Dose Assessment for Natural Radioactive Nuclides in Tile as Decorative Building Material

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Abstract

A total of 60 kinds of tiles available in Taiwan were sampled and their natural radioactivities were measured by gamma spectroscopy. A model for estimating indoor dose rate based on QAD-CG computer code package was used to evaluate additional ambient gamma dose induced by these samples. Typical parameters of rooms in Taiwan are simulated, such as room size, wall thickness, occupancy of bedroom, living room and bathroom. The energies and gamma intensities of uranium and thorium series were taken from the newest NUDAT database. The results show that the samples for wall decoration induce less dose than those for floor. The radioactivity is mostly from zircon used in glaze that varies due to different purity and formula recipe. Self-absorption effect is about 3% to 15% and can be neglected . Tiles that used in next room contribute about 16% for the dose increase. Dose rate on infinite tile surface is about 3.7 times of that at 1 m away, and only the later was used for ambient dose equivalent evaluation. The evaluated results were compared to the associated activity indices that were defined by former USSR and West Germany. Activity indices of about 27% of samples exceed suggested limit that seems to be much more than other country's experiences. However, the annual dose induced by all samples are below 0.1 $mSva^{-1}$, and the evaluated annual doses show good correlation with activity indices. Compared with constructive building material, the decorative material induced much less dose due to less amount of buildup material was used.

INTRODUCTION

After the steel contaminated event, people in Taiwan concern the radiation level from building material recently. In a large scale investigation of indoor gamma dose rate for those house built in 1983 and 1984, those houses with indoor ambient dose rate more then three-fold of average indoor dose rate are due to the usage of materials containing higher natural radioactive nuclides. For example, some of them have radium painted dial, live beside the fertile storage or in most cases are using granite or tile as decorative building material.

In the ICRP report no.60, it recommends to take intervention for high natural radioactive condition. In Taiwan, more and more people question about how to make choice for lower level decorative material, although the AECROC declare that there is no obvious risk by using these higher natural radioactive material. Refer to the experience of other countries (1-7), granite samples had been well investigated due to geological survey. However, little report mentioned about the radioactivity in grazed tile. Unlike other countries that most people living in wooden house, typical house in Taiwan is build of reinforced concrete. And tile usage for wall or floor decoration is quite common for its advantage of easy cleaning and good looking. To understand the actual radioactivity contained and the dose induced by using tile, 60 tile and 6 glaze are sampled .

Method

Tile samples were collected from domestic products (47 samples) and imported ones (13 samples, imported from Italy, Spain and Malaysia). All tile samples were cut into 15cmW×15cmL for gamma spectroscopy using a high purity germanium detector. Glaze powder are meshed and canned in the 2 inch diameter sample holder. Specific activities of U-series and Th-series are determined by the calibrated Bi-214 (609 keV) and Tl-208 (583 keV) peaks respectively. Gamma yield of each peaks is referred to the newest NUDAT database.(8)

A computer package QAD-CG was used for gamma source flux and dose rate calculation.(9) The first step was to divide the source into lots of small cell and assume each cell as a point source. The dose rate at measurement point is calculated by kernal algorithm that is described as

$$D = \sum_j \sum_i Q_j \frac{S_{ij}}{4\pi R_j^2} \exp(-\sum_k (\mu_j t)_k) B_{jk}$$

where

j: energy index
I: source index

K: region index
Q: flux to dose rate conversion factor
S: gamma source strength
R: distance from source point to measurement point
B: buildup factor
μ: linear attenuation coefficient
t: sub-distance of R through region *k*

Number of energy peaks for QAD-series package is limited within thirty while the energy peaks for U-series or Th-series are more then three hundred. Those peaks within 100 keV are simplified into one effective energy and related intensity. The simplified results are shown in Table 1.

Geometry of the simulated typical living room in Taiwan is 4mW×4mL×3mH with wall thickness of 15cm. Three rectangles were used for combinatorial geometry. The outer material is cement and the inner one is air. A thin layer of 1.5 cm located at inner bottom is the decorative material as shown in figure 1. Decorative material is assumed to be only material paved on floor. Buildup factor is set to be concrete equivalent. An smaller room with size only 2mW×2mL×3mH is used to simulate the bathroom that is also shown in figure 1. Occupancy factor for living room plus bedroom is 5/6 while for bathroom, kitchen and balcony summing together is 1/6. Dose rate is calculated at the location one meter above ground .

