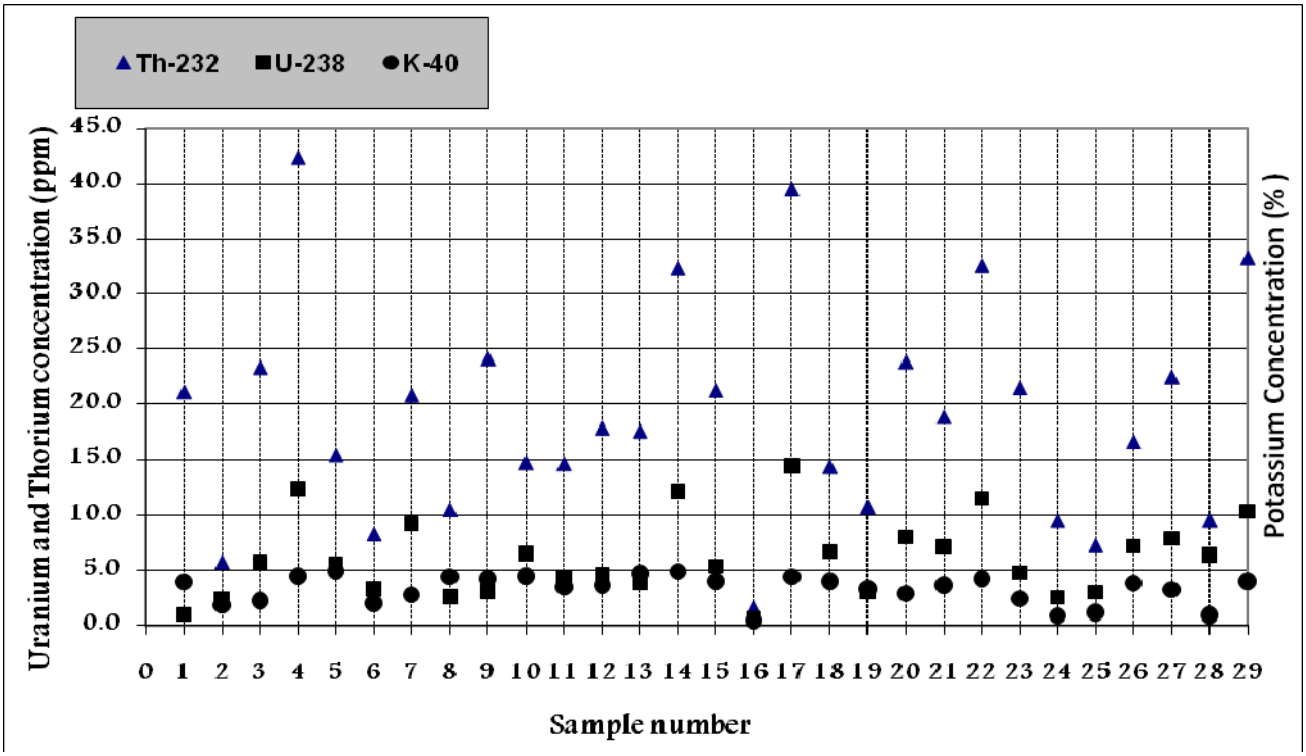

Fig. 1 Th, U and K elemental concentrations in the samples of "granite" rocks (Table 1) imported and used in Iran

