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EXISTENCE OF RADIONUCLIDES AND THEIR CONCENTRATION IN SOME SELECTED POLISHED GEMSTONES

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ABSTRACT

Gems are frequently subjected to various treatments in order to improve their appearance in terms of color and transparency, 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. In the present study, an investigation on the existence and level of radioactive nuclides on five samples of stone beads has been done using Thallium Activated Sodium iodide detector gamma ray spectroscopy. The Radium Equivalent Activity was also calculated to access the radiation hazard of the samples. The results show that, the radionuclides detected are ^{226}Ra, ^{232}Th and ^{40}K, it was found that the highest and the lowest activity concentration of ^{226}Ra, is 74.20 and 6.23Bqkg^{-1} in Amethyst and Quartz respectively. The highest activity of ^{232}Th was 78.30Bqkg^{-1} in Amethyst and its lowest activity was 9.37 Bqkg^{-1} in Quartz, while the highest activity of ^{40}K was 721.46 Bqkg^{-1} in Coral with the lowest activity of 29.81 Bqkg^{-1} in Quartz. Their Radium Equivalent Activities were found to be 229.51 Bqkg^{-1} for Amethyst, Aquamarine, Agate, Coral and Quartz having 153.88, 139.95, 122.44 and 29.81 Bqkg^{-1} respectively. The Radium Equivalent Activities values are below the recommended value of 370 Bqkg^{-1}, this shows that, the radiations emitted by these samples are not hazardous. Even though, sight should not be loose due to the fact that some of these polished stones may be radioactive. A routine check should persist as to be conscious of radiation hazards from stone beads and ornaments.

Keywords: Gemstones, Radiological Index, specific activity, Stone beads.

1. INTRODUCTION

Radioactivity is a natural phenomenon and natural sources of radiation are features of the environment. Radiation and radioactive substances have many beneficial applications, ranging from power generation to uses in medicine, industry, and agriculture. The radiation risks to workers and the public and to the environment that may arise from these applications have to be assessed and, if necessary, controlled. Activities such as the medical uses of radiation, the operation of nuclear installations, the production, transport, and use of radioactive material, and the management of radioactive waste must, therefore, be subject to standards of safety. Regulating safety is a national responsibility. However, radiation risks may transcend national borders (International Atomic Energy Agency - IAEA, 2014). 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 that is worn around the neck or wrist. The Stone bead is made from gemstone or precious or jewel stone. A gemstone is a piece of crystal which is cut into different smaller sizes, polished and used as jewelry or ornamentals. These crystals are obtained from rocks; they are hard, translucent, luster etc. (Don, 2014). The chemical compositions of gemstones include; Silicates, Oxides, Carbonates, Sulfides, Halides, Phosphate, Sulphate etc. Apart from adornment, gemstones have industrial applications such as glass making, abrasives, foundry sand etc. (King, 2014). 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, of course, a 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 (U.S.N.R.C, 2007). A research was done by Ashbaugh (1992) on some reactor irradiated gemstones and found them to contain radioactive elements in which their cooling time was calculated to be almost 500 years. Another investigation shows that some reactor irradiated gems contain radioactive elements with a half-life of 5 years (Salama et al, 2012). In the present work, the existence and concentration of radionuclides activity in five samples of gemstones materials that are commonly used as pieces of jewelry 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 activity.

2. Materials and method

2.1 List of materials

The materials used in this work include the five samples of the fashioned stone beads: Agate, Amethyst, Aquamarine, Coral and Quartz. The pulverizer, Radon impermeable cylindrical plastic containers having dimension of 7.6cm by 7.6cm, Vaseline Jelly, Candle Wax, Masking tape and sodium Iodide detector gamma ray spectrometer.

2.2 Sample Collection and Preparation

450g for each of the samples were purchased at the shop opposite Grand Central Hotel, in Kano State, Nigeria. Each of the 0.45kg gems samples collected was washed, dried and crushed to fine powder with the use of pulverizer. Packaging of the samples 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 (geometry) was also carried out. To prevent ^{222}Rn 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 (Ibrahim et al., 2013).

2.3 Instrumentation

The analysis was carried out using 7.6cm by 7.6cm Thallium doped Sodium Iodide **NaI (Tl)** detector crystal which 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. The detector crystal is optically coupled (via photocathode) to a photomultiplier tube (PMT). The PMT is externally supplied with one kilovolt (1kV) of electricity; this PMT is incorporated into amplifier which is connected to Analog to digital converter (ADC). The ADC is coupled with a Multi Channel Analyzer (MCA) which is in data acquisition software. A graphic display screen is incorporated.

