

**The Henryk Niewodniczański  
INSTITUTE OF NUCLEAR PHYSICS  
Polish Academy of Sciences  
152 Radzikowskiego str., 31-342 Kraków, Poland**

www.ifj.edu.pl/reports/2004.html  
Kraków, July 2004

---

**REPORT No 1945/PN**

**Sensitivity of the thermal neutron time decay to the hydrogen  
content in a rock sample**

*K. Drozdowicz, J. Dąbrowska, B. Gabańska, A. Igielski, W. Janik,  
E. Krynicka, A. Kurowski, K. Niedźwiedź, U. Wiącek, U. Woźnicka*

The work has been partly sponsored by the Polish Committee for Scientific Research  
in the frame of the Research Project No. 8 T12B 046 21 (2001 – 2004).

**Abstract**

A pulsed neutron method to measure the water or hydrogen content in a rock material has been tested on the experimental set-up at the fast neutron generator in the IFJ PAN. A dedicated pulsed thermal neutron source has been designed, built and added to this set-up. The test experiments have been done using dry crumbled granite as the rock matrix. The hydrogen content in samples has varied due to an addition of a defined amount of polyethylene. The time decay constant of the pulsed thermal neutron flux has been measured as a function of polyethylene content in granite+polyethylene samples. The experimental results have been supplemented with Monte Carlo simulations of the experiments. Analytical estimations of the time decay constant in the examined geometry have also been done. Difficulties of the proposed experimental method at low values of the hydrogen content are discussed. The proposed method using the pulsed neutron source to determine the hydrogen content which is less than 10 %, can be applied for rock samples of volume about 30 dm<sup>3</sup>. For higher hydrogen content the volume of the sample can be lower – about 7 dm<sup>3</sup>.

## 1. Introduction

The theoretical principles of a pulsed neutron method to measure the water or hydrogen content in a rock material were tested (Drozdowicz *et al.*, 2003c) on the experimental set-up at the fast neutron generator in the IFJ PAN. The time decay constant  $\lambda$  of the thermal neutron flux in the samples was measured as a function of the hydrogen content  $w$ . Dry crumbled granite was used as a rock material. The hydrogen content varied due to an addition of a defined amount of polyethylene (0 to 20 %). A few geometry systems (neutron source + sample) were tested to optimize the measured signal, *i.e.* the decay constant  $\lambda$  of the thermal neutron flux  $\varphi(t)$  in the sample.

The experimental set-up consisting of a special thermal neutron pulsed source and a cylindrical stainless steel container ( $H = 2R = 9.6$  cm) for the bulk sample was chosen as the best possible arrangement. The difficulty of realisation of the mentioned experiment was in obtaining a high thermal neutron flux in the sample (the rock sample, which contains a small amount of hydrogen, is a weak moderator of neutrons: if the fast neutron source is used, the thermal neutron field is very poor). The proposed system ensures the thermal neutron flux high enough in the sample.

Here in the report new series of the  $\lambda(w)$  experiments are presented. The experiments have been planned on the base of conclusions obtained in the paper mentioned above. The scheme of the chosen experimental geometry is shown in Fig.1. The elemental composition of the selected portion of granite has been determined by Geochemical Laboratory XRAL, Canada. Knowledge of the elemental composition has given us the possibility to compare the experimental results of the  $\lambda(w)$  measurements to the Monte-Carlo simulations and some analytical evaluations.

## 1. Samples

Samples of bulk granite (Granite S) from the Strzegom-Żbik deposit (Poland) have been used as basic rock material in experiments. In such type experiments with thermal neutrons the water content in the sample is fully equivalent to the hydrogen content in it. Thus, polyethylene,  $-\text{CH}_2-$ , has been chosen as the material sufficiently well simulating water,  $\text{H}_2\text{O}$ . Granulated polyethylene has been available as a technical product, Stavrolen™ (Russian production). The thermal neutron diffusion parameters have been experimentally tested for the portion of Stavrolen used in the experiments. The typical theoretical neutron data for pure

–CH<sub>2</sub>– can be used unreservedly (Drozdowicz et al., 2003c). The dried granulated Stavrolen has been mixed with the rock material in required proportions: in this way the hydrogen content has been well defined. This method ensures the homogeneous mixture of the components in the samples. The equivalence of the hydrogen and water contents to the 1 % of polyethylene content is presented in Table 1.

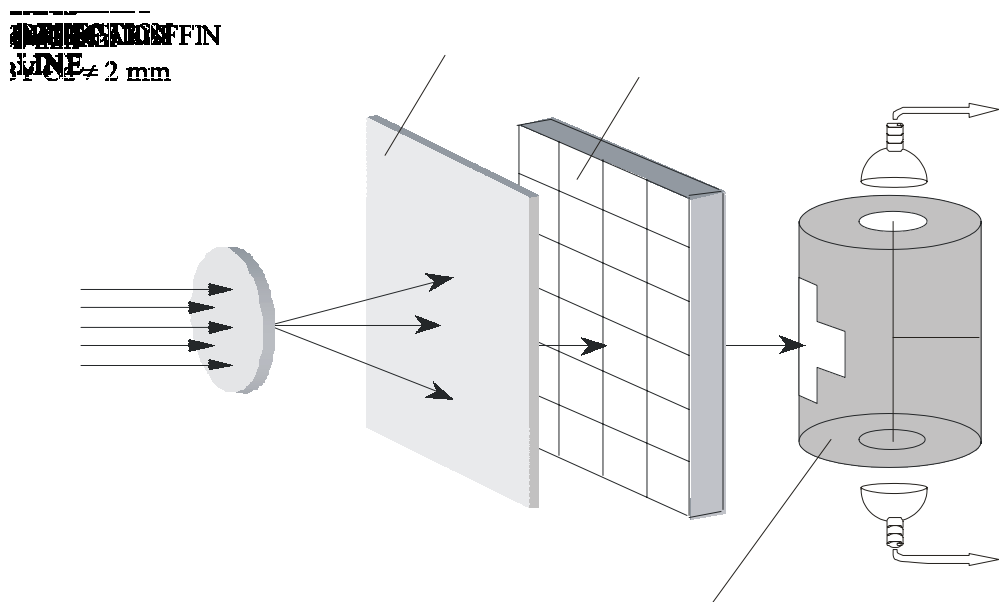


Fig.1. The experimental set–up with the thermal neutron pulsed source.

