



MEASUREMENT OF NATURAL RADIOACTIVITY IN PORCELAIN STONEWARE TILES

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Abstract

This paper presents the results of the measurement of natural radio activity present in the Porcelain stoneware tiles samples collected from the ceramic firms of Government Ceramic institute, Virudhachalam. Natural radioactivity in the porcelain stoneware tiles samples is mainly due to ^{226}Ra , ^{232}Th and ^{40}K and their daughter products. In this study, these measurements have been estimated in Gamma-ray spectrometry and their levels for six samples are compared and also the Radium equivalent concentration (Ra_{eq}) is calculated. The gamma index is calculated and compared well with the reported values.

Key Words: Gamma-ray spectrometry; Natural radio activity; Porcelain stoneware tiles.

Introduction

All building materials contain various amount of natural radioactive nuclides materials derived from rock and soil contain mainly natural radionuclides of the uranium (^{238}U) and thorium (^{232}Th) series, and the radioactive isotope or potassium (^{40}K). In the uranium series the decay chain segment starting from radium (^{226}Ra) is radiologically the most important and, therefore, reference is often made to radium instead of uranium [3]. The knowledge of the natural radioactivity of building materials is important for the determination of population exposure to radiations, as most of the people spend ~80% of their time indoors [5]. High levels of radioactivity in construction materials can increase external and internal indoor exposure currently; a worldwide effort is underway to measure the activity concentrations in building materials.

Porcelain stone ware tiles are one of the commonly used decorative building materials: They are made of a mixture of earthly materials that has been pressed into shape and fired at high temperature. The body of ceramic tiles may then be glazed or left unglazed. Dust-pressed ceramic tiles with water absorption levels $<0.5\%$, and high mechanical and chemical characteristics are known as, 'Fully vitrified stone ware or porcelain stoneware'.

Porcelain stoneware tiles can show natural activity concentration significantly lower than the average values of Earth's crust.

This report contains a summary of result obtained on porcelain stone ware tiles (six) collected from Govt. ceramic Institute (manufacturing firms), Virudhachalam. The survey consisted of measurement of ^{226}Ra , ^{232}Th and ^{40}K activity concentration by HpGe gamma-spectroscopy. The results of this investigation are compared with the finding of similar studies carried out in other countries.

Materials and Methods

Sample collection and preparation

A total of 6 samples of porcelain stoneware tiles were collected from Govt. Ceramic Institute, Virudhachalam (Cuddalore District, Tamilnadu) for the measurements of concentrations. The tiles samples were powdered to obtain even grain size. The dry samples were transferred to uniform (250ml) containers. Weighed, sealed and kept for 4 weeks so that a secular equilibrium between ^{226}Ra and ^{232}Th and their progeny could be

reached (The sealed counting vials were stored for 30 days to allow ²²⁶Ra and its short lived decay products to reach radioactive equilibrium)

Experimental Technique

The concentration of the natural radioactivity (²²⁶Ra, ²³²Th and ⁴⁰K) in the tiles samples, were measured using the gamma ray spectrometer in the Laboratory of Health and Safety Division, IGCAR, Kalpakkam, Tamilnadu NAI(TI) crystal detector of size 3”x3” combined with 8K multi channel analyzer. The technique used for measurement is a direct γ-counting method. The counting time fixed for each sample was 20,000 seconds. The activity of standard sources used were KCL (329 grams), R-226(0.09μci) and Ra28 (6000pci) with the counting time of 20,000 seconds, the minimum detectable activity limits were 13.25 Bq/Kg for K-40, 8.5 Bq/Kg for ²²⁶Ra and 1Bq/Kg for Th-232. In-site dose rate measurement was done by using digital environmental radiation dosimeter (ERDM Type 107, nucleonix).

Results and Discussion

The activity concentration (in Bq Kg⁻¹) of ²²⁶Ra, ²³²Th and ⁴⁰K in the porcelain stone ware tiles samples (S1-S6) were analyzed and the activity values obtained in this study are given in Table.1 and Fig.1.

