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Theoretical and experimental estimation of self-attenuation corrections in determination of ^{210}Pb by gamma-spectrometry with well Ge detector.

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Abstract

This paper aims at giving a practical method for routine measurements of the activity of ^{210}Pb in solids by gamma spectrometry with a well Ge crystal. Since the gamma rays emitted by ^{210}Pb have a low energy, 46.5 keV, the proportion of pulses counted in the crystal depends strongly on the sample composition trough self-attenuation within the sample itself. Self-attenuation can be calculated by use of a Monte Carlo method based on a model. In the present work two codes, namely GEANT 4 and a Laboratory code gave results in agreement. However, both necessitate a good knowledge of the sample composition, of the detector geometry and of the computing programs. In order to circumvent such drawbacks, was developed an experimental technique, where the overall counting efficiency can be derived directly from the transmission of 46.5 keV γ–rays through the sample, by means of a linear relationship. Such correlation was observed experimentally with samples of known activity and the approach was validated by verification that a linear relation preliminary exists between the transmission and the calculated self-absorption.

Key-words

Well Ge detector; gamma spectrometry; lead 210; self-attenuation

1. Introduction

^{210} Pb is a very useful radioactive element for environmental studies (Ivanovich and Harmon, 1992). It has been used extensively for measuring the sedimentation rates in lakes, estuaries and the coastal marine environment during the past century, since Godlberg (1963) introduced it as a geochronometer. Measuring its activity in air and in surface soils will afford quantitative information about the flux of ^{222}Rn and its daughters in the atmosphere. ^{210}Pb can help in uranium exploration, through the radon yield, and in monitoring transfers of nuclides of the uranium series in soils and aquatic systems. In the context of luminescence dating (i.e., our concern), the ^{210}Pb/ ^{226}Ra activity ratio can give the proportion of ^{222}Rn that can escape from a given sediment, such data being of importance in the calculation of annual radiation dose rate (Aitken, 1985).

There are three basic approaches to the measurement of ^{210}Pb activity (Ivanovich and Harmon, 1992): by alpha spectrometry of ^{210}Po, assuming radioactive equilibrium between the two nuclides; by beta spectrometry, observing the growth of its daughter ^{210}Bi; and by gamma-ray spectrometry, which allows direct measurement in various media, including water, rocks and soils (see e.g., Allisy et al., 1994) . It consists in counting, using an adapted detector, the specific gamma rays that are emitted at 46.5 keV by the nuclide, during a certain number of hours, for a given amount of sample sealed in a container. The number, N (counts.s^{-1}), of such photons, that are observed per time unit is directly connected to the activity A (Bq) of ^{210}Pb in the sample. The ratio $\varepsilon = N/A$ (global efficiency) depends on :

- The "branching ratio" (4.25 %) of the gamma ray, that is the proportion of disintegrations that give rise to a 46.5 keV photon (data from the National Nuclear Data Center, Brookhaven National Laboratory).

- The attenuation in the sample; this means that some of the gamma rays that are emitted will lose partially or totally their energy in the sample itself before leaving the container, resulting

in a diminution of the number of 46.5 keV gamma rays that can reach the detector. This effect is called self-attenuation. The self-attenuation factor η, is defined here as the ratio:

$$\eta = N/N_0 \quad [1]$$

where N_0 (counts.s^{-1}) is the number of 46.5 keV photons per unit time that would be counted for the same sample if no attenuation occurred within the sample.

- The efficiency ε_0 of the counting system, that is the proportion of initially emitted 46.5 keV photons that would be counted if no self-attenuation occurred. It depends on the physical properties and geometry of the detector and sample container, but not on the sample characteristics.

$$\varepsilon_0 = N_0/N'_0 \quad [2]$$

where N'_0 (counts s^{-1}) = total number of emitted 46.5 KeV photons = 0.0425A

We finally have :

$$A = N/\varepsilon = N/(0.0425\ \varepsilon_0\ \eta) \quad [3]$$

For evaluating the activity of ^{210}Pb in various samples in routine measurements, assessment of ε needs evaluation of ε_0, once for all, and η for each individual sample. ε_0 can be obtained by means of calibration using reference samples of well known activity. And its evaluation needs also, first, evaluation of η for the reference samples themselves. The problem is that η can significantly vary from sample to sample (see Table 1). Actually, for a given container volume and geometry, the self-attenuation depends strongly on both the composition and apparent density of the sample (Cutshall et al., 1983).

