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Reducing of radiation activity index for quarry dust waste located in industrial zone for marble and granite Shaq El Thobaan Egypt

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Abstract

Building materials industry like marble and granite, ceramics, cement, and bricks produce a huge amount of wastes that have an adverse effect on the environment. The protection of the environment and inhabitants from harmful wastes received more attention from environmentalists all over the world. Recycling and reuse of industrial wastes in different industries is a very important trend to keep the environment and inhabitants from harmful effects. The aim of the present study is to evaluate the activity index of radionuclides of quarry dust (marble and granite) from Shaq El Thobaan region Cairo, Egypt. The samples of marble and granite were analyzed for U, Th, and K in a radiation measurement laboratory (Institute of graduated studies and researches) at Alexandria university using a pre-calibrated high-resolution gamma-ray spectrometer. The radiation activity for marble waste samples equal to 0.296 less than 1 but for granite samples equals 1.488 more than 1 the permissible limit. So, samples of granite dust mixed with a mixture of cement and sand (1:3) with 10%, 20%, 30%, 40%, 50%, and 60%. the results showed that the activity index for radiation 0.552, 0.656, 0.760, 0.864, 0.967and 1.07. So, the granite can have used up to 50% in cement mixtures.

Key Words

Marble dust, granite dust, quarry dust, Radiation activity, Shaq El Thobaan Region.

1- Introduction

Human beings are continuously exposed to radiation of natural origin. The main sources of such exposure are the cosmic rays and the terrestrial natural radionuclides (such as ^{238}U radionuclides) that are occurring at trace levels in the Earth's crust. The exposure to radiation of terrestrial origin depends mainly on the nature, origin and geology of the ground formation. The main contributors to the radiation exposure from terrestrial origin are the long-lived ^{238}U-series and ^{232}Th-series, in addition to ^{40}K [1]. Humans are daily exposed to radionuclides that occur naturally in the environment. The distribution of naturally occurring different radionuclides depends on two main factors, the first one is the distribution of originating rocks and the second is processes which result to their removal and extraction from the soil and their migration [2]. Since the Earth formed and life developed, background radiation has been our constant companion. Primordial radionuclides are found around the globe in igneous and sedimentary rock. These radionuclides migrate from rocks into soil, water, and even air. Human activities such as uranium mining have also redistributed these radionuclides. Primordial radionuclides include the series of radionuclides produced when uranium (^{238}U) and thorium decay (^{232}Th) as well as potassium (^{40}K) [3]. Natural radiation refers to ionizing radiation originating either from high energy cosmic rays entering the earth's atmosphere from outer space or from Naturally Occurring Radioactive Materials (NORM) present in the crust of the earth. This radiation is distinguished from artificial radiation produced through man-made nuclear or atomic transformations. The exposure of human beings to a background of natural radiation is a continuing and inescapable feature of life on earth. The effective dose due to this ionizing radiation for members of the public varies substantially depending on where they live, occupation, personal habits, diet, building type and house utilization pattern [4].

Humans are continuously exposed to ionizing radiation which is widely spread in the earth 'environment due to naturally occurring radioactive materials (NORM)which exists everywhere; earth crust, soils, rocks, water, air and building materials. NORM is the main source of external and internal radiation exposures due to ɣ- rays and α-particles emitted from uranium series (^{238}U), thorium series (^{232}Th), and radioactive potassium nucleus (^{40}K). The materials that maintain high concentration of NORM, have a theoretical potential to be carcinogenic. Therefore, measuring of radioactive materials in environmental samples is of primary importance from the view point of radiation protection in the environment [5].

All building materials and products derived from rock and soil contain various amount of mainly natural radionuclides of ^{238}U (Uranium), ^{232}Th (Thorium) and ^{40}K (Potassium) [6]. Ceramic tiles may be glazed or unglazed. Zircon is a common opacifying constituent of glazes applied to ceramic tiles and is also used as an opacifier in porcelain tiles by incorporating due to the presence of zircon in the glaze, ceramic tiles can show elevated concentration of natural radioactivity. The possibility of the activity concentration associated with ceramic tiles to elicit anomalous response from body tissues was assessed by determining the corresponding Gonadal dose equivalent as well as the excess lifetime cancer risk [7].
Problems in Industrial Zone of marble and granite in Shaq El Thobaan regions are outbreak of a group of diseases (e.g.: tinea, intestinal colic and chest disease) among workers in Shak El Thobaan as a result of drinking water and food contamination. Marble and granite industry have stone waste in generally a highly polluting waste due to both its highly alkaline nature and its manufacturing and processing techniques, which impose a health threat to the surroundings [4]. The internal radiation exposure, affecting the respiratory tract, is due to radon and its decay products which emanate from building materials. Because most of the people spend 80% of their time indoors, so we pay attention to the low-level exposure from naturally occurring radionuclides. It is well known that, the long exposures to low levels of ionizing radiation can seriously increase health risks to humans [8]. Long term exposure to elevated concentration of radon gas and its daughter products can lead to functional changes in respiratory organs and lung cancer [9]. Radiation exposure has been associated with many forms of leukemia and with cancers of many organs such as lung, breast and thyroid [10].

