

# Evaluation and test of the response matrix of a multisphere neutron spectrometer in a wide energy range \*

## Part I. Calibration

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A multisphere neutron spectrometer has been developed. It consists of a cylindrical <sup>3</sup>He thermal neutron counter and a set of 11 spherical moderators made of polyethylene with diameters ranging from 2 to 15 in. The spectrometer has been calibrated in two thermal and eight monoenergetic neutron beams (8.2 keV–14.8 MeV). The spectrometric system is described, and the results of the calibration are discussed. The thermal and high energy regions are well covered but there is a significant gap at intermediate energies. A comparison of the results with those of similar works is presented. The differences obtained underline the necessity for a proper specific calibration for each multisphere spectrometric system.

### 1. Introduction

The spectrometric information on the neutron radiation field is very important in radiation protection since the neutron's quality factor depends on its energy and because the response of existing neutron monitors and personnel dosimeters is energy-dependent. The need for a good knowledge of the neutron spectral distribution has been emphasized by the recent ICRP publication 60 [1], which recommends a reduction of the annual dose limit and an increase of the neutron's quality factor (now energy weighting factor), for certain energies. These changes lead to a decrease of the required detection limit of neutron dosimeters which underlines the importance of the precise determination of the neutron dose equivalent.

In areas where the neutron spectra present a great variability, neutron spectrometry is the only method that allows to understand the measuring devices' behaviour, and which permits the accurate evaluation of the dosimetric quantities relevant for radiation protection.

Different systems have been developed for neutron spectrometry; the most commonly used are based on threshold detectors (activation or fission), <sup>3</sup>He propor-

tional counters, LiI scintillators enriched in <sup>6</sup>Li, recoil proton detectors, and the time-of-flight method [2]. For radiation protection purposes, particularly inside nuclear facilities, the multisphere spectrometer is the best suited instrument in the thermal and intermediate energy range.

The aim of the present work is to establish as accurately as possible the response matrix of a Bonner spheres system using a cylindrical <sup>3</sup>He counter. This paper deals with the description of the system and its calibration in thermal and monoenergetic beams; it will be followed by two companion papers, one presenting the calculation of the spheres' response functions over the whole energy range of interest and their adjustment to the experimental points, and the other one presenting the experimental validation of the response matrix in reference neutron spectra.

### 2. The multisphere spectrometer

The multisphere spectrometer, called also Bonner spheres system [3], has been the subject of several reviews [4–6]. The spectrometer is based on the use of a thermal neutron detector surrounded by moderating spheres of different diameters, usually made of polyethylene. The fast neutrons are slowed down in the moderator and reach the detector as thermal ones, while the thermal neutrons initially present in the field are mostly captured in the moderator. Therefore when

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the moderating sphere's diameter increases, the maximum sensitivity of the system moves to higher energies, making possible to perform neutron spectrometry.

The count rate  $M_i$  from the sphere  $i$  in a given neutron field is obtained by integration of the product of the sphere's response function  $F_i(E)$  with the neutron fluence spectral distribution  $\Phi_E(E)$ . The use of a number  $N_d$  of spheres of different diameters in an unknown neutron field leads to the following set of equations:

$$M_i = \int_{E_{\min}}^{E_{\max}} F_i(E) \Phi_E(E) dE, \quad i = 1, \dots, N_d. \quad (1)$$

The resolution of the system with respect to  $\Phi_E(E)$ , called unfolding, is performed by means of numerical methods.

The thermal neutron detector used in the first Bonner spheres system [3] was a small volume scintillator of lithium iodide enriched in  $^{6}\text{Li}$ . Due to the small volume of the detector the neutron sensitivity of the system was very low, and an increase of the crystal dimensions was restricted because it leads to an increase in the relative sensitivity to photons.

The replacement of the scintillator by proportional counters filled with  $^{10}\text{B}$  trifluoride [7–10] or  $^{3}\text{He}$  [11–17] of different geometric shapes allowed an increase of the neutron sensitivity by a factor of 10, with a very low photon sensitivity.

Passive detectors have also been used in Bonner spheres, in order to measure very intense pulsed neutron fields such as those encountered around particle accelerators [18–20], or in the case where a low intensity neutron field requires a very long integration time such as in some environmental measurements. The types of passive detectors employed include activation detectors sensitive to thermal neutrons, pairs of  $^{6}\text{Li}$  and  $^{7}\text{Li}$  fluoride thermoluminescent detectors, and track detectors with radiators made of  $^{10}\text{B}$ ,  $^{6}\text{Li}$  or  $^{235}\text{U}$ .

