A generalized exponential function model of the integrity loss of stones subjected to laboratory weathering cycles

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Abstract

Ultrasonic measurements are widely used as NDT method in historical structures for evaluation of stone material strength properties. Actual stone degradation in situ is estimated based on various laboratory tests which allow researchers to correlate the number of artificial ageing cycles of stone specimens with ultrasonic wave velocity measured on these specimens. Paper presents the results obtained for granite, marble, limestone, travertine and sandstone which undergone various cyclic ageing tests including freezing and thawing, high temperature and salt crystallisation. Analysis of the obtained results shows that independent of the stone type tested and independent of the ageing test applied a rate of change of the stone elastic properties is described by an ordinary differential equation which solution is an exponential law analogues to the Newton's law of cooling. The degradation function model can be used for further research on expected residual strength and dynamics of the heritage materials degradation processes.

Keywords

stone deterioration, decay function model, monuments, ultrasonic measurements, cyclic ageing

Introduction

Stone material ageing is a process of natural degradation of mechanical and decorative properties of stones due to long lasting environmental processes (ICOMOS ISCS, 2008)[1]. In geology these processes are related to a weathering of stone material (Irfan and Dearman, 1978) [2].

Susceptibility of a stone to weathering depends on its morphological and physical-mechanical properties, environmental conditions and exposure time. The ageing process dynamics can hardly be observed in natural conditions because of its slowness. An important role of engineering geology is the assessment of actual and future dynamics of stone material degradation and its susceptibility to environmental loadings. The deterioration processes adversely change structural and aesthetic properties of stone material. This problem is economically important also in Poland which has a long tradition of using stone structural and decorative elements in heritage buildings. Nowadays civil engineering and aesthetic use of stones also pursue the research on the dynamics and modelling of deterioration processes.

Engineering geology developed geomechanical research methods for non-invasive diagnostics of stone susceptibility to environmental risks. These methods can be studied in laboratories [3,4,5] and advantageously forecast stone integrity based on geomechanical tests of accelerated ageing. Measured values of e.g. ultrasonic wave velocity, reboundness hardness, strength etc. can be plotted against number of cycles of the ageing and observed trend extrapolated to predict further decrease of stone elastic properties [6], There are also papers presenting deeper insight in stone deterioration mechanisms however they publish fresh and degraded stone material properties after one time period or one number of cycles without intermediate stages which does not inform about a dynamics of deterioration processes [7,8,9] or valuable works showing deterioration trends in graphical or mathematical form however with an insufficient information about measurement data to allow a reader to analyse the results by himself [10].

Comparing an impact of various deterioration processes it is possible to determine dominating and less important environmental factors for various building materials. To this purpose more interesting than simple mathematical extrapolation of the test results is having a physical model of a stone deterioration process and of degradation of its properties. The interesting idea was presented by Mutlutürk et al. in 2004 [11] of adopting to this purpose well known radioactive decay function. The same decay function was used to describe a decrease of the compressive strength of natural hydraulic lime mortar due to varying leaching time [12].

In 2005 the Mutlutürk model was considered to describe stone durability under various ageing processes within EC Project MCDUR and generalised version of the decay function called fatigue law there, with non-zero asymptotic stone integrity level (residual integrity), was proposed by Exadaktylos and analysed by the Project Partners. According to the authors best knowledge, it has not been published except in MCDUR Final Report [13].

In the paper a resilience of stones from Mediterranean Basin and Poland to deterioration processes and influence of moderate and Mediterranean climates environmental conditions on these processes is analysed. The process is modelled by the exponential law proposed in MCDUR being the general exponential decay function analogues to the Newton’s law of cooling.

Tested stone materials

Tested stone materials from Mediterranean Basin are represented by Gioia Marble (MG), Italy and travertine from Pamukkale (PA), Turkey. These stones have been used as structural and decorative materials both in ancient times and nowadays. The stones from Poland represented by sandstone from Szydłowiec (SZ), granite from Strzegom (ST) and limestone from Raciszyn (RA) known under technical name “Polish travertine” are the stones used in today constructions for internal and outside cladding. Travertine, marble, sandstone and limestone were fresh from quarry while granite was displaced from XIX c. grave tombs during conservation works. The petrographic characteristics of the tested stone materials are presented in Table 1.

