Using γ-Rays for Characterizing the Radioactivity in Raw Materials and Wastewater

Daniel Marcos Bonotto

***Abstract*—**Gamma rays of variable energy are emitted after the alpha and beta decays from a number of naturally occurring and man-made radioactive sources, constituting important records of the presence of radioactivity in different matrices as reported in this study. Raw materials consisting on tantalite ore and several rock types potentially useful for use as dimension stones have been analyzed by γ-rays spectrometry, as well a wastewater sample providing from a Brazilian hospital dedicated to cancer treatment. In the solid samples, it has been identified a major contribution from ^{40}K and radionuclides generated through the decay sequence of the three alpha emitting radionuclides (^{232}Th, ^{238}U, and ^{235}U), whereas artificial ^{131}I was characterized in the sample of liquid effluent.

***Keywords*—**Gamma rays, radioactivity, natural and man-made sources, raw materials and wastewater

I. Introduction

Environmental radiation originates from a number of naturally occurring and man-made sources. Radioactive materials in the environment have several sources, among them: primordial radioisotopes of uranium-thorium series, potassium, etc. in the earth´s crust; continuous production by cosmic radiation; production by nuclear explosions or from the nuclear fuel cycle; release by nuclear installations or medical/mining activities.

These radionuclides become a part of different components of nature. By their radioactivity they label particular components on the local, regional, or global scale making it reliable to study physical, chemical, and biological processes in the atmo-, geo-, hydro- and biospheres.

Most of the cosmic rays originate from deep in interstellar space whilst some are released from the sun during solar flares. Naturally, the levels of terrestrial radiation differ from place to place around the world, as the concentrations of these materials in the earth´s crust vary. The major contribution to the terrestrial gamma radiation field comes from ^{40}K and from radionuclides generated through the sequence of decay transformations of three alpha emitting primeval radionuclides, i.e. ^{232}Th, ^{238}U, and ^{235}U.

^{40}K decays directly to ^{40}Ca in the ground state through β^- emission (89.3%) and also to ^{40}Ar in a 1.46 MeV excited state followed by a prompt 1.46 MeV gamma emission through EC-electron capture (10.7 %) [1].

^{232}Th is precursor of the natural mass number 4n decay series that finishes at stable ^{208}Pb, according to [1], [2]: ^{232}Th (14.0 Gy, α) → ^{228}Ra (5.8 y, β^-) → ^{228}Ac (6.2 h, β^-) → ^{228}Th (1.9 y, α) → ^{224}Ra (3.7 d, α) → ^{220}Rn (55.6 s, α) → ^{216}Po (0.14 s, α) → ^{212}Pb (10.6 h, β^-) → ^{212}Bi (60.6 min, β^--64.1% or α-35.9%) → ^{212}Po (0.3 μs, α) or ^{208}Tl (3.0 min, β^-)→ ^{208}Pb.

^{238}U is precursor of the natural mass number 4n+2 decay series that finishes at stable ^{206}Pb, according to [1], [2]: ^{238}U (4.47 Gy, α) → ^{234}Th (24.1 d, β^-) → ^{234m}Pa (1.17 min, β^-) → ^{234}U (0.246 My, α) → ^{230}Th (75.4 ky, α) → ^{226}Ra (1.6 ky, α) → ^{222}Rn (3.82 d, α) → ^{218}Po (3.10 min, α) → ^{214}Pb (26.8 min, β^-) → ^{214}Bi (19.9 min, β^-) → ^{214}Po (0.16 ms, α) → ^{210}Pb (22.3 y, β^-) → ^{210}Bi (5.0 d, β^-) → ^{210}Po (138.4 d, α) → ^{206}Pb.

^{235}U is precursor of the natural mass number 4n+3 decay series that finishes at stable ^{207}Pb, according to [1], [2]: ^{235}U (0.70 Gy, α) → ^{231}Th (25.5 h, β^-) → ^{231}Pa (32.8 ky, α) → ^{227}Ac (21.8 y, β^-- 98.6% or α-1.4%) → ^{227}Th (18.7 d, α) or ^{223}Fr (21.8 min, β^-) → ^{223}Ra (11.4 d, α) → ^{219}Rn (4.0 s, α) → ^{215}Po (1.8 ms, α) → ^{211}Pb (36.1 min, β^-) → ^{211}Bi (2.14 min, α) → ^{207}Tl (4.8 min, β^-) → ^{207}Pb.

Gamma radiation is simply a high energy form of electromagnetic radiation. Gamma rays have their origin in a nuclear decay process rather than an atomic electron decay process (X-rays) or a thermal electron decay process (light rays). Gamma rays can be detected by non-destructive methods that have a lot of advantages from the technical point of view over alpha and beta spectrometric techniques, inclusive allowing the identification and quantification of alpha and beta-emitters radionuclides.

The NaI(Tl) scintillation and high-purity germanium (HPGe) detectors have been extensively used for characterizing the natural gamma radiation, which interacts with the crystal atoms through three major processes: photoelectric effect, Compton effect, and pair production.

