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THERMAL NEUTRON PULSED PARAMETERS IN NON-HYDRROGENOUS SYSTEMS. EXPERIMENT FOR LEAD GRAINS

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ABSTRACT

In Czubek's method of measurement of the thermal neutron macroscopic absorption cross section a two-region geometry is applied where the investigated sample is surrounded by an external moderator. Both regions in the measurements made up till now were hydrogenous, which means the same type of the thermal neutron transport properties. In the paper a theoretical consideration to use non-hydrogenous materials as the samples is presented. Pulsed neutron measurements have been performed on homogeneous material in a geometry of the classic experiment with the variable geometric buckling. Two decay constants have been measured for different cylindrical samples of small lead grains (a lead shot).

1. INTRODUCTION

Thermal neutron material parameters are measured in the Laboratory of the Neutron Transport Physics at the Institute of Nuclear Physics using the 14 MeV neutron pulsed generator. The fast neutrons are slowed down in the sample volume investigated and the thermal neutron decaying flux is observed. The material parameters obtained finally are called the pulsed thermal neutron parameters: the absorption rate, the diffusion constant, and the diffusion cooling coefficient. The system which contains hydrogen is a good moderator and provides a good efficiency of the measurement considering the thermal neutron flux. But hydrogen is an extraordinary element in certain respect: its properties for the thermal neutron

transport (the scattering cross section and the average cosine of the scattering angle as a function of the incident neutron energy) vary for different materials depending on the molecular bindings. Thus, the cross section of gas hydrogen is very different from that in water or in Plexiglass, *etc.*

In the Lab a measurement method was elaborated where a two-region sample-moderator system was used for the determination of the thermal neutron absorption cross section Σ_a of the sample. The same type of the solutions of the neutron diffusion equation and the same way of the interpretation of the experimental results can be assumed when media in both regions are hydrogenous. Difficulties can appear when one medium is hydrogenous and the second is not.

Problems of the neutron transport in hydrogenous and non-hydrogenous media will be investigated in a future work in the Lab. Therefore, a usefulness of available non-hydrogenous materials for experiments with pulsed neutron fields should be considered. A relevant material of such a type could be also used for additional new tests for CZUBEK's (1981, 1996) method of the Σ_a measurement on small samples.

Basing on the analysis made, lead in grains (of the diameter $\varnothing$ = 2 mm) was chosen as a non-hydrogenous material. Two values of the fundamental mode decay constant λ_0 of the thermal neutron flux at two values of the geometric buckling B_g^2 were measured for the samples prepared of the lead shot.

2. REVIEW OF SELECTED NON-HYDROGENOUS MATERIALS.

Materials of different types were selected (as listed in Table 1). They can be divided in three groups.

1) Glass.

Glass consists of silica SiO_2 and various admixtures. Therefore, it is possible (at least in a theory) to find such a composition of glass to get the thermal neutron parameters which will be most relevant for the mentioned experiments planned. Among the glasses quoted in Table 1 such 'theoretical' glasses are the nickel glass (containing 50% of SiO_2 and 50% of NiO) and the boric glass (containing 0.05 % of B).

Table 1. Thermal neutron parameters of various materials calculated on the basis of the elemental composition ($\upsilon_0 = 2200\,m/s$).

Material	$\Sigma_a^M(\upsilon_0)$	$\Sigma_s^M(\upsilon_0)$	$\Sigma_{tr}^M(\upsilon_0)$	ρ	$\langle\upsilon\Sigma_a\rangle$	$\langle D\rangle$	D_0
	[cm^2/g]	[cm^2/g]	[cm^2/g]	[g/cm^3]	[s^{-1}]	[cm]	[cm^2s^{-1}]
Glasses							
SiO_2	0.00172	0.09587	0.09302	1.8	680	1.9563	485650
SiO_2+50%NiO	0.01896	0.13483	0.13138	~ 5	20851	0.4525	112327
SiO_2+0.05%B	0.02308	0.09594	0.09309	1.8	9139	1.6607	412266
architectural glass	0.00313	0.09475	0.09191	2.8	1931	1.2556	311689
white glass	0.00329	0.09274	0.08999	2.8	2027	1.2796	317658
amber glass	0.00341	0.09282	0.09007	2.8	2099	1.2771	317030
bottle glass	0.00351	0.09308	0.09031	2.8	2159	1.2726	315915
dense flint	0.01305	0.06407	0.06265	3.87	11109	1.1759	291904
Metals and alloys							
Monel metal	0.04522	0.14664	0.14500	~ 6	59694	0.3083	76532
cupro-nickel	0.03787	0.09550	0.09449	8.9	74152	0.3042	75506
brass: CuZn16Si3	0.03070	0.06712	0.06638	~ 8	54030	0.4670	115937
AlSi21CuNi	0.00617	0.03654	0.03566	~ 2.9	3938	2.8198	700006
AlCu2Mg2NiSi	0.00666	0.03685	0.03596	~ 2.9	4251	2.7739	688602
Al	0.00516	0.03155	0.03077	2.72	3085	3.4969	868082
Al + 30%Ni	0.01548	0.06492	0.06358	~ 4.5	15525	0.9613	238644
Bulk materials (porosity 40%)							
Ni	0.04606	0.18258	0.18051	5.01	50762	0.30280	75169
Cu	0.03583	0.07374	0.07296	5.36	42229	0.60488	150158
bottle glass	0.00353	0.09382	0.09104	1.008	777	3.54082	878985
Co	0.37992	0.06131	0.06062	5.34	446331	0.17682	43893
Mn	0.14579	0.02412	0.02382	4.44	142406	0.55168	136952
Sn	0.00318	0.02491	0.02477	4.32	3019	2.80425	696138
Na	0.01388	0.07924	0.07694	0.588	1796	6.37911	1583572
Zn	0.01023	0.03759	0.03720	4.278	9624	1.69785	421480
Fe	0.02760	0.12239	0.12093	4.716	28641	0.48913	121423
lead shot	0.00157	0.03248	0.03237	6.85	2362	1.4421	357986
Teflon	0.00027	0.14477	0.13871	2.150	129	1.1158	276991
Plexiglass							
$C_5H_8O_2$	0.01611	3.07496	2.45997	1.18	4183	0.1141	28326

2) Metals and alloys.