Table 1. General petrographic characteristics of tested stone materials

Stone	Quarry / Country (material)	Geological period	Petrographic characteristics
MG	Carrara / Italy (marble)	Lower Jurassic	marble with typical granoblastic polygonal microstructure compact texture, non-directional (calcite 99%, dolomite traces)
PA	Pamukkale / Turkey (travertine)	Pleistocene	crystalline limestone composed of carbonate sediment, consisting mainly of calcite, sparite, porous sedimentary stone , there are remains of plants in the stone
SZ	Szydłowiec / Poland (sandstone)	Lower Jurassic	fine-grained and medium-grained sandstone with siliceous binder
ST	Strzegom / Poland (granite)	Carboniferous	medium-grained granite, light grey colour, full-crystalline, non-directional texture; it consists mainly of plagioclase, quartz, biotite, rarely hornblende
RA	Raciszyn / Poland (limestone)	Upper Jurassic	limestone with ooid, with a biomorphic structure, macroporous with a stoney background composed of organic fragments, pseudooolites combined with fine-grained calcite

Physical and mechanical properties of the tested stone materials are given in Table 2. The selected stones represent sedimentary, metamorphic and igneous stones and cover a broad range of these properties. For example uniaxial compressive strength varies from16.8 MPa to 109.1 MPa for Mediterranean stones and from 42.8 MPa to 120.8 MPa for stones from Poland. Similar observation can be done in the case of other stone properties. These allowed to analyse dynamics of degradation processes for relatively large variety of stone materials and present conclusions regarding possibility of efficient modelling of stone integrity loss being an interesting and practically important geomechanical problem.

Table 2. Physical and-mechanical properties of tested stone materials

Stone	Real density [kg/m^3]	Bulk density [kg/m^3]	Total porosity [%]	Water absorption [%]	Longitudinal wave velocity [m/s]	Uniaxial compressive strength [MPa]
MG	2716	2708	0,76	0,28	1900	109,1
PA	2010	1790	23,7	5,75	3950	16,8
SZ	2670	1940	27.38	9.60	2980	46.0
ST	2670	2590	2,88	0,52	2680	120,8
RA	2680	2300	9,89	2,72	4050	42,8

Laboratory methods of deterioration testing

Non-destructive testing (NDT) of stones has a main advantage of using the same specimens throughout the whole research programs – from origin stone state and after each cycle of laboratory accelerated ageing. Thus measurement results in each cycle are performed on the same material and not influenced by possible differences between various specimens.

Ultrasonic measurements are among the most often used NDT methods because of their reliability, ease of use and direct relation of the sound propagation velocity V to modulus of elasticity E of the material ($V^2 \sim E$). Assuming the Continuum Damage Mechanics damage model of the material [20] its integrity I_N after N deterioration cycles is equal to E_N/E_0 and hence

$$I_N = \frac{E_N}{E_0} = \left(\frac{V_N}{V_0}\right)^2 \qquad (1)$$

where E_N, V_N are the elasticity modulus and wave propagation velocity after N cycles and E_0, V_0 represents the elasticity modulus and the ultrasonic wave velocity recorded at the beginning of ageing experiments.

Non-destructive ultrasonic measurements of V_P and V_R velocities were made to tests weathering behaviour of five stones with known physical and mechanical properties. Longitudinal waves V_P were measured in transition mode on granite, sandstone, travertine and limestone along the height of the cylindrical specimens having 50 mm in diameter and the slenderness equal to 1. Ultrasonic V_P probes had frequency of 1 MHz and 0.5 MHz.

