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## Presence of uranium and thorium in zircon assemblages separated from beach sands of Cox's Bazar, Bangladesh

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### Article info.

### ABSTRACT

#### Key Words:

Radioactive minerals, Beach placers, Separation flow chart, XRD, Neutron activation analysis

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*Beach sands harbor appreciable amount of radioactive elements such as zircon which contains some important and radioactive elements (e.g. uranium, thorium). Very limited quantitative information is available of such elements in the sand of the beaches of Cox's Bazar in Bangladesh. The present study presents application of Neutron Activation Analysis (NAA) to quantify uranium and thorium elemental concentration in zircon assemblages from beach placers of Cox's Bazar. Zircon was separated from beach sands in a mineral processing pilot plant. The separated zircon assemblages were examined by a polarizing petrographic microscope and X-ray diffraction techniques and found that it contained more than 75% pure zircon. Thereafter, uranium and thorium concentrations in zircon were measured by NAA and revealed that zircon contained 94-141 ppm uranium and 127-506 ppm thorium. Therefore, this noticeable amount of uranium and thorium in zircon should be taken in consideration during any mining attempt of the placer minerals present in Cox's Bazar beaches.*

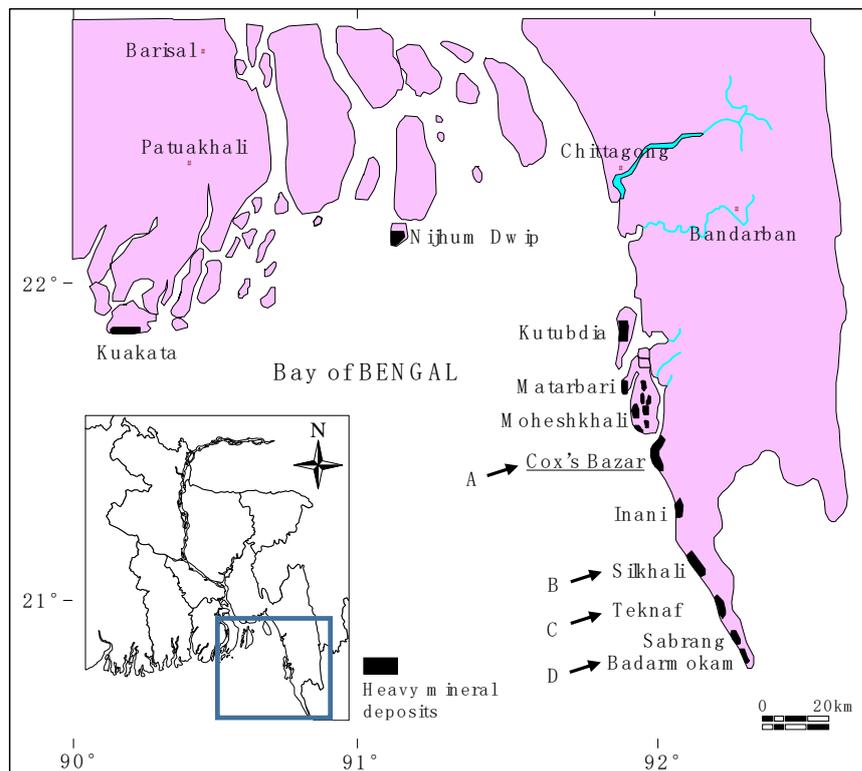
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## I. Introduction

Radioactive minerals (e.g. monazite, zircon, etc.) containing uranium (U) and thorium (Th) are very much associated with beach placer minerals as found in many places in Australia, India, Brazil, etc. In

