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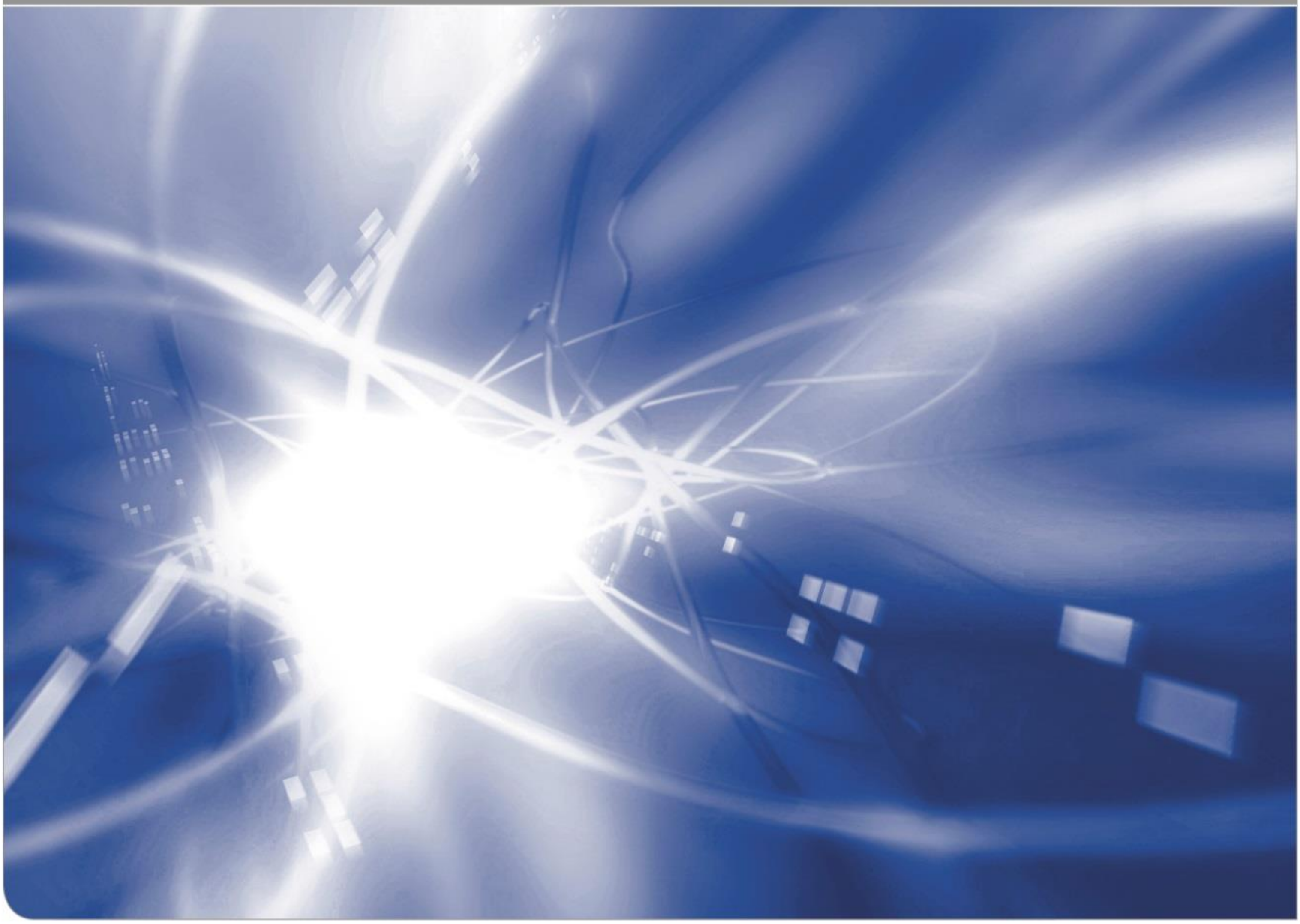
KIT SCIENTIFIC WORKING PAPERS 203



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Impressum

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2022

ISSN: 2194-1629

KFK γ-ray leakage Iron sphere benchmark with Cf source: entry for SINBAD and analysis.

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Abstract

An information necessary for the new entry in the SINBAD database was collected and thoroughly analyzed. It covers the measurement of the γ-ray leakage spectra from three iron spheres of diameter 20, 25 and 35 cm with a Cf source in the center. The experiment was performed at Kernforschungszentrum Karlsruhe (KFK) around 1977 and was an extension of measurements of the neutron leakage spectra, the latter were already compiled in SINBAD. This report includes the detailed description of the KFK experiment, the numerical γ-ray leakage spectra and other information necessary for the nuclear data validation. The MCNP input deck was assembled and used for the sample calculations of the radiation transport and sensitivities with the ENDF/B-VIII.0 neutron reaction cross section data. The spontaneous fission of ^{252}Cf was modelled as a source of neutrons and gammas, both prompt and delayed. If the neutron emission data are rather well established, the knowledge of the γ-ray data is much poorer. In the present work the prompt γ-ray spectrum and multiplicities were combined from known measurements and theoretical calculations. The knowledge of the delayed gamma-ray multiplicities and spectra from ^{252}Cf(s.f.) are still relying on the old and scarce measurements or compilations. The KFK experiment was compared with similar measurements performed at IPPE a few years later. Agreement between them, confirmed by direct comparison and similar conclusions derived from Monte-Carlo analysis, proves the reliability of both experiments.

November 2022

Content

Introduction

Validation of the evaluated cross sections data files against experimental benchmarks is the substantial and inevitable part of the reliable nuclear data evaluation process. Integral benchmarks in comparison with the cross section measurement experiments usually have much more complicated geometry and material composition. Consequently, they require essentially more efforts to collect necessary information about experimental set-up, measuring methods, and benchmark responses necessary for re-simulation and getting a feedback on the quality of the tested evaluation. For such purpose the complete compilation of experiment details and creation of the input deck for the radiation transport codes have a large value.

All such information is intended to be stored in the relevant databases such as Shielding Integral Benchmark Archive and Database (SINBAD) [1], ICSBEP [2], or others, preventing the potential users from searching through the original publications, laboratory reports or private communications. Similar objectives were formulated by dedicated subgroup 47 (SG47) “Use of Shielding Integral Benchmark Archive and Database for Nuclear Data Validation” of the Working Party on International Nuclear Data Evaluation Co-operation (WPEC) under auspicious of the Nuclear Energy Agency (NEA) in Paris during time period 2019 – 2021 [3], in which the authors took part.

The main goal of present report is a compilation for SINBAD of the γ-ray spectra leaking from three iron spheres of diameter 25, 30 and 35 cm with a ^{252}Cf(s.f.) neutron source located at the center. The measurements were performed at Kernforschungszentrum Karlsruhe (KFK), Germany, around 1977 [4 - 6]. It is worth mentioning that these measurements were an extension of a preceding experiment in which the neutron leakage spectra were measured and analysed [7 - 9]. The neutron leakage data for 15, 20, 25, 30, 35 and 40 cm diameter iron spheres were already compiled in the SINBAD as entry NEA-1517/43 [10].

It has to be noted that another similar but independent experiment was carried out at Institute of Physics and Power Engineering (IPPE) in Obninsk, Russian Federation approximately in the same time period. The neutron and γ-ray leakage spectra from six iron spheres with diameter 15, 20, 25, 30, 35 and 40 cm were measured by proportional hydrogen counter and stilbene crystal scintillator [11 - 15]. This experiment was compiled in the ICSBEP handbook under entry “ALARM-CF-FE-SCHIELD-001” [15].

