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**Measurements of the thermal neutron absorption  
 $\Sigma_a$  of model systems (reference KCl solutions)  
in two-region geometry**

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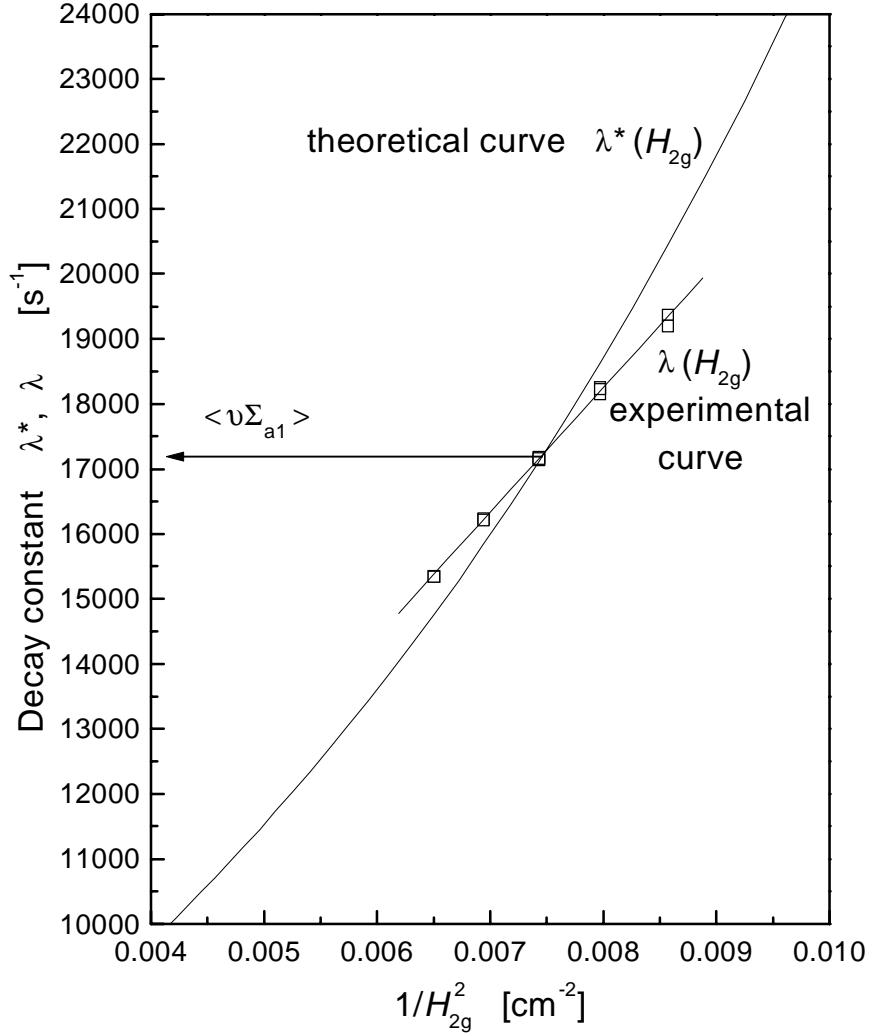
**Abstract**

Reference experiments for the thermal neutron macroscopic absorption cross section  $\Sigma_a$  measurements are described. Water solutions of high-purity potassium chloride have been used as the reference  $\Sigma_a$  samples. Czubek's pulsed neutron measurement method has been applied. It is based on a series of experiments in a two-region geometry (inner zone: sample, outer zone: moderator) combined with a theoretical function  $\lambda^*$  dependent on the thermal neutron parameters of the moderator and on the system geometry. The accuracy of this measurement method can be improved by adding the assumption that the diffusion cooling coefficient  $C$  of the outer medium is not only a pure material parameter but is also dependent on the function  $\lambda^*$ . This dependence  $C^*$  has been determined from the experiments presented.

