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Estimating granite roughness using systematic random sampling for the evaluation of radon gas emanation from ornamental granite rocks

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There are three natural radioactive families according to their decay, which are: the uranium series (^{238}U decreasing to stable ^{206}Pb), the actinium series (^{235}U decreasing to stable ^{207}Pb) and the thorium series ($^{236}\text{U} \rightarrow {}^{232}$Th decreasing to stable ^{208}Pb). The three series all have radon gas as an intermediary element, but each with a different atomic mass (^{222}Rn, ^{219}Rn and ^{220}Rn). The three isotopes are inert gases at ambient conditions and all are alpha particles emitters. Soils naturally emanate these radioactive gases in variable concentrations depending on composition and location. The radon radioactive emanation is a mass flow composed of radionuclides emitted to the atmosphere from the surface of the material, or transported to it. Emanation depends on the amount of radon atoms formed from the decay of radium and on the surface roughness of the material. Treatment such as polishing can be used to decrease radon gas emanation by closing open surface pores and reducing the specific surface area. This study aims at evaluating granite roughness of experimental plates of ornamental rocks using a systematic random sampling approach in order to minimise analysis time. To validate the systematic minimum area sampling results these were compared to measurements made over the whole reference area. It is concluded that measurements can be conducted in just a few locations using systematic random sampling, significantly reducing the time for obtaining estimates of the granite's roughness by factors 150–200.

Introduction

This study addresses development of a fast method to obtain a granite plate's roughness using systematic random sampling as a tool to minimize the measurement time without quality loss. Granite roughness is an important parameter for a correct evaluation of radon gas emanation. Estimating granite roughness is part of test regimens for characterizing gas emanation. This test is by far the slowest of all characterisation tests. It takes 16 days to obtain relevant data from a 20 × 20 cm plate. This motivated the authors to use a sampling method to reduce the time needed for estimating granite roughness.

Among the available surface treatments aiming to decrease radon gas emanation, there is granite polishing which is a cheap and efficient method since it reduces the specific area and closes open surface pores. Figure 1 shows the difference of emanation between polished and rough surfaces measured by a radon meter, which has a scintillation cell as operating principle.

The EU (European Union) published a Council Directive in December of 2013 (2013/59 EURATOM) that compels all member states to present a national action plan to address long-term risks from radon exposures by February of 2018. Guidance on methods and tools for measurements, identification of building materials with significant radon emanation are on the list of items to be considered in preparing this action plan. In view of this EU document, the authors feel that methodological studies in this field are well motivated.

Radon emanation

There are three radon isotopes (^{222}Rn, ^{219}Rn and ^{220}Rn) which are alpha particles emitters, all of which are inert gases at ambient conditions. The radioactive emanation is a mass flow composed of radionuclides transported to the atmosphere from the material, depending on the amount of radon atoms formed from decay from radium and the physical characteristics e.g. surface roughness[1]. The amount of gas that reaches the surface is directly proportional to the specific area of the material.

Inhalation of radon gas and its decay products is a health risk to humans. Alpha particles from the radioactive decay may reach lung tissue and cause damage that can lead to lung cancer. Most of radon gas exits the human body by exhalation before the decay process however, so most of the radioactive dose comes from the decay products that are inhaled as dust and become lodged in the lung tissue. These radionuclides decay quickly which results in further damage of the lung tissue[2].

Surface metrology – Interferometry

Surface metrology is a branch of engineering related to measurement of roughness, sharpness, waviness and other surface parameters, which are dependent on a given engineering application. Methods available to characterize surface texture can be classified as contacting and non-contacting[3]. While contacting methods demand physical contact to assess the surface topography, non-contacting methods, as the name implies, do not require any such. Surface interferometry is a non-contacting technique, based on a superposition principle. Two waves with no phase shift, identical w.r.t. amplitude and frequency, when combined, will result in a wave with the same frequency but the amplitude will be doubled. This effect is known as constructive interference. Two waves with a phase shift of 180° will result in a wave with zero amplitude. This effect is known as destructive interference. The interaction between different waves in general results in patterns, known as fringes, showing constructive and destructive interference. Figure 2 shows