This study reports the use of gamma ray spectrometry for characterizing naturally occurring and man-made radionuclides in three different matrices of great environmental concern: raw materials for dimension stones use, raw material for components utilized in the electronic industry, and hospital wastewater.

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II. Gamma Rays Detection

A. The Spectrometric Systems

Three types of detectors have been utilized in this study. The first is a planar and well-type 7.5 cm length × 7.5 cm diameter NaI(Tl) scintillation detector, whereas the second is a 8.4 cm length × 6.7 cm diameter HPGe coaxial detector. Fig. 1 illustrates a typical γ-spectrometric system similar to those used in this study. The major differences in the case of the system employing the HPGe detector are: the operating bias is negative 4500 V rather than positive 1000 V; a X-Cooler III unit specifically designed is used to provide the cooling required by the detector (77K at STP); an ASPEC-927 ORTEC dual 16k multichannel buffer (MCB) is used instead of the ACE 2k ORTEC MCB; the resolution (FWHM) at 1.33 MeV, ^{60}Co, is 2.1 keV against 123 keV of the NaI(Tl) detector; the efficiency at 1.33 MeV, ^{60}Co, is 63% contrarily to 1.3% of the scintillation detector. Despite the superior technical characteristics of the system using the HPGe detector, the low cost and reliable responses of the NaI(Tl) gamma spectrometers to the designed essays have justified their use in the radioactivity characterization of the materials.

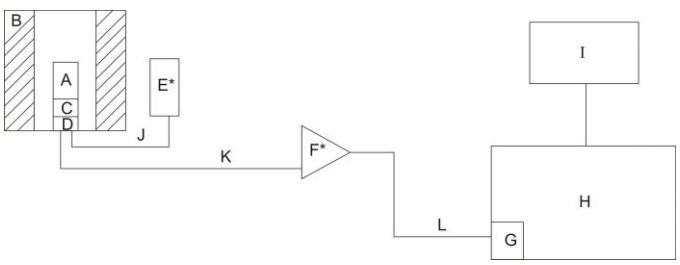

Fig. 1 A simplified block diagram of a typical spectrometric system for detecting gamma rays based on a NaI(Tl) scintillation detector. A=NaI(Tl) detector; B=Lead shielding; C=Photomultiplier; D=Pre-Amplifier; E=HV Power Supply; F=Amplifier; G=ACE 2k MCB; H=Microcomputer; I=Printer; J,K,L=Cable; *NIM BIN powering

B. Systems Calibration in Energy

The spectrometric systems for performing gamma readings have been calibrated in energy by the use of the following radioactive sources: ^{133}Ba solution (γ-rays energy = 0.36 MeV), ^{137}Cs (γ-rays energy = 0.66 MeV), ^{60}Co (γ-rays energy = 1.17 and 1.33 MeV), and pure powdered KCl (52 wt% in K) as a source of ^{40}K (γ-rays energy = 1.46 MeV). Figs. 2 to 5 illustrate the gamma spectra obtained for these radionuclides in the spectrometric system utilizing the planar NaI(Tl) scintillation detector and a 2,048-channels MCB provided by ORTEC ACE 2k hardware controlled by MAESTRO software. Table I reports the photopeak channel identified in the gamma spectra recorded in the MCB and the corresponding energy. These parameters allowed generate the energy calibration curve of the gamma spectrometer (Fig. 6) that is expressed by:

$$E = 0.0016 - 0.0255\ Ch \quad (1)$$

where: E is the energy (in MeV) and Ch is the channel number in the MCB.

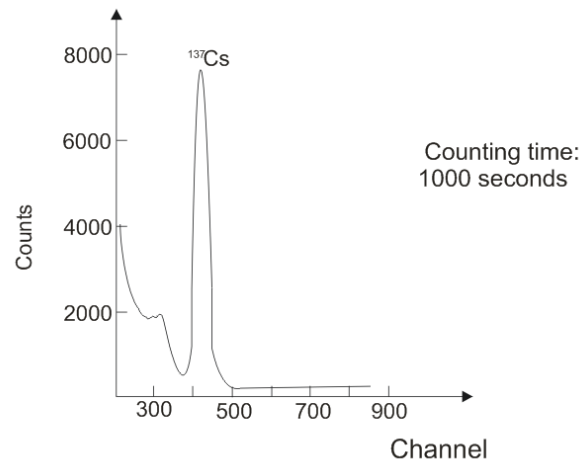

Fig. 2 ^{137}Cs spectrum in the planar NaI(Tl) gamma spectrometer.