**Table 1.** Hydrogen and water contents equivalent to the 1 % polyethylene content.

Compound	Molecular mass [u]	Content [wt.%]
–CH <sub>2</sub> –	14.027	1.000
H <sub>2</sub>	2.016	0.144
H <sub>2</sub> O	18.015	1.284

The important physical and thermal neutron diffusion parameters of water, polyethylene, and granite, are presented in Table 2, where  $v_0 = 2200 \text{ ms}^{-1}$  is the most probable velocity of thermal neutrons, and  $\sigma(x)$  – here and in all subsequent tables – denotes the standard deviation of the  $x$  value. The table is supplemented with the parameters of quartz which is later used as a theoretical reference material.

**Table 2.** Thermal neutron and physical parameters of materials under study.

Material	Thermal neutron cross-sections		Solid material density $\rho$ $\sigma(\rho)$ [g cm <sup>-3</sup> ]	Granulation
	Absorption $\Sigma_a(\nu_0)$ $\sigma(\Sigma_a)$ [cm <sup>-1</sup> ]	Scattering $\Sigma_s(\nu_0)$ $\sigma(\Sigma_s)$ [cm <sup>-1</sup> ]		
H <sub>2</sub> O	0.02224 0.00005	~3.985 0.166	~1	—
–CH <sub>2</sub> –	0.02726 0.00006	~4.900 0.202	0.9495 <sup>P</sup> 0.0015	Spherical grains 2R ≈ 3 mm
Granite S	0.01050 0.00017	0.2901 0.0020	2.6381 <sup>P</sup> 0.0005	Sieve mesh 2R ≈ 0 ÷ 4 mm
SiO <sub>2</sub>	0.00455 0.00008	0.2541 0.0003	2.65	—

<sup>P</sup>) dried material measured in a helium pycnometer at 20 °C.

The thermal neutron diffusion parameters have been calculated with the SIGSA code (Drozdowicz and Krynicka, 1995), using a certain approximation for hydrogenous materials (Drozdowicz, 1998). The elemental composition of granite used for these calculations is specified in Table 3.

**Table 3.** Chemical composition of Granite S according to analysis by XRAL Laboratories Geochemical Exploration and Research Analysis (Canada) and recalculation to the elemental composition.

Chemical compound	Content [wt. %]	Element	Content [wt. %]
SiO <sub>2</sub>	74.45	O	49.0278
Al <sub>2</sub> O <sub>3</sub>	13.03	Si	34.85
CaO	1.22	Al	6.9
MgO	0.23	Fe	1.53
Na <sub>2</sub> O	3.39	Ca	0.8720
K <sub>2</sub> O	4.7	Mg	0.1387
Fe <sub>2</sub> O <sub>3</sub>	2.19	Na	2.515
MnO	0.04	K	3.9
TiO <sub>2</sub>	0.23	Ti	0.1380
P <sub>2</sub> O <sub>5</sub>	0.04	Mn	0.031
Cr <sub>2</sub> O <sub>3</sub>	<0.01	P	0.0175
LOI	0.45	H	0.05 <sup>*)</sup>

<sup>\*)</sup> if LOI = H<sub>2</sub>O

### 3. Measurements of the time decay constant $\lambda$ of granite+polyethylene samples

The measurements have been done on the experimental set-up shown in Fig. 1. The samples have been placed in the stainless-steel cylindrical container of the internal size:  $H = 2R = 9.6$  cm. The container has been enveloped by a 2 mm thick cadmium foil. Two round openings in the top and bottom cadmium shield have been used as the windows for the thermal neutron detectors. The thermal neutron decay curves have been registered by two independent multiscaler lines. The time decay constants  $\lambda$  have been determined independently from each detecting line. The mean values of  $\lambda$  obtained from the both lines are collected in Table 4. The experiments have been repeated few times for a given polyethylene content,  $w$ . The bulk density of samples of the given  $w$  value can slightly differ from one case to another. It happens when the whole sample is prepared of few times: a repetition of the same bulk density is, in principle, impossible.

The results are also presented in Fig.2. The twofold standard deviation  $2\sigma(\lambda)$  is marked as the uncertainty of the experimental results. The functional dependence  $\lambda(w)$  is difficult for analysing. The  $\lambda$  values for  $w = 0$  and  $w = 5$  wt.% is near the same. Then some maximum between 10 and 12 wt.% may be expected, and finally the decreasing slope of the curve is observed. The behaviour of the  $\lambda(w)$  function in the range of polyethylene content from 0 up to 10% should be tested with the denser step. However, it is experimentally very difficult, which was indicated in the report by Drozdowicz et al. (2003c).

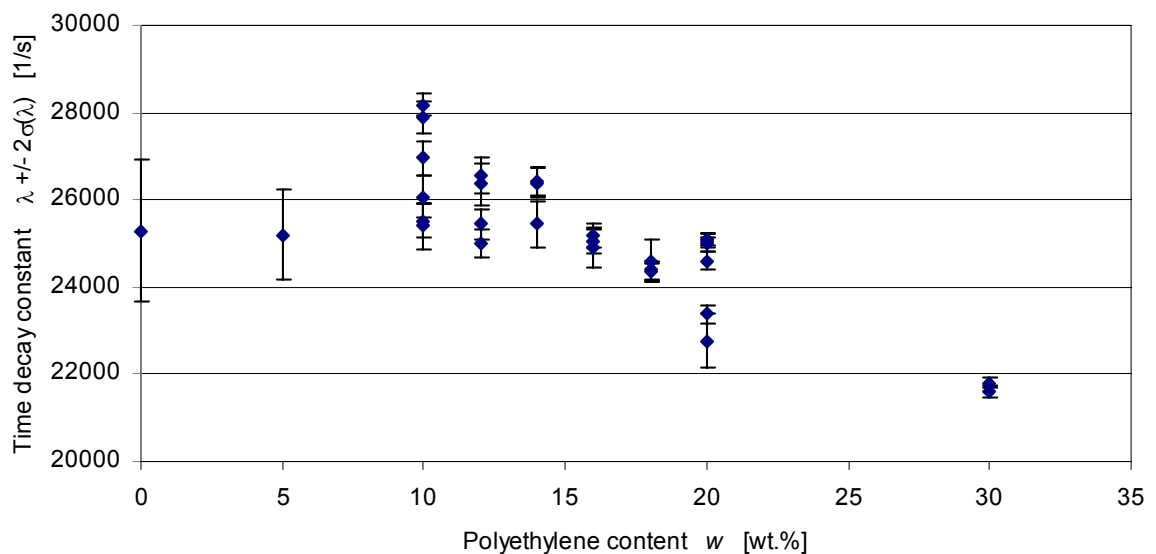


Fig.2. Experimental time decay constant  $\lambda$  vs. polyethylene content  $w$  in the Granite S bulk samples.