The multisphere spectrometer shows several characteristics useful for radiation protection measurements in nuclear facilities (power plants, experimental reactors and particle accelerators). Its main advantages are:

- the functional simplicity;
- the wide energy domain (from thermal energies to several MeV);
- the quasi-isotropic response;
- the high neutron sensitivity, allowing the measurement of low dose equivalent rates encountered in radiation protection (down to  $1 \mu\text{Sv/h}$ );
- the good discrimination of electronic noise and photon counting by a suitable choice of the counter and adjustment of the associated electronics.

Due to the shape of the response functions, the energy resolution of the system is rather low, but judged satisfactory for the evaluation of the dosimetric quantities used in radiation protection.

The accurate evaluation of the response matrix is essential for a correct use of the spectrometric system. The impact of the matrix uncertainty on the different dosimetric quantities computed from the spectrum has been emphasized by several authors [21–23].

The experimental calibration of the system with neutron beams of known energy, usually monoenergetic, represents an important step in the determination of the spectrometer's response matrix.

### 3. Materials and methods

#### 3.1. Description of the multisphere spectrometer

The spectrometric system studied in the present work [24] consists of 11 moderating spheres made of polyethylene with diameters of: 2, 2.5, 3, 4.2, 5, 6, 8, 9, 10, 12 and 15 in. (from 5.08 to 38.1 cm). The polyethylene density is  $0.916\text{--}0.918 \text{ g cm}^{-3}$  according to the

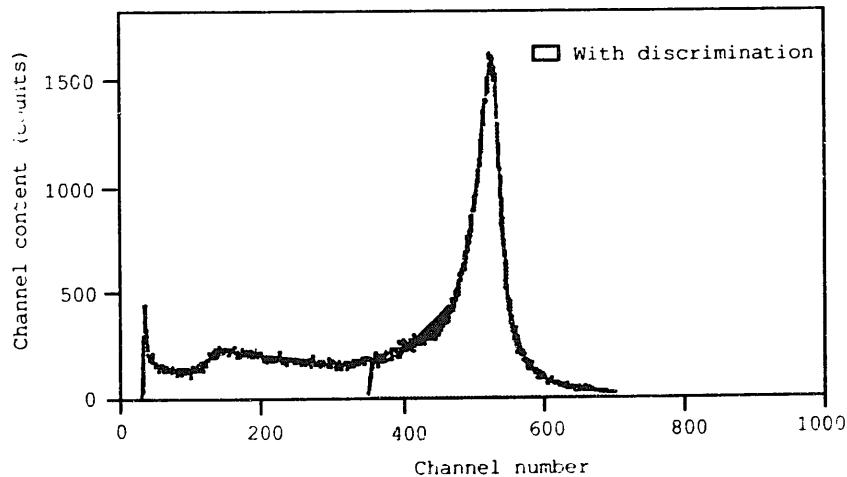


Fig. 1. Pulse spectrum of the  $^{3}\text{He}$  bare counter irradiated with thermal neutrons.

manufacturer (SEIV Atmostat, France). The  $^3\text{He}$  counter is introduced in a radial cylindrical duct. The counter's active volume is located in the centre of each sphere. A second 5 in. sphere is used to monitor the variation of the neutron fluence.

The spheres with diameters smaller than 6 in. can be covered by a cylindrical cadmium box with a diameter of 200 mm, a height of 200 mm and a thickness of 1.4 mm, in order to absorb the thermal component of the incident neutron spectrum.

The thermal neutron detector used with the spheres is a miniaturized proportional counter filled with  $^3\text{He}$  (type 0.5NH10, produced by LCC, Déetecteurs Nucléaires Thomson-CSF, France). Its active volume is an orthocylinder with a diameter of 9 mm and a height of 10 mm. The outer wall is made of monel (copper and nickel alloy) with a density of  $8.4\text{--}8.8\text{ g cm}^{-3}$ . The filling pressures given by the manufacturer are:  $8 \times 10^5\text{ Pa}$  for  $^3\text{He}$ ,  $2 \times 10^5\text{ Pa}$  for Kr and traces of a quenching gas. The manufacturer also guarantees a  $^3\text{He}$  content of 99.7% and a maximum tritium content of  $2 \times 10^{-9}\text{ vol\%}$ .