Self-attenuation can be theoretically calculated using physicals models of interaction between gamma rays and matter, computed with a Monte Carlo technique. Such techniques, in which stochastic phenomena can be simulated by means of random draws generated by the computer, have been adapted to the assessment of the counting efficiency for germanium

detectors in various configurations (e.g., Nakamura, 1975 ; Kamboj and Kahn, 2003). In the present work, two different Monte Carlo codes have been used specifically for evaluating self attenuation in the well geometry: (i) a home-made code (Laboratory code, hereafter) and (ii) a program that is widespread in the field of nuclear physics, namely GEANT 4 (Agostinelli et al., 2003). Results will be presented.

Alternatively, some experimental approaches have been proposed. Wilson (1980) based his method for determination of absolute activity of large volume geological samples, without being hindered by self-attenuation, on replicate counting of increasing volumes of the unknown samples. He obtained good results but the method was time consuming. Cutshall et al. (1983) have reported a method based on an evaluation of the transmission of low energy γ-rays from a point source placed on an aluminium container in the presence and absence of sample. Further, Joshi (1989) attempted to establish a direct correspondence between the measured count rate for the sample and the count rate expected for material identical to that used for efficiency calibration using γ-ray transmission; this was for measuring ^{241}Am (at 59.6 keV), however the situation is nearly the same as for ^{210}Pb. Bolivar et al. (1996) generalized the method of Joshi, by describing the transmission factor as a single function of the gamma-ray energy, sample density and geometry. Using a derived approach, in the present paper, we show that the transmission factor for ^{210}Pb is strongly correlated to the self-attenuation that is calculated using a Monte Carlo method. This allowed to develop and justify an experimental method of direct evaluation of the overall counting efficiency for unknown samples using a calibration curve made by plotting the efficiency vs the transmission factor for a set of reference samples.

2. Experimental setting

We deal here with spectrometry of rocks and soils by means of an intrinsic germanium cylindrical crystal (Canberra GCW3523, 193 cm^3 ; 65x65 mm) in a well configuration (54x23 mm) (Fig.1). The thickness of the dead layers is not accurately given by the manufacturer, and this is discussed further.

The samples to be measured are finely ground (< 100 μm) before being poured and compressed by hand in a container made of PETP (polyethylene-terephtalate). Use of PETP is intended to get a radon-tight container with a view to evaluating not only ^{210}Pb, but also ^{226}Ra from its short-lived post-radon daughters ^{214}Pb and ^{214}Bi, using the gamma ray emissions at 195.2, 351.9 and 609.3 keV (measurements after a delay of 15 days for getting secular equilibrium); it has been experimentally observed that commercially available containers that would geometrically suit the well are generally made of polymers that are porous to radon (e.g., polystyrene crystal). The volume of the container, 4.35 cm^3, is always completely filled up with powder. The samples that are routinely measured are common rocks and sediments, with a low ^{210}Pb activity, typically in the range 10-100 Bq/kg. In this context, counting is usually performed for at least three days, so that a statistically satisfying number of counts (usually, more than ~1200, resulting in an uncertainty of less than 10%) is got in the 46.5 keV photo-peak (after subtraction of Compton background). Evaluation of counts in the peak is derived from a commercially available software from Canberra (Genie 2000).

The reference samples comprise: commercially available powdered rock standards and various powdered rocks that have been dispatched within several laboratories for comparisons of radioactivity measurements (Faïn et al., 1997) (Table 2). ε_0 can be averaged over several reference samples.

3. Determination of self-attenuation corrections

3.1 Monte Carlo methods

The aim is to evaluate the mean self-attenuation factor for the gamma rays that are able to reach the crystal detector and that are prone to be entirely absorbed for finally giving rise to a pulse in the photo-peak. For this purpose it is necessary to have a good knowledge of the detector geometry and characteristics. However, as mentioned earlier, the thickness of the germanium dead layers is not accurately known. Such information is absolutely necessary for calculation of the overall efficiency, ε, of the detector, but has a very weak impact on the self-attenuation; this is because such attenuation occurs within the sample, before reaching the Ge-crystal. Actually, attempts have been made to assess the thickness of the dead layer in the well (work in progress) and preliminary estimations have been tentatively taken into account in the calculation of the self attenuation with both codes. Results indicated that a dead layer of 25μm would give the same self-attenuation as without dead layer within statistical uncertainty.