**Table 4.** Measured  $\lambda$  values in Granite S+polyethylene bulk samples.

Polyethylene content $w$ [wt. %]	Bulk density $\rho_B$ [g cm <sup>-3</sup> ]	Decay const. $\lambda \pm \sigma(\lambda)$ [s <sup>-1</sup> ]	Measurement code
30	1.079	21794 ± 59	01676
	1.079	21614 ± 66	01677
20	1.172	25084 ± 74	01622
	1.172	25013 ± 58	01624
	1.172	24607 ± 103	01625
	1.157	23372 ± 102	01652
	1.157	22761 ± 306	01654
	1.190	25028 ± 105	01663
18	1.206	24338 ± 108	01664
	1.206	24397 ± 101	01665
	1.234	24610 ± 237	01669
16	1.294	25174 ± 139	01659
	1.294	25057 ± 137	01660
	1.248	24912 ± 223	01668
14	1.276	26431 ± 158	01657
	1.276	26392 ± 173	01658
	1.301	25447 ± 265	01666
12	1.336	26558 ± 197	01661
	1.336	26355 ± 239	01662
	1.318	25441 ± 171	01672
	1.318	25009 ± 164	01673
10	1.430	25395 ± 264	01655
	1.430	25511 ± 196	01656
	1.336	28178 ± 131	01670
	1.336	27890 ± 189	01671
	1.381	26072 ± 237	01674
	1.381	26967 ± 196	01675
5	1.419	25203 ± 510	01617
0	1.479	25286 ± 810	01629

#### 4. Numerical simulation of thermal neutron flux in the granite+polyethylene samples

The experiments presented in the report are arduous and difficult especially if a good accuracy of the results is needed. The hard experimental conditions of the measurements are the reason why no more measurements have been done. The experiments have to be done at extreme pulsed neutron generator parameters to keep a very high fast neutron beam in order to

get the thermal neutron field sufficient for measurement. This is time- and target-consuming. In this situation, an additional support to the experimental data has been obtained by computer simulations of the measurements.

The thermal neutron transport in the investigated samples has been simulated using the numerical computer code MCNP (Briesmeister, 2000). The pulsed Maxwellian thermal neutron source has been assumed. The obtained numerical data have been fully comparable with the thermal neutron decay curves registered in the real experiments. The  $\lambda_{\text{MCNP}}$  values have been calculated using the same computer software as for the interpretation of the real experiment. The results are presented in Table 5 and in Fig. 3. The results for simulations for the sample of the size  $H = 2R = 16$  cm are discussed in the Conclusions.

**Table 5.** Thermal neutron time decay constants obtained from the MCNP simulations for the granite+polyethylene samples.

Polyethylene cont. $w$ [wt. %]	Density $\rho$ [ $\text{g cm}^{-3}$ ]	Decay const. $\lambda_{\text{MCNP}} \pm \sigma(\lambda)$ [ $\text{s}^{-1}$ ]	Measurement code
$H = 2R = 9.6$ cm			
20.0	1.065	$24278 \pm 40$	sgpe020
	1.190	$23427 \pm 41$	s1000q04
10.0	1.381	$26301 \pm 64$	sgpe023
5.0	1.419	$25088 \pm 167$	s1000q03
	1.419	$24802 \pm 180$	sgpe024
3.0	1.561	$23988 \pm 149$	sgpe026
2.0	1.554	$22958 \pm 167$	sgpe025
0.0	2.638	$26925 \pm 91$	sgpe022
	1.480	$23117 \pm 168$	sgpe021
$H = 2R = 16$ cm			
20.0	1.157	$11904 \pm 18$	s1000q04d
10.0	1.382	$14160 \pm 43$	s1000q04c
8.0	1.380	$14998 \pm 32$	s1000q03d
6.0	1.370	$15818 \pm 42$	s1000q04b
	2.000	$14548 \pm 19$	s1000q03c
5.0	1.428	$16056 \pm 41$	s1000q02d
	2.000	$15162 \pm 41$	s1000q02c
4.0	1.500	$16220 \pm 63\text{a)}$	s1000q03b
	1.500	$16223 \pm 63\text{a)}$	s1000q02b
3.5	1.535	$16239 \pm 77$	s1000q04a
3.0	1.561	$16311 \pm 47$	s1000q02a
2.0	1.554	$16536 \pm 68$	s1000q03a
0.0	1.440	$16082 \pm 35$	s1000q05
	2.638	$17378 \pm 76$	s1000q04e

a) using different time widths of the source square pulse

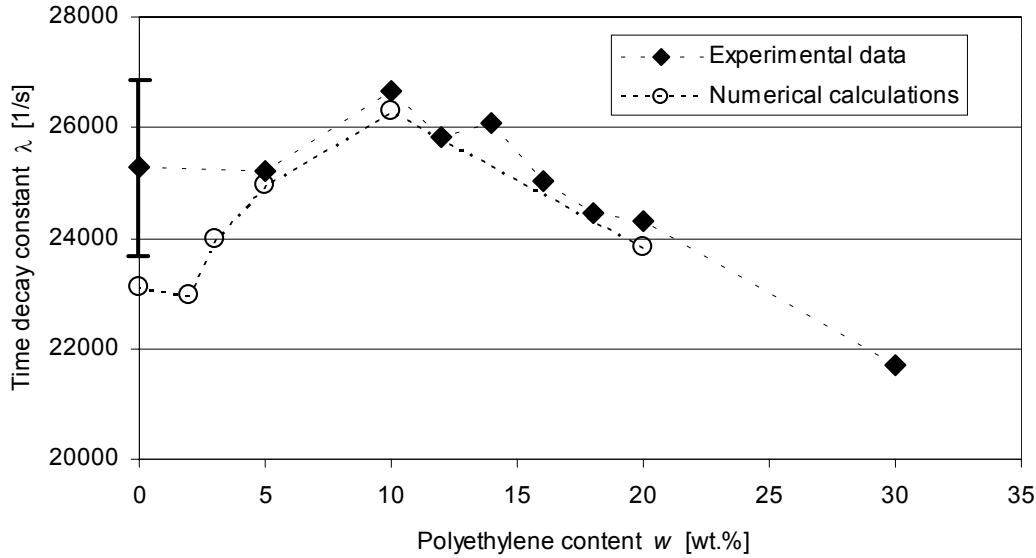


Fig.3. Comparison of the time decay constant  $\lambda$  vs. polyethylene content  $w$  from the real and simulated experiments (samples  $H = 2R = 9.6$  cm).

