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CHARACTERISTICS OF PREPARED GAMMA-RAY
CALIBRATION SOURCES FOR RADIOACTIVITY MEASUREMENT
OF ENVIRONMENTAL AND RADIATION CONTROL SAMPLES

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Characteristics of Prepared Gamma-ray Calibration Sources for
Radioactivity Measurement of Environmental and Radiation Control Samples

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Several types of calibration source having different density were prepared using one or combinations of those materials, namely foam cement, liquid, glass beads, polystyrene foam bead and hard plastic bead for gamma-ray spectrometry of the samples with different densities and shapes(variable height with constant base area). For each type of the source, a few sources were prepared to examine characteristics in such cases as (a) different heights but constant density, and (b) constant height and constant density. For the foam cement source, several sources with different densities and a constant height were prepared. All the sources were measured with a gamma-ray spectrometry system and the results were discussed. This report also presents the results obtained from the experiments for the evaluation of (1) the variation of detector efficiency-energy with gamma-ray energy, and (2) the variation of detector efficiency with density of the sources.

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Keywords: Gamma-ray Calibration Sources, HPGe Detector, Efficiency-energy Curve, Efficiency-density Curve, Radioactivity Measurement, Environmental and Radiation Control Samples, Foam Cement, Glass Bead, Polystyrene Foam Bead, Hard Plastic Bead.

環境及び放射線管理試料の放射能測定のために調整した γ 線校正用線源の特性

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密度及び形状（底面積を一定とし試料高さを変化）の異なる試料のガンマ線スペクトロメトリーの校正のために、気泡セメント、液体、ガラスビーズ、発泡ポリスチレンビーズ及び硬質プラスチックビーズを材料とし、これらを単品または組合せて密度の異なる数種類の校正用線源を作製した。個々の種類の線源毎に（a）同じ密度で異なる高さ、（b）同じ高さで同じ密度、にした場合の特性を調べるため線源は数個ずつ用意した。気泡セメント線源については、異なる密度で同一の高さのものを数個用意した。全ての線源は γ 線スペクトロメトリー装置で測定し、その結果の検討を行った。本報告では、また、（1） γ 線エネルギーによる検出効率曲線の変化、及び（2）特定の γ 線エネルギー毎に密度による検出効率の変化、の評価のために行った実験結果について述べる。

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1. INTRODUCTION

In JAERI, a centralized system of radioactivity measurement has been utilized for radiation control purposes since 1980. In this system, various types of samples collected at various sampling points are measured with equipments such as alpha/beta counters, alpha-, beta-, x- and gamma-ray spectrometers, and liquid scintillation counters. Quality control measures for these equipments are regularly practiced for the purpose of achieving reliability of results, both in precision and accuracy.

The types of samples collected from the controlled areas and the environment for the monitoring of radioactivity can be categorized into several types by their original forms. For example; smear papers, carbon cartridge filters, waste water and solid samples such as soil, chemically processed marine and land products.

1.1 Samples and calibration sources

To determine accurate value of radioactivity in the samples, suitable calibration sources which have a similar shape, composition and density to the measured sample are necessary¹⁾. The quality control scheme in practice is shown in Fig.1.1 for example.

1.2 Foam cement calibration source

The foam cement source is prepared in such that it has the same properties in shape and density as the sample to be measured. The status of the source is a mixture of foam and cement. In the preparation of this source, which will be explained in more detail later (in Section 2.2.2(2)), the amount of introduced foam can be adjusted so that the resultant source density is equal to the sample density. The possible range of density is from 0.2 to 2.1 gcm⁻³.

The suitability of this foam cement source as a calibra-

tion source for radioactivity measurement has been tested precisely and accurately²⁾. Although the preparation of the foam cement source is convenient and not a problem to this laboratory, it is somewhat complicated in other laboratories. This is especially true when it comes to the stage of homogenizing the diluted radioactive source in the foam cement.

For this reason, it is thought useful (besides this foam cement source) to have other types of calibration sources, which are more convenient to prepare, for the purpose of radioactivity measurements.

1.3 Objectives of this work

The main objectives of this experimental work are (1) to study the characteristics of self-prepared calibration sources depending on their shape, composition and density, and (2) to study the reproducibility of source preparation method.

Three sets of calibration sources, from six types (or compositions), namely foam cement, liquid, calibration sources with matrix of glass bead³⁾ of 6 mm in diameter, glass bead of 2 mm in diameter, polystyrene foam bead and hard plastic bead respectively are prepared. Set A has characteristics of constant density but different heights, Set B of different densities and constant height (only applicable to cement source), and Set C of constant density and constant height.

The study for the characteristics of the calibration sources depending : (a) on shape and composition, is done by evaluating the efficiency-energy curve of Set A, and (b) on density, is done by evaluating that of Set B. For the study of the reproducibility of source preparation, the efficiency-energy curve of Set C is evaluated.

For a constant source height, calibration sources from sets A, B and C are used to study further the characteristics of the calibration sources depending on composition and density, by evaluating the efficiency-density curve for each

gamma-ray energy.

On the basis of the understanding of these characteristics, a simple preparation task of suitable calibration source as a standard for accurate radioactivity determination of any environmental and radiation control samples will be preferable. This work is also aimed to find the most suitable calibration source as intercomparison material among laboratories.

The factors that determine the suitability of a calibration source may be as follows : (1) the availability of material to be used as the composition, (2) the reproducibility of the preparation method, (3) radionuclide uniformity (homogeneity) and (4) precision and accuracy. Factors (1) and (2) are especially needed for other laboratories. For factor (3), as our laboratory has previously worked and experienced with foam cement source, which radionuclide homogeneity is the prime concern due to its manner of preparation, it is also the purpose of this work to check its validity by comparing its results with the results obtained from other calibration sources. The second and final factors are also taken into account in this work.

Finally, it is hoped that some of the prepared calibration sources, if not all, will be routinely used as calibration standards in the radioactivity measurement of environmental and radiation control samples.

2. PREPARATION OF CALIBRATION SOURCE

2.1 Radioactivity standard

2.1.1 Radioactivity standard solution

Throughout this work, a mixed-source radioactivity standard solution supplied by DAMRI/LMRI (Ref no: 9ML01-ELMB60), comprising ten radionuclides and thirteen major gamma-ray energies was used. On receiving this source, its volume is 5 cm³ and it is contained in a flame-sealed glass ampoule. In this report, this source is sometimes referred as a master solution.

The certified data of this standard source, which yields the type of radionuclides, their half-lives, energies (keV), absolute intensities (%), photon emission rate per unit mass of solution (4π steradians g⁻¹s⁻¹) is given in appendix A. The necessary values used in the calculation of activity of any nuclide are taken from this data.

2.1.2 Handling of ampoules

Before using this source, it is however the usual practice of this laboratory to transfer the whole amount of the source into several polyethylene ampoules (of volume 3 cm³), for the purposes of safety, sampling convenience and sampling accuracy. This is especially true since gravimetric sampling is applied for this type of source, due to its activity certification (per unit mass of solution). The types of ampoules used in this work are shown in Photo 1. On receiving the source, it is transferred to several ampoules of P-1. Then the top section of P-1 is heated to form P-2. It is in this form of P-2 that the source is kept, before it is in use. When later the source is to be used, the top section of P-2 ampoule is heated to have the neck drawn out to form a capillary as P-3. Similar method is used to transform P-4 to P-5. The form of P-3 and P-5 is respectively called pycnometer of 3 cm³ and 50 cm³ in volume. The application and advantage of these pycnometers will be

described in the later section. All the work of transforming the polyethylene ampoules from P-1 to P-2 and P-2 to P-3 is carried out in the draft (fumed-cupboard) of the hot laboratory.

2.2 Calibration source preparation

The types of all the calibration sources to be prepared are shown in Table 2.1. It is obvious from the compositions that the process of preparing the sources involves four stages of work, namely (1) calculations prior to preparation, (2) preparation of matrix materials, (3) preparation of diluted radioactive source, and (4) introduction of the diluted source to the matrix material. These stages of work can be divided into two main tasks, i.e. preparation work carried out outside a hot laboratory (stages 1 and 2), and inside a hot laboratory (stages 3 and 4). This section describes these two main tasks.

The complete form of all types of the calibration sources will only be achieved after going through these stages. Photo 2 shows a set of the complete form of all types. Before these sources are ready to be measured, each of them with matrix of beads needs to be stuck with an outer and inner acrylic lids (the shape of this lid is shown in Fig.2.1) with the use of epoxy adhesive, then wrapped with a plastic bag. The procedures are intended to prevent radioactive contamination from the sources.

The same containers of 9.5 cm in diameter that are currently been used for measuring environmental samples such as sand, soil and milk are used. Notice that for this type of container, the heights of 1.5 cm, 3.0 cm and 4.5 cm correspond respectively to the volumes of 100 cm³, 200 cm³ and 300 cm³ (as shown by the x-axis of Fig.2.2). These volumes do not correspond to the formula for calculating the cylindrical volume, i.e. $\pi D^2 H / 4$, where D and H are the respective diameter and height in cm. This is due to the smaller diameter at the container bottom, in comparison with the

container top (of which diameter of 9.5 cm is quoted). It will be seen in the next section that this work considers these three heights. Also in this work, the term of height simply represents the meaning of volume or shape.

2.2.1 Work carried-out outside hot laboratory

(1) Calculations prior to preparation

Since this work mostly involves with quantities like mass, density and volume, it is most helpful to write the relationship of these quantities, as it would be seen later, the understanding of the calculation would become more clear, i.e.

$$\text{density} = \text{mass/volume} \quad (1)$$

Eqn (1) yields that if a constant density is to be obtained for volumes of 100, 200 and 300 cm³, the exact proportion of masses must be achieved. To see the exact proportion of masses to these volumes, Fig.2.2 gives the graphical meaning of eqn (1) for six different densities. In the preparation of the sources (shown in Table 2.1 as T1, T2, T3A, T3B, T4 and T5), a target has been set, that is to produce calibration source sets with following three properties :

(a) to prepare three calibration sources (from each type) which represent three different heights of 1.5, 3.0 and 4.5 cm, that have a constant density. Fig. 2.2 shows examples of six constant densities for these three heights. The number of sources to be prepared here is 18.

(b) to prepare five calibration sources from the foam cement type only which represent five different densities, that have a constant height of 4.5 cm. The number of sources to be prepared here is 5.

(c) to prepare three calibration sources for each type, that have a constant height of 4.5 cm to examine reproducibility. The number of sources to be prepared is 18.

But if the value of density in this case is made equal to that in the case (a) of height of 4.5 cm, the number of

sources that now need to be prepared becomes 12 (=18-6). In addition, if one of the foam cement sources in case (b) (of course they are all 4.5 cm in height) is made to have this value of density, the number of sources to be prepared becomes 11 (=12-1).

The total number of calibration sources henceforth that need to be prepared is 34 (=18+5+11). The three characteristics of (a), (b) and (c) simply represent the main objectives of this work, i.e. in respect to (1) study the variation of the efficiency-energy curve with composition and height of the calibration sources, for a constant density, (2) study variation of the efficiency-energy curve with density of the foam cement source, for a constant height, and (3) study the reproducibility of the efficiency-energy curve with composition of the calibration source, for a constant density and height.

(2) Problems in preparing sources with a constant density

In practice it is not simple to prepare sources that have characteristic of a constant density. As described above, this involves cases of (a) a constant density but different heights, and of (c) a constant density and a constant height. Examples of cases in (a) are given in Fig.2.2 with the same symbols. As seen in this figure and Table 2.1, to make a constant density, it also means : in the case of (a), the source net mass (i.e. the sum of mass of matrix materials plus the mass of diluted master solution) of the three calibration sources must be accurately determined in proportion with the intended heights of 1.5, 3.0 and 4.5 cm, and in the case of (c), the source net mass of the three calibration sources must be accurately determined to be equal (for a constant height of 4.5 cm). A slight deviation of total mass will result in three values of density and not a constant. However, since the obtained values of density are quite close to each other (almost equal), it is assumed that they have a constant density value.

The above problem is not encountered in the case of (b) described above, i.e. constant height but different densities, since a constant height can be achieved by method of direct measurement. However, to have different densities and constant height, it simply depends the availability of a matrix material that is capable of changing its mass only, without changing its volume (since a constant volume is needed here). This is only possible in the case of foam cement source, since foam can be used to change its mass, without changing its volume. Other calibration sources are not possible since changing their mass also means changing their volume.

(3) Preparation of matrix material

This stage of work only concerns with the matrix material of the bead sources³⁾. The actual form of the variety of beads used in this work is shown in Photo 2. The glass beads are made of soda-lime glass (SiO_2 65-75, Al_2O_3 0.5-4, Fe_2O_3 <2, CaO 5-15, MgO 0.5-4 and Na_2O 10-20 %).

This work plans to make several types of samples (T3A, T3B, T4 and T5) to have the properties of (a) constant density at different heights (namely, 1.5 cm, 3.0 cm and 4.5 cm), and (c) a constant density and at a constant height (4.5 cm). Beads (i.e. T3A, T3B, T4 and T5) are filled tightly into the container until it reached the intended height. When this process is over, the mass of beads (in g) is then determined by the use of a digital balance (Mettler PM 4000). To prevent beads from moving or floating (especially in the case of polystyrene foam when the diluted radioactive solution is introduced later) a piece of PVC mesh and an inner lid are put on top of the beads. This procedure is illustrated in Fig.2.1.

2.2.2 Work carried-out inside hot laboratory

(1) Dilution of radioactivity standard solution

Dilution of the radioactivity standard solution means a process of dispensing a known amount of the master solution (in g) to a 1000 cm³ polyethylene bottle that contains a diluting solution (i.e. of distilled water with 0.5 mol/l HCl). The bottle is then shaken slowly and carefully to homogenize the solution.

When the diluting solution has been prepared, the dilution of master solution may be started before or after the preparation of the matrix material. However, in the case of foam cement source, the dilution work must be done earlier, for the reason described in section 2.2.2(2) below.

In this dilution work, a gravimetric sampling method⁴⁾ is used, with the help of two digital chemical balances (Mettler:PC 2200 and AE 163). The shape of pycnometer P-2 which contains the master solution is transformed into pycnometer P-3. The end of the capillary is then cut, and a known amount of master solution (in g) is dispensed to the diluting solution.

It is also useful to have a formula written here for the purpose of calculation of the activity (disintegration s⁻¹) of a known amount of diluted master solution dispensed to a certain calibration source, in terms of known amount of master solution and known amount of diluting solution. If, in the preparation, M_{ms} g of master solution was dispensed to a polyethylene bottle that contains M_{ds} g of diluting solution, then a diluting factor D_f for the diluted master solution exists, which equals to M_{ms}/(M_{ms}+M_{ds}). If in the preparation the condition of M_{ms} << M_{ds} is valid, then D_f approximately becomes M_{ms}/M_{ds}. If now M_{dms} g of diluted master solution (from the polyethylene bottle) is dispensed to a calibration source, the activity A_c of the calibration source is given by

$$A_c = M_{dms} \times (A_c^* \times D_f) \text{ s}^{-1}$$

where A_c* is the initial activity at time zero of a certain radionuclide that is contained in the master solution. In

practice, correction for radioactive decay are necessary.

A list of parameters for reference solution is presented in Appendix A with a numeral example. In Appendix A, notice that the value given in column 5 is the activity of a certain radionuclide emitting gamma-ray with a particular energy.

(2) Introduction of diluted radioactive solution to the matrix material

The diluted radioactive solution is now ready to be poured to the matrix materials of (i) foam cement and (ii) beads. The following sections describe the procedure of introducing the diluted master solution into these matrices.

The steps for the liquid source are simply a method of directly filling of the diluted source to the container, using a P-5 pycnometer. The amount (in g) of the diluted solution is determined by the use of the chemical balance.

(i) Cement as matrix material (plus water and foam)

In the case of the foam cement, since cement powder added with water will get hard quickly, it is thought wiser to prepare the matrix material of this type of source in the hot laboratory. For reference the element of cement and its contents are : SiO_2 20-25, Al_2O_3 4-6, Fe_2O_3 2-4, CaO 62-66, MgO 1-2 and SO_3 1-2 %.

The method of preparation for foam cement source is similar to the one described by Kawasaki et. al²⁾ and Manabe⁵⁾.

The foam is first made. By letting water ready in a beaker, chemical agents making the foam solution (S-100 and S-200) are dropped into it, followed by smoother (which is introduced in stages, a little amount at a time), to make a foam solution. A known amount of this solution is put into a smaller bowl. Foam can be obtained from this solution by putting a mixer into the bowl and drive it (to allow the intake of air) at high speed. Photo 3 shows the form of foam which is ready to be transferred (with known amount) to

the cement paste.

Secondly the foam cement is prepared. A known amount of both cement powder and water are put together into the large bowl. By the use of mixer and spatula, they are mixed slowly, until they form a cement paste. This stage of work is shown in Photo 4. By mixing homogeneously a known amount of foam to the cement paste, the foam cement is formed. Notice that a different amount of foam added to the cement paste will produce a different density of the foam cement.

When this matrix material is ready, a known amount of diluted master source (in g) is dispensed to the known amount of the foam cement to produce a calibration source.

Prior to this preparation, it is important to know beforehand the amount of component material (i.e. water, cement, and foam) that is needed for the intended amount of source volume and density. The recipe for this calculation is given in Appendix B.