3.1.1 Laboratory code

The self-attenuation factor η is calculated as the ratio:

$$\eta = (\Sigma p_a)/(\Sigma p_w) \quad [4]$$

where :

p_a = probability of interaction within the crystal with attenuation within the sample taken into account; p_w = probability of interaction in the crystal without attenuation within the sample. Summation is made over a great number of draws (typically, 10^4). For the sake of simplification, it is assumed that every 46.5 keV photon that may interact in the crystal will be

entirely absorbed and give rise to a pulse in the peak. This is justified due to the short range of the low energy photons in germanium (less than 1mm).

Basically, the successive steps of the calculation are as follows :

- 1: Draw at random an emitting point within the sample
- 2: Draw at random a direction for the emission of the gamma ray
- 3: Test possible intersection of this direction with the crystal : if not, go back to step 1
- 4: Calculation of the geometrical path lengths in the different media (sample, container walls, aluminium shield, germanium crystal). The parameterisation uses cylindrical coordinates.
- 5: Calculation of p_a and p_w.

p_w is calculated as follows:

$$p_w = [\exp(- \delta_p\mu_p\rho_p - \delta_k\mu_k\rho_k - \delta_d\mu_g\rho_g)] [1 - \exp(-\delta_g\mu_g\rho_g)] \qquad [5]$$

where δ_p, δ_k, δ_g and δ_d (cm) are the geometrical path lengths in the container walls (p), aluminium shield (k), dead layer (d) and "active" germanium (g), respectively and μ_i ($cm^2.g^{-1}$) and ρ_i ($g.cm^{-3}$) are the corresponding attenuation coefficients and densities. The formula [5] is valid whatever the energy; however, as noted earlier, the second term, $[1 - \exp(-\delta_g\mu_g\rho_g)]$, which relates to absorption in germanium, equals 1 at 47 keV. The coefficients μ_i are computed from the major element composition of the samples and the attenuation coefficients for those elements are extracted from the NIST tables (http://physics.NIST.gov/PhysRefData/XrayMassCoef/cover.html). Densities ρ_i are simply derived from the mass and volume (4.35 cm^3) of the sample in the container.

p_a is given by:

$$p_a = p_w [\exp(- \delta_s\mu_s\rho_s)] \qquad [6]$$

where the subscript s is related to the sample characteristics.

The results for a set of 15 different samples are shown on Table 1 (for δ_d = 25 μm). The standard deviation for successive runs is around 0.3 %, one run of 10^4 draws taking 30s with a personal computer (equipped with a Pentium II, 233 MHz).

3.1.2 GEANT 4

GEANT 4 is a general toolkit for simulating the passage of particles through matter (Agostinelli et al., 2003). It includes all possible physical interactions and corresponding data tables. In the present work, the version GEANT 4.5.2 was applied, together with the Low Energy Electromagnetic Processes option. In the frame of the present work, the code allowed one to follow and list, step by step, all the interactions of the gamma rays and the resulting electrons in the different media : sample, container walls, aluminium, dead layer, germanium. For evaluating the global efficiency ε for a given sample, an homogeneous source was simulated in the sample container, that emitted at random, in all directions, a simple 46.5 keV photon. In term, the total number of gamma rays that let all their energy in the crystal was compared to the total number of created photons. The ratio gave the overall efficiency. The computation was performed with various theoretical compositions of the sample in the container, including air only. The self-attenuation factor η was then evaluated as the ratio between the efficiency calculated with a rock or sediment sample and the efficiency calculated with an "air sample". Note that all the created photons were tracked; thus, more draws were necessary than using the Laboratory code which selects the photons that can give a pulse in the photopeak. Routinely, processing one sample (10^6 draws) takes around 20 minutes, leading to a reproducibility of successive runs within ± 0.2 %. It can be seen (Table

1) that the results are not much different from the results obtained using the Lab. code, the correlation between the results of both methods being: $\eta_{Geant} = 1.012\ (\pm\ 005)\eta_{Lab.} + 0.003\ (\pm\ 0.002)$ ($R^2 = 0.998$).