The electronics consist of a charge preamplifier located close to the counter, linked through 20 m long cables to a set of standard NIM modules (linear amplifier, single channel analyzer, counter, timer, high voltage supply). This allows the user to stay in a safe area during the measurements.

### 3.2. Electronic noise and $\gamma$ ray discrimination

The measurements performed in order to determine an optimal discrimination level which ensures a good rejection of noise and  $\gamma$  rays without affecting the neutron sensitivity too much have shown that the level corresponding to the beginning of the second plateau in the counter's pulse spectrum is satisfactory. Fig. 1 shows the superposition of the pulse spectra obtained with and without discrimination, after an irradiation of the bare counter with thermal neutrons.

The discrimination threshold indicated in fig. 1 produces a loss in the neutron sensitivity of 24% relative to the situation where the whole first plateau of the counter's pulse spectrum is also retained. At the same threshold level the counter's  $\gamma$  ray sensitivity is very low (0.01 counts/ $\mu\text{Sv}$ ) for  $^{60}\text{Co}$  and much lower for  $^{137}\text{Cs}$ . The nominal value for the counter's neutron sensitivity given by the manufacturer is 0.5 counts/( $\text{n cm}^{-2}$ ) for thermal neutrons. This corresponds to  $6 \times 10^4$  counts/ $\mu\text{Sv}$  and leads to a relative neutron-gamma sensitivity of  $6 \times 10^6$ . Even in the worst case when the counter works in a hard spectrum such as that of Am-Be and the measured neutron sensitivity is as low as 1 counts/ $\mu\text{Sv}$ , the relative neutron-gamma sensitivity of the counter remains of the order of 100.

Table 1

Monoenergetic neutron beams used for the calibration of the multisphere spectrometer at PTB

Reaction	Energy [MeV]
$^{45}\text{Sc}(\text{p}, \text{n})^{45}\text{Ti}$	0.0082
$^7\text{Li}(\text{p}, \text{n})^7\text{Be}$	0.144, 0.250, 0.570
$\text{T}(\text{p}, \text{n})^3\text{He}$	1.2, 2.5
$\text{D}(\text{d}, \text{n})^3\text{He}$	5
$\text{T}(\text{d}, \text{n})^4\text{He}$	14.8

### 3.3. Calibration beams

#### 3.3.1. Monoenergetic beams

The multisphere spectrometer has been calibrated with several monoenergetic neutron beams of energies between 8 keV and 14.8 MeV produced by a Van de Graaff accelerator at the Physikalisch-Technische Bundesanstalt (PTB) in Braunschweig (Germany). The reactions used are presented in table 1.

The measurements were performed 6 m above the concrete ground of a low scatter room of dimensions  $25 \times 30\text{ m}^2$ . The shadow cone technique was used in order to account for the scattered radiation. The neutron fluence was monitored by means of a De Pangher long counter, a cylindrical monitor ( $\varnothing 288 \times 310\text{ mm}^2$ ) equipped with two  $\text{BF}_3$  counters and another cylindrical monitor ( $\varnothing 50 \times 233\text{ mm}^2$ ) equipped with a  $^3\text{He}$  counter [24].

#### 3.3.2. Thermal neutron beams

The response of the Bonner spheres at thermal energies has been measured in two different thermal neutron beams: at PTB and at the Centre d'Etudes Nucléaires (CEN) in Cadarache (France).

The thermal neutron reference beam of the PTB research reactor is produced by placing a Be scatterer in the primary beam. The neutrons are then guided outside the reactor by a collimating system. A 20 cm thick Bi filter is inserted into the beam in order to reduce its contamination with high energy neutrons and  $\gamma$  rays generated by neutron-capture reactions in the scatterer and the collimator. The Cd difference method is used to separate the residual 2% contribution of intermediate and high energy neutrons. The thermal beam has a mean energy of 0.028 eV [25]. An external 1.35 m boron-loaded wood collimator provides a beam with a well-defined exit profile ( $\varnothing 32\text{ mm}$  at 90% of the maximum counting rate). The spheres' cross section being much larger than that of the beam, a uniform irradiation in a broad parallel beam has been simulated by scanning. The beam position is fixed in space and the spheres are placed on a mobile table. The table movements and the data acquisition are

controlled by computer. A more detailed description of the whole system is presented in ref. [26].

