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### Evaluation of the Natural Radioactivity and the Radiation Hazard Indices of Some Granitic Masses at Raniah Area, El-Taif, Kingdom of Saudi Arabia.

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#### ABSTRACT

Five granitic masses in the Raniah area were a matter of concern regarding their suitability for domestic uses in the term of their natural radioactivity and the hazard indices. Raniah area is located between latitudes 28°15'- 28° 18'N and longitudes 42° 30' - 42° 50' E about 350 km east of El-Taif city, Kingdom of Saudi Arabia. The granitic masses are named as; JabalMaslukh, JabalWaridat, JabalKawr, JabalHarashef and Jabal Al-Khathami granites. Petrographically, they were assigned as alkali feldspar granites to alkali granites with perthite, quartz and plagioclase as the major mineralogical components together with sensible amounts of biotite and alkali amphiboles. Zircon appeared as the common accessory mineral in addition to the apatite and opaques. Geochemically, they exhibited the alkalinity and per-alkalinity affinities and originated from high differentiated magma in the within-plate tectonic environment as A-type granites. The radiometric measurements pointed to that the granitic masses of J. Maslukh, J. Kawr and J. Harashef have the lowest <sup>226</sup>Ra and <sup>232</sup>Th concentrations while the highest values were provided by J. Al-Khathami granite. The calculated radium-equivalent values were ranged between 134.07 Bqkg<sup>-1</sup> (J. Maslukh) and 255.13 Bqkg<sup>-1</sup> (J. Al-Khathami) while the adsorbed dose rate recorded its lowest value as 64.39 nGyh<sup>-1</sup> and its highest value as 122.83 nGyh<sup>-1</sup> by the granites of J. Maslukh and J. Al-Khathami respectively, also the same behavior were found in the values of the external and internal hazard indices. The annual indoor effective dose, the annual outdoor effective dose and the total annual effective dose values of all the examined granitic rocks recorded their highest values as 0.603, 0.151 and 0.754 (mSv) respectively in J. Khathami which are lesser than the safe criterion limit (1 mSv) for general public. Although some values of the excess lifetime cancer risk are slightly elevated but they are generally either within the safe limit  $(1.45 \times 10^{-3})$  or very close.

Keywords: Natural radioactivity, Hazard indices, Granite, Raniah, El-Taif.

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#### INTRODUCTION

The term "Granitic rocks" refers to those igneous rocks of mineralogical composition mainly consists of alkali feldspar,quartz, plagioclase, mica and occasionally the hornblende. The granitic rocks occupy large areas of the Arabian-Nubian Shield including the western part of the Kingdom of Saudi Arabia (KSA). Due to its durability and decorative appearance, the granite is a popular building material in homes and buildings. Generally, all the rocks in the nature contain naturally occurring radioactive elements like radium, uranium and thorium. In particular, some kinds of the granitic rocks encountered more concentrations of these radioelements more than the other rocks.

In terms of the natural radioactivity in granitic rocks, the main source of radiation is represented in the radionuclides of the uranium (<sup>238</sup>U) and thorium (<sup>232</sup>Th) series, and the radioactive isotope of potassium (<sup>40</sup>K). In the <sup>238</sup>U series, the decay chain segment starting from radium (<sup>226</sup>Ra) is radiologically the most important, therefore reference is often made to <sup>226</sup>Ra instead of <sup>238</sup>U<sup>[1]</sup>. These radionuclides are sources of the external and the internal radiation exposures in dwellings [1]. The internal exposure happens through the inward breath of radon gas and the external exposure occurs through the outflow of entering gamma rays from the radioactive sources [2]. Accordingly and from the natural risk point of view, it is necessary to know the dose limits of public exposure and to estimate human exposure to natural radiation sources. Also, knowledge of the common radioactivity level is suitable valuable to set the gauges and national rules used for providing recommendations. This is the reason that excessive considerations have been paid to deciding radionuclide fixations in building materials in many countries [3-15].

Since the extensive using of granites into houses and buildings as tiles, walls and/or decorative materials and as the most individuals spend 80% of their time indoors, a great attention has been paid to determining radionuclide concentrations in building materials in many countries [16-25]. The average indoor absorbed dose rate in air from terrestrial sources of radioactivity is estimated to be 70 nGyh<sup>-1</sup> [26].

The KSA government devoted an interest to the ornamental stones as an important economic source to fulfillment the requirements of the domestic market especially in the western region (Jeddah – Mecca – Raniah), south Nagd, Dakhna, Rass, Albjadih, south, Asir, Najran, northwest Hejaz, Madina and Yanbu.

The physical characteristics of the rock (e.g. the hardness, the mechanical characteristics, the attractive appearance, resistance to weathering and pollution) are tightly relate to the mineral composition, degree of crystallization, level of firmness and degree of hardness of the rock which, in turn, depend on its origin (magmatic, metamorphic and sedimentary).