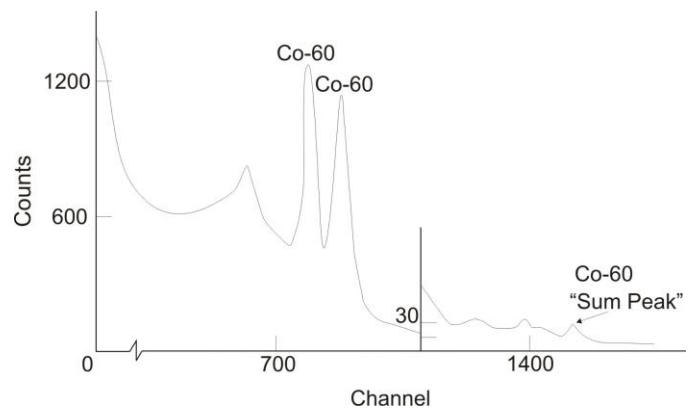

Fig. 3 ^{60}Co spectrum in the planar NaI(Tl) gamma spectrometer. Counting time = 1000 s

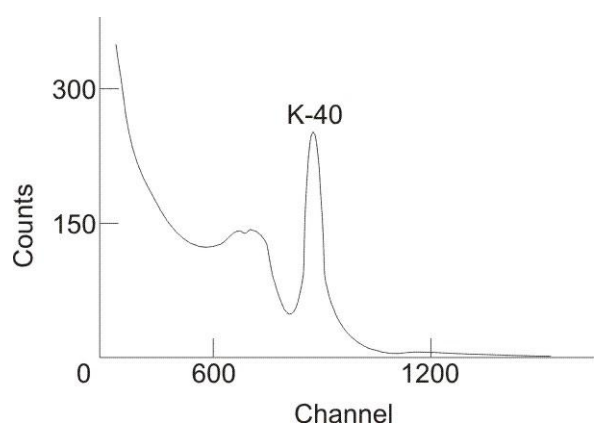

Fig. 4 ^{40}K spectrum in the planar NaI(Tl) gamma spectrometer. Counting Time = 1000 s.

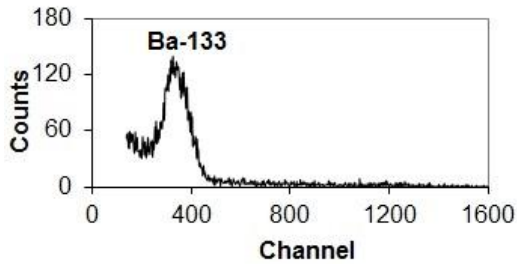

Fig. 5 ^{133}Ba spectrum in the planar NaI(Tl) gamma spectrometer. Counting Time = 1000 s

TABLE I
MCB READINGS IN THE PLANAR NAI(TL) GAMMA SPECTROMETER

Radionuclide	Photopeak Channel	Energy (MeV)
^{133}Ba	295	0.356
^{137}Cs	416	0.661
^{60}Co	736	1.173
^{60}Co	832	1.332
^{60}Co (sum peak)	1568	2.405

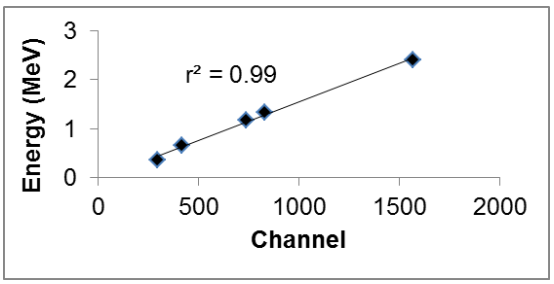

Fig. 6 Calibration curve in energy of the planar NaI(Tl) gamma spectrometer

C. Systems Calibration in Concentration

The gamma spectrometers have been also calibrated in concentration for performing adequate measurements of the radionuclides present in the matrices analyzed. Pitchblende and monazite sand standards having different uranium and thorium concentrations (NBL101A, NBL102A, NBL103A, NBL104A, NBL105A, NBL106A, NBL107A, NBL108A, NBL109A, and NBL110A) and providing from New Brunswick Laboratory, U.S. Department of Energy, Argonne, Illinois, USA, were submitted to gamma readings after waiting ^{222}Rn to reach secular radioactive equilibrium with ^{226}Ra (at least 25 days). Two well-homogenized samples of stream sediments (SS1 and SS2) were also used in this calibration step in order to provide lower Th concentrations. Pure KCl (52 wt% in K, standard S1) and different mixtures prepared from this matrix and additions of pure SiO_2 were utilized to obtain variable potassium concentration. The following standards were prepared: S2 (54.16 g SiO_2 + 28.9 g KCl), S3 (80.12 g SiO_2 + 5.8 g KCl), S4 (82.11 g SiO_2 + 2.9 g KCl), and S5 (84.72 g SiO_2 + 0.5 g KCl).

Fig. 7 shows a spectrum of a pitchblende standard obtained in the planar NaI(Tl) gamma spectrometer, where several ^{214}Bi photopeaks have been identified, as this radionuclide is a ^{238}U-descendant with many gamma ray emissions. Fig. 8 illustrates a gamma spectrum of a monazite sand standard obtained in the same system, in which several ^{208}Tl photopeaks have been identified since this radionuclide is a ^{232}Th-descendant with several gamma ray emissions too.