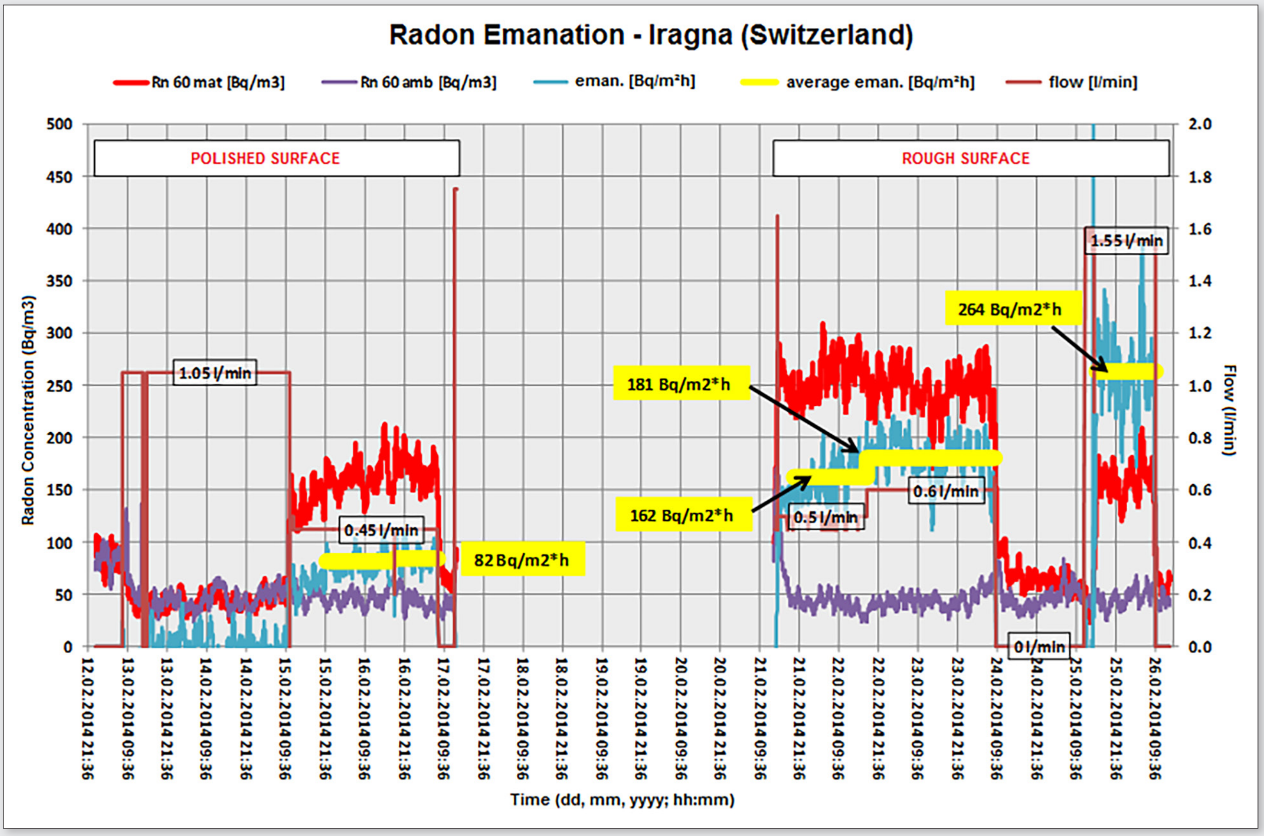

Figure 1. Difference of radon gas emanation from polished (left) and rough (right) surfaces as measured by a radon meter (based on a scintillation cell).

the reference and test beams combined, resulting in constructive interference[4].

An interferometer emits a single light beam which is split into two by a beam splitter. The two beams are destined to interact with each other resulting in the mentioned fringe pattern *after* interaction with the surface whose texture is to be characterised. One light beam is directed to the sample surface, the test beam. The other beam will be directed to a reference mirror. The two beams are reflected and reach the detector. The device then analyses the coherence of the resulting signal, which is a measure of the correlation of two beams in the resulting wave, separated by a given delay. The device finds the proper height at which the coherence value for each pixel reaches a maximum and the resulting fringe pattern is used to calculate the surface height. Figure 2 also shows the interferometer scanning process, which finds the surface height by analysing the fringe patterns[4].

