IEEE NPSS (Toronto), UOIT, Oshawa, ON, Canada, 25 & 26 June, 2010
International Workshop on Real Time Measurement, Instrumentation & Control [RTMIC]

Rapid, Efficient, Reliable and Cost Effective Method to Assess Radiation Safety of Granite Tiles Materials with Specified Dimensions Used in Building Construction

Sadek Zeghib [a] and A. Sh. Aydarous [a,b]

[a] Physics Department, Faculty of Science, Taif University, Al-Hawiah, Taif, P.O. Box 888, Kingdom of Saudi Arabia

[b] Faculty of Energy Systems and Nuclear Science, University of Ontario Institute of Technology, 2000 Simcoe St North , Oshawa, ON, Canada, L1H 7K4
E-mail: Abdulkadir.Al-Aydarous@uoit.ca

Abstract

In assessing the radiation safety of granite tiles for use in building construction in accordance with UNSCEAR(1) & EC(2) criteria, HPGe measurements for 9 (nine) granite samples were carried out to calibrate a portable scintillation gamma radiameter (PGR). The HPGe measurements of the activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K were used to deduce the absorbed dose rate that emanated from granite tiles having specified dimensions of 60cm x 30cm x 2cm. The correlation between the two measurements was surprisingly very high. In particular, the dose rate response shows a linear dependence with a correlation coefficient exceeding 98%. The validity of the method has been successfully tested on 6 (six) granite samples spanning the 50-200 nGyh^{-1} dose rate range. The expected dose rates for the type (material) of granite tiles extracted using the "calibration" curves was compared to the actual HPGe dose measurements. The results show an accuracy ranging from 1% for relatively elevated radioactivity to 16% for low radioactivity. Since our purpose is to look for suspicious material of relatively elevated radioactivity, the method seems effective, reliable, and efficient.

Keywords: Granite, radioactivity concentration, radium equivalent, dose rate, annual effective dose.

1. Introduction

All building materials including granite contain various amounts of natural radioactive nuclides. Materials derived from rock and soil contain mainly natural radionuclides of the uranium (^{238}U) and thorium (^{232}Th) series, as well as potassium (^{40}K). Granites, in particular, exhibit an enhanced elemental concentration of these natural radionuclides in comparison to the very low abundance of these elements observed in the mantle and the crust of the Earth. The igneous rocks of granitic composition are strongly enriched in U and Th (on an average 5ppm of U and 15ppm of Th), compared to rocks of basaltic or ultramafic composition (<1 ppm of U) (3-6). In the uranium series, the decay chain segment starting from radium (^{226}Ra) is radiologically the most important element and therefore reference is often made to radium instead of uranium. The world-wide average concentrations of radium, thorium and potassium in the earth's crust are about 50 Bq kg^{-1}, 50 Bq kg^{-1} and 500 Bq kg^{-1}, respectively (7-8).

Over the last decade, Saudi Arabia has increasingly become a large market for local and foreign marble and granite usage. New practices have been introduced in house construction that include the usage of granite(9) in different fashions and different amounts. It was observed that granite tiles of 2cm to 5cm thickness are widely used as covering and building materials for counters, cashier desks, shelves, benches, tables, and counter-tops. The concern is the external radiation dose from the granite tiles available in the local market. Most of these samples contain Radium-226 (^{226}Ra), Thorium-232 (^{232}Th) and Potassium-40 (^{40}K) in different amounts. Even though these radionuclides in building materials are natural, it is important to assess their radiological risks (external exposure) to the population when they are used for the construction of houses.

In order to measure the radioactivity concentration and estimate the radiation hazards from building materials, gamma spectroscopy is usually applied using different detectors such as HPGe detectors and scintillation detectors. Although these techniques are widely recognized in measuring activity concentrations and provide the required accuracy, they are time consuming, tedious, and costly especially when the number of samples is high. On the other hand, portable dose rate meter survey instruments such as PGR scintillation radiameter are widely used for area monitoring in hospitals, industry, and environment. They may sometimes be the only source of information available for a preliminary assessment of radiation hazard. Because of their simple operation, they can normally be used by non-specialized personnel. However, the readings from these instruments can significantly underestimate the dose rate because they either use large area detectors such as scintillators or large volume detectors such as air ionization chambers.