recent studies, radioactivity levels in heavy mineral rich beach sands are mostly focused due to their environmental impact, effect on human health, potentiality of commercial exploitation, etc. (Gandhi *et al.*, 2014; Arnedo *et al.*, 2013; Mahawatte and Fernando, 2013; Lu and Zhang, 2008; Veiga *et al.*, 2006; Ramasamy *et al.*, 2004; Kannan *et al.*, 2002). Presence of radioactivity in Cox's Bazar beach of Bangladesh was first reported by Schimdt & Asad (1963). Heavy placer deposits of Bangladesh coast contain several heavy minerals, in which monazite is radioactive mineral because of thorium in its composition (Zaman *et al.*, 2009). Zircon is also radioactive to some extent because it contains radioactive elements in its composition (Bazzari *et al.*, 1986; Evans *et al.*, 2005; Natarajan *et al.*, 2012). However, there is very limited information about the quantification of radioactive elements (particularly U and Th) in zircon from Cox's Bazar except very recent findings from Sasaki *et al.* (2015) and Zaman *et al.* (2012). The presence of such radioactive elements in zircon or other minerals could hamper the mining attempt of heavy minerals in coastal areas. If anomalous radioactivity arises from zircon, it will create occupation hazards for mine workers. Besides, they could be potential source for radioactivity release in the marine environment by the processes of weathering, erosion, leaching through groundwater recharge, etc. Therefore, it is necessary to characterize the zircon with respect to the presence of radioactive elements.



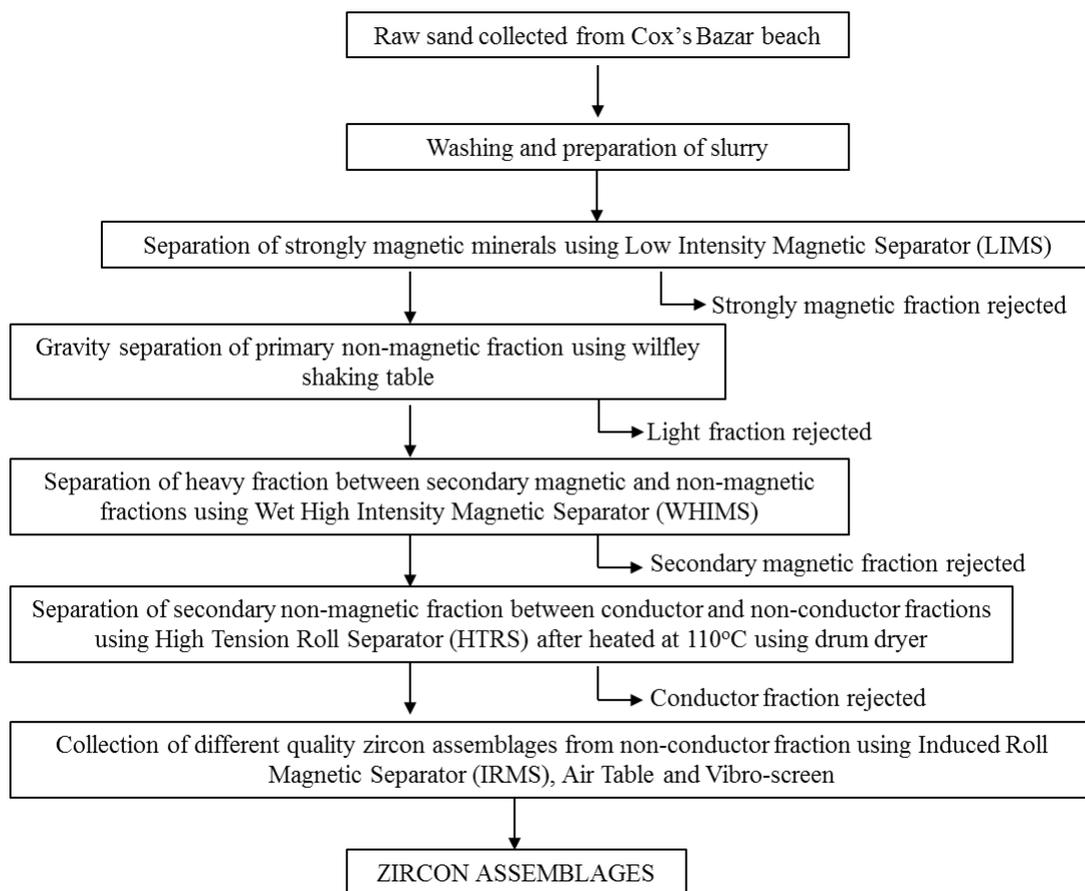
**Figure 01. Location of heavy mineral deposits along the coastal belt of Bangladesh as explored by BAEC through geological exploration at BSMEC (adopted from Sasaki *et al.*, 2015).**