#### Aim of the study

The current work will elaborate on the mineralogical, chemical and, in a particular interest, the radioactive characteristics of some granitic masses in the western area of KSA to evaluate their suitability for using in different application specially the houses construction, ornamentation and decoration. Consequently, two benefits are intended from this work; (1) Effective contribution to create a radioactive exposure map overall the KSA land which support the human being and environmental protection issue and (2) Evaluate the minerals, ores and rocks as economic resources at Taif region which might support and integrate with the national development programs.

To conduct these aims various field, petrographical, chemical and radiometric studies were carried out on the examined granitic rocks and some important radiation indices were calculated to evaluate the human exposure dose could be imposed by these granites.

#### MATERIALS AND METHODS

#### The petrographic investigation

Twenty-five fresh samples representing the five granitic masses in Raniah area (JabalMaslukh granite, JabalWaridat granite, JabalKawr granite, Jabal Al-Khathami granite and JabalHarashef granite) were selected



for the thin-sections preparation. All the thin-sections were carefully studied using the polarizer microscope which established in Taif University, KSA.

#### The geochemical analysis

Twenty-three granitic samples were picked up as they the highly fresh varieties based on the microscopic survey. These samples were crushed then ground to –200 mesh size then the appropriate weights (0.5 and 0.05 g) of each sample were completely dissolved using the acid attack (15 ml HF + 5 ml HNO<sub>3</sub> + 5 ml HClO<sub>4</sub>) and the alkali fusion (NaOH pellets) techniques. After the appropriate heating till complete dryness, the residue of each attack was dissolved using dilute HCl acid (1:1) then up to volume (100 ml) using de-ionized water. The major element constituents were measured by the spectrophotometer instrument (Unicam brand, model UV2/100 of dual UV/Visible beams), the flame photometry device (Jenway brand model) or the traditional titration method. The analytical technique and steps were employed based on the kind of the measured element [27-28]. On the other hand, the trace elements concentrations were determined by the non-destructive X-ray fluorescence technique (model; PHILIPS brand, X'Unique II model).The analytical processes were carried out in Al-Taif University, KSA.

#### The radiometric measurement

Ten samples from the concerned granitic masses were chosen for the radiometric measurements. They were crushed, ground, homogenized and sieved to make them in the size of  $\leq 0.2$  mm which is the appropriate size enriched in heavy minerals (usually contain the radioelements). Each sample was dried in the dryness oven at 110°C for 24 hours for complete moisture removal. The desired weight of each sample was placed in a polyethylene cup of 350 cm<sup>3</sup> volume. The beakers were tightly sealed for 4 weeks to reach the secular equilibrium where the progeny decay rate comes to equilibrium with that of the parent (radium and thorium) within the volume and the progeny will also remain in the sample.

By the end of the four weeks, the sealed cups were put in a stand-alone high-resolution spectroscopic system which used for measuring 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 55 mm in diameter and 73 mmin length) with an efficiency of 30%, relative to a 33.33 Nal(TI) scintillator. The detector is mounted on a cryostat which is dipped into a 30-litreDewar filled with liquid nitrogen. Advanced Multi-Channel Analyzer (MCA) emulation software (MAESTRO-32) allows data acquisition, storage, display and online analysis of the acquired  $\gamma$ -spectra.

The photo-peaks of <sup>214</sup>Pb (352 keV) and <sup>208</sup>Tl (2615 keV) were considered to assess the activity concentrations (Bqkg<sup>-1</sup>) of <sup>226</sup>Ra and <sup>232</sup>Th respectively while the <sup>40</sup>K activity was measured directly from its 1460 keV gamma ray line.

#### Geological, petrographical and geochemical features

The Raniah area (the area of this study) is located between long.  $42^{\circ} 30' - 42^{\circ} 50'$  E and lat.  $28^{\circ} 15' - 21^{\circ}$  18' N about 350 km to the east of Taif and 140 km NE of Bishah and about 30 – 200 m above plain level (figure 1).

The Raniah area consists of a mountainous series extends for a distance of 15 Km. including JabalMaslukh, JabalWaridat, JabalKawr, Jabal Al-Khathami and JabalHarashef. Tectonically, they are late originated granites which are a favorable situation for using ornamental stones.

**Jabal Maslukh** granite forms an isolated small hill of massive and dense rock forming large blocks ( $\approx$  5m<sup>3</sup> in volume) and sometimes it is found as smaller loose blocks. It is a coarse-grained texture with a dominant pink color where the feldspar crystal up to 0.7 cm in length.

**Jabal Waridat** forms an isolated pure pale green mountain and far about 12 km to the NNE of Raniah town. Its granite appears free from cleavages with consolidated medium- to coarse-grained grains and has high mechanical properties. *It is found with great reservoir and is exploited under the name "silver pearl"*.





Fig (1): Location map of Raniah area showing the spread granitic quarries.