The elemental compositions of the alloys for which the neutron parameters were calculated were assumed after Mechanics Handbook (1976). Among the alloys quoted in Table 1 are the brass CuZn16Si3 (containing 79-81% Cu, 2.5-4% Si), the Monel metal (containing 67-69% Ni+Co, 28-31% Cu, 1-2% Mn, 0.5% Zn, 2.0% Fe), the cupro-nickel (containing 18-20% Ni+Co and 79-81% Cu), the alloy AlSi21CuNi (containing 20-23% Si, 1.1-1.5 % Cu, 0.5-0.9 % Mg, 0.1-0.3% Mn, 0.8-1.1% Ni) and the alloy AlCu2Mg2NiSi (containing 1.9-2.5% Cu, 1.4-1.8% Mg, 0.5-1.2% Si, 0.8-1.3% Ni, 0.8-1.3% Fe).

3) Bulk materials (of the same porosity).

It was assumed for the calculation that a given crushed material is homogeneous of a porosity equal to 40 %. This defines a significantly lower bulk density (quoted in Table 1) than the mass density of the solid material.

The thermal neutron parameters listed in Table 1 were estimated basing on their elemental compositions and microscopic absorption and scattering cross sections, using the following dependencies (*e.g.* DROZDOWICZ and KRYNICKA 1995).

The thermal neutron mass absorption cross section:

$$\Sigma_{\mathrm{a}}^{\mathrm{M}}(E) = N_{\mathrm{A}} \sum_{j=1}^{J} \left[\sum_{i=1}^{I} \frac{\sigma_{ai}(E)}{M_j} n_{ij} \right] q_{ij}, \tag{1}$$

where:

N_{A} – Avogadro number,

$\sigma_{ai}(E)$ – thermal neutron microscopic absorption cross section of the i-th element (energy function),

$M_j = \sum_{i=1}^{I} n_{ij} A_{ij}$, and A_{ij} is the gram-atom of the i-th element in the j-th compound,

q_{ij} – weight content of the j-th compound in the mixture, and $\sum_{j=1}^{J} q_j = 1$.

The thermal neutron mass scattering cross section:

$$\Sigma_{\mathrm{s}}^{\mathrm{M}}(E) = N_{\mathrm{A}} \sum_{j=1}^{J} \left[\sum_{i=1}^{I} \frac{\sigma_{si}(E)}{M_j} n_{ij} \right] q_{ij}, \tag{2}$$

where $\sigma_{si}(E)$ is the thermal neutron microscopic scattering cross section of the *i*-th element (energy function).

The thermal neutron mass transport cross section:

$$\Sigma_{tr}^{M}(E) = \left[1 - \mu(E)\right]\Sigma_{s}^{M}(E) \quad , \tag{3}$$

where $\mu(E)$ is the average cosine of the scattering angle (as a function of energy).

The corresponding *linear macroscopic cross sections* (*absorption, scattering, transport*) are related by

$$\Sigma(E) = \rho(T)\Sigma^{M}(E) \quad , \tag{4}$$

where $\rho(T)$ is the material density at temperature T.

The thermal neutron absorption rate, averaged over the neutron energy flux distribution:

$$\langle \upsilon\Sigma_a \rangle = \int_0^\infty \upsilon\Sigma_a(E) M_\Phi(E) \mathrm{d}E \, , \tag{5}$$

where:

υ – neutron velocity, $\upsilon = \sqrt{2E/m_n}$ (where m_n is the neutron mass),

M_Φ – Maxwellian distribution for the thermal neutron flux.

The diffusion coefficient:

$$D(E) = \frac{1}{3\left[\Sigma_a(E) + \Sigma_{tr}(E)\right]} \, . \tag{6}$$

The diffusion constant:

$$D_0 = \frac{\langle D(E) \rangle}{\langle 1/\upsilon \rangle} \quad , \tag{7}$$

where $\langle x \rangle$ is the Maxwellian-weighted average of $x(E)$.

Program SIGSA has been used (DROZDOWICZ and KRYNICKA 1995) to calculate the neutron parameters. It includes the microscopic absorption σ_a and scattering σ_s cross sections after MUGHABGHAB *at al.* (1981, 1984).

3. USE OF MATERIALS FOR THE Σ_a TEST MEASUREMENTS WITH CZUBEK'S METHOD.

The absorption rate $\langle \upsilon \Sigma_a \rangle$ or – after a recalculation – the mass or linear macroscopic absorption cross section can be determined experimentally by CZUBEK's (1981,1996) method. The measuring set-up for the method exists at the pulsed 14 MeV neutron generator in the INP (DROZDOWICZ *et al.* 1993). The experiment can be performed in a spherical or cylindrical geometry.

All the measurements up till now were made in the sample-moderator systems in which both the media were hydrogenous. Water solutions or crushed rocks saturated with water solutions were the samples. Plexiglass was always used as the outer moderator. In those cases the thermal neutron scattering properties of the sample and moderator were similar. In the case when the inner sample is of a material which does not contain hydrogen the scattering properties are significantly different from the properties of Plexiglass. Therefore, it is interesting to discuss a possibility to do test measurements for Czubek's method in the case when the inner sample is not hydrogenous.