## 1. Introduction

The fast neutron generator in the Neutron Transport Physics Laboratory is among other used for measurements of the thermal neutron absorption cross section  $\Sigma_a$  of various materials. Recently the experimental set-up was modernised and two independent registration lines with two fast multiscalers are now used to register the time decay of the pulsed thermal neutron flux in the investigated system. Geometrical conditions at the measuring position were also changed by an installation of a thermostatic chamber for future experiments related to temperature dependences of the thermal neutron diffusion parameters. The macroscopic absorption cross section  $\Sigma_a$  is measured on this experimental set-up using Czubek's pulsed neutron method [1]. The measurement is based on a comparison of certain experimental results with a theoretical description of the physical phenomenon. The basic result of the measuring procedure, *i.e.* the absorption rate  $\langle v\Sigma_{a1} \rangle$  of the sample material, is obtained as the ordinate of the intersection point of two curves. One of them is obtained experimentally and the other is theoretically calculated (The idea is presented in Fig. 1). For that reason the theoretical curve ought to be verified to eliminate a possible influence of the environment on the results of the measurement. Tests can be done only on the samples with the absorption cross section known with a very high accuracy.

Czubek's method uses a two-region geometry. The system consists of the investigated cylindrical sample of size  $H_1 = 2R_1$  surrounded entirely by a cylindrical moderator ( $H_{2g} = 2R_{2g}$ ) covered with a cadmium shield which assures the vacuum boundary conditions for thermal neutrons. The sample-moderator system is irradiated by bursts of 14 MeV neutrons which are slowed down, and the time decay of the resulting thermal neutron flux is observed. The decay constant  $\lambda$  of the fundamental exponential mode of the flux is determined from the registered curve [2]. The experiment is repeated using the outer moderators of different sizes  $H_{2g}$ . In this way the experimental dependence  $\lambda = \lambda(H_{2g}; H_1 = const)$  is obtained. Another curve  $\lambda^* = \lambda^*(H_{2g}; H_1 = const)$  is calculated based on the diffusion approximation for the thermal neutron flux in the two-region cylindrical system [3], [4]. The position of the theoretical curve  $\lambda^*(H_{2g})$  depends on the thermal neutron parameters of the external moderator and, therefore, they have to be known with a high accuracy. The function  $\lambda^*(H_{2g})$  includes the absorption rate  $\langle v\Sigma_{a2} \rangle$ , the diffusion constant  $D_{02}$ , and the diffusion cooling



**Fig. 1.** Absorption rate of the sample  $\langle v\Sigma_{a1} \rangle$  determined from the intersection of the theoretical and experimental curves.

coefficient  $C_2$ . Values  $\langle v\Sigma_{a2} \rangle$  and  $D_{02}$  are usually well established. The coefficient  $C_2$  results from the diffusion cooling of the thermal neutron energy spectrum in a finite system. For the same material, different data can be found. Theoretical values depend on a theoretical approach assumed (cf. [5], [6]) and experimental ones depend on some details in an interpretation procedure (cf. [7]). The problem was reviewed in [8]. Plexiglas is routinely used as the outer moderator in our measurements. The values of its diffusion cooling coefficient reported in the literature are:

experimental – between 2800 and 7149 cm<sup>4</sup>s<sup>-1</sup>,

theoretical – between 4150 and 6310 cm<sup>4</sup>s<sup>-1</sup>

(collected by Drozdowicz [8]). The thermal neutron parameters, based on a re-interpretation of the raw data from the variable buckling experiment [9], are following:

$$\begin{aligned}\langle v\Sigma_{a2} \rangle &= 4\,169 \pm 10 \text{ s}^{-1}, \\ D_{02} &= 36\,542 \pm 125 \text{ cm}^2\text{s}^{-1}, \\ C_2 &= 4\,377 \pm 140 \text{ cm}^4\text{s}^{-1},\end{aligned}\tag{1}$$

when the 3-parameter fit has been used. The absorption rate and the diffusion constant, specified in Eq.(1), are directly used in the calculation of the theoretical curve  $\lambda^*(H_{2g})$ . The coefficient  $C_2$  in the theory of the  $\Sigma_a$  measurement mentioned above should include also a correction which takes into account a deviation of the theory from the experimental reality. Such an approach to the diffusion cooling coefficient was already used by us during elaboration of the  $\Sigma_a$  measurement method [10], [11]. Then a constant value of  $C_2$ , different from the value of the material parameter, was adjusted.