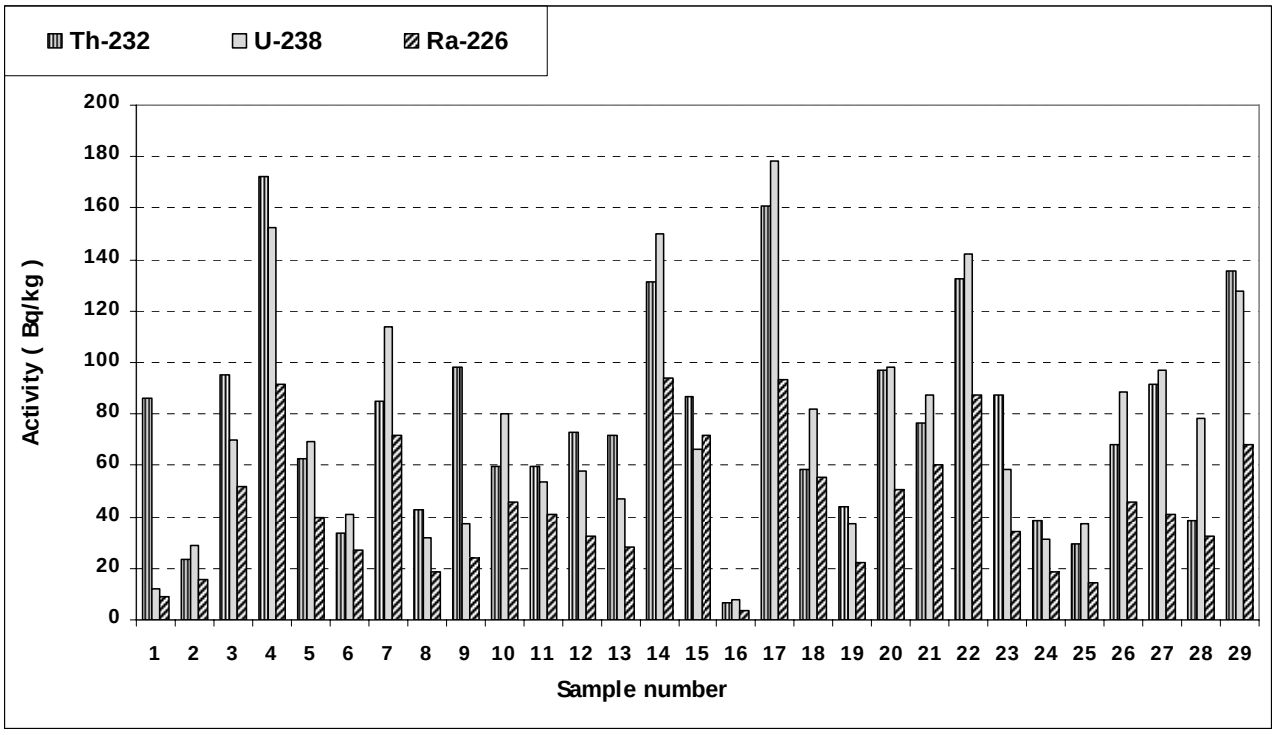

Fig. 2 Thorium (^{232}Th) and uranium (^{238}U) decay series and Radium (^{226}Ra) activity in the "granite" samples (Table 1).

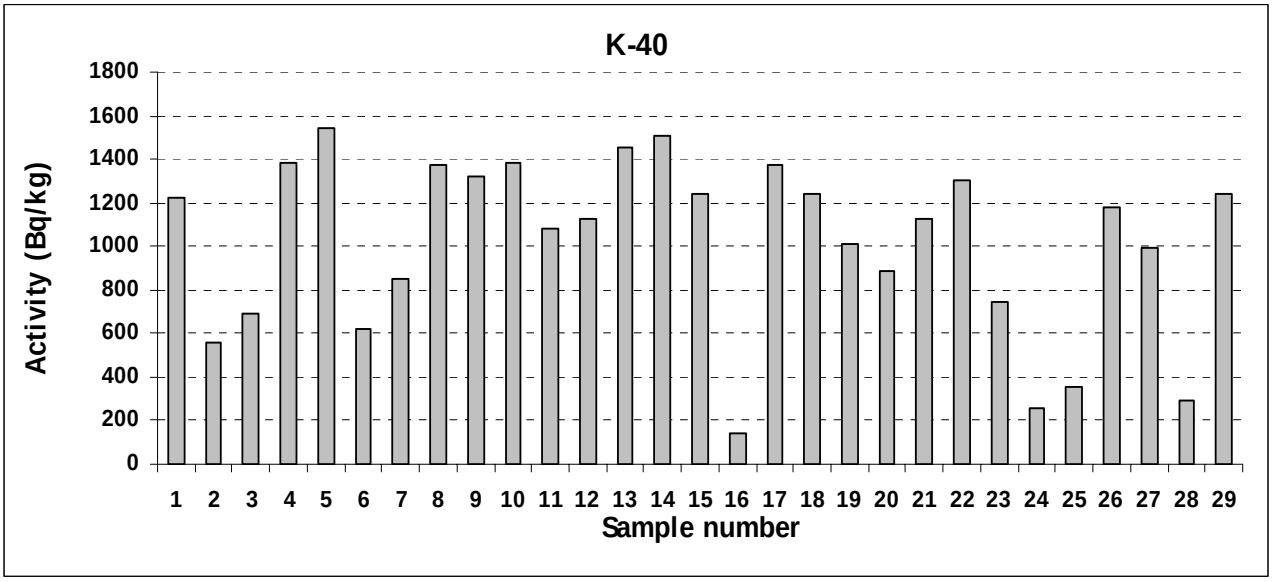

Fig. 3 Potassium (^{40}K) activity in the "granite" samples (Table I)