Jabal Kawr contains varieties of alkali granite to alkali feldspar granite. The alkali granite forms the main exposed type. It ranges in color from pinkish red to greenish blue of coarse-grained to less medium-grained crystals. Weathering is everywhere shallow, but the rocks are sound below. The fresh variety is of pale green color and intersected by two sets of vertical joints tending WNW and NNW parallel to the faults intersecting the rocky block. The fresh rock is homogeneous in terms of color and grain size and has been commercially known as "green emerald".

**Jabal Al-Khathmi** composed of massive granitic blocks (Figure 7) of large size ( $\approx 7m^3$ ) with variable colors. It is a favorable occurrence for bench or open-bit quarrying. Al-Khathami granite is alkali feldspar granite varies in color from beige, brown to greenish color with medium- to coarse-grained size. The rock is homogeneous in terms of color and texture and is exploited under the name "golden plant leaf".

**Jabal Harashef** occupies the northern part area of JabalKawr. Its granites exhibit various colors (grey, green and blue) which probably attributed to their variable contents of the amphibole and pyroxene minerals

The petrographic description of the studied granites indicated the approximate similarity in their composition and textures with minor differences in the minerals abundance, the accessory and secondary minerals varieties (figures 2&3). The potash feldspars, quartz and plagioclases are the main constituents with sensible presence of the alkali amphibole and sometimes the biotite minerals. Zircon is the common accessory mineral in all the examined granites and in lesser amount the apatite, opaques and, occasionally, sphene



minerals. The secondary minerals are usually represented by sericite, kaolinite, epidote, iron oxides and chlorite. The Hypidiomorphic texture is a characteristic texture in these granites with occasional presence of myrmekitic, granopheric and prophyritic textures vary from granite rock to another. Generally, all the studied granites were categorized either as alkali feldspar granite or alkali granite and displayed the presence of arfvdsonite and/or riebeckiteminerals.



Fig. (2): A- irregular blebs and patches within the orthoclase giving rise to granopheric texture, the quartz corrodes into the k-feldspar, (J. Maslukh granite).

B- albite twinned plagioclase with thin lamellae and arfvedsonitesubhedral crystals, (J. Maslukh granite). C- brownish green elongate prismatic amphibole crystals (arfvedsonite) and large perthite crystal engulfed blebs of quartz showing poikilitic texture, (J. Waridat granite).

D- Chloritizedbiotite flakes with elongated zircon and opaque crystals (J. Wardiat granite). E- quartz anhedral crystals with faint undulose extinction and epidot crystals, (J. Kawr granite). F- k-feldspar of string perthite type and myrmekitic texture, (J. Kawr granite).

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Fig. (3): A- alkaline granite with brownish riebeckite crystal, (J. Harashef granite).
B- string perthite engulfed quartz blebs showing poikilitic texture, lamellar twinned plagioclase and dark blue amphibole crystals, (J. Harashef granite).
C- large quartz crystal with myrmekite texture developed when plagioclase crystals replace the potash feldspar, (J. Al-Khathami granite).
D- subhedralbiotite plates and alkali amphiboles with hypdiomorphic texture, (J. Al-khathami

granite).

**The chemical composition** of Raniah granitic masses was identified through the concentrations of their major and trace constituents (tables 1&2).

Eleme S.No.	ent	SiO <sub>2</sub>	TiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub>	MgO	CaO	Na <sub>2</sub> O	K <sub>2</sub> O	P <sub>2</sub> O <sub>5</sub>	LOI	Total
M1	_	71.20	00.05	13.10	04.10	00.50	02.70	02.00	03.60	00.20	00.36	97.81
M2	lukh nite	72.10	00.10	16.00	00.50	00.40	02.00	03.70	03.20	00.50	00.47	98.97
M3	Mas grat	75.00	00.20	13.80	00.50	00.90	01.50	03.50	03.60	00.60	00.40	100.0
M4	2	75.00	00.15	13.80	03.60	01.40	01.70	03.50	03.40	00.20	00.36	103.1
W1		74.30	00.35	15.00	00.50	00.50	01.60	03.10	03.60	00.40	00.20	99.55
W2	idat 1ite	75.10	00.60	10.20	03.30	00.50	01.80	02.60	03.00	00.09	00.92	98.11
W3	Nar grai	70.00	00.20	14.00	03.70	00.56	03.00	03.40	03.50	00.08	00.25	98.69
W4	-	71.10	00.20	14.20	03.50	00.60	02.60	03.00	03.20	00.10	00.45	98.95
Khath11	e e .	70.90	00.05	15.80	01.50	00.40	01.40	03.50	03.50	00.30	00.30	97.65
Khath12	주 문	75.00	00.20	11.20	03.30	01.10	01.40	03.30	03.40	00.06	00.05	99.01

Table (1): Major elements constituents (%) of the investigated granites.