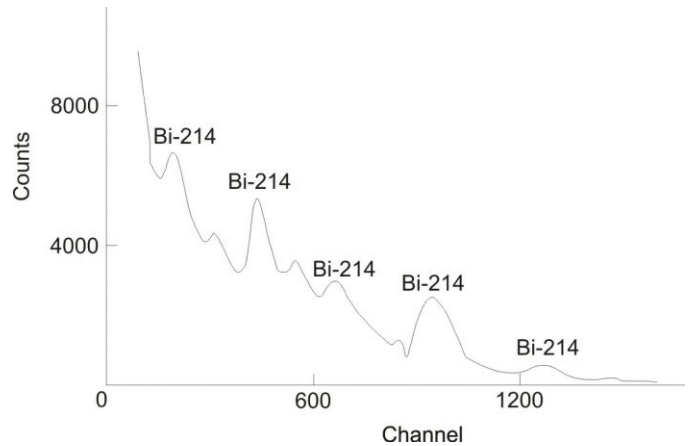

Fig. 7 Spectrum of a pitchblende standard in the planar NaI(Tl) gamma spectrometer. Counting Time = 1140 s

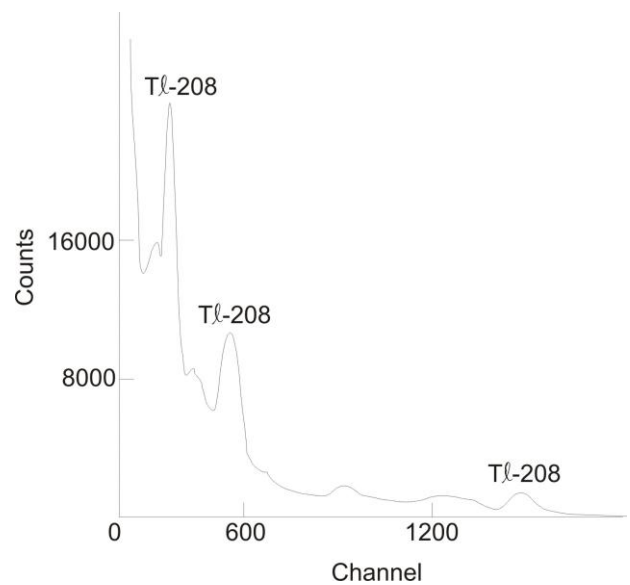

Fig. 8 Spectrum of a monazite sand standard in the planar NaI(Tl) gamma spectrometer. Counting Time = 30000 s

From the standards readings in the planar NaI(Tl) gamma spectrometer, it was possible to plot calibration curves for the concentration of natural U, Th, and K, as shown in Figs. 9, 10 and 11, respectively, which may be expressed by:

$$logC_U = 1.057 \times log\ I_U + 2.578 \quad (2)$$

$$logC_{Th} = 1.075 \times log\ I_{Th} + 3.273 \quad (3)$$

$$logK = 0.953 \times log\ I_K + 1.459 \quad (4)$$

where: C_U (in ppm or μg/g), C_{Th} (in ppm or μg/g) and C_K (in %) is the equivalent U, Th and K concentration, respectively, and its corresponding effective intensity (I_U, I_{Th} and I_K, in cpm/g).

Different matrices have been chosen for applying the

methods described. Raw materials consisting on several rock types potentially useful for use as dimension stones have been analyzed, as well tantalite for utilization in the electronic industry due to its resistance to heat, among other favorable aspects. Additionally, wastewater providing from a hospital has been also subjected to the radionuclides analysis.

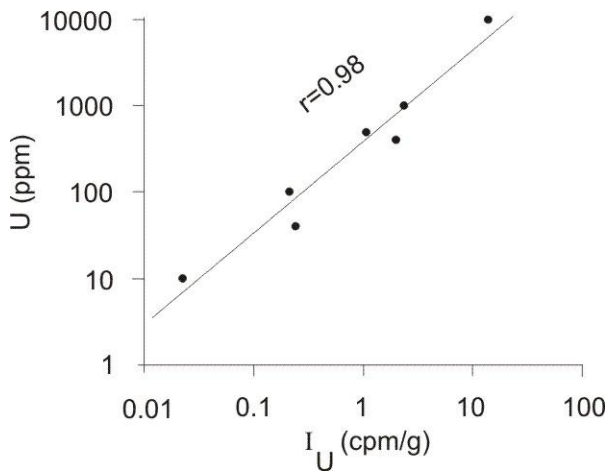

Fig. 9 Calibration curve of the U concentration in the planar NaI(Tl) gamma spectrometer

