Natural Gamma Emitters after a Selective Chemical Separation of a TENORM Residue: Preliminary Results

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An analytical procedure was established in order to obtain selective fractions containing radium isotopes (^{228}Ra), thorium (^{232}Th), and rare earths from RETOTER (REsíduo de TÓrio e TErras Raras), a solid residue rich in rare earth elements, thorium isotopes and small amount of natural uranium generated from the operation of a thorium pilot plant for purification and production of pure thorium nitrate at IPEN ï CNEN/SP. The paper presents preliminary results of ^{228}Ra, ^{226}Ra, ^{238}U, ^{210}Pb, and ^{40}K concentrations in the selective fractions and total residue determined by high-resolution gamma spectroscopy, considering radioactive equilibrium of the samples.

Keywords: solid waste, gamma emitter, natural radioactivity, TENORM.
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INTRODUCTION

TENORM stands for *Technologically-Enhanced Naturally Occurring Radioactive Material* and the majority of radionuclides in TENORM are found in the uranium and thorium decay chains [1].

There are many areas of high natural radioactivity throughout the world. Between them we can mention the mineralized volcanic intrusions in the Brazilian State of Minas Gerais and the monazite-bearing sands found in the Kerala coast in India and in the coastal regions of the Brazilian State of Espírito Santo [2].

Thorium takes a vanguard place as nuclear fuel in breeders reactors or regenerators In Brazil, one of sources of thorium originates from monazite, which contains 65% of rare earths oxide, 5 - 6% of thorium oxide and 0,15 - 0,20% of uranium oxide [3, 4].

In the early ´1970s, at IPEN-CNEN/SP, a pure thorium nitrate purification plant was established, in order to supply several national industries, like electrical, ceramic, etc., and external markets with thorium [5, 6, 7]. A solid residue, RETOTER (REsíduo de TÓrio e TErras Raras) was derived after the wet process production of Pure Thorium Nitrate, being considered as nuclear waste meaning a financial cost of

storage, management and safeguards facing the Brazilian radioprotection regulatory rules. Nowadays, there are about 25 MT of RETOTER stored at IPEN-CNEN/SP facility safeguards [8] and can be considered as TENORM. This study intends to develop an analytical method for the residue decontamination, separating and characterization of similar chemical groups, containing some specific radioactive isotope [9].

METHODS

The residue was digested with nitric acid [9] and the ^{228}Ra, major radionuclide, was separated by $BaSO_4$ co-precipitation procedure [10]. After, the thorium was separated by the use of peroxide precipitation and the rare earth elements group were recovered in the peroxide filtrate solution [9, 11], as can be seen in Figure 1.