The principle of the present consideration is modified as follows. The measurements performed on a reference sample of the known value of the absorption rate  $\langle v\Sigma_{a1} \rangle^{\text{ref}}$  determine the experimental curve  $\lambda(H_{2g})$ . The intersection of this curve with the theoretical one,

$$\lambda^*(H_{2g}) = \lambda^*(H_{2g}; \{H_1, \langle v\Sigma_{a2} \rangle, D_{02}, C_2\} = \text{const}),\tag{2}$$

should return the value  $\langle v\Sigma_{a1} \rangle^{\text{ref}}$ . If the theoretical curve  $\lambda^*(H_{2g})$  describes with a deviation the physical reality, the „experimental” result  $\langle v\Sigma_{a1} \rangle^{\text{exp}} \neq \langle v\Sigma_{a1} \rangle^{\text{ref}}$ . The curve  $\lambda^*(H_{2g})$  can be corrected by changing the parameter  $C_2$  until the correct result,  $\langle v\Sigma_{a1} \rangle^{\text{exp}} = \langle v\Sigma_{a1} \rangle^{\text{ref}}$ , is attained. This corresponds to a certain new value  $C_2^*$  of this coefficient. However, this value can be different at different absorption rates  $\langle v\Sigma_{a1} \rangle$ . Then a repetition of the measurements for different samples determines the dependence  $C_2^* = f(\langle v\Sigma_{a1} \rangle^{\text{ref}})$ , which can be used in the calculation of the theoretical curve:

$$\lambda^*(H_{2g}) = \lambda^*(H_{2g}; \{H_1, \langle v\Sigma_{a2} \rangle, D_{02}\}) = const, C_2^* = f(\langle v\Sigma_{a1} \rangle) \quad (3)$$

within the tested interval of the  $\langle v\Sigma_{a1} \rangle^{\text{ref}}$  values.

## 2. The absorption reference material

As said, the test of the theoretical curve  $\lambda^*(H_{2g})$  ought to be done at various neutron absorption cross sections of the sample. Solutions of a chosen material are convenient to get easily a required absorption. The selection of a reference material, which has the thermal neutron macroscopic absorption cross section  $\Sigma_a$  accurately known, is difficult. The material must not contain absorbing admixtures or their contents have to be known with a very high accuracy, sometimes up to single ppm's (where  $1 \text{ ppm} = 10^{-6}$ ).

**Table 1.**  
Contents of the trace elements in  
99.999 % potassium chloride  
(Aldrich Chemical Company).

Element	Weight content [ppm]
Ca	6.2
Si	1.6
Na	1.2
B	1.1
Rb	0.7
Cs	0.5
Fe	0.3
Pb	0.2
Li	0.1
Mn	0.1
Cr	0.03
Sn	0.03
Ge	0.02
Ba	0.01
Pd	0.01
Tl	0.01

Water solutions of different concentrations of boric acid  $H_3BO_3$  might be used as the reference samples, considering the  $\langle v\Sigma_{a1} \rangle$  interval which should be covered. However, the isotope ratio  $^{10}\text{B} / ^{11}\text{B}$  in natural boron B slightly fluctuates [12]. This finally influences the absorption cross section  $\Sigma_a$  of boron because the microscopic absorption cross sections of the isotopes  $^{10}\text{B}$  and  $^{11}\text{B}$  differ extremely:  $\sigma_a(^{10}\text{B}) = 3837 \pm 9 \text{ b}$  and  $\sigma_a(^{11}\text{B}) = 5.5 \pm 3.3 \text{ mb}$  [13]. Unfortunately, knowledge of the boron isotopic ratio with a sufficient accuracy is a serious problem in the case of each series of the material.