Rayleigh surface wave V_R [14] was measured on side walls of the prismatic marble specimen using edge probes emitting 2 MHz ultrasonic wave [15]. The time of flight of ultrasonic waves was recorded and sound velocity calculated for known travel distance. In each measurement case several recordings were done and the average ultrasound velocity values were calculated. The ultrasound velocity values before laboratory weathering are presented in Table 2.

The following geomechanical accelerated ageing tests were applied:

(A) freezing and thawing (F/T) of the marble, granite, limestone and travertine in the temperature range (-25 ^{o}C ÷ 20 ^{o}C) according to Standard (PN-EN 12371, 2010) [16];

(B) artificial insolation of the sandstone, granite, limestone and travertine, at 40 ^{o}C to simulate sun operation at Mediterranean and moderate climatic zones according to originally developed procedure by the second author (AB) [17];

(C) salt crystallisation for the limestone and travertine according to Standard (PN-EN 12370, 2001) [18].

Theoretical model of stone deterioration

Mathematical modelling of stone deterioration processes can be done using thermo-physical models [19] requiring many material parameters and extensive computer calculation time. Opposite to rigorous theoretical analysis it is reasonable to look for phenomenological description of stone deterioration. The main reason is operational suitability to geotechnical analysis, ease of use and neglecting the local variations of material parameters influencing the complicated thermo-physical-mechanical modelling results. A good phenomenological model can describe material deterioration with statistically acceptable accuracy at relatively low experimental and calculation costs. However it should not be simply a function approximating experimental data. For a model it is required to describe the law governing observed deterioration phenomena, otherwise deterioration processes of various stones under various environmental loadings could not be compared among each other.

An interesting and simple phenomenological model was suggested by Mutlutürk and co-author [11] and verified for 10 various stone types (limestones, marbles, travertines and diabase) using Shore hardness SH index (scleroscope method) as a measure of stone integrity. The authors observed that decay law known e.g. for describing a decay of radioactive elements can well describe their experimental observations. In this case the rate of loss of a stone integrity I is proportional to its integrity at the beginning of the each ageing cycle N

$$-\frac{dI}{dN} = \lambda I \qquad (2)$$

where $-dI/dN$ is a disintegration rate (integrity loss rate) and λ a positive constant number called decay constant. Integrity I equals to 1 for non-deteriorated material and equals to zero in the case of the complete degradation.

The solution of this differential equation is

$$I_N = \mathrm{e}^{-\lambda N} \qquad (3)$$

where λ is a decay constant, shows that stone integration I_N after N ageing cycles diminishes exponentially with the number of cycles. The law assumes that a stone completely deteriorates and losses its full integrity which is not always the case and depends on a stone properties and environmental degradation factors.

The mathematical model of the generalized deterioration law [13] is based on experimental observations that in some cases the final residual integrity is not zero and a continuation of the stone ageing process does not result in its further deterioration. Such a generalized model is analogous to the Newton's law of cooling and is expressed as

$$-\frac{dI}{dN} = \lambda(I - A) \qquad (4)$$

which means that in general the disintegration rate $-dI/dN$ is proportional to the difference between the actual integrity I and its asymptotic value $A \geq 0$ which is the residual integrity. Obviously when there is no residual integrity and a stone completely deteriorates then $A = 0$ and equation (3) becomes the same as (2). The solution of equation (4)

$$I_N = A + (1 - A)\mathrm{e}^{-\lambda N} \qquad (5)$$

allows to describe and compare degradation of stones with zero and non-zero residual asymptotic integrity A.

In the present research integrity model was calculated based on measurements of elastic waves propagation velocity using equation (5) and experimentally measured stone integrity is calculated as Material Elasticity Index (MEI)

$$MEI = \left(\frac{V_N}{V_0}\right)^2 \qquad (6)$$

The wave propagation velocity measurements were performed for longitudinal ultrasonic wave for every tested stone except of Gioia Marble for which ultrasonic surface Rayleigh wave was used.

Results and discussion

Measured ultrasonic wave velocities before and after deterioration cycles for the tested stones and various applied degradation tests performed in the laboratory of Institute of Hydrogeology and Engineering Geology of Warsaw University are presented in the below tables. MEI values calculated according to (6) are rounded to 4 meaningful digits.

