The pulsed neutron experiment (the variable geometric buckling experiment) in spherical geometry has been simulated using the MCNP code. The time decay of the thermal neutron flux has been observed as a function of the sample size. The thermal neutron diffusion cooling coefficient $C$ with the correction $F$ has been determined for three basic rock minerals at the given specific densities $\rho$:

- Quartz ($\text{SiO}_2$, $\rho = 2.65 \text{ g/cm}^3$): $C = 2 117 000 \text{ cm}^4\text{s}^{-1}$, $F = 7 060 000 \text{ cm}^6\text{s}^{-1}$
- Calcite ($\text{CaCO}_3$, $\rho = 2.71 \text{ g/cm}^3$): $C = 1 341 000 \text{ cm}^4\text{s}^{-1}$, $F = 3 610 000 \text{ cm}^6\text{s}^{-1}$
- Dolomite ($\text{CaMg(CO}_3)_2$, $\rho = 2.87 \text{ g/cm}^3$): $C = 778 000 \text{ cm}^4\text{s}^{-1}$, $F = 1 380 000 \text{ cm}^6\text{s}^{-1}$.

The corresponding density-removed parameters have been also obtained.
1. Introduction

The diffusion cooling effect appeared significant in considerations of the pulsed thermal neutron flux in finite rock media [1]. Unfortunately, no data on the subject exist. A certain approximation for the diffusion cooling coefficient was assumed in the mentioned theoretical study. In this report we present Monte Carlo simulations of the pulsed neutron experiments for basic rock minerals (quartz, calcite, dolomite). The variable geometric buckling experiments have been simulated in the spherical geometry. The decay constant \( \lambda \) of the fundamental exponential mode, \( e^{-\lambda t} \), of the thermal neutron flux decaying in time in a given finite sample is related to the neutron parameters of the medium and the size of the system

\[
\lambda = \langle \nu \Sigma_a \rangle + D_0 B^2 - C B^4 + F B^6 - \ldots ,
\]

where \( \langle \nu \Sigma_a \rangle \) is the absorption rate (\( \Sigma_a \) – the macroscopic absorption cross section, \( \nu \) – the thermal neutron speed), \( D_0 \) is the diffusion constant, \( C \) is the diffusion cooling coefficient, and \( F \) is a correction term. The geometric buckling \( B^2 \) for the spherical system is defined as

\[
B^2 = \left( \frac{\pi}{R} \right)^2 ,
\]

where \( R \) is the extrapolated radius,

\[
R = R_g + d_{\text{sph}},
\]

i.e. the geometric radius \( R_g \) increased by the extrapolation length \( d_{\text{sph}} \). At the spherical surface, the latter can be expressed by

\[
d_{\text{sph}} = d \left( \frac{1}{1 + \frac{d}{R_g}} \right) ,
\]

where \( d \) is the extrapolation length defined in the Milne problem for the thermal neutron vacuum boundary conditions at the half-space with the flat boundary surface:

\[
d = 0.71 \langle l_{\text{tr}} \rangle = 2.13 \langle D \rangle = 2.13 \langle 1/\nu \rangle D_0 .
\]

In Eq.(5), the dependences between the thermal neutron transport mean free path \( l_{\text{tr}} \), the
diffusion coefficient $D$, and the diffusion constant $D_0$ are utilised, and the bracket $\langle \rangle$ denotes the values averaged over the thermal neutron flux energy spectrum, described by the Maxwellian distribution (cf. [2], [3], [4]).

2. Simulated pulsed-neutron experiment

In the variable geometric buckling experiment, the thermal neutron flux decaying in time is registered for a series of samples of different sizes, i.e. of different bucklings $B_i^2$. The fundamental mode decay constant $\lambda_i$ is found in each case, and the function defined in Eq.(1) is fitted to the obtained set of the experimental data, $(B_i^2, \lambda_i)$. The thermal neutron parameters are then determined. The geometric buckling depends through the extrapolation length on the diffusion constant $D_0$ being just determined, Eqs (2) to (5), and an iterative fit has to be performed until a convergence of the diffusion constant is achieved.