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Khath13		72.10	00.10	15.10	02.50	00.20	02.00	03.10	03.40	00.04	00.25	98.79
Khath14		75.00	00.20	13.80	00.25	00.20	01.80	03.70	03.50	00.30	00.16	98.91
Har16	h f	69.10	00.30	13.50	06.80	00.95	01.20	02.00	03.60	00.10	00.42	97.97
Har17	anit	75.00	00.20	13.80	00.60	01.20	01.20	02.5	03.60	00.60	00.20	98.90
Har18	Br B	69.10	00.20	13.50	05.70	01.20	01.00	01.80	04.25	00.10	00.10	96.95
Kor2		69.2	00.25	14.20	04.10	01.01	02.60	04.30	03.20	00.20	00.25	99.31
Kor4		72.10	00.05	14.50	02.40	01.10	02.60	01.70	03.40	00.20	00.10	98.15
Kor5	ite	73.20	00.15	14.10	03.10	00.10	01.60	02.00	03.30	00.10	00.03	97.68
Kor6	gran	72.50	00.40	15.60	00.20	00.10	01.70	03.30	03.40	00.50	00.12	97.82
Kor7	N 8	72.40	00.35	15.10	01.40	00.10	01.70	03.50	03.40	00.60	00.04	98.59
Kor8	Kav	69.10	00.10	13.50	02.60	00.10	01.40	03.40	03.50	001.0	00.02	93.82
Kor9		69.10	00.06	13.50	02.50	00.40	01.20	03.50	03.50	00.10	00.06	93.92
Kor10		74.10	00.10	14.00	02.00	00.40	01.20	02.40	02.90	00.30	00.05	97.45

Table (2): Trace elements constituents (ppm) of the investigated granites.

Eleme S.No.	ent	Cr	Ni	Cu	Zn	Zr	Rb	Y	Ва	Pb	Sr	Ga	V	Nb
M1	_	31	5	9	90	498	158	228	286	15	18	13	6	84
M2	te ukh	31	7	12	74	270	123	170	224	5	14	10	4	64
M3	lasl	18	3	4	71	361	113	164	217	9	13	5	4	62
M4	Σŵ	24	6	7	60	337	124	154	227	10	12	7	4	58
W1		27	7	9	66	344	166	157	266	16	16	13	5	59
W2	dat ite	31	6	8	51	342	116	112	201	14	9	9	4	41
W3	/ari ani	25	7	12	69	348	133	155	410	13	13	14	9	59
W4	2 20	21	7	11	60	312	168	140	297	10	12	10	6	53
Khath11	5	29	6	10	87	534	217	242	242	25	20	17	4	90
Khath12	har 1ite	25	5	7	80	494	180	242	251	19	20	8	4	91
Khath13	hat grai	21	6	11	79	529	200	231	229	16	19	14	4	87
Khath14	<u> </u>	23	6	12	91	504	181	225	252	17	18	19	4	83
Har16	. <u>.</u>	25	4	8	131	328	56	417	333	5	35	9	6	157
Har17	ara 'ani	21	5	10	136	337	128	350	339	8	2	12	5	131
Har18	тლ	28	5	7	111	732	65	331	313	4	27	10	6	125
Kor2		22	7	11	51	552	150	247	463	14	21	11	9	93
Kor4		23	5	6	52	294	264	136	157	18	10	8	3	50
Kor5		27	4	9	54	347	249	162	209	12	13	12	3	59
Kor6	te	26	7	11	63	290	242	113	163	19	8	16	3	41
Kor7	ani	21	6	11	60	365	311	170	162	22	13	17	3	63
Kor8	10	25	5	11	71	312	303	147	172	21	1	15	3	55
Kor9	awi	52	7	12	59	423	273	192	209	20	15	14	3	72
Kor10	Ÿ	18	5	9	74	338	276	157	186	21	12	13	3	57

In terms of the obtained major and trace elements data, some geochemical features of the examined granitic masses can be adopted as following:

i- The alkalinity and peralkalinity nature of these granites was indicated by their high concentrations of Nb, Y and Zr which ranged between 41-157 ppm, 112-417ppm and 243-928 ppm respectively. These values are in consistence with their corresponding of alkaline granites in the Arabian-Nubian Shield [29]. This conclusion is in compatible with presence of the sodic amphiboles (riebeckite and arfvedsonite) in these granites as indicated from their petrographic investigation.



- ii- They are assigned as A-type granite (figures 4&5) [30-31]. Also, these granites show the typical geochemical characteristics of the A-type post-collision granites such as; high values of SiO<sub>2</sub>, Ga/Al, Zr, Nb, Ga and Y and low values of CaO, MgO, Ba, and Sr.
- iii- Tectonically, they are consistent with the within-plate tectonic setting (figure 6) [32]. This conclusion is in integration with the field observations where the studied granites cross-cut the Late Neoproterozoiccalcalkaline rocks and represent the youngest igneous activity in the studied area as well as their petrographic studying pointed to presence of the sodic amphiboles indicating their alkaline and/or peralkaline affinities of within-plate tectonic setting







Fig (5): Binary variation diagram of Nb vs. Ga/Al for the studied granites (Whalen et al., 1987).