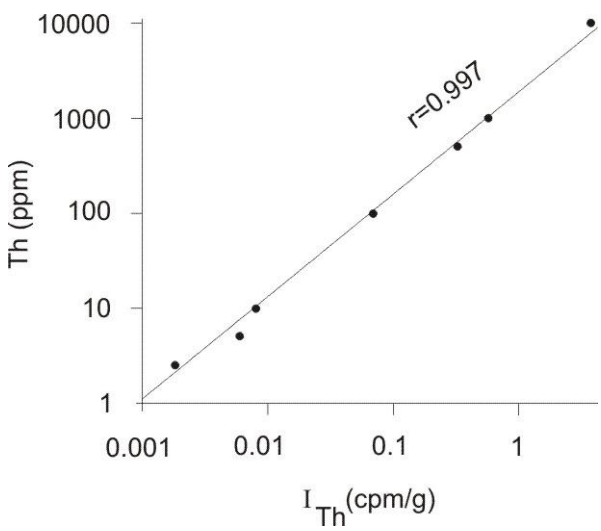

Fig. 10 Calibration curve of the Th concentration in the planar NaI(Tl) gamma spectrometer

III. Natural Radioelements In Raw Materials For Dimension Stones

Because natural radioelements are widely spread in the environment, they also occur in materials used to build human inhabitation. Much attention has been given in the last years to decorative stones used especially as flooring and countertops inside homes since the rocky materials may sometimes possess high levels of radioactivity. This is particularly true if granitic rocks are used for such purpose as natural U and Th are lithophile elements distributed in crustal rocks that concentrate preferentially in acid igneous rocks compared with intermediate, basic, and ultrabasic varieties.

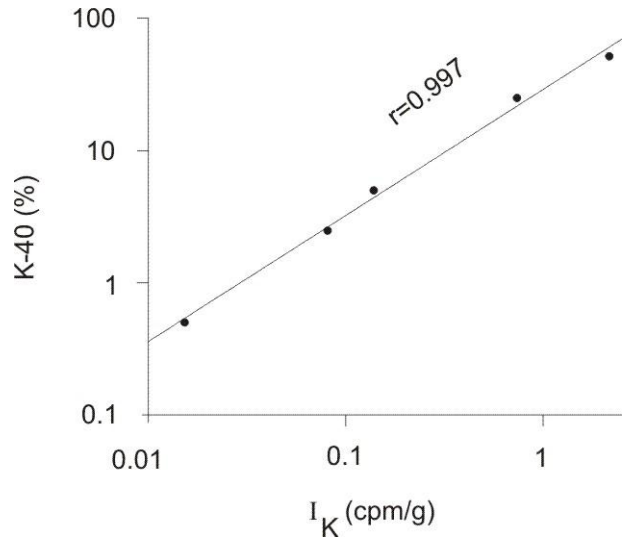

Fig. 11 Calibration curve of the K concentration in the planar NaI(Tl) gamma spectrometer

Uranium occurs in crustal rocks at an average concentration of 2.5 µg/g [3]. Other rock types exhibit the following average U concentration (in µg/g) [4]: sandstone = 1.4; grayish schist = 4.2; carbonaceous schist = 53; limestone = 1.9; riolite = 5.0; granite = 3.6; phonolite and syenite = 6.5; alkali basalt = 0.99; gabbro = 0.84; andesite =0.79; peridotite = 0.01. Thus, U may reach a maximum enrichment of 500 times in granite and 650 times in syenite relatively to rocks representing the mantle composition like amphibolite, granulite, eclogite and dunite [4].

In crystalline rocks, the most of the U is incorporated into accessory minerals such as monazite, allanite, sphene, and zircon so that U is not readily accessible for solution and available to secondary mineralization processes. The typical U concentration in some minerals is (in µg/g) [5]-[8]: quartz = 1.7; feldspars = 2.7; biotite = 8.1; muscovite = 2.8; hornblende = 0.2–60; pyroxene = 0.1–50; olivine = 0.05; allanite = 30–1000; apatite = 10–100; epidote = 20–200; garnet = 6–30; huttonite = 3–70000; magnetite = 1–30; monazite = 500–3000; titanite = 10–700; xenotime = 300–40000; zircon = 100–6000.

The presence of U, Th and K in construction materials offers radiation exposure both in outside environments and inner buildings due to gamma radiation of ^{40}K and members of the U and Th decay series. Additionally, radon (^{222}Rn, half-life 3.84 d) has been of a general health concern as can be a health risk for indoor users of building materials containing U [9], [10].

Igneous rocks have been extensively used in Brazil as building materials, with granites representing the majority of them. The exportation of granites as decorative stones constitutes an important economic activity in Brazil. In general, granites are widely recognized to exhibit high levels of U and Th due to the characteristics of the genetic magma and associated tectonic environment. Rocks generated in the

crust are often more enriched in radioelements relatively to those formed in the mantle, as a consequence of magma partial fusion and fractioned crystallization processes that concentrated them in the liquid phase enhanced in silica.

Rock samples of different geological units occurring in the states of São Paulo, Minas Gerais and Rondônia, Brazil, have been collected in outcrops and quarries and subjected to γ-ray analysis by the planar NaI(Tl) scintillation detector. According to the genetic classification, they have been recognized as toleiitic diabases, deep sienites, shallow sienites, among other. This lithologic diversity influenced the results of the natural radioactivity.