The aim of the present study:

- Determination of activity index in both marble and granite dust wastes.
- Reducing the activity concentration of marble and granite dust wastes to achieve the allowable limits.
- Protecting inhabitants healthy from the harmful exposure to radioactive materials.
- Reducing environment pollution from serious industrial waste by products through its utilization as additives in building materials.

2- Material and Method

2-1 Materials

2-1-1 Marble dust waste

Marble dust waste is produced from the manufacture of stone blocks which accumulated waste with a great amount. Marble waste was obtained from industrial zone for marble and granite Shaq El Thobaan Region Cairo -Egypt.

2-1-2 Granite dust waste

Granite dust waste is produced from the manufacture of stone blocks which accumulated waste with a great amount. Granite waste was obtained from industrial zone for marble and granite Shaq El Thobaan Region Tura El Cairo -Egypt.

2-1-3 Sand

The sand used in the present study was tested according to standard Egyptian specification [11].

2-1-4 Cement

The cement used in all mixes was tested according to standard Egyptian specification [12]., British standard specification [13] and A.S.T.M. standard specification [14].

2-2 Methods

Five samples of marble dust waste, four samples of granite dust waste were analyzed for measuring specific activity of radio-nuclides e.g. ^{234}Th, ^{226}Ra, ^{214}Pb, ^{214}Bi (^{238}U), ^{212}Bi,^{208}Tl (^{232}Th), ^{40}K and ^{137}Cs. Also, samples of cement and sand (1:3) were loaded with 10%, 20%, 30%, 40%, 50% and 60% of granite dust waste as a replacement of cement and sand and analyzing for measuring the pre-mentioned radio-nuclides. Samples were measured using a pre- calibrated high-resolution gamma ray spectrometer in radiation measurement laboratory (Institute of graduated studies and researches) Alexandria university. The spectrometer was based on closed – end coaxial. Hyper pure germanium detector (Model CS20-APTEC) with 108 cm^{3}sensitive volume and 5.40cm diameter.

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3- RESULTS AND DISCUSSION

Concentration of radio-nuclides in both marble and granite dust wastes are shown in table (1). Activities indexes are used to assess whether the safe requirements are being fulfilled. For construction materials the activity indexes are calculated on the measured concentrations of (^{40}Ra), Thorium(^{232}Th) and potassium(^{40}K) [15]. The result of radiation activity index in for both marble and granite dust wastes were calculated using the following equation:

$$I = \frac{CTh}{200} + \frac{CRa}{300} + \frac{CK}{3000}$$

Where: I: Activity index

C_{Th}: Concentration of Thorium (in Bq kg-1)

C_{Ra}: concentration of Radium (in Bq kg-1)

C_{K}: Concentration of Potassium (in Bq kg-1)

Table (1) Concentration of radio-nuclides in Mable dust and granite dust samples

Nuclide	Concentration activity Bq Kg^{-1}	
	Marble dust waste	**Granite dust waste**
^{40}K	62.527 ± 0.264	1165.313 ± 1.138
^{137}Cs	0.801 ± 0.030	0.210 ± 0.015
^{208}Th	2.332 ± 0.051	15.27 ± 0.130
^{210}Pb	32.959 ± 0.191	12.39 ± 0.117
^{212}Bi	9.433 ± 0.069	15.40 ± 0.131
^{212}Pb	4.29 ± 0.142	64.505 ± 0.268
^{214}Bi	18.029 ± 0.140	32.58 ± 0.190
^{214}Pb	17.725 ± 0.140	30.92 ± 0.185
^{226}Ra	38.47 ± 0.210	95.098 ± 0.325
^{228}Ac (^{232}Th)	29.48 ± 0.181	156.536 ± 0.417
^{234}Th	10.532 ± 0.110	4.529 ± 0.071

The main nuclides in building materials are ^{232}Th, ^{226}Ra and ^{40}K

$$\text{Activity index (I)} = I = \frac{CTh}{200} + \frac{CRa}{300} + \frac{CK}{3000}$$

$$\text{I marble dust waste} = \frac{29.48}{200} + \frac{38.47}{300} + \frac{62.527}{3000} = 0.296 < 1$$ (Markkanen, 1995)[14]

$$\text{Activity index (I)} = \frac{CTh}{200} + \frac{CRa}{300} + \frac{CK}{3000}$$

I granite dust waste = $\frac{156.536}{200} + \frac{95.098}{300} + \frac{1165.315}{3000}$ = 1.488> 1 (Markkanen, 1995)[14]

From the previous calculations the activity index of marble dust waste equal to 0.296 < 1 within the permissible limits but of granite dust waste equal to 1.488 > 1 that exceeds the permissible limits. The obtained results are satisfied with that carried by [13] and [16].