The thermal neutron calibration at CEN (Cadarache) took place near the SIGMA reference pile. The facility consists of a graphite cube (150 cm side) crossed by an irradiation channel with a diameter of 8 cm. Up to 12 calibrated Am-Be sources (592 GBq each) can be disposed around the central cavity. The pile is mounted on an elevated metallic floor. The Cd difference method is also used to separate the contribution of intermediate and high energy neutrons.

#### 4. Results and discussion

The calibration results are presented in fig. 2 for each energy separately and the numerical values are given in the appendix; they represent the spheres' response expressed in number of counts to a unitary neutron fluence. The results of the measurements per-

formed in the reference neutron thermal beams at CEN and at PTB are presented together; the differences between them are significant. This is due to the fact that the two beams do not have the same neutron temperature. These results are discussed in detail in Part 3 (Validation) of the present paper.

The fluence values communicated by the PTB were already corrected for the attenuation in air. The results are also corrected for the dead time and the contribution of scattered radiation (evaluated by the shadow cone method) has been subtracted. The angle subtended by the spheres was less than 5° with respect to the beam axis and thus the anisotropy effect was judged to be negligible. For neutron beams with energies of 144 keV and upwards, the uncertainty in the results is determined by the systematic uncertainty of the fluence monitors ( $\pm 2.5\%$ ) and the statistical uncertainty in their counting rate. The latter is situated between 1 and 3% depending on the beam energy. One should add to these two components the uncertainty in the