The mode values in the granitic rocks of Rondônia State (n = 55) were: K = 11%; U = 29 µg/g; Th = 85 µg/g. Significant Pearson correlation coefficient was found between K and U (r = 0.71), K and Th (r = 0.72), and U and Th (r = 0.72), indicating congruency of their accumulation processes in the minerals of the rocks analyzed.

The data in µg/g (ppm) and % in the granitic rocks of São Paulo and Minas Gerais states (n = 14) were converted to activity concentration (in Bq/kg) on using the following factors [11]: 1% K = 317 Bq/kg; 1 ppm U = 13 Bq/kg; 1 ppm Th = 4.08 Bq/kg. Thus, the following activity concentration range was found: U = 12.2-251.9 Bq/kg; Th = 9.6-347.5 Bq/kg; K = 407.5-1615.0 Bq/kg.

Another useful parameter is the absorbed radiation dose rate (*DR*, in nGy/h per Bq/kg) in air above 1 m of the terrain surface. It can be expressed by [12]:

$$DR = 0.0414\, A_K + 0.461\, A_U + 0.623\, A_{Th} \quad (5)$$

where: A_K, A_U and A_{Th} (in Bq/kg) is the K, U, and Th specific activity, respectively; 0.0414, 0.461 and 0.623 is the conversion factor of the gamma dose rate (in nGy/h per Bq/kg) for K, U, and Th, respectively.

The *DR* data may be converted to effective dose using the fator 0.7 Sv/Gy [12]. It ranged from 0.45 to 7.19 mSv/yr in the granitic rocks of Rondônia State, whilst the mode value of 2.7 mSv/yr is higher than the majority of the data [11]. It is also slightly higher than the global average value of 2.4 mSv/yr [13].

In fact, the gamma readings for U is essentially based on the measurements of the ^{214}Bi photopeaks (Fig. 7). For this reason, they have been named equivalent uranium, eU (^{214}Bi = ^{226}Ra), providing information on the ^{222}Rn supported by its parent ^{226}Ra in the rocks [14]. Thus, the ^{226}Ra, ^{232}Th and ^{40}K activity concentration have allowed estimate three different indices: the radium equivalent activity, Ra_{eq} [15]; the index of external radiation hazard, H_{ex} [16]; and the gamma activity concentration index, I_γ [11]. They are expressed by:

$$Ra_{eq} = C_{Ra} + 1.43\, C_{Th} + 0.077\, C_K \quad (6)$$

$$H_{ex} = (C_{Ra}/370) + (C_{Th}/259) + (C_K/4810) \quad (7)$$

$$I_\gamma = (C_{Ra}/300) + (C_{Th}/200) + (C_K/3000) \quad (8)$$

where: C_{Ra}, C_{Th} and C_K is the specific activity concentration (in Bq/kg) of ^{226}Ra, ^{232}Th and ^{40}K, respectively. Ra_{eq} is expressed in Bq/kg, whereas H_{ex} and I_γ are dimensionless indices.

The following range of values was found in the granitic rocks of São Paulo and Minas Gerais states: Ra_{eq} = 57.21-752.81 Bq/kg; H_{ex} = 0.15-2.03; and I_γ = 0.23-2.67. Significant Pearson correlation coefficient was found between Ra_{eq} and H_{ex} (r = 1.0), Ra_{eq} and I_γ (r = 0.98), and H_{ex} and I_γ (r = 0.98), as expected due to the common specific activity concentrations utilized on their evaluation. The I_γ range was higher in the granitic rocks of Rondônia State, varying between 0.57 and 8.99.

The values estimated of Ra_{eq}, H_{ex} and I_γ in the granitic rocks of São Paulo and Minas Gerais states are consistent with the magmatic origin of the rocks analyzed. The highest values of Ra_{eq} (752.8 and 597.7 Bq.kg^{-1}) and H_{ex} = (2.03 and 1.61) were found in two rocks that suffered superimposed geological processes (shear zones), despite their common origin, which caused enrichment or depletion in radioelements, depending if they had been brittle or ductile. Beyond the importance of the crystallization history, hydrothermal alteration processes may also affect the radionuclides distribution as often reported in other igneous rocks [17]. The highest I_γ value (2.67) corresponded to a rock that suffered a process of ductile-brittle deformation that caused it a microbrecciated shape and an enhancement in radioelements. The processes of shearing or brittle deformation possibly have created paths through with fluids enriched in radioelements moved and were subsequently deposited in these host rocks.