In order to reduce the activity index of granite dust waste, samples of cement and sand (1:3) were prepared and loaded with 10%, 20%, 30%, 40 %, 50% and 60% of granite dust waste and analyzing for measuring radio-nuclides. The concentration activity for samples are shown in table (2) and table (3).

Table (2) Concentration of radio-nuclides in cement and sand with 10%, 20%, 30% granite dust waste

Nuclide	Concentration activity Bq Kg^{-1}		
	10% granite D W	**20% granite D W**	**30% granite D W**
^{40}K	205.25 ± 0.478	311.923 ± 0.590	418.597 ± 0.682
^{137}Cs	0.652 ± 0.027	0.603 ± 0.026	0.554 ± 0.025
^{208}Th	9.655 ± 0.104	10.279 ± 0.107	10.903 ± 0110
^{210}Pb	16.523 ± 0.135	16.331 ± 0.135	15.838 ± 0.133
^{212}Bi	30.190 ± 0.183	28.519 ± 0.178	26.879 ± 0.173
^{212}Pb	22.016 ± 0.156	26.737 ± 0.172	31.458 ± 0.187
^{214}Bi	45.618 ± 0.226	43.677 ± 0.220	42.290 ± 0.217
^{214}Pb	45.538 ± 0.225	43.913 ± 0.221	42.289 ± 0.217
^{226}Ra	91.550 ± 0.319	91.944 ± 0.320	92.338 ± 0.320
^{228}Ac (^{232}Th)	35.723 ± 0.200	49.147 ± 0.234	62.570 ± 0.264
^{234}Th	29.990 ± 0.183	49.147 ± 0.234	21.998 ± 0.156

Table (3) Concentration of radio-nuclides in cement and sand with 40%, 50%, 60% granite dust waste

Nuclide	Concentration activity Bq Kg^{-1}		
	40% granite D W	**50% granite D W**	**60% granite D W**
^{40}K	525.271 ± 0.764	631.945 ± 0.840	738.618 ± 0.906
^{137}Cs	0.505 ± 0.024	0.456 ± 0.023	0.406 ± 0.021
^{208}Th	11.527 ± 0.113	12.151 ± 0.116	12.774 ± 0.119
^{210}Pb	15.346 ± 0.131	14.853 ± 0.128	14.360 ± 0.126
^{212}Bi	25.239 ± 0.167	23.600 ± 0.162	21.960 ± 0.156
^{212}Pb	36.179 ± 0.200	40.900 ± 0.213	45.621 ± 0.225
^{214}Bi	40.903 ± 0.211	39.516 ± 0.210	38.128 ± 0.206

^{214}Pb	40.665 ± 0.213	39.041 ± 0.210	37.416 ± 0.204
^{226}Ra	92.732 ± 0.321	93.127 ± 0.322	93.521 ± 0.322
^{228}Ac (^{232}Th)	75.994 ± 0.291	89.418 ± 0.315	102.842 ± 0.338
^{234}Th	19.503 ± 0.147	17.007 ± 0.137	14.511 ± 0.127

Activity index (I) = $\frac{CTh}{200} + \frac{CRa}{300} + \frac{CK}{3000}$

I for 10% granite = $\frac{35.723}{200} + \frac{91.55}{300} + \frac{205.25}{3000}$ = 0.552

I for 20% granite = $\frac{49.147}{200} + \frac{91.944}{300} + \frac{311.923}{3000}$ = 0.656

I for 30% granite = $\frac{62.57}{200} + \frac{92.338}{300} + \frac{418.597}{3000}$ = 0.76

I for 40% granite = $\frac{75.994}{200} + \frac{92.732}{300} + \frac{525.721}{3000}$ = 0.864

I for 50% granite = $\frac{89.418}{200} + \frac{93.127}{300} + \frac{631.945}{3000}$ = 0.968

I for 60% granite = $\frac{102.842}{200} + \frac{93.521}{300} + \frac{738.618}{3000}$ = 1.07

Table (4): The activity index for mixtures of granite and mixture of cement and sand for different ratios