Table 3a. Ultrasound velocity (Rayleigh surface wave) in F/T tests for Gioia Marble

Cycle	0	40	60	80	100	120
$V_{N\,[m/s]}$	1900	1600	1500	1450	1300	1500
$(V_N/V_0)^2$	1	0,7091	0,6233	0,5824	0,4681	0,6233
Model parameters	Decay constant **λ = 0.0251** | Residual asymptotic Integrity **A = 0.53** | Correlation coefficient **Rsq = 0.9099**					

Table 3b. Ultrasound velocity in F/T test for Strzegom Granite

Cycle	0	5	10	15	20
$V_{N\,[m/s]}$	4359	3792	3613	3391	3072
$(V_N/V_0)^2$	1	0,7568	0,6870	0,6052	0,4967
Model parameters	Decay constant **λ = 0.087** | Residual asymptotic Integrity **A = 0.42** | Correlation coefficient **Rsq = 0.9765**				

Table 3c. Ultrasound velocity in F/T tests for Raciszyn Limestone

Cycle	0	5	10	15	20	25
$V_{N\,[m/s]}$	4942	4895	4816	4836	4805	4788
$(V_N/V_0)^2$	1	0,9811	0,9497	0,9576	0,9453	0,9386
Model parameters	Decay constant **λ = 0.0814** | Residual asymptotic Integrity **A = 0.93** | Correlation coefficient **Rsq = 0.9289**					

Table 3d. Ultrasound velocity in F/T tests for Pamukkale Travertine

Cycle	0	5	10	15	20	25
$V_{N\,[m/s]}$	3882	3821	3781	3662	3644	3662
$(V_N/V_0)^2$	1	0,9688	0,9486	0,8897	0,8811	0,8897
Model parameters	Decay constant **λ = 0.0508** | Residual asymptotic Integrity **A = 0.83** | Correlation coefficient **Rsq = 0.9250**					

Table 4a. Ultrasound velocity in Insolation tests for Syzdłowiec Sandstone

Cycle	0	7	14	21	28	35	42	49	56
$V_{N\,[m/s]}$	3406	3368	3368	3339		3333	3333	3327	3339
$(V_N/V_0)^2$	1	0.9778	0.9778	0.9610		0.9576	0.9576	0.9541	0.9610
Model parameters	Decay constant **λ = 0.0983** | Residual asymptotic Integrity **A = 0.96** | Correlation coefficient **Rsq = 0,9300**								

Table 4b. Ultrasound velocity in Insolation tests for Strzegom Granite

Cycle	0	7	14	21	28	35	42	49	56	63	70
$V_{N\,[m/s]}$	4132	4063	3987	3755	3583	3488	3258	3222	3250	3190	3210
$(V_N/V_0)^2$	1	0.9669	0.9310	0.8258	0.7519	0.7126	0.6217	0.6080	0.6187	0.5960	0.6035
Model parameters	Decay constant **λ = 0.0178** | Residual asymptotic Integrity **A = 0.39** | Correlation coefficient **Rsq = 0.9565**										

Table 4c. Ultrasound velocity in Insolation tests for Raciszyn Limestone

Cycle	0	7	14	21	28	35	42	49	56	63	70	77	84	91	98
$V_{N\,[m/s]}$	4703	4627	4568	4457	4461	4500	4526	4525	4512	4475	4445	4399	4390	4390	4371
$(V_N/V_0)^2$	1	0.9679	0.9434	0.8981	0.8997	0.9155	0.9261	0.9257	0.9204	0.9054	0.8933	0.8749	0.8713	0.8713	0.8638
Model parameters	Decay constant **λ = 0.0395** | Residual asymptotic Integrity **A = 0.88** | Correlation coefficient **Rsq = 0,7669**														