Fig (6): Binary variation diagram of Y+Nb vs. Rb for the studied granites (Pearce et al., 1984).

#### The radiometric characteristics

As the<sup>226</sup>Ra and <sup>232</sup>Th are not directly gamma emitters, their activity concentrations ( $A_{Ra}$  and  $A_{Th}$ ) were identified through the gamma rays of their decay products <sup>214</sup>Pb and <sup>208</sup>Tl respectively. The  $\gamma$ -ray photo peaks<sup>214</sup>Pb (351 keV) and <sup>208</sup>Tl (2614.5 keV) were to identify the  $A_{Ra}$  and  $A_{Th}$  respectively [33]. On the other hand, the activity concentration of 40K was directly recognized from its single peak of 1460 keV [34].

The activity concentration (A) in Bqkg<sup>-1</sup>of each radionuclide in the granitic samples was calculated according to the following equation [35].

$$A = Np / (e x \eta x m)....(1)$$

where **Np** = the (cps) sample - (cps) background, "**e**" is the abundance of the  $\gamma$ -line in a radionuclide, "**n**" is the measured efficiency for each gamma-line observed for the same number of channels either for the sample or the calibration source and "**m**" is the mass of the sample in kilograms. The calculated activity concentrations (Bqkg<sup>-1</sup>) of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K are shown in table (3).

Rock type	S. Code	<sup>226</sup> Ra	<sup>232</sup> Th	<sup>40</sup> K
Alkali feldspar pink granite	M2	27.44	28.74	1233.38
	M4	38.83	30.07	678.42
Syenogranite	W2	53.16	32.74	926.05
	W3	61.73	51.00	1154.87
Alkaline granite	KOR6	32.46	27.72	1455.1
Alkali feldspar granite to alkali	KATH12	71.26	50.68	1315.04
granite	KATH14	74.39	53.26	1275.58
	KATH15	73.47	55.85	1322.60
greenish blue grey alkaline	HAR16	35.34	27.47	1228.93
granite	HAR17	37.95	31.31	1253.99

Table (3): The activity concentration (Bqkg<sup>-1</sup>) of the investigated samples

• M2 & M4 (JabalMaslukh), W2 &W3 (JabalWaridat), KOR6 (JabalKawr), KATH 12, 14 & 15 (Jabal Al-Khathami) and HAR 16 & 17 (JabalHarashef).

Distribution of the measured activity concentrations showed that Al-Khathami granite contains the highest recorded radioactivity while both Waridat and Kawr granites revealed the lowest activity

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concentrations (figure 7). Such distribution seems to be compatible with the relative abundance of the accessory minerals in the studied granites as indicated from the petrographic study where some of these minerals (particularly zircon) are known by their hosting for the radioactive elements into their crystal lattices.



Fig (7): Distribution of the radioactivity concentration of <sup>226</sup>Ra and <sup>232</sup>Th (Bq kg<sup>-1</sup>) in the investigated granites

#### Origin of the radioactivity

Usually, the radionuclides are related to some minerals in the granitic rocks either those identified as uranium and thorium minerals (e.g. uraninite, uranophane, thorite....etc.) or some accessory minerals such as zircon, monazite and sphene. The current petrographic investigation pointed to absence of any uranium and thorium minerals while it recorded sensible presence of the zircon mineral in all the studied granitic rocks. In similar regard, the geochemical study revealed relative high Zr concentrations in these granites. Consequently, the binary relations between Zr-<sup>226</sup>Ra and Zr-<sup>232</sup>Th were constructed (figures 8 & 9) to testify the role of zircon mineral on the radioactivity of the Raniah granitic masses. The achievable correlation values of Zr-<sup>226</sup>Ra and Zr-<sup>232</sup>Th were 0.92 and 0.87 respectively which strongly support that zircon mineral is the main source of the measured radioactivity in the investigated granites. Also, it represents respectful evidence on the magmatic origin of the parent uranium and thorium elements.



Fig (8): The binary relation of Zr-<sup>226</sup>Ra in the studied granites

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Fig (9): The binary relation of Zr-<sup>232</sup>Th in the studied granites

#### Radium equivalent activity (Bqkg<sup>-1</sup>)

The <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K emit different  $\gamma$ -doses even if present in the same amount in any material. So the radiation hazards of a material are estimated by calculating the net effect of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K present in the material as radium equivalent activity (Ra<sub>eq</sub>) which is calculated according to the following equation [36]:

 $Ra_{eq} = 370x[(A_{Ra}/370)+(A_{Th}/259)+(A_{K}/4810)]...(2)$ 

Where  $A_{Ra}$ ,  $A_{Th}$  and  $A_K$  represent the activity concentrations of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K as (Bq kg<sup>-1</sup>) respectively. It is based on the assumption that 370 Bq kg<sup>-1</sup> of <sup>226</sup>Ra, 259 Bq kg<sup>-1</sup> of <sup>232</sup>Th, and 4810 Bq kg<sup>-1</sup> of <sup>40</sup>K produce the same  $\gamma$ -radiation dose rate [37]. The highest value of the (Ra<sub>eq</sub>) was given by Al-Khathami granite (255.13 Bqkg<sup>-1</sup>) while the lowest value was recorded byMaslukh granite (134.07 Bqkg<sup>-1</sup>). Generally, the calculated Ra<sub>eq</sub> values of all the studied granitic samples (table 4) are less than the world's average of 370 Bq kg<sup>-1</sup>[38] and meet the recommended limit [37].