This study describes the use of HPGe gamma spectroscopy detector and a portable scintillation gamma radiameter (PGR scintillation radiameter) for evaluating the dose rate from granite tiles materials. The study aims at providing a correlation between the estimated dose rate measured by the PGR portable survey meter and the other quantities such as radium equivalent, dose rate, and effective dose rate measured by the HPGe detector. If a correlation between the two systems is established, the task of measuring the dose rate and hazard indices quantities could considerably be alleviated. This will enable rapid, efficient, reliable, and cost effective measurements to be made to assess radiation safety of granite tiles materials used in building construction.

2. Experimental methods

2.1 Gamma spectroscopy measurements with HPGe

The specific activities (in Bq kg^{-1}) of ^{226}Ra, ^{232}Th and ^{40}K in some (ten) granite samples of wide use locally have been determined in our previous work (10) by gamma spectroscopy using Canberra Reverse electrode ~50% HPGe detector. In order to estimate their radiological effects, the total absorbed dose rate (D), the effective dose rate (E), the radium equivalent (Ra_{eq}), the external hazard index (H_{ex}), and internal hazard index (H_{in}) have been estimated (10) in order to compare them with the criteria set by the European Union recommendation No 112 (Radiological protection principles concerning the natural radioactivity of building materials)(2).

2.2 Dose rate estimation with PGR for granite tile materials (60cm x 30cm x 2cm)

PGR scintillation radiameter is a multi-purpose survey meter and it can be used for geology, mining, environmental monitoring, and industrial and health sectors. The scintillation crystal is 2x2˝ NaI(Tl). It operates under a wide range of gamma energies (0.5-2.5 MeV). The data can be stored and transferred to a PC for further analysis.
Dose rate measurements have been performed using PGR scintillation radiameter. The following steps and requirements have been applied:

1- The background radiation was measured in an appropriate flat area far from both walls and any scattering objects (to avoid scattered radiation from surrounding objects). Five measurements of 120s each were taken and averaged.
2- The dose measurements were then taken at the same place where the background had been measured. The PGR instrument was placed above the granite tile with its sensitive area approximately on the center of the tile. The dose rate measurements were obtained by taking 3 readings of 120s each. These measurements were taken for granite tiles having the same dimensions of (60cm x 30cm x 2cm).
3- Background correction was made by subtracting the average background reading obtained in step 1 from the actual reading obtained in step 2.

3. Results and Discussion

3.1 Correlation between PGR and HPGe measurements

This section describes the use of granite tile materials to provide a correlation between dose quantities estimated by HPGe detector and dose rate measurements obtained by the PGR scintillation radiameter. The PGR survey instrument's responses versus the actual dose rate, effective dose rate, radium equivalent, and hazard indices measured by the HPGe detector are presented in Figure 1 to Figure 4. The correlation between the two measurements is reasonably consistent and shows good linear dependence with a correlation coefficient exceeding 98%. By using these curves, the expected HPGe measured absorbed dose rate of any granite tile material with the same specified dimensions can be estimated without performing detailed measurements using HPGe gamma spectroscopy detector. The latter can be extracted from the so called "dose rate calibration curve" of Figure 1, by just performing PGR measurements in 5 minutes only (instead of 15 hours) with an acceptable degree of reliability and accuracy regarding our main goal which is to estimate the radiation safety of granite tile materials used in building construction. The reliability and accuracy of the method has been verified by performing PGR and HPGe measurements (using the same pair of instruments) on six test granite tile materials.

