Radioactivity levels and concentrations of heavy elements in black sand

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Abstract. The radioactivity levels and concentrations of heavy elements in Langkawi black sand have been studied through gamma ray spectroscopy. Direct gamma counting was used for measuring the activity of the natural radionuclides uranium, thorium and potassium, and neutron activation analysis for detecting the presence of other heavy elements. Our results show the activity of uranium is 44.5 pCi g⁻¹, which is four times higher than that of thorium (11.6 pCi g⁻¹) and eight times that of potassium (5.5 pCi g⁻¹). Ytterbium, thorium, europium, cesium, antimony, scandium, gallium, iron, chromium, cobalt, cerium, arsenic, lanthanum, samarium and hafnium were detected; their concentrations range from 4.9 to 16072 ppm.

Abstrak. Aras keradioaktifan dan kepekaian unsur berat di pasir hitam telah dikaji secara spektroskopisinar gamma. Penghitungan terus gamma digunakan untuk penentuan keaktifan radionuklid semula jadi uranium, thorium dan kalium, dan analisis pengaktifan neutron untuk mengesan kehadiran unsur berat lain. Hasil kami menunjukkan bahawa aktifan uranium adalah 44.5 pCi g⁻¹, yang empat kali aktifan thorium (11.6 pCi g⁻¹) dan lapan kali aktifan kalium (5.5 pCi g⁻¹). Ytterbium, thorium, europium, cesium, antimoni, scandium, gallium, iron, chromium, kobalt, sirium, arsenik, lanthanum, samarium dan hafnium telah dikesan; jutal kepekaan adalah 4.9 ke 16072 ppm.

Introduction

Radioactivity in the environment results from the decay of radionuclides that are present in the ground; the distribution of radionuclides in the ground has been well documented in the literature [1]. The main natural radionuclides include potassium, thorium and uranium. Although most of the gamma rays emitted by the nuclides are absorbed by the ground, those originating near the surface pose a health hazard, and the extent of human exposure to the radiation depends on factors such as air-flow pattern and time of exposure.

Our work is a study on the level of radioactivity in the black sand found at Pasir Hitam beach on Langkawi Island (100° E and 6° N), Malaysia. The sand contains monazite (CeLaYThPO₄), a mixed phosphate salt of thorium and other heavy metals [2]. The Pasir Hitam (literally, black sand) beach stretches for almost two kilometers, and the sand is black owing to the high content of ilmenite (FeTiO₃).

A study the natural radioactivity of the black sand may be of geological interest as a sand having an elemental concentration above 10 ppm indicates possible mineralisation. In this study, a direct gamma-ray counting method was used for the activity measurements whereas the concentration of heavy elements was determined by neutron activation analysis.

Experimental

The black sand (top 5 cm) was collected from four different sites on Pasir Hitam beach into plastic tubes (3 cm diameter, 15 cm long) that were cleaned with 10% nitric acid. The samples were then homogenised and air dried. Representative samples were used for the radioactivity measurements and for the analysis of heavy elements.

Direct gamma counting

The gamma ray spectra of the samples were recorded on a high-resolution HPGe gamma ray...
detector (20% efficiency) that was coupled to a
PC-based multichannel analyser. The detector
was maintained in vertical position inside a
cylindrical 12-cm x 1 m lead shield. The
resolution of $^{60}$Co as measured by the FWHM at
1332 keV energy is 1.90 keV. The absolute
efficiency of the detector was calibrated by
standard sources and a standard reference
material IAEA-368 (sediment), which took into
account the geometry of the samples.

The samples were placed in sealed
cylindrical 300 mL plastic containers and then
set aside for four weeks to enable the radon gas
in the U series to equilibrate with the other
daughter nuclei. A series of measurements was
performed by placing the sample approximately
10 cm from the detector. For each measurement,
the observed counting rate was corrected for the
background contribution. The counting time for
all samples was 10 hours.

