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Baseline evaluation for natural radioactivity level and radiological hazardous parameters associated with processing of high grade monazite

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ABSTRACT

Egyptian monazite is a promising resource and investment attractive for production of valuable metals of industrial or nuclear interest such as rare earth elements (REEs), thorium (Th) and uranium (U). The study was focused to establish a baseline framework in viewpoint of radiation protection for the workers in production of REEs from high-grade monazite treated by sodium hydroxide (NaOH) solutions. Radiological hazard indices (cancer, gonadal and other risks) were evaluated, due to emissions (α-, β- and γ-radiations) of radium-isotopes (^{228}Ra, ^{226}Ra, ^{223}Ra) and lead (^{210}Pb). The values of the estimated radiological hazard indices were higher than the permissible safe limits, worldwide average and varied with those reported in other countries. It was found that more than 70% of radioactivity and radiological hazardous indices resulted from emissions of ^{228}Ra, while the rest was attributed to ^{226}Ra, ^{223}Ra and ^{210}Pb. Therefore, processing of the Egyptian monazite can cause a significant radiological impact on workers through external exposure from γ-radiations and/or internal exposure through inhalation or ingestion airborne contaminated by the radionuclides. Thus, the results recommended that protection rules could be considered to prevent the radiation hazards associated with the production of the REEs from the high grade monazite attacked by caustic method.

1. Introduction

All living things are continuously exposed to ionizing radiation from naturally occurring radioactive materials (NORM), technically enhanced naturally occurring radioactive materials (TENORM), man-made radionuclides or nuclear accidents. Some studies on radiation levels and radionuclide distribution in the environment provide vital radiological information (Sowole, 2014; Ozdis et al., 2017; Mikhailovskaya et al., 2017; Kumamoto et al., 2017). Such information is significant in understanding human exposure from the natural and/or man-made sources of radiation and necessary in establishing rules and regulations relating to radiation protection programme (Sowole, 2014; Hesham et al., 2016). About 90% of human radiation exposure originates from natural sources such as cosmic radiation, exposure to the short-lived radionuclides of radon gas (^{222}Rn, ^{220}Rn, ^{219}Rn) and long-lived radionuclides of uranium (^{238}U, $t_{1/2}$. 4.5×10^9y) and thorium (^{232}Th, $t_{1/2}$. 1.4×10^{10}y) decay chains as well as singly long-lived radionuclides of potassium (^{40}K, $t_{1/2}$. 1.2×10^{10}y) and rubidium (^{87}Rb, $t_{1/2}$. 4.9×10^{10}y) (Lee et al., 2004).

The geological formations and human activities are important factors enhancing the background levels of the natural radiation. The continuity in increasing these radionuclides in the environment may be attributed to several factors such as mining and milling operations, successive utilization of phosphate fertilizer, burning of fossil fuels (crude oil and coal), and building materials. Ingesting or inhaling such levels of radionuclides contribute significantly to the radiation dose that people receive (Colmenero et al., 2004). In addition, mining and milling of both nuclear and non-nuclear materials may cause significant environmental and occupational radiological impacts, and, typically, TENORM residue in commercial and industrial products has the potential to expose workers and members of the public to some fraction of the

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recommended annual radiation exposure limit. Gamma radiation emitted from ^{232}Th and ^{238}U series and their decay products, and ^{40}K, which exists as trace levels in all ground formations, represents the main external source of irradiation to the human body (UNSCEAR, 2000; Harb et al., 2008; Attia et al., 2015; Petra et al., 2016).

The effects of the radiation emitted by different radionuclides depend on the physical and chemical properties of the NORM and/or TENORM residue (El Afifi and Awwad, 2005; El Afifi et al., 2009; Beliverimis et al., 2010). The interaction of ionizing radiation with human body leads to various serious biological effects (ICRP, 2010, 2014). The exposure to the natural and man-made radiation can cause damage, modifications and/or death to living cells. These alterations may lead to most forms of leukemia and cancers of many organs such as lung, breast and thyroid. There is an increased chance of developing cancer as a result of exposure to radiation about the global average level of natural radiation. The radiation induced cancer may manifest decades later after exposure, which, does not differ from cancers that arise as a result of other factors. This could emanate from natural sources such as cosmic rays and naturally occurring radioactive substances which may be internally or externally (UNSCEAR, 2008). Industry of the REEs production has become an indispensable integral of green technology, their extraction and processing can pose a serious environmental challenge and health risks to plant workers and the entire public in terms of radiation exposure. The raw materials of REEs are a concentrated mixture of REEs and radionuclides of Th, U, and their decay products such as radionuclides of radium-isotopes (^{228}Ra, ^{226}Ra, ^{224}Ra, ^{223}Ra), radon gas, ^{210}Pb, ^{210}Bi and ^{210}Po as well as stable lead isotope (^{206}Pb and ^{208}Pb) (Schmidt, 2013). Until recently, monazite, xenotime and bastnasite were the most important resources for extraction of the rare earth elements (REEs) (Paschoa, 2008).