The numerical results confirmed the trend of the curve  $\lambda(w)$  and fill up the range of low  $w$  values showing the strong changeability of the function. The MCNP calculations for the polyethylene content  $w = 0$  and  $w = 20$  wt.% (in the sample  $H = 2R = 9.6$  cm) have been repeated twice at two different densities. The obtained results testified that the spread of the  $\lambda$  values for  $w = \text{const}$  cannot be explained as a dependence of a change of sample bulk density only. In the case of  $w = 0$  the decay constant  $\lambda$  is higher for the higher density, contrary to the case for the  $w = 20$  wt.%. The observed differences are caused by the complex problem of the thermal neutron transport in the bounded medium having different hydrogen contents.

## 5. Analytical estimation of the $\lambda(w)$ function for the granite+polyethylene samples

The  $\lambda(w)$  function can be estimated on the base of the diffusion approximation of the thermal neutron transport in bounded media when the pulsed neutron diffusion parameters are known. Unfortunately, there are no sufficient theoretical and/or experimental data for such kind of mixtures as used in the discussed experiment, i.e. for rock + water (or other hydrogenous compound). Only sparse data, generally based on the numerical simulation of the buckling experiments are available (e.g. for moisturized quartz or dolomite (Drozdowicz et al., 2002b, 2003b)).



### 5.1. Thermal neutron diffusion parameters for the granite+polyethylene medium

The theoretical  $\lambda$  values in a bounded medium can be calculated according to the formula:

$$\lambda = \alpha + D_0 B^2 - C B^4 + F B^6, \quad (1)$$

where  $\alpha \equiv \langle \nu \Sigma_a \rangle$  is the thermal neutron absorption rate,  $D_0$  is the diffusion constant,  $C$  is the diffusion cooling coefficient, and  $B^2$  is the geometric buckling of the sample. Parameter  $F$  includes some corrections to the  $C$  value.

The absorption rate for rock and for mixtures rock+polyethylene can be exactly calculated basing on the elemental composition of the medium. The  $D_0$  and  $C$  values can be calculated from the formulae existing only for the dry rock (Drozdowicz et al., 2002a). Neither analytical formulae nor experimental data are known for the given mixture Granite S +hydrogenous component.

The determination of the neutron diffusion parameters  $D_0$  and  $C$  for the mixture of two types of ingredients – one containing hydrogen bounded in the molecule and a second one which is built of heavier elements – is difficult both from the theoretical and experimental points of view. The determination of the  $D_0(w)$  and  $C(w)$  dependencies has been first time done by Drozdowicz et al. (2002b, 2003b) for moisturized dolomite and quartz by the Monte-Carlo simulation of the pulsed neutron experiment (the variable buckling experiment) with the method used previously for dry rocks (Drozdowicz et al., 2003a).

In order to estimate the  $D_0(w)$  and  $C(w)$  parameters for the mixture of Granite S +polyethylene we can adopt the results obtained for the moisturized quartz. Both the  $D_0$  and  $C$  values are dependent on the thermal neutron scattering cross-section. The scattering cross-sections of granite and quartz are nearly the same. Water and polyethylene have this parameter close to each other if compared to the low value for granite (Table 2).

For the simplicity of calculations the so-called density-removed thermal neutron diffusion parameters (cf. Granada et al., 1987; Czubek, 1997) are used:

$$\langle \nu \Sigma_a \rangle^M = \rho^{-1} \langle \nu \Sigma_a \rangle, \quad D_0^M = \rho D_0, \quad C^M = \rho^3 C, \quad F^M = \rho^5 F. \quad (2)$$

The  $D_0^M$  and  $C^M$  parameters for  $\text{SiO}_2$  and Granite S have been calculated according to the formulae given by Drozdowicz et al. (2002a) and the results are presented in Table 6.

**Table 6.** Density-removed diffusion parameters for the media of interest.

	SiO <sub>2</sub>	Granite S
$D_0^M$ [cm <sup>2</sup> s <sup>-1</sup> (g cm <sup>-3</sup> )]	817 500	801 800
$\sigma(D_0^M)$	±6 400	
$C^M$ [cm <sup>4</sup> s <sup>-1</sup> (g cm <sup>-3</sup> )]	39 400 000	31 935 000
$\sigma(C^M)$	±1 360 000	

The relevant values of the parameters for given mixtures, Granite S+polyethylene, have been estimated using the following approximations:

$$[D_0^M(w)]_{\text{Granite S}} = \frac{(D_0^M)_{\text{Granite S}}}{(D_0^M)_{\text{SiO}_2}} [D_0^M(w)]_{\text{SiO}_2} = 0.98 [D_0^M(w)]_{\text{SiO}_2} \quad (3)$$

$$[C^M(w)]_{\text{Granite S}} = \frac{(C^M)_{\text{Granite S}}}{(C^M)_{\text{SiO}_2}} [C^M(w)]_{\text{SiO}_2} = 0.81 [C^M(w)]_{\text{SiO}_2} \quad (4)$$

where the values for the SiO<sub>2</sub> + H<sub>2</sub>O mixtures have been taken from the paper by Drozdowicz et al. (2003b). The possible differences resulted from the differences between the  $D_0$  and  $C$  parameters for water and polyethylene have been neglected. The neutron diffusion parameters as a function of  $w$  for the Granite S+polyethylene mixtures are collected in Table 7.