A single sample of well defined size $\widetilde{R}_1$ (a sphere, a cylinder) is needed for a determination of the Σ_a of the investigated material. The sample is surrounded by the Plexiglass moderator. The decay constant λ_0 of the thermal neutron flux in the system is measured at a few different external sizes $\widetilde{R}_2$ of the outer moderator. The curve $\lambda_0(\widetilde{R}_2)$ obtained is a base for the determination of the Σ_a for the material of which the sample is made. Namely, the absorption rate $\langle \upsilon \Sigma_a \rangle$ of the sample is obtained from the intersection of the curve $\lambda_0(\widetilde{R}_2)$ with another one, $\lambda_0^*(\widetilde{R}_2)$, which is calculated theoretically for the system under the assumption that the dynamic material buckling in the inner region is equal to zero (cf. CZUBEK 1981). This makes the curve $\lambda_0^*(\widetilde{R}_2)$ being dependent only on the neutron parameters of the outer moderator. A condition of the measurement method is that the absorption cross section of the investigated

sample is higher than of the moderator. Because the moderator set of the experimental set-up is made of Plexiglass the condition to be fulfilled is

$$\Sigma_a > \Sigma_{a\,\mathrm{Plexi}}\ , \tag{8}$$

that is, expressing in the absorption rates:

$$\langle \upsilon \Sigma_a \rangle > 4200\ \mathrm{s}^{-1}. \tag{8a}$$

Because of the apparatus possibilities the highest well-measured decay constant is $\lambda_{max} \approx 40\,000\ \mathrm{s}^{-1}$ (cf. DROZDOWICZ *et al.* 1993). Thus, the test material should be characterized by the absorption rate being in the range:

$$4200\ \mathrm{s}^{-1} < \langle \upsilon \Sigma_a \rangle < 40\,000\ \mathrm{s}^{-1}\ . \tag{9}$$

The above limitation makes that only nickel glass, boric glass, dense flint and the alloy of Al + 30%Ni can be considered among the materials listed in Table 1.

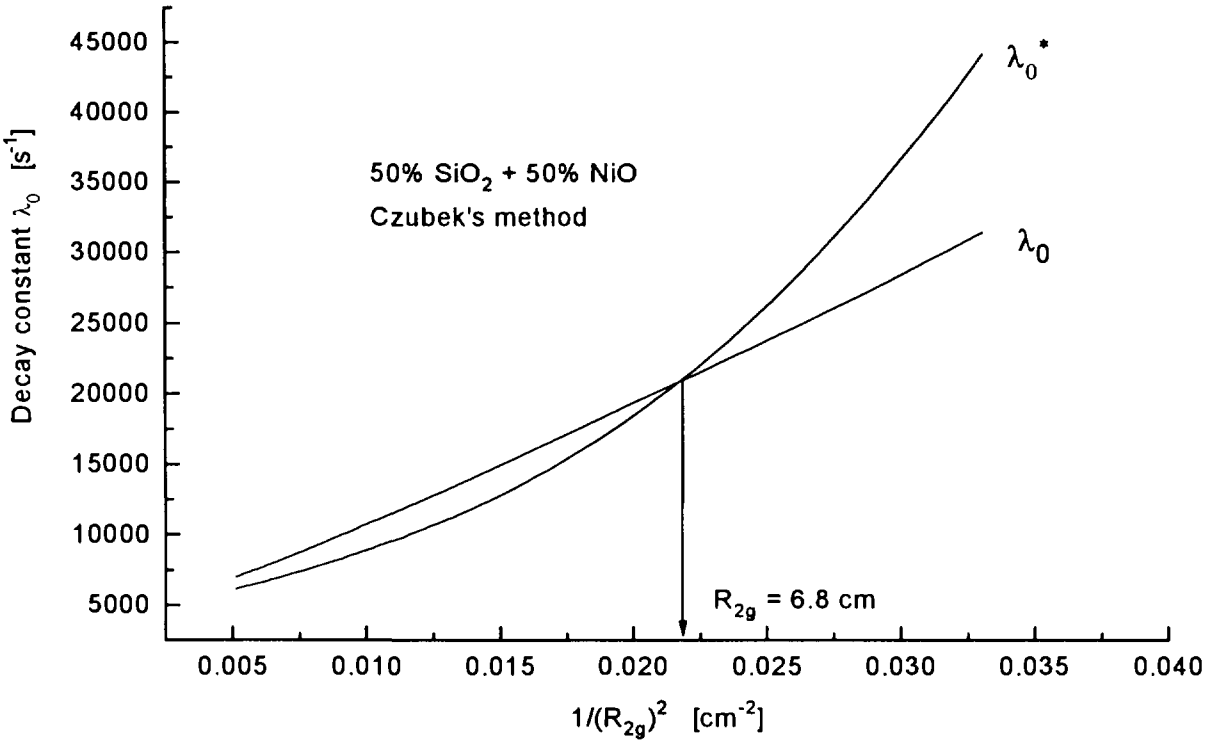

Fig.1. Curve $\lambda_0^*(1/R_{2g}^2)$ for Plexiglass moderator and calculated experimental curve $\lambda_0(1/R_{2g}^2)$ for the system: moderator with the sample of nickel glass (spherical geometry, $R_{1g} = 4.5$ cm).

In Fig.1 and Fig.2 the calculated dependencies $\lambda_0(1/R_{2g}^2)$ and $\lambda_0^*(1/R_{2g}^2)$ are shown which would be measured in Czubek's method for nickel glass and alloy Al + 30% Ni. The sample of the material was assumed as a sphere of radius R_{1g} = 4.5 cm which is surrounded by the spherical moderator of a variable size. The dependence $\lambda_0(1/R_{2g}^2)$ is plotted where R_{2g} is the external radius of the Plexiglass moderator.

For the sample of nickel glass (Fig.1) the intersection point of the curves gives the absorption rate $\lambda = \langle \upsilon \Sigma_a \rangle$ = 20 748 s^{-1} which corresponds to the external radius of the moderator R_{1g} = 6.8 cm. Measurements with moderators of the size in the range of R_{2g} = 8.2 cm to R_{2g} = 6.1 cm would be necessary to determine the curve $\lambda_0(1/R_{2g}^2)$ in a real experiment (The values of R_{2g} quoted correspond to the available set of the moderators).