%ge of granite dust	%ge of cement and sand (1:3)	I
60%	40%	1.07
50%	50%	0.968
40%	60%	0.864
30%	70%	0.760
20%	80%	0.656
10%	90%	0.552

When adding 60% of granite dust waste to the mixture of cement and sand (1:3) as replacement reduces the activity index from 1.488 to 1.07. Replacing 50% of granite dust waste with the mixture of cement and sand (1:3) reduces the activity index from 1.07 to 0.968. Replacing 40% of granite dust waste with the mixture of cement and sand (1:3) reduces the activity index 0.968 to 0.864. Replacing 30% of granite dust waste reduces the activity index to 0.760, Replacing 20% granite dust waste reduces the activity index to 0.656. Replacing 10% of granite dust waste reduces the activity index to 0.552.

Radiological effects of external exposure can be assessed for naturally occurring radioactive material (NORM) ^{238}U. ^{232}Th and ^{40}K by deducing the radium equivalent (Ra_{eq}) calculated from the following equation:

$Ra_{eq} = C_{Ra} + 1.43\, C_{Th} + 0.077\, C_K$ according to [17] .

Where:

Ra_{eq}: Radium equivalent in Bq Kg^{-1}

C_{Ra}: The specific activity of ^{226}Ra (^{238}U) in Bq Kg^{-1}

$C_{Th:}$ The specific activity of ^{232}Th in Bq Kg^{-1}

C_k: The specific activity of ^{40}K in Bq Kg^{-1}

Radium equivalent for marble dust waste:

Ra_{eq} = 38.470 + 1.43* 29.480 + 0.077* 62.527 = 85.441 Bq Kg^{-1}

Radium equivalent for granite dust waste:

Ra_{eq} = 95.098 +1.43* 156.536 + 0.077* 1165.313 = 408.67 Bq Kg^{-1}

Radium equivalent for 60% granite and 40% cement and sand (1:3)

Ra_{eq} = 93.521 + 1.43* 103.842 + 0.077* 738.618 = 297.46 Bq Kg^{-1}

Radium equivalent for 50% granite and 50% cement and sand (1:3)

Ra_{eq} = 93.127 + 1.43* 89.418 + 0.077* 631.945 = 269.655 Bq Kg^{-1}

Radium equivalent for 40% granite and 60% cement and sand (1:3)

Ra_{eq} = 92.732 + 1.43* 75.994 + 0.77* 525.271 = 241.850 Bq Kg^{-1}

Radium equivalent for 30% granite and 70% cement and sand (1:3)

Ra_{eq} = 92.338 + 1.43* 62.57 + 0.077* 318.597 = 206.345 Bq Kg^{-1}

Radium equivalent for 20% granite and 80% cement and sand (1:3)

Ra_{eq} = 91.944 + 1.43* 49.147 + 0.077*311.923 = 186.242 Bq Kg^{-1}

Radium equivalent for 10% granite and 90% cement and sand (1:3)

Ra_{eq} = 91.550 +1.43* 35.723 + 0.077*205.250 = 158.438 Bq Kg^{-1}

Table (5): The activity index for mixtures of granite and mixture of cement and sand for different ratios

%ge of granite dust	%ge of cement and sand (1:3)	Ra_{eq}
100%	0%	408.670
60%	40%	297.46
50%	50%	269.655
40%	60%	241.850
30%	70%	206.345
20%	80%	186.242
10%	90%	158.438

Radiological assessment of radiation level in pure granite dust waste sample exceeds 370 Bq Kg^{-1} [18] which is the acceptable limit for safe use while, radiation level in pure marble dust waste sample is below the acceptable limit for safe use. Also, radiation level in cement and sand (1:3) and granite dust waste as replacement of 10% to 50% is below the acceptable limit. The results are in agreement with that calculated using the equation listed in (Markkanen.1995). The activity index for 60% granite as replacement of cement and sand (1:3) exceeds the acceptable limit (1Bq Kg^{-1}) while, the equivalent radium does not exceed the acceptable limit so, the use of granite dust waste with 50% as replacement of cement and sand (1:3) satisfy the accepted limit radiation.

4- Conclusion

Granite dust waste occupies a large area of land.

Granite dust waste is a main source of radiation where the activity index equal to 1.488 more than the acceptable limit (1).

The frequent exposure to granite dust waste during manufacturing processes leads to a lot of severe health hazards to workers and inhabitants around the manufacturing plants.

Protecting Environment from granite dust waste through its utilization as an ingredient in the production of building materials.

The use of granite dust waste with 50% as replacement of cement and sand (1:3) achieve the acceptable limit for both activity index and equivalent radium.

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