This report contains a short description of the KFK Fe γ-ray leakage benchmark and relevant numerical data with their uncertainties. The reliability of KFK data were confirmed by direct comparison with the similar benchmark carried out at IPPE. A model of the KFK experiment was created as input for the MCNP code [16] and included in SINBAD allowing the users to repeat the coupled neutron-photon transport simulation. Finally, the intensive analysis of the KFK γ-ray leakage benchmark and its representativeness for validation of evaluated cross section data were demonstrated using library ENDF/B-VIII.0 [17]. The latter was processed in the ACE format by code NJOY21 [18].

The preliminary results were presented at several meetings of Subgroup 47 “Use of Shielding Integral Benchmark Archive and Database for Nuclear Data Validation” of WPEC [19 - 21].

The measured numerical data of the KFK Iron sphere γ-ray leakage benchmark and MCNP model will be distributed with next upgrade of the SINBAD database.

1. Description of the KFK experiment

The measurements of the γ-ray leakage spectra from the iron spheres with a ^{252}Cf(s.f.) neutron source were carried out in period 1975 - 1977 at Kernforschungszentrum Karlsruhe (KFK) [4 - 6]. These measurements were an extension of the preceding experiments in which the neutron leakage spectra were measured and analysed [7 - 9].

Following sub-sections will describe the details of the KFK experiment with a specific attention to the parameters necessary for the MCNP input creation and Monte Carlo modelling. The simulation results will be presented in the subsequent sections.

1.1. Iron spheres and experimental room

Three iron spheres having outer diameters 25, 30, and 35 cm were investigated in the KFK experiment. The set-up arrangement is shown in Fig. 1.1. The spheres were hung below the top of tripod (three-feet holder) located in the hall. The minimal distance was ≈ 2 m to the floor and ≈ 3 m to the nearest walls [7].

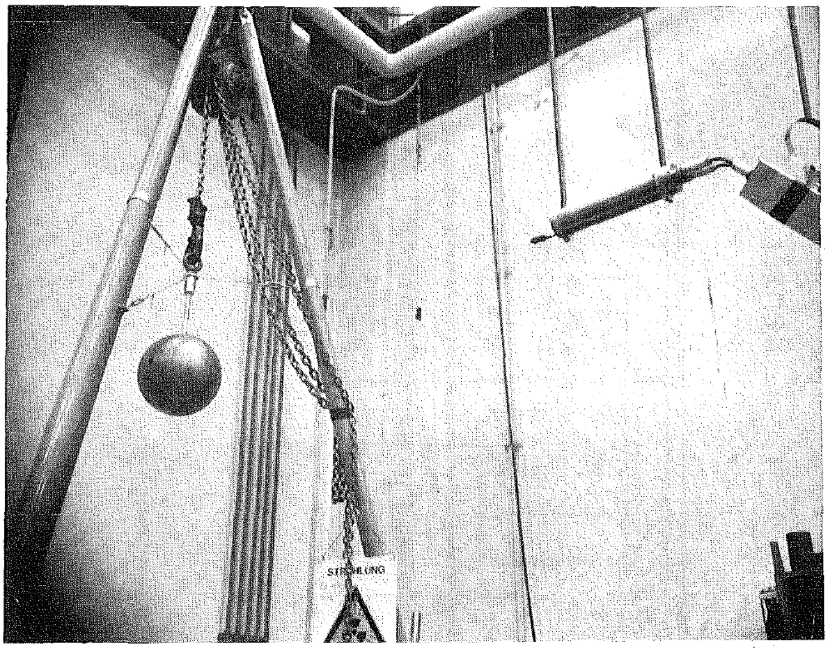


Fig. 1.1. The experimental set-up of the KFK measurements of the neutron or gamma–ray leakage spectra from the Iron spheres with a ^{252}Cf source in the center (figure is copied from Report KFK-2219 [7]).

The spheres were fabricated from very pure soft iron cylindrical discs. The performed chemical analysis of iron composition has shown following impurities: C - 0.07, Mn - 0.05, P - 0.009 and S - 0.007 wt%. The spheres had the central cylindrical channel Ø1.6 cm for inserting capsule with a Cf-source as shown in Fig. 1.2. During the measurements this channel was filled up from both sides by the cylinder plugs made of the same iron material.

1.2. ^{252}Cf source of neutrons and gammas

In the time period 1975 to 1977 the KFK laboratory used two ^{252}Cf sources one of them was called "small" and another - "large" that reflected their neutron intensity. For the described experiments with iron spheres the small ^{252}Cf neutron source was used. It consisted of ≈ 30 μg of ^{252}Cf distributed in the SiO_2/MgO matrix. The matrix was contained in the two capsules: internal one made of Zircaloy (Zr + 1.5 wt% Sn) and external – AlMg3 (Al + 3 wt% Mg). Fig. 1.2 shows the geometrical configuration, sizes and material components of the "small" Cf-source.

The source strength was determined by three independent methods: comparison with standard ^{252}Cf source, measuring by the calibrated ^{235}U fission chamber and by proton-recoil neutron spectrometer. These three independent calibrations resulted to the well agree values that allowed to estimate the absolute strength of Cf-source as 5.51 10^{+7} n/sec with uncertainty ± 5%.

The isotopic composition of the Cf-source was not reported.

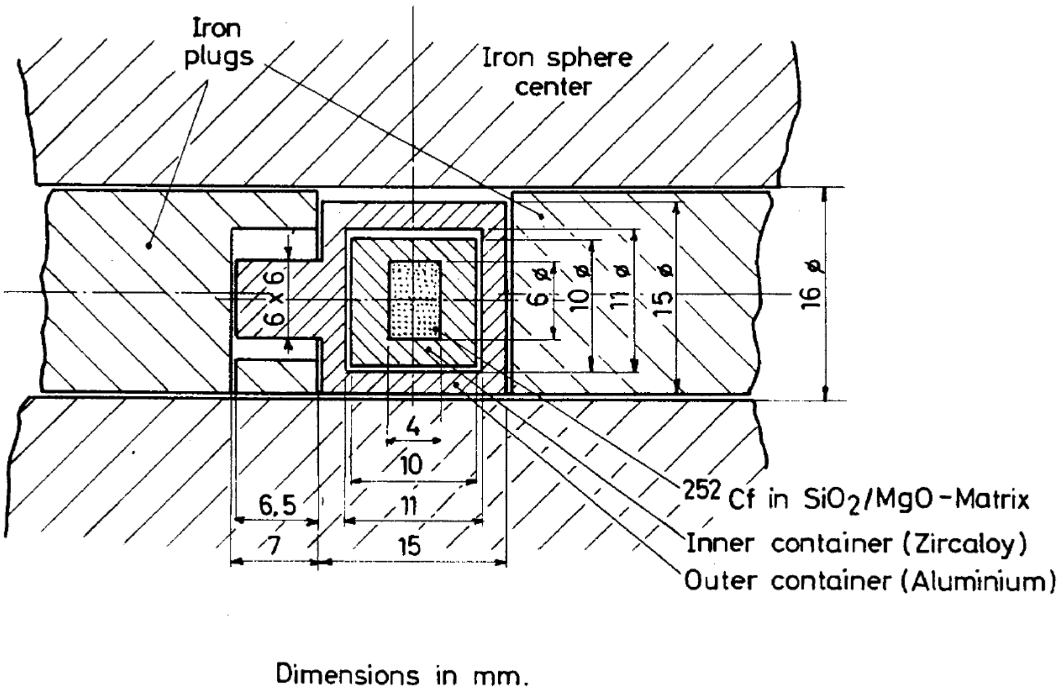


Fig. 1.2. The geometrical configuration and material composition of the Cf-252 source and its location in the central region of iron spheres. The sizes are given in mm. Figure is a copy from Report KFK-2444 [4].