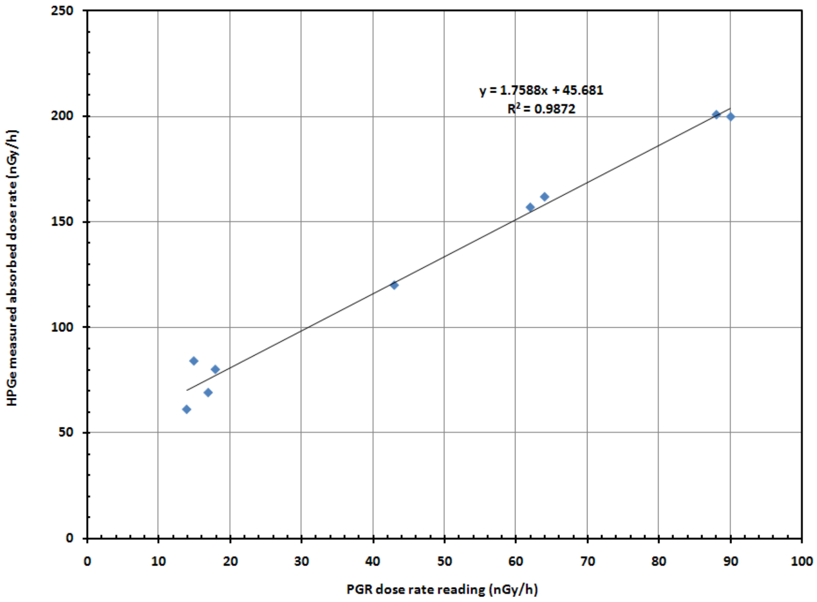

Figure 1: HPGe measured absorbed dose rate versus PGR dose rate.

Also similar sort of "direct calibration" curves have been drawn for the other four quantities (effective dose rate, radium equivalent, Hex and Hin) directly as a function of the PGR dose rate reading **(** Figure 2 to Figure 4**),** drawn from the nine calibrating granites . By using these curves, the expected four quantities of any granite tile material with the same specified dimensions can be estimated directly.

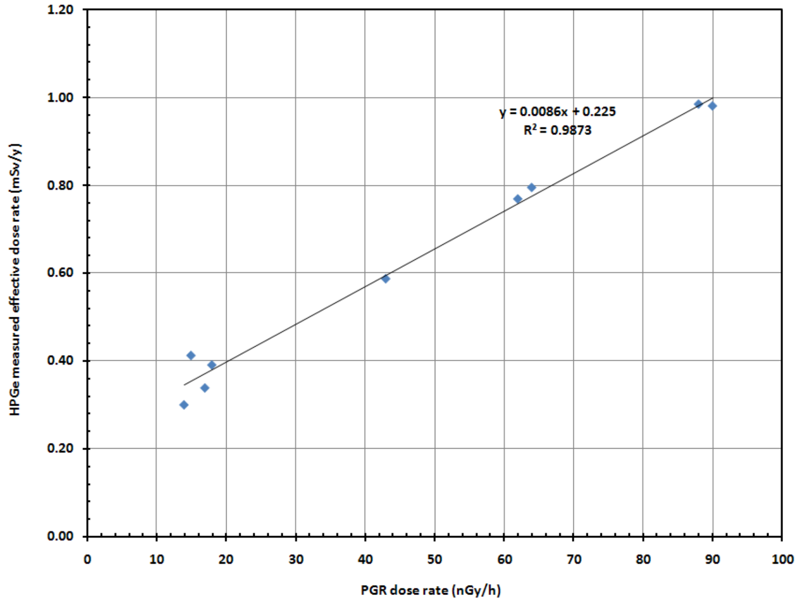

Figure 2: HPGe measured effective dose (E.D.) rate versus PGR dose rate.

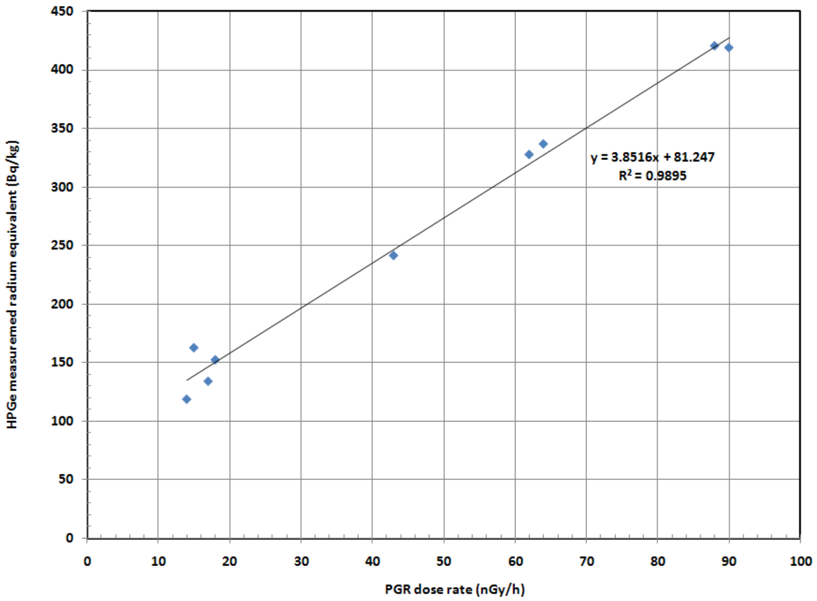

Figure 3: HPGe measured Radium equivalent (Ra_{eq}) versus PGR dose rate.