A real experiment with a rock material is extremely difficult both due to the problem of the bulk density of individual samples [5] and to a possible varying contamination with trace elements in different portions of the material. The Monte Carlo method was found very helpful for simulations of the pulsed neutron experiments [6], [7]. In such simulation a pure rock material can be assigned, keeping exactly the same elemental composition and mass density for all samples.

The ideal pulsed neutron experiment for the rocks has been simulated by using the MCNP 4B code [8]. The thermal neutron source has been used. In order to simulate a square neutron burst, neutrons have been generated within a 100 $\mu$s interval with a constant probability. Their initial positions $r$ have been sampled from the probability density function $p(r) \sim r^2$ for $r \leq R_g$, which means that neutrons are uniformly generated inside the sample. This corresponds to the conditions in a source-free medium after the neutron burst. The source has been isotropic. The initial neutron energies have been sampled from the Maxwellian distribution at the room temperature, $kT = 0.0253$ eV. For lack of the $S(\alpha,\beta)$ thermal scattering cross sections for the materials of the rocks being investigated, the free gas model of scattering has been applied, which is fully relevant for the contributing elements, relatively heavy for the neutron collisions. The cross sections have been taken from the rmecs, endf5u, and endf5p standard MCNP tables.
In order to determine the time decay constant $\lambda$ for a given sample, the thermal neutron flux has been scored in time intervals after the source pulse. The number of time channels and their widths have been adjusted individually for each sample, according to the rate of the time decay of the neutron flux. The number of histories in the MCNP runs has been chosen for each sample to assure the relative error of the thermal neutron flux not exceeding 10 % for the smallest contents in the time channels (i.e. in the final part of the registered curve).

Three rock minerals have been investigated: quartz – SiO$_2$, calcite – CaCO$_3$, and dolomite – CaMg(CO$_3$)$_2$. The range of the geometric buckling and the total number of the samples have been chosen individually for each rock, from an observation of values of the consecutive decay constants, depending on the thermal neutron parameters of a particular rock.

Fits of Eq.(1) to the simulated experimental results have been performed essentially neglecting terms $O(B^8)$, i.e. not only the diffusion cooling coefficients $C$ but also the corrections $F$ have been determined. Some questions on the relation between the thermal neutron parameters and the coefficients resulting from the fitting procedure were discussed in [7]. Therefore, here in all cases the fits without the terms $O(B^6)$ have been also tested.

The thermal neutron absorption in a substance of the well defined elemental composition does not create any doubts. Therefore, the absorption rates $\langle \nu \Sigma_a \rangle$ of the considered minerals have been calculated from their stoichiometric formulae, their mass densities $\rho$, and the microscopic absorption cross sections [9] of the contributing elements, using a computer code [10]. Thus, this parameter has been assumed as known in the fits.

3. Thermal neutron parameters of quartz

Simulations of the variable geometric buckling experiment for quartz, SiO$_2$, have been performed under the following conditions:
– the rock density $\rho = 2.65$ g cm$^{-3}$,
– the range of the radius of spheres $R_8 = (8 \div 45)$ cm.

The decay constants $\lambda$ of the fundamental mode thermal neutron flux have been obtained from the registered decaying curve in the same way as is used at the interpretation of real pulsed experiments [11]. The results are shown in Table 1.
Table 1. Decay constants $\lambda$ obtained from the simulated experiment for quartz.