(ii) Bead as matrix material

The diluted radioactive solution is poured carefully through the hole of the inner lid of the container, by using the pycnometer of 50 cm³ in volume (shown as P-5 in Photo 1). This process is stopped when the solution reaches the PVC mesh. The amount of the diluted master solution (in g) poured into the container is determined by the use of the balance. The outer lid is then used to cover the top of container(see Fig.2.1). As a safety precaution, this outer lid is stucked to the container by the use of epoxy adhesive.

2.2.3 Illustration of data in the preparation work

From Fig.2.2, the source net mass (y-axis) refers to the sum of component masses of the compositions of the calibration sources. The compositions of the calibration sources are given in Table 2.1. However, for the purpose of illustrations, these composition are repeated here, i.e. (1)

Net mass of the foam cement type (T1) = sum of masses of cement, water, foam, carrier HCl, distilled water and master solution (2) Net mass of the liquid type (T2) = sum of masses of HCl, distilled water and master solution, and (3) Net mass of bead types (T3A or T3B or T4 or T5) = sum of masses of beads, HCl, distilled water and master solution.

In the case of T1, if for example a constant density of 0.97 gcm^{-3} for source heights of 1.5, 3.0 and 4.5 cm is needed, Fig.2.2 gives the exact values of source net mass for these heights. This means for each of this height, the net mass (sum of masses of component compositions) must be precise. However, as been mentioned earlier about the problem of constant density, it can be seen that in Appendix C, data on this work has several density values, and not a constant. This is due to the slight deviation in values of source net mass (from the exact one, as shown in Fig.2.2). But since these values are very close to each other, it is assumed that a constant density is obtained.

The data in Appendix C also shows the source net masses for T2, T3A, T3B, T4 and T5 and their respective densities, for different heights.

The values in Appendix C can be represented graphically, as shown in Fig.2.3 (a), (b) and (c), in terms of heights, densities and types (or compositions) of calibration sources. Notice that only foam cement source can have different densities, and not other sources, as shown in Fig.2.3(b).

3. MEASUREMENTS

3.1 Gamma-ray spectrometry system

The measuring system consists of a vertical coaxial HPGe detector(SEIKO EG&G, Model GMX-45200, SN 32-N 30628A, 45% relative efficiency at 1332 keV of Co-60) connected to a 3.5 keV negative HV power supply(EG&G ORTEC 459), amplifier (EG&G ORTEC 572), and PC-based MCA(NEC-PC-9801-VX). The computer is installed with analysis software for data manipulation. A radiation shielding constructed with lead of 10 cm thickness is used around the detector to minimize contribution from background radiation.

In all measurements, the cylindrical source is placed axially above the detector, as shown in Photo 5, where the source to the detector distance is fixed at 5 cm to avoid the effect of cascade summing⁶⁾ and to achieve a very low counting dead-time (0.5%). The measuring time is also fixed to 5000s for the purpose of accuracy (uncertainty less than 1%) of the accumulated area under the photo-peak.

The automatic sample changer system, which is routinely used for radioactivity measurement of samples, is also applied, for the purpose of checking the accuracy of the results (obtained from the manual system). This system is shown in Photo 6.

3.2 Data analysis

3.2.1 Spectrum-analysis software

An analysis software of SEIKO EG&G Co., Ltd. (1987) GMX Version 3.30 is used for the manipulation of measurement data. This software will perform the necessary calibrations and yield the empirical function of : (1) energy as a function of channel number, (2) FWHM as a function of energy, and (3) channel number as a function of energy. It is also capable of performing the peak search, and eventually yielding the information on identification of the nuclides, their photo-peak net area and uncertainty.

3.2.2 Efficiency-energy and efficiency-density curves of calibration source

From the experiment the information on the registered net gamma-ray counts A_n in the peak area of energy E accumulated in live time T (sec) will be obtained. Consequently from Appendix A, the number of emitted gamma-rays A_t from the diluted master solution in time T can be obtained. The value of A_t may be calculated from $A_c f T$, where A_c is the source activity at the time of measurement and f is the gamma emission rate at energy E . The absolute efficiency E_{abs} for energy E (keV) may then be obtained from $E_{abs} = A_n / A_t$. Notice that this equation is similar to the equation mentioned in Fig. 1.1.

Since the diluted master solution yields thirteen major gamma-ray energies, this also means that for each calibration source, thirteen pairs of efficiency-energy data (E, E_{abs}) will be obtained. And since for a constant height of 4.5 cm, all types of calibration source yield ten density (D) values (as seen in Fig. 2.3), thirteen pairs efficiency-energy data (E, E_{abs}) for each density will also be obtained. These data pairs however can be rearranged in such that ten pairs of efficiency-density data (D, E_{abs}), for each gamma-ray energy, are obtained.

3.2.3 Fitting function for the curves

In plotting the experimental data pairs of (E, E_{abs}) for each calibration source and (D, E_{abs}) for each gamma-ray energy, a software of Ngraph Version 5.3⁷⁾ was used. This software is also capable of plotting the fitting curve to the data and yielding the empirical function of E_{abs} as a function of E , for $60 \text{ keV} < E < 1836 \text{ keV}$; and E_{abs} as a function of D , for $0.21 \text{ gcm}^{-3} < D < 2.04 \text{ gcm}^{-3}$. These functions are very useful as the calculation E_{abs} for any energy and density within these ranges will be made possible.

4. RESULTS AND DISCUSSION

Figures 4.1, 4.2, 4.3, 4.4, 4.5 and 4.6 respectively show the variation of three efficiency-energy curves with heights (for a constant density), for calibration sources of foam cement, liquid, glass bead (6 mm in diameter), glass bead (2 mm in diameter), polystyrene foam bead, and hard plastic bead. The plotted data for the gamma-ray energies of 136.5 and 320.1keV differ from the curves, compared with those for the other energies. The result is based on analysis of the small peak area due to the short half-life and weak gamma-ray emission rate of corresponding nuclides. Therefore, the uncertainty should be taken into account to estimate the efficiency-energy curves. It can be seen in all the figures that the efficiencies decrease when the height is increased, as expected.

Fig.4.7 shows the variation of five efficiency-energy curves with density (for a constant height), for five foam cement sources. It can be seen that the efficiencies decrease when the density is increased, as expected. It can also be noted that the efficiency separation at low energy is wider than at the high energy, which implies the significance of the self-attenuation in the source, at low energy.

Figures 4.8, 4.9, 4.10, 4.11, 4.12 and 4.13 respectively show the variation of three efficiency-energy curves with heights (for a constant density), for calibration sources of foam cement, liquid, glass bead (6 mm in diameter), glass bead (2 mm in diameter), polystyrene foam bead, and hard plastic bead. From all the figures it can be seen that the three efficiency curves are situated very close to each other, as if they are obtained from a single source. This implies that the reproducibility of our method of source preparation is good.

As seen in Fig.2.3, for a height of 4.5 cm, all the prepared calibration sources have a density in the range of 0.21 to 2.04 gcm⁻³. Henceforth if the appropriate experimental data shown in figures 4.1 to 4.13 are taken to plot the

efficiency versus density for this constant height, the variation of efficiency-density curves with the thirteen gamma-ray energies in the region of 59.54 to 1836 keV, for all types of calibration sources, can be seen. For this purpose, all the thirteen efficiency-density curves were first plotted, then for each curve, the quality of the correlation (i.e. the agreement of data from all the types of source to make a curve) was evaluated. It is found that quality of correlation can be grouped into three categories, namely, bad (for gamma-ray energy of 59.5 keV, as shown in Fig.4.14), poor (for energies of 88.03, 122.1, 136.5, 165.9 and 320.1 keV, as shown by samples in Fig.4.15), and good (for energies of 391.7, 514.0, 661.7, 898.0, 1173, 1332 and 1836 keV, as shown in Fig.4.16). In the case of the poor correlation, since all the curves are close to each other, only two curves with small uncertainty (for the energies of 122.1 and 165.9keV) are selectively shown in Fig.4.15.

The bad correlation as shown in Fig.4.14 is first discussed. The types (or compositions) of the calibration source which contributed to this data are also indicated in the figure. Notice that the dotted line shows the fitting of data due to the foam cement source only. It can be seen in this figure that the efficiencies of foam cement source are fairly smaller at any corresponding density than those of the other sources. In the case of density 1 gcm^{-3} , the efficiency of the foam cement source is relatively 24 % smaller than that of the liquid source. This implies that if a sample which has a characteristic like the liquid source (for example, a sample of waste solution) is to be measured for its gamma-ray radioactivity, the application of foam cement as the calibration source is not recommended here.

The difference of efficiency between the foam cement and the other sources is due to the elements contained in their matrix materials. The interaction of low energy photons with materials is mainly determined by the photoelectric effect that strongly depends on the atomic number of element. Polystyrene foam and plastic beads are composed of such

elements as hydrogen, oxygen and carbon, and water is of the elements of hydrogen and oxygen. And also, the dominant element in glass beads is silica. On the other hand, the foam cement source contains large amount of calcium. The atomic number of calcium is higher than those of the elements contained in the sources of polystyrene foam bead, plastic bead, glass bead and liquid. Therefore, the decrease of the efficiency for the foam cement source is clearly observed in the low gamma-ray energy region corresponding to the bad correlation case.

The case of the poor correlation is shown in Fig.4.15. It can be seen that this poor case is much better than the bad case but is not so good case. Therefore, this gamma-ray energy region is considered to be a transitive region.

In the case of the good correlation (regardless of the types of the calibration source) as shown in Fig.4.16, this implies that efficiency dependence on the types (or composition) of the source is less significant for gamma-ray energy higher than 391.7 keV. In addition to this, the same figure also shows that when gamma-ray energy keeps increasing, the variation of efficiency-density curves becomes less significant (this fact will be discussed later in this section).

From Fig.4.14, although it is noticed that the efficiency-density correlation at this low energy is bad for all types of sources, it can be seen however (from the dotted line) that the correlation yielded by the foam cement source alone is good, and acceptable. For this reason, the efficiency-density correlation due to the foam cement source only is plotted for all the gamma-ray energy, as shown in Fig.4.17. It can be seen that good correlation is obtained, which implies the validity and suitability of this foam cement source to be used as a calibration source in the gamma-ray radioactivity measurements for limited compositions.

In Fig.4.16, if the efficiency variation at a particular gamma-ray energy is defined as the ratio of the efficiency value at density 0.21 gcm^{-3} (at certain energy) to the

efficiency value at 2.04 gcm^{-3} (at the same energy), and if this efficiency variation versus gamma-ray energy is plotted, the significance of this efficiency variation (in this density range) as a function of the gamma-ray energy may be seen. Notice that if the ratio equals to one, it means that there is no variation of the efficiency at the particular energy. This ratio is also obtained for Fig.4.17. As it can be seen from Fig.4.18, it implies that efficiency variation is very significant for low energy gamma-ray, and becomes less significant when the energy increases.

It is also worth mentioning here about the usefulness of the liquid and the bead sources. Although the foam cement calibration source alone is very useful for the purpose of getting a good correlation between the efficiency and density for all the gamma-ray energy (Fig.4.17), it is also thought that for the same purpose, the liquid and the bead sources are also useful, but only at the gamma-ray energy above 100 keV (Figs 4.15 and 4.16).

Notice that accuracy of all the results (discussed in this section) have been checked by the use of the alternative detection system, i.e. the automatic sample changer system for routine radioactivity measurement of samples.

5. CONCLUSION

Based on the results obtained, it may be concluded from this work that:

- (1) the common expectations of this work have been fulfilled,
- (2) the reproducibility of the source preparation method is good,
- (3) the accuracy of the results obtained with the present system is good,
- (4) the foam cement source is the most suitable calibration source for gamma-ray radioactivity measurements for limited compositions,
- (5) the liquid and the four types of bead source are not very

efficiency value at 2.04 gcm^{-3} (at the same energy), and if this efficiency variation versus gamma-ray energy is plotted, the significance of this efficiency variation (in this density range) as a function of the gamma-ray energy may be seen. Notice that if the ratio equals to one, it means that there is no variation of the efficiency at the particular energy. This ratio is also obtained for Fig.4.17. As it can be seen from Fig.4.18, it implies that efficiency variation is very significant for low energy gamma-ray, and becomes less significant when the energy increases.

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- (5) the liquid and the four types of bead source are not very

suitable or useful for use as the calibration sources for gamma-ray radioactivity measurement over all the energy and density ranges of interest,

(6) however, for gamma-ray energy above 100 keV, the liquid and the four other types of the calibration sources are also useful for the calibration work,

(7) for gamma-ray energy lower than 100 keV, in the case of equal shape, the significance of types (or compositions) and density of the calibration sources is prominent, and

(8) the use of the foam cement source as a calibration source for the purpose of radioactivity measurement of low gamma-ray energy (59.54 keV) of samples like water (density value about 1 gcm^{-3} , for example, the waste solution) is not recommended.

ACKNOWLEDGEMENTS

The authors are extremely grateful to the collaboration given by the staff of Radiation Counting Section of Radiation Dosimetry Division Dep. of Health Physics JAERI, namely Mr. Koji Takahashi and Mr. Masato Ishizawa, that greatly contributed to the success of this work. One of us (SBS) is grateful to The Science and Technology Agency (STA), Government of Japan for the award of STA Scientist Exchange Programme Fellowship, which has made leave from Universiti Kebangsaan Malaysia at Tokai-mura for three months 29.3.1994-25.6.1994 possible.

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Table2.1 Calibration sources : type, composition and identification

Type	Composition			Identification
	Matrix material	Carrier	Diluting solution	
Foam cement	cement + water + foam	used*	distilled water (with 0.5mol/l HCl)	T 1
Liquid	none	none	same as above	T 2
Beads	glass, of 6mm in diameter	none	same as above	T 3 A
	glass, of 2mm in diameter			T 3 B
	foamed polystyrene			T 4
	hard plastic			T 5

* : Cs, Sr, Y, Ce, Co, each 10mg/ml in 4N HCl

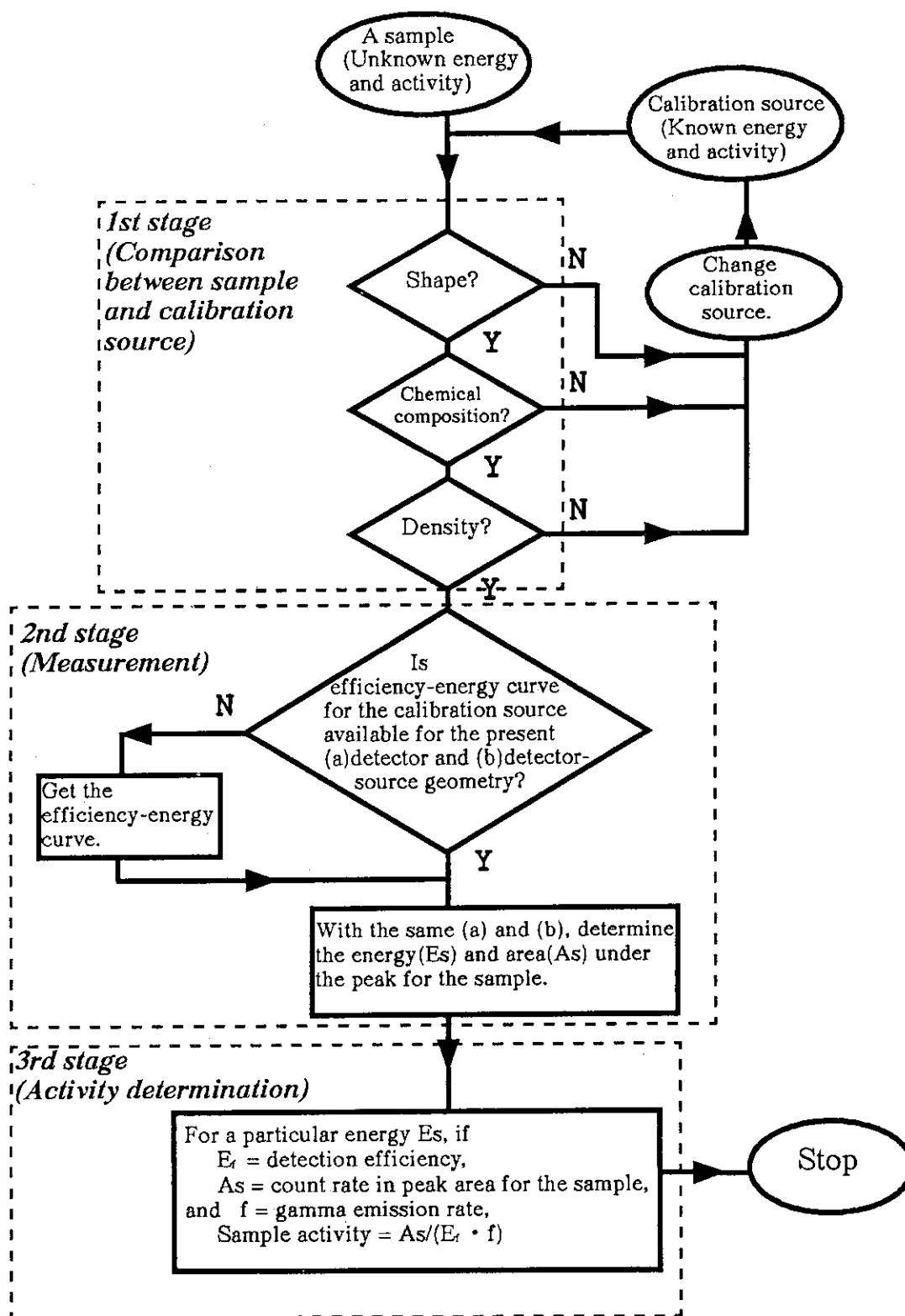


Fig1.1 Standard procedures for qualitative and quantitative determination of a sample, with emphasis given on the shape, chemical composition and density.