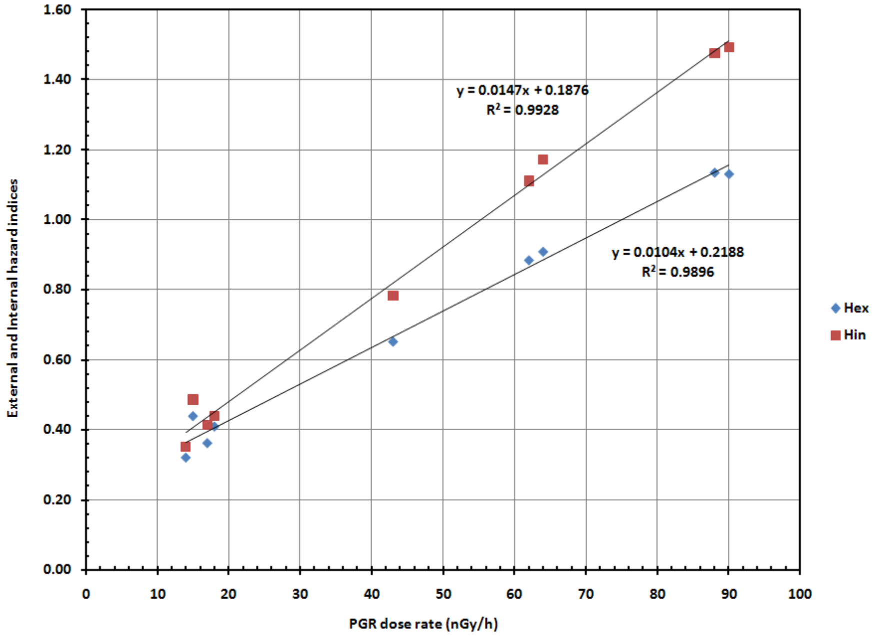

Figure 4: HPGe estimated external and internal hazard indices versus PGR dose rate

Table 1 presents PGR dose rate measurements for the six test sample materials as well as the expected HPGe absorbed dose rate (extracted from the calibration curve of Figure 1), and also compared with their actual HPGe absorbed dose rate measurements for comparison. The results show an accuracy ranging from 1% for relatively elevated radioactivity (sample: Balmoral Italy) to 16% for low radioactivity (sample: Malaga China). Since the purpose is to look for suspicious material of relatively elevated radioactivity the method seems promising, reliable, and efficient.

Table 2, Table 3, Table 4 and Table 5 present the expected annual effective doses, radium equivalent, H_{ex} and H_{in} (extracted directly) for the six test samples materials as well as their corresponding actual HPGe measurements for comparison including the deviation (accuracy). The results show an accuracy ranging from 0% to 16% for all quantities overall. Some remarks worth mentioning are given below:

1- HPGe activity concentration measurements and PGR dose rate measurements were performed for the same type of granite material.
2- Apart from one sample (Rosa Beta), all the investigated samples (tiles) are considered to be homogenous as long as they are from the same brand. Therefore, PGR instrument should produce the same results if measurements have to be done with other samples of the same brand.
3- Rosa beta granite tile which showed a clear non homogeneity in radioactivity distribution has been excluded from the dose rate calibration curve. It was clear from its appearance that the distribution of black grains was non-homogenous all over the tile, which leads to different activities and was confirmed by PGR readings.
4- We relied on materials measured by our instruments (~50% HPGe and PGR), so the "calibration curves" are only appropriate to these instruments. Nevertheless, the method (not the curves) could be applied to any other appropriate pair of instruments (lab instrument and portable).
5- It is very important to mention that the calibration curves mentioned in this work apply only to granite tiles having the specific dimensions (60cm x 30cm x 2cm). This size is widely used as covering material and is sold the most in the markets of Saudi Arabia. However, for a more complete work, this method will be expanded to other appropriate tile dimensions such as (60cm x 30cm x 3cm) and (60cm x 30cm x 5cm) having larger thickness, which are available in the market to a lesser extent. This would lead to different calibration curves (mainly different slopes) but will lead to the same conclusion about the granite material radiation safety in hand.