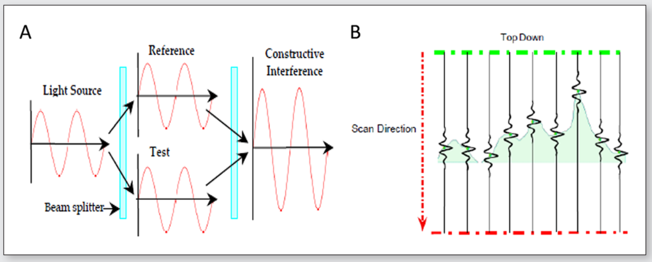

Figure 2[3]. (A) Superposition principle on an interferometer showing the combination of reference and test beams, resulting in constructive interference[4] (B) Interferometer scanning process, which finds the surface height by analysing the fringe patterns.

Systematic Random Sampling (SRS)

This sampling method is probabilistic and involves a regular, pre-established pattern for selecting sampling locations. The sampled area was divided into sectors, strata and substrata, where[5]:

- Sector: is a fraction of the total area, whose size could e.g. be based on recommendation from statisticians. Or, as in this study, based on reverse calculation to define this size, using the attainable substrata (explained bellow) size.
- Stratum: each sector is submitted to the preselected sampling mode (random systematic). This is performed by dividing each sector into a systematic grid.
- Substratum: each stratum must be divided into a certain number of basic units called substrata. It is suggested that division of each stratum in a number of substrata must be at least 20 times the number of measurements that should be collected within one sector.

The location of the initial sampling point is randomly selected in one preselected stratum of an area which is divided into smaller equal size substrata. In general, data acquisition and data analysis for each substratum should be managed independently. Figure 3 illustrates this kind of systematic sampling.

Methodology

The test sample was a granite from the city of Iragna, Switzerland. The material is commonly commercialised in plates, which have one polished surface and one natural surface (this is the 'rough surface'

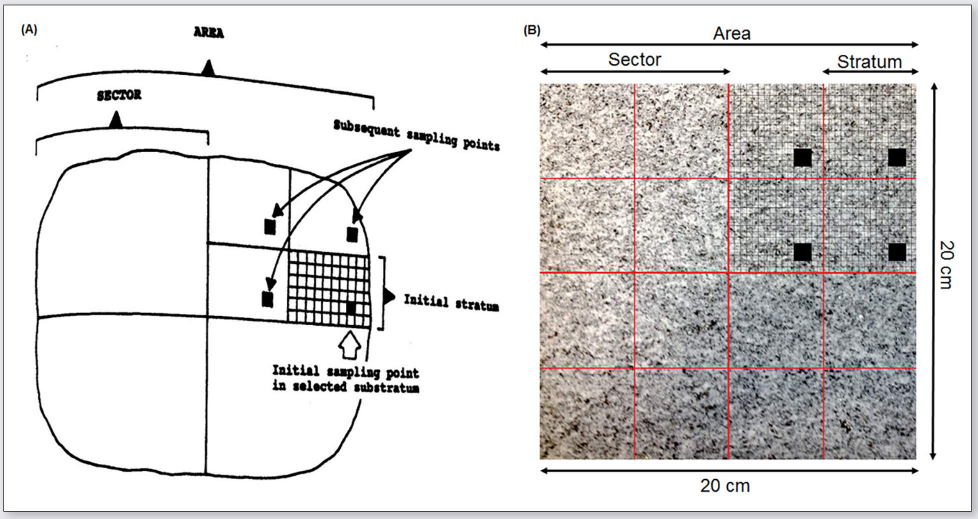

Figure 3. (A) Diagram showing initial selection of one random substratum in the initial stratum of one sector (SRS)[5] (B) Grid used to guide roughness assessment of Iragna's granite plate, the location of the initial sampling point is randomly selected in one preselected strata of an area which is divided into smaller equal size sectors, called substrata.

in this study). Both sides were tested to evaluate the accuracy of the data obtained by SRS. The whole area measurement was taken as reference. In this case, the sectors were set as a systematic grid following an adapted interpretation of the Theory of Sampling for contaminated areas[5].

In order to achieve good measurements we setup our study to fulfil these three pre-requirements[5]:

- Each sector must have a surface smaller than the local limit. In this study, the local limit is one 20 × 20 cm granite plate.
- The number of strata per sector must be at least four.
- Each sector must be characterized by their average and variances.

Figure 3 shows the grid used to stratify the granite plate surface (20 × 20 cm). The original sample was cut in 16 smaller pieces (5 × 5 cm) so that each stratum would fit in the object holder for the optical interferometer, used for the surface measurements from which 3D surface parameters were obtained (Figure 4).