1.3. Si(Li) Compton spectrometer of γ-rays

The energy distributions of the γ-rays were measured by the Si(Li) semiconductor Compton spectrometer. Such detector was selected by authors of this experiment as one already proved to be suitable for measurements in the mixed gamma–neutron fields [22, 23]. The γ-rays with energies 0.3 – 3.0 MeV deposit their energy mainly to Compton electrons. Unfolding of the electron energy distribution yields the absolute energy spectra of the incident γ-rays.

The configuration of the used Si(Li) semiconductor is depicted in Fig. 1.3. It had a sensitive volume with area 110 mm^2 and depth 5 mm and was packed under vacuum in the Al capsule. The distance between the outer front surface of detector and the middle plane of the active zone was 1.55 cm.

The detector response functions, i.e. the electron pulse height distributions induced by the mono-energetic γ-rays, which are required for unfolding, were determined experimentally. For this a set of seven mono-energetic γ-ray sources which cover the energy range 0.511 to 2.754 MeV was used. Since the response of the Si(Li) detector depends on the incident angle, two response functions were determined: one with the γ-sources on the axis of the detector (denoted by the authors as "axial" response) and the other with the γ-sources in the radial symmetry plane of the detector ("radial" response). Below the level of the electronic noises (≤ 200 keV), the measured response functions have been extrapolated, that was a main contributor to the systematic error.

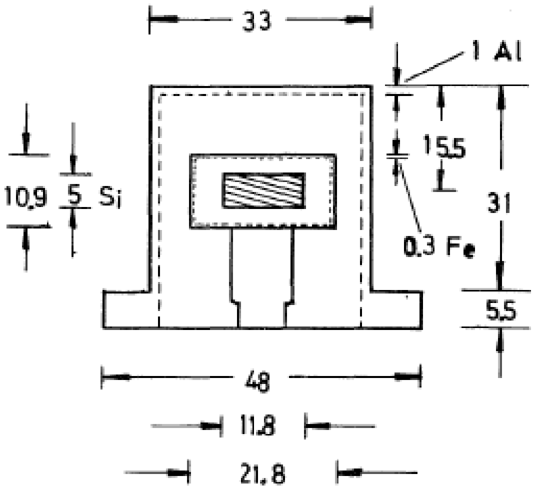


Fig. 1.3. The geometrical configuration and material components of the Compton Si(Li) γ-ray detector. The sizes are given in mm. Figure is a copy from Report KFK-2444 [4].

The absolute efficiency of the detector was determined with a help of absolutely calibrated ^{137}Cs source. The absolute energy calibration of the pulse amplitude distribution was carried using the position of the photo-peaks observed with several calibrated γ-ray sources.

Since the energy resolution of the Si(Li) spectrometer is not given in numerical form, we found it from the full width of the peaks (FWHM) observed during calibration of the detector with calibrated γ-ray sources and presented in Fig. 2.10 of Report FZK-2444 [4] (the FWHM uncertainties are supposed to be ≈ 30% due to insufficient quality of the figure). The experimental relative energy resolution of the Si(Li) spectrometer derived in such a way is plotted in Fig. 1.4. We fitted the measured points by expression $a + b/\sqrt{E}$ which was then used for Gaussian smoothing of the γ-ray tallies of MCNP. It is seen that energy resolution decreases from 20 to 7% when γ-ray energy increases from 0.5 to 3.0 MeV.

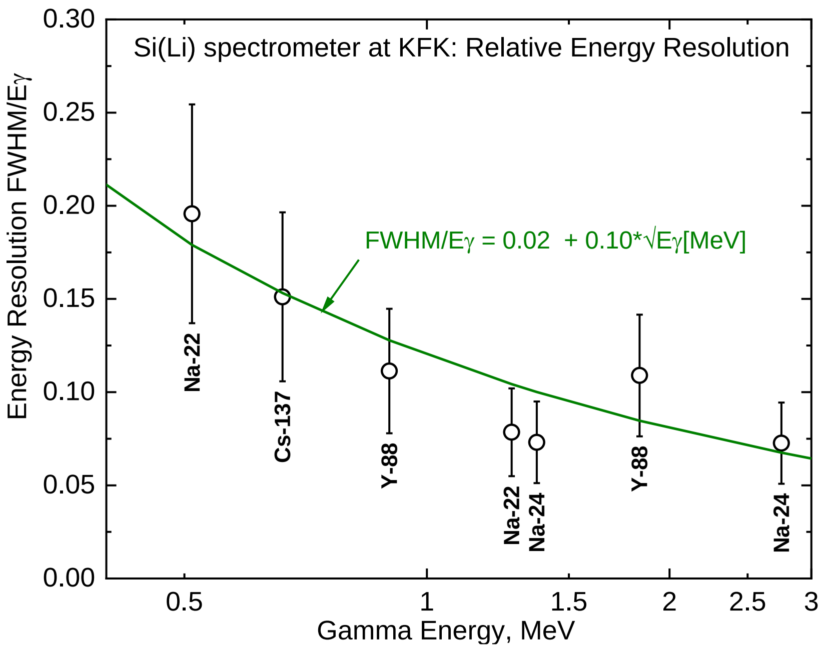


Fig. 1.4. The relative energy resolution of the Si(Li) Compton spectrometer. Symbols – experimental data derived from Fig. 2.10 of Report KFK-2444 [4], curve – fit, which we use for Gaussian smoothing of γ-ray tallies of MCNP. Isotope labels indicate the calibrated γ-ray sources used at KFK.

1.4. Measured γ-ray energy spectra from the bare ^{252}Cf source and Iron spheres

The Compton electron energy distributions induced by the γ-rays were recorded by the Si(Li) detector. The spectrometer was positioned at distance 102.5 cm from the bare Cf-capsule to measure the source γ-ray spectrum or on the outer surface of the iron spheres to measure the leaking γ-rays. The energy spectra of the γ-rays were unfolded from measured electron spectra employing both the axial and istoropic response functions (the latter one being the average of axial and radial). The difference between results fluctuates within (3 – 5)%, that is lower than other uncertainties (see next section). ***NB: the authors of KFK experiment have stated that the axial evaluation of response function should be a better approximation and thus they recommended to use the leakage γ-ray spectra obtained with an axial response functions***.

For the unfolding of the γ-ray spectra from the measured Compton electron energy distributions modified version of the code SPEC-4 [24, 25] was used. During this procedure the contribution of γ-rays with energies above ≈ 3 MeV were not considered. The width of the energy bins for unfolded results was chosen to be ≈ 7% of γ-ray energy.

The numerical measured data were made available in Oct 2019 by Prof. Shiang-Huei Jiang for the author (S.S.) of present report. After their comparison with data digitized in Fig. 2.20 of Report KFK-2444 [4] it was found that the γ-energy scale in the received tables has to be shifted downwards by 4 energy bins. After this, the coincidence with γ-energies available in tables for the iron pile experiment with ^{228}Th γ-source could be considered as an additional confirmation.

The received (and eventually included in SINBAD) numerical data contains γ-rays spectra for:

- bare Cf-source obtained with the help of the axial response function of the Si(Li) detector and
- three iron spheres of outer diameters 25, 30 and 35 cm unfolded with axial and isotropic response functions of the Si(Li) detector.

The isotropic response is, in turn, an averaging of the axial and radial responses and was used for measurements inside the iron pile.

These data are plotted in the top part of Fig. 1.5, i.e. the energy spectra of γ-rays emitted by the bare Cf source and the spectra of γ-rays leaking from three Fe spheres of diameter 25, 30 and 35 cm. The latter are double: unfolded from the measured electron distributions by axial and isotropic response functions of the Si(Li) detector. The relative difference between them, as seen in the bottom part of Fig. 1.5, fluctuates within ± (0 – 5)%.

Following the recommendation of the authors, we used in the further analysis the data obtained with Si(Li) detector response function calibrated with γ-sources on the its axis. Additionally, for the comparison with transport calculations we used the angle integrated γ-ray leakage spectra, i.e. after multiplication by factor $4\pi L^2$. Where the distance between Cf-source and Si(Li) detector $L = R_{out} + 1.55$ cm, R_{out} being the outer radius of iron sphere.