<table>
<thead>
<tr>
<th>$R_g$ [cm]</th>
<th>$\lambda$ [s$^{-1}$]</th>
<th>$\sigma(\lambda)$ [s$^{-1}$]</th>
<th>$R_g$ [cm]</th>
<th>$\lambda$ [s$^{-1}$]</th>
<th>$\sigma(\lambda)$ [s$^{-1}$]</th>
<th>$R_g$ [cm]</th>
<th>$\lambda$ [s$^{-1}$]</th>
<th>$\sigma(\lambda)$ [s$^{-1}$]</th>
<th>$R_g$ [cm]</th>
<th>$\lambda$ [s$^{-1}$]</th>
<th>$\sigma(\lambda)$ [s$^{-1}$]</th>
</tr>
</thead>
<tbody>
<tr>
<td>8.0</td>
<td>17 653</td>
<td>259</td>
<td>9.5</td>
<td>15 148</td>
<td>135</td>
<td>12.0</td>
<td>11 849</td>
<td>42</td>
<td>18.0</td>
<td>7 327</td>
<td>29</td>
</tr>
<tr>
<td>8.2</td>
<td>17 175</td>
<td>198</td>
<td>10.0</td>
<td>14 348</td>
<td>145</td>
<td>13.0</td>
<td>10 790</td>
<td>21</td>
<td>20.0</td>
<td>6 387</td>
<td>22</td>
</tr>
<tr>
<td>8.5</td>
<td>16 652</td>
<td>221</td>
<td>10.5</td>
<td>13 653</td>
<td>121</td>
<td>14.0</td>
<td>9 879</td>
<td>55</td>
<td>25.0</td>
<td>4 756</td>
<td>9</td>
</tr>
<tr>
<td>9.0</td>
<td>15 913</td>
<td>206</td>
<td>11.0</td>
<td>13 063</td>
<td>31</td>
<td>16.0</td>
<td>8 468</td>
<td>33</td>
<td>45.0</td>
<td>2 340</td>
<td>16</td>
</tr>
</tbody>
</table>

Values of the thermal neutron diffusion parameters, resulting from the fits of Eq.(1), are presented in Table 2. In Case q1, the full available range of bucklings has been used and the $F$ correction has been obtained with a very good accuracy. The function $\lambda(B^2)$ is plotted in Fig. 1, together with the experimental points. Case q2 shows a change of the interpreted neutron parameters while neglecting the correction $F$. Although the values of the diffusion constant $D_0$ and of the diffusion cooling coefficient $C$ do not correspond in this case to the physical magnitudes, they can be helpful in certain circumstances. For example, in any theoretical approach in which the correction $F$ is not included, it is better to use these approximate parameters, $D_0$ and $C$, from Case q2 than those more accurate from Case q1 with the correction $F$ neglected. Case q3 presents the values of the parameters obtained in a smaller range of variation of the geometric buckling. As expected, an influence of the $F$ correction becomes insignificant and the values $D_0$ and $C$ tend to the result obtained in Case q1.

Table 2. Thermal neutron diffusion parameters determined for quartz.

<table>
<thead>
<tr>
<th>Case</th>
<th>Range of $B^2$ [cm$^2$]</th>
<th>Range of $R_g$ [cm]</th>
<th>Fixed $\langle\nu\Sigma_a\rangle$ [s$^{-1}$]</th>
<th>Fitted $D_0$ [cm$^2$s$^{-1}$]</th>
<th>Fitted $C$ [cm$^4$s$^{-1}$]</th>
<th>Fitted $F$ [cm$^6$s$^{-1}$]</th>
</tr>
</thead>
<tbody>
<tr>
<td>q1</td>
<td>0.004 ± 0.10</td>
<td>8 ± 45</td>
<td>1 001 ± 18</td>
<td>308 500 ± 2 400</td>
<td>2 117 000 ± 73 000</td>
<td>7 060 000 ± 530 000</td>
</tr>
<tr>
<td>q2</td>
<td>0.004 ± 0.10</td>
<td>8 ± 45</td>
<td>1 001 ± 18</td>
<td>271 000 ± 3 700</td>
<td>1 094 000 ± 44 000</td>
<td>—</td>
</tr>
<tr>
<td>q3</td>
<td>0.004 ± 0.04</td>
<td>13 ± 45</td>
<td>1 001 ± 18</td>
<td>309 700 ± 1 100</td>
<td>1 905 000 ± 51 000</td>
<td>—</td>
</tr>
</tbody>
</table>
Fig. 1. Results of the simulation of the variable buckling experiment for quartz.