Three strata from the 16 were randomly chosen to be tested (sample 1, 2, 3) both rough and polished surfaces, six tests total. The estimated time for measuring a 5 × 5 cm surface is one day using the interferometer (16 days to measure the whole 20 × 20 cm granite plate). This is considered too long and consequently too resource demanding for obtaining this crucial parameter to evaluate emanation correctly, especially since the interferometer is rented by the hour. Furthermore, estimating roughness is only one of many tests (e.g. porosimetry, permeability, X-ray diffraction) that should be performed in order to characterise the sample fully before the radon emanation measurement. It is currently the slowest test and it delays the final result of the radon emanation analysis seriously.

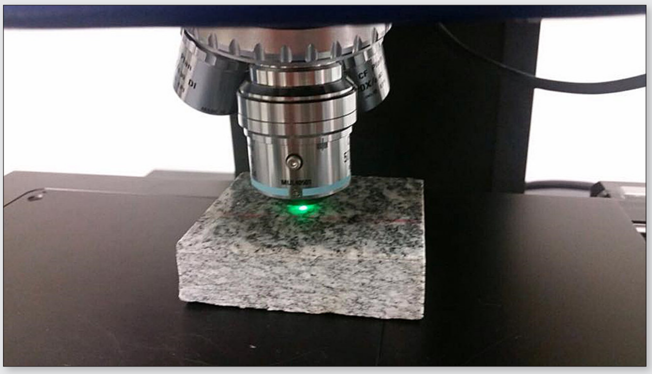

Figure 4. Interferometer in active measuring, also showing object holder.

The statistical analysis (Wilcoxon Signed Rank Test) of the results was conducted using SPSS for Windows statistical software (version 22).

Results and discussion

The three samples analysed were obtained by randomly choosing three strata of the granite plate. The area of each sample was analysed completely (whole area) and was subsequently partitioned to apply Systematic Random Sampling on a much smaller area to assess the same roughness manifestation. Figure 5 illustrates the SRS applied for one of the 5 × 5 cm plates (sample 1). The schematic drawing covers only a half plate but measurements were conducted in full, thus totalling 16 substrata on each surface.

[Figure 5]

The output parameters of the interferometer software are:

- Height parameters (ISO 25178): Sq (root mean square height - µm); Ssk (skewness) and Sku (kurtosis).
- Volume functional parameters (ISO 25178): Vm (material volume – µm^3/µm^2); Vv (void volume – µm^3/µm^2); Vmp (peak material volume - µm^3/µm^2); Vmc (core material volume – µm^3/µm^2); Vvc (core void volume – µm^3/µm^2); Vvv (pit void volume – µm^3/µm^2).
- Functional parameters (ISO 25178): Smr (area material ratio - %); Smc (inverse area material ratio - µm).
- Hybrid parameters (EUR 15178N): Sdr (developed interfacial area ratio - %).

It is difficult to select a most suitable parameter for roughness characterisation and it is therefore common to use a combination of two or three, dependent on the material type.

Tables 1 and 2 show the results from a relative error analysis between the SRS data and data based on the whole area (reference).