Bangladesh Atomic Energy Commission (BAEC) has long been involved in the exploration and research activities on heavy placer deposits along the coastal belt of Bangladesh. As a result, as many as 18 heavy mineral deposits were identified locating 16 at Teknaf, Inani, Sonarpara, Cox's Bazar, Moheshkhali and Kutubdia at the south, and Nizhum dwip and Kuakata at the northern coast (Figure01). The aim of the exploration of heavy minerals through BAEC was because of inclusion of radioactive materials. In course of time, due to both natural and artificial physiographic changes like, natural erosion and shifting of heavy mineral deposits, excavation of deposits for construction purposes, etc. didn't let the commercial exploitation of those economic minerals. Hence, maximum economic value can be achieved if the radioactive materials can be extracted from these beach minerals (Sasaki *et al.*, 2015). Therefore, the objective of the present study was to separate zircon from beach sand by the processes of specific gravity, magnetic and electric separations, to verify their presence by microscopic and X-ray Diffraction

(XRD) technique, and to measure the concentrations of both uranium and thorium in zircon using Neutron Activation Analysis (NAA).

## II. Materials and Methods

Bulk sand was collected randomly for processing in a pilot plant from the fore-dune part (geological name of the recent deposition of the beach just above the interaction of tidal waves) of Cox's Bazar heavy mineral deposits. The collected sand was dark brown to black color and ranged from 0.25 to 0.063 mm in grain size having more than 80% heavy minerals as separated arbitrarily by liquid gravity separation. The raw sands were processed in the pilot plant of Beach Sand Minerals Exploitation Centre (BSMEC). The plant is equipped with various gravity, electric, magnetic and grain size separators in order to separate five economically potential heavy minerals (magnetite, ilmenite, zircon, garnet, rutile). Using pilot plant processing technique, zircon assemblages were separated from heavy sands according to a separation flow sheet (Figure02).



**Figure 02. Flow chart of the separation technique of zircon assemblage from raw sand using mineral processing pilot plant.**

**Mineralogical composition of separated zircon assemblages:** From zircon assemblages, zircon crystals (about 300) were mounted on glass slides using medium of Canada balsam. These slides were taken for microscopic studies for the identifications and quantifications of zircons along with other mineral components in each sample. Using trinocular polarizing microscope (Model-Meiji Techno ML9000 Series), 85 grain slides of 5 sample populations were undergone grain counting analysis in order to determine the individual mineral percentage. The individual mineral percentage in a particular sample was determined following the procedure of [Macdonald \(1972\)](#).

**X-Ray Diffraction:** The presence of zircon was tested by X-ray diffraction, as this technique is one of the most reliable mineral identification and characterization techniques used by the geologists ([USGS](#),

1997). XRD peaks were recorded using a Philips PW 3040 X-Pert PRO XRD system with Cu-K $\alpha$  radiation, operated at 40 KV and 30mA, with angular range  $10^{\circ} \leq 2\theta \leq 90^{\circ}$ . Analysis of the XRD peaks was performed that enabled absolute determination of the minerals in the samples. This was achieved by comparing XRD recorded curves of the samples with known spectra from a large mineral database and by identifying the observed peaks.

**Neutron Activation Analysis (NAA) to get radioactive concentration:** Neutron activation analysis is a nuclear technique to determine the concentration of elements based on the excitation by neutrons so that the treated samples emit gamma-rays. It is a very much reliable technique in the field of chemistry, geology, archeology, medicine, environmental monitoring and even in the forensic science. The technique is based on the conversion of a stable nucleus into a radioactive one by neutron capture. In Bangladesh, NAA has been used for different sensitive investigations with the irradiation by a 3 MW TRIGA Mark II Research Reactor of BAEC. Since the technique is very sensitive, therefore, used for minor elements present in very low concentrations and trace element analysis. Application of NAA to determine U and Th is thus very trustworthy.