4. Thermal neutron parameters of calcite

The simulations of the variable geometric buckling experiment for calcite, CaCO₃, have been performed under the following conditions:
– the rock density $\rho = 2.71 \text{ g cm}^{-3}$,
– the range of the radius of spheres $R_g = (7.5 \div 45) \text{ cm}$.

The obtained decay constants $\lambda$ of the fundamental mode of the thermal neutron flux are listed in Table 3 and plotted in Fig. 2.
Table 3. Decay constants $\lambda$ obtained from the simulated experiment for calcite.

<table>
<thead>
<tr>
<th>$R_g$ [cm]</th>
<th>$\lambda$ [s$^{-1}$]</th>
<th>$\sigma(\lambda)$ [s$^{-1}$]</th>
<th>$R_g$ [cm]</th>
<th>$\lambda$ [s$^{-1}$]</th>
<th>$\sigma(\lambda)$ [s$^{-1}$]</th>
<th>$R_g$ [cm]</th>
<th>$\lambda$ [s$^{-1}$]</th>
<th>$\sigma(\lambda)$ [s$^{-1}$]</th>
</tr>
</thead>
<tbody>
<tr>
<td>7.5 19 887 45</td>
<td></td>
<td></td>
<td>10.0 14 905 28</td>
<td></td>
<td></td>
<td>16.0 8 491 19</td>
<td></td>
<td></td>
</tr>
<tr>
<td>8.0 18 696 195</td>
<td></td>
<td></td>
<td>11.0 13 282 22</td>
<td></td>
<td></td>
<td>18.0 7 375 30</td>
<td></td>
<td></td>
</tr>
<tr>
<td>8.5 17 664 147</td>
<td></td>
<td></td>
<td>12.0 12 029 48</td>
<td></td>
<td></td>
<td>20.0 6 475 29</td>
<td></td>
<td></td>
</tr>
<tr>
<td>9.0 16 622 120</td>
<td></td>
<td></td>
<td>13.0 11 019 67</td>
<td></td>
<td></td>
<td>25.0 4 910 9</td>
<td></td>
<td></td>
</tr>
<tr>
<td>9.5 15 657 84</td>
<td></td>
<td></td>
<td>14.0 10 076 62</td>
<td></td>
<td></td>
<td>45.0 2 755 9</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Fig. 2. Results of the simulation of the variable buckling experiment for calcite.

Values of the thermal neutron diffusion parameters, resulting from the fit of Eq.(1), are presented in Table 4. Also here it has been possible to determine not only the diffusion cooling coefficient, $C$, but also the correction, $F$ (Case c1). Similarly as in the preceding section, other variants of the fit are shown here in Table 4 (Cases c2, c3).
Table 4. Thermal neutron diffusion parameters determined for calcite.

<table>
<thead>
<tr>
<th>Case</th>
<th>Range of $B^2$ [cm$^2$]</th>
<th>Range of $R_g$ [cm]</th>
<th>Fixed</th>
<th>Fitted</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>$\langle \nu \Sigma_a \rangle$ [s$^{-1}$]</td>
<td>$D_0$ [cm$^2$s$^{-1}$]</td>
</tr>
<tr>
<td>c1</td>
<td>0.004 ÷ 0.12</td>
<td>7.5 ÷ 45</td>
<td>1 557 ± 72</td>
<td>265 800 ± 1 800</td>
</tr>
<tr>
<td>c2</td>
<td>0.004 ÷ 0.12</td>
<td>7.5 ÷ 45</td>
<td>241 700 2 700</td>
<td>755 000 30 000</td>
</tr>
<tr>
<td>c3</td>
<td>0.004 ÷ 0.06</td>
<td>11 ÷ 45</td>
<td>262 900 1 300</td>
<td>1 118 000 28 000</td>
</tr>
</tbody>
</table>

5. Thermal neutron parameters of dolomite

The simulations of the experiment for dolomite, CaMg(CO$_3$)$_2$, have been performed under the following conditions:

– the rock density $\rho = 2.87$ g cm$^{-3}$,
– the range of the radius of spheres $R_g = (6 ÷ 45)$ cm.