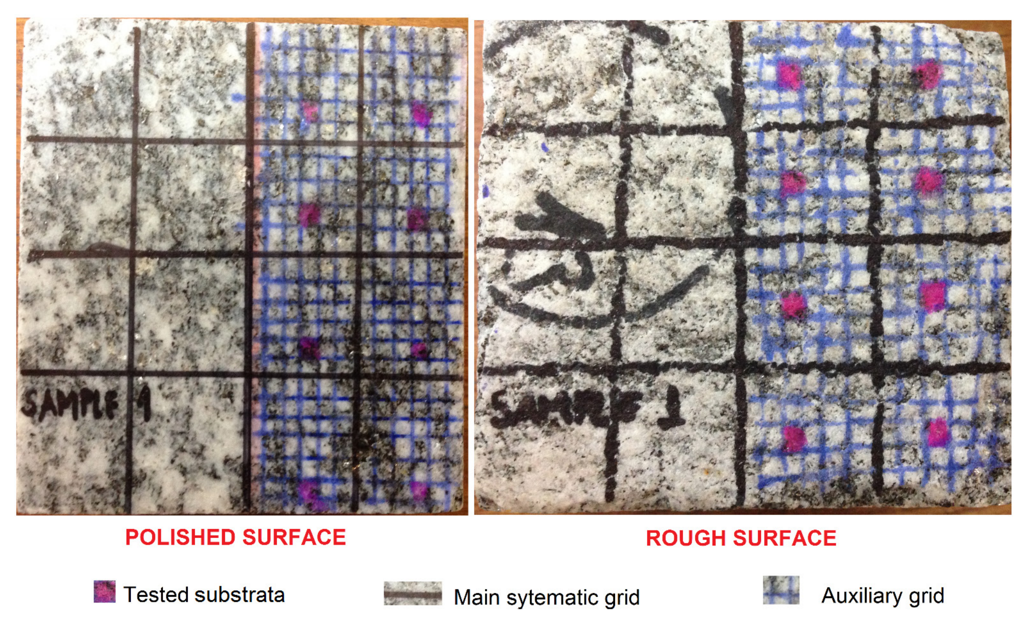

Figure 5. SRS applied to 5 × 5 cm plates of both polished and rough surfaces – Sample 1. Schematic drawing covers only half plates but the measurements were conducted in full, totalling 16 substrata on each surface.

The results show that:

- There is, not surprisingly, a consistent roughness difference between the polished and the rough surface (Figure 6). The parameter used for this comparison is Sq, the one displaying the smallest relative error for both surface type.
- Some height and volume parameters are to be used with caution as they are sensitive to isolated peaks and pits which may not be significant. They could be used if extreme peaks and valleys are removed or a threshold is applied[6]. Therefore, considering the high interference from small distortions in the analysis, it can be concluded that there is a small systematic error (bias) between whole area analysis and SRS for some parameters, such as Sq, Vv and Vvv.
- The other parameters tend to show a higher systematic error (bias) between the two analyses because they are more sensitive to small distortions that usually are averaged out when analysing a bigger area.

Figure 7 illustrates small distortions of this kind.

Method validation

To validate the SRS approach a Wilcoxon Signed Rank Test was used (for paired samples) to determine whether there were

Table 1. Analysis of surface data for polished surfaces.

Parameter	Polished surfaces								
	Whole Area	SRS	Relative Error	Whole Area	SRS	Relative Error	Whole Area	SRS	Relative Error
	Sample 1	Sample 1	Sample 1	Sample 2	Sample 2	Sample 2	Sample 3	Sample 3	Sample 3
Sq	5.6315	6.254	-11%	3.1155	3.102	0%	4.264	4.318	-2%
Ssk	-7.1535	-4.264	40%	-1.492385	-1.25	16%	-0.095	-0.105	-1%
Sku	47.83	60.234	-26%	21.79	22.015	-1%	15.012	16.04	-5%
Vm	0.05677	0.016	72%	0.18275	0.182	0%	0.654	0.54215	61%
Vv	2.7395	2.84	-4%	1.51385	1.235	18%	2.661	2.4065	21%
Vmp	0.05677	0.063	-11%	0.18275	0.152	17%	0.778	0.54215	155%
Vmc	1.00055	1.121	-12%	0.64605	0.524	19%	1.75	0.8487	172%
Vvc	1.2945	1.125	13%	0.7887	0.624	21%	1.523	1.60435	-13%
Vvv	1.4447	1.332	8%	0.72515	0.654	10%	0.745	0.80185	-9%
Smr	0.000080365	0.0005042	-527%	0.000066345	0.00006624	0%	0.00004032	0.000008035	49%
Smc	2.6825	2.745	-2%	1.331	1.223	8%	1.564	1.864	-25%
Sdr	13.796	12.052	13%	142.24	145.321	-2%	646.21	332.9	216%

Table 2. Analysis of surface data for rough surfaces.

	Rough surfaces								
Parameter	Whole Area	SRS	Relative Error	Whole Area	SRS	Relative Error	Whole Area	SRS	Relative Error
	Sample 1	Sample 1	Sample 1	Sample 2	Sample 2	Sample 2	Sample 3	Sample 3	Sample 3
Sq	27.67	25.21	9%	27.4	31.7	-16%	36.36	42.21	-18%
Ssk	0.028265	0.1120215	-296%	0.052936	0.023549	56%	0.04564	0.03215	57%
Sku	2.436	2.315	5%	2.6115	1.5362	41%	2.452	3.111	-43%
Vm	1.0685	1.5445	-45%	1.1662	1.3641	-17%	1.428	0.987	32%
Vv	37.485	35.046	7%	37.58	39.24	-4%	49.365	53.321	-10%
Vmp	1.0685	2.354	-120%	1.1662	1.3214	-13%	1.428	1.564	-10%
Vmc	26.965	27.165	-1%	25.775	27.664	-7%	35.37	37.35	-7%
Vvc	34.845	36.011	-3%	34.79	36.21	-4%	45.92	47.36	-4%
Vvv	2.6385	2.7892	-6%	2.7945	2.654	5%	3.447	3.664	-8%
Smr	0.00003764	0.001214	-3125%	0.000020098	0.000032151	-60%	0.0001084	0.000632	-1629%
Smc	36.415	36.154	1%	36.415	39.215	-8%	47.935	42.635	14%
Sdr	18594.5	18596.5	0%	7041.5	7951.3	-13%	31870.5	36321.5	-56%