Table 4d. Ultrasound velocity in Insolation tests for Pamukkale Travertine

Cycle	0	7	14	21	28	35	42	49	56	63	70	77	84	91	98	105
$V_{N\,[m/s]}$	4505	4481	4482	4499	4474	4468	4462	4448	4453	4436	4468	4425	4413	4400	4398	4391
$(V_N/V_0)^2$	1	0,9894	0,9898	0,9973	09863	0,9836	0,9810	0,9749	0,9770	0,9696	0,9864	0,9848	0,9596	0,9539	0,9531	0,9500
Model parameters		Decay constant **λ = 0,0005** | Residual asymptotic Integrity **A = 0,07** | Correlation coefficient **Rsq = 0,8893**														

Table 5a. Ultrasound velocity in salt crystalization tests for Raciszyn Limestone

Cycle	0	3	6	9	12	15
$V_{N\,[m/s]}$	5021	5009	5004	5001	4999	4997
$(V_N/V_0)^2$	1	0.9952	0,9932	0,9920	0,9913	0,9905
Model parameters	Decay constant **λ = 0.1906** | Residual asymptotic Integrity **A = 0.99** | Correlation coefficient **Rsq = 0.9956**					

Table 5b. Ultrasound velocity in salt crystalization tests for Pamukkale Travertine

Cycle	0	5	10	15	20	25
$V_{N\,[m/s]}$	3627	3607	3599	3598	3594	3589
$(V_N/V_0)^2$	1	0,9890	0,9846	0,9841	0,9819	0,9792
Model parameters	Decay constant **λ = 0.2352** | Residual asymptotic Integrity **A = 0.98** | Correlation coefficient **Rsq = 0.9778**					

The presented figures (at the end of the manuscript) are graphical representations of data shown in the Tables 3, 4 and 5. In the Figure 1 and Figure 5 a lack of one measurement data point can be seen. There is no experimental point for 20th F/T cycle of Gioia Marble and for 28th insolation cycle of Szydłowiec Sandstone. In both cases the measured wave velocities fallen outside the 95% error confidence bounds and were rejected. Altogether 85 experimental data recorded for five stones were analysed. The results are presented in Figures 1-10 with continuous lines representing integrity model function calculated according to equation (5) and discrete points show experimental MEI indices calculated using equation (6). Asymptotic integrity values *A* predicted by the model and correlation coefficient Rsq between the data and the model are also given in each figure.

In nine cases out of 10 the residual integrity A of the tested stones is moderate (0.39÷0.53) or high (0.88÷0.99). Only Pamukkale Travertine undergoing ageing due to artificial insolation (test type B) shows the residual integrity as low as A = 0.07 but degrades very slowly – only about 5% of integrity loss after 105 artificial insolation cycles. On the other hand Pamukkale Travertine sustains in-situ tens of thousands of real insolation cycles and still has the integrity greater than zero (monuments still exist). This suggests that developed artificial insolation test is probably much more aggressive than natural environmental loads.

Correlation of MEI with the model is very high in the most cases. The lowest value of Rsq = 0.7669 is for Raciszyn Limestone under insolation ageing.

Calculated model integrity functions allowed us to compare the dynamics of degradation processes under F/T for marble, travertine, granite and limestone. The comparison is presented in Figure 11. Without a well correlated model such a comparison would not be possible.

Conclusions

Laboratory experiments of artificial ageing of granite, marble, limestone, travertine and sandstone using freezing and thawing, insolation and salt crystallization tests combined with measurement of ultrasonic wave velocities allowed to observe deterioration of elastic properties of stones. The measured ultrasound velocities were used to define Material Elasticity Index (MEI) which describes experimentally detected loss of stone material integrity. The mathematical model of degradation of stone elastic properties analogues to the Newton's law of cooling shows good correlation with experimental MEIs. Calculated model degradation functions can be used for further research on expected residual strength and dynamics of the stone cultural heritage buildings, sculptures and artifacts degradation processes. As an example the comparison of dynamics of degradation of four different stones under cyclic freezing and thawing is presented. It can be seen that both a susceptibility of the stone to the environmental loading and the expected residual degradation level can be qualitatively described by the model.