It may be seen from the table that for porcelain stoneware tiles samples, the values of ²³²Th has been found to be varying from 24.68 to 35.74 BqKg⁻¹. The activity concentration of ²²⁶Ra varies from 13.87 BqKg⁻¹ to 19.09 BqKg⁻¹. The activity concentration of ⁴⁰K varies from 341.09 BqKg⁻¹ to 415.83 BqKg⁻¹

An index Ra_{eq} called the radium equivalent activity is also presented in Table.1 to compare the specific activities of material containing different amounts of radium, thorium and potassium. This is a widely used radiological hazard index and was calculated according [3]

$$Ra_{eq} = C(Ra) + 1.43C(Th) + 0.077C(K) \quad \text{----- (1)}$$

Where, C(Ra),C(Th),and C(k) are the activity of ²²⁶Ra, ²³²Th and ⁴⁰K respectively in BqKg⁻¹. The value of Ra_{eq} of porcelain stoneware tiles samples (S1-S6) is ranging from 76.65 to 98.39 BqKg⁻¹ (Table.1) From the result obtained, there’s an evidence of considerable

variation in the Ra_{eq} of the Six different (proportions) properties of tiles, but also using the same type.

A number of indexes dealing with the assessment of the excess gamma radiation originating from building materials have been proposed [8-12]. In this study, the gamma- index was calculated as proposed by the European Commission [7]. The commission suggests that building materials should be exempted from all restrictions concerning their radioactivity if the excess gamma radiation originating from them increases the annual effective dose of a member of the public by 0.3mSv at the most. On the contrary, does higher than 1mSv should be accepted only in some very exceptional cases where materials are used locally. The European Commission recommends that controls should be based on a dose in the range 0.3-1mSv y⁻¹. This is the excess gamma does to that received outdoors. The European Commission has proposed the following activity concentration index (I) for identifying whether a dose criterion is met:

$$I = \frac{C_{Ra}}{300Bqkg^{-1}} + \frac{C_{Th}}{200Bqkg^{-1}} + \frac{C_K}{3000 Bqkg^{-1}} \quad \text{----- (2)}$$

Where C_{Ra}, C_{Th}, and C_K are the activity concentration of ²²⁶Ra, ²³²Th and ⁴⁰K (Bqkg⁻¹) respectively, in the building material. The activity concentration index shall not exceed the values (0.3 -1 mSv y⁻¹) shown in Table (1). Note that the activity concentration index should be used only as a screening tool for identifying materials which might be of concern. Any actual decision on restricting the use of a material should be based on a separate dose assessment. Such assessment should be based on scenarios where the materials is used in typical way for the type of material in question.

Large variation in Ra_{eq} activities has been reported in many studies on natural radioactivity in building materials. For examples, in 1985 Beretka and Mathew [4] calculated Ra_{eq} values ranging between 15 and 883BqKg⁻¹ in Australian building materials. The values obtained by Amrani and Tahtat [5] in Algeria building materials ranged from 28 to 190 BqKg⁻¹. In Egypt, Ahmed [6] recorded the lowest values in mud and clay bricks (about 100 BqKg⁻¹), and the highest ones granites and marbles (about 400BqKg⁻¹).

The recommended maximum levels of radium equivalent for building materials to be used for homes, i.e., to keep the external dose below 1.5mGyy⁻¹, is less than 370BqKg⁻¹. It is worth noting that the average

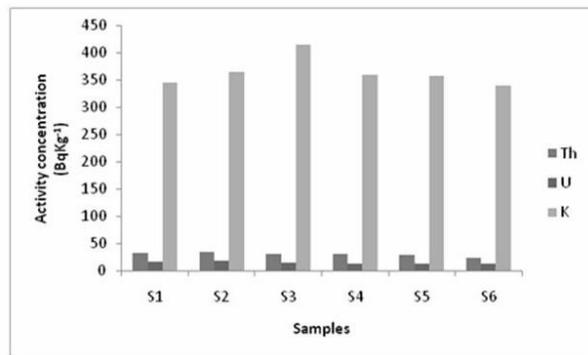
radium equivalent values are within the limit proposed by the OECD countries. Nevertheless, it is important to note that this recommended value is calculated for materials used in bulk amount, and it is not appropriate for decorative building materials such as tiles.