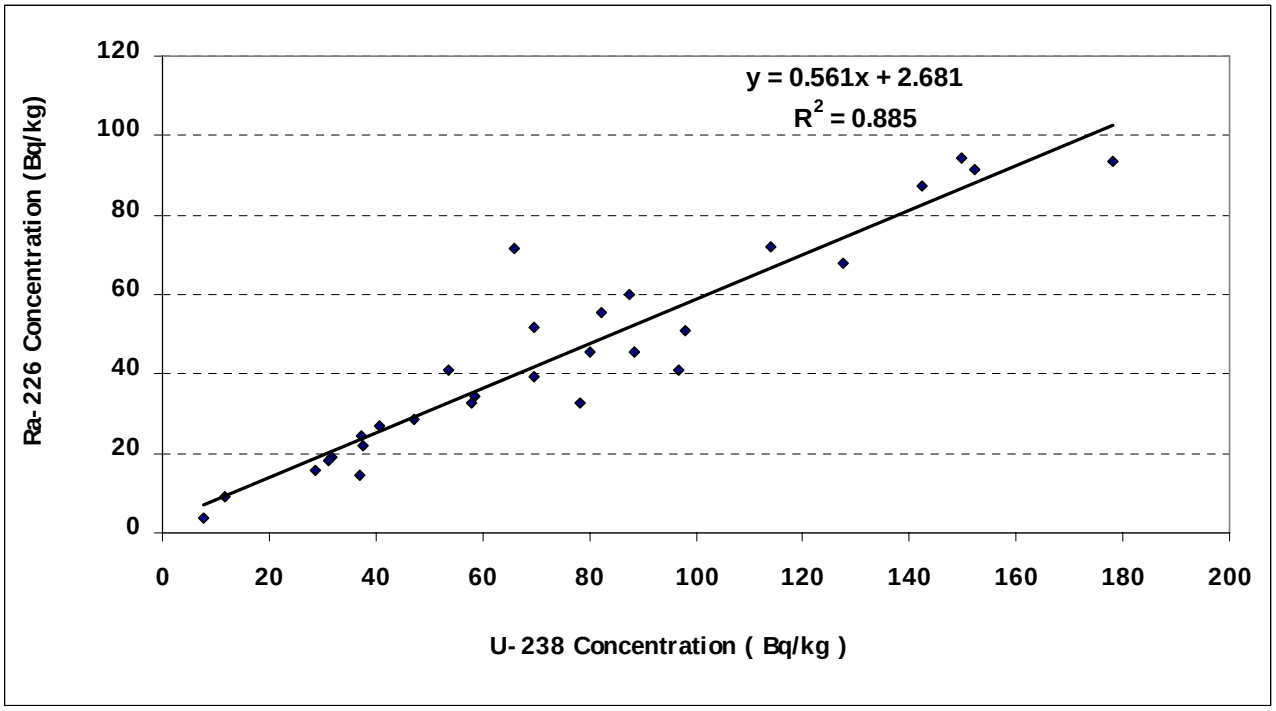

Fig. 4 The correlation between ^{238}U and ^{226}Ra concentration in samples

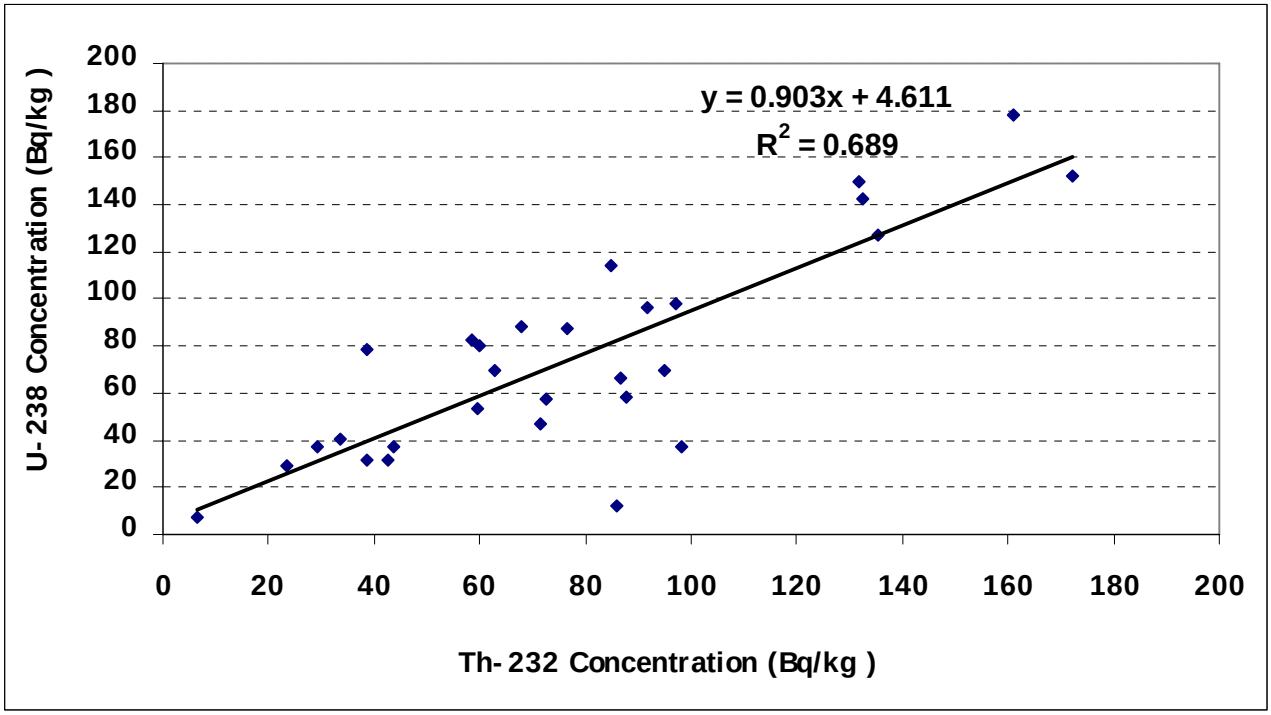

Fig.5 The correlation between ^{238}U and ^{232}Th concentration in samples