Potassium chloride is another quite convenient material to get solutions of the required values of the absorption cross section. This substance of a very high purity, 99.999 %

KCl, has been used in our reference measurements. The trace elements are specified in Table 1. The calculated mass macroscopic absorption cross sections  $\Sigma_a^M = \Sigma_a / \rho$  for 100 % KCl and for the used material with the trace elements are given in Table 2. The values are very close to each other (equal within the uncertainty interval), which shows that an influence of the trace element contents is here very weak. If it is taken into account during calculation of the cross section  $\Sigma_a^M$ , the 99.999 % KCl substance may be used as a good reference material.

**Table 2.** Mass absorption cross sections  $\Sigma_a^M$  calculated for potassium chloride (at the most probable thermal neutron velocity  $v_0 = 2\,200$  m/s).

Compound	$\Sigma_a^M \pm \sigma(\Sigma_a^M)$ [cm <sup>2</sup> /g]
100 % KCl	$287.55 \pm 2.55$
99.999 % KCl with trace elements from Table 1	$287.60 \pm 2.55$

### 3. Experimental results

The thermal neutron absorption rate  $\langle v\Sigma_{a1} \rangle$  of KCl solutions of six different concentrations has been measured. The maximum dissolution of potassium chloride in water (at the normal conditions) corresponds to the KCl weight concentration  $k = 24.0\%$  in the solution. The highest concentration, used in our experiments, was  $k = 23.6\%$ . The sample was prepared by a direct dissolution of potassium chloride in distilled water. Solutions of required lower KCl concentrations were obtained by consecutive dilutions of the samples already existing. Based on the data on the density  $\rho$  of the solutions, given in [14] for several concentrations  $k$ , the function  $\rho(k)$  has been fitted:

$$\rho(k) = A + B_1k + B_2k^2 + B_3k^3. \quad (4)$$

If  $k$  is expressed in per cents, the obtained coefficients are:

$$\begin{aligned} A &= 0.99824 \text{ g cm}^{-3}, & B_1 &= 0.00636 \text{ g cm}^{-3}, \\ B_2 &= 0.114786 \cdot 10^{-4} \text{ g cm}^{-3}, & B_3 &= 0.350287 \cdot 10^{-6} \text{ g cm}^{-3}. \end{aligned}$$

Additionally, the density  $\rho$  of each prepared solution was checked with a densimeter. The specification of the samples is given in Table 3. For each sample the thermal neutron decay constants  $\lambda$  were measured in the two-region cylindrical geometry as a function of the size of the outer moderator. These results, which define the experimental curves  $\lambda(H_{2g})$ , are listed in Table 4 (where  $\sigma(\lambda)$  is the standard deviation of the determined value  $\lambda$ ).

**Table 3.** List of the samples.

Sample	$k$ $\sigma(k)$ [wt %]	$\rho(k)$ [g cm <sup>-3</sup> ]
POTX236	23.600 0.007	1.1593
POTX210	21.000 0.007	1.1401
POTX180	18.000 0.007	1.1185
POTX150	15.000 0.006	1.0974
POTX130	13.000 0.006	1.0836
POTX115	11.500 0.005	1.0734

#### 4. The dependence $C_2^*(\lambda^*)$

The absorption rate  $\langle v\Sigma_{a1} \rangle$  of a solution of the given concentration  $k$  is defined by the elemental composition, the microscopic absorption cross sections, and the mass density  $\rho(k)$ . The reference absorption rates  $\langle v\Sigma_{a1} \rangle^{\text{ref}}$  have been calculated for all samples (using a computer program [15] which includes the cross section library based mainly on [13] and [16]). The intersection point