The chemical attack of the monazite mineral separates concentrates of REEs and valuable elements of nuclear interest as Th and U (Xhixha et al., 2013). The processing of the ore can therefore concentrate these NORMs in the wastes (TENORM), which, if not handled effectively, can become a channel of public exposure. Thorium dust, which is a known cancer-inducing agent, is easily blown by the wind and carried by water over long distances, thereby create radiation hazards over a large span of areas. Thus, all the steps of REEs production, from mining through transportation, processing and waste disposal stages, are potential pathways for contamination of soil and water by radioactive and hazardous chemicals (Kolo et al., 2015). In literature, there are few studies have been carried out on the radiological hazard indices in soil, sediment, water samples, or TENORM residues associated with many industrial activities in different parts of the world (Colmenero et al., 2004; El Afifi et al., 2006; Kolo et al., 2015; Zubair et al., 2013; Srilatha et al., 2015; Mohammad et al., 2015; Karim, 2016; Hamed et al., 2016; Arabi et al., 2016; Rangaswamy et al., 2016; Wang et al., 2016; Borai et al., 2017).

Recently, due presence significant amounts of monazite in Egypt, extraction and production of the rare earth oxides from monazite ores can be considered as one of the promising industries and attractive for the exterior or interior investment. Therefore, the main objective of this study is attempting to establish baseline for the possible activity concentration and radiological hazardous indices associated with the processing of monazite digested by caustic soda. Activity concentrations of the radium-isotopes (^{228}Ra, ^{226}Ra and ^{223}Ra), lead (^{210}Pb) and potassium (^{40}K) were measured and evaluated in monazite ore, hydroxide cake, sodium phosphate liquor (Na_3PO_4) and washing effluents (I and II). The results obtained were used to estimate different ten radiological hazardous indices (I_γ, I_α, H_{ex}, H_{in}, D_R, E, AGDE and ELCR) to predict the possible health risks affect on the workers in the recent strategic industry in Egypt, i.e., production of lanthanides.

2. Experimental

2.1. Samples and caustic digestion

The monazite used in this study was of a high grade brand, i.e., monazite (92%). Monazite was separated from black sand deposits obtained from the Rosetta beach (El Behaira Governorate, 263 km North of Cairo, **N**: 31° 24′ 05″, **E**: 30° 25′ 10″) on the Mediterranean Sea by the Nuclear Material Authority, Cairo, Egypt. Monazite samples were digested by the caustic method using a hot solution of sodium hydroxide (NaOH) through main two steps. An accurate 50 g monazite ore was added to 250 ml of 40% NaOH with stirring for 4 h at 140 ± 2 °C. In the second step, 20% NaOH was added while stirring for 1 h at 140 ± 2 °C. The solid phase was separated from the supernatant of sodium phosphate (Na_3PO_4) and excess of NaOH by decantation and washed twice with hot distilled water. The produced hydroxide cake was dried in oven at 110 ± 1 °C, then cooled to room temperature, crushed, sieved, homogenized and prepared for analysis. Liquors of sodium phosphate and washing effluents (I and II) were collected and prepared for analysis by γ-ray spectrometry. Conversion of monazite to the respective hydroxide cake by caustic method is illustrated in Fig. 1.

2.2. Preparation of the samples for the radiometric analysis

Samples of monazite and hydroxide cake were dried in an oven at 110 °C until constant dry weight to remove the moisture content. The samples were cooled at room temperature, crushed, pulverized, homogenized and sieved to <100 μm. For the radiometric measurements, 50 g of each sample were accurately weighed and packed in rounded polyethylene bottles (150-cm^3). An exact 100 ml of the liquors was packed in bottles. The bottles were closed tightly, sealed by molten wax to prevent escape of radon gas and preserved for 30 days to attain the secular equilibrium between radium-isotopes (^{228}Ra and ^{226}Ra) and their respective decay daughters (Hilal et al., 2015; El Afifi et al., 2015; Borai et al., 2017).

2.3. Nondestructive gamma ray measurements

Radiometric analysis of samples was done using a high purity germanium (HPGe) detector (Model: GX1020-10863 with liquid nitrogen (LN_2) monitor, Operating voltage: +3500 V, Relative efficiency: 10%, Resolution at full width half maximum (FWHM): 2 keV at 1332.5 keV peak of ^{60}Co, Data acquisition system: GIENE-2000 and Multichannel Analyzer (MCA): 8192 k). The measuring system was provided by the Canberra Industries Inc., USA. The energy calibration of the HPGe-detector was performed by sealed point source containing 0.1 μCi (3.7 × 10^3 Bq) of ^{60}Co, ^{133}Ba and ^{137}Cs (Amersham, England), while the efficiency calibration was carried out by certified reference material-stream sediment, namely IAEA-314 containing natural radionuclides of ^{238}U, ^{232}Th and ^{226}Ra (AQCS-IAEA, Seibersdorf, Austria) and certified reference

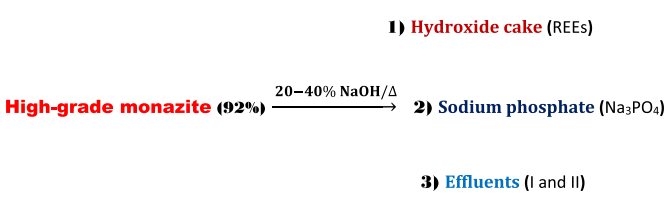

Fig. 1. Flowchart shows digestion of monazite by caustic method.