Determination of Radioactivity and its Level in Gemstones Beads used in Nigeria

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Abstract: The radioactivity concentrations of some stone beads used in Kano state, Nigeria, were determined using NaI (TI) gamma ray spectrometry. The radionuclide detected were found to be that of natural ones normally associated with rocks which are ²²⁶Ra, ²³²Th and ⁴⁰K. The radiation hazards associated with these radionuclide were accessed in which the radium equivalent activities of the samples were found to be lower than the maximum recommended limit of 370 Bqkg⁻¹, the representative level index and the external hazard values were lower than unity except for the Amethyst and Topaz sample which is having 1.62 and 1.4 respectively. This study shows that the samples are found to be radiation hazard free. Stone beads with higher RLI values should have special management and treatment to reduce the radioactive hazard that will comply with the environmental protection regulations and to maintain the indexes values within the accepted levels. **Keywords:** Stone beads, Radioactivity, Radiological indices.

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I. Introduction

Radiation is a transmission of energy either as an electromagnetic wave (γ -rays, X-rays, ultraviolet rays, microwaves and radio waves) or as a stream of moving particles (alpha, beta, and neutrons) [1]. As these particles pass through matter, they lose energy, in some situations, breaking molecular bonds and creating ions (hence the term ionizing radiation). It is well known that excessive exposure to radiation can destroy tissues. In mild cases it results in a burn, as with common sunburn. Greater exposure can cause very severe illness or death by a variety of mechanisms, including massive destruction of tissue cells, alteration of genetic material and destruction of the components in bone marrow that produced red blood cells [2].

Bead is a small piece of hard material with a hole through it, used for putting together with others on a string for sewing onto material worn around the neck or wrist. The Stone bead is made from Gemstone or precious or jewel stone. Gemstone is a piece of crystal which is cut into different smaller sizes, polished and used as jewelry or Ornamentals (International Gemologist Society [3].These crystals are obtained from rocks; they are hard, translucent, luster etc. [3].The chemical compositions of gemstones include; Silicates, Oxides, Carbonates, Sulfides, Halides, Phosphate, Sulphate, etc. [4].

Gems are frequently subjected to various treatments in order to improve their appearance in terms of color and transparency, and hence increase their commercial value. Color is the most important property of gemstones; they are frequently treated to alter their color to good or excellent results. Usually by different techniques such as dyeing, heating and irradiation treatment. Irradiated gems especially those irradiated through bombardment with either neutrons or electrons can make the gems radioactive. After irradiation, the stones are typically required to set aside for a couple of months to allow any radioactivity to decay. Many gemstones currently are imported and distributed by a number of companies without any regulation or distribution license according to United State Nuclear Regulatory Commission [5].

In the present work, the level and concentration of radionuclide's activity in seven samples of gemstones materials that are commonly used as jewelries in Kano state, Nigeria, were determined by means of gamma-ray spectrometry. The potential radiological hazards associated with these materials were also assessed by calculating their Radium equivalent (R_{eq}) activity, Representative level index (RLI) and external hazard index (H_{ex}).

II. Materials and Method

Sample collection and preparation

A total of seven samples of stone beadscommonly sold and used in Kano state of Nigeriawere purchased from the sellers. The samples are: Agate, Amethyst, Cat's eye, Coral, Topaz and Quartz. The mass of each sample is 450g which were washed, dried in the sun and crushed to fine powder with the use of pulverizer. the powdered samples were homogenized and dried at 110°C in an oven for complete removal of moisture. The samples were catalogued and Packaged into radon-impermeable cylindrical plastic containers which were

selected based on the space allocation of the detector vessel which measures 7.6cm by 7.6cm in dimension. To prevent ²²²Rn escaping, the packaging in each case was triple sealed. The sealing process included smearing of the inner rim of each container lid with Vaseline jelly, filling the lid assembly gap with candle wax to block the gaps between lid and container, and tight-sealing lid-container with masking adhesive tape. Radon and its short-lived progenies were allowed to reach secular radioactive equilibrium by storing the samples for 30 days prior to gamma spectroscopy measurements [6],[7].

Radiometric analysis

The samples were subjected to gamma spectral analysis with a counting time of 29000 seconds. A 76x76mm sodium iodide NaI (Tl) detector crystal optically coupled to a photomultiplier tube (PMT) was used. The detector is enclosed in a 6cm lead shield with cadmium and copper sheets. This arrangement is aimed at minimizing the effects of background and scattered radiation. By performing an energy calibration, a relationship between channel number and gamma ray energy is determined. Each radionuclide has one or more identifying energy levels. Each peak produced and measured by the Gamma-Spec analyzer is matched to a radionuclide energy level. The detected concentrations of various radio nuclides activity were determined and reported in Becquerel per kilogram (Bq/Kg).