Flux to dose conversion factor for indoor dose rate assessment is adapted from ANSI. To compare assessed results with measured one, another set for flux to ambient dose rate conversion are also alternative because survey meters used in this work are calibrated to ambient dose.

Table 1 Effective energy peaks for each 100 keV division.

Energy group keV	Th-series E_{eff} (keV)	Th-series Intensity(%)	U-series E_{eff} (keV)	U-series Intensity(%)	K-40 E_{eff} (keV)	K-40 Intensity(%)
0 - 100	48.44	78.11	36.10	105.66		
100 - 200	138.80	5.01	177.45	4.22		
200 - 300	240.54	58.59	279.78	28.05		
300 - 400	329.88	18.87	352.36	38.81		
400 - 500	448.00	7.51	466.45	1.83		
500 - 600	566.50	41.16	551.76	1.03		
600 - 700	653.91	0.87	611.47	48.05		
700 - 800	756.99	16.87	763.23	8.13		
800 - 900	852.83	8.20	819.35	2.22		
900 - 1000	936.50	48.45	936.94	3.74		
1000 - 1100	1068.36	1.38	1026.99	1.61		
1100 - 1200	1131.45	0.65	1123.91	17.33		
1200 - 1300	1253.59	0.85	1244.44	7.75		
1300 - 1400	1349.23	0.13	1376.00	5.00		
1400 - 1500	1475.18	1.82	1406.92	3.49	1461.00	10.67
1500 - 1600	1571.98	4.95	1538.07	3.89		
1600 - 1700	1631.82	4.14	1664.27	1.44		
1700 - 1800	1728.98	0.20	1758.91	18.35		
1800 - 1900	1842.90	0.34	1852.73	3.04		
1900 - 2000	1945.54	0.12	1938.17	0.06		
2000 - 2100			2050.49	0.20		
2100 - 2200			2120.39	1.29		
2200 - 2300			2209.71	5.43		
2300 - 2400			2339.12	0.04		
2400 - 2500			2447.77	1.58		
2500 - 2600			2505.40	0.01		
2600 - 2700	2614.53	35.70	2695.09	0.03		
2700 - 2800			2769.80	0.03		
2800 - 2900			2877.82	0.02		
2900 -			3002.24	0.06		

*$E_{eff}=\Sigma E_i \times I_i / \Sigma I_i$

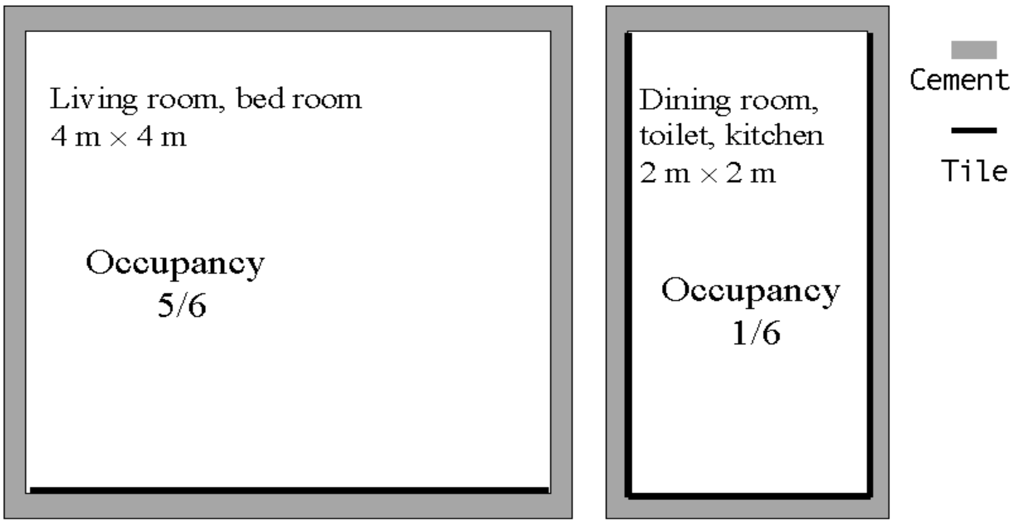

Figure 1 Models of specific room in typical house of Taiwan

Result and discussion

The ranges of specific activities of U-series and Th-series contained in tiles are shown in figure 2 and figure 3. Compare with the cases of other countries, the range of U-series activity is wider while that of Th-series is comparable. This implied the high radiation level may come from the higher U-series concentration.