The obtained decay constants $\lambda(R_g)$ of the thermal neutron flux are given in Table 5, and plotted in Fig. 3. The interpreted thermal neutron diffusion parameters are presented in Table 6.

Table 5. Decay constants $\lambda$ obtained from the simulated experiment for dolomite.

<table>
<thead>
<tr>
<th>$R_g$ [cm]</th>
<th>$\lambda$ [s$^{-1}$]</th>
<th>$\sigma(\lambda)$ [s$^{-1}$]</th>
<th>$R_g$ [cm]</th>
<th>$\lambda$ [s$^{-1}$]</th>
<th>$\sigma(\lambda)$ [s$^{-1}$]</th>
<th>$R_g$ [cm]</th>
<th>$\lambda$ [s$^{-1}$]</th>
<th>$\sigma(\lambda)$ [s$^{-1}$]</th>
<th>$R_g$ [cm]</th>
<th>$\lambda$ [s$^{-1}$]</th>
<th>$\sigma(\lambda)$ [s$^{-1}$]</th>
</tr>
</thead>
<tbody>
<tr>
<td>6.0</td>
<td>24 089 94</td>
<td>8.5 16 783 67</td>
<td>11.0</td>
<td>12 243 23</td>
<td>16.0 7 415 4</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>6.5</td>
<td>22 463 34</td>
<td>9.0 15 731 62</td>
<td>12.0</td>
<td>11 001 49</td>
<td>18.0 6 312 7</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>7.0</td>
<td>20 873 34</td>
<td>9.5 14 737 49</td>
<td>13.0</td>
<td>9 822 31</td>
<td>20.0 5 484 22</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>7.5</td>
<td>19 334 153</td>
<td>10.0 13 895 76</td>
<td>14.0</td>
<td>8 886 20</td>
<td>25.0 4 055 10</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>8.0</td>
<td>17 950 87</td>
<td>10.5 13 001 8</td>
<td>15.0</td>
<td>8 120 33</td>
<td>45.0 2 092 16</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
**Fig. 3.** Results of the simulation of the variable buckling experiment for dolomite.

**Table 6.** Thermal neutron diffusion parameters determined for dolomite.

<table>
<thead>
<tr>
<th>Case</th>
<th>Range of $B^2$ [cm$^{-2}$]</th>
<th>Range of $R_g$ [cm]</th>
<th>Fixed $\langle \nu \Sigma_a \rangle$ [s$^{-1}$]</th>
<th>Fitted $D_0$ [cm$^2$s$^{-1}$]</th>
<th>$C$ [cm$^4$s$^{-1}$]</th>
<th>$F$ [cm$^6$s$^{-1}$]</th>
</tr>
</thead>
<tbody>
<tr>
<td>d1</td>
<td>0.005 ÷ 0.18</td>
<td>6.0 ÷ 45</td>
<td></td>
<td>225 200 ± 1 300</td>
<td>778 000 ± 24 000</td>
<td>1 380 000 ± 100 000</td>
</tr>
<tr>
<td>d2</td>
<td>0.006 ÷ 0.18</td>
<td>6.0 ÷ 45</td>
<td>1 033 ± 42</td>
<td>205 100 ± 2 000</td>
<td>443 000 ± 15 000</td>
<td>—</td>
</tr>
<tr>
<td>d3</td>
<td>0.005 ÷ 0.06</td>
<td>11 ÷ 45</td>
<td></td>
<td>227 200 ± 1 300</td>
<td>739 000 ± 28 000</td>
<td>—</td>
</tr>
</tbody>
</table>
6. Density-removed thermal neutron parameters of the minerals

The thermal neutron diffusion parameters are expressed by the macroscopic cross sections (the absorption, total and differential scattering cross sections) and, thus, depend on the mass density $\rho$ of the substance. The MC simulations have been done using the mass densities of the solid rock materials. In any experiment or a calculation for a crushed rock samples, the bulk density appears and it changes the values of the parameters. Therefore, values of the density-removed parameters $\langle (\Sigma_a)^M, D_0^M, C^M, F^M \rangle$ are more general. They have been obtained here from the determined parameters $\langle (\Sigma_a), D_0, C, F \rangle$, according to the theoretical definitions in which the dependence on $\rho$ is explicit (e.g. [3], [12]). The results are collected in Table 7.