III. Results

The radioactivity concentration

The radionuclide detected were only that of natural radioactive isotopes which consist of: 226 Ra, 327 Th and 40 K, as shown in table 1.While the activity concentration, Radium equivalent, Representative level index and the External Radiation hazard were calculated using equation (1), (2), (3) and (4) as shown in table 2. The actual quantity of radioactivity is the amount of each radionuclide present in a gemstone is calculated as follows: For each spectral peak, the activity in Becquerel per kilogram for the radionuclide responsible for producing that peak is:

$$A = \frac{n}{t\gamma\epsilon ms}$$
(1)

Where; **A** is the concentration in Becquerel per kilogram, **n** is the number of counts (area under the spectral peak), **t** is the time (duration of the live count in seconds) and γ is the gamma yield (fraction of gamma rays released at the particular energy level per radioactive disintegration). ε is the efficiency (fraction of gamma rays at the specified energy that is completely absorbed into scintillator crystal), **m** is the gemstone sample mass in grams and **s** is the self-shielding correction factor (compensates for the density difference between the gemstone and the calibration source) [8].

Radium Equivalent Activity (R_{eq})

Radium equivalent concentration is the quantity representative of external γ irradiation dose associated with the material. In order to compare the specific activity of materials containing different amounts of ²²⁶Ra, ²³²Th, and ⁴⁰K, the radium equivalent activity \mathbf{R}_{eq} is used as defined by: [9] $R_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_{k}$ (2)

Where A_{Ra} re, A_{Th} and A_k are the activity concentrations in Bq kg⁻¹ of ²²⁶Ra, ²³²Th, and ⁴⁰K, respectively. The value of R_{eq} must be less than 370 Bqkg⁻¹ for the radiation hazard to be negligible [10].

Representative level index (RLI)

This is another radiation hazard index primarily used to estimate the gamma radiation associated with different concentrations of some specified radionuclide and can be expresses as:[11]

$$RLI = \frac{A_{Ra}}{150 \text{ BqKg}^{-1}} + \frac{A_{Th}}{100 \text{ BqKg}^{-1}} + \frac{A_k}{1500 \text{ BqKg}^{-1}}$$
(3)

This index is very important for quality control of γ -radiation annual effective doses and in monitoring radiation inside human body. The assessed values of RLI must be less than or equal to 1 to make sure that the stone beads are generally safe and hazard-free [12].

External Radiation Hazard (H_{ex})

The external radiation hazard (H_{ex}) The external radiation hazard, H_{ex} , for ²²⁶Ra, ²³²Th, and ⁴⁰K radionuclide was calculated using the equation:[11]. $H_{ex} = \frac{A_{Ra}}{370 \text{ BqKg}^{-1}} + \frac{A_{Th}}{258 \text{ BqKg}^{-1}} + \frac{A_{k}}{4810 \text{ BqKg}^{-1}}$ (4)

The result of H_{ex} should be less than or equal to unity (i.e. $H_{ex} \le 1$). Normally, the value of H_{ex} is equal to one, corresponds to the upper limit of \mathbf{R}_{eq} (370 Bqkg⁻¹). in order to make the radiation hazard insignificant, the values of H_{ex} should be lower than one[11],[12].

Table 1: The Radionuclide obtained in the samples						
S/N	Samples	Radionuclide				
1	Agate	²²⁶ Ra,				
2	Amethyst					
3	Aquamarine	²³² Th				
4	Cat's eye	40				
5	Coral	⁴⁰ K				
6	Topaz					
7	Quartz					

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Table 2: Activity concentration,	Radium equivalent,	Representative lev	el index :	and External	hazard index fo	r
	the selecte	ed Stone beads.				

		Activity concentration in (Bq/Kg)			
S/N	Samples	²²⁶ Ra ²³² Th ⁴⁰ K	R _{eq} in (Bq/Kg)	RLI	H _{ex}
1.	Agate	38.60 52.31 421.30	145.85	1.06	0.39
2.	Amethyst	77.40 71.60 583.40	224.71	1.62	0.61
3.	Aquamarine	27.10 54.41 612.21	154.05	1.01	0.41
4.	Cat's eye	42.50 23.72 349.20	103.33	0.75	0.28
5.	Coral	12.73 41.46 734.38	128.57	0.99	0.35
6.	Topaz	51.6267.43572.36	192.12	1.40	0.52
7.	Quartz	15.61 8.9733.62	31.03	0.22	0.08