For the sample of alloy Al + 30% Ni (Fig.2) the intersection point of the curves is λ = = $\langle \upsilon \Sigma_a \rangle$ = 15 779 s^{-1} which corresponds to the size R_{2g} = 7.5 cm of the moderator. In this

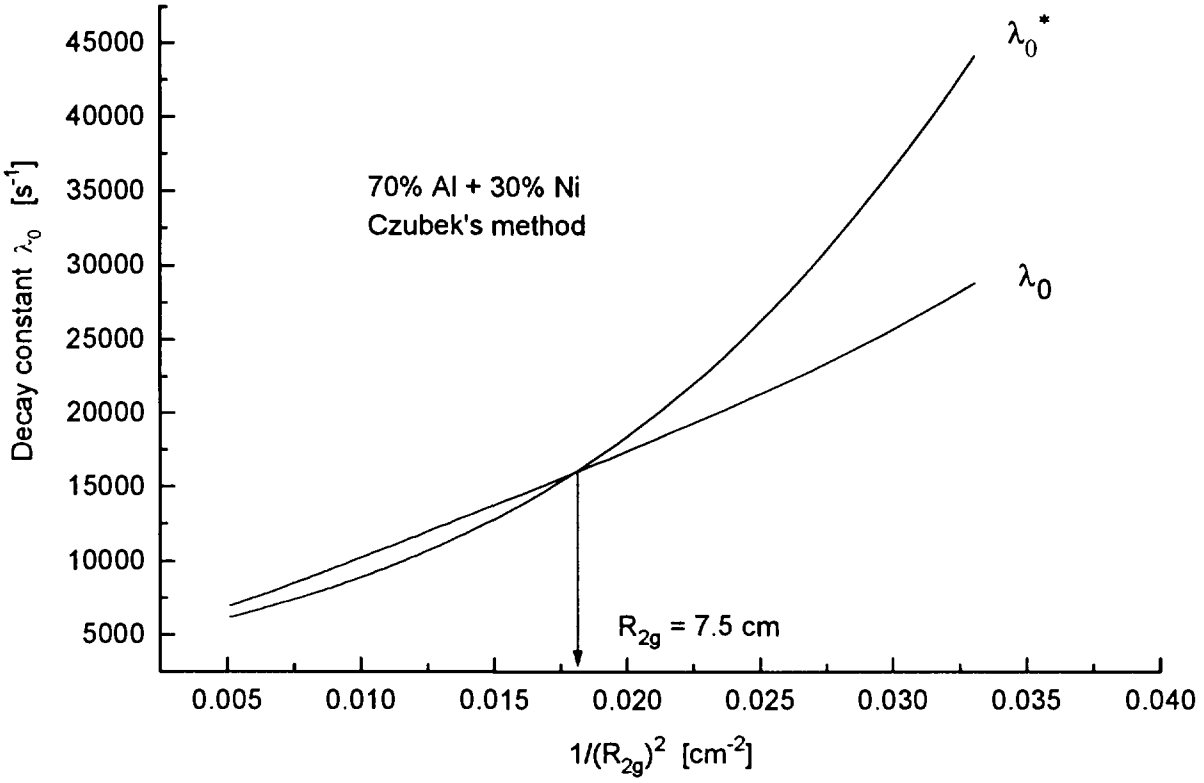

Fig.2. Curve $\lambda_0^*(1/R_{2g}^2)$ for Plexiglass moderator and calculated experimental curve $\lambda_0(1/R_{2g}^2)$ for the system: moderator with the sample of alloy Al + 30 % Ni (spherical geometry, R_{1g} = 4.5 cm).

case the real experiment for a determination of the curve $\lambda_0(1/R_{2g}^2)$ should be performed using moderators of the size from $R_{2g} = 9.1$ cm to $R_{2g} = 6.6$ cm.

It results from the calculation that the absorption rates of both the materials are in the measurable range.

Another test of the use of the selected materials for Czubek's method is an estimate of the intersection angle between the curves $\lambda_0(1/R_{2g}^2)$ and $\lambda_0^*(1/R_{2g}^2)$.

Formulae for a calculation of the intersection angle in the spherical geometry have been given by CZUBEK (1981). The calculation shows that the intersection angles for hydrogenous and non-hydrogenous samples (surrounded by a hydrogenous moderator) are close to each other and vary in the range of 5 to 6 degrees depending on the size of the sample. When the sample of the radius R_{1g} = 4.5 cm is made of the nickel glass 50% SiO + 50% NiO and surrounded by the moderator of Plexiglass the angle is equal to 5.8 degree.

4. USE OF MATERIALS FOR THE VARIABLE BUCKLING EXPERIMENT.

An idea of the classic experiment based on the variable geometric buckling is following. The medium is irradiated with the fast neutron burst and the thermal neutron flux is observed after the fast neutrons are slowed down in the system. This flux decays in time due to the neutron absorption and scattering (leakage). The fundamental mode decay constant λ_0 is connected to the thermal neutron diffusion parameters in the medium and to the geometric buckling (*e.g.* BECKURTS and WIRTZ 1964):

$$\lambda_0 = \langle \upsilon \Sigma_a \rangle + D_0 B_g^2 - C B_g^4 + \ldots \quad , \tag{10}$$

where:

C – diffusion cooling coefficient, [cm^4 s^{-1}],

B_g^2 – geometric buckling [cm^{-2}] which for a sphere of radius R_g is

$$B_g^2 = \left(\frac{\pi}{R_g + d_R} \right)^2 \tag{11a}$$

and for a cylinder of height H_g and radius R_g is

$$B_g^2 = \left(\frac{\pi}{H_g + 2d}\right)^2 + \left(\frac{j_0}{R_g + d_R}\right)^2 \qquad (11b)$$

where j_0 is the first zero of Bessel function of the first kind ($j_0 \approx 2.405$) and d and d_R are the extrapolated lengths.[1]

When the measurement of the decay constant λ_0 is performed at different values of the geometric buckling B_g^2 (*i.e.* at different sizes the sample) the thermal neutron parameters are obtained from a fit of Eq.(10).