$$\lambda(H_{2g}) = \lambda^*(H_{2g}) = \langle v\Sigma_{a1} \rangle \quad (5)$$

**Table 4.** Decay constants  $\lambda$  measured on samples of different concentrations of KCl.

a) Sample POTX236		b) Sample POTX210		c) Sample POTX180	
$H_{2g}$ [cm]	$\lambda$ $\sigma(\lambda)$ [s <sup>-1</sup> ]	$H_{2g}$ [cm]	$\lambda$ $\sigma(\lambda)$ [s <sup>-1</sup> ]	$H_{2g}$ [cm]	$\lambda$ $\sigma(\lambda)$ [s <sup>-1</sup> ]
10.0	23 972 59	10.4	21 544 84	10.8	19 370 62
	24 079 83		21 532 67		19 204 37
	23 939 77		21 547 117	11.2	18 258 110
	23 921 104		21 538 111		18 254 53
10.4	22 259 98	10.8	20 095 122	11.6	18 157 86
	22 334 116		20 176 90		18 225 128
	22 305 69		20 157 77	12.0	17 137 53
	22 249 104		20 152 89		17 185 62
10.8	20 791 76	11.2	18 944 85	12.4	17 183 94
	20 787 76		18 924 77		17 160 59
	20 774 160		18 883 128	12.0	16 225 50
	20 827 71		18 893 66		16 232 35
11.2	19 490 37	11.6	17 797 102	12.4	16 213 76
	19 512 62		17 755 100		16 216 32
			17 812 58	12.4	15 351 46
			17 859 57		15 344 48



**Table 4.** (continued).**d) Sample POTX150**

$H_{2g}$ [cm]	$\lambda$ $\sigma(\lambda)$ [s <sup>-1</sup> ]
11.6	16 474 49
	16 453 39
	16 416 56
	16 534 85
	16 462 97
	16 534 94
12.0	15 595 67
	15 578 44
	15 595 59
	15 583 19
12.4	14 764 51
	14 874 66
	14 760 84
	14 889 97
12.8	14 125 41
	14 104 114
	14 077 50
	14 162 53

**e) Sample POTX130**

$H_{2g}$ [cm]	$\lambda$ $\sigma(\lambda)$ [s <sup>-1</sup> ]
12.4	14 423 75
	14 410 46
	14 428 71
12.8	13 757 86
	13 752 46
	13 725 52
13.2	13 145 84
	13 144 62
	13 173 62
13.6	12 590 39
	12 598 63
	12 574 78

**f) Sample POTX115**

$H_{2g}$ [cm]	$\lambda$ $\sigma(\lambda)$ [s <sup>-1</sup> ]
12.8	13 482 23
	13 494 22
13.2	12 887 42
	12 872 103
13.6	12 399 60
	12 352 38
14.0	11 898 50
	11 885 51

was determined for each sample by fitting the value of the coefficient  $C_2^*$  in order to obtain the result  $\langle \nu \Sigma_{a1} \rangle^{\text{exp}} = \langle \nu \Sigma_{a1} \rangle^{\text{ref}}$ . The reference absorption rates  $\langle \nu \Sigma_{a1} \rangle^{\text{ref}}$  of the samples and the  $C_2^*$  values fitted are listed in Table 5.

**Table 5.**

Coefficient  $C_2^*$  fitted at different absorption rates  $\langle \nu \Sigma_{a1} \rangle$ .

$k$ [wt %]	$\langle \nu \Sigma_{a1} \rangle^{\text{ref}}$ $\sigma(\langle \nu \Sigma_{a1} \rangle^{\text{ref}})$ [s <sup>-1</sup> ]	$C_2^*$ [cm <sup>4</sup> s <sup>-1</sup> ]
23.6	21 643 154	6 447
21.0	19 554 135	6 770
18.0	17 224 114	7 460
15.0	14 978 93	8 155
13.0	13 525 80	8 740
11.5	12 458 70	9 010

The obtained  $C_2^*$  values are individual at different measured absorption rates, *i.e.*  $C_2^* = C_2^*(\langle \nu \Sigma_{a1} \rangle)$ . In the intersection point, Eq.(5), the measured value  $\langle \nu \Sigma_{a1} \rangle$  belongs also to the function  $\lambda^*$ . Thus, the dependence of the coefficient  $C_2^*$  can be defined as