1.5. Experimental uncertainties of the γ-rays spectra

Several components of uncertainties, resulting from the Cf-source neutron strength and γ-ray energy spectra, were quantified by the authors of the experiment. They are plotted in the bottom part of Fig. 1.5.

The accuracy of the ^{252}Cf-source neutron source strength was estimated to be ± 5%. This value is based on the scattering of the values received with the various calibration methods (see section 1.2).

The systematic errors of γ-ray spectrum measurements stemmed mainly from the errors in the response function. Its components, as specified in [4, 5], were: uncertainty of the absolute efficiency determination with a help of calibrated ^{137}Cs source (≈ 2%); energy dependence of the γ-rays absorption in detector walls (≈ 2%); extrapolation of the measured response function to zero energy (< 10%). The total uncertainty caused by the response functions was estimated to be ≈ ± 10% that was derived from comparison of measurements with several calibrated gamma sources.

Additional uncertainty resulted from the electron spectrum unfolding, namely from neglecting of the γ-rays contribution with energies above ≈ 3 MeV which are produced by the neutron capture and inelastic scattering. Such influence of the high-energy γ-rays and corresponding systematic error were found by varying the upper energy limit from 2.7 to 1.8 MeV. The authors of benchmark have concluded that the high-energy γ-rays will not disturb the measured γ-ray spectra below ≈ 1 MeV. Between 1 and 2 MeV, the errors up to 10 and 20% were expected for all spheres. We linearly interpolated the latter uncertainty between 1 and 2 MeV.

The statistical error of the single experimental point (energy dependent uncertainty) varies between 5 and 10%.

In addition, it was stated that below 0.5 MeV the neutron background disturbs the gamma spectra in a mixed neutron-gamma field. ***Finally, the authors of KFK experiment have recommended to restrict the quantitative interpretation of the measurements to the energy region between 0.5 and 2 MeV***.

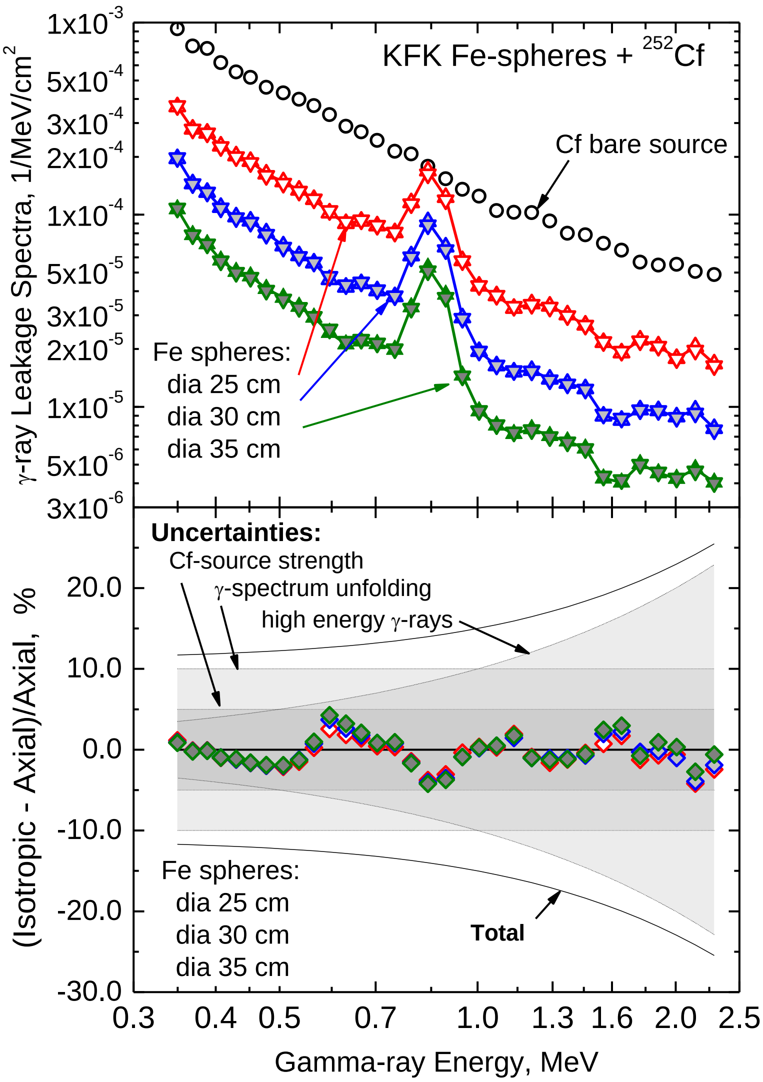


Fig. 1.5. Top: spectra of γ-rays leaking from the KFK bare Cf-source and from three iron spheres of outer diameters 25, 30 and 35 cm, obtained with axial (up-forward symbols) and isotropic (down-forward symbols) response functions of the Si(Li) detector. Bottom: relative difference of these γ-rays leakage spectra for each sphere as well as the total uncertainty and its components.

2. Comparison of the KFK and IPPE measurements for Ø30 cm Fe sphere

It is extremely seldom when benchmark responses measured in the independent laboratories could be directly compared. In the present case it becomes possible since practically identical iron sphere, namely of the outer diameter 30 cm (the configuration of the inner holes are comparable), was also measured in IPPE, Obninsk [11 - 15]. The IPPE experiment is sketched–out in Fig. 2.1. The γ-ray spectra have been measured by stilbene scintillation detector located at distance approximately equal of three outer radius of sphere in the energy range 0.4 to 11 MeV. The shadow cone has allowed to measure the background.

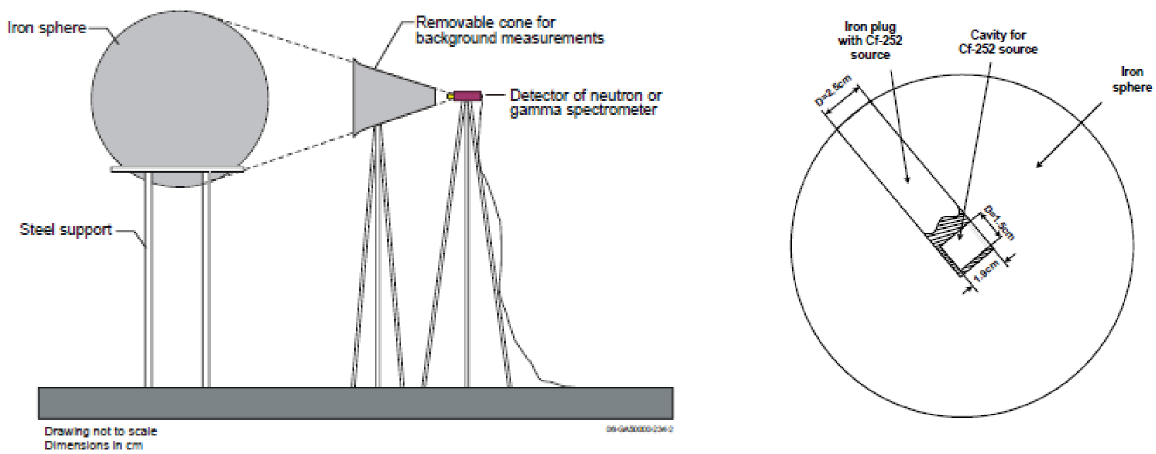


Fig. 2.1. The lay-out of the IPPE experiment (left) and arrangement of the iron sphere with cavity for ^{252}Cf source closed by the iron plug (right) [11 - 15].