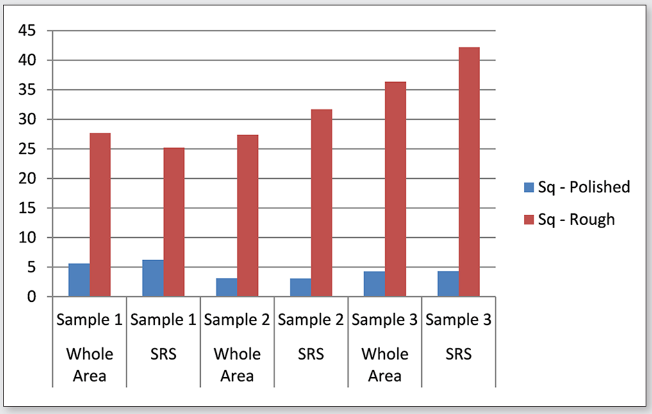

Figure 6. Root mean square height (Sq parameter) for rough and polished surfaces showing data consistency. This parameter was chosen because it displays the smallest relative error for both rough and polished surfaces

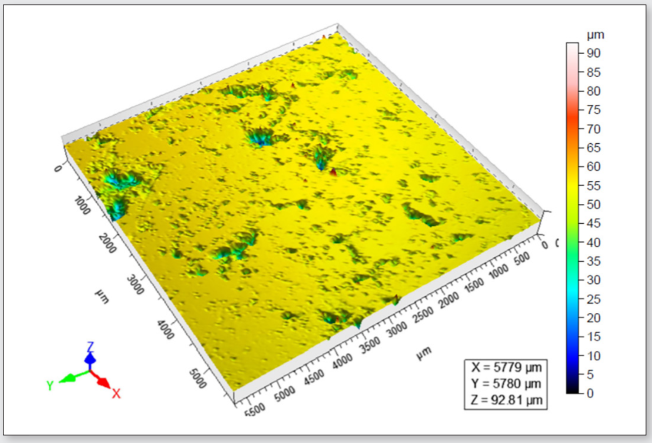

Figure 7. Surface morphology image obtained through the interferometer software. Note "small distortions".

significant differences between the whole area and the 6SRS measurements. The signed rank test compares the median of the values with a hypothetical population median (represented as the reference whole area in this study). Both the difference between these two values and the confidence interval of the difference are compared. The test leads to accepting the null hypothesis when there is no significant difference between the two values. Since the nonparametric test works with ranks, it is usually not possible to get a confidence interval with exactly 95% confidence[7].

Out of six Wilcoxon Signed Rank Tests, five retained the null hypothesis and one test rejected the null hypothesis. It is however believed that this is due to an experimental error and it is currently being re-measured.

The tests refers to three selected strata measured both rough and polished surfaces. The main results are:

- There is no significant difference between the whole area analysis and Systematic Random Sampling analysis (84% adherence) despite having one rejected case. In order to confirm this result more tests are currently being carried out amongst others varying number of sampled points in the SRS design.
- Thus it is possible to validate the method, at a first stage. The SRS approach will be very useful to reduce the time needed for estimating granite roughness. The new method provides the final result for one 20x20 cm granite plate in less than one hour, unbelievably fast compared to 16 days with the current approach.

Conclusions

Radon gas is formed from natural materials that have one of its natural precursors as part of its compositional make up. Granites tend to have a high radon gas emanation rate and since they are used as ornamental rocks inside and outside buildings it is important to assess the concentration reliably. This study presents a method that substantially decreases the time and resources needed to perform these important assessments. Analysis of the experimental results demonstrates the feasibility of a Systematic Random Sampling approach to obtain reliable estimates of granite roughness, an approach that is much faster than analysing the entire target area (factor of 192). As a consequence of the reduced measurement time, the cost of the test also decreases substantially since the interferometer is expensive when rented by the hour.

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