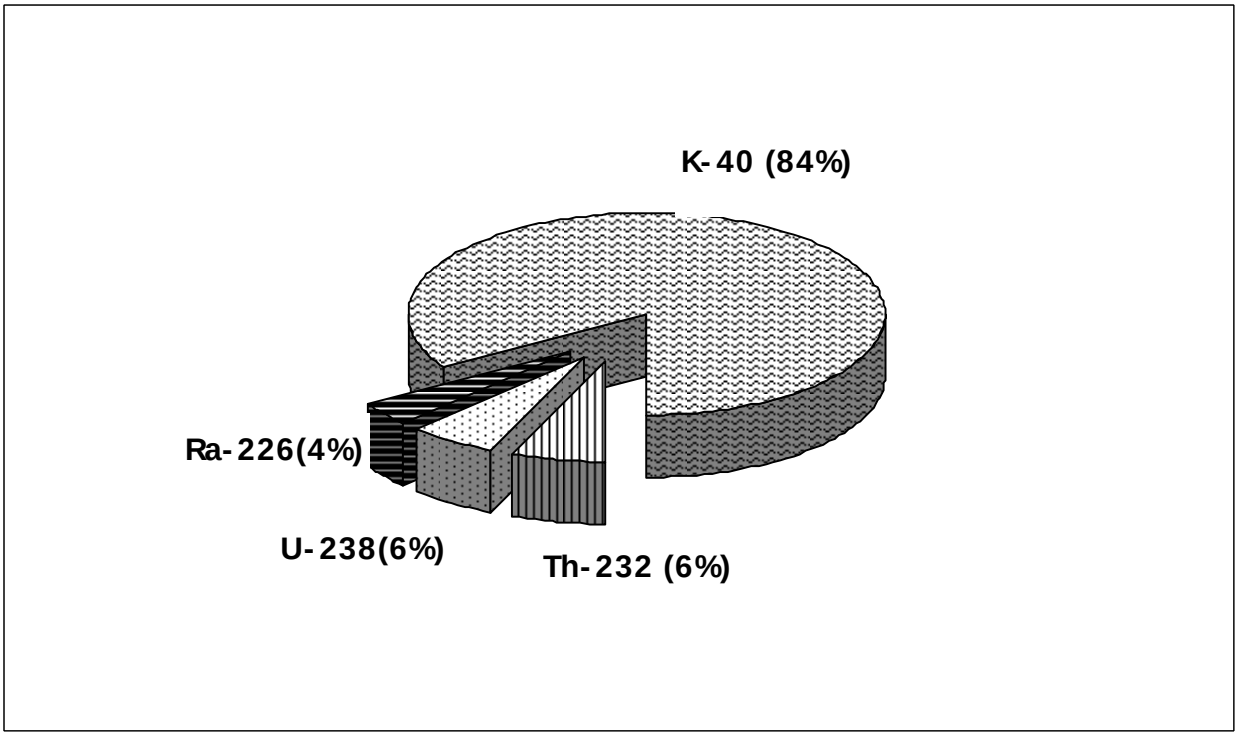

Fig.6 Relative concentration radioactivity due to thorium (^{232}Th) , uranium (^{238}U) , radium (^{226}Ra) and potassium (^{40}K) in "Granite" samples