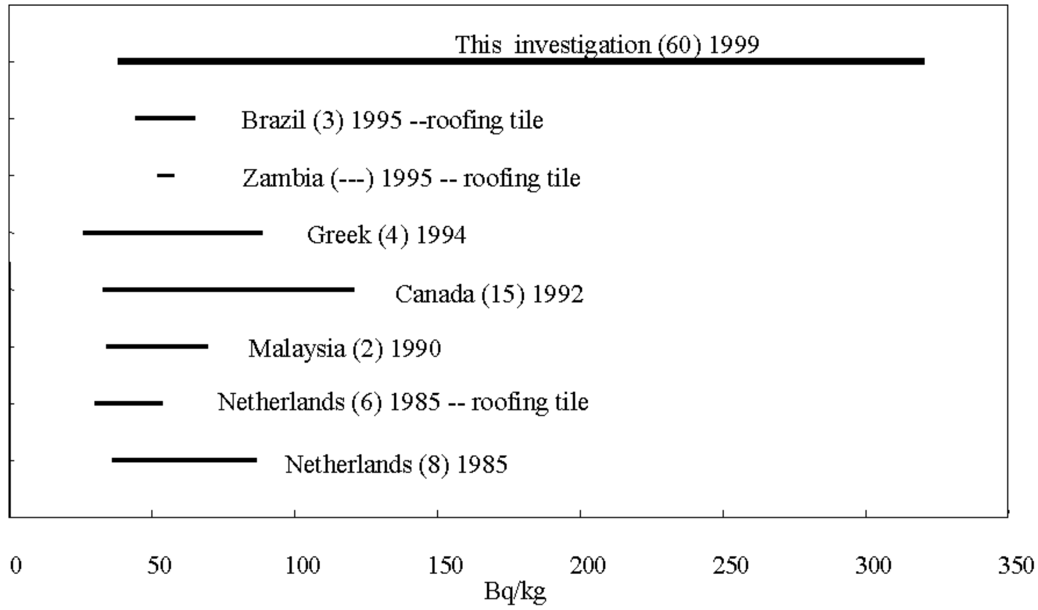

Figure 2 Range of specific activity of U-series contained in Tile. (sample number)

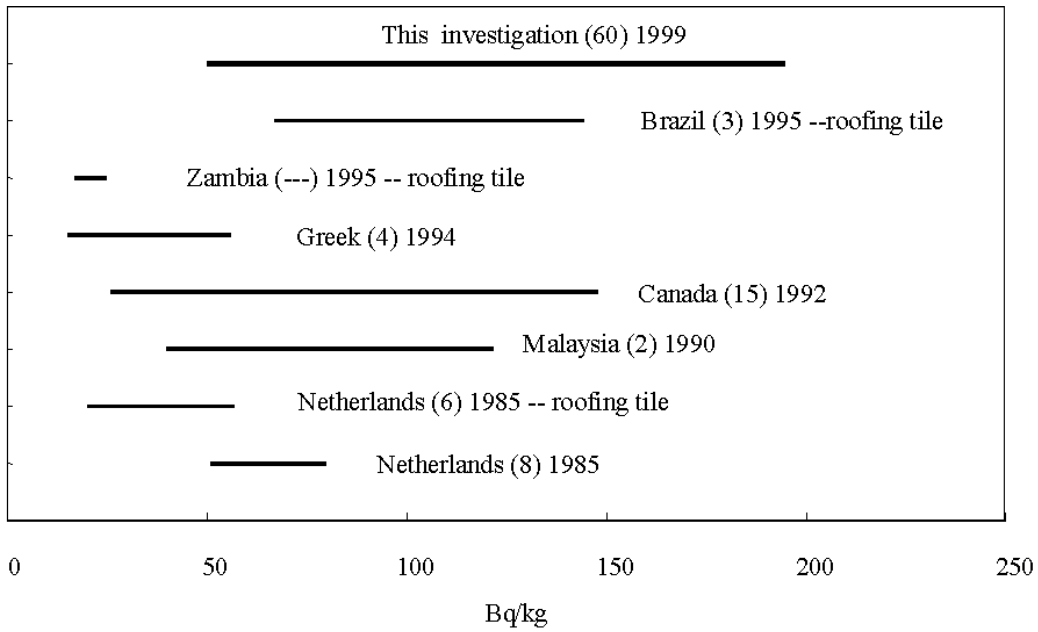

Figure 3 Range of specific activity of Th-series contained in Tile. (sample number)

The specific activities of U-series and Th-series of glaze and soil in Taiwan is list in table 2. The glaze samples show the trend that concentration of U-series is higher then Th-series. Obviously, the high radiation level of tile is due to the glaze on the surface. Zircon, $ZrSiO_4$ sand, is added to materials used to made the glaze.(10) Due to the La series shrine property, uranium is easier to penetrated and trapped in the lattice of zircon crystal.