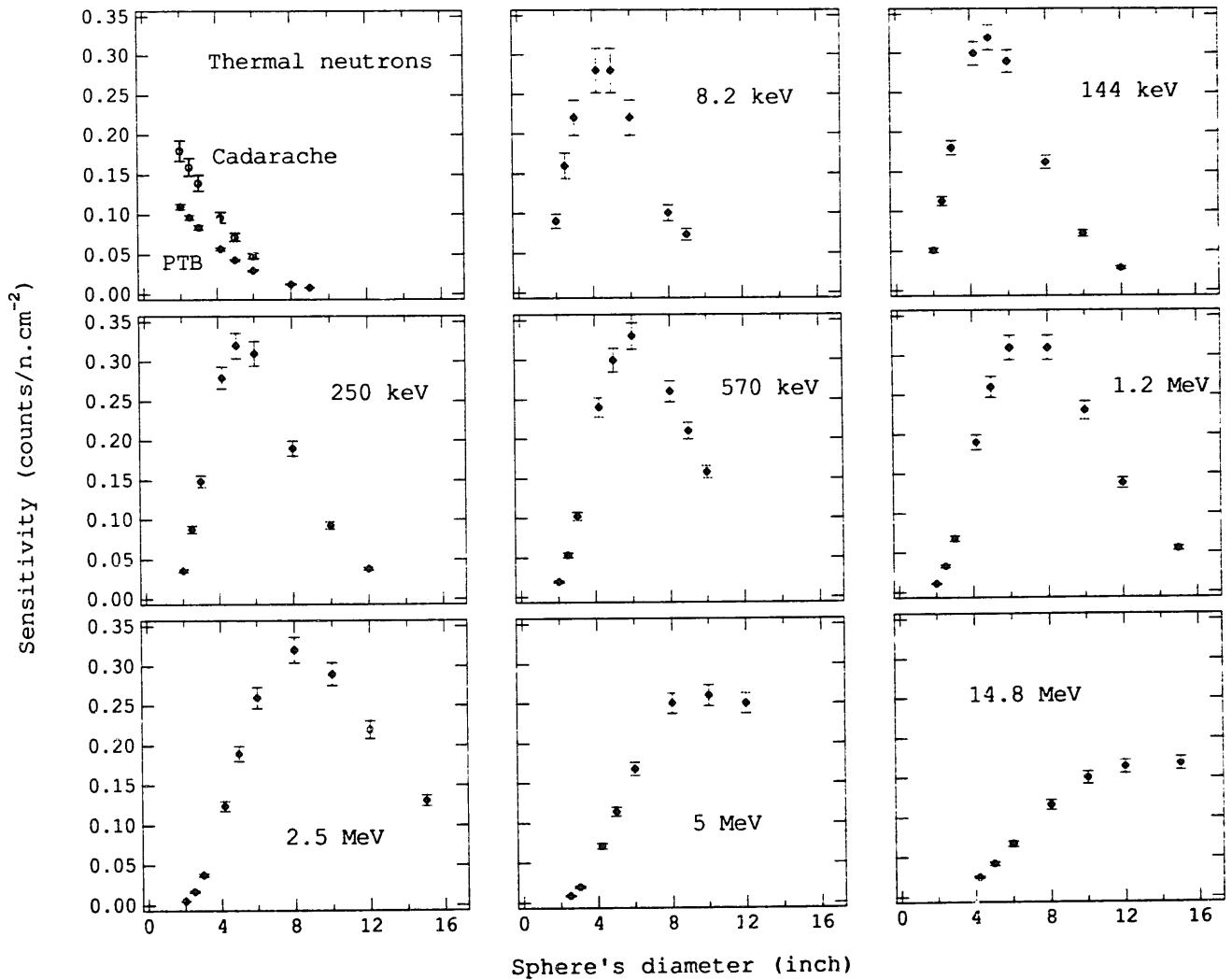


Fig. 2. Results of the experimental calibration of the Bonner spheres system.

spheres' counting rate, which was always less than 1% (typically 0.3%). Consequently, the global uncertainty in the results for beams with energies of 144 keV and above can be considered to be of the order of  $\pm 5\%$ .

For the 8.2 keV beam the fluence rate was very low; this has led to larger statistical uncertainties in the spheres' counting rates (up to 2%) and a relatively low distance between the target and the spheres. In this geometry the contribution of the neutrons scattered by the spheres to the monitor's indication ( $\approx 3\%$ ) could not be neglected and the fluence values given by the PTB have been corrected for this effect. The global uncertainty in the neutron fluence for this beam energy is of the order of  $\pm 10\%$ .

The uncertainty in the neutron fluence made in the thermal beam at CEN is of the order of  $\pm 6.5\%$ . In this beam the measurements were performed on spheres with diameters up to 6 in. because the Cd box cannot accommodate larger spheres. The uncertainty in the thermal neutron fluence at PTB is of the order of  $\pm 3\%$ . The calibration was performed on spheres with diameters up to 9 in.

Fig. 3 presents the calibration results versus neutron energy for five spheres. The shift of maximum sensitivity on the energy scale appears clearly. The lack of experimental points between thermal energies and the keV region is obvious. This problem, common to all the Bonner spheres' calibration works reported in the literature, underlines the importance of the calculation of the spheres' response functions in order to fill the gap. This will be discussed in detail in Part 2 (Simulation) of the present paper.

Among the calibration works performed so far, those of Caizerques et al. [12] and Alcvra et al. [17] are particularly interesting since they deal with the same kind of neutron thermal counter as ours ( $^3\text{He}$ , cylindrical, type 0.5NH10). Ref. [17] presents the results of a

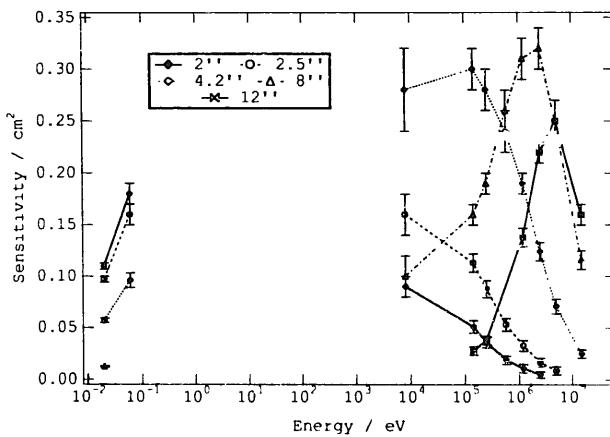


Fig. 3. Experimental response of five Bonner spheres versus neutron energy.