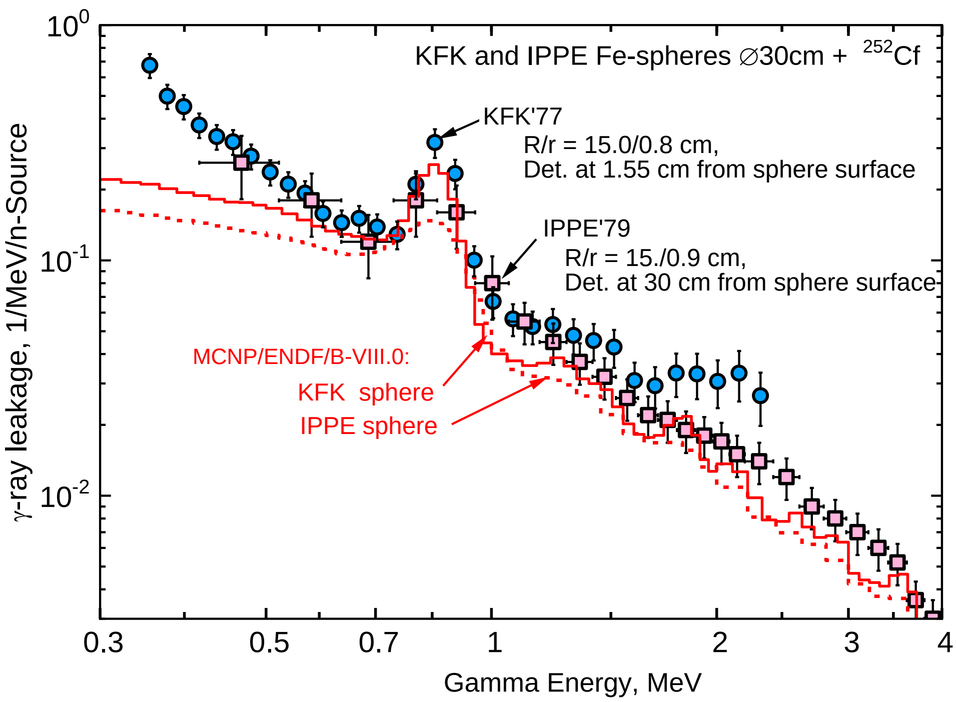


Fig. 2.2. Comparison of the spectra of γ-rays leaking from two Fe spheres of the same outer diameter 30 cm with ^{252}Cf source in the center, which were measured in KFK and IPPE. Symbols – experimental data: circle – KFK sphere, squares - IPPE; curves – MCNP transport calculations with the ENDF/B-VIII.0 data: solid – for KFK sphere, dashed - IPPE.

The γ-ray spectra from the IPPE and KFK experiments for the iron shells of diameter 30 cm are compared in Fig. 2.2. It is clear seen, that in energy interval 0.5 - 2 MeV the both experiment data agree within declared uncertainties 10 – 20%. The results of the MCNP simualtions with ENDF/B-VIII.0 show the difference (≈ 10%) which should be between the KFK and IPPE benchmarks if all their experimental configuration details are considered.

3. Physical parameters used to model KFK iron shell experiment with a ^{252}Cf source

This section describes the underlying physics and data which are needed to simulate the KFK experiment.

The basic properties of the radiations which follow the ^{52}Cf disintegration are listed in Table 3.1. ^{252}Cf decays with probability (96.914 ± 0.008)% emitting α-particles and also undergoes spontaneous fission with probability (3.086 ± 0.008)%. The corresponding half-lives are (2.6470 ± 0.0026) and (85.76 ± 0.23) years. These data are taken from the Decay Data Evaluation Project (DDEP) [26].

Table 3.1. Main parameters of the neutron and γ-ray radiations from disintegration of ^{252}Cf. Multiplicities are normalized per spontaneous fission event (f). The origin of the energy spectra used in present simulations is referenced.

	Neutrons			Gammas		
	Multiplicity ν, n/f	$\langle E_n \rangle$, MeV	Origin of Spectrum	Multiplicity M_γ, γ/f	$\langle E_\gamma \rangle$, MeV	Origin of Spectrum
	Spontaneous Fission: P_{sf} = (3.086 ± 0.008)%, $T_{1/2}$ = (85.76 ± 0.23) y					
Prompt	3.7590 [17]	2.122	Standards [27]	11.0	0.730	present work
Delayed	0.0086 [29,17]	0.464	ENDF/B-VIII.0 [17]	10.7	0.774	Stoddard [30,31]
Total	**3.7676** [28,17**]**			**21.7**		
	Alpha Decay: P_α = (96.914 ± 0.008)%, $T_{1/2}$ = (2.6470 ± 0.0026) y					
Total				**0.0089**	**0.070**	DDEP [26]

Spontaneous fission of ^{252}Cf is a source of the neutrons and γ-rays. There are two mechanisms of their emission:

- prompt (within the first ≈ 10^{-8} - 10^{-6} s), i.e. during the fission of ^{252}Cf and fast de-excitation of the primary fission products;
- delayed (time scale > 10^{-8} - 10^{-6} s), i.e. after β-decay or isomeric de-excitation of the fission products and residual nuclei.

Alpha decay of ^{252}Cf is an additional source of the γ-rays. This process populates the ground state of ^{248}Cm with probability 81.7% and the first excited levels with probability 15.33% [26]. The de-excitation of these levels proceeds mainly via internal electron conversion, but also and with lower rate – via emission of the discrete γ-rays with energies (and probabilities per ^{252}Cf disintegration): 43.40 keV (0.0152%), 100.2 keV (0.0119%) and 154.5 keV (0.00051%) [26]. The γ-ray multiplicity $M_\gamma(\alpha)$ caused by alpha decay being normalized per one ^{252}Cf(s,f.) event equals 0.0089 γ/f or only 0.043% of the total spontaneous fission γ-multiplicity M_γ. Due to this reason the gammas from ^{252}Cf(α)^{248}Cm were excluded from our further consideration and from the assembled MCNP input.

Since KFK experiment did not use any time coincidence technique, both prompt and delayed mechanisms of ^{252}Cf spontaneous fission should be represented in the MCNP model. In other words: the source description cards should sample corresponding multiplicities and energy distributions of emitted neutrons and gammas. The modelling details will be described in the next sub-sections.

3.1. ^{252}Cf(s.f.) as a source of neutrons

The prompt fission neutron spectrum (PFNS) from spontaneous fission of ^{252}Cf was rather well measured. In the emission neutron energy range from 0.1to 15 MeV it was adopted with uncertainties 1 – 12% as a standard [27]. For our Monte-Carlo simulations the numerical PFNS data in the 725 groups presentation were taken from database IRDFF-II [32]. The prompt neutron multiplicity, i.e. the number of prompt neutrons per spontaneous ^{252}Cf fission, ν_p was taken to be equal to 3.7590 ± 0.0048 [17].

The delayed neutron spectrum (DFNS) was extracted from ENDF/B-VIII.0 evaluation [17]. The number of delayed neutrons per spontaneous ^{252}Cf fission, ν_d = 0.0086 ± 0.0010 [29, 17].

Both spectra normalized per total neutron multiplicity $\nu_{tot} = \nu_p + \nu_d$ = 3.7676 ± 0.0047 are displayed in Fig. 3.1. The fraction of delayed neutrons ν_d / ν_{tot} = 0.23% is relatively small, however the average delayed neutron energy $\langle E_d \rangle$ = 0.464 MeV is several times less than average energy of prompt neutrons $\langle E_p \rangle$ = 2.122 MeV. Consequently at neutron energies below 0.1 MeV the fraction of the delayed neutrons amounts already more than 20% of the total neutron yield, Fig. 3.1.

Fig. 3.1 also depicts two neutron spectra from the bare Cf sources (^{252}Cf within capsule) measured at KFK and IPPE, which are the sum of the prompt and delayed neutrons. It is seen that the KFK data have rather large fluctuations and overestimate PFNS above 10 MeV. The both measured spectra are cut-off below 0.1 - 0.2 MeV.