The material used for these measurements should fulfil the following conditions:

a) the diffusion constant D_0 being possibly low, which involves a low slope of curve $\lambda_0(B_g^2)$,

b) the decay constants measured should not exceed $\lambda_0 > 40\ 000\ s^{-1}$ due to the mentioned possibilities of the apparatus set-up,

c) the values of the geometric buckling have to correspond to the range of geometric sizes of models which have a physical sense from the point of view of the thermal neutron diffusion, *i.e.*:

– the minimum size of the sample should be not less than $3l_D$ (cf. BELL and GLASSTONE 1970) where l_D is the thermal neutron diffusion mean free path:

$$l_D = \left\langle \frac{1}{\Sigma_a + \Sigma_{tr}} \right\rangle = 3\langle D \rangle, \qquad (12)$$

– the maximum size of the sample should be not greater than $(35 \div 40)\ l_D$; the samples of a greater size are close to an infinite medium considering the thermal neutron diffusion and an isolation of the fundamental mode decay constant is then difficult.

Estimates of the size of samples, of their masses and of the expected decay constant were made for the spherical geometry for a sake of simplicity. Eq.(10) has been approximated by

$$\lambda_0 \approx \langle \upsilon \Sigma_a \rangle + D_0 B_g^2 \qquad (13)$$

---

[1] The extrapolated distance d is proportional to the diffusion coefficient, $d = k\ \langle D \rangle$. The proportionality coefficient is dependent on the medium in which the thermal neutron diffusion occurs. For a flat boundary (the Milne problem) the following values are assumed: $k = 2.28$ for a hydrogenous medium and $k = 2.13$ for a non-hydrogenous one. If the boundary has a curvature c the extrapolated distance can be approximated by $d_c = d/(1+cd)$, which for the side surface of cylinder gives for the extrapolated distance accompanied with its radius R the expression: $d_R = d/[1+d/(2R)]$.

because the diffusion cooling coefficients of the materials considered are usually unknown. The minimum size $2R_{min}$ of samples of the selected materials has been estimated basing on the calculated mean free path l_D. A list is given in Table 2.

Four examples of the dependence of the decay constant λ_0 on the radius R_g of the spherical sample were calculated from the simplified formula Eq.(13): silica SiO_2 (100 %), nickel glass (50% SiO_2 + 50% NiO), aluminium alloy (70% Al + 30% Ni), and lead shot ∅ = 2 mm. They are plotted in Fig.3.

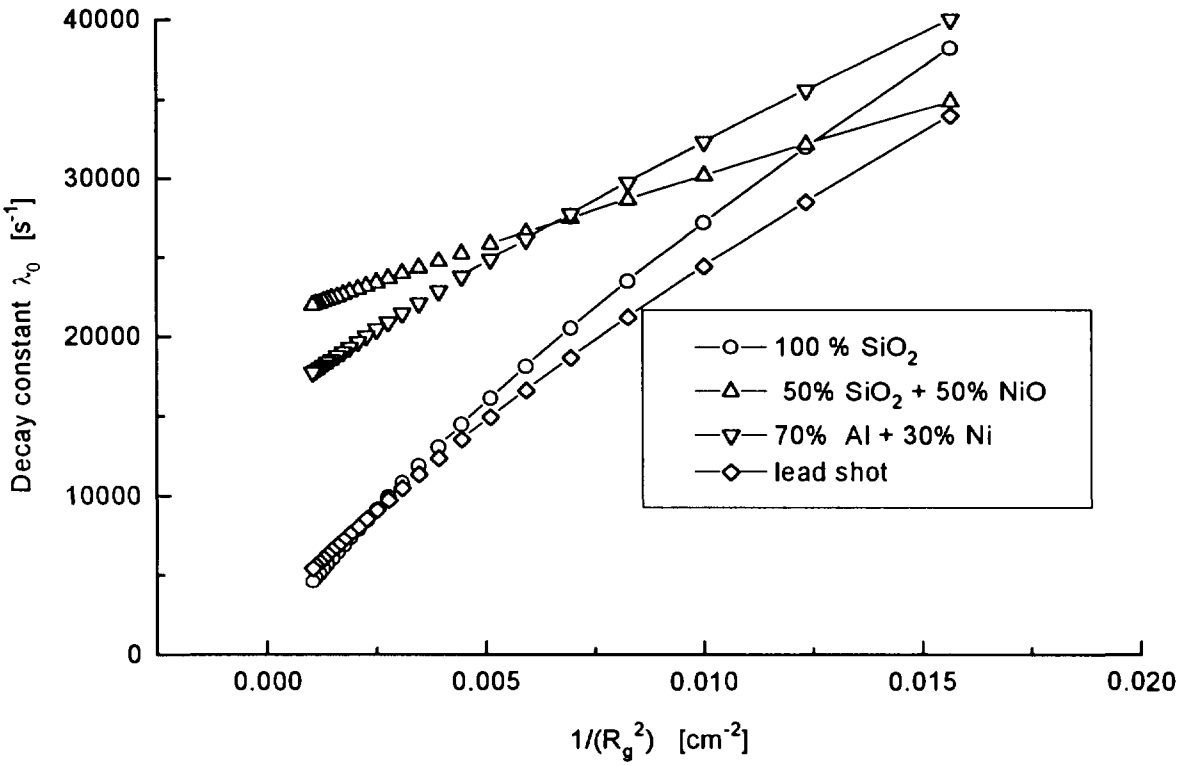

Fig.3. Decay constant λ_0 *vs* B_g^2 in spherical geometry for four selected materials.