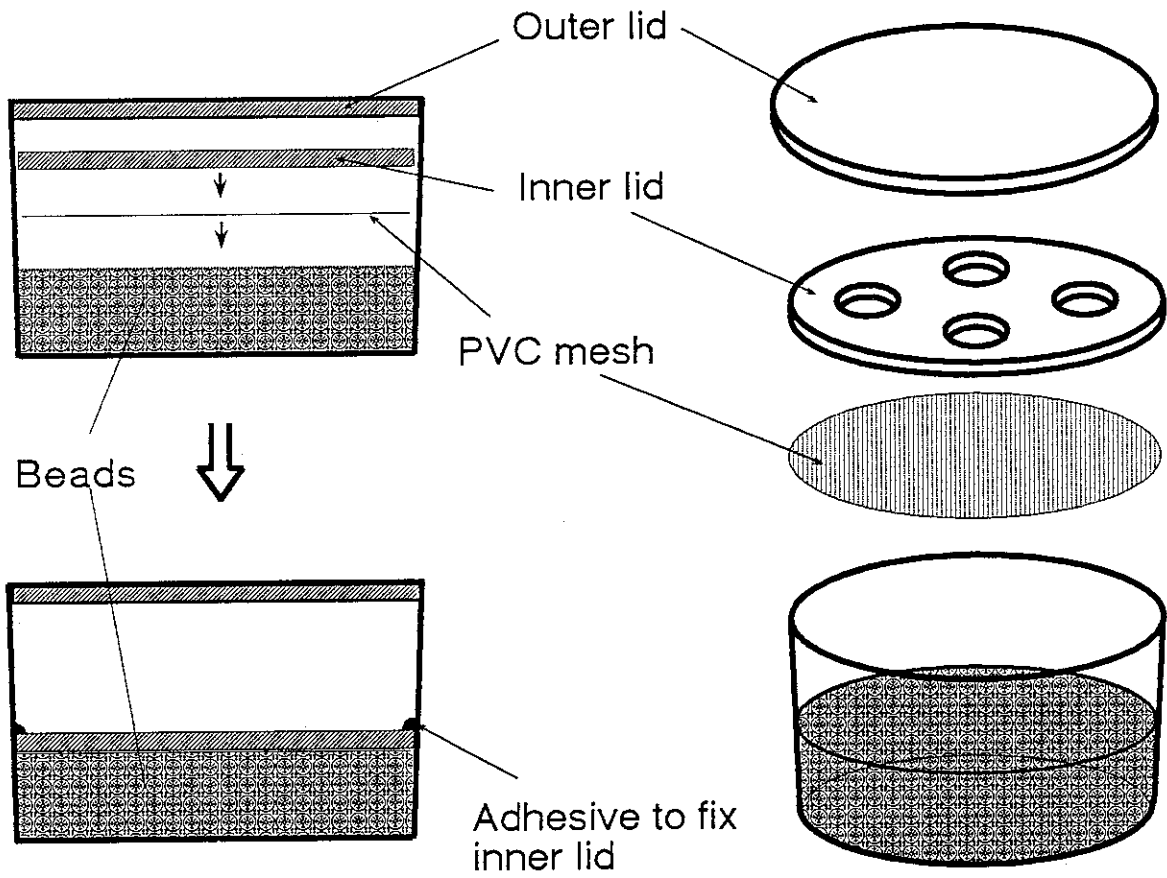


Fig2.1 Containment for matrix materials of bead calibration source.

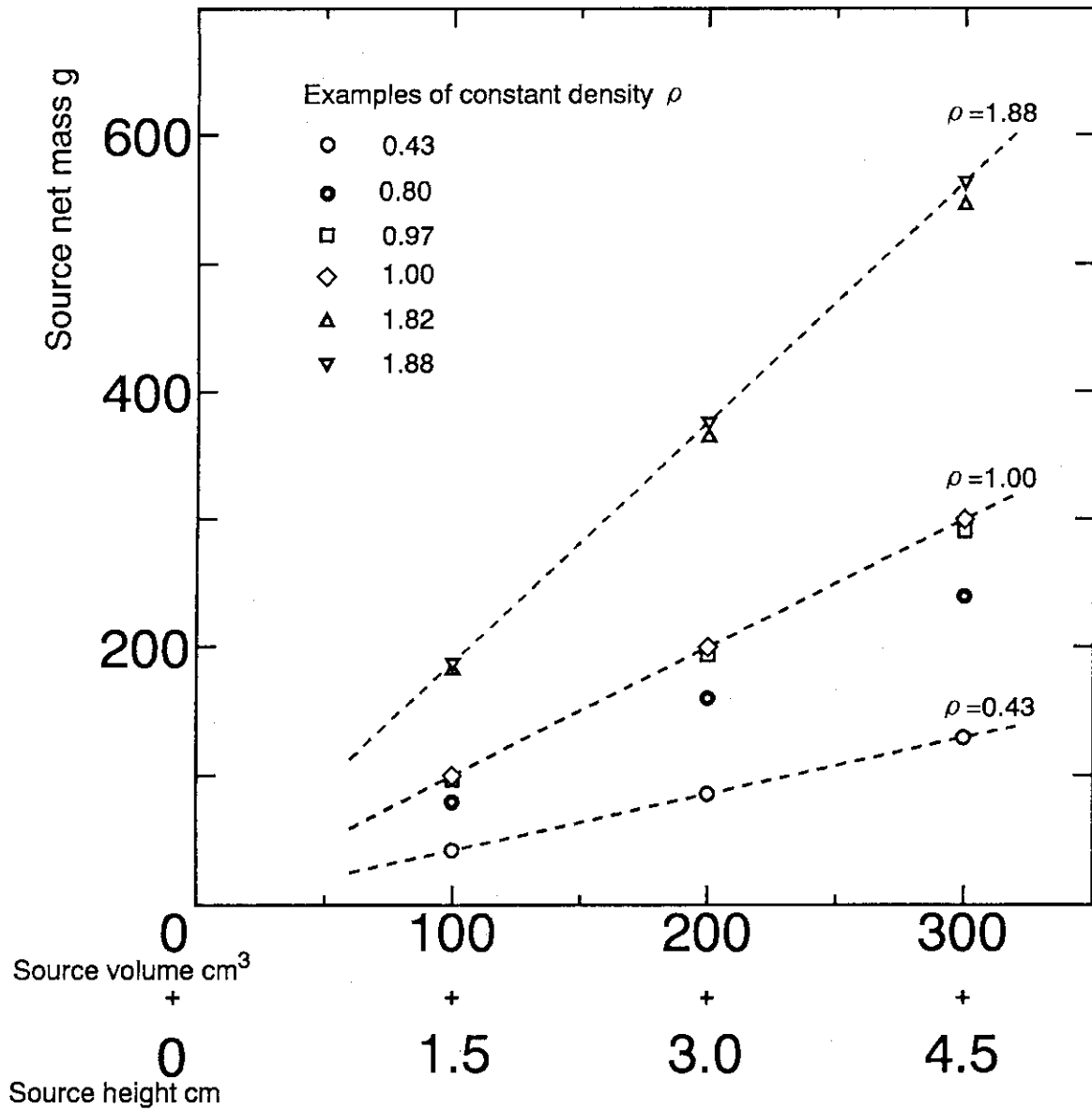
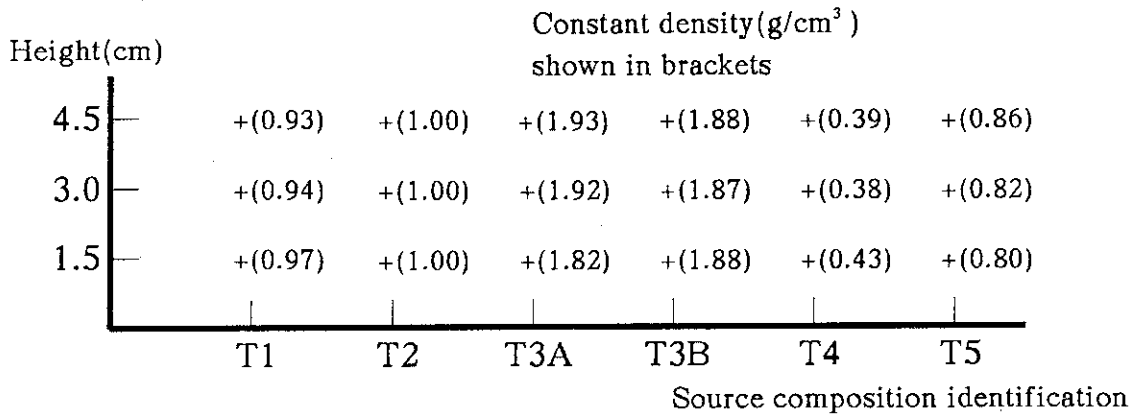
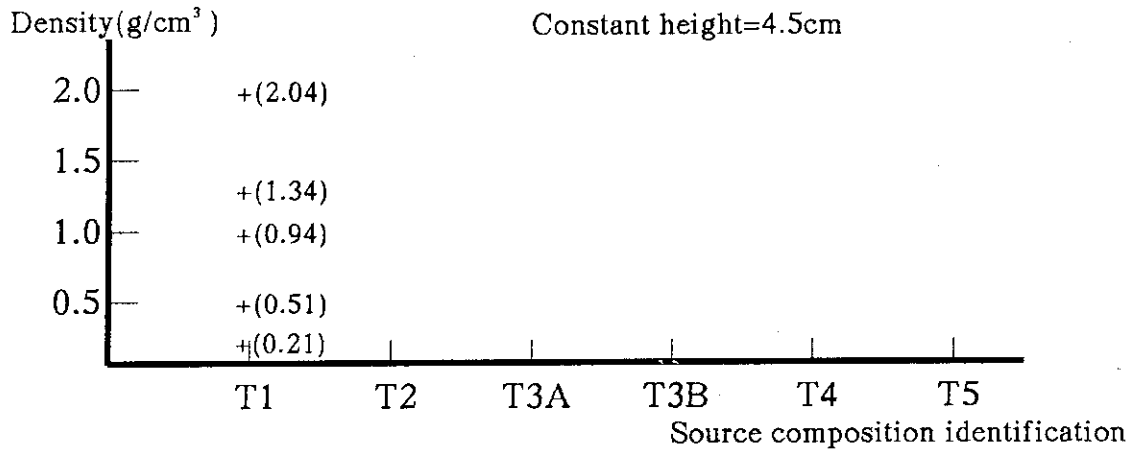


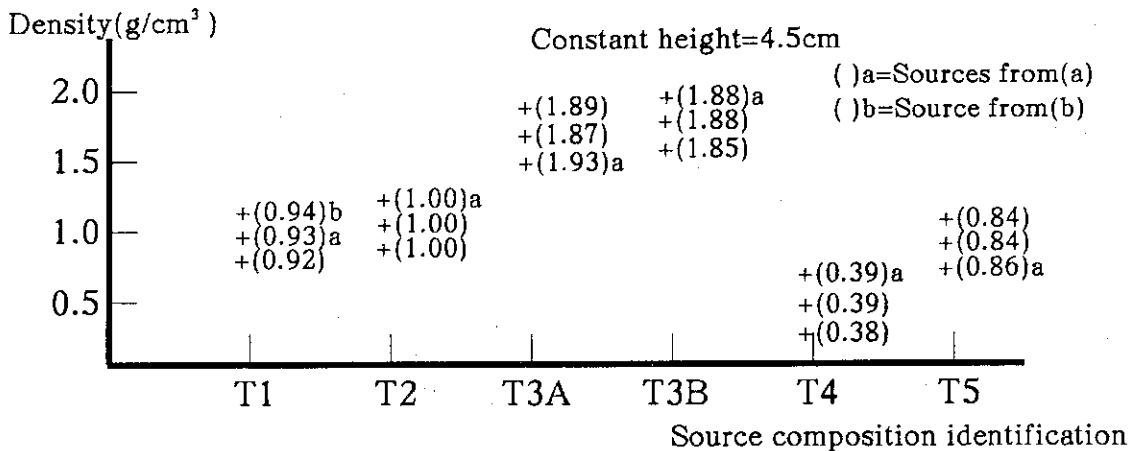
Fig2.2 Guide for the preparation of calibration sources with constant density.



(a) To study the variation of efficiency curve with composition and height of calibration sources.



(b) To study the variation of efficiency curve with density of foam cement sources.



(c) To study the reproducibility of source preparation method.

Fig2.3 Objectives of this work. All the sources in (a), (b) and (c) use the same plastic container of 9.5cm in diameter. Details of the composition identification is given in Table2.1

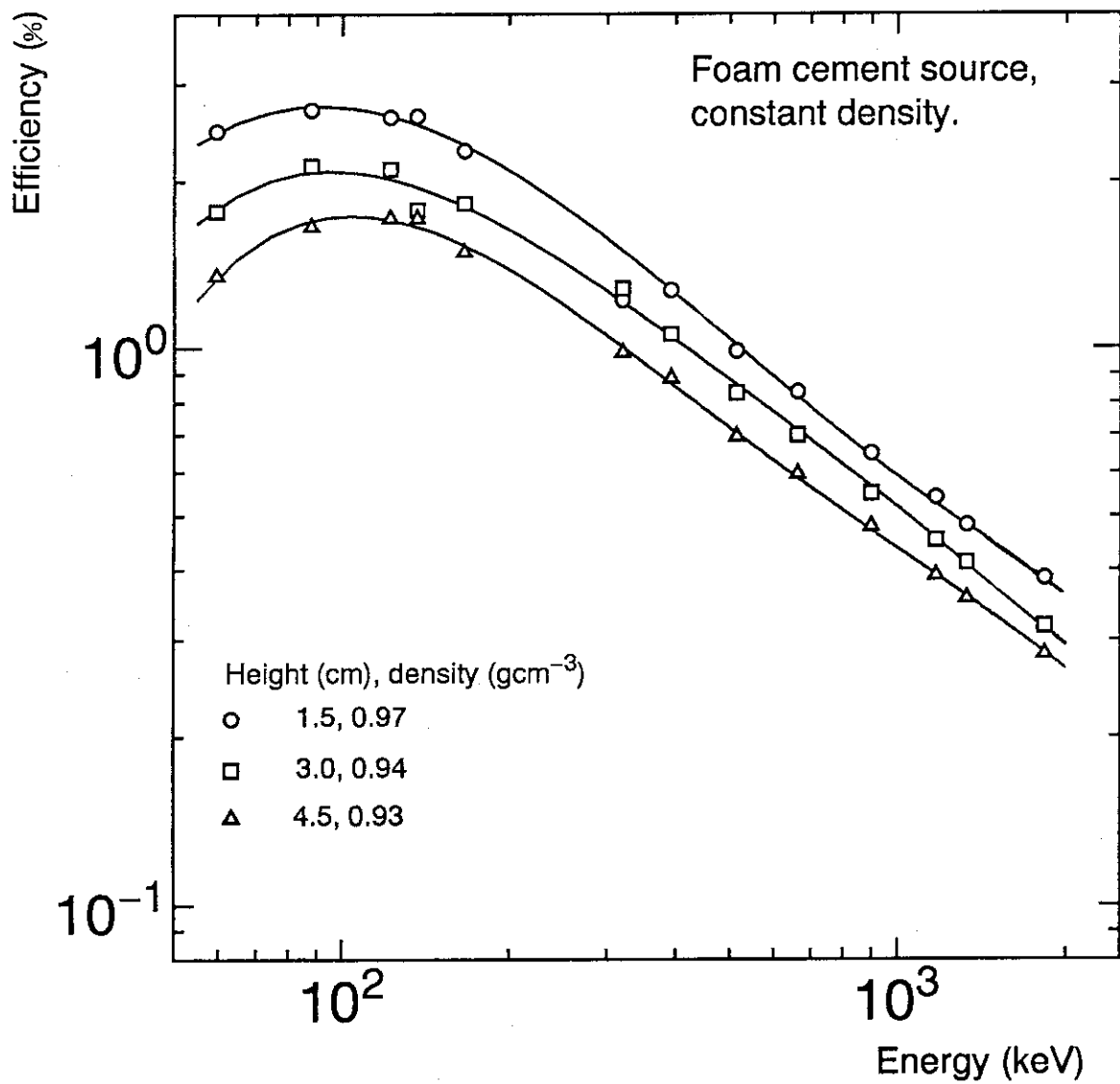


Fig4.1 Variation of efficiency-energy curves with heights of foam cement source for a constant density.