**Table 7.** Estimated density-removed thermal neutron diffusion parameters for the Granite S+polyethylene mixtures.

$w$ [wt.%]	$\langle v\Sigma_a \rangle^M(w)$ [s <sup>-1</sup> /(g cm <sup>-3</sup> )]	$D_0^M(w)$ [cm <sup>2</sup> s <sup>-1</sup> (g cm <sup>-3</sup> )]	$C^M(w)$ [cm <sup>4</sup> s <sup>-1</sup> (g cm <sup>-3</sup> ) <sup>3</sup> ]
0	875	801 800	31 935 000
2	984	539 200	8 608 000
4	1093	413 600	4 089 000
6	1202	335 000	2 281 000
8	1311	281 000	1 367 000
10	1420	243 100	927 000
20	1963	144 200	219 000

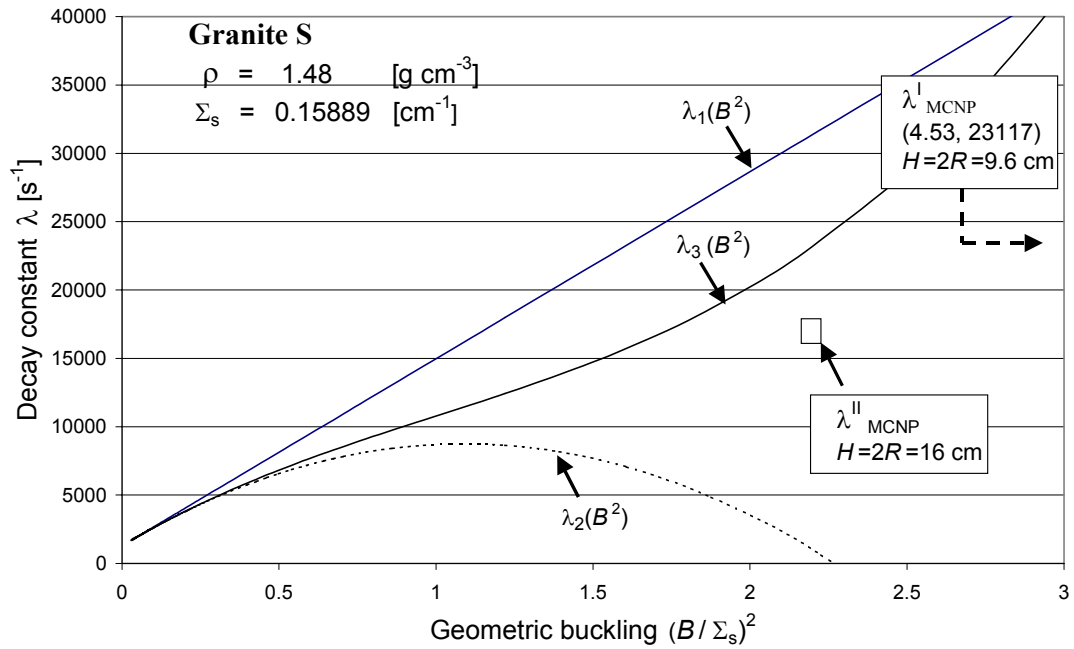
The term  $FB^6$  has to be used in Eq.(1) in some cases when the geometric buckling reaches high values. The coefficient  $F$  has no special physical interpretation in the thermal neutron transport theory and is treated as a correction term to the diffusion cooling coefficient. Its estimated values are obtained from the fit of expression (1) to real or simulated experimental data as was done e.g. for the basic rock minerals (Drozdowicz et al., 2003a). There are neither theoretical nor experimental expectations for this value for the tested mixture of Granite S+polyethylene.

Some check calculations of the function  $\lambda = \lambda(B^2)$  have been done with and without the term  $F$ . The value  $F_{\text{SiO}_2}^{\text{M}} = 922 \cdot 10^6 \text{ [cm}^6\text{s}^{-1}(\text{g cm}^{-3})^5]$ , which has been estimated from the simulated buckling experiment for  $\text{SiO}_2$  (Drozdowicz et al., 2003b), has been used here in the first approximation. Two examples of the function  $\lambda(B^2)$  are presented in Fig. 4. Both sets of curves have been calculated for the Granite S: once for the solid density  $\rho = 2.638 \text{ g cm}^{-3}$  (Fig.4a) and second for the bulk density  $\rho = 1.48 \text{ g cm}^{-3}$  (Fig.4b). The difference in densities (of the same material) involves the significant differences in the neutron scattering properties of the medium. An expression of the geometric buckling  $B^2$  in units of the scattering mean free path  $(Bl_s)^2 = (B / \Sigma_s)^2$  makes possible a direct comparison of the  $\lambda(B^2)$  functions at different material densities.

Three curves have been calculated at each density:  $\lambda_1(B^2)$  – pure diffusion approximation,  $\lambda_2(B^2)$  – including the diffusion cooling coefficient  $C$ , and  $\lambda_3(B^2)$  - the full development given in Eq.(1), i.e. introducing the correction  $F$ , important for very small samples. (Note: the small sample in that discussion means the size which is comparable to the scattering mean free path in the given medium).

The values obtained from the Monte-Carlo simulations for the Granite S samples of two sizes ( $\lambda_{\text{MCNP}}^{\text{I}}$  for  $H = 2R = 9.6 \text{ cm}$ , and  $\lambda_{\text{MCNP}}^{\text{II}}$  for  $H = 2R = 16 \text{ cm}$ ) are marked on the plots.

a)



b)