2.4 Experimental Procedure

When a gamma ray from a radioactive sample enters the NaI crystal, some combination of three physical processes can occur: Photoelectric emission of an electron that absorbs all of the gamma’s energy, Compton scattering of the gamma ray photon off electrons in the crystal, or Pair-production of an electron–positron pair. In order for the last process to occur with any likelihood, the incoming gamma must have an energy that is at least twice the rest mass energy of the electron (2 X 0.511 MeV = 1.022 MeV). Although a couple of the radioactive samples will emit gammas in this range, unless the gamma is substantially more energetic than 1.022 MeV, the pair-production mechanism is not observable. The electron liberated by the photoelectric effect is quite likely to scatter around in the NaI crystal, losing energy, until it is captured by an atom in the crystal with an electron vacancy. In the process of scattering, photons in the visible and ultraviolet (UV) region of the electromagnetic spectrum are emitted. Likewise with the Compton scattering process, the recoil electron will ultimately deliver most of its energy as visible and UV photons. The low frequency (visible and UV) photons produced when a gamma interacts with the scintillator crystal, enter a photomultiplier tube (PMT), in which a cascade of electrons is generated, again via the photoelectric (and secondary electron) effect and then reaching the amplifier. This has the effect of turning a light pulse into a current pulse, which is then converted into a voltage pulse as the current flows through the resistor at the anode. In general, the more energy the original gamma ray had, the larger the voltage pulse that the PMT will produce. The ADC converts the voltage pulse into digits (number), the pulse height analyzer (MCA) divides the range of all possible voltages into bins, or channels, and keeps a running count of how many pulses arrive in each bin, thus producing a graphical spectrum of the number of counts (Knoll, 2010). The detection limit of NaI (TI) detector system for ^{226}Ra, ^{232}Th and ^{40}K are 3.84, 9.08 and 14.54 Bq/kg respectively for a counting time of 29000 seconds, for each sample. The peak area of each energy in the spectrum was used to compute the activity concentrations. The detected nuclides activity levels are reported in Becquerel/kilogram (Bq/kg) using equation (1).

2.5 Radionuclides Concentration

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 (Ashbaugh, 1992);

$$A = \frac{n}{t\gamma\varepsilon ms} \tag{1}$$

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 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 are completely absorbed in to 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).

2.6 Radium Equivalent Activity (R_{eq})

Radium equivalent concentration is the quantity representative of external γ irradiation dose associated with material. In order to compare specific activity of materials containing different amounts of ^{226}Ra, ^{232}Th and ^{40}K, the radium equivalent activity $\mathbf{R_{eq}}$ is used as defined by Nwankwo *et al (*2015).

$$R_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_k \tag{2}$$

Where A_{Ra}, A_{Th} and A_k are the activity concentrations in Bqkg^{-1} of ^{226}Ra, ^{232}Th and ^{40}K, respectively. The value of R_{eq} must be less than 370 Bqkg^{-1} for the radiation hazard to be negligible (UNSCEAR, 2000).

3. RESULTS AND DISCUSSION

3.1 RESULTS

The radionuclides obtained in the samples include ^{226}Ra, ^{232}Th, and ^{40}K as shown in table 1. The concentration of the radionuclides and the common radiological index used to compare the specific activities has been given in Table 2. Figure 1 to 5 is charts representing the concentration of the radionuclides obtained in each of the samples. In figure 6 to 8 are charts showing the comparison of the concentration of a particular radionuclide in all the samples. In figure 9, is a chart, representing the R_{eq} of each sample and R_{eq} given by the international organization on radiation.

Table 1: Types of the Radionuclides detected in the samples

S/N	MATERIAL	RADIONUCLIDES
1	Amethyst	^{226}Ra ^{232}Th ^{40}K
2	Aquamarine	
3	Agate	
4	Coral	
5	Quartz	

Table 2: Activity concentration and associated radiological hazards in different types of selected gemstones

S/N	Materials	Activity concentration in (Bq kg^{-1})			R_{eq} (Bq kg^{-1})
		^{226}Ra	^{232}Th	^{40}K	
1	Amethyst	74.20	78.30	562.43	229.51
2	Aquamarine	25.40	56.23	624.35	153.88
3	Agate	36.37	49.14	432.60	139.95
4	Coral	10.50	39.41	721.46	122.44
5	Quartz	6.23	9.37	29.81	29.81