For NAA analysis, samples were dried, ground and powdered, and 50 mg of each sample was used. For standardization, the Standard Reference Material (SRM) IAEA Soil-7 and for quality assurance, IAEA S-1 was used. Then the samples and standards were simultaneously put into the PE (Poly Ethylene) irradiation tube and irradiated using the reactor. After irradiation, the samples and standards were allowed to decay 3~5 days for uranium and 7 weeks for thorium before gamma ray counting by a High Purity Germanium (HPGe) detector coupled with a PC based S-100MCA master board packages. The 277 keV and 312 keV gamma ray peaks were used for the determination of uranium and thorium, respectively. The concentration of uranium and thorium in the samples were calculated using the relative standardization method.

### III. Results and Discussion

Microscopic identification of separated zircon samples from pilot plant reveals that the percentage of zircon in most of the samples are more than 75% followed by minor amount of rutile, garnet and unidentified minerals (Table 01).

Microscopic observation also identified the presence of numerous secondary materials as inclusions in zircon grains (Figure 03). Zircons of Bangladesh show some typical prismatic shape containing different types of inclusions, and transparent in cross polarized light, whereas some others are mostly sub-rounded and not as transparent as typical zircon grains. Inclusions in zircon grains may contain uranium whereas later types of grains sometimes include the mineral monazite (Sasaki *et al.*, 2015) of which Th is a constituting element.

**Table 01. Modal analysis of five sample populations collected from Cox's Bazar beach**

Sample ID	Zircon	Rutile	Garnet	Unidentified minerals
Zrn-1	85.93	0.00	0.42	13.65
Zrn-2	75.08	4.05	0.78	20.09
Zrn-3	89.47	0.20	0.13	10.20
Zrn-4	73.87	4.49	0.91	20.73
Zrn-5	84.38	1.07	0.71	13.84