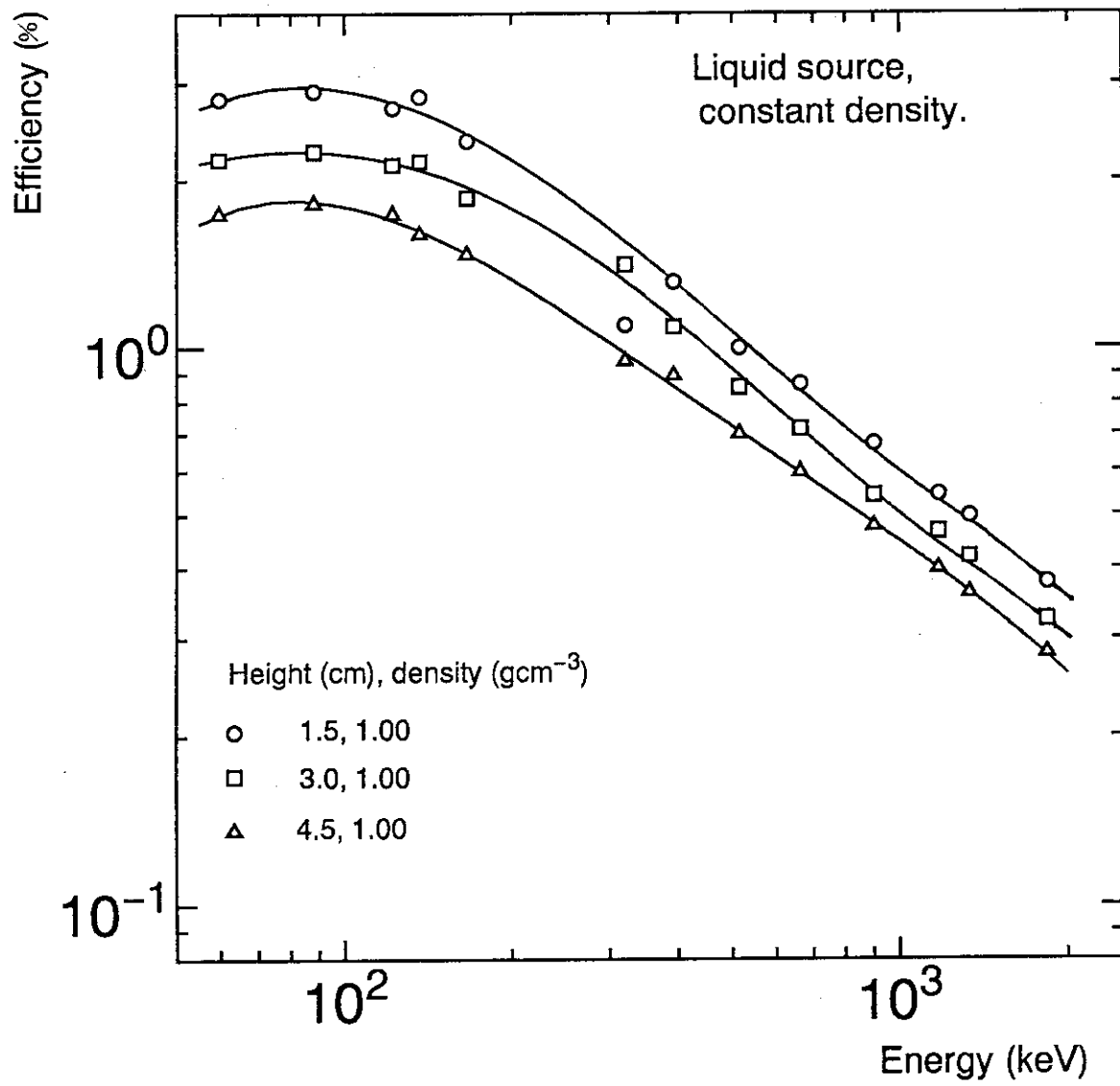


Fig4.2 Variation of efficiency-energy curves with heights of liquid source for a constant density.

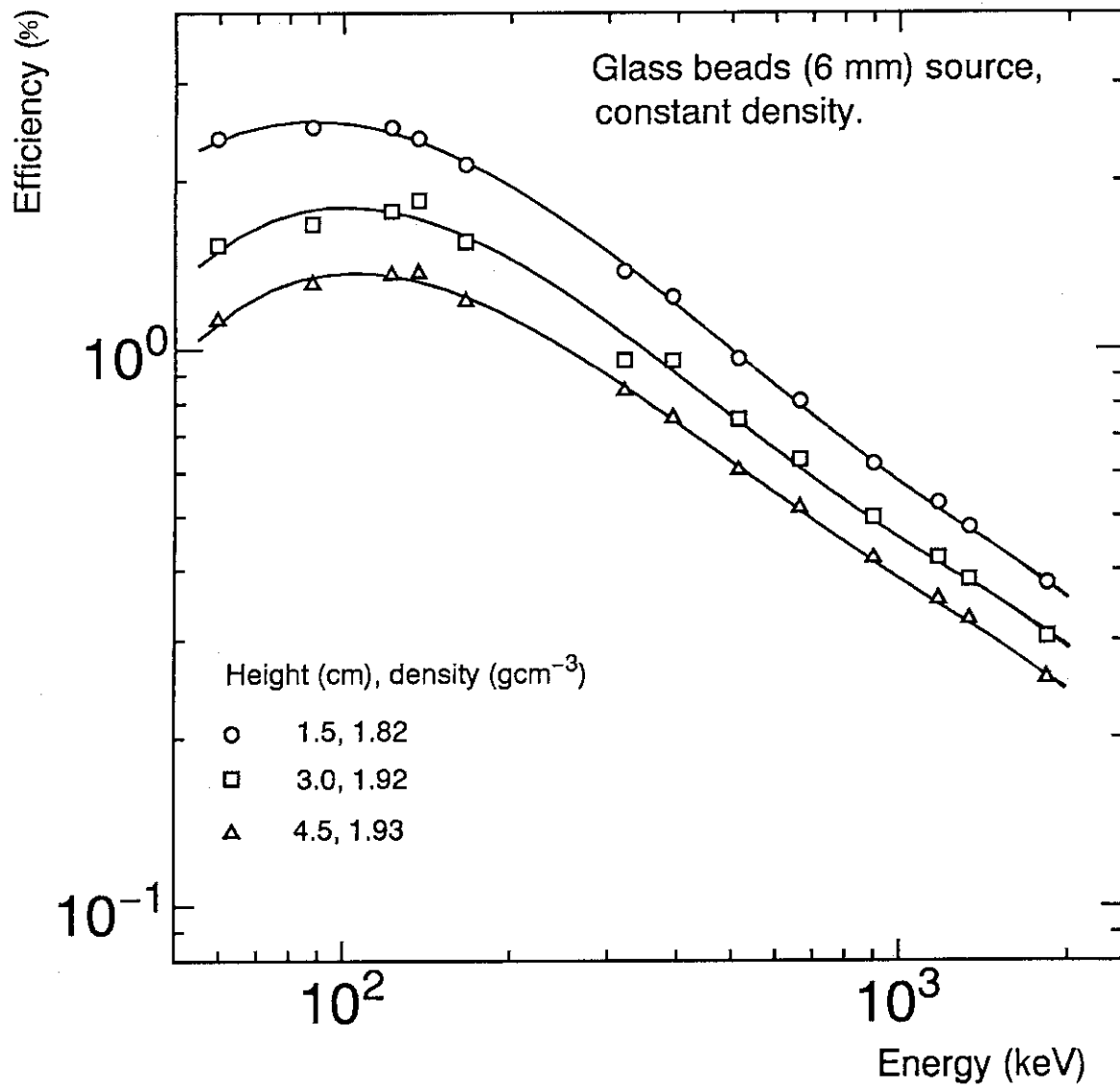


Fig4.3 Variation of efficiency-energy curves with heights of glass bead (6mm in diameter) source for a constant density.

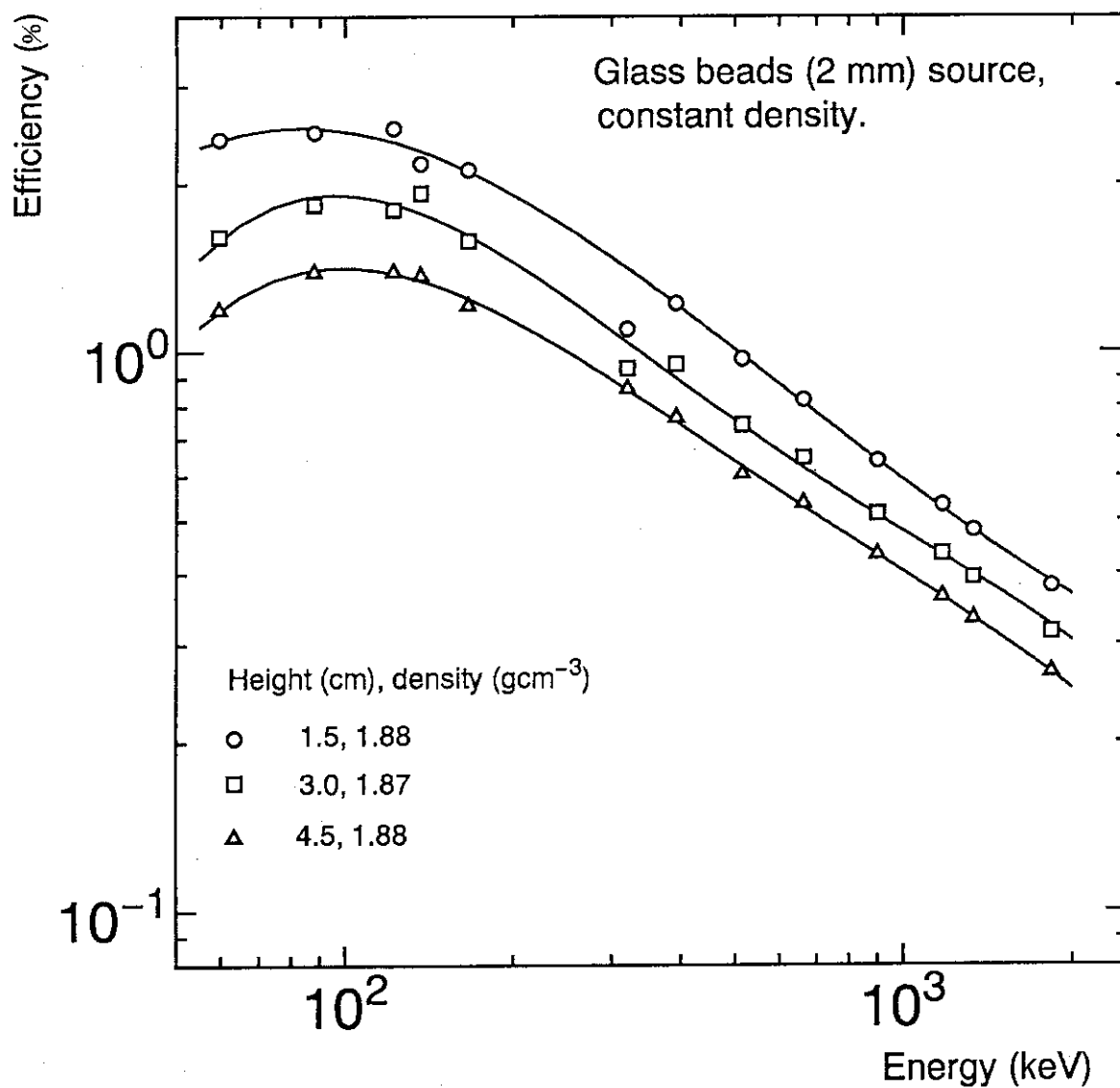


Fig4.4 Variation of efficiency-energy curves with heights of glass bead (2mm in diameter) source for a constant density.

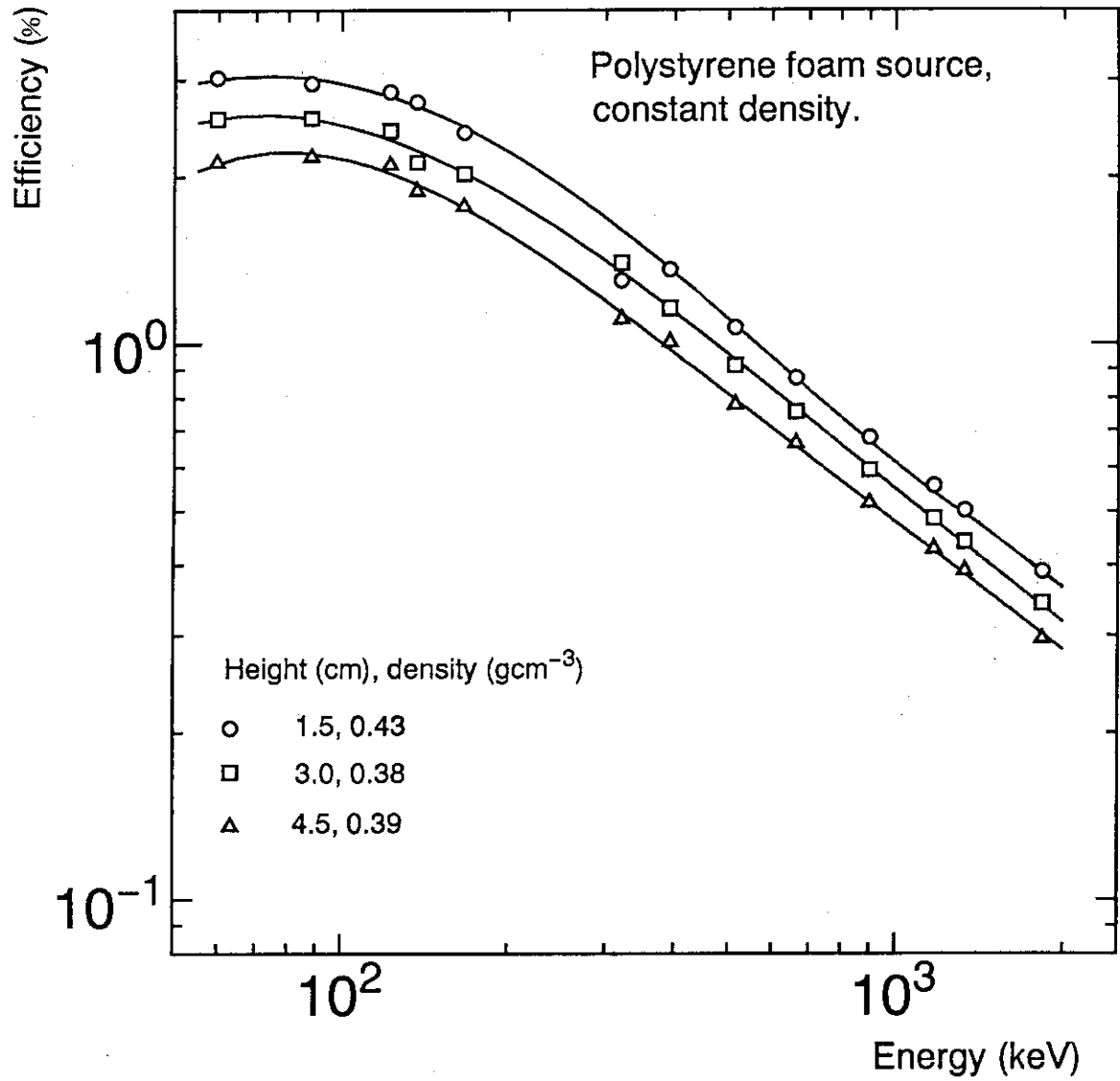


Fig4.5 Variation of efficiency-energy curves with heights of polystyrene foam source for a constant density.

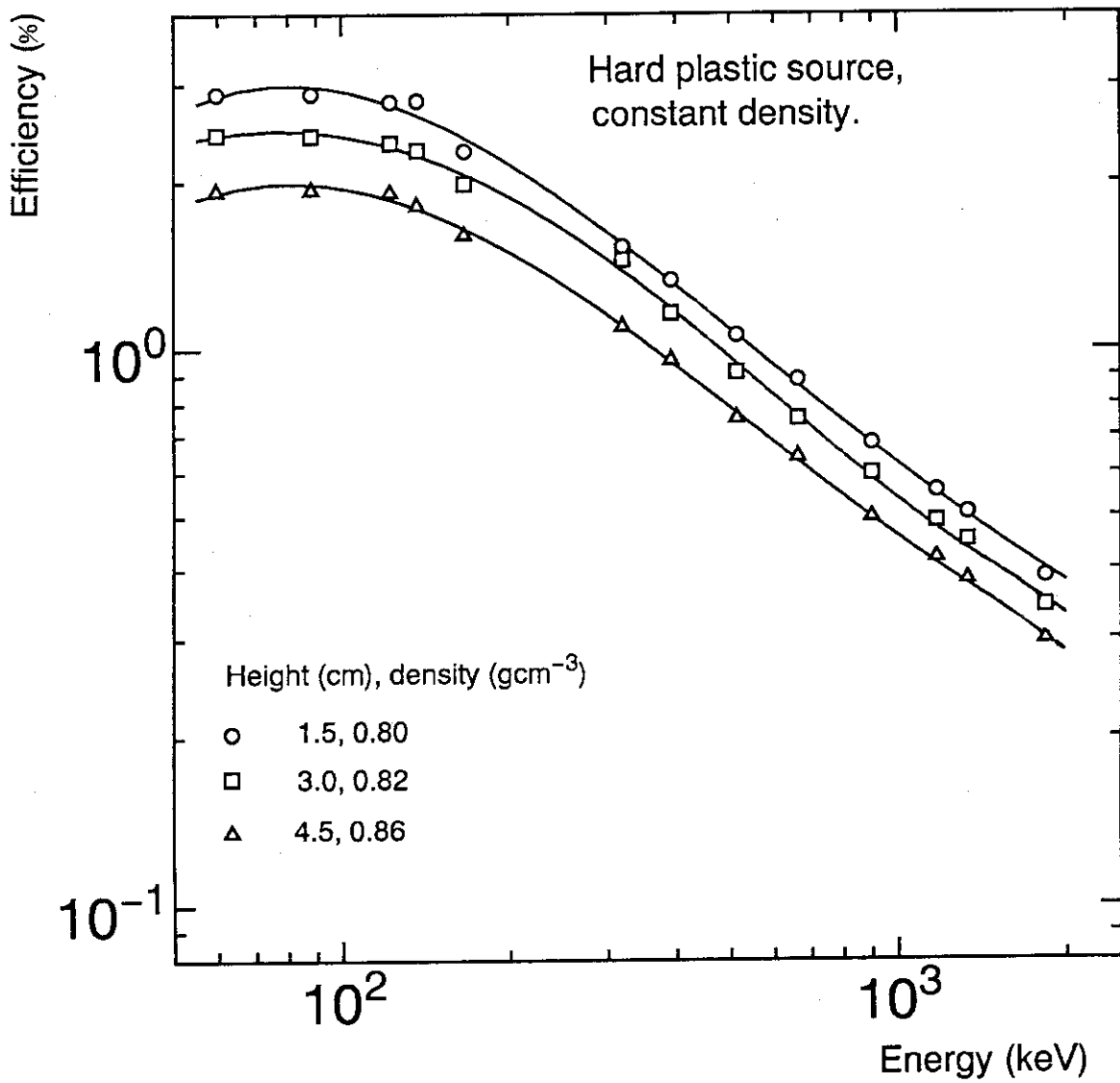


Fig4.6 Variation of efficiency-energy curves with heights of hard plastic source for a constant density.