3.2 Experimental

3.2.1 Self-attenuation and efficiency vs transmission

The experiments presented below are derived from the transmission method proposed by Cutshall et al. (1983) and developed by Joshi (1989) and Bolivar et al. (1996). The basic idea was to look for a correlation between the experimental attenuation through the whole sample, measured with an external gamma source, and the self-attenuation coefficient. For that purpose, the experimental setting of Fig.2 was used. The source is made of ^{210}Pb (283 Bq in December 2003) diluted in a polymer matrix, that forms a disk (ϕ=9mm) deposited onto an aluminium holder. An aluminium mounting allows insertion of a PETP tube between the source and the top surface of the aluminium shield of the detector. The attenuation is calculated by the ratio $n_s/n_{0,}$ with n_s = [counting rate with a tube containing the sample] and n_0 = [counting rate with an empty tube]. Actually, n_s can be corrected as n'_s, taking into account the gamma rays at 46.5 keV that come from the sample itself. n'_s is only different from n_s for very active samples; it can be easily derived from a single measured ratio between the counts in the well position and the counts in the position of Fig. 2. The plot of n'_s/n_o for various samples of rock and sediment against the self-attenuation coefficient calculated for the same samples gives a linear correlation in the field of usual rock and sediment compositions, as well for the η calculated with GEANT 4 as for η calculated with the Laboratory code (Fig. 3). The correlation looks linear ($R^2 = 0.988$) but for an outlying sample that has been shown for comparison: a non-natural manganese rich sand (lower black triangle).

The above results suggested that the overall efficiency should be also linearly correlated to the transmission. This was verified using a set of 8 reference samples, which are common rocks of known ^{210}Pb activity, by plotting ε vs n'_s/n_0 (Fig. 4). The precision on ε was limited by the low signal/background ratio around 47 keV; however, this is true whatever the approach that is adopted with gamma spectroscopy of ^{210}Pb. This limited the correlation coefficient to 0.89 in the present example.

3.2.2 Practical method for ε assessment

For a practical purpose, i.e., estimating the efficiency at 46.5 keV for unknown samples, it is sufficient to know the correlation between the transmission n'_s/n_0 and the efficiency ε. For ordinary rocks and sediments samples, the ^{210}Pb activity A (Bq) can be estimated by use of a calibration curve, where A/N (Bq.s.counts^{-1}) is plotted vs n'_s/n_0. This is done routinely in our laboratory. With the ^{210}Pb source that is used, a statistical uncertainty of less than 1 % on the experimental attenuation (horizontal axis on Fig.4) can be attained within around one day of counting; this lengthens the total counting time for one sample by around 25%.

Since the available samples of known ^{210}Pb activity were used for plotting the calibration curve (Fig. 4), it was not possible to test the method with such samples. We tested it another way: samples C396bis and C397 (Table 1), not used for the calibration curve, should show secular equilibrium between ^{226}Ra and ^{210}Pb according to geological considerations (sediments included in a lava flow, not allowing radon migration). Because the activity of radium can be evaluated with a good accuracy using gamma spectroscopy, an agreement between the activity of radium and the activity of ^{210}Pb measured as above would be a test. ^{226}Ra was basically measured by the same method as for ^{210}Pb; however, since the self attenuation at 295 keV and above is significantly less important and less composition-dependant than at 46 keV, it is simply accounted for by use of a factor $\exp(-\delta_m \mu_m \rho_s)$, where

δ_m is the mean path length in the tube (0.65cm, evaluated experimentally) and μ_m is the attenuation coefficient at the energy of interest for a standard sediment sample. We finally found activities ratios $^{210}Pb/^{226}Ra$ of 1.00 ± 0.15 and 1.08. ± 0.11 (1σ) respectively. This illustrates the potentiality of the technique in one of the fields of interest (see section 1).

4. Resume and conclusions

The use of a simulation code, either a home-made code or a widespread code like GEANT 4, can allow evaluation of the self-attenuation coefficients for the 46.5 keV gamma ray from ^{210}Pb emitted within a sample. This can be illustrated by the agreement that is obtained between the self-attenuation coefficients which were obtained using the two different approaches. However, such a method imposes the control of the code and its language (C++), a good knowledge of the detector geometry, including parts that can not be seen externally (data necessary for the simulation), and an assessment of the composition in major elements and water content for every sample to be measured. The evaluated value for the attenuation coefficient is not error free; this is illustrated by the difference that can sometimes be observed between the results obtained from two different approaches (Table 1), the origin of which is unknown (not accounting for errors in weighting, chemical analysis and geometrical data). For circumventing such problems, we have developed an experimental technique, derived from the transmission technique initiated by Cutshall et al. (1983), in which the counting efficiency is directly derived from the measured transmission at 46.5 keV, through a linear correlation obtained by use of reference samples. In the present work, the technique was limited to ordinary rocks and sediments in a certain range of elemental compositions (Table 2). We have indications that extending the range of compositions would result in a correlation that is no longer linear (see triangle on Fig. 3); however, a simplified model based on exponential attenuation (not reported here) indicated that the correlation cannot be strictly

linear and, therefore, it would not be irrelevant to use, e.g., polynomial fits for extending the range of measured samples.