#### The absorbed dose rate (D)(nGy h<sup>-1</sup>)

The measured activity of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K were converted into doses (nGyh<sup>-1</sup>Bq<sup>-1</sup>kg<sup>-1</sup>) by applying the factors 0.462, 0.604 and 0.0417 for radium, thorium and potassium, respectively [39]. These factors were used to calculate the total absorbed gamma dose rate in air at 1m above the ground level for the uniform distribution of the naturally occurring radionuclides using the equation 3. Where;  $A_{Ra}$ ,  $A_{Th}$  and  $A_{K}$  are the activity (Bqkg<sup>-1</sup>) of radium, thorium and potassium in the samples respectively [35].

 $D (nGyh^{-1}) = 0.462A_{Ra} + 0.604A_{Th} + 0.0417A_{K}$ .....(3)

The calculated values of absorbed dose rates in the examined granitic samples (table 4) range between 64.39 nGyh<sup>-1</sup> in Maslukh granite and 122.83 nGyh<sup>-1</sup> in Al-Khathami granite. Six D-values are found to be within the global range of 18-93 nGyh<sup>-1</sup> [39] and the other calculated D values (one sample from Waridat granite and three samples from Al-Khathamy granite) are relatively higher than the global range.

Table (4): The estimated radioactive indices of Raniah granitic masses; radium equivalent (Raeq), adsorbed
dose rate (D), external hazard index ( $H_{ex}$ ) and internal hazard index ( $H_{in}$ ).

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	Акан тегозраг рітк	IVIZ	103.51	81.47	0.442	0.516	
	Rock type	S. No.	Ra(eq)	(D) (nGy <sup>-1</sup> )	Hex	Hin	
		<b>a</b>	- / \	(-)			



granite	M4	134.07	64.39	0.362	0.467
Syenogranite	W2	171.28	82.79	0.463	0.606
	W3	223.58	107.48	0.604	0.771
Alkaline granite	KOR6	184.14	92.42	0.497	0.585
Alkali feldspar	KATH12	244.99	118.37	0.662	0.854
granite to alkali	KATH14	248.77	119.73	0.672	0.873
granite	KATH15	255.13	122.83	0.689	0.888
greenish blue grey	HAR16	169.25	84.17	0.457	0.553
alkaline granite	HAR17	179.28	88.74	0.484	0.587

#### The internal hazard index (H<sub>int</sub>.)

Several indices have been proposed to assess the exposure level due to radon inhalation originating from building materials [40]. The internal hazard index is defined as:

 $H_{int} = (A_{Ra}/185) + (A_{Th}/259) + (A_{K}/4810)...(4)$ 

where  $A_{Ra}$ ,  $A_{Th}$  and  $A_K$  are the activity concentration (Bqkg<sup>-1</sup>) of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K respectively. The recommended value of the H<sub>int</sub> should be  $\leq 1[40]$ .

The studied granitic samples showed  $H_{int}$  values range between 0.467 and 0.888 (table 4) which reflect their suitability in the civilian uses.

#### The external hazard index (Hext.)

The external hazard index ( $H_{ext}$ ) due to the emitted gamma rays is defined in order to examine the applicability of using materials in construction. For a typical material it is given by the following expression [40]:

$$H_{ext.}=(A_{Ra}/370) + (A_{Th}/259) + (A_{K}/4810)...(5)$$

where  $A_{Ra}$ ,  $A_{Th}$  and  $A_K$  are the activity concentration (Bqkg<sup>-1</sup>) of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K respectively. The recommended value of the H<sub>ext</sub>should be  $\leq 1$ .

The studied granitic samples showed H<sub>int</sub> values range between 0.362 and 0.689 in JabalMaslukh and JabalKhathamy respectively (table 4) which support the applicability of the examined granites in construction, decoration and ornamentation purposes.

#### The annual effective dose

The annual effective dose is of two types. The annual outdoor effective dose ( $E_{out}$ ) and annual indoor effective dose ( $E_{in}$ ). To estimate annual effective doses, account must be taken into consideration (i) the conversion coefficient from absorbed dose in air to effective dose and (ii) the outdoor and indoor occupancy factors. The annual estimated average effective dose equivalent received by a member is calculated using a conversion factor of 0.7 Sv Gy<sup>-1</sup>, which is used to convert the absorbed rate to annual effective dose with an outdoor occupancy of 20% and 80% for indoors [41].