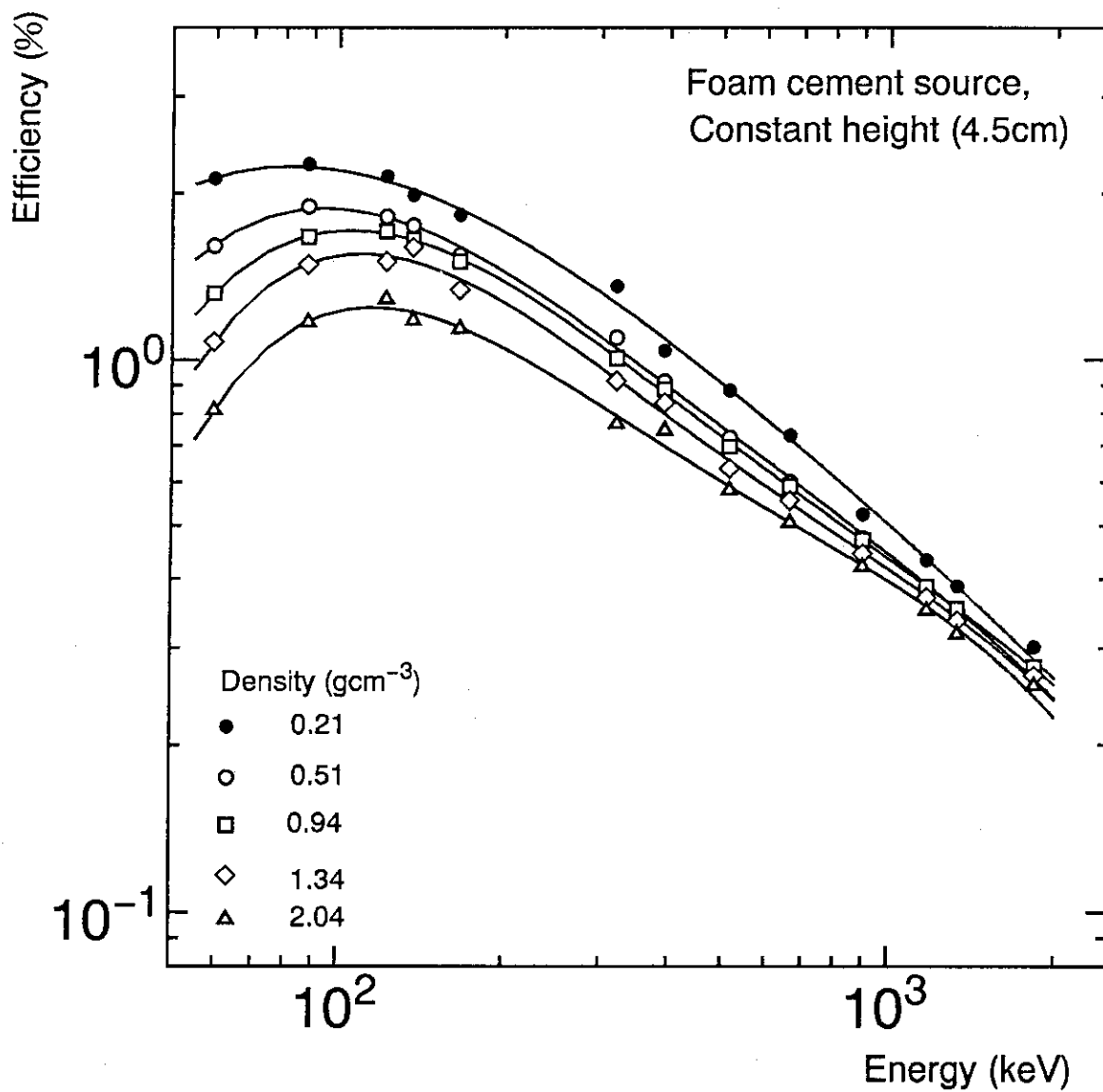


Fig4.7 Variation of efficiency-energy curves with densities of foam cement source for a constant height.

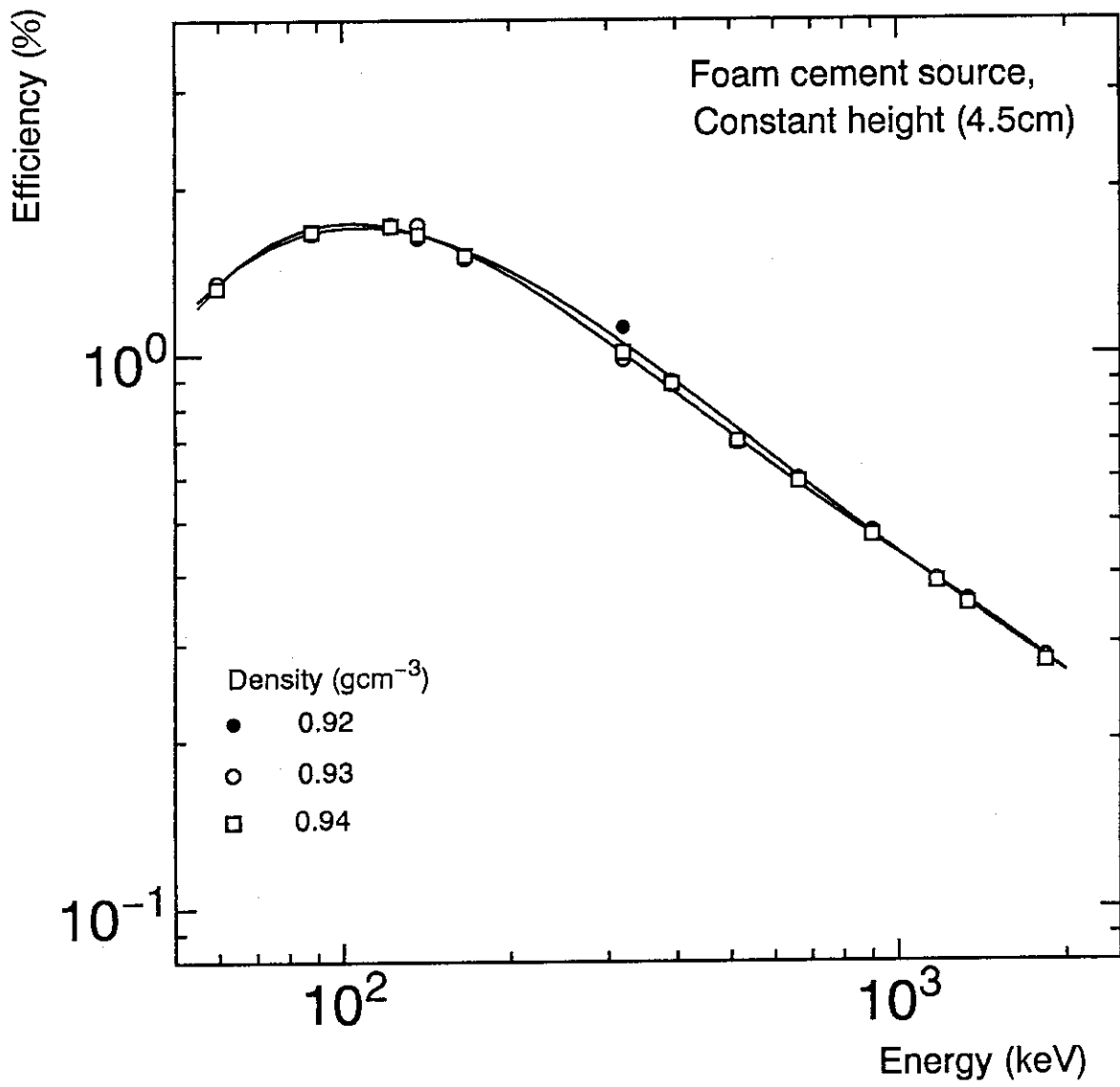


Fig4.8 Reproducibility of foam cement source preparation
(with constant height and density).

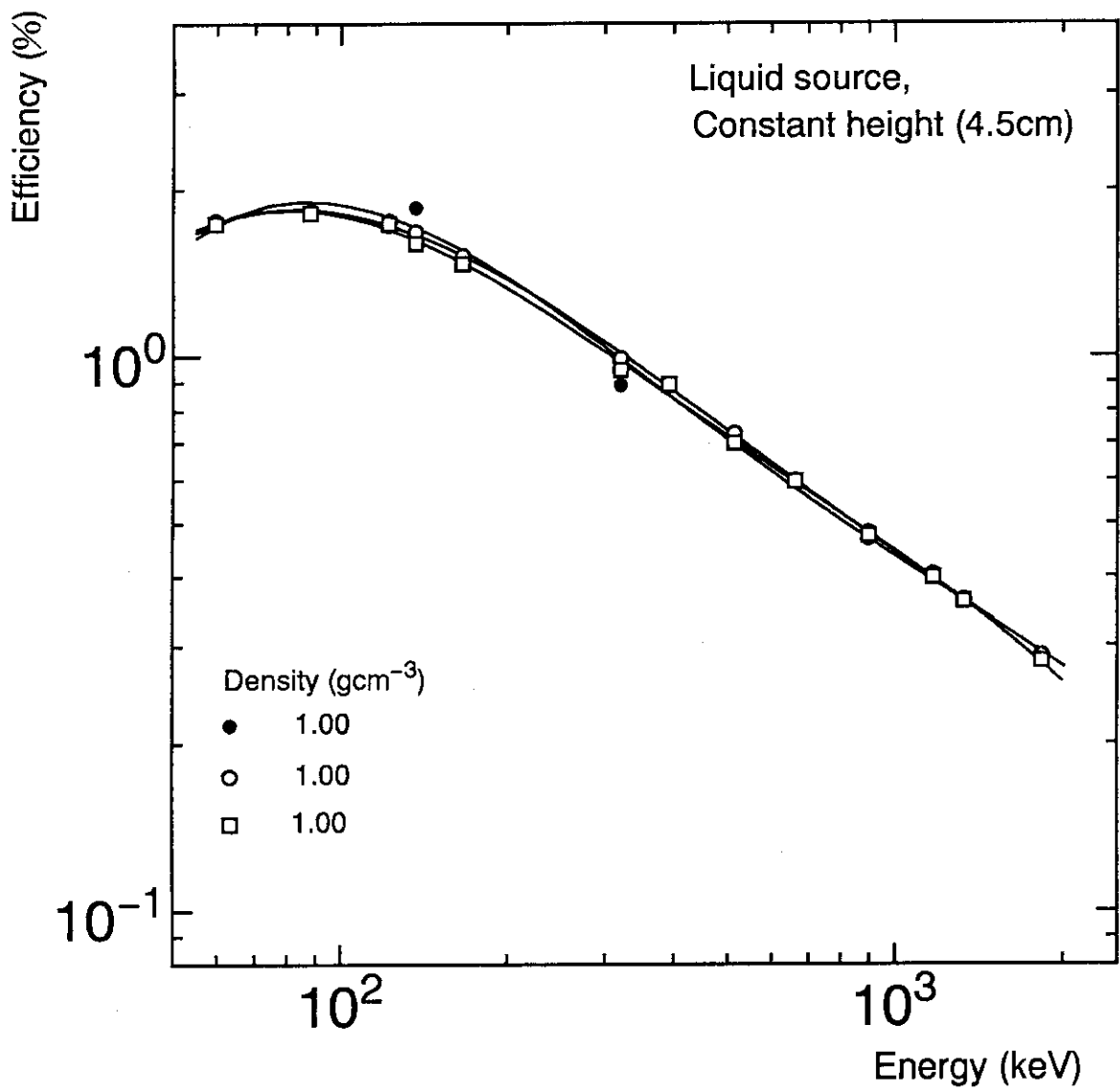


Fig4.9 Reproducibility of the liquid source preparation (with constant height and density).

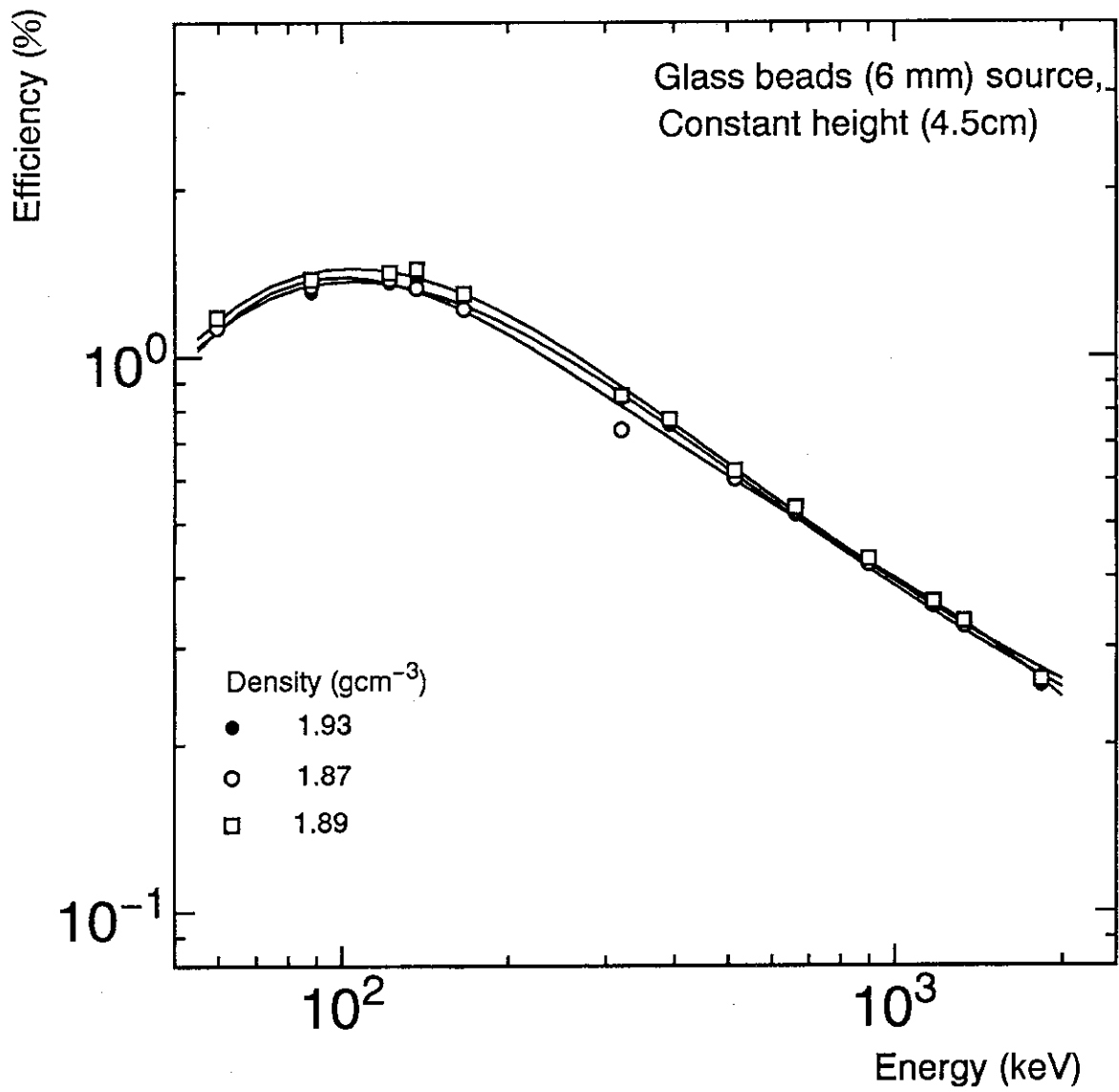


Fig4.10 Reproducibility of the glass bead (6mm in diameter) source preparation (with constant height and density).