Table 2. Thermal neutron mean free path and the minimum sample size for selected materials.

Material	l_D [cm]	$2\,R_{min}$ [cm]
Plexiglass	0.42	1.26
SiO_2	5.86	17.58
50%SiO_2+ 50%NiO	1.33	3.99
Al	10.23	30.70
70%Al +30%Ni	2.77	8.32
Fe (*porosity 40%*)	1.43	4.28
Teflon	3.35	10.04
Lead shot	4.30	12.9

5. ASSUMPTIONS FOR THE EXPERIMENT TO DETERMINE THE DEPENDENCE $\lambda_0(B_g^2)$ FOR THE LEAD GRAINS.

A certain problem exists to perform the classic experiment with the variable geometric buckling for a non-hydrogenous material when the system is irradiated by the fast neutron bursts and the resulting thermal neutron flux is observed. A non-hydrogenous material is usually a poor moderator (excepting a few elements) and the neutron mean free path is quite long. Therefore, the size of the medium has to be relatively large and the efficiency of the experiment is very low. However, we decided to use such a material to get an experience for that case. The aim of the experiment was to measure the decay constant λ_0 for at least two models of a chosen material of two different geometric sizes (*i.e.* of two different values of the geometric buckling).

From the third part of Table 1 the lead shot was selected as assuring conditions for our measuring system. The elemental composition of the lead alloy is given in Table 3. It was assumed after the producer certificate taking an average contents of elements from the given ranges and estimating the standard deviations from the limits of the range. The lead shot of grains of the diameter $\varnothing$ = 2 mm was available. Two decay constants λ_0 to be measured at different geometric buckling B_g^2 were estimated from Eq.(13) for two regular cylinders

Table 3. Elemental composition of the lead shot in weight per cent.

Element	[wt. %]
Pb	96.079 ± 0.3
Sb	3.0 ± 0.25
As	0.9 ± 0.02
Cu	0.02 ± 0.05
Sn	0.0003 ± 0.0001
Fe	0.0004 ± 0.0001

(H_g=2R_g). The thermal neutron parameters for the shot, calculated basing on the elemental composition from Table 4 at the minimum bulk density $\rho = 5.94$ g cm^{-3}, are:

$$\langle \upsilon \Sigma_{aPb} \rangle = 2048 \text{ s}^{-1}, \qquad D_{0Pb} = 412\,826 \text{ cm}^2\text{s}^{-1}.$$

The corresponding decay constants for two cylindrical samples are given in Table 4.

Table 4. Estimated decay constants for two cylinders of the lead shot.

Radius of the cylinder R_g [cm]	Mass of the sample m [g]	Decay constant λ_0 [s^{-1}]
14	71 900	14 625
7	17 130	30 954

Two cylindrical samples, Pb1 and Pb2, were prepared. Two cylindrical containers were filled with the lead shot. On the inner side they were covered with the 2 mm layer of cadmium to fix the vacuum boundaries for thermal neutrons. In the bottom an opening in the cadmium shield was made for the thermal neutron detector. The main parameters of the samples are given in Table 5.

Table 5. Geometrical and physical parameters of two cylindrical samples of the lead shot.

Parameter	Pb1	Pb2
$\overline{H}_g \pm \sigma(\overline{H}_g)$ [cm]	23.7 ± 0.1	14.6 ± 0.1
$\overline{R}_g \pm \sigma(\overline{R}_g)$ [cm]	11.8 ± 0.1	7.3 ± 0.1
$\overline{V} \pm \sigma(\overline{V})$ [cm^3]	10 410 ± 91	2461 ± 69
$m_{Pb} \pm \sigma(m_{Pb})$ [g]	71 312 ± 0.3	17 294 ± 0.1
$\rho_{Pb} \pm \sigma(\rho_{Pb})$ [g cm^{-3}]	6.85 ± 0.06	7.03 ± 0.20

5.1. Problem of the bulk density.

The bulk densities of samples Pb1 and Pb2 are different a little. Generally, a behaviour of the bulk materials causes that it is very difficult to get two samples of a different size and of the same bulk density. This results from the fact that an arrangement of grains in a volume is unrepeatable. Let us consider an isometric medium, *i.e.* grains being small spheres of equal diameters, made of the same homogeneous material of the solid material density ρ_0. Our lead grains fulfil this condition. The following two limiting cases of the arrangement of spheres exist (cf. CZUBEK 1968).

a) Simple cubic arrangement.

This is a metastable arrangement in which each sphere contacts with 6 neighbours. The pores created in this way are regular concave octahedrons. Then a porosity of the medium is maximal:

$$\phi_{max} = \frac{8R^3 - \frac{4}{3}\pi R^3}{8R^3} = 0.4764 \qquad (14)$$

which involves the minimal bulk density:

$$\rho_{min} = 0.5236\ \rho_0 \,. \qquad (15)$$

In the case of the lead shot considered this gives $\rho_{min} = 5.944$ g cm^{-3}.

b) Compact arrangement (cubic or hexagonal).

In this arrangement each sphere is tangential to 12 neighbours and the pores have a shape of regular concave octahedrons and tetrahedrons (the number of the second ones is twice greater). The resulting porosity is minimal:

$$\phi_{\min} = \frac{4\sqrt{2}R^3 - \frac{4}{3}\pi R^3}{4\sqrt{2}R^3} = 0.2595 \qquad (16)$$

and the bulk density is maximal:

$$\rho_{max} = 0.7405\ \rho_0\ . \qquad (17)$$

The density for the shot considered is $\rho_{max} = 8.405$ g cm^{-3}.