Table 1: Comparison of measured and expected HPGe dose rate values for the six test granite samples using measurements from PGR instrument.

Sample Name	PGR Dose rate (nGy/h)	Measured HPGe absorbed dose rate (nGy/h)	Expected HPGe absorbed dose rate (nGy/h)	Deviation (%)
Balmoral Italy	91	205.7	204	0.8
Saudi Pink	18	77.3	76.2	1.4
Gandola China	25	89.7	88.3	1.6
Malagua China	6	56.2	48.4	16
Rosa Porrino (Italy)	28	94.9	87.5	8.4
Golden leaf (Saudi)	21	82.6	81.2	1.7

Table 2: Comparison of measured and extracted HPGe effective dose rates for the six test granite samples using measurements from PGR instrument.

Sample Name	PGR Dose rate ($nGyh^{-1}$)	Measured HPGe effective dose rate ($mSv.y^{-1}$)	Expected HPGe effective dose rate ($mSv.y^{-1}$)	Deviation (%)
Balmoral Italy	**91**	**1**	**1.01**	**1**
Saudi Pink	**18**	**0.37**	**0.38**	**2.7**
Gandola China	**25**	**0.43**	**0.44**	**2.3**
Malagua China	**6**	**0.24**	**0.27**	**13**
Rosa Porrino (Italy)	**28**	**0.43**	**0.46**	**7.0**
Golden leaf (Saudi)	**21**	**0.4**	**0.40**	**0**

Table 3: Comparison of measured and extracted HPGe radium equivalent (Ra_{eq}) values for the six test granite samples using measurements from PGR instrument.

Sample Name	PGR Dose rate ($nGyh^{-1}$)	Measured HPGe Ra_{eq} ($Bq.kg^{-1}$)	Expected HPGe Ra_{eq} ($Bq.kg^{-1}$)	Deviation (%)
Balmoral Italy	**91**	**423.7**	**431.9**	**1.9**
Saudi Pink	**18**	**147.00**	**150.0**	**2.0**
Gandola China	**25**	**176.9**	**177.1**	**0.1**
Malagua China	**6**	**93.4**	**103.7**	**11**
Rosa Porrino (Italy)	**28**	**174.3**	**188.7**	**8.3**
Golden leaf (Saudi)	**21**	**158.4**	**161.6**	**2.0**

Table 4: Comparison of measured and extracted HPGe external hazard index (H_{ex}) values for the six test granite samples using measurements from PGR instrument.

Sample Name	PGR Dose rate ($nGyh^{-1}$)	Measured HPGe H_{ex}	Expected HPGe H_{ex}	Deviation (%)
Balmoral Italy	**91**	**1.14**	**1.17**	**2.6**
Saudi Pink	**18**	**0.40**	**0.40**	**0**
Gandola China	**25**	**0.48**	**0.48**	**0**
Malagua China	**6**	**0.25**	**0.28**	**12**
Rosa Porrino (Italy)	**28**	**0.47**	**0.51**	**8.6**
Golden leaf (Saudi)	**21**	**0.43**	**0.44**	**2.3**

Table 5: Comparison of measured and extracted HPGe internal hazard index (H_{in}) values for the six test granite samples using measurements from PGR instrument.

Sample Name	PGR Dose rate ($nGyh^{-1}$)	Measured HPGe H_{in}	Expected HPGe H_{in}	Deviation (%)
Balmoral Italy	**91**	**1.4**	**1.52**	**8.6**
Saudi Pink	**18**	**0.45**	**0.45**	**0**
Gandola China	**25**	**0.56**	**0.56**	**0**
Malagua China	**6**	**0.28**	**0.28**	**0**
Rosa Porrino (Italy)	**28**	**0.56**	**0.60**	**7.1**
Golden leaf (Saudi)	**21**	**0.49**	**0.50**	**2.0**

It is important to stress upon the word "material" of the granite tile because all estimated quantities concern the material of the tile and not the tile itself. One has to be aware that the aim of this method is not to assess the dose rate from the granite tile itself, but from the material composing it through applying an appropriate calibration factor that correlates HPGe and PGR measurements. It is also worth noting that the produced calibration curves are so far applicable to the dose rate range estimated by the two detection systems for the nine granite calibrating samples.