Table. 1 The activity concentrations of ²³²Th, ²²⁶Ra and ⁴⁰K in porcelaine stoneware tiles samples

S.No	Samples	²³² Th (Bqkg ⁻¹)	²²⁶ Ra (Bqkg ⁻¹)	⁴⁰ K (Bqkg ⁻¹)	Radium equivalent activity	Activity concentration index
1	S1	33.90	17.07	345.94	92.8143	0.34381
2	S2	35.74	19.09	366.18	98.3940	0.36439
3	S3	32.62	15.78	415.83	94.4455	0.35431
4	S4	32.61	13.87	361.30	88.3224	0.32971
5	S5	30.06	14.75	358.93	85.3734	0.319109
6	S6	24.68	15.10	341.09	76.6563	0.287426

Gamma indexes of the sample are shown in Table.1. The gamma index (I) has been found to be verifying from 0.28 to 0.36. The European commission [2] suggests that building materials should be exempted from all restrictions concerning their radioactivity if the excess gamma radiation originality from them increases the annual effective dose of a member of the public by 0.3 mSv as the most, corresponding to a gamma index <1 (Table.1) on the contraly, closes higher than 1m(corresponding to a gamma index >6) should be accepted only in some very exceptional cases, when materials are used locally six porcelain stone ware tiles show gamma indices lower than 1.

Fig. 1. Activity concentrations of ²³²Th, ²²⁶Ra and ⁴⁰K for six porcelaine stoneware tiles samples (S1-S6)



Activity concentration and radium equivalent activities for porcelain stone ware tiles are compared with results obtained by various authors in Tables.2. It is possible to observe, activity concentrations results lower than those reported by various authors.

Table. 2. Comparison of activity concentrations and radium equivalents (Bq kg⁻¹) in porcelain stoneware tiles

Country	²²⁶ Ra (Bqkg ⁻¹)		²³² Th (Bqkg ⁻¹)		⁴⁰ K (Bqkg ⁻¹)		Ra _{eq} (Bqkg ⁻¹)		Reference
	Mini	Max	Mini	Max	Mini	Max	Mini	Max	
South korea	44	82	34	96	310	1019	124	264	Leeetal [20]
Algeris	55	-	41	-	410	-	145	-	Ammari& Tahat [5]
India	28	-	64	-	24	-	121	-	Kumar et al [14]
China	64	131	55	107	561	867	200	331	Xinwazi [15]
Egypt	40	230	10	130	80	600	-	-	Ahmed [6]
Egypt	61	118	55	98	730	1050	267	-	Elafifi et al [16]
Greece	25	174	29	47	411	786	-	-	Kretic et al [17]
Spain	75	191	68	76	507	490	-	-	Sernadell et al [18]
Italy(porous fired tiles)	27	88	42	69	544	977	-	-	Bruzzi et al [19]
Italy(porela in stone ware tiles)	39	247	40	92	528	1000	-	-	Bruzzi et al [19]
Italy(porous fired tiles)	36	87	38	86	411	996	130	261	F ₁
Italy(porela in stone ware tiles)	20	708	33	145	158	830	93	943	F ₁
India (porcelain stoneware tiles)	7.14	26.14	18.9	42.13	307.4	452.03	59.4	117.3	Present Study

Conclusion

Activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K were measured and next radium equivalent activities and gamma indexes were calculated in a six number of porcelain stoneware tiles. Results confirm that mean values measured in porcelain stoneware tiles are comparable with mean worldwide value in earth crust. From a radiological point of view, the results indicate that the use of these materials in construction of dwelling could be considered safe for inhabitants. As matter of that, all samples shown gamma indexes much lower than the limit indicated by the European Commission and the radium equivalent activity is within the limit set by the OECD.

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