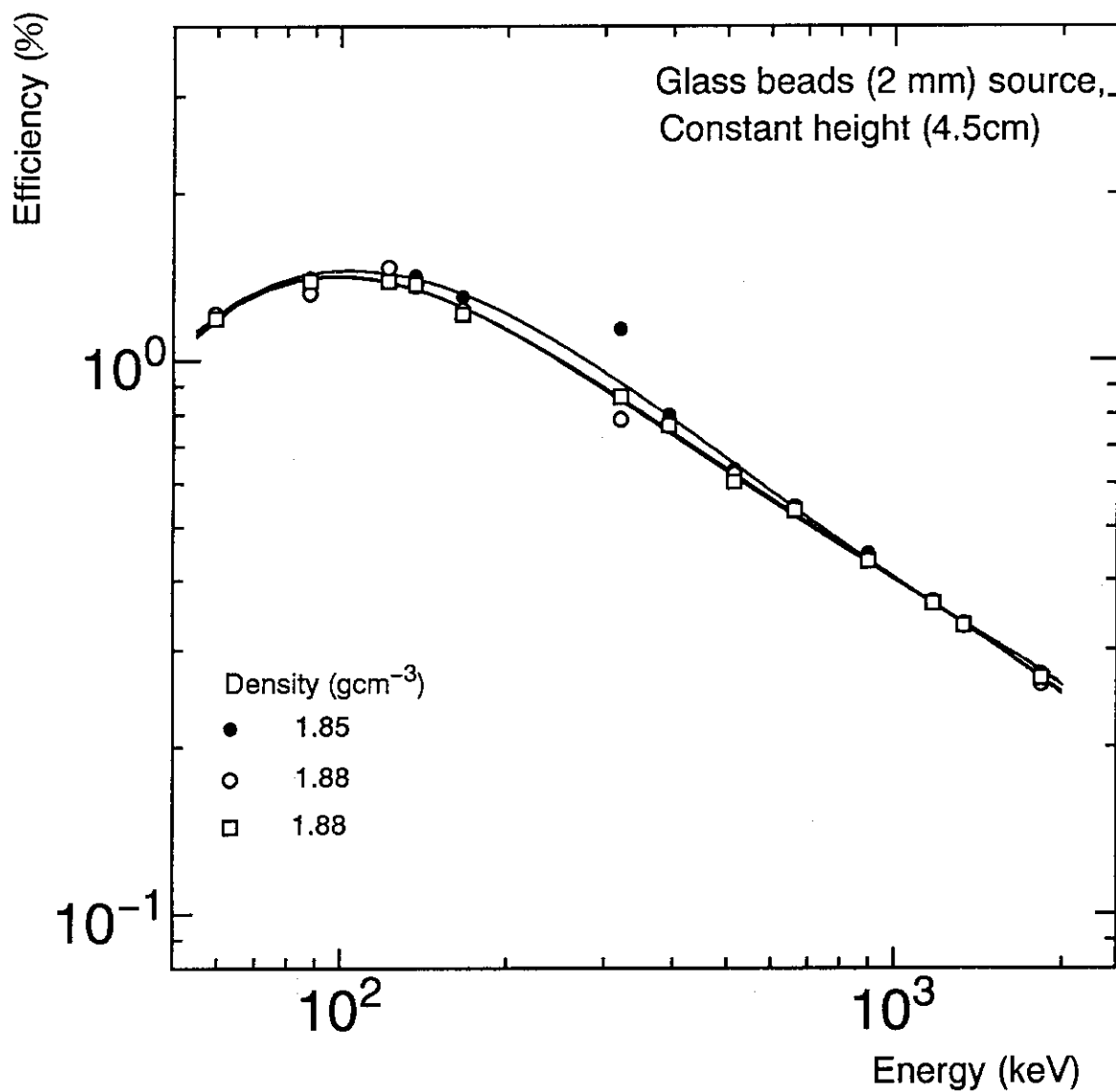


Fig4.11 Reproducibility of the glass bead (2mm in diameter) source preparation (with constant height and density).

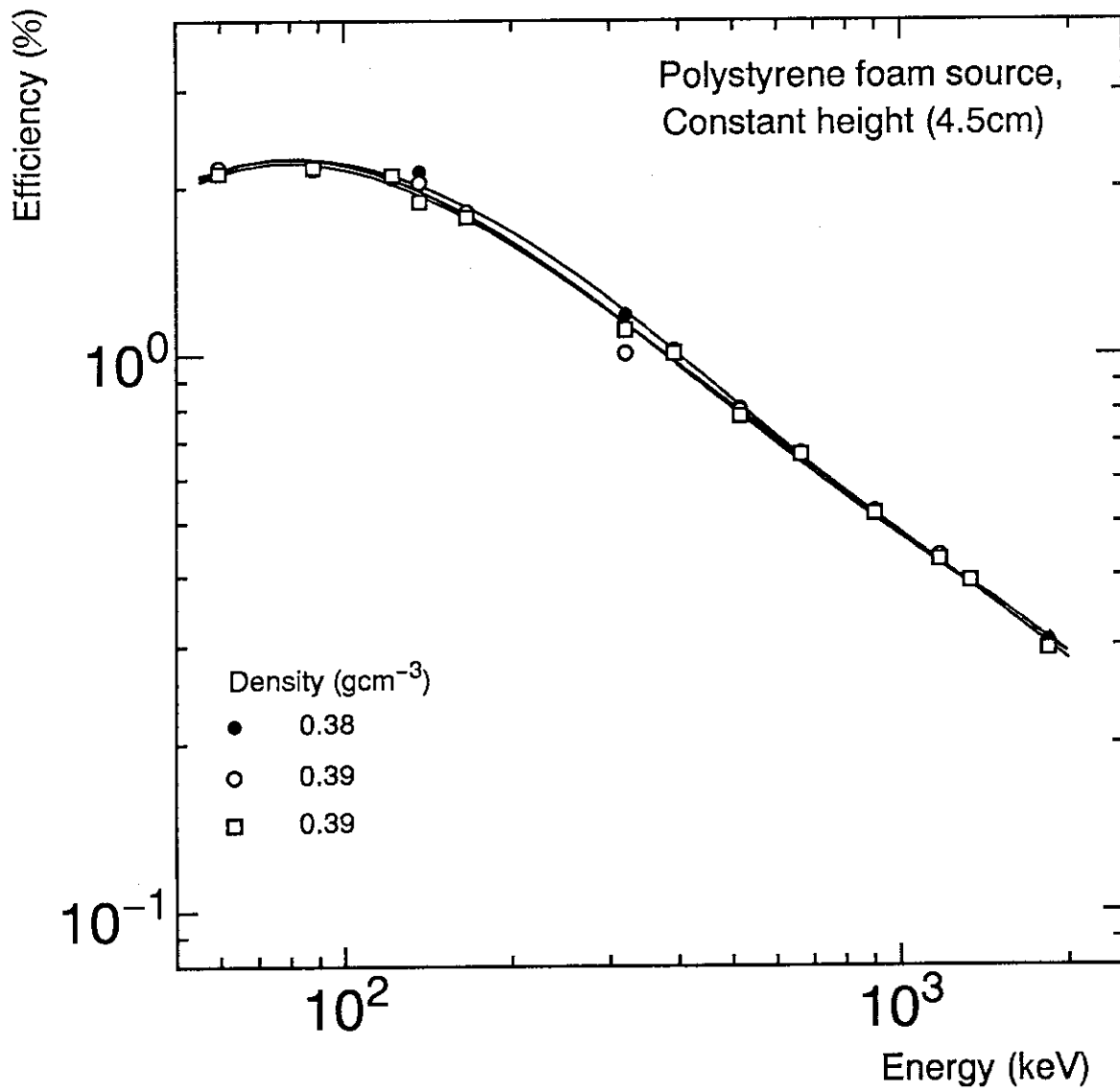


Fig4.12 Reproducibility of the Polystyrene foam source preparation (with constant height and density).

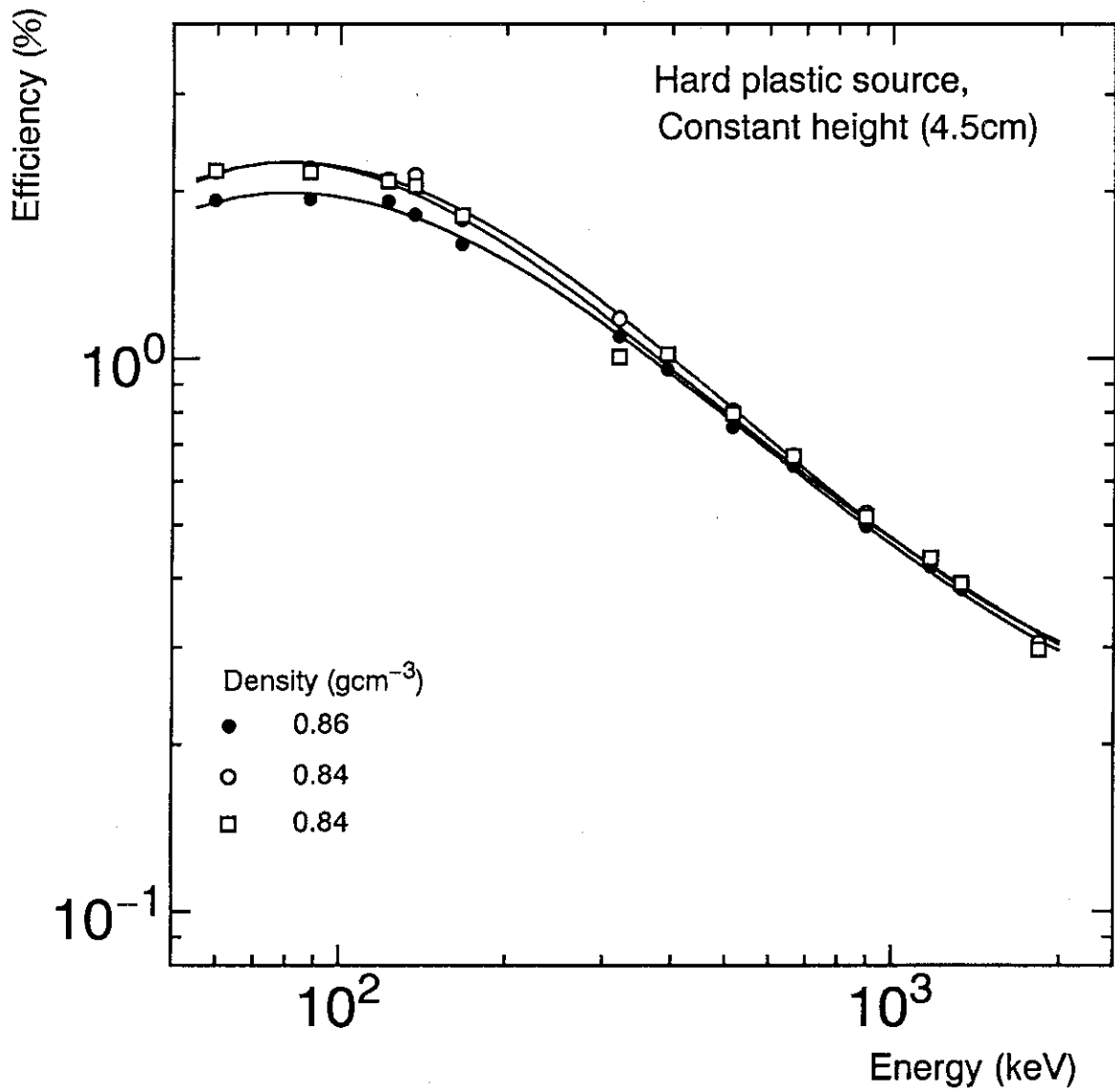


Fig4.13 Reproducibility of the hard plastic source preparation (with constant height and density).

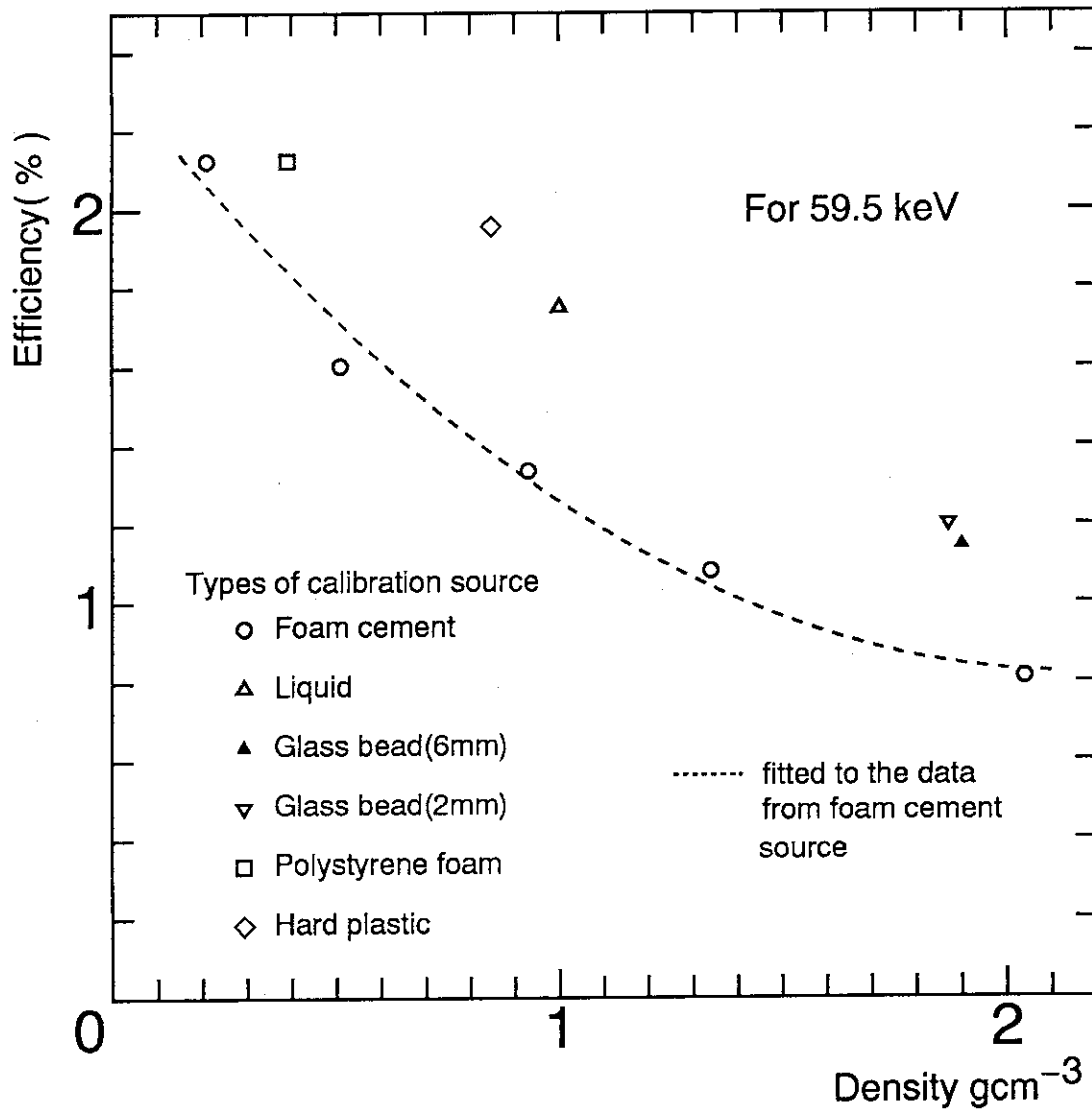


Fig4.14 Variation of efficiency-density curve with gamma-ray energy of 59.54keV. The types of the calibration source are indicated here. The broken line fits to the data from the foam cement source.

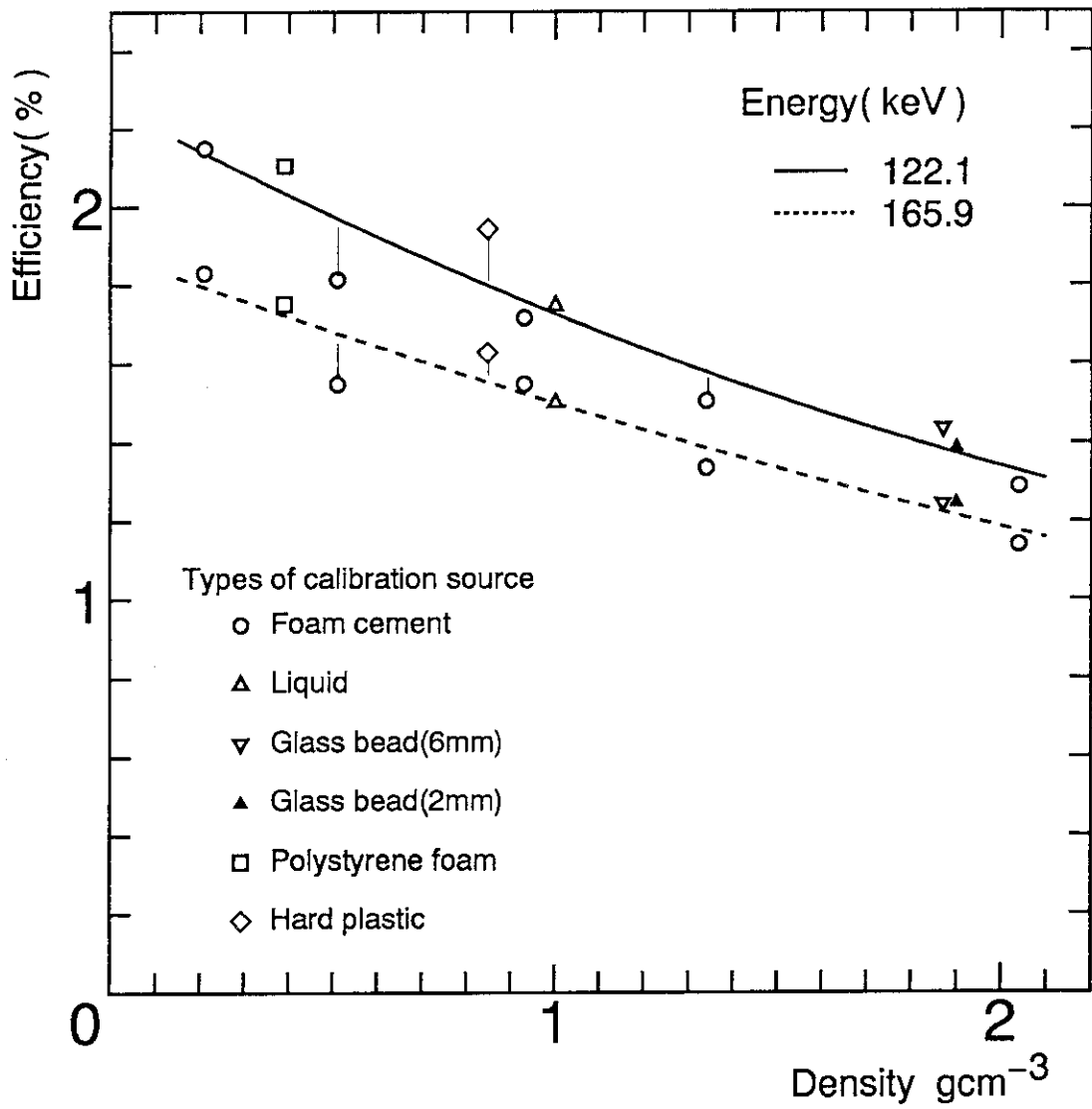


Fig4.15 Variation of efficiency-density curves with gamma-ray energies of 122.1 and 165.9keV. The types of the calibration source are indicated here.