Due to these reasons the MCNP model created for the KFK experiment presents the ^{252}Cf(s.f.) as a source of prompt and delayed neutrons with energy distributions and multiplicities from the aforementioned Standards and ENDF/VIII.0 evaluation. To account for impact of the Cf-source capsule its geometry and material composition was fully modelled following the description given in section 1.2 and in Fig. 1.2.

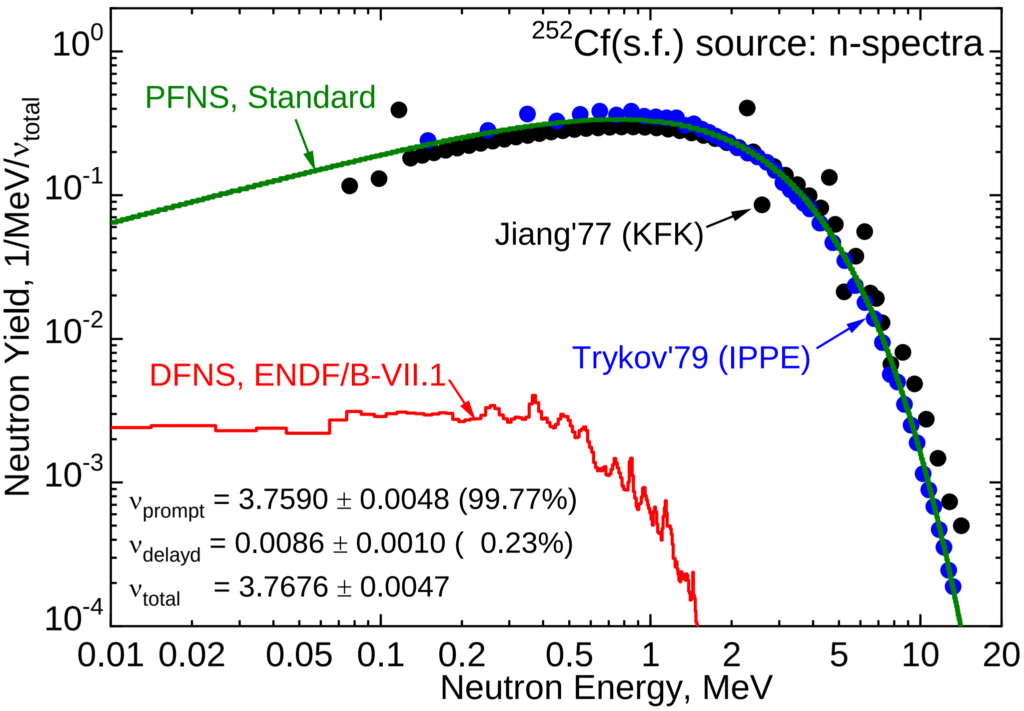


Fig. 3.1. Total neutron spectra from the bare Cf-sources measured in KFK [4] and IPPE [11]: black and blue circles. The prompt (PFNS) and delayed (DFNS) fission neutron spectra from spontaneous fission of ^{252}Cf(s.f.) from IRDFF-II (Standards) [32] and ENDF/B-VIII.0 [17]: curves.

3.2. ^{252}Cf(s.f.) as a source of gammas

To represent the ^{252}Cf(s.f.) as a source of γ-rays in the MCNP model it is necessary to know the yield of gammas per fission event as well as their energy distribution. Similar to the neutron emission the prompt and delayed mechanisms of the γ-radiation emission are expected. The sources of delayed γ-rays are de-excitation of isomeric states in the fission products and gammas which follow the betta-decay. The prompt γ-ray emission is usually measured by the gamma-fission (γ–f) coincidence technique.

The collection and analysis of the experimental and evaluated data for the γ-ray multiplicities and spectra from ^{252}Cf(s.f.) were presented by authors of this report at series of expert meetings [19 - 21, 33, 34].

Gamma-ray Multiplicities. EXFOR data base [35] contains rather many independent experiments carried out in different labs during a several decades [36 - 50]. Such measurements are usually made with a tiny layer of ^{252}Cf deposited on the thin electrode of the fast fission chamber. Ionization induced by the fission fragments in the filling gas is used to count the number of fission events as well as a stop signal for the time window (typically of order of several μs) in which the prompt γ-ray flash is selected (γ–f coincidence). In this way only the prompt γ-ray multiplicity is measured.

The γ-ray spectrometers have the individually defined low energy cut-off (threshold), which will impact on the measured integral value M_γ(prompt). The experimental Mγ(prompt) found in the literature are plotted in Fig. 3.2 versus the γ-ray detection threshold used. The decreasing tendency is clearly visible, that allows an extrapolation to zero threshold and estimate total multiplicity:

- Experimental M_γ(prompt, $E_\gamma > 0$) = 11.0 γ/f or = 2.9 γ/n (after division by ν_n(tot) = 3.7676 n/f).

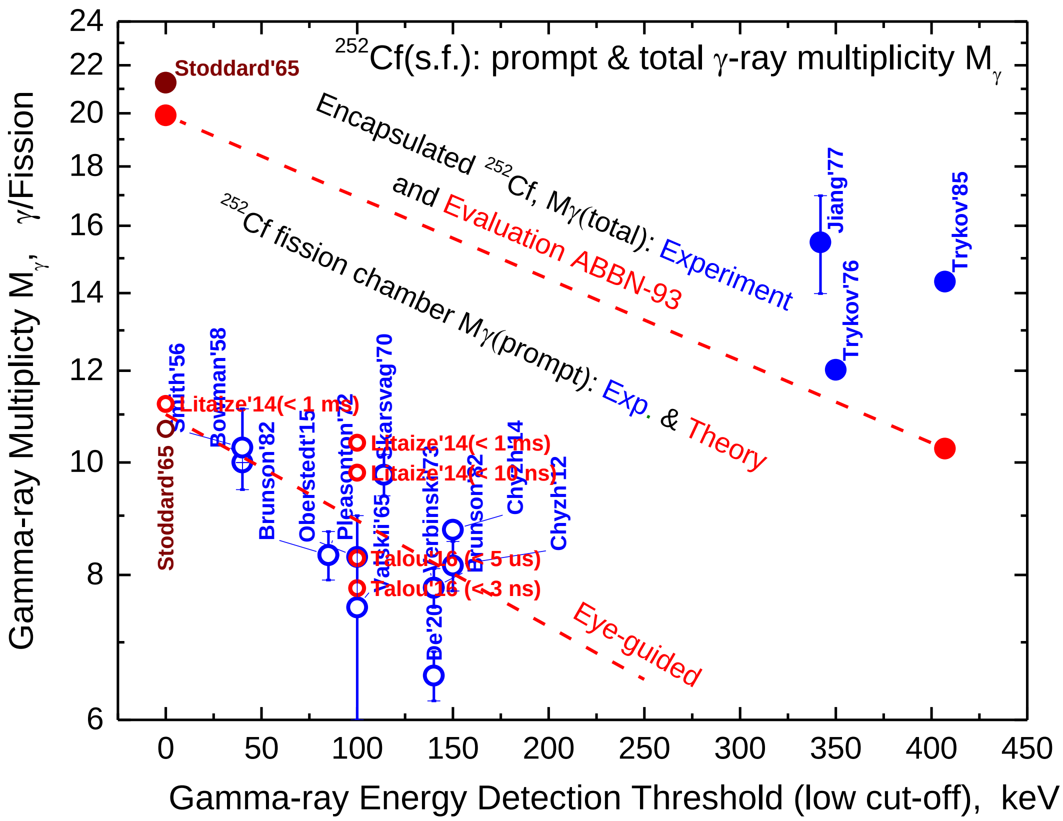


Fig. 3.2. Total and prompt gamma-ray multiplicities for ^{252}Cf(s.f.) source versus the detection threshold. The known measurements are shown by blue symbols: open – prompt emission, closed – total. Theoretical and evaluated M_γ – red or dark red symbols (the time integration interval considered by theory after ^{252}Cf fission is given). Dashed red curves are the eye guided trend.