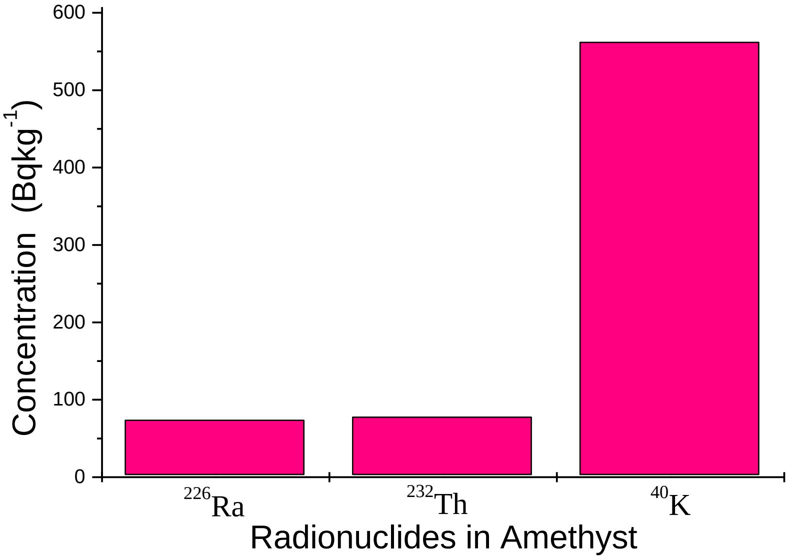

Figure 1: Radionuclides and their concentration in Amethyst

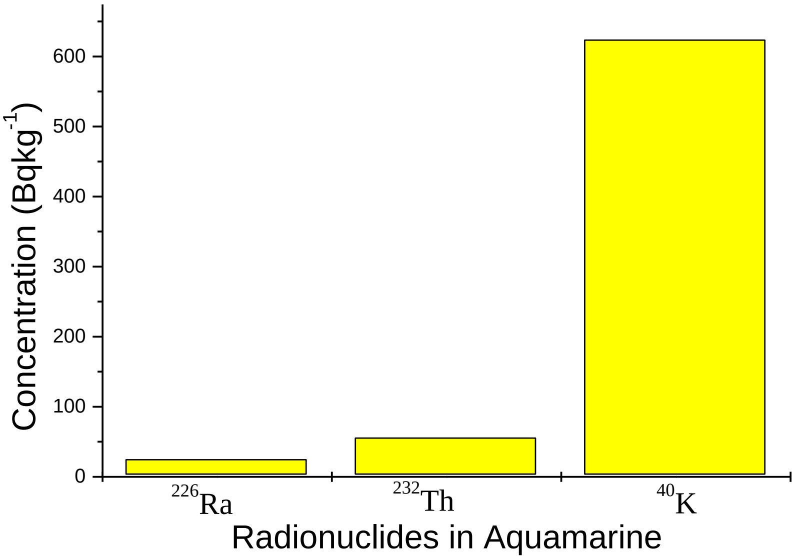

Figure 2: Radionuclides and their concentration in Aquamarine

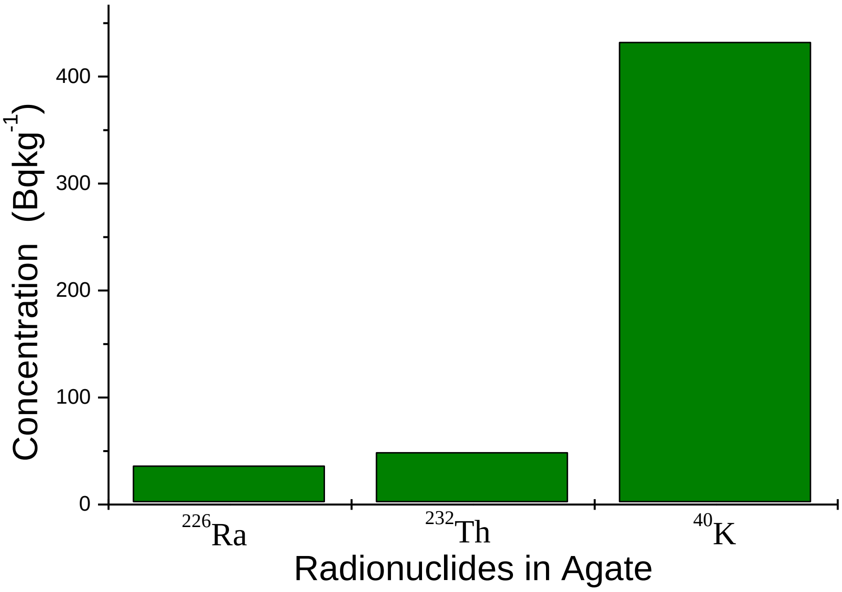

Figure 3: Radionuclides and their concentration in Coral

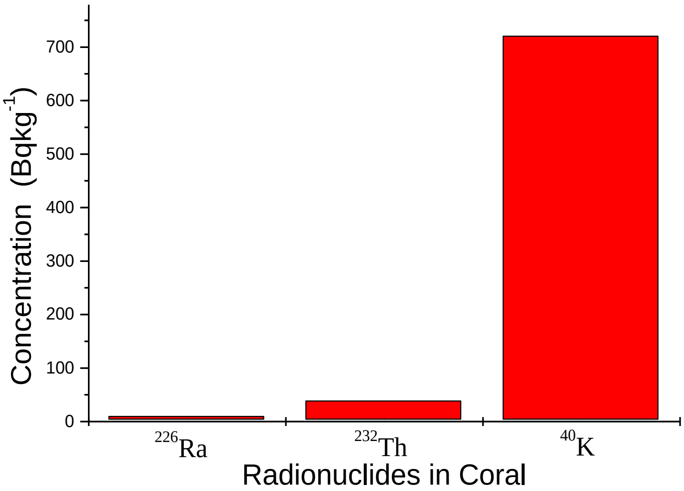

Figure 4: Radionuclides and their concentration in Coral

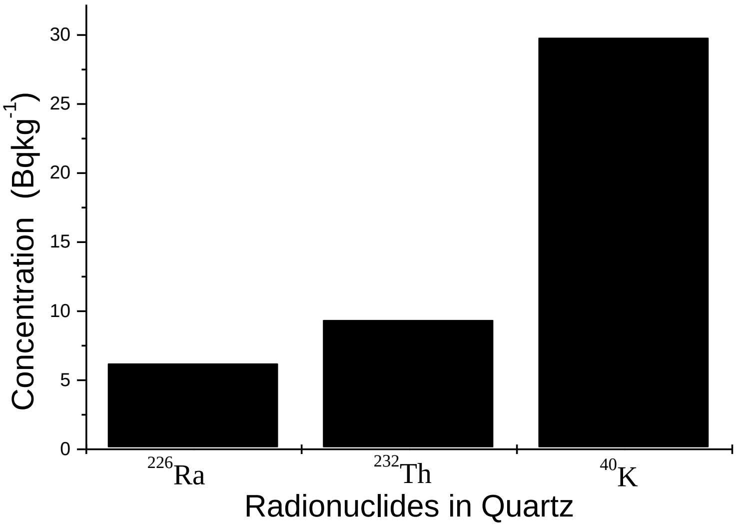

Figure 5: Radionuclides and their concentration in Quartz

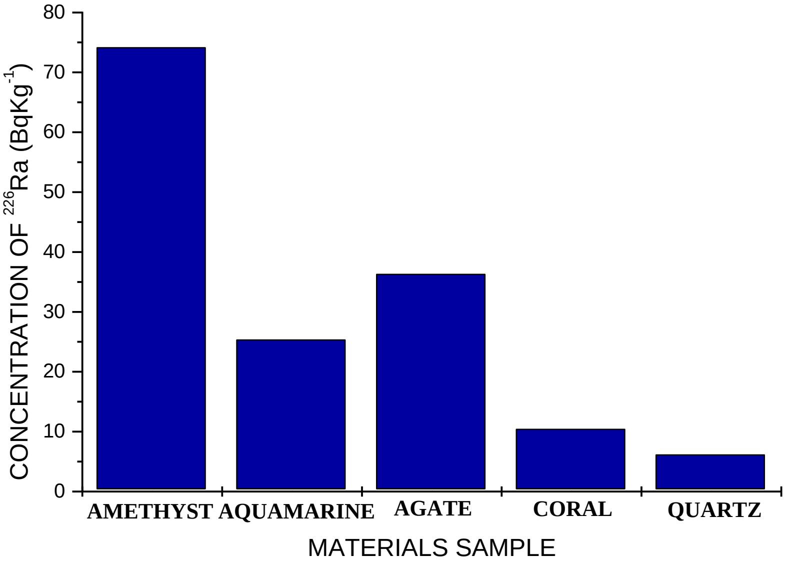

Figure 6: The concentration of Ra-226 radionuclide in different samples

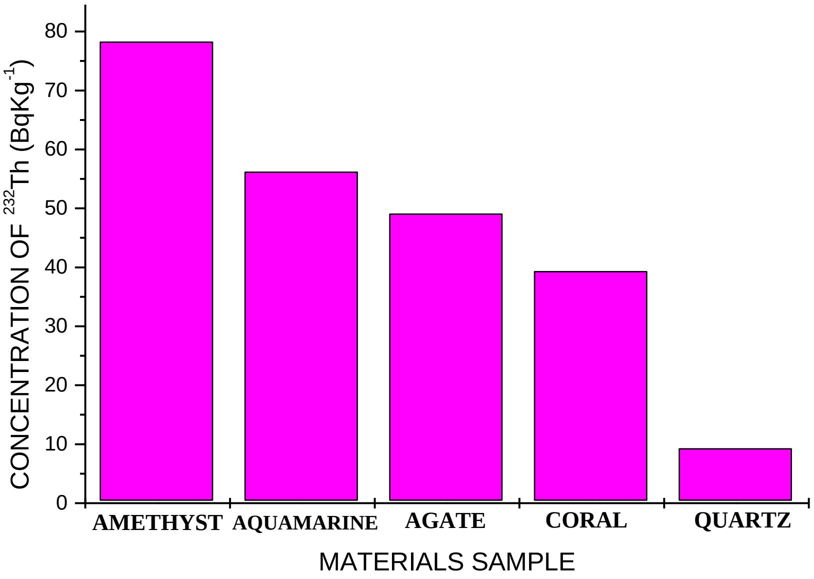

Figure 7: The concentration of the Th-232 radionuclide in different samples

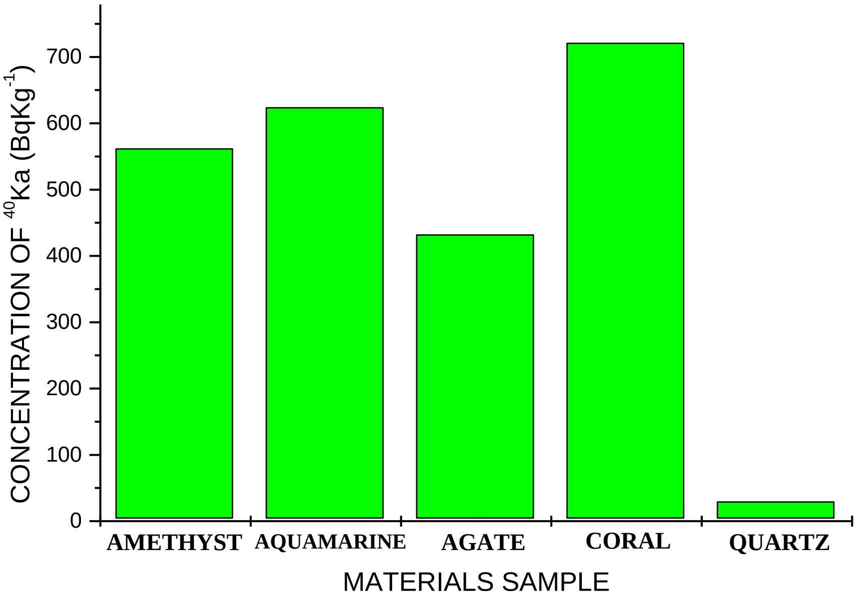

Figure 8: The concentration of Ka-40 radionuclide in different samples

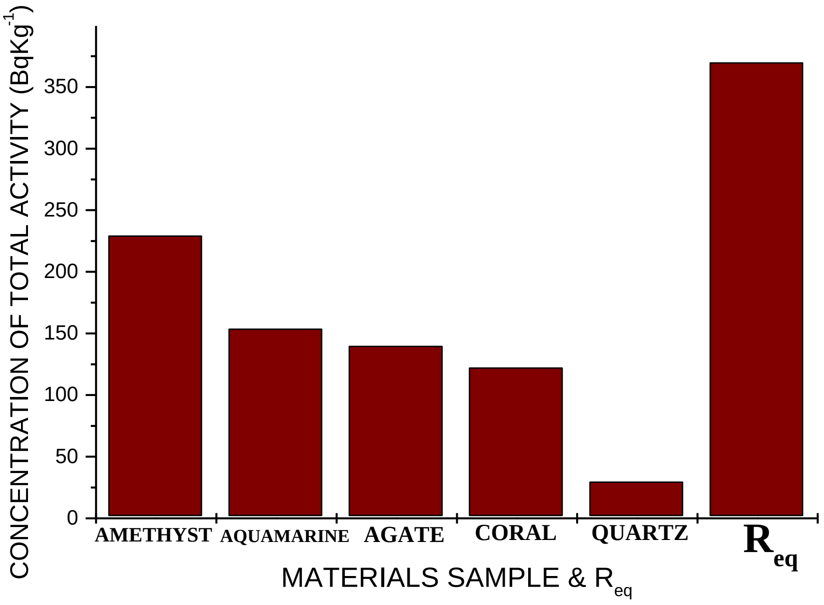

Figure 9: The comparison between the Req of each sample gemstone beads and that recommended by UNSCEAR.

3.2 DISCUSSION