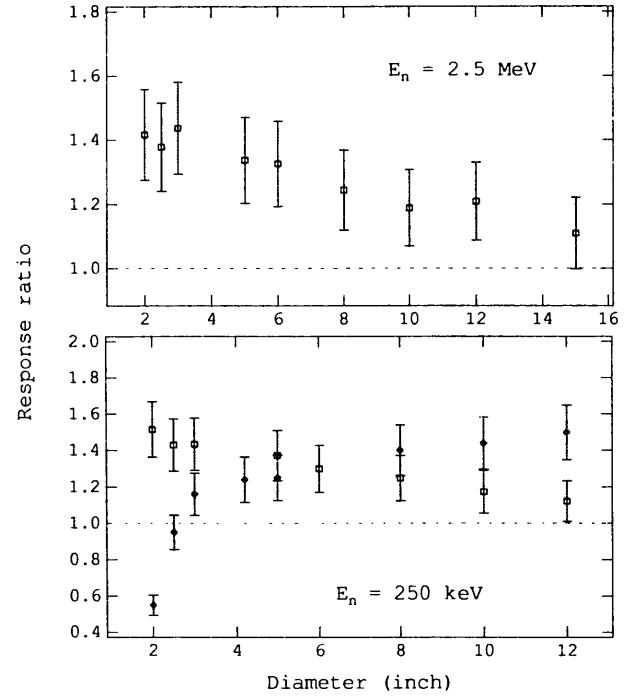


Fig. 4. Comparison of the present experimental results with others reported in the literature (see main text).  $\square$  Alevra et al./present work.  $\blacklozenge$  Caizerques et al./present work.

joint programme for the experimental calibration with monoenergetic neutrons of four multisphere spectrometers involving the PTB (Germany), National Physical Laboratory (NPL, England), and Gesellschaft für Strahlen- und Umweltforschung (GSF, Germany). Only one of the four systems studied uses a cylindrical  $^3\text{He}$  counter. A comparison of the experimental data obtained with this system showed that its response is higher than our system's. At all energies, the difference between the two systems is more pronounced for small sphere diameters. Two typical curves indicating the variation of the systems' response ratio versus the sphere diameter are shown in fig. 4; the ratio varies from about 1.1 for large diameters to about 1.5 for small ones.

The comparison of Caizerques et al. results with ours is possible only for the unique common calibration energy (250 keV). The variation of the systems' response ratio versus the sphere diameter is also shown in fig. 4. It indicates that for this energy the Caizerques et al. system is about 50% more sensitive than ours for the 12 in. sphere, of equal sensitivity around a diameter of 2.5 in. and about 50% less sensitive for the 2 in. sphere.

The difference in response for various multisphere spectrometers using the same thermal neutron counter can be attributed to several factors such as:

- a possible variation of the counter's filling pressure;

- a difference in the polyethylene density ( $0.946 \text{ g cm}^{-3}$  for the system of Alevra et al. and  $0.916 \text{ g cm}^{-3}$  for ours, 3% difference);
- a different discrimination threshold for the counter's pulses, which might be the most important factor (section 3.2).

The effect of the first two factors on the system's response will be discussed in detail in Part 2 (Simulation) of the present paper.

## 5. Conclusion

The multisphere spectrometer developed in the Institute for Applied Radiophysics has been calibrated with thermal neutrons both at CEN (Cadarache) and PTB (Braunschweig) and with several monoenergetic neutron beams at PTB. The number of calibration points is reasonably high and covers the main available energies, which should improve the determination of the system's response matrix, but there is a significant gap at intermediate energies. The results of the calibrations were presented with an evaluation of the experimental errors, and compared with other results reported in the literature. The comparison revealed that a number of parameters can influence the system's response. This underlines the necessity for a proper specific calibration for each multisphere spectrometric system. In the second part of this paper the results presented here will be used together with calculated response functions in order to establish the system's response matrix.

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## Appendix

**Table A**  
Numerical results of the experimental calibration of the Bonner spheres; sensitivity in counts/n cm<sup>-2</sup>

$\emptyset$ [in.]	Thermal beam at CEN	Thermal beam at PTB	8 keV	144 keV	250 keV	570 keV	1.2 MeV	2.5 MeV	5 MeV	14.8 MeV
2	0.18	0.11	0.09	0.051	0.036	0.019	0.011	0.0052	-	-
2.5	0.16	0.097	0.16	0.113	0.088	0.053	0.033	0.0172	0.009	-
3	0.14	0.084	0.22	0.18	0.149	0.102	0.068	0.038	0.020	-
4.2	0.096	0.057	0.28	0.30	0.28	0.24	0.19	0.124	0.071	0.025
5	0.072	0.043	0.28	0.32	0.32	0.30	0.26	0.19	0.114	0.042
6	0.048	0.03	0.22	0.29	0.31	0.33	0.31	0.26	0.168	0.067
8	-	0.0126	0.10	0.162	0.19	0.26	0.31	0.32	0.25	0.116
9	-	0.0083	0.073	-	-	0.21	-	-	-	-
10	-	-	-	0.072	0.093	0.158	0.23	0.29	0.26	0.150
12	-	-	-	0.028	0.038	-	0.138	0.22	0.25	0.164
15	-	-	-	-	-	-	0.055	0.131	-	0.168