$$C_2^* = C_2^*(\lambda^*) \quad (6)$$

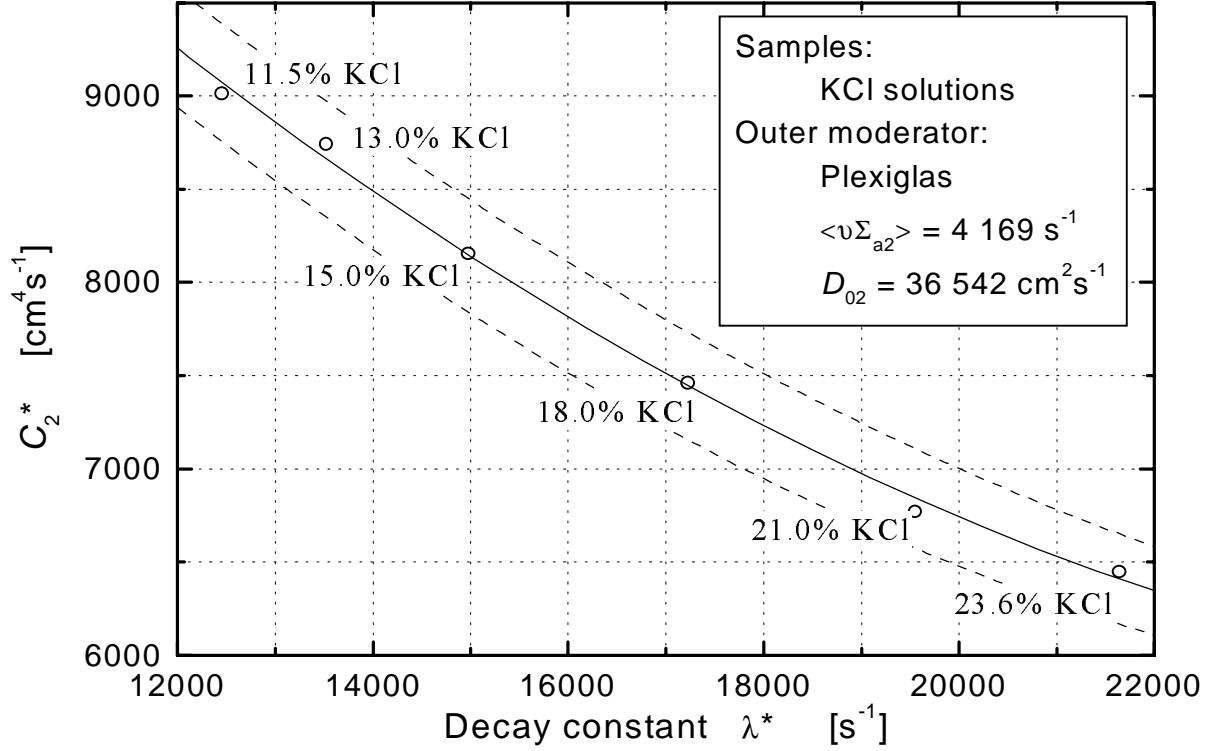
This function in the investigated interval of  $\lambda^*$  has been approximated by the parabola:

$$C_2^* = a + b_1 \lambda^* + b_2 \lambda^{*2}, \quad (7)$$

and the parameters obtained from the fit are:

$$\begin{aligned} a &= 15\,816 \pm 1\,066 \text{ cm}^4 \text{ s}^{-1}, \\ b_1 &= -0.68578 \pm 0.12916 \text{ cm}^4, \\ b_2 &= 1.1603 \cdot 10^{-5} \pm 0.3787 \cdot 10^{-5} \text{ cm}^4 \text{ s}. \end{aligned} \quad (8)$$

The experimental data and the fitted function are plotted in Fig. 2. The correlation coefficient  $r(C_2^*, \langle \nu \Sigma_{a1} \rangle^{\text{ref}})$  is 0.998. The function given in Eq.(7) has been accepted as the correction that eliminates a discrepancy between the experiment and the theory used.