Guidelines on the radiological protection principles concerning the natural radioactivity of building materials have been proposed [18]. Doses to members of the public should be kept as low as reasonably achievable. Within the EU, doses exceeding 1 mSv/y should be taken into account from the radiation protection point of view. The index I_γ has been correlated to the annual dose due to external gamma radiation generated by building materials. If the materials are used in bulk amount (concrete, etc.) then, I_γ should be lower than 1, but if they have more restrict uses like dimension stones, covering layers (surfacing), tiles, boards, etc., then, $I_\gamma \leq 6$ [18]. Therefore, the I_γ index establishes a dose criterion that can be preliminarily utilized as a useful tool for identifying materials appropriate for use as surfacing in civil construction, where materials with $I_\gamma > 6$ should be avoided for such purpose as could generate dose rates higher than 1 mSv/y [11]-[13], [18].

The highest $I\gamma$ value found in the granitic rocks of São Paulo and Minas Gerais states corresponded to 2.67 that is much below the threshold limit value of 6 for superficial and other materials with restricted uses [18]. However, in the case of the granitic rocks of Rondônia State, six specimens exhibited $I\gamma$ values above 6, suggesting they are not suitable as building materials of more restrict uses like dimension stones, covering layers (surfacing), tiles, boards, etc.

IV. Natural Radioelements In Raw Material For The Electronic Industry

The mineral group tantalite [(Fe,Mn)(Ta,Nb)$_2$O$_6$] is the primary source of the chemical element tantalum. Iron-rich

tantalite is the mineral tantalite-(Fe) or *ferrotantalite* and manganese-rich is tantalite-(Mn) or *manganotantalite*. Tantalite is black to brown in both color and streak. Manganese-rich tantalites can be brown and translucent. Tantalite has been found in Australia, Brazil, Canada, Colombia, Egypt, Madagascar, Namibia, Nigeria, Rwanda, Zimbabwe, the United States, and northern Europe.

Tantalite is a well-valued ore extensively used in the electronic industry. Beyond its application in cell phones, the solid state Ta capacitors are utilized in computer circuits, video, and cameras, including as well the automotive electronic and equipment for military and medical uses. Additional applications for Ta include the tantalum carbide in cutting devices, superalloys in the aeronautics industry for manufacturing special turbines, laminate products and wires resistant to corrosion and high temperatures. Some lower efficiency substitutes for tantalum are: Nb (in superalloys and carbides); Al and ceramics (in capacitors).

Brazil has the world's largest reserve of tantalite (52.1%) and this material has been exported mainly to USA and Mexico. Because natural radioelements U, Th and K may be incorporated in the crystalline structure of tantalite, radioactivity essays must be realized in the raw material for exportation. This was the case for one sample exhibiting the following chemical composition: NbO = 19.97%; Ta_2O_5 = 44.91%; Fe_2O_3 = 13.47%; MgO = 6.29%; P_2O_5 = 4.86%; Al_2O_3 = 4.55%; MnO = 4.31%; ZrO_2 = 1.11%; ZnO = 0.50%. The amount of the raw material to be exported by the company corresponded to two lots of 315 kg each that would be subjected to the radioactivity control for checking if they met the government packing requirements for the safe transport of radioactive material.

In Brazil, the nuclear issues and subjects are entirely managed by the Federal administration, meaning that only the Federal Government (represented by the Republic President) can make decisions, ranging from mining affairs until the control of the nuclear activities. The National Nuclear Energy Commission (CNEN – *Comissão Nacional de Energia Nuclear*), established in 1962, is the agency in the country responsible for the nuclear energy peaceful use, rulings on uranium production and nuclear issues, the supervision and radiological protection, the waste management and nuclear safety. The transport of radioactive materials in Brazil is regulated by Rule CNEN-NE-5.01 established by Resolution CNEN 13/88 published in 1st August 1988, which followed the Safety Series No. 6 (Regulations for the Safe Transport of Radioactive Materials) published in 1985 by the IAEA (International Atomic Energy Agency), Vienna. According to these guidelines, the "Radioactive Materials" (Class 7) are those exhibiting specific activity higher than 70 kBq/kg, and, under such circumstance, they would need very special packing requirements for their safe transport.

One crushed sample (200 mesh, 98 g) of the raw material has been submitted to the radionuclides analysis during 9.1 h by the use of the γ-rays spectrometer employing the HPGe coaxial detector. Similarly to (2)-(4), the calibration curves for the U, Th, and K concentration were:

$$\log C_U = 1.065 \times \log I_U + 4.447 \quad (9)$$

$$\log C_{Th} = 1.078 \times \log I_{Th} + 4.766 \quad (10)$$

$$\log K = 1.082 \times \log I_K + 3.405 \quad (11)$$

Equations (9)-(11) allowed to find the following values: C_U = 509.0 μg/g; C_{Th} = 495.7 μg/g; C_K = 4.8%. When converted to activity concentration, these data yielded 6.6 kBq/kg for U, 2.0 kBq/kg for Th and 1.5 kBq/kg for K. Therefore, the total activity concentration corresponded to 10.1 kBq/kg, thus, implying that the exported tantalite is not Class 7 i.e., "Radioactive Material".