Table 2 Specific activity of glaze and soil samples.

Unit : Bq kg^{-1}

Sample	K-40	Th-series	U-series
Glaze #1	<MDA	564	3125
Glaze #2	127	666	5079
Glaze #3	96	618	2869
Glaze #4	<MDA	733	3918
Glaze #5	558	103	215
Glaze #6	1202	185	241
Sand	266	30	14
Cray	382	33	22
Soil (yellow)	471	36	33
Soil (gray)	443	45	28
Soil (dark brown)	490	45	31
Soil (brown)	515	47	30
Soil (light brown)	520	54	35
Soil (dark red)	607	71	45

In a house, we measure the indoor dose rate at the center of the room is about 0.101 μGyh^{-1} while the calculated dose rate to be 0.094 μGyh^{-1}. The 7% lower is due to the measured result including radiation origin of cosmic rate and soil and the calculated one considering building material only.

Maximum dose rate increased for living room (bedroom) is at the center of the room because of the assumption that only floor is tile paved. The assessed maximum indoor dose rate increased by using these 60 samples are 0.002~0.006 μSvh^{-1}. Annual dose assessment are 0.012 ~ 0.045 $mSva^{-1}$.

For bathroom in Taiwan, tiles are used for all walls and floor. The maximum dose rate is about 3 to 5 cm to the wall surface. Although the real occupancy at the wall surface is much smaller then mentioned above, maximum dose rate is adapted for conservative assessment. The maximum dose rate increased by using these 60 samples are 0.009~0.033 μSvh^{-1}. Annual dose assessment are 0.013 ~ 0.048 $mSva^{-1}$ for less occupancy factor.

For the dose rate range increased by the tile paved on floor of upstairs room is within 0.0003 ~0.001 μSvh^{-1} . Annual dose assessment are 0.0023 ~ 0.0074 $mSva^{-1}$. In practical, such a small increasing could hardly be detected. It is suggested to neglect.

To reduce statistical variation, dose rate measurement near ground surface of floor building material is preferred but not one meter above ground. That will lead to overestimate annual dose if derived directly from surface dose rate. The average dose rate ratio between one meter and surface is about 0.27 for natural radioactive nuclide in these samples. That is, the annual dose derived from ground surface dose rate should multiply the correction factor of 0.27 to reasonable one.

The shielding effect of tile to dose rate from concrete about 1.2% for floor and 6% for walls pavement. For some sample containing little natural radioactivity, it shows the fact of using tile can reduce indoor dose rate. However, for conservative consideration, this effect is neglected.

Activity index had been define as (2) $AI = \frac{C_U}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810}$ ..And for building material, it suggest that AI should not exceed 1. In this work , 16 samples, 26.7% of total samples, are found to be AI over 1. And all of them are made in Taiwan. This is more then other countries' experience. The maximum one of these samples is AI=1.55.

The AI distribution of different purpose tile is shown in figure 4. The tiles for floor have higher AI in general but with widely spread distributing. Most tile for wall with AI smaller then 1 due to different glaze recipe for different purpose.