Experimental data recorded and analysed for Pamukkale Travertine suggest that developed artificial insolation test is probably much more aggressive than natural environmental loads.

FIGURES

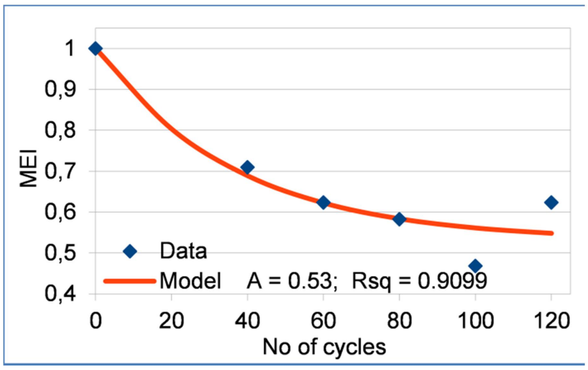

Fig. 1 Gioia Marble (IT); Freeze and Thaw test.

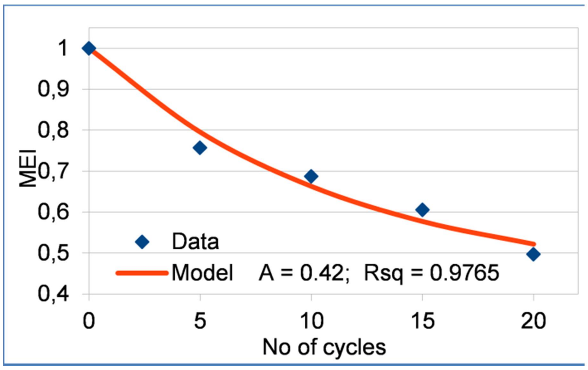

Fig. 2 Strzegom Granite (PL); Freeze and Thaw test.

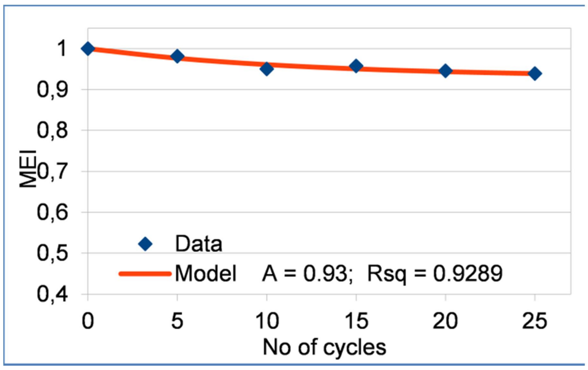

Fig. 3 Raciszyn Limestone (PL); Freeze and Thaw test.

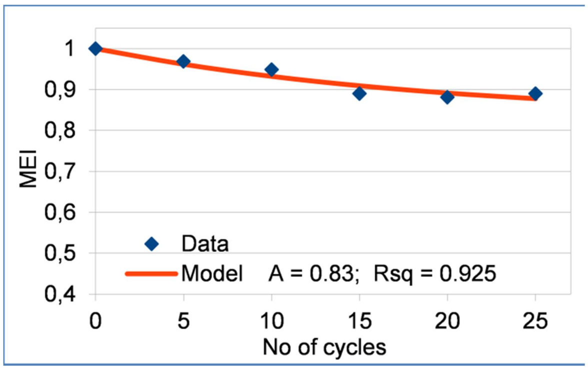

Fig. 4 Pamukkale Travertine (TR); Freeze and Thaw test.

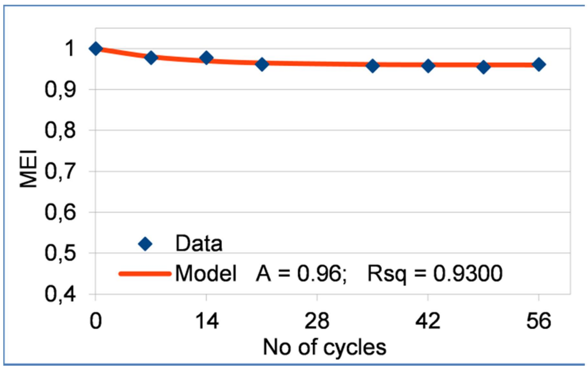

Fig. 5 Szydłowiec Sandstone (PL); Insolation test.