Fig. 3.2 also displays results of the theoretical nuclear fission modelling [51, 52]. The latter slightly depend on the time interval (typically nano – milli seconds) during which the gamma emission is considered. As seen in figure, the theoretical predictions do agree with the experimental data trend.

For comparison, Fig. 3.2 also shows the total γ-ray multiplicities M_γ(total) derived from the γ-ray spectra measured with the bare encapsulated Cf-sources at KFK [4] and IPPE [13]:

- KFK Cf-source (sizes = 2 - 3 mm SiO_2/MgO matrix + 2 capsules 2 - 3 mm ZrSn and 2 mm Al):
  Mγ(total, E_γ > 500 keV) = 3.2 γ/n or (after multiplying by ν_n(tot) = 3.7676 n/f) = 12.1 γ/f;
- IPPE Cf-source (capsule sizes = 3.2 mm Fe + 2.3 mm Cu):
  Mγ(total, E_γ > 400 keV) = 3.8 γ/n or (after multiplying by ν_n(tot) = 3.7676 n/f) = 14.3 γ/f.

The known evaluated total γ-ray multiplicities (with zero threshold) from D. Stoddard [30, 31] and ABBN-93 (taken data from [15]) are:

- Stoddard: M_γ(total, E_γ > 0) = 5.6 γ/n or (after multiplying by ν_n(tot) = 3.7676 n/f) = 21.7 γ/f.
- ABBN-93: M_γ(total, E_γ > 0) = 5.3 γ/n or (after multiplying by ν_n(tot) = 3.7676 n/f) = 19.9 γ/f.

As seen in Fig. 3.2 these evaluated data generally reproduce the data measured with encapsulated Cf sources.

In general, the γ-rays multiplicities derived from the KFK and IPPE encapsulated Cf-sources are about 2 - 3 times as large as ones measured with the miniaturized ^{252}Cf fission chamber sources within ≈ 1 μs time window after fission for selection of the γ-rays. The reason is that these two types of experiments deliver different γ-ray multiplicities, namely total and prompt, correspondingly.

Gamma-ray Spectra. The top of Fig. 3.3 plots all known measured Prompt Gamm-ray Fission Spectra (PFGS) measured with miniaturized Cf fission chambers (open symbols). The numerical data are taken from EXFOR and are usually normalized per one spontaneous fission event (f). There are several exceptions - data were presented by authors in arbitrary units. The Total Fission Gamma Spectra (TFGS) measured at KFK and IPPE with encapsulated Cf sources are also displayed in Figure as solid symbols.

To demonstrate (dis)agreement of measured data in reasonable scale the bottom part of Fig. 3.3 displays the ratio of measured spectra over the analytical function which is a sum of two Maxwellian distributions. It was difficult to find an optimal representation of the fission gamma spectra, thus the ratio still oscillates around unity. It is seen that: (i) PFGS measured at different labs are rather reasonable agree each other at γ-ray energies Eγ between 0.1 and 10 MeV; (ii) TFGS measured at KFK and IPPE agree in the shorter interval from 0.5 MeV to 2 – 3 MeV; and (iii) the TFGS absolute values are about 3 times as large as PFGS.

Fig 3.3 also shows that the experimental prompt fission gamma spectra are reasonable reproduced by the theoretical modelling of P. Talou et al. [52]. The total fission γ-ray spectra from ABBN-93 evaluation reproduces the IPPE spectra only below ≈ 5 MeV, but above - substantially overestimates. Evaluation of D. Stoddard et al. [30, 31] well predicts PFGS up to 7 MeV, whereas his Delayed Fission Gamma Spectra (DFGS) is limited by ≈ 2 MeV at highest energy.

Generally, the DFGS is rather poor studied. There are several measurements of the yields of delayed (or total) discrete γ-rays, however the maximum energy reported is below ≈ 1.8 MeV [53. 54] (that confirms the high energy cut-off form the Stoddard' evaluation).

Relying on information gathered above we have eventually selected following data to be used for simulation of the ^{252}Cf(s.f.)γ multiplicities and energy spectra:

- for prompt emission: γ-ray energy spectra as a combination of experiments of S. Oberstedt (0.3 MeV < Eγ < 10 MeV) and D. Pandit (Eγ > 10 MeV), theory of P. Talou (0.1 MeV < Eγ < 0.3 MeV) and eye guided interpolation (Eγ < 0.1 MeV);
  multiplicity Mγ(prompt) = 11.0 γ/f;
- for delayed emission: γ-ray energy spectra from D. Stoddard (0.0 MeV < Eγ < 2.0 MeV);
  multiplicity Mγ(delayed) = 10.7 γ/f.

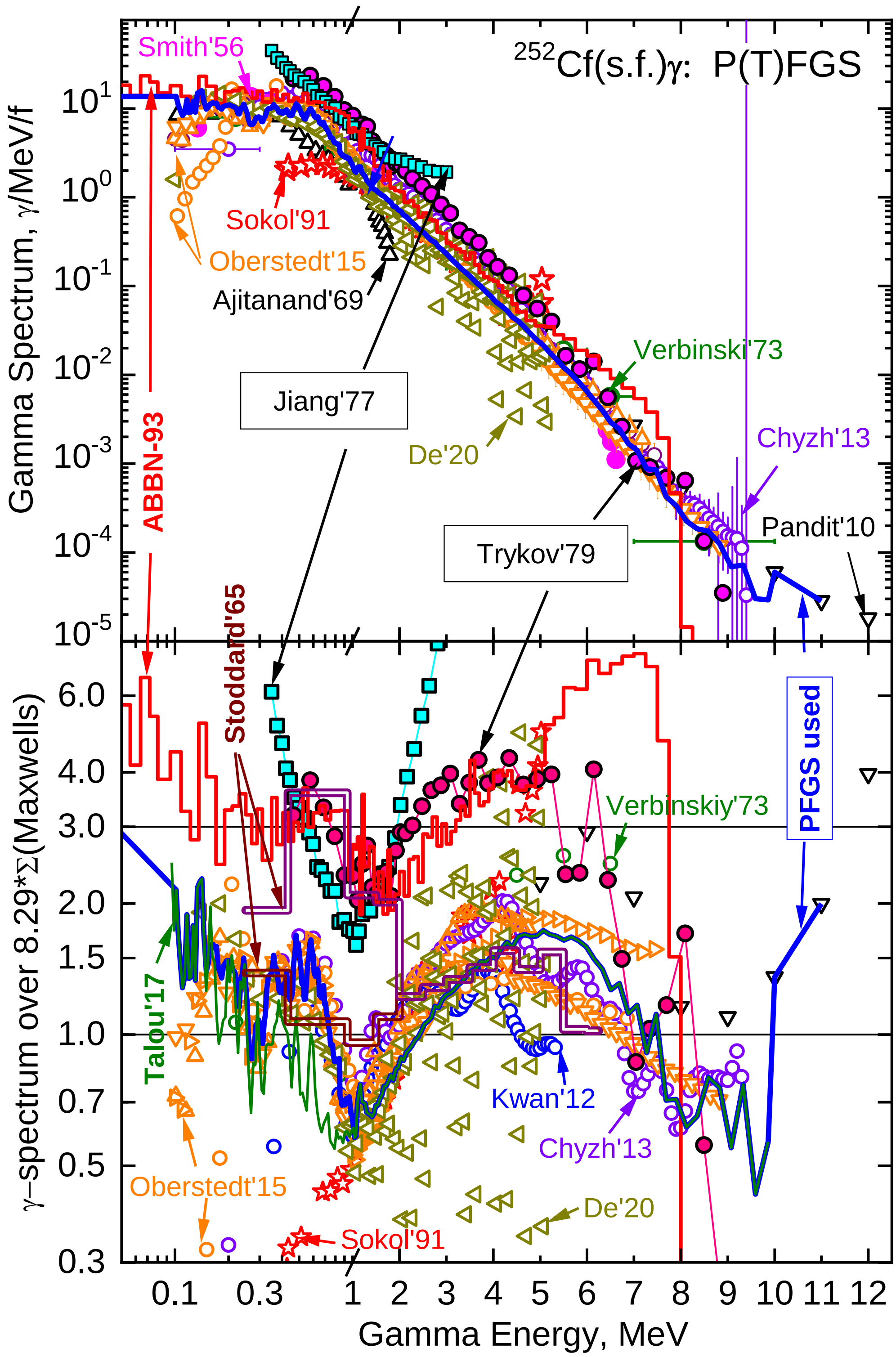
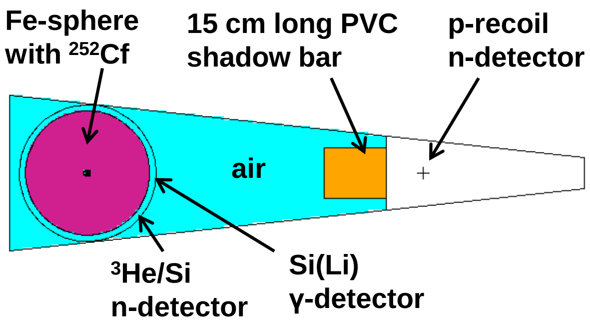