Acknowledgements

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Figure captions

Figure 1.
Cross section of the gamma detector (cylindrical geometry). 1: germanium crystal; 2: powdered sample; 3: aluminium alloy shield; 4: sample container.

Figure 2.
Experimental setting for measuring attenuation through the whole container using a ^{210}Pb source. 1: germanium crystal ; 2: powdered sample; 3: aluminium alloy shield; 4: sample container; in black: ^{210}Pb source. The mounting was not drawn for sake of simplification.

Figure 3.
Black dots : experimental attenuation n'_s/n_0 using the setting of Fig. 2, vs calculated self-attenuation η (using the Laboratory code), for various natural rocks and sediments (including those of Table 1). The lower point (black triangle) is a non natural manganese sand and the upper one is water. Statistical error limits to within 2σ.

Figure 4.
Plot of the measured global efficiency ε, for a set of samples of known ^{210}Pb content vs experimental attenuation n'_s/n_0, using the setting of Fig. 2. Error bars are statistical + systematic (2σ).

Sample reference	η (Lab. code)	η (GEANT 4)	ε (experimental)
C341	0.563 ± 0.002	0.576 ± 0.001	
C347	0.719 ± 0.002	0.734 ± 0.001	0.52 ± 0.07
C396bis	0.712 ± 0.002	0.729 ± 0.001	
C397	0.704 ± 0.002	0.721 ± 0.001	
65SR300	0.813 ± 0.003	0.825 ± 0.001	0.61 ± .05
Al2O3	0.775 ± 0.002	0.790 ± 0.001	
SiO2	0.761 ± 0.002	0.773 ± 0.001	
water	0.871 ± 0.002	0.878 ± 0.001	
MAZ	0.579 ± 0.002	0.593 ± 0.001	
GOU	0.638 ± 0.002	0.650 ± 0.001	0.46 ± 0.06
MPX	0.751 ± 0.003	0.765 ± 0.001	
BEN	0.591 ± 0.002	0.597 ± 0.001	0.41 ± 0.06
1081	0.706 ± 0.002	0.720 ± 0.001	0.52 ± 0.04
1603	0.617 ± 0.002	0.623 ± 0.001	0.44 ± 0.07
1906	0.648 ± 0.002	0.657 ± 0.001	0.41 ± 0.05
2149	0.657 ± 0.002	0.664 ± 0.001	0.48 ± 0.06
623B	0.619 ± 0.002	0.629 ± 0.001	0.51 ± 0.15
CASC110	0.373 ± 0.001	-	

Table 1

Compared values of the self-attenuation coefficient η, obtained by modelization using a local code (10^4 draws) and GEANT4 (10^6 draws); errors are statistical only (1σ). Experimental values of the global efficiency ε for the samples which content of ^{210}Pb is known; overall uncertainty at 1σ.