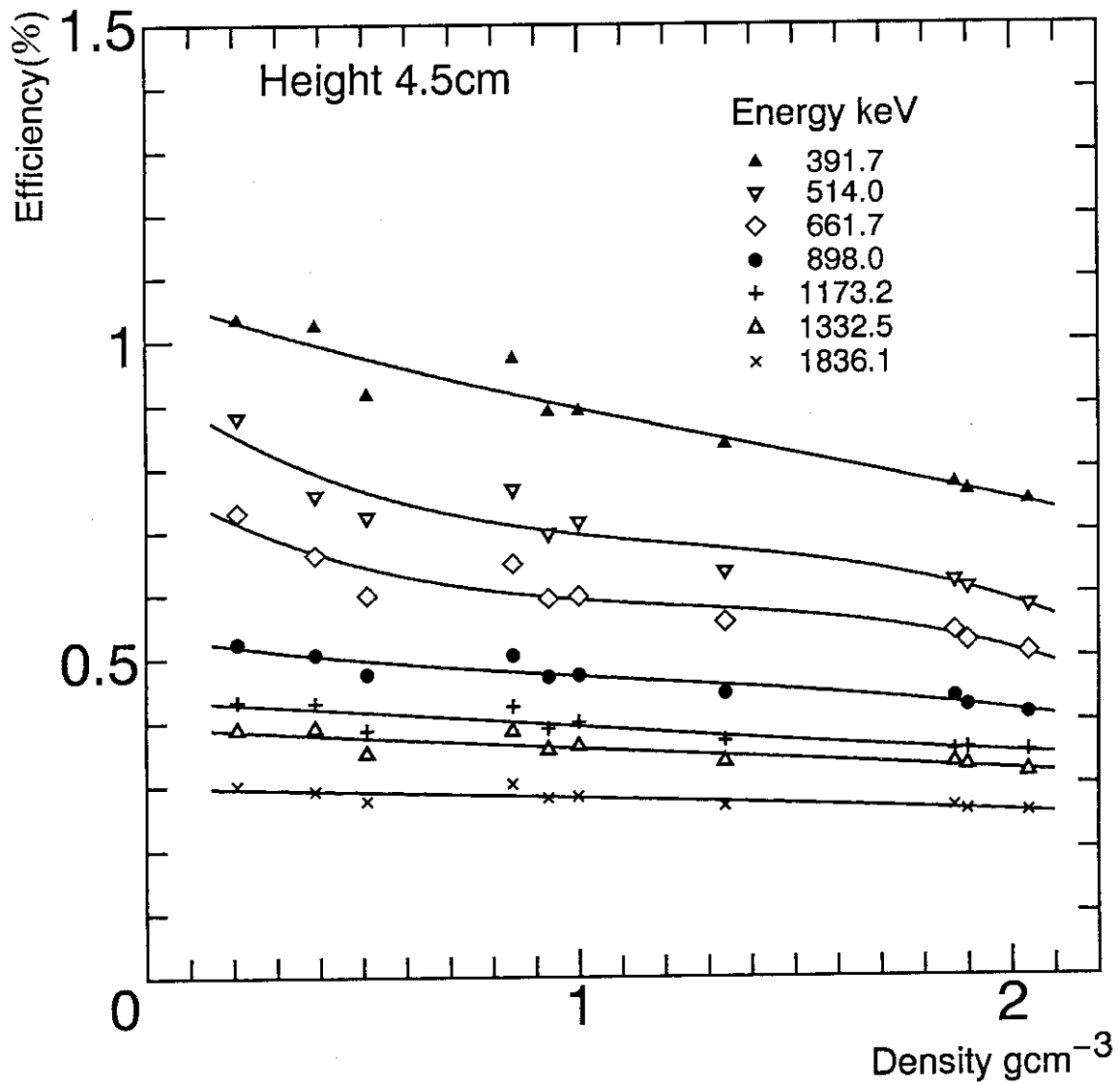


Fig4.16 Variation of efficiency-density curves with seven gamma-ray energies, for all the calibration source.

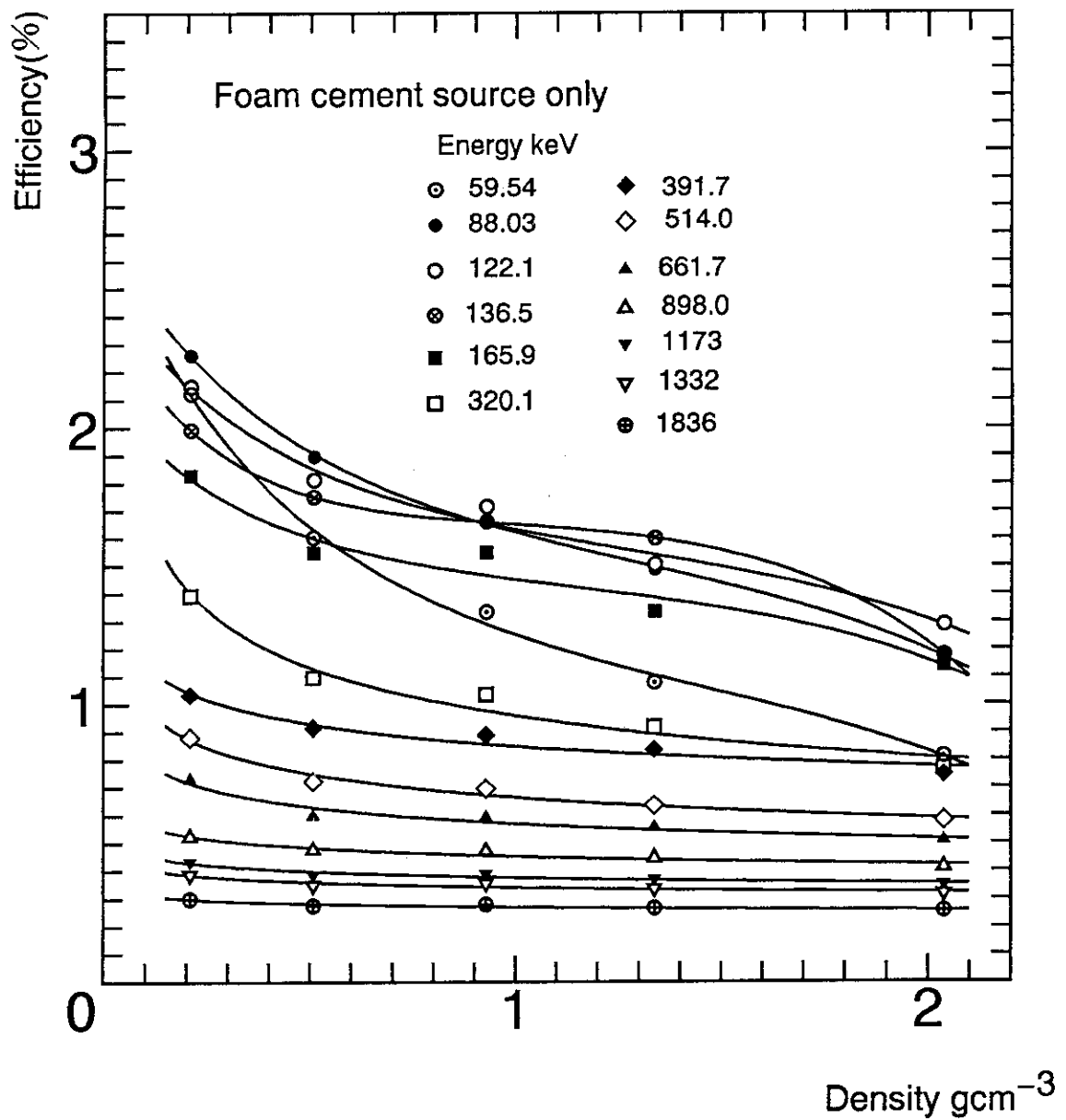


Fig4.17 Variation of efficiency-density curves with all the thirteen gamma-ray energies, for the calibration source of foam cement only.

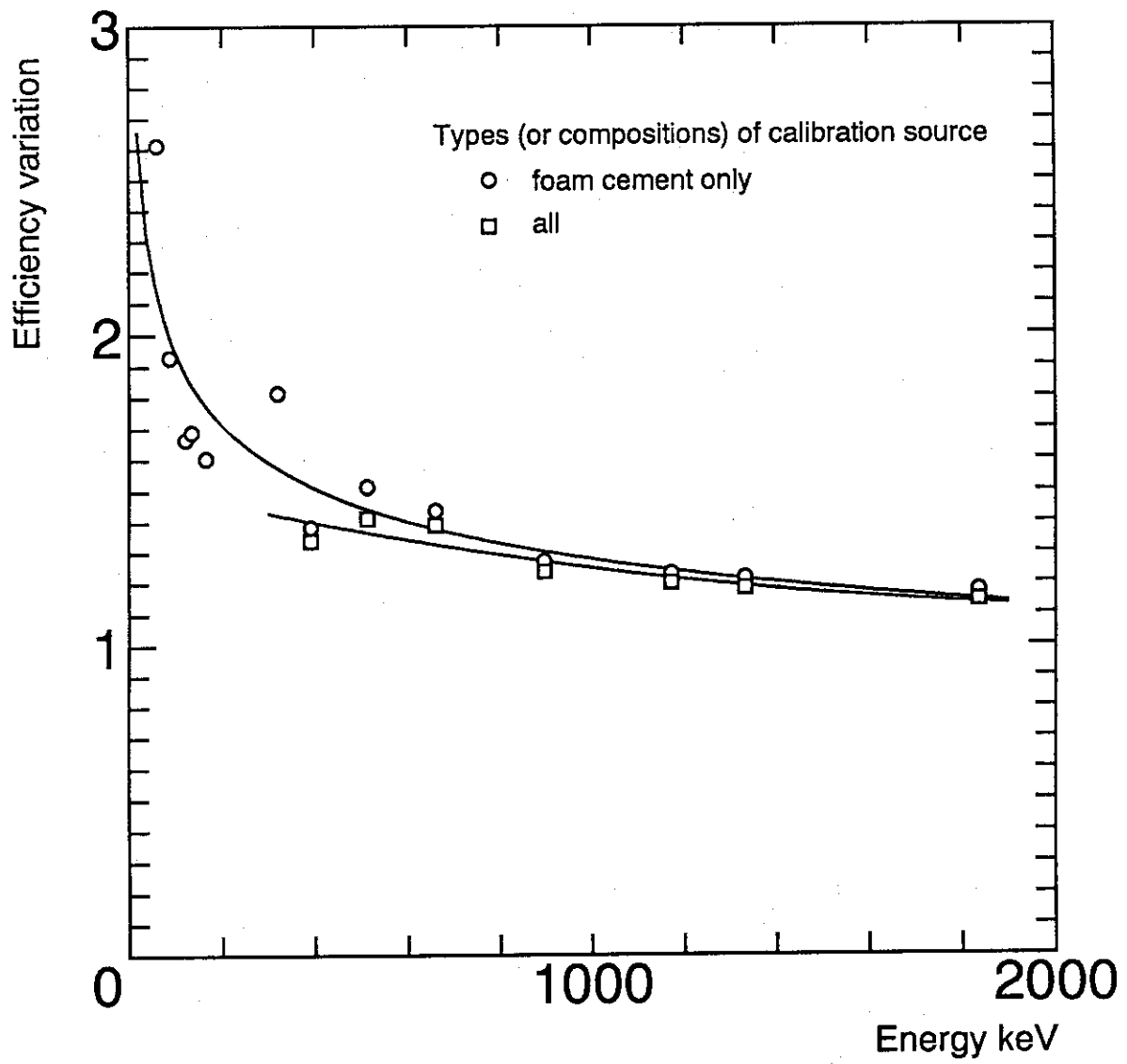


Fig4.18 Variation of efficiency with gamma-ray energy.

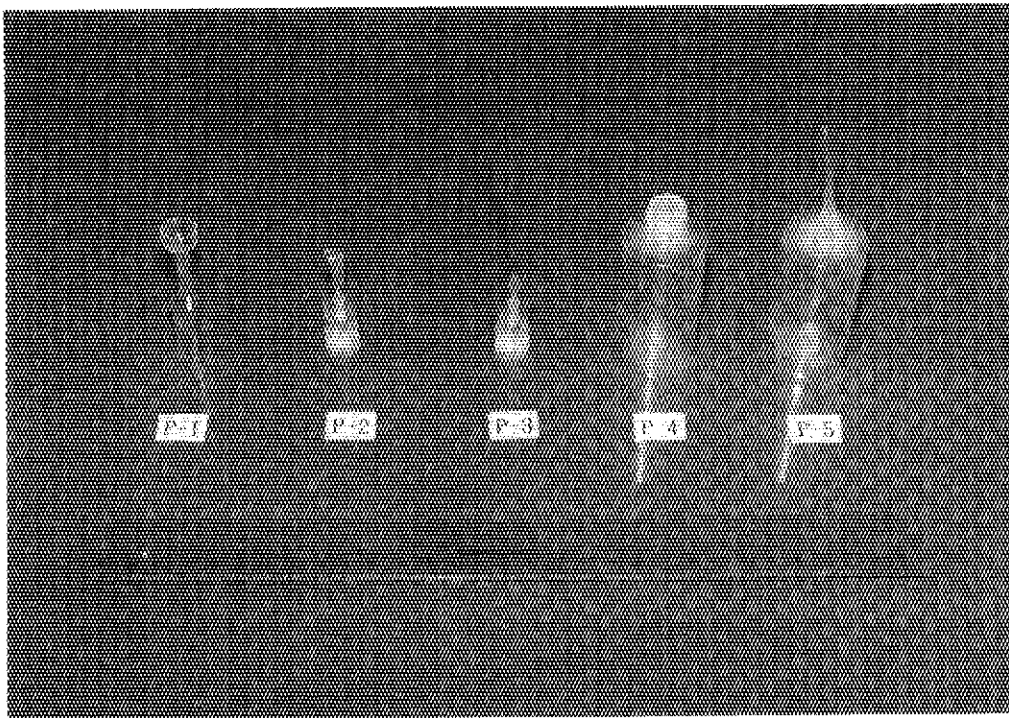


Photo1 Types of polystyrene ampoules (pycnometers) used in this work.

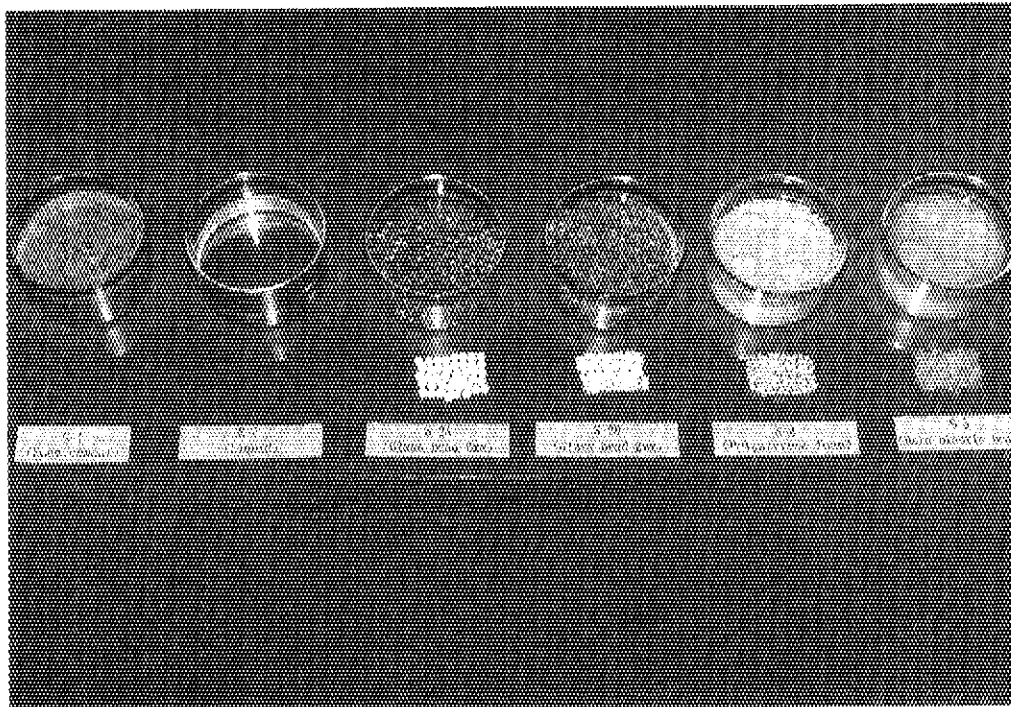


Photo2 Types of calibration sources prepared for this work.