The radionuclides measured were $^{238}$U,
$^{232}$Th and $^{40}$K. The $^{226}$Ra (or $^{238}$U for samples
at radioactive equilibrium) and $^{232}$Th
radioactivities were estimated from the 609.3
keV and 583.1 keV gamma lines of $^{214}$Bi and
$^{208}$Tl, respectively. The $^{40}$K radioactivity was
determined by using the 1460 keV gamma line.

The equation for the specific activity is:

$$A_S = C_S \cdot e \cdot Pr \cdot Ms$$

where

$C_S = \text{count rate of the analytical line}$
$e = \text{detector efficiency of the specific line}$
$Pr = \text{absolute transition probability of}$
$\text{gamma decay through the line, and}$
$Ms = \text{mass of the sample (kg)}$

In order to estimate the doses 1 m above ground,

$$D = aC$$

where

$D = \text{dose rate, mrad yr}^{-1}$
$C = \text{concentration, pCi g}^{-1}$, and
$a = 17.8 \text{ for } ^{238}U \text{ (+decay products)} \text{ and}$
$25.5 \text{ for } ^{232}Th \text{ (+decay products)} \text{ and}$
$1.6 \text{ for } ^{40}K.$

**Neutron Activation Analysis (NAA)**

The sand samples were placed inside
polyethylene vials (2/5 drag), sealed and then
inserted into irradiation containers and irradiated
with neutron fluxes of $4 \times 10^{12} \text{ ncm}^{-2} \text{s}^{-1}$ at the
Malaysian Institute of Nuclear Technology
(MINT) nuclear reactor TRIGA MK II.
Irradiation time was set at 10 hours, with a
cooling time of two weeks. The presence of
heavy elements in the samples was deduced from
the detection of of gamma rays emitted by
induced radionuclide activities [3]. The method
of comparison was used to calculate the
concentration level of each element and the
comparator used was SRM Coal Ash 1632a.

**Results and Discussion**

Table 1 shows the result of the activity
measurements by the method of direct gamma
counting that are expressed as pCi g$^{-1}$. Our
results indicate the presence of natural
radionuclides; U, Th and K in Langkawi black
sand. We observed that the activity of uranium
is higher than the activities contributed by
thorium and potassium. Generally, the
percentage error from our measurements are
relatively small between 5 - 7 per cent which are
attributed from the counting statistics.

The second and third row of the table show
the respective activity values obtained from
Egyptian black sand [4] and sandstone [5]. The
activity values obtained from our samples are
greater than the activities of the other two types
of sand. For instance, the activity of uranium in
Langkawi black sand is more than 100 times
higher than the activity found in sandstone and
approximately 8 times greater than that of
Egyptian black sand.

The last column of Table 1 show the
exposure rate calculated for the three types of
sand as expressed in microrad per hour. Again
we observed that the rate derived from our black
sand is higher almost 4 times greater than that of
Egyptian sand and 30 times that of the normal
sandstone. Table 2 shows the concentrations of
heavy trace elements in Langkawi black sands as
obtained by the method of instrumental neutron activation analysis.

Fifteen heavy elements; Yb, Tb, Eu, Cs, Sb, Sc, Ga, Fe, Cr, Co, Ce, As, La, Sm and Hf were detected by NAA with concentrations ranging from 5.4 ppm to as high as 16072 ppm dry weight. Our analysis have shown that Langkawi black sand contains considerable amount of europium, approximately 16 mg g\(^{-1}\) followed by hafnium and cerium. With this high concentration of heavy elements there is a possibility that mineral extraction can be carried out in future which on the other hand has to be weighed against the economic costs.

Langkawi black sand has been shown to contain naturally occurring radioisotopes \(^{232}\)Th, \(^{238}\)Ra and \(^{40}\)K as well as several important heavy elements. The main radioactivity is contributed by uranium and the overall exposure rate was found to be small.

References


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<th>Table 1. Activities of radionuclides expressed as pCi g(^{-1})</th>
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<tr>
<td>Langkawi black sand</td>
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<td>Egyptian black sand</td>
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<td>Sandstone</td>
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<th>Table 2. Concentrations of heavy elements in Langkawi black sand</th>
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<td>Element</td>
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