sample reference	nature	SiO2	Al2O3	Fe2O3	MnO	FeO	MgO	CaO	Na2O	K2O	TiO2	P2O5	H2O	d	μ_0
C341	lava	46.95	16.67	12.15	.19		6.16	9.59	3.71	1.68	2.44	.57	.17	1.94	0.537
C347	granite	73.09	15.05	1.45	< LD		.38	.89	3.62	4.17	.13	.19	.82	1.54	0.362
C396bis	sediment	67.51	15.66	3.63	0		0.77	0.91	1.83	5.06	0.48	0.35	3.7	1.45	0.396
C397	sediment	71.94	15.2	3.16	0		0.72	0.78	1.83	5.23	0.34	0.24	0.45	1.51	0.393
SiO_2	silica	100												1.43	0.318
65SR300	sediment	45.74	13.93	3.64	0.02		0.46	0.48	0.73	2.56	0.54	0.2	30.61	0.94	0.353
Al_2O_3	alumina		100											1.42	0.292
MAZ	lava	49.19	17.13	12.12	0.22		4.69	8.47	4.36	2.29	2.39		0.33	1.82	0.537
GOU	lava	54.9	18.2	7.91	0.22		2.25	5.47	5.4	3.14	1.38		0.03	1.69	0.467
MPX	sandstone	86.00	6.89	0.83	0.01		0.11	0.07	0.19	2.99	0.05		0.1	1.38	0.345
BEN	lava	38.2	10.07	12.08	0.2		13.2	13.9	3.18	1.39	2.61	1.05	0.5	1.72	0.548
1081	lava	62.43	16.55	2.45	0.16	2.35	1.59	2.91	6.27	3.38	1.1	0.31	0.27	1.41	0.381
1603	lava	47.53	18.21	3.76	0.18	6.68	5.48	10.7	4.16	1.1	1.77	0.65	0.13	1.66	0.416
1906	lava	51.02	18.63	4.25	0.14	5.09	2.98	8.51	4.69	2.08	1.66	0.71	0.21	1.52	0.420
2149	lava	49.94	20.38	5.43	0.14	2.75	3.24	9.29	4.58	1.16	1.51	0.62	0.68	1.53	0.433
623B	lava	48.65	14.73	2.92	0.16	7.51	10.3	9.55	3.62	0.7	1.55	0.39	0.19	1.65	0.396
water													100	1.00	0.217
CASC110	Mn sand	2.83	6.81	6.5	64.98		0.47	0.33	0.18	1.09	0.27	0.34	15.11	1.70	0.120

Table 2

Elemental composition (%) of the samples and their density d ($g.cm^{-3}$) in the container. Most compositions were obtained by ICPMS. The mass attenuation coefficient, μ_0, is in cm^2g^{-1}.

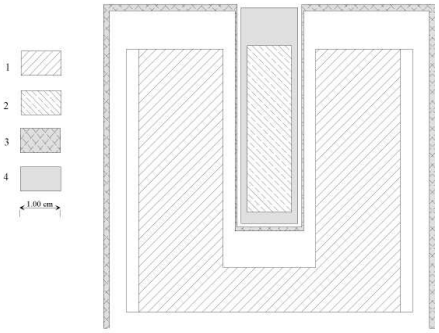

Figure 1

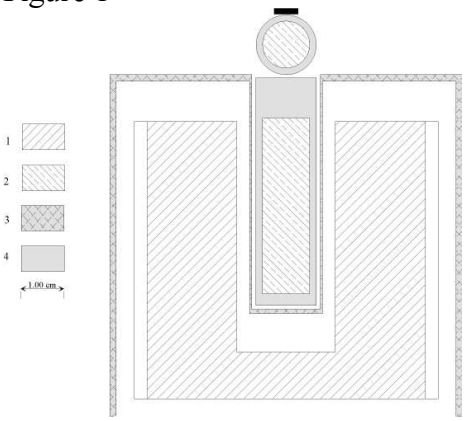

Figure 2

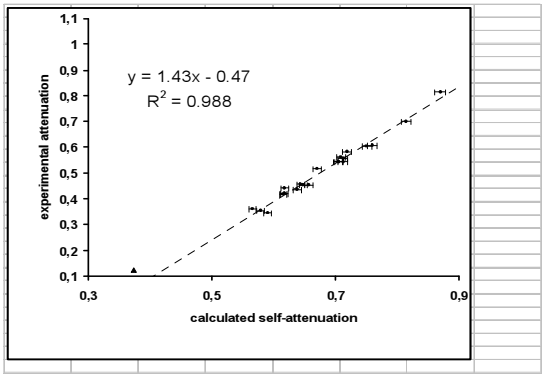

Figure 3

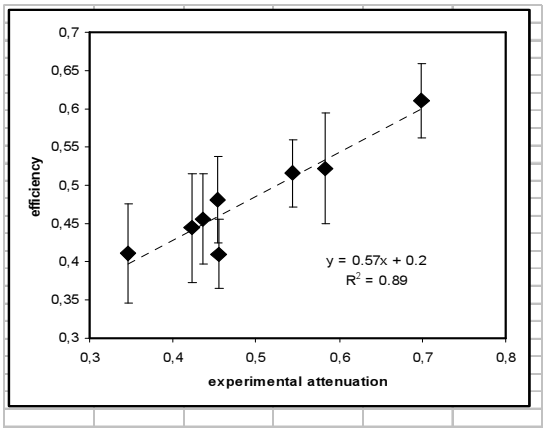

Figure 4