As can be seen from table 1, the types of radionuclides determined in the samples include; ^{226}Ra, ^{232}Th and ^{40}K respectively. This investigation has proven the existence of radionuclides in the samples, the result agrees with the existing knowledge that; "All materials composed of rock and soil contains natural radioactive isotopes; ^{226}Ra, ^{232}Th and ^{40}K (Senthilkumar et al., 2014). These radionuclides are all primordial and are also called naturally occurring radioactive materials (NORM), i.e., they have mineral origin not induced by a nuclear reactor. Each of the samples contains the same types of radionuclides but with different concentration. In table 2, the concentration of ^{226}Ra, ^{232}Th, and ^{40}K were calculated as 74.20, 78.30 and 562.43 Bq kg^{-1} respectively in Amethyst, with the ^{40}K having the highest concentration. In Aquamarine, 25.40, 56.23 and 624.35 Bq kg^{-1} of ^{226}Ra, ^{232}Th and ^{40}K respectively were obtained with also, ^{40}K having the highest concentration. In Agate, the concentration of the radionuclides ^{226}Ra, ^{232}Th, and ^{40}K was also calculated to be 36.37, 49.14 and 432.60 Bq kg^{-1} respectively. In Coral, 10.50, 39.41 and 721.46 Bq kg^{-1} of ^{226}Ra, ^{232}Th and ^{40}K respectively were calculated wherein Quartz, 6.23, 9.37 and 29.81 Bq kg^{-1} of ^{226}Ra, ^{232}Th