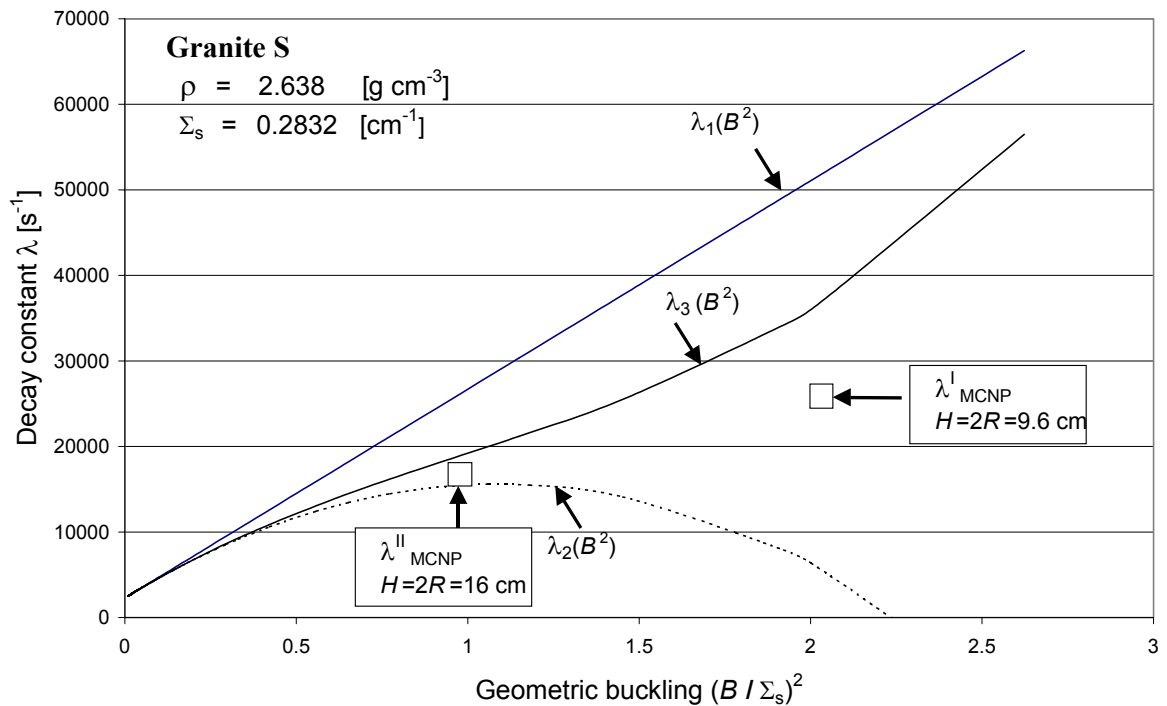


Fig.4. The  $\lambda(B^2)$  functions for Granite S: a) for the density  $\rho = 2.638 \text{ g cm}^{-3}$ ; b) for the density  $\rho = 1.48 \text{ g cm}^{-3}$ . The  $\lambda_{MCNP}$  results (see Table 5) are marked in the plots. The neutron data of Granite S are taken from Table 7 for  $w = 0$ . Parameter  $F = F_{SiO_2}$ .

The plots in Fig.4a correspond to the rock sample of the solid material density. The experimentally expected  $\lambda_{MCNP}^{\text{II}}$  value for the larger sample is situated in the point when the discrepancy between the  $\lambda_2(B^2)$  and  $\lambda_3(B^2)$  curves starts to be visible. The position of the  $\lambda_{MCNP}^{\text{I}}$  point for the smaller sample implies that the  $F$  parameter has to be used as the important correction to the  $C$  value. Plots in Fig.4b show analogous examples – also for Granite S – but for a smaller density which corresponds to the typical bulk density obtained for a loose sample. The mean free path is longer in a medium of a smaller density. Therefore, the respective  $\lambda_{MCNP}$  points are shifted to the right, to the larger values of the  $(B / \Sigma_s)^2$ .

The positions of the  $\lambda_{MCNP}$  values and the  $\lambda_3(B^2)$  curves imply that the  $F$  parameter assumed in these calculations is too high. The set of curves  $\lambda(w)$  calculated on the base of the functions  $\lambda_{1,2}(B^2)$  and  $\lambda_3(B^2)$  at different values of parameter  $F$ ,

$$[F^M(w)]_{\text{Granite S}} = k_F [F^M(w)]_{\text{SiO}_2}, \quad 0.5 \leq k_F \leq 2.0 \quad (5)$$

is plotted in Fig.5. The shape of the curve  $\lambda(w)$  suggested by the experimental and simulated data can be achieved when the  $F$  parameter is taken into consideration. The data of  $[F^M(w)]_{\text{Granite S}}$ , which are chosen for a further consideration, are presented in Table 8 They have been obtained on the base of the results for moisturized  $\text{SiO}_2$  and for  $k_F = 0.5$ .

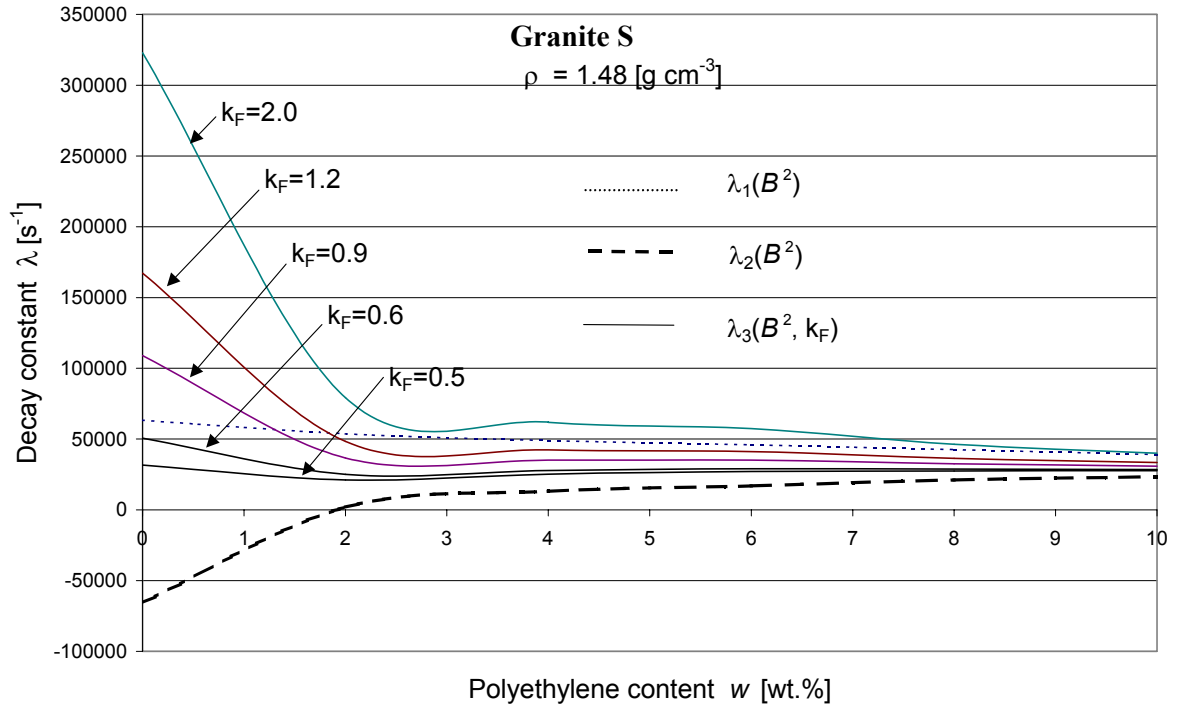


Fig.5. Function  $\lambda(w)$  calculated for the Granite S+polyethylene samples ( $H = 2R = 9.6$  cm) on the base of the estimated neutron parameters. The variation of the shape of the curves corresponds to the variation of the  $F$  parameter:  $F = k_F F_{\text{SiO}_2}$ .