As mentioned previously, granite tiles are becoming widely used in the construction of buildings in various ways (coverings, tiles, desks, shelves, benches, counter-tops, cabinets, etc.) and in various amounts and in various geometries. Because of these varieties, this study stressed upon granite material as one of the many types of building material regardless of its use. If one would like to assess accurately the external exposure of these materials, all previously cited factors (mainly quantity, geometry, location, and time factors) have to be taken into account.

Other advantages of this method lie on the fact that the problem of non-homogeneity is to overcome by a simple rapid scan of tiles with PGR instrument. Finally, when a sample shows relatively high dose rate using the rapid PGR measurement, it is recommended that this sample should be subject to precise HPGe measurements, if further clarification is needed.

4. Conclusion

A rapid, efficient, reliable, and cost effective method has been investigated to assess radiation safety of granite tiles materials used in building construction. This method is based on comparing dose rate measurements from HPGe gamma spectroscopy system with a conventional counting system (PGR). Using the calibration curves the expected dose rates, effective dose rate, radium equivalent, and external and internal hazard indices for each test granite tile materials have been compared successfully with the actual HPGe measurements. Using the PGR instrument, the dose rate measurements and subsequent expected quantities for each granite material sample, are obtained in only ~15 min instead of ~15 hours of hard work using complete gamma spectroscopy measurements besides sample preparation and system calibration. The results showed accuracy on the deduced dose rate ranging from 1% for relatively elevated radioactivity to 16 % for low radioactivity. Since the purpose is to look for suspicious material of relatively elevated radioactivity, the method seems promising, reliable, and efficient. The range of absorbed dose rates available from the study in our work covers from 50 to 200 n Gyh^{-1}

In order for this work to be more complete, the calibration curves will be extended to other tiles thicknesses present in the market especially those with 3cm and 5cm thicknesses. It is also worth expanding the dose rate range to cover granite tiles materials having higher dose rates if they are available in the market, and not just relying on extrapolation process of the calibration curves.

5. References

1. UNSCEAR. *United Nation Scientific Committee on Effects of Atomic Radiations, pp 140, Annex B.* (2000).
2. EC. Nuclear Safety and Civil Protection. European Commission (EC). *Radiological Protection Principles Concerning the Natural Radioactivity of Building Materials*. Radiation Protection 112. (1999).
3. Faure, G. *Principles of Isotope Geology* (John Wiley & Sons, 1986).
4. Me`nager, M. T., Heath, M. J., Ivanovich, M., Montjotin, C., Barillon, C. R., Camp, J. & Hasler, S. E. *Migration of uranium from uranium-mineralised fractures into the rock matrix in granite: implications for radionuclide transport around a radioactive waste repository.* In Fourth International Conference of Chemistry and Migration Behaviour of Actinides and Fission Products in the Geosphere 47–83 (Charleston, USA, 1993) (47–83)
5. Pavlidou, S., Koroneos, A., Papastefanou, C., Christofides, G., Stoulos, S. & Vavelides, M. *Natural radioactivity of granites used as building materials*. Journal of Environmental Radioactivity, **89** (1) 48-60 (2006).
6. Tzortzis, M., Tsertos, H., Christofides, S. & Christodoulides, G. *Gamma radiation measurements and dose rates in commercially-used natural tiling rocks (granites)*. Journal of Environmental Radioactivity, **70** (3) 223-235 (2003).
7. UNSCEAR. United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR). *Exposures from Natural Sources of Radiation*. Annex A, A/Ac., 82/R. United Nations, New York, (1993).
8. UNSCEAR. United Nations Scientific Committee on the Effects of Atomic Radiation. *Sources, Effects and Risks of Ionizing Radiation*. United Nations, New York, (2000).
9. Al-Saleh, F. S. & Al-Berzan, B. *Measurements of natural radioactivity in some kinds of marble and granite used in Riyadh region*. Journal of Nuclear and Radiation Physics,, **2** (1) 25-36 (2007).
10. Aydarous, S. A., Zeghib, S. & Al-Dughmah, M. *Measurements of natural radioactivity and resulting radiation doses from commercial granites*. Radiation Protection Dosimetry, **Submitted** (2010).