and ^{40}K were calculated respectively. In the results, it is observed that all the radionuclides have a varying concentration in the samples i.e. the concentration of ^{226}Ra varies in all the samples, likewise, ^{232}Th concentration differs in all the samples and also ^{40}K has a varying concentration in all the samples. Studies show that the variation of the activity concentration in the rock material is due to the mineral content (Senthilkumar et al., 2014). Although the level of radionuclides activity of the selected samples was determined, it is not enough to just know the level of activity concentration unless if it can be compared with the level of specific activity which determines the radiation as hazardous or non-hazardous. This comparison is achieved using R_{eq} given by equation (2). From table 2, column 3, the values of Radium Equivalent Activity R_{eq} for each sample was recorded. It is obvious that Amethyst has the R_{eq} of 229.51Bqkg^{-1}, Aquamarine, Agate, Coral, and Quartz having 153.88, 139.95, 122.44 and 29.81Bqkg^{-1} respectively. Surely, none of the samples posed a threat of hazardous radiation when compared to a given recommended value of 370Bqkg^{-1} by the UNSCEAR, 2000.

4. CONCLUSION

The radionuclide content, Activity concentration and Radium equivalent activity (R_{eq}) of the selected stones beads commonly used as jewelleries in Kano state, Nigeria were determined. The radionuclides obtained in the samples were naturally occurring which consists of ^{226}Ra, ^{232}Th and ^{40}K. Their concentration in the samples shows that, they are not hazardous to human health as calculated using R_{eq}. The values of R_{eq} are below the criterion limit of 370Bqkg^{-1}by the UNSCEAR. All the samples investigated are within the recommended safety limit and hence do not pose any radiation hazard to the users. From the result of the present study, it is observed that the selected samples of the fashioned gemstones do not contain any radionuclides due to reactor irradiation; this verified the fact that the selected samples are not reactor irradiated. Even though, sight should not be loose due to the fact that some of these polished stones may be radioactive. A routine check should persist as to be conscious of radiation hazards from stone beads and ornaments.

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