V. Dissolved Radionuclides In Hospital Wastewater

In nuclear medicine, radioisotopes are used both for diagnostic and therapeutic purposes. Currently, about 20 radioisotopes are produced for use in nuclear medicine, such as ^{131}I (8 d, β^-), ^{99m}Tc (6.01 h, γ), ^{51}Cr (27.7 d, EC), ^{68}Ga (67.8 min, β^+ or EC), ^{58}Co (70.96 d, β^+ or EC), ^{137}Cs (30.07 y, β^-), and ^{133}Ba (10.7 y, EC), among other [19]. Several hospitals and other medical facilities daily use some of these radioisotopes delivered by main radiopharmaceutical suppliers. From this use, hospital solid wastes and liquid effluents containing radioactivity have been produced. Radio protection measures have been implemented in the medical facilities according to international standards, to prevent or reduce the irradiation and contamination of the staff and facilities, including procedures for solid waste segregation and safe disposal, and procedures for liquid waste management like special bathrooms for patients under treatment with radiopharmaceuticals [19].

The radioactive liquid effluents produced at the hospital facilities mainly dedicated to cancer treatment (e.g., from patient bathrooms and laboratory sinks) can contain relatively high levels of radioactivity depending on the type of disease treated in the facilities, amount of radioisotopes applied, and number of patients treated. The discharge of radioactive liquid effluents from hospital and medical facilities to the environment has been a matter of some concern and investigation in several large European cities [19].

One wastewater sample (1 L) from a hospital dedicated to cancer treatment and situated at Barretos city, São Paulo State, Brazil, was evaporated up to 12 mL, inserted in an appropriate glass vial and submitted to the radionuclides analysis during 7.8 h by the use of the γ-rays spectrometer employing the well-type NaI(Tl) scintillation detector. Fig. 11 shows the spectrum obtained, where two photopeaks (^{133}Ba and ^{137}Cs) had been initially identified from an energy calibration curve similar to that in Fig. 6.

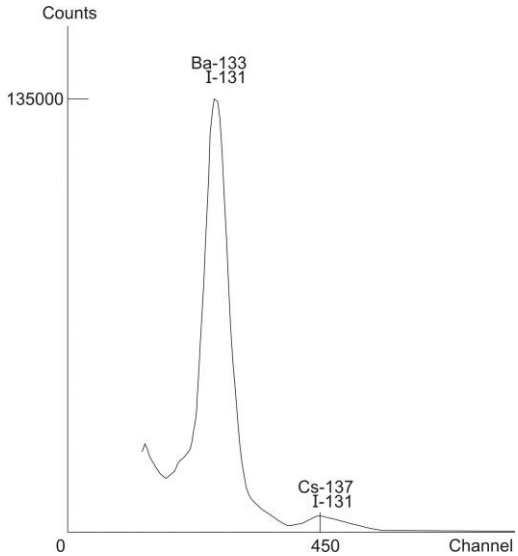

Fig. 11 Spectrum of the hospital wastewater sample in the well-type NaI(Tl) gamma spectrometer

However, careful evaluation of the health activities developed in the hospital indicated that ^{131}I was in fact the radionuclide present in the liquid effluent. The thyroid gland is one of the body's regulators, controlling and regulating the metabolism, but sometimes the thyroid is overactive (or hyperthyroidism) and sometimes it is affected by cancer. In both cases treatment with radioactive iodine (^{131}I therapy) may be required. In the case of an overactive thyroid, the radioactive iodine dose destroys part of the thyroid gland so that the remaining part of the thyroid functions at a normal level. In the case of cancer, following removal of the thyroid, a large dose of radioactive iodine may be prescribed to completely ablate (destroy) any remaining thyroid tissue in the thyroid area. It will also destroy any cancerous thyroid tissue that may have moved elsewhere in the body.

^{131}I is produced by the fission of uranium atoms during operation of nuclear reactors and by Pu (or U) in the detonation of nuclear weapons. It suffers β^--decay to ^{131}Xe, a process that is accompanied by the emission of γ-rays possessing the following energies [19]: 284 keV (6.12%), 364 keV (81.5%), and 637 keV (7.16%). Thus, the γ-rays energy of the photopeaks in Fig. 11 attributed to ^{133}Ba (356 keV) and ^{137}Cs (661 keV) are very similar to those of ^{131}I, explaining the initial wrong identification. In general, the radionuclides ^{137}Cs and ^{133}Ba are used in hospitals as standards or sealed sources for irradiation, unlike ^{131}I that is supplied to patients in capsules to swallow with water. When ^{131}I is ingested, some of it concentrates in the thyroid gland, the rest passes from the body in urine, and, then, to urban wastewater (sewage).

The ^{131}I activity concentration in the liquid effluent analyzed was 5 Bq/L that is considered LBN (low radiation level) according to Rule CNEN-NE-6.05 established by Resolution CNEN 19/85 published in 17th December 1985. The same resolution points out that, except for ^{3}H and ^{14}C, the total annual amount of radionuclides released into the sewage net should not exceed 3.7×10^{10} Bq. Under this scenario, it would be possible daily release about 20 kt of the wastewater analyzed.

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