#### a- The annual outdoor effective dose (Eout)

The yearly estimated outdoor effective dose ( $E_{out}$ ) received by the public depends on the adsorbed dose rate (D), the dose conversion factor (0.7 Sv Gy<sup>-1</sup>) and the approximate staying time in the outdoor (20% of the total hours per a year). The annual indoor effective dose ( $E_{out}$ ) is calculated according to following equation [39];



#### $E_{out}(mSv) = D \times 0.7 \times 0.2 \times 8760 \times 10^{-6}...(6)$

For the measured granitic samples, the lowest  $E_{out}$  value (0.079 mSv y<sup>-1</sup>) was recorded in JabalMuslukh granite while the highest value (0.151 mSv y<sup>-1</sup>) was related to JabalKhathamy granite (table 5).

# Table (5): The estimated radioactive indices of Raniah granitic masses; outdoor annual effective dose (E<sub>out</sub>), indoor annual effective dose (E<sub>in</sub>) total annual effective dose (E<sub>total</sub>), gamma index (I<sub>γ</sub>), alpha index (I<sub>α</sub>) and excess lifetime cancer risk (ELCR).

Rock type	S. No.	E <sub>out</sub> (mSv)	E <sub>in</sub> (mSv)	E <sub>total</sub> (mSv)	$I_{\hat{Y}}$	lα	ELCR
Alkali feldspar pink	M2	0.099	0.399	0.498	0.646	0.137	1.62
granite	M4	0.079	0.316	0.395	0.506	0.194	1.28
Syenogranite	W2	0.102	0.406	0.508	0.650	0.266	1.65
	W3	0.132	0.527	0.659	0.846	0.309	2.14
Alkaline granite	KOR6	0.113	0.453	0.566	0.732	0.162	1.84
Alkali feldspar	KATH12	0.145	0.581	0.726	0.929	0.365	2.35
granite to alkali	KATH14	0.147	0.587	0.734	0.939	0.372	2.38
granite	KATH15	0.151	0.603	0.754	0.965	0.367	2.45
greenish blue grey	HAR16	0.103	0.413	0.516	0.665	0.177	1.68
alkaline granite	HAR17	0.109	0.435	0.544	0.701	0.190	1.77

#### b- The annual indoor effective dose (Ein)

The ( $E_{in}$ ) is the dose which a person receives in the indoor environment. Like in  $E_{out}$ , the ( $E_{in}$ ) depends on the adsorbed dose rate (D) and the dose conversion factor (0.7 Sv Gy<sup>-1</sup>) but the staying time in the indoor was estimated to be 0.8 (80% of the total hours per a year). Accordingly, the annual indoor effective dose ( $E_{in}$ ) is calculated according to following equation [39];

$$E_{in}(mSv) = D \times 0.7 \times 0.8 \times 8760 \times 10^{-6}...(7).$$

The calculated  $E_{in}$  in the studied granites ranged from 0.316 mSv y<sup>-1</sup> for JabalMuslukh granite and 0.603 mSv y<sup>-1</sup> in Khathamy granite **(table 5).** 

Based on the estimated  $E_{out}$  and  $E_{in}$  values in the Raniah granitic masses, the total annual effective dose ( $E_{out} + E_{int}$ ) was found to be ranged between 0.395 mSv y<sup>-1</sup> (Muslukh granite) and 0.754 mSv y<sup>-1</sup> (Khathamy granite).

All the calculated  $E_{out}$ ,  $E_{in}$  and the total annual effective doses of the investigated granitic masses showed their lowering than the safety criterion limit of 1 mSv y<sup>-1</sup> as recommended safety limit for general public [42](figure 10).





Fig (10): Showing the annual outdoor effective dose (E<sub>out</sub>), the annual indoor effective dose (E<sub>in</sub>) and the annual total effective dose (E<sub>total</sub>) relative to the recommended safety criterion of annual effective dose (1 mSv y<sup>-1</sup>).

#### Gamma index (I $\gamma$ ) and alpha index (I $\alpha$ ):

To verify whether the natural building materials could/couldn't be an additional source of exposure to the population, two radiological parameters called the gamma index  $(I_{\gamma})$  and alpha index  $(I_{\alpha})$  were proposed [43-44] where;

 $I_{Y}=(A_{Ra}/300) + (A_{Th}/200) + (A_{K}/3000) \le 1....(8)$  $I_{\alpha} = A_{Ra}/200 \le 0.5....(9)$ 

On the otherwise, most countries applied their own control on the upper limit, the Europe commission (EC) introduced a two dose criteria for the gamma dose of building materials: an exception basic of 0.3 mSv year<sup>-1</sup> and a maximum cut-off of 1 mSv year<sup>-1</sup> [43]. In the event that the exclusion level of 0.3 mSv year<sup>-1</sup> is viewed, the estimates values of  $I_{\gamma}$  ought to be beneath 0.5 for materials used in bulk like granite, cement, sand....etc., while if the upper level of 1 mSv year<sup>-1</sup> is observed, the values of  $I_{\gamma}$  ought to be underneath 1 for such materials. Generally, the advisable values of  $I_{\alpha}$  and  $I_{\gamma}$  are beneath 0.5 and 1, respectively [45&43].