In Fig.4 the function $\lambda_0(1/R_{2g}^2)$ is plotted for two cylindrical models of the same material but different bulk densities. The ratio of the cylinder height to diameter is $H_g/2R_g = 1$. The curves in Fig.4 have been determined from the simplified dependence, Eq.(13), assuming the neutron parameters for the lead shot at the minimum and maximum bulk densities, respectively. The plot is pictorial, the diffusion cooling effect [cf. Eq.(10)] has not been included. However, the influence of the bulk density on the value of the decay constant of the thermal neutron flux is visible to be important and has to be considered in the experiments on the samples for which the bulk density can differ at different sizes of the samples.

5.2. Thermal neutron diffusion parameters for samples of different bulk densities.

A method to avoid an influence of fluctuations of the bulk density of the sample on the measurement of the function $\lambda_0(B_g^2)$ is shown below (cf. CZUBEK 1997). Dividing both sides of Eq.(10) by the density ρ one obtains:

$$\frac{\lambda_0}{\rho} = \frac{\langle \upsilon \Sigma_a \rangle}{\rho} + \rho D_0 \frac{B_g^2}{\rho^2} - \rho^3 C \frac{B_g^4}{\rho^4} + \ldots \qquad (18)$$

which can be written as

$$\tilde{\lambda}_0 = \langle \upsilon \tilde{\Sigma}_a \rangle + \tilde{D}_0 \tilde{B}_g^2 - \tilde{C} \tilde{B}_g^4 + \ldots \qquad (19)$$

using the following notations:

$\langle \upsilon \tilde{\Sigma}_a \rangle = \dfrac{\langle \upsilon \Sigma_a \rangle}{\rho}$ is the density-removed absorption rate (or the 'mass absorption rate', following the nomenclature of the cross section), independent of the material density [cf. Eqs (1) and (5)];

$\tilde{D}_0 = \rho D_0$ is the density-removed diffusion constant which defined in this way is also independent of the material density because the diffusion coefficient D [Eq.(6); (4)] and the related diffusion constant D_0 [Eq.(7)] are inversely proportional to the material density;

$\tilde{C} = \rho^3 C$ is the density-removed diffusion cooling coefficient because the diffusion cooling coefficient itself, C, is proportional to $1/\rho^3$ (*e.g.* BECKURTS and WIRTZ 1964).

In a similar way the generalized decay constant:

$$\tilde{\lambda}_0 = \frac{\lambda_0}{\rho} \qquad (20)$$

and the generalized geometric buckling:

$$\tilde{B}_g^2 = \frac{B_g^2}{\rho^2} \qquad (21)$$

are defined. Then the thermal neutron pulsed parameters obtained from a fit of Eq.(19) to the experimental data are the density-removed parameters (or the mass parameters), $\langle \upsilon \tilde{\Sigma}_a \rangle$, $\tilde{D}_0$, $\tilde{C}$, for a material and can be re-calculated to the linear parameters, $\langle \upsilon \Sigma_a \rangle$, D_0, C, for a given density of the material.

A change of the bulk densities cannot be too big because the external cases (*e.g.* pure lead and lead shot) involve a significant change of the scattering properties of the medium. In the experiment referred the bulk densities of samples Pb1 and Pb2 were different of 2.6 % which is admissible.

5.3. Measurement of the thermal neutron decay constant λ_0 of samples Pb1 and Pb2.

A bloce scheme of the measurement system is shown in Fig.5. The thermal neutrons from the volume of the sample investigated find their way through the opening in the cadmium