**Fig. 2.** The dependence  $C_2^*(\lambda^*)$ .

## 5. Final remarks

In order to check the accuracy of the obtained fit  $C_2^* = C_2^*(\lambda^*)$ , a selftest has been done. The sets of the experimental points  $\{\lambda(H_{2g})\}$ , which were measured for each reference sample and used in the procedure of determination of the function  $C_2^*(\lambda^*)$ , have been treated as the experimental results obtained for unknown samples. The absorption rate for each sample  $\langle \nu \Sigma_{a1} \rangle^{\text{test}}$  has been determined with Czubek's method using in the theoretical curve, Eq.(3), the obtained dependence  $C_2^*(\lambda^*)$ . These experimental results  $\langle \nu \Sigma_{a1} \rangle^{\text{test}}$  have been compared in

Table 6 with the reference data  $\langle \nu \Sigma_{a1} \rangle^{\text{ref}}$ . The relative disagreement  $|\varepsilon|$  does not exceed 0.2 %, which allows us to accept the obtained fit. The value of  $C_2^*$  changes approximately between 9200 and 6400  $\text{cm}^4 \text{s}^{-1}$  in the interval presented in Fig. 2. It is on average lower than a constant value assumed in the former conditions [11] but, as previously, still higher than the material parameter (the thermal neutron diffusion cooling coefficient  $C_2$  of Plexiglas).

**Table 6.** Results of the selftest.

Sample	$\langle \nu \Sigma_{a1} \rangle^{\text{ref}}$ $\sigma(\langle \nu \Sigma_{a1} \rangle^{\text{ref}})$ [s <sup>-1</sup> ]	$\langle \nu \Sigma_{a1} \rangle^{\text{test}}$ $\sigma(\langle \nu \Sigma_{a1} \rangle^{\text{test}})$ [s <sup>-1</sup> ]	$\varepsilon = \frac{\langle \nu \Sigma_{a1} \rangle^{\text{test}} - \langle \nu \Sigma_{a1} \rangle^{\text{ref}}}{\langle \nu \Sigma_{a1} \rangle^{\text{ref}}}$ [%]
POTX236	21 643 154	21 618 153	- 0.12
POTX210	19 554 135	19 591 142	+ 0.19
POTX180	17 224 114	17 219 115	- 0.03
POTX150	14 978 93	14 977 97	- 0.007
POTX130	13 525 80	13 506 110	- 0.14
POTX115	12 458 70	12 473 104	+ 0.12

A separate problem is to estimate a possible systematic error which can come from the inaccuracy of the microscopic cross sections used to calculate the reference material cross section  $\Sigma_a^M$ . They are known with their individual standard deviations and the resulting standard deviation  $\sigma(\Sigma_a^M)$  is given in Table 2. All the samples were prepared of the same material. Thus, all the calculated cross sections can be deviated only in one direction from the true (unknown) cross section and they do not fluctuate within the interval defined by the standard deviation  $\sigma(\Sigma_a^M)$ . Therefore, it is possible to find a belt around the fitted function  $C_2^*(\lambda^*)$  when the reference cross section is assumed as  $\Sigma_a^M + \sigma(\Sigma_a^M)$  and  $\Sigma_a^M - \sigma(\Sigma_a^M)$ .

The fitting procedure, which has been presented in paragraph 4, was repeated for the two cases. The results are plotted with a dashed line in Fig 2. In this way the standard deviation was finally found as possible to be approximated by the relative deviation  $\sigma(C_2^*)/C_2^* \approx 3.36\%$ .

We would like to mention an experimental problem which we met at this work. It is related to the reliability of the reference material for the thermal neutron absorption. Our first measurements on potassium chloride have been performed with use of another portion of the material, defined as the 99.9% KCl purity. The specified contents of the trace elements allowed us to calculate the mass absorption cross section of the substance,  $\Sigma_a^M = 287.27 \pm 2.55 \text{ cm}^2/\text{g}$ , which is very close to the cross section of the pure compound (see Table 2, for 100 % KCl). However, the measured fundamental decay constants  $\lambda$  were lower than expected and they indicated that the true absorption cross section was lower (which could suggest a contamination by low-absorbing components which were not specified). Thus, that 99.9 % KCl material had to be excluded from the use as a reference material. The problem of the purity of standards for  $\Sigma_a$  measurements we will discuss elsewhere.

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