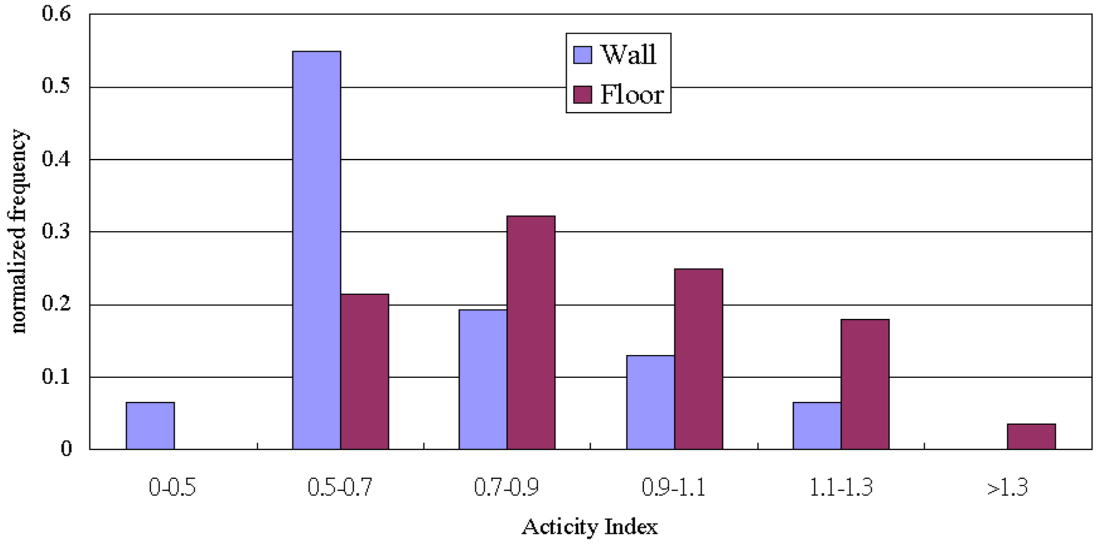

Figure 4 AI distribution of tiles for different purpose.

The activity index can be related to dose by regression analysis as shown in figure 5. The correlation coefficient for living room and bathroom are R^2= 0.989 and R^2= 0.987 respectively. The strong relationship imply that the dose can be assessed by AI multiply a conversion factor. That will be more easy and convenient then running complex QAD code. Because the amount of decorative material is much smaller the that of constructive building material, like concrete or brick, the dose induce by those samples with AI over 1 are not serious.

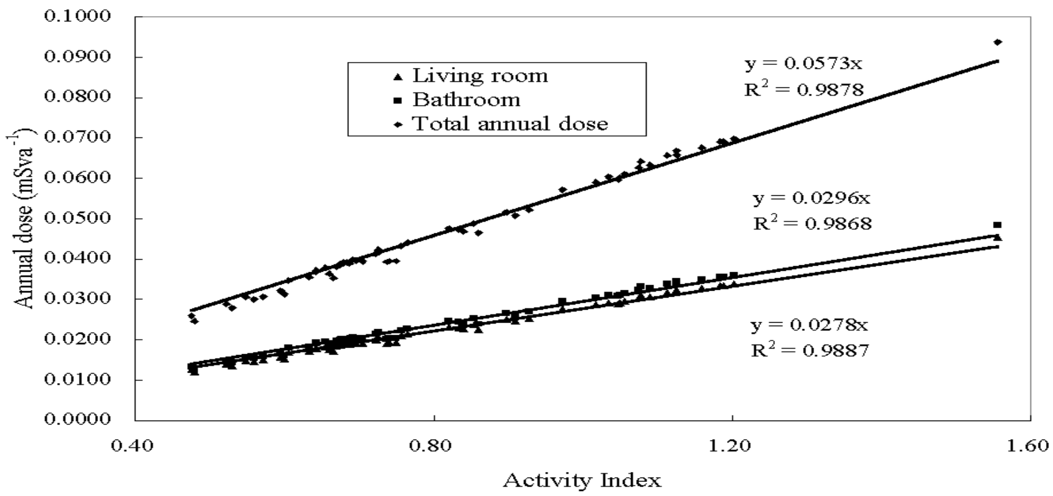

Figure 5 Correlation analysis between annual dose and activity index.

Conclusion

Due to the results obtained, we conclude that ,

1. The higher natural radioactivity contained in tile is due to the glaze that used on the tile surface. If any intervention should take, we suggest paying more attention on zircon related process.
2. The annual doses induced by the 60 samples in this work are all within 0.05 $mSva^{-1}$. Compared to the average indoor gamma dose in Taiwan (11) 0.57 $mSva^{-1}$, that is only within 10% of general background. We suggest not to take any action on such radiation level.
3. Dose induced by the tiles used on upstairs or neighbor's floor could be neglected.
4. There is strong correlation between assessed dose and activity index. Activity index is much easier and more convenient to calculation. The annual dose can be derived by multiplying a conversion factor 0.057

mSva^{-1} to AI.
5. Surface dose will lead to overestimation of annual dose. A correction factor of 0.27 should be applied.
6. The average AI of tiles for floor is higher then those for wall because different recipe for different purpose.
7. For some tiles containing little radioactivity will reduce indoor dose rate because of shielding effect. But most tile is not thick enough to make such reduction obvious.

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