**Table 8.** Estimation of parameter  $F^M$  for the Granite S+polyethylene medium.

$w$ [wt.%]	$F^M(w)$ [cm <sup>6</sup> s <sup>-1</sup> (g cm <sup>-3</sup> ) <sup>5</sup> ]
0	922·10 <sup>6</sup>
2	103·10 <sup>6</sup>
4	364·10 <sup>5</sup>
6	165·10 <sup>4</sup>
8	748·10 <sup>4</sup>
10	445·10 <sup>4</sup>
20	67·10 <sup>4</sup>

## 5.2. Comparison of the experimental, simulated and theoretical $\lambda(w)$ results

The comparison of the data obtained for the function  $\lambda(w)$  is presented in Fig. 6. A complicated course of the curve for the low  $w$  values has been followed out by the theoretical consideration.

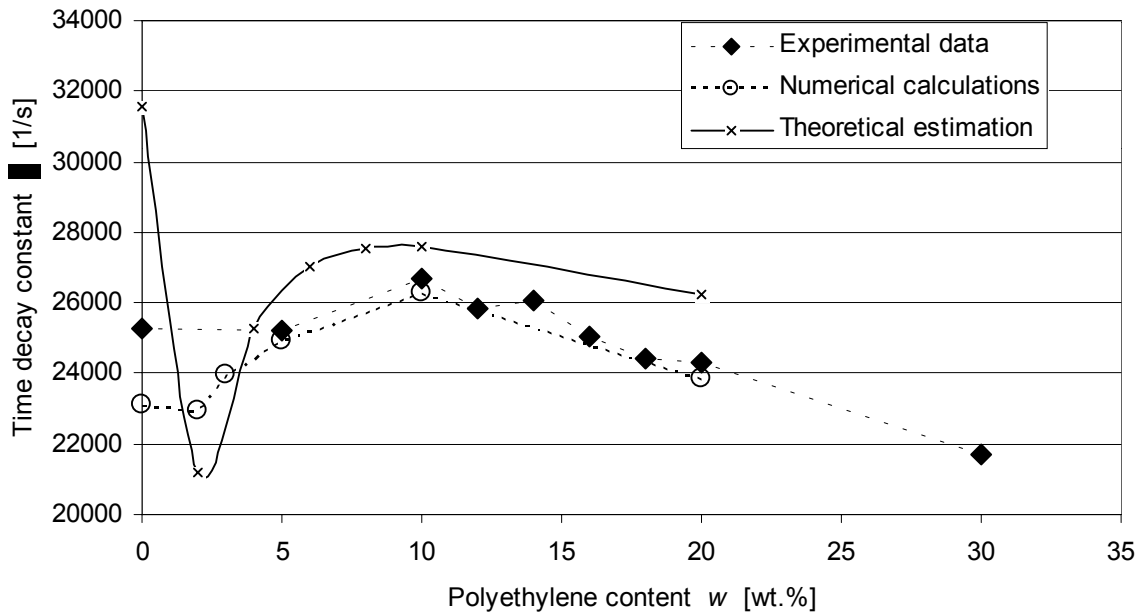


Fig.6. Comparison of the theoretical, experimental and simulated prediction of the  $\lambda(w)$  function for the Granite S+polyethylene sample (size  $H = 2R = 9.6$  cm).

A better conformity of the results should be obtained when the better prediction of the thermal neutron diffusion data for the Granite S+polyethylene media are done. This is possible by the numerical simulation of the buckling experiments for a large set of the Granite S+polyethylene samples. It seems not important at the present stage of the elaboration of the task.

## 6. Conclusions

The problem of proper determination of the  $\lambda(B^2)$  function presented above for the dry rock samples extends on samples containing a small amount of hydrogen. The complex shape of the  $\lambda(w)$  function in the range of  $0 < w < 10$  wt.% results from the thermal neutron diffusion process in small samples at a varying hydrogen content. This causes that the  $\lambda(w)$  function is not monotonical for small contents of polyethylene in the rock material. This conclusion is in agreement with the previous theoretical estimations (Drozdowicz et al., 2002a) made for spherical rock samples of 10 cm radius. From those preliminary calculations the important role of the diffusion cooling coefficient for any neutron experiments with dry rock samples was concluded. Here the experimental and numerical confirmation is obtained.

A higher hydrogen (water/polyethylene) content in the rock sample causes that the neutron scattering characteristics of the medium are dominated by the hydrogen scattering properties. This was confirmed for moisturized dolomite (Drozdowicz et al., 2002b) and quartz (Drozdowicz et al., 2003b). The amount of about 10 % of water in the rock material significantly brings the  $D_0$  and  $C$  parameters nearer to those for water.

If the monotonical run of the  $\lambda(w)$  curve is expected for samples containing less than 10 % of water, bigger samples should be used. The example of  $\lambda_{\text{MCNP}}(w)$  curves for two Granite S+polyethylene samples of different sizes is presented in Fig. 7.

One can expect that a large difference between neutron scattering properties of hydrogen and of a typical rock material gives the possibility to estimate hydrogen (water and/or polyethylene) content  $w$  from the time decay constant  $\lambda$  measured in a moisturized sample. Unfortunately, weak abilities of rock material to scatter thermal neutrons cause difficulties in a realization of pulsed thermal neutron experiment.

Thermal neutron field in the sample of interest is insufficient if  $w$  is low. The primary fast neutron pulsed source has to be high to generate the enough high thermal neutron field in the sample. The application of the proposed thermal neutron pulsed source (paraffin in the Cd grid) is very interesting solution for such a kind of experiments. The construction of the source can be still better optimized, but no spectacular improvement may be achieved in

comparison to the obtained neutron yield.