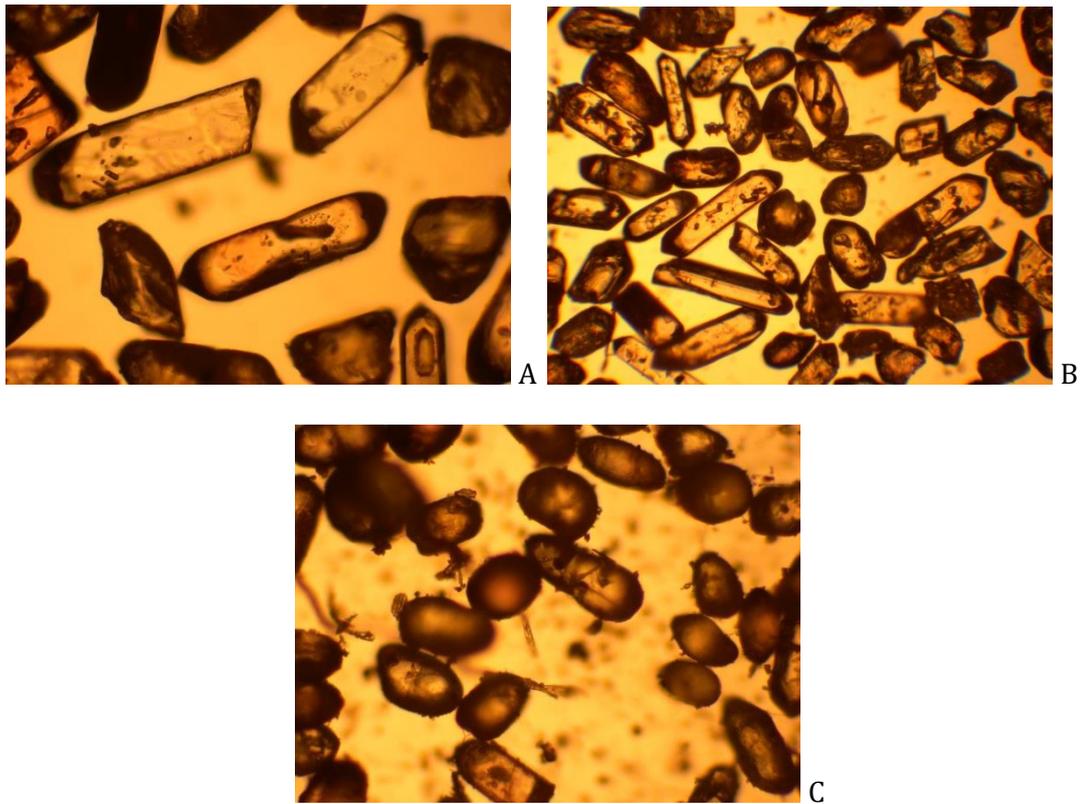


Figure 03. Photomicrographs showing fresh zircon grains (A), zircon with numerous inclusions (B) and monazite grains (C). Photographs were taken with Petrographic Ore Microscope at plane polarized light at  $\times 10$  (A&C),  $\times 4$  (B).

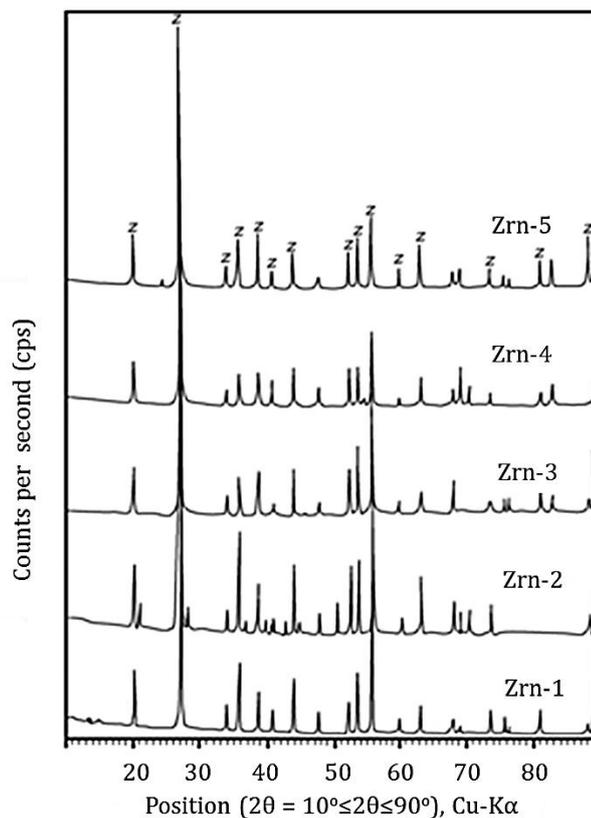
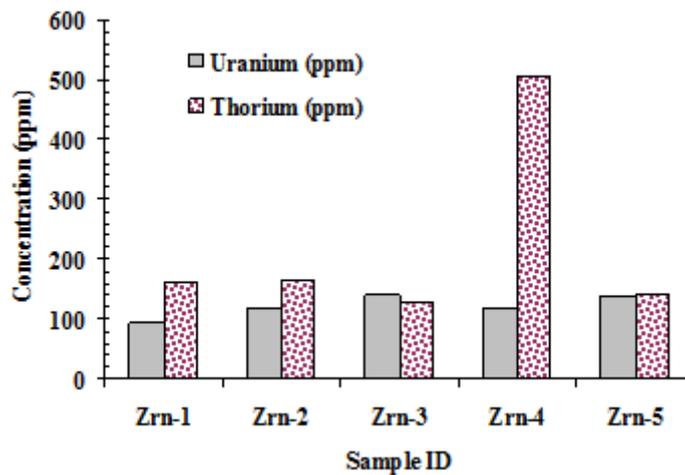
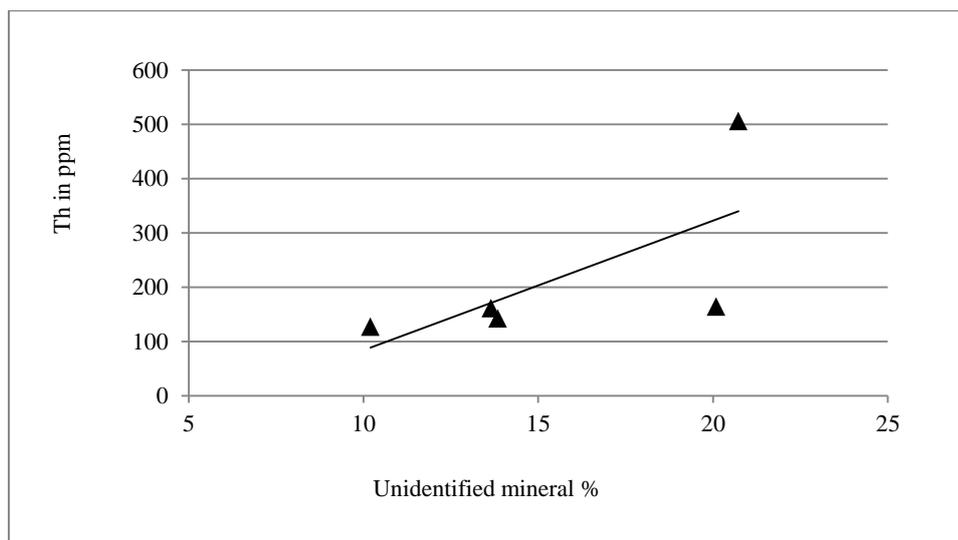