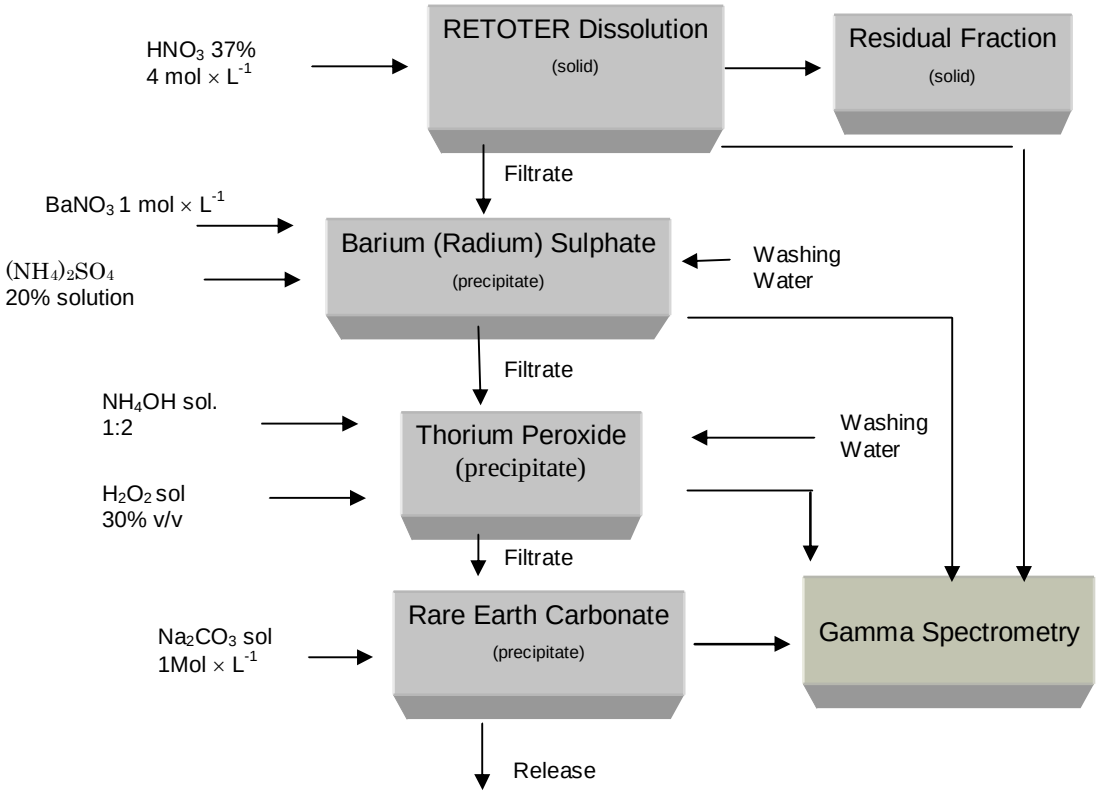


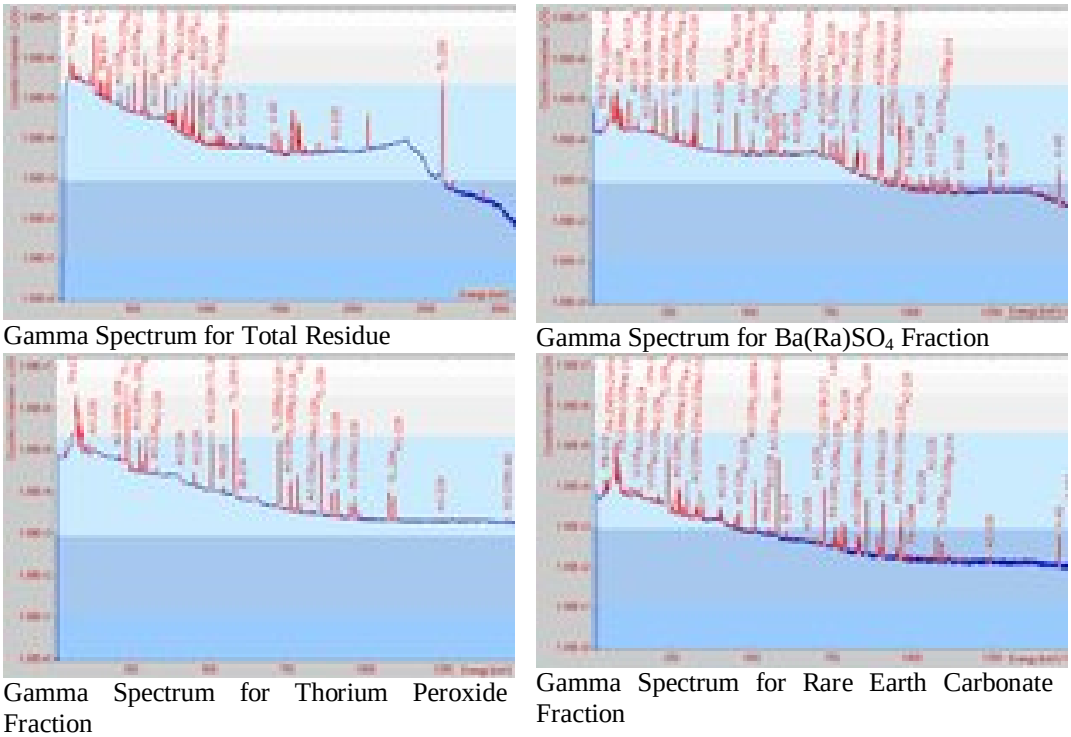
FIGURE 1. Flowchart of the Analytical Procedure for Selective Fractions Obtain and Gamma Emitter Measurement.

Sealed samples of the total residue and analytical selective fractions were measured by high-resolution gamma spectrometry, after reaching radioactive equilibrium [12].

RESULTS

The procedure results for total residue and analytical fraction of $Ba(Ra)SO_4$ had showed photopics of gamma energy from ^{228}Ac, demonstrating that the radioisotope ^{228}Ra was separated as co-precipitate in the barium sulphate one selective fraction, being the main radioisotope gamma emitter in the total residue and fractions $Ba(Ra)SO_4$.

The gamma spectra of each sample are showed in Figure 2. Main gamma energies transitions are identified for several radioisotopes.



Gamma Spectrum for Total Residue

Gamma Spectrum for $Ba(Ra)SO_4$ Fraction

Gamma Spectrum for Thorium Peroxide Fraction

Gamma Spectrum for Rare Earth Carbonate Fraction

FIGURE 2. Gamma Spectra of Total Residue and Selective Fractions.

Table 1 presents the quantitative results for gamma emitters in the samples. The uncertainty is specified for 1 sigma, 95% of confidence.

TABLE 1. Massic Activity of Analyzed Gamma Emitters in Total Residue and Selective Fractions.

	Activity ($Bq.g^{-1}$)				
Sample	**^{228}Ra**	**^{226}Ra**	**^{210}Pb**	**^{238}U**	**^{40}K**
Total Residue	5675 ± 4	nd	nd	nd	165 ± 5
Barium (Ra) Sulphate	34520 ± 410	667 ± 43	38 ± 9	1229 ± 140	1344 ± 54
Thorium Peroxide	194 ± 8	nd	nd	nd	nd
Rare Earth Carbonate	407 ± 5	nd	208 ± 10	107±26	16 ± 3

nd: not determined

CONCLUSION

As can be seen from Figure 2 and Table 1, the preliminary results obtained by high-resolution gamma-spectrometry allow to conclude that the analytical procedure established in order to obtain selective fractions containing radium isotopes (^{228}Ra), thorium (^{232}Th), and rare earths from RETOTER (REsíduo de TÓrio e TErras Raras), is effective when used to separate radium isotopes from solid residue.

That presents the possibility to improvement in future studies for decontamination and commercial use for the obtained fractions.

In future work, strategies in the management and possible economic and commercial use of this residue also will be considered, as the chemical sequential methodology presented can lead to an improvement in possible studies for decontamination strategies and commercial use for the obtained selective fractions.

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