<table>
<thead>
<tr>
<th>Mineral</th>
<th>$\rho_0$ (g cm$^{-3}$)</th>
<th>$\langle (\Sigma_a)^M \rangle$ s$^{-1}$/(g cm$^{-3}$)</th>
<th>$D_0^M$ cm$^2$ s$^{-1}$ (g cm$^{-3}$)</th>
<th>$C^M$ cm$^4$ s$^{-1}$ (g cm$^{-3}$)$^3$</th>
<th>$F^M$ cm$^6$ s$^{-1}$ (g cm$^{-3}$)$^5$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Quartz SiO$_2$</td>
<td>2.65</td>
<td>378 ± 7</td>
<td>817 500 ± 6 400</td>
<td>39 400 000 ± 1 360 000</td>
<td>922 000 000 ± 68 000 000</td>
</tr>
<tr>
<td>Calcite CaCO$_3$</td>
<td>2.71</td>
<td>575 ± 27</td>
<td>720 300 ± 4 800</td>
<td>26 680 000 ± 930 000</td>
<td>528 000 000 ± 43 000 000</td>
</tr>
<tr>
<td>Dolomite CaMg(CO$_3$)$_2$</td>
<td>2.87</td>
<td>360 ± 15</td>
<td>646 400 ± 3 700</td>
<td>18 400 000 ± 560 000</td>
<td>269 000 000 ± 19 000 000</td>
</tr>
</tbody>
</table>

The values of the parameters at a given density of the material can be then easily obtained:

$$\langle \Sigma_a \rangle = \rho \langle (\Sigma_a)^M \rangle, \quad D_0 = D_0^M/\rho, \quad C = C^M/\rho^3, \quad F = F^M/\rho^5.$$  

(6)

In Table 7, there are given the rock densities $\rho_0$ at which the parameters, $\langle (\Sigma_a), D_0, C, F \rangle$, have been obtained. This is a certain reference because results of a recalculation of the parameters for a material of the density $\rho$, which is very different from $\rho_0$, have to be used attentively, as mentioned in [6].
7. Conclusions

At neutron laboratory experiments with rock materials, finite samples of the material are always used (either small crushed samples or rock blocks). The thermal neutron diffusion cooling effect cannot be neglected in certain cases, as stated in [1]. Here, the results of the simulated pulsed-neutron experiment for three basic rock minerals have been presented. Not only the thermal neutron diffusion coefficients, $C$, but even the corrections, $F$, have been obtained with a high accuracy. Approximations for the $C$ coefficients have been given for the cases when the $F$ correction is not included into a calculation. The density-removed thermal neutron parameters have been also determined in order to make the presented results possible to apply for a calculation at the material density different from the specific one used in the simulations.

The investigated pure minerals (quartz SiO$_2$, calcite CaCO$_3$, dolomite CaMg(CO$_3$)$_2$) are major constituents of the respective rocks (sandstone, limestone, dolomite). These natural rocks contain some admixtures of other substances. The admixtures can significantly change only the macroscopic absorption cross section $\Sigma_a$ of a material if they contain highly absorbing elements. The scattering cross section $\Sigma_s$ is practically insensitive to such a contamination. The thermal neutron diffusion cooling is defined just by the scattering properties of the material and, in consequence, is also almost independent of the trace elements content. Therefore the diffusion cooling coefficients determined for pure rock minerals can be used for the corresponding rocks.

A serious problem exists in another case, namely when rock samples contain natural humidity. The thermal neutron scattering properties of hydrogen and of the elements present in the rock minerals are extremely different (regarding the dependence on the neutron energy). The diffusion cooling coefficient is not additive in respect to the scattering cross sections of the components of a material (cf. the theoretical definitions in [4], [12]) and is not a linear combination of the respective coefficients of the contributing substances. The problem for a wet rock material has been indicated in [1]. A determination of the diffusion cooling coefficient for such materials needs a separate study.
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