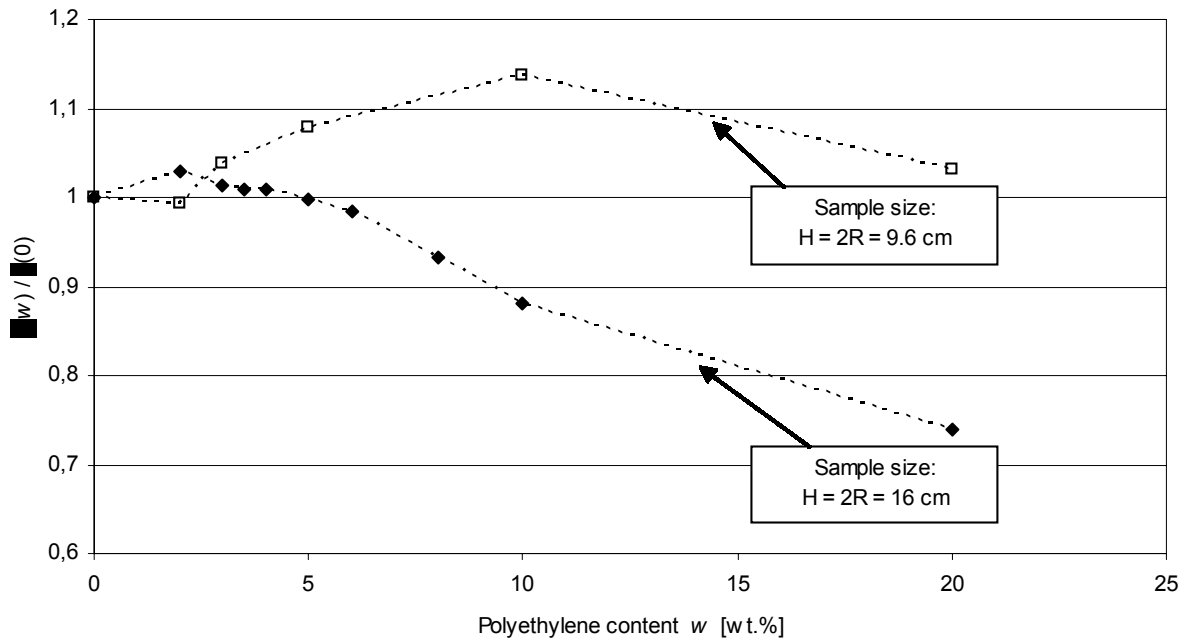


Fig.7. The  $\lambda(w)$  dependence for two Granite S+polyethylene samples of different sizes. Results from the numerical simulations (MCNP).

The time decay constant  $\lambda$  (measured in bulk rock samples of volume about  $7 \text{ dm}^3$  and containing less than 10 % of water) is described by a very complicated function of the pulsed diffusion neutron parameters  $\lambda(B^2)$ . The proper interpretation of the water content from the measured  $\lambda$  value requires knowledge of the theoretical prediction of the  $\lambda(w)$  function, *i.e.* knowledge of the pulsed neutron parameters  $\Sigma_a$ ,  $D_0$ ,  $C$ ,  $F$  of the given rock material. From those parameters only  $\Sigma_a$  can be calculated from the elemental composition or known from the laboratory measurement (using e.g. Czubek's method). The others, especially their dependence on hydrogen content is known in very limited cases only from the Monte-Carlo simulation of the pulsed experiments.

At the present stage of investigation of the possibility to determine water content  $w$  by the measurement of  $\lambda$  value, the final conclusions are:

1. For the water content  $w < 10$  % the bulk sample volume should not be less than  $30 \text{ dm}^3$ . Sample of the volume about  $7 \text{ dm}^3$  is sufficient if some pressing procedure were applied to increase the material density to  $\rho > 2.5 \text{ g cm}^{-3}$ .
2. If the water content  $w > 10$  %, the proposed measurement method gives the acceptable results for samples of volume about  $7 \text{ dm}^3$ . Some optimization of the measurement method is still possible.



Regardless of the problem of the measurement of the hydrogen content in rocks, the research done during this investigation gives numerous important informations and posed interesting questions in the matter of the thermal neutron transport in media of weak scattering properties. The role of the  $F$  parameter in describing the diffusion process of thermal neutron in bounded media should be further continued. Thermal neutron diffusion pulsed experiments on small bulk samples (*i.e.* of sizes of a few diffusion lengths) are very helpful in an elaboration of the theoretical consideration of neutron transport in media. The Monte Carlo calculations of the neutron transport process are very useful tool provided that neutron data are accurate enough. The time decay constant, which can be measured with a high accuracy, is a very sensitive tool in testing of analytical solutions of neutron transport phenomena or in testing of different numerical simulations of those processes.

## References

- Briesmeister J.F. (Ed.) (2000), MCNP – A general Monte Carlo n-particle transport code Version 4C. LA-13709-M Manual. Los Alamos National Laboratory.
- Czubek J.A. (1997), A new idea of pulsed neutron experiments with bulk materials. *Appl. Radiat. Isot.* **48**, 237-239.
- Drozdowicz K. (1998), A method to calculate thermal neutron diffusion parameters for hydrogenous mixtures. *Nucl. Instrum. Meth.* **A 411**, 121-129.
- Drozdowicz K., Gabańska B., Woźnicka U. (2002a), Possibility of  $\Sigma_s$  measurement on rock samples by a pulsed neutron method. Rept. INP No. **1899/PN**, Institute of Nuclear Physics, Kraków. <http://www.ifj.edu.pl/reports/2002.html>
- Drozdowicz K., Krynicka E. (1995), Thermal neutron diffusion parameters in homogeneous mixtures. Rept. INP No. **1694/PN**, Institute of Nuclear Physics, Kraków, 1995.
- Drozdowicz K., Krynicka E., Dąbrowska J. (2002b), Influence of the water content in rock on the thermal neutron diffusion and diffusion cooling coefficients (by Monte Carlo simulations). I: - Dolomite. Rept. INP No. **1917/PN**, Institute of Nuclear Physics, Kraków. <http://www.ifj.edu.pl/reports/2002.html>
- Drozdowicz K., Krynicka E., Dąbrowska J. (2003a), Diffusion cooling of thermal neutrons in basic rock minerals by Monte Carlo simulation of the pulsed neutron experiments. *Appl. Radiat. Isot.* **58**, 727-733.
- Drozdowicz K., Krynicka E., Dąbrowska J. (2003b), Influence of the water content in rock on the thermal neutron diffusion and diffusion cooling coefficients (by Monte Carlo simulations). II: - Quartz. Rept. INP No. **1933/PN**, Institute of Nuclear Physics, Kraków. <http://www.ifj.edu.pl/reports/2003.html>
- Drozdowicz K. and Drabina A., Dąbrowska J., Gabańska B., Igielski A., Janik W., Krynicka E., Kurowski A., Wiącek U., Woźnicka U. (2003c), Feasibility study of experimental detection of the hydrogen content in rock material by a pulsed neutron method. Rept. INP No. **1935/PN**, Institute of Nuclear Physics, Kraków. <http://www.ifj.edu.pl/reports/2003.html>
- Granada J.R., Gillette V.H., and Mayer R.E. (1987): Calculation of neutron cross sections and thermalization parameters for molecular gases using a synthetic scattering function. II. Applications to H<sub>2</sub>O, D<sub>2</sub>O, and C<sub>6</sub>H<sub>6</sub>. *Physical Review A*, **36**, No 12, 5594-5605.