Figure 04. X-Ray Diffraction peaks of zircon assemblages (Z is for zircon).



**Figure 05. Concentration of uranium and thorium in zircon assemblages determined by NAA.**

Monazite grains are very fine and sometimes resembles with a variety of zircon. Pleochroic halos (i.e., ring of color produced around a radioactive impurity included in a mineral by alpha particles emitted from the radioactive elements in the inclusion) were also seen in some zircon grains. XRD study of those samples reflected that the samples are principally composed of zircon with some minor amount of other minerals (e.g. garnet, rutile, etc.) (Figure 04). NAA revealed that the average concentrations of uranium and thorium were 122 ppm and 220 ppm, respectively. The maximum concentrations of uranium and thorium were found as 141 ppm and 506 ppm, respectively (Figure05). There was also somewhat increasing trend with Th content with the increase of unidentified mineral content (Figure06). That may indicate the presence of monazite in the unidentified mineral fraction from where Th radioactivity is coming. However, the identification of monazite with sophisticated technology is needed to confirm this.



**Figure 06. Thorium concentration in the un-identified minerals of the zircon assemblages.**

Radioactivity in beach placers of Bangladesh are already reported quite high when properly separated from other non-radioactive fractions (Sasaki *et al.*, 2015; Zaman *et al.*, 2012). The main source of the radioactivity is coming from mostly monazite and zircon as suggested by those researches. However, as separation of monazite is still not possible in bulk amount in BSMEC, all the monazites are probably accumulating in the zircon fraction of the present separation flow chart and producing different grades of zircon. Few other studies have quantified the amount of U and Th along with other radioactive

elements like K by the gamma spectrometry in some soils and heavy minerals of Cox's Bazar region (Alam *et al.*, 1999; Mollah *et al.*, 1987; Chowdhury, 2003; Kabir *et al.*, 2010). However, quantification of U and Th in the regular fractions of zircon assemblages in the processing of pilot plant of BSMEC has not been conducted before. The specific concentration of U and Th in these different graded zircons is thus necessary to quantify as those zircons are sometimes used for general purposes without considering their radioactive content.

Uranium and thorium can be extracted from the placer sand when the radioactive zircon and monazite are enriched with laboratory processing. Although, ore deposits contain much higher amount of U and Th comparing to the present values in placer sands, less concentration can be considered economically extractable as found in several deposits of Australia (Mernagh and Miezitis, 2008). Considering the estimated heavy mineral reserve of 1.76 million tons along Bangladesh coast, the quantity of U and Th obtained from present study could be motivating to consider for commercial exploitation. The production of other heavy minerals during such exploitation is already proven as economically feasible (BSEC, 1994).

#### IV. Conclusion

Beach sands in Cox's Bazar, Bangladesh contain zircons that have significant amount of uranium and thorium. This estimation would help in planning and proper management of entire heavy mineral deposits, especially for any industrial use with the minerals. As the coastal areas of Bangladesh have several heavy mineral deposits in which zircon is one of the major mineral components, the presence of uranium and thorium in zircon should be taken into consideration during any mining attempt of those heavy minerals.

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