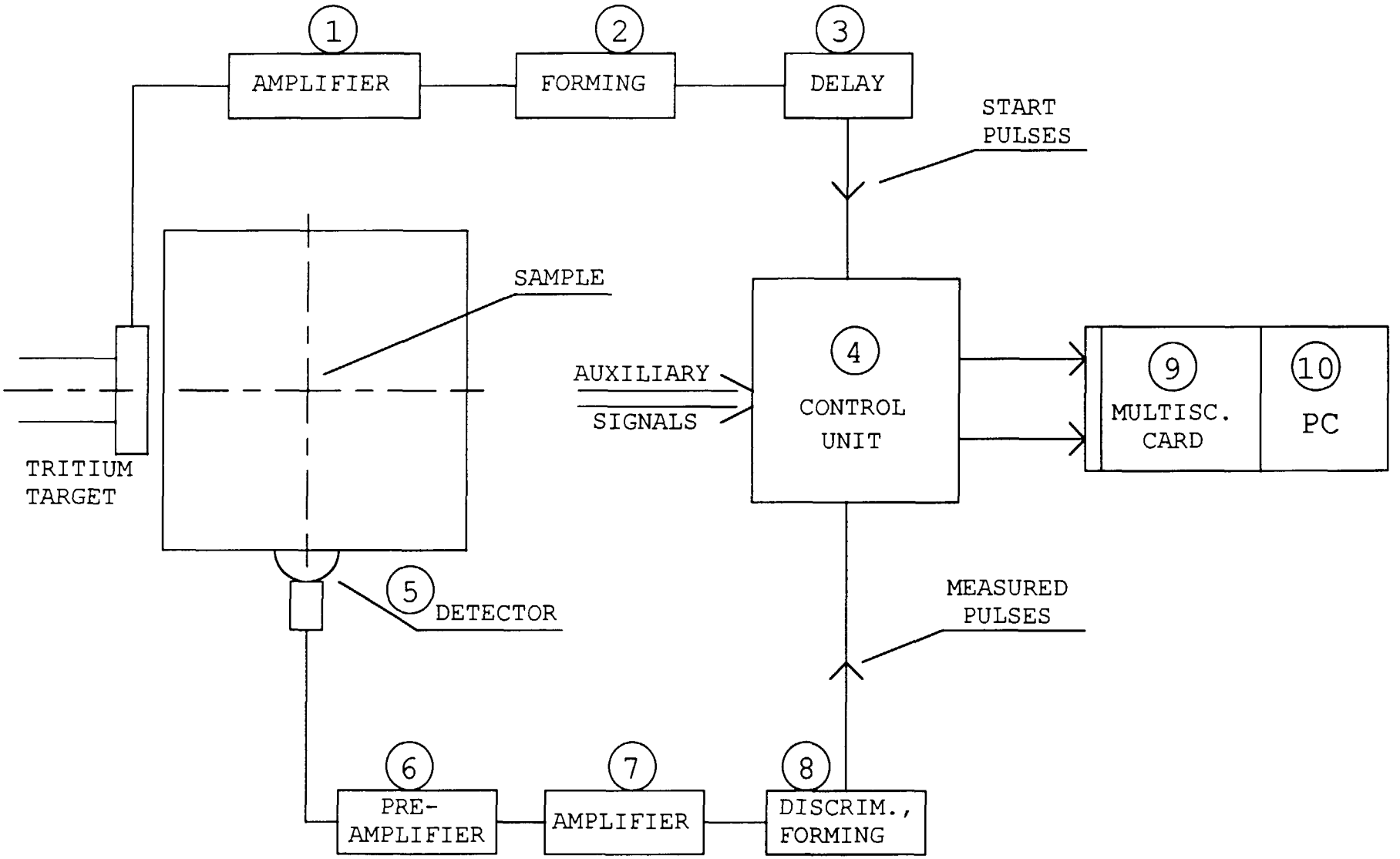

Fig5. Block scheme of the measurement system

shield into the ^{3}He detector (5). The pulses from the detector are amplified and shaped in the preamplifier (6) and amplifier (7). After the amplitude discrimination (8) the standardized pulses come into the input of the time analyser (9) (a multiscaler regime). A PC card is used, the AccuSpec FMS Canberra. The pulse for a start of each analysis run is obtained from the ion current of the fast neutron generator when the deuteron burst reaches the target. These pulses, after the amplification (1) and shaping (2), are relevantly delayed by the delay unit (3) and sent to the control unit (4) of the measurement system and from it to the start input of the multiscaler (9). The control system (4) allows the start pulses to be transmitted only if proper conditions of the measurement are fulfilled (the temperature of the sample is kept, the average thermal neutron beam is in the required range, the background is sufficiently low, etc., and some auxiliary technical devices at the neutron generator work properly). In this way the measuring cycles being not of a full value are excluded. The control system allows also to stop the experiment when either a given time or a given number of cycles or a given total number of measured pulses are reached.

The final results of the measurements of the decay constants for two cylindrical models of the lead grains are given in Table 6. Eight measurements of the decay constant were performed for sample Pb1 (Table 7). As the final result the weighted mean λ_0 of these measurements was taken with the estimator of the standard deviation σ of the mean λ_0 (cf. DROZDOWICZ *et al.* 1995). For sample Pb2 only three measurements were made (Table 8). The average λ_0 from those measurements was taken as the final result and the standard deviation was estimated from the data range.

Table 6. Decay constants measured for samples Pb1 and Pb2.

Sample	$\overline{H}_g \pm \sigma(\overline{H}_g)$ [cm]	B_g^2 [cm^{-2}]	λ_0 [s^{-1}]	$\sigma(\lambda_0)$ [s^{-1}]
Pb1	23.7 ± 0.1	0.0337	13 473	324
Pb2	14.6 ± 0.1	0.0705	18 458	1253

Table 7. Individual results for sample Pb1.

λ_0 [s^{-1}]	13 792	13 451	13 913	13 057	13 294	13 615	13 397	12 302
$\sigma(\lambda_0)$ [s^{-1}]	262	338	276	212	1 077	941	257	389

Table 8. Individual results for sample Pb2.

$\lambda_0[s^{-1}]$	19 291	18 962	17 120
$\sigma(\lambda_0)\ [s^{-1}]$	1 497	435	2 027

6. CONCLUSIONS.

A difficulty to find a proper non-hydrogenous material for the time-dependent thermal neutron experiments is visible from the review presented in the paper. Such a testing material has to be characterized by the neutron parameters relevant for the experiment and also has to have a very precisely defined composition to be able to compare any calculational and experimental results.

At the moment it was not possible to make of a non-hydrogenous material a test sample for Czubek's method of the absorption cross section measurement. Pure materials, as cooper or iron, are omitted in the discussion because of the absorption cross section which is too high to be measured on the existing experimental set-up. Only artificial mixtures like those presented in Figs 1 and 2 would be useful in the experiment. The cost of manufacturing the materials is high because of a unique production.

The buckling experiment with non-hydrogenous material appeared more realistic. However, it was also difficult to find a proper material. Pure metal samples are to heavy when a set of samples of different dimensions has to be used. One of the lead shot samples used in the experiment was about 71 kg. A pure lead sample of the same size is more than twice heavier. The selection of lead shot to measure the buckling curve was a compromise to get the material cheep enough and having the neutron parameters sufficiently proper for the experiment.

The experiment was difficult because of a very poor counting statistics. Bad slowing-down properties of the lead were a reason of a very small population of thermal neutrons in the sample volume. A very small amount of thermal neutrons was detected by a neutron counter after each fast neutron burst. An isolation of the fundamental mode time decay constant of thermal neutrons was difficult and loaded with a high error. An accumulation of data in a multiscaler took many hours, which caused that the apparatus error had an important impact.

Two experimental points measured are of course not enough to determine three pulsed neutron parameters: the absorption rate, the diffusion constant, the diffusion cooling coefficient. The last one is of the most interest because of lack of experimental data and of a theoretical prediction.

A future research is planned. Further points on the buckling curve can be calculated using a Monte-Carlo program to simulate the time decay process of neutrons in the given sample size. The points obtained from the present real experiment will be test points for the computer simulation. When the buckling curve is obtained the calculation of diffusion cooling coefficient will be possible. Later it will be possible to use for other non-hydrogenous materials the checked simulation procedure and the pulsed neutron parameters can be determined. This will also give an opportunity to simulate Czubek's method with non-hydrogenous samples.

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