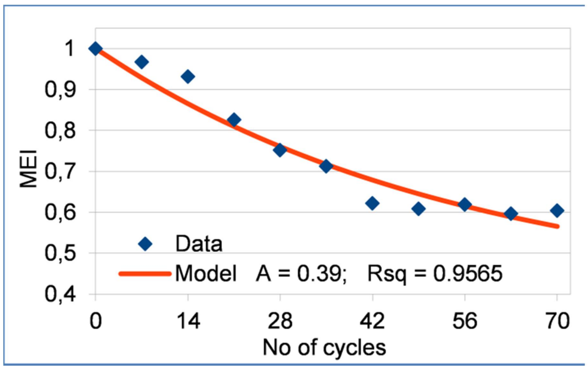

Fig. 6 Strzegom Granite (PL); Insolation test

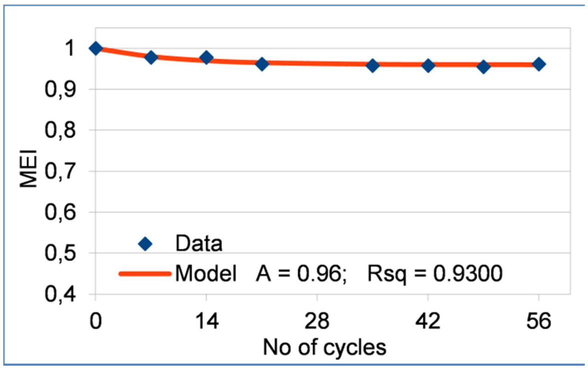

Fig. 5 Szydłowiec Sandstone (PL); Insolation test.

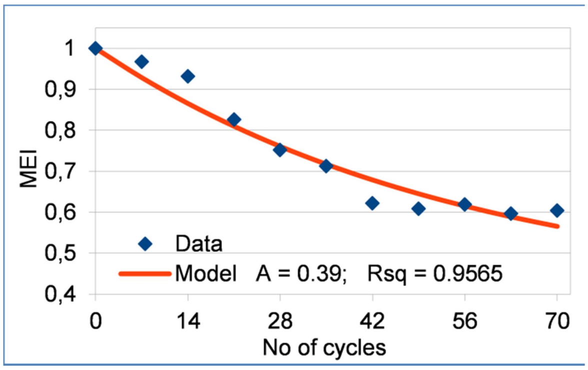

Fig. 6 Strzegom Granite (PL); Insolation test

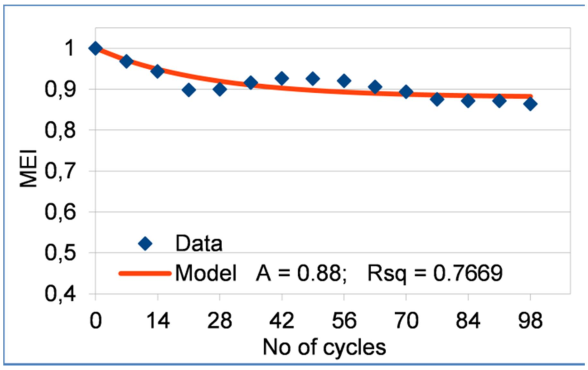

Fig. 7 Raciszyn Limestone (PL); Insolation test

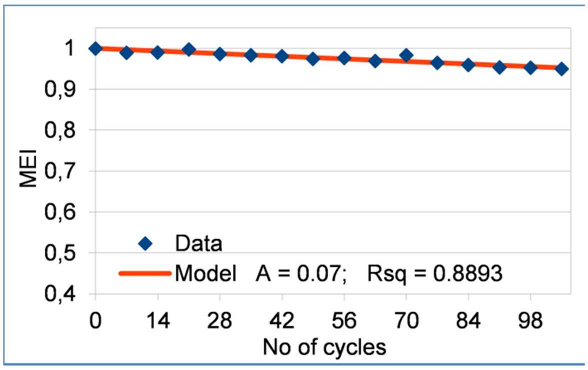

Fig. 8 Pamukkale Travertine (TR); Insolation test.