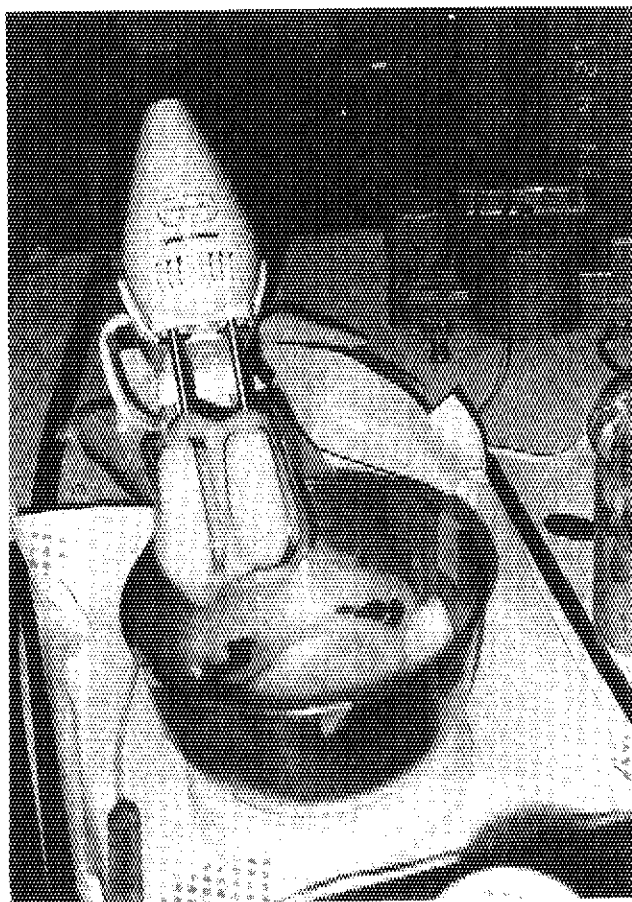


Photo3 Preparation of foam.



Photo4 Mixing water and cement.

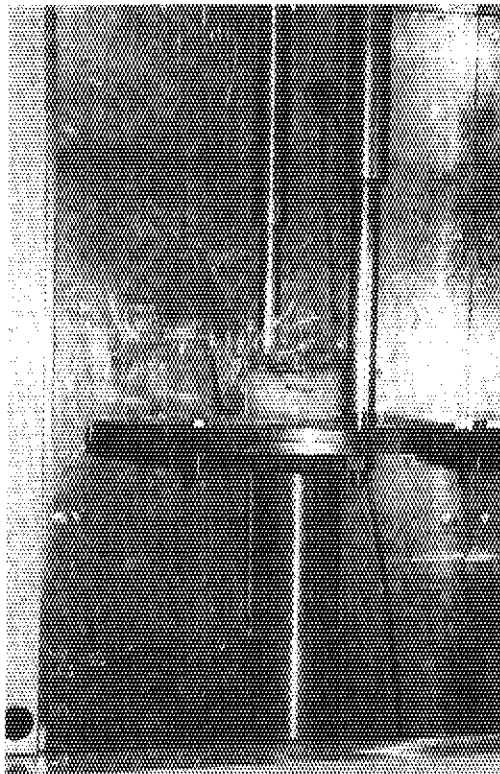


Photo5 Calibration source to the HPGe detector geometry.

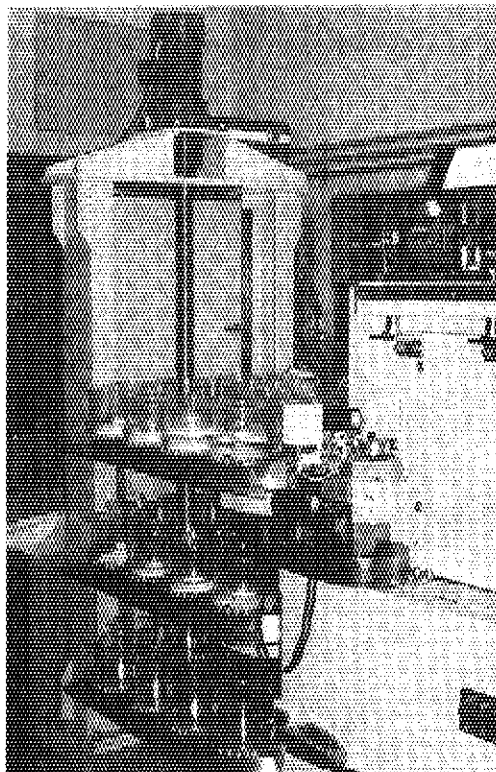


Photo6 Automatic sample changer system. This system is also utilised in this work for the purpose of checking the accuracy of results obtained from the manual system.

Appendix A

The radioactivity standard solution supplied by DAMRI/LMRI at reference date 6 Dec 1993, at 12:00

Radionuclide	Half-life	Energy (keV)	Absolute Intensity (%) $\pm \sigma$	Photon Emission rate 4π sr \cdot s $^{-1}$ g $^{-1}$	Uncertainty (%) $\pm 3\sigma$
^{241}Am	432.7y	59.537 \pm 0.001	35.9 \pm 0.4	1.462E+04	3
^{109}Cd	462.6d	88.034 \pm 0.002	3.65 \pm 0.06	8.479E+03	4
^{57}Co	271.8d	122.0614 \pm 0.001	85.68 \pm 0.13	8.782E+03	3
^{57}Co	271.8d	136.4743 \pm 0.001	10.67 \pm 0.13	1.094E+03	3
^{139}Ce	137.6d	165.857 \pm 0.006	79.87 \pm 0.06	8.126E+03	3
^{51}Cr	27.7d	320.0842 \pm 0.001	9.85 \pm 0.09	2.868E+04	3
^{113}Sn	115.1d	391.702 \pm 0.004	64.89 \pm 0.17	4.080E+04	3.5
^{85}Sr	64.8d	514.009 \pm 0.012	99.29 \pm 0.04	3.605E+04	3
^{137}Cs	30.25y	661.660 \pm 0.003	85.2 \pm 0.2	4.395E+04	3
^{88}Y	106.6d	898.042 \pm 0.004	94.1 \pm 0.5	5.170E+04	3
^{60}Co	5.27y	1173.240 \pm 0.004	99.89 \pm 0.02	6.143E+04	3
^{60}Co	5.27y	1332.500 \pm 0.005	99.983 \pm 0.001	6.149E+04	3
^{88}Y	106.6d	1836.060 \pm 0.013	99.36 \pm 0.05	5.459E+04	3

As a numerical example, a case is quoted : 1.3382×10^{-2} g of master solution is diluted and dispensed to the foam cement, and the activity at energy 122.1 keV of a Co-57 radionuclide is to be determined at reference data of 10 May 1994. From Appendix A, the activity at reference data of 6 Dec 1993 at this energy is $8.782 \times 10^3 \text{ s}^{-1} \text{ g}^{-1}$. The activity on 10 May 1994 may be obtained (by half-life correction) as $5.9157 \times 10^3 \text{ s}^{-1} \text{ g}^{-1}$. The activity of foam cement source at this energy is $1.3382 \times 10^{-2} \text{ g} \times 5.9157 \times 10^3 \text{ s}^{-1} \text{ g}^{-1}$, which equals to $7.916 \times 10^1 \text{ s}^{-1}$.

Appendix B

Calculation involving in the preparation of foam cement source.

If

$$\begin{aligned}
 D &= \text{Intended density of the source} && \text{gcm}^{-3} \\
 V &= \text{Intended volume of the source} && \text{cm}^3 \\
 V_1 &= \text{Prepared volume (resultant in practice)} && \text{cm}^3 \\
 &\approx V + 0.2V \text{ (where } 0.2V \text{ is the loss, as waste)} \\
 &= 1.2V \text{ cm}^3
 \end{aligned}$$

and if

$$\begin{aligned}
 M_w &= \text{Mass of water} && \text{g} \\
 M_c &= \text{mass of cement} && \text{g} \\
 M_f &= \text{Mass of foam} && \text{g} \\
 V_f &= \text{Volume of foam} && \text{cm}^3
 \end{aligned}$$

the estimation of M_w , M_c , M_f and V_f can be made from the following relationships, which are gained experimentally.

$$\begin{aligned}
 M_w &= (15 \times V_1 \times D - V_1) / 62.5 && \text{g} \\
 M_c &= 3.3 \times M_w && \text{g} \\
 M_f &= V_1 \times D - (M_w + M_c) && \text{g} \\
 V_f &= 15M_f && \text{cm}^3 \\
 V_2 &= (M_w + M_c) / 2.15 + V_f && \text{cm}^3
 \end{aligned}$$

where it can be seen that $V_2 = V_1$.

As an example, for the preparation of a foam cement source of volume $V = 300 \text{ cm}^3$, the amount of M_w , M_c , M_f and V_f as a function of the source density is given in Fig. APP-B-1. Notice that from the figure, when density is increasing, M_w and M_c are increasing too, but M_f and V_f are decreasing.

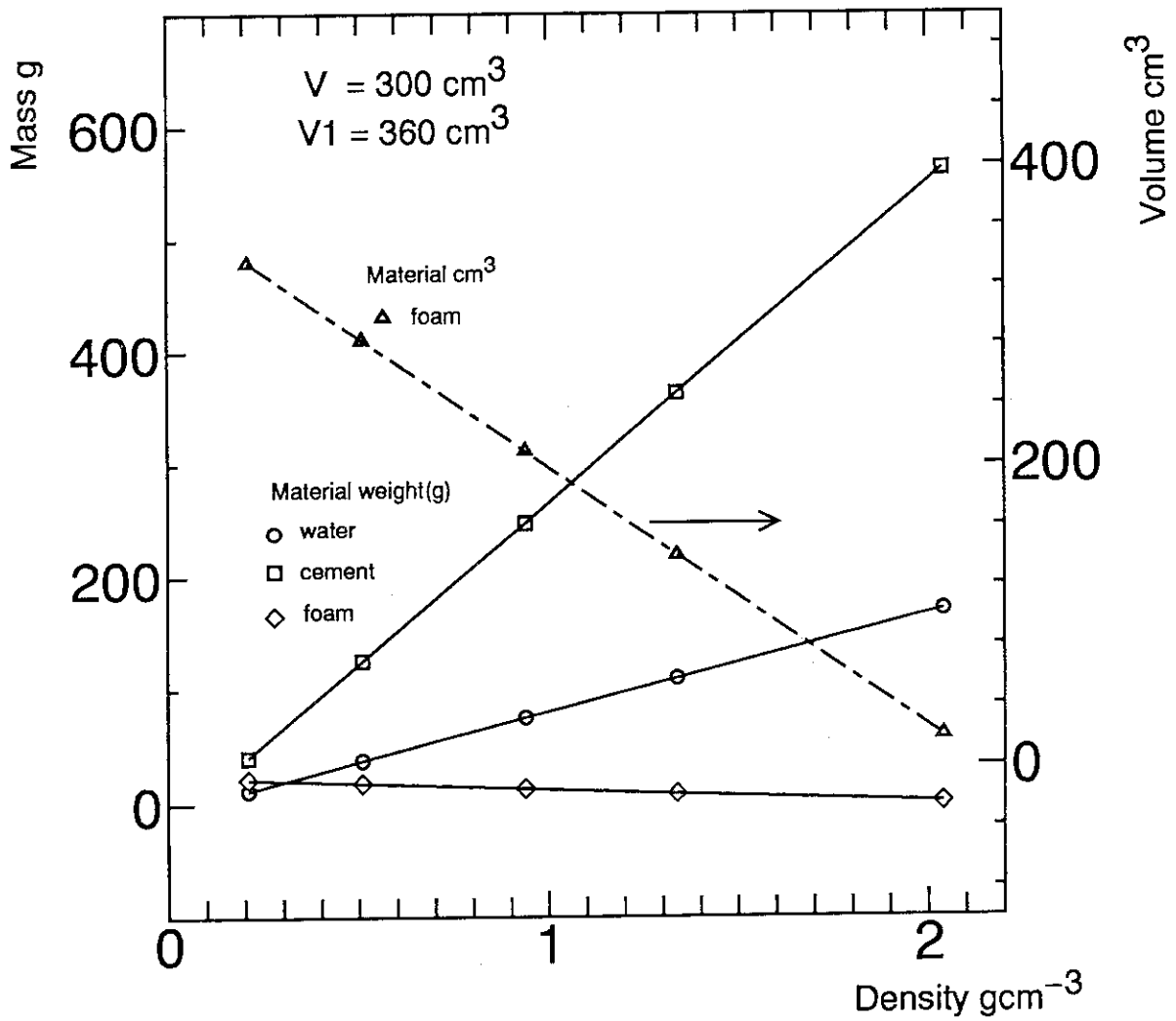


Fig.App-B-1: A guide for estimating the amount of water (g), cement (g) and foam (g and cm^3) for a certain density of the foam cement calibration source.

Appendix C

Data on the work for preparation of calibration sources

- C1 = Type
- C2 = Height
- C3 = Intended density
- C4 = Volume
- C5 = Mass of container
- C6 = Mass of beads only
- C7 = Mass of HCl+distilled water
- C8 = Mass of beads+HCl+distilled water+master solution
- C9 = Mass of cement+carrier+water+foam+HCl+distilled water+master solution
- C10 = Density of sources

Data on the work for preparation of calibration sources

C 1	C 2 (mm)	C 3 (g/cm ³)	C 4 (cm ³)	C 5 (g)	C 6 (g)	C 7 (g)	C 8 (g)	C 9 (g)	C 10 (g/cm ³)
Type-1-1	15	1.00	100	36.55	—	—	—	96.75	0.97
Type-1-2	30	1.00	200	36.51	—	—	—	188.09	0.94
Type-1-3	45	1.00	300	36.75	—	—	—	279.15	0.93
Type-1-4	45	0.20	300	35.97	—	—	—	62.03	0.21
Type-1-5	45	0.60	300	36.04	—	—	—	153.96	0.51
Type-1-6	45	1.00	300	36.53	—	—	—	282.77	0.94
Type-1-7	45	1.60	300	36.49	—	—	—	401.91	1.34
Type-1-8	45	2.20	300	36.15	—	—	—	611.85	2.04
Type-1-9	45	1.00	300	36.03	—	—	—	275.97	0.92
Type-2-1	15	—	100	36.69	—	100.00	—	—	1.00
Type-2-2	30	—	200	36.57	—	200.00	—	—	1.00
Type-2-3	45	—	300	36.09	—	300.00	—	—	1.00
Type-2-4	45	—	300	36.66	—	300.00	—	—	1.00
Type-2-5	45	—	300	36.57	—	300.00	—	—	1.00
Type-3-1A	15	—	100	36.62	144.78	37.24	182.02	—	1.82
Type-3-2A	30	—	200	36.67	304.62	79.70	384.32	—	1.92
Type-3-3A	45	—	300	35.59	460.20	119.20	579.40	—	1.93
Type-3-4A	45	—	300	35.97	453.81	113.20	567.01	—	1.89
Type-3-5A	45	—	300	36.19	446.57	115.60	562.17	—	1.87
Type-3-1B	15	—	100	36.09	152.12	35.80	187.92	—	1.88
Type-3-2B	30	—	200	36.80	300.68	73.50	374.18	—	1.87
Type-3-3B	45	—	300	36.58	450.34	113.50	563.84	—	1.88
Type-3-4B	45	—	300	37.94	444.76	110.10	554.86	—	1.85
Type-3-5B	45	—	300	37.93	454.00	110.00	564.00	—	1.88
Type-4-1	15	—	100	36.55	1.58	40.92	42.50	—	0.43
Type-4-2	30	—	200	36.02	3.38	72.20	75.58	—	0.38
Type-4-3	45	—	300	36.63	5.17	110.71	115.88	—	0.39
Type-4-4	45	—	300	36.09	5.22	110.94	116.16	—	0.39
Type-4-5	45	—	300	36.17	5.15	109.75	114.90	—	0.38
Type-5-1	15	—	100	36.56	49.19	30.48	79.67	—	0.80
Type-5-2	30	—	200	36.51	109.79	54.30	164.09	—	0.82
Type-5-3	45	—	300	36.63	164.93	93.20	258.13	—	0.86
Type-5-4	45	—	300	36.06	166.48	86.60	253.08	—	0.84
Type-5-5	45	—	300	36.17	167.28	85.00	252.28	—	0.84