In the Raniah granitic masses, the calculated values of  $I_y$  were found to be ranged between 0.506 mSv year<sup>-1</sup> (Maslukh granite) and 0.965 mSv year<sup>-1</sup> (Khathmi granite) while the estimated values of  $I_\alpha$  were ranged between 0.137 mSv y<sup>-1</sup> (Maslukh granite) and 0.372 mSv y<sup>-1</sup> (Khathami granite) (table 5). In general, the obtained data points to that both the  $I_\alpha$  and  $I_y$  values are less than the advisable values which mean that the investigated granitic rocks do not cause additional source of radioactive exposure for the public.

#### Excess lifetime cancer risk (ELCR)

Based upon calculated values of annual effective dose excess lifetime cancer risk (ELCR) was calculated using the following equation [46];

 $ELCR = E_{total} \times LE \times RF.....(10)$ 

where  $E_{total}$  is the annual effective dose, LE is the life expectancy (65 years) and RF (Sv<sup>-1</sup>) is fatal risk factor per Sievert, which is 0.05 [52]. The ELCR ranged between 1.28 x 10<sup>-3</sup> (Maslukh granite) and 2.45 x 10<sup>-3</sup>

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(Khathami granite). The ELCR values of the studied rocks (table 5) are approximately similar to or slight higher than the world's average of  $1.45 \times 10^{-3}$  [46].

## Table (6): Comparison between the activity concentrations (Bqkg<sup>-1</sup>) of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K in the studied granites and other worldwide granites.

	<sup>226</sup> Ra	<sup>232</sup> Th	<sup>40</sup> K	Reference
Raniah granites (average values)	50.6	38.9	1184.4	Present study
France	90	80	1200	[47]
Taiwan	42	73	1055	[48]
India	82	112	1908	[49]
Greece	67	95	1200	[50]
Egypt	18	24	350	[51]
Brazil	48.6	288.2	1335	[52]
KSA	23	30	340	[53]
Earth crust	35	30	400	[39]

### Table (7): Comparison between the estimated values of radium equivalent (Ra<sub>eq</sub>), adsorbed dose rate (D), total annual effective dose (E<sub>total</sub>) gamma index (I<sub>Y</sub>) in the studied granites and other worldwide granites

	Ra <sub>eq</sub> (Bqkg <sup>-1</sup> )	D (nGyh⁻¹)	E <sub>total</sub> (mSv)	Ι <sub>γ</sub>	Reference	
Raniah granites (average values)	197.4	96.24	0.59	0.76	Present study	
Russian Federal (typical-maximum values)	93-269			0.34-0.98		
Europe (typical-maximum values)	196-1263			0.71-4.55		
India (typical-maximum values)	80-399			0.27-1.47		
Egypt (typical-maximum values)	81-226			0.30-0.82	[54]	
Saudi Arabia	69-165			0.25-0.60		
Vietnam (local granite)		224.6	1.1	0.94	[55]	
Spain (Rosa porrino)		153.9	1.4	2.96		
Brazil (Topazio)		95.7	0.8	1.51	[56]	

**To evaluate** the radioactivity risks of the studied granitic masses, the radioactivity concentrations of the measured radionuclides as well as some calculated radioactive indices were compared to their corresponding values of other granitic rocks in many countries around the world where they are used as natural building materials (tables 6 & 7). The comparison pointed to those radioactive concentrations and the estimated indices of the Raniah granitic bodies are clearly lower than most of the compared granites which supports the safe using of the studied granites in the different construction and ornamentation purposes.

#### CONCLUSION

**B**ased on the obtained criteria, it can be concluded that all the studied granitic masses will not impose excessive radiation exposure for public and will not increase the radiation levels of the surrounding environment. Consequently, these granites are suitable to the economical exploitation without needing for more restriction actions and/or precautions and they are quite safe from the radioactivity point of view.

Finally, some recommendations shall be delivered based on this study:

- 1- Extending the radiometric survey to involve all the granitic masses found on the KSA land particularly those at the eastern part of the Arabian-Nubian Shield.
- 2- Construction of a radioactive map for the KSA land and formulate the own criteria of the radiometric indices based on the local conditions and guided by the international permissible levels.



- 3- Delivering more attention and studies to cover all the natural resources e.g. sand, cement, gravel, marble...etc. as well as the water resources that are domestically used to evaluate their suitability from the radioactivity point of view.
- 4- Encouraging El-Taif University to extend its contribution to similar environmental trends which will support its contribution in serving the national development and the society issues.

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