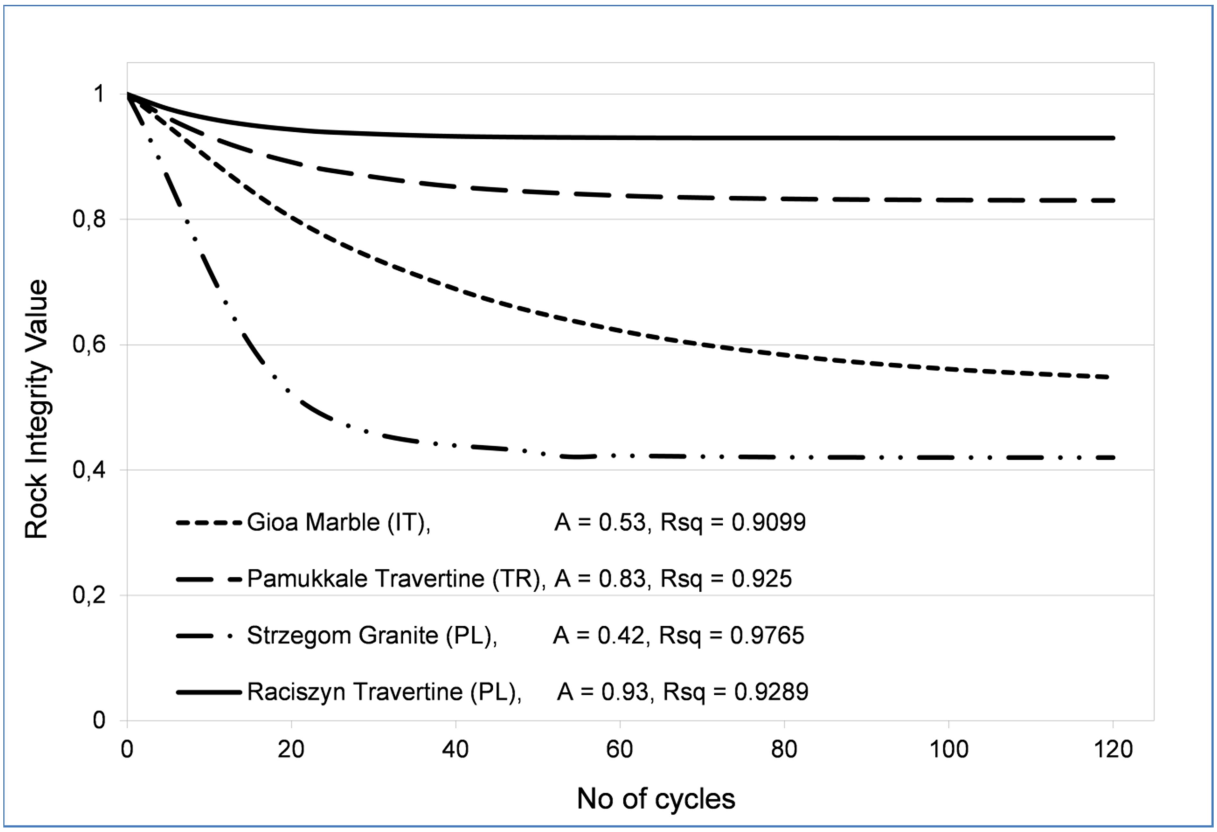

Fig. 11 Comparison of generalized decay functions of four stones under freeze and thaw action based on ultrasound R-wave (Marble) and P-wave (Granite, Travertine) velocity measurements

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