Table 3: Comparison of activity concentration and Radium equivalent of the same materials from	n different
etudy	

study						
S/N	Samples	Activity concentration in (Bq/Kg) (Reference: Present study, 2019)		R _{eq} in (Bq/Kg) Reference Present	Activity concentration in (Bq/Kg) (Reference: [13])	R _{eq} in (Bq/Kg) (Reference: [13])
		²²⁶ Ra ²³²	² Th ⁴⁰ K	Study, 2019	226 Ra 232 Th 40 K	
1.	Agate	38.60 52.3	31 421.30	145.85	36.37 49.14 432.60	139.95
2.	Amethyst	77.40 71.	60 583.40	224.71	74.20 78.30 562.43	229.51
3.	Aquamarine	27.10 54.4	41 612.21	154.05	25.40 56.23 624.35	153.88
4.	Cat's eye	42.50 23.	72 349.20	103.33	Sample not used	Sample not used
5.	Coral	12.73 41.4	46 734.38	128.57	10.50 39.41 721.46	122.44
6.	Topaz	51.62 67.4	43 572.36	192.12	Sample not used	Sample not used
7.	Quartz	15.61 8.	97 33.62	31.03	6.23 9.37 29.81	29.81

Discussion IV.

The available radionuclides detected in the samples are ²²⁶Ra, ²³²Th, and ⁴⁰K, as shown in Table 1. This revealed that, all the stone beads contains only naturally occurring radionuclide, as stone beads are materials composed of rocks they also contains natural radioactive isotopes ²²⁶Ra, ²³²Th, and ⁴⁰K [11]. The samples contain similar radionuclide of different proportion.

The activity concentrations were given in table 2; it shows that, the concentration of ²²⁶Ra is higher in Amethyst having 77.40 Bqkg⁻¹ while Coral having the least of 12.73 Bqkg⁻¹. Also, the concentration of 232 Th in the samples is manifested in Amethyst to be the highest with the value of 71.60 Bqkg⁻¹, whereas, Quartz having the least of 8.97 Bqkg⁻¹. In the case of ⁴⁰K, the Coral is having the highest concentration of 734.38 Bqkg⁻¹ while the Quartz having the lowest of 33.62 Bqkg⁻¹. These variations in concentration of the samples may be due to the mineral content of the rocks in which they are made-up [11].

The Radium equivalent activity of the stone beads were given in table 2, the highest value was obtained in amethyst having 224.71 Bqkg⁻¹ which is lower than the maximum permissible limit of 370 Bqkg⁻¹[10].It also found in table 2, that, the representative level index of Amethyst is 1.62 and Topaz 1.40 which are greater than the recommended level. In Agate and Aquamarine, they have 1.06 and 1.02 respectively which are slightly higher than the recommended value of 1. The remaining samples (Coral, Cat's eye and Quartz) have RLI lower than unity.

In the case of external hazard, all the samples were found to have their H_{ex} below the criterion value (less than one) as shown in table 2. In comparison with the existing study, Table 3 has given information in which comparison was made on activity concentration and Radium equivalent activity for the same materials with the exception of Cat's eye and Blue Topaz from different study. The comparison reveals that, there is varying activity concentration in the samples; this may be due to mineral content of the rocks. However, the comparison of R_{eq} 's, shows that, all the samples have their R_{eq} below the recommended maximum limit.

V. Conclusion

The radionuclide content, Activity concentration, Radium equivalent activity (R_{eq}), Representative Level Index (RLI) and the External radiation hazard (H_{ex}) of the selected stones beads commonly used as jewelries in Kano state, Nigeria, were determined. The radionuclides obtained in the samples are²²⁶Ra, ²³²Th and ⁴⁰K. This shows that, there are no induced radionuclide in the samples, the detected radionuclide are only naturally radioactive nuclei which occurs in rock materials. Their concentration in the samples shows that they are not hazardous to human health asthe values of R_{eq} are below the criterion limit of 370Bqkg⁻¹given by the UNSCEAR. All the H_{ex} values are below the recommended limit of unity which makes the radiation hazard insignificant. Also, the RLI values are below the unity except for the Amethyst and Topaz sample which is having 1.62and 1.4 respectively. This study is in agreement with existing one as the samples are found to be radiation hazard free.

Stone beads with higher RLI values should have special management and treatment to reduce the radioactive hazard that will comply with the environmental protection regulations and to maintain the indexes values within the accepted levels. As importation of stone beads is a continual process, it is not wise to make a recommendation on the use of all the available stone beads in the market; rather, a policy makers should devise a means of investigating radiation hazard in all imported stone beads jewelries and ornaments for the safety of its citizens.

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