Fig. 3.3. γ-ray spectra from ^{252}Cf(s.f.) (top) and ratio over the sum of two Maxwellian distributions (bottom). **Experiment**: prompt emission - measured with a ^{252}Cf fission chamber (open symbols) and total – measured with encapsulated Cf (closed symbols: ■ - KFK and ● - IPPE). **Theory or Evaluation**: histograms or curves show the prompt γ-ray spectrum theoretically calculated by P. Talou [52], prompt and total spectra evaluated by D. Stoddard [30, 31]; total spectrum from evaluation ABBN-93 [15]. Thick blue curve – ^{252}Cf(s.f.) prompt γ-ray spectrum compiled in the present work and used in the Fe sphere benchmark simulations. *Note the change of the energy scale at 1 MeV.*

4. MCNP input deck created for the KFK iron spherical benchmark

This section describes the input deck assembled for the KFK iron benchmark. The features most essential for calculation of the relevant responses and nuclear data benchmarking are presented. The other details are available in the input deck in the form of the self-explaining comments. The MCNP geometry model of the KFK experiment is displayed in Fig. 4.1.



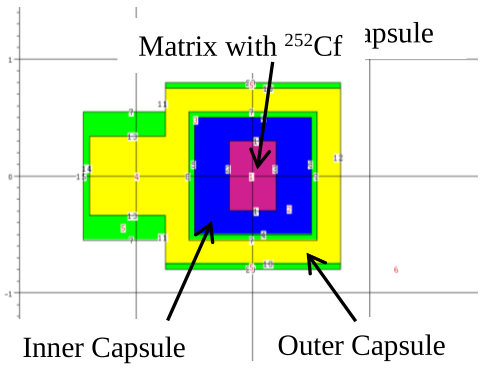


Fig. 4.1. The MCNP geometrical model of the KFK experiment set-up (left) and Cf-source representation (right) for simulation of the γ-ray and neutron leakage spectra from the iron spheres with a Cf source in the centre.

<u>The Cf-source of the KFK experiment</u> was modelled in the MCNP deck with a help of the SDEF and several distribution cards. The prompt and delayed neutrons and gammas from spontaneous fission of ^{252}Cf are sampled simultaneously as four independent particles. The real physical emission probabilities, i.e. multiplicities, are applied to the sampled particle weights after renormalization to one total neutron multiplicity ν(tot) = 3.7686. MCNP reads the energy distributions of the neutrons and gammas from the external separated text files:

1) Cf252_nPrompt_725g.dat – Prompt Fission Neutron Spectrum (PFNS) in 725 groups,
2) Cf252_nDelayed.dat – Delayed Fission Neutron Spectrum (DFNS),
3) Cf252_gPrompt.dat – Prompt Fission Gamma Spectrum (PFGS),
4) Cf252_gDelayed.dat – Delayed Fission Gamma Spectrum (DFGS).

The origin of the corresponding neutron and γ-ray multiplicities and their energy spectra are described in the previous Section 3.

It was supposed that isotope ^{252}Cf is uniformly distributed in the SiO_2/MgO matrix which is surrounded by two capsules as outlined in Figs. 1.2 and 4.1. Their chemical compositions and weights are given in Report KFK-2219 [7]. Regrettably but authors have reported the elemental composition only for the whole Cf-source structure, that makes difficult to model separately the matrix and two capsules. To overcome this we used the given weights and sizes of each source components to calculate their mass and atom densities. The elemental and isotopic fractions were calculated with a help of the MCNP utility *mattool* [16]. The results are listed in Table 4.1 showing that the calculated sum density of all elements is in a good agreement with those given by the authors of KFK experiment.

<u>The iron sphere</u> is presented by sphere with the variable outer radius R = 12.5, 15.0 and 17.5 cm to choose the desired diameter of sphere 25, 30 or 35 cm for the γ-ray leakage experiment simulation. Additionally, user can select diameters 15, 20 or 40 cm to compute the neutron leakage which was also measured at KFK.

Iron atom density 0.0844E+24 atoms/cm^3 and elemental composition are taken from Report KFK-2219 [7]. Isotope fractions were computed from elemental composition employing the MCNP utility *mattool* [16].

Table 4.1. Material composition and elemental fractions of the Cf source components presented in Report KFK-2219 [7]. *The atom fractions calculated by us are highlighted by italic font.*

Cf-source component	Matrix with Cf	Inner Capsule	Outer Capsule		
Material	SiO_2 / MgO	Zr+1.5wt%Sn	Al+3.0wt%Mg		
Weight, g	0.1400	4.5500	4.8700		
Volume, cc	0.1131	0.6723	1.8414		
Mass density, g/cc	1.2379	6.7678	2.6447		
Atom dens., 10^{+24} a/cc	0.03710	0.04452	0.05923		
	Atom fractions, 10^{+24} atoms/cc			**Sum**	KFK-2219 [7]
O, 10^{+24} a/cc	*0.022262*			***0.022262***	**0.002600**
Mg, 10^{+24} a/cc	*0.007421*		*0.001966*	***0.009387***	**0.003900**
Al, 10^{+24} a/cc			*0.057264*	***0.057264***	**0.105500**
Si, 10^{+24} a/cc	*0.007421*			***0.007421***	**0.000800**
Zr, 10^{+24} a/cc		*0.044009*		***0.044009***	**0.029400**
Sn, 10^{+24} a/cc		*0.000515*		***0.000515***	**0.000400**
Sum of Atoms	***0.037103***	***0.044524***	***0.059230***	***0.140857***	**0.142600**

Photon and neutron tallies. The MCNP input deck simulates ^{252}Cf(s.f.) as a source of both neutrons and γ-rays. This allows us to perform the validation of neutron and gamma leakage spectra in one run with single MCNP input file. Because of such option, both neutron and photon tallies were included in the input deck for iron spheres.

Two photon tallies are available there to score the absolute energy flux of the γ-rays leaking:

1) from bare Cf-source - point detector (tally 105) located in the air at distance 102.5 cm (the iron sphere, i.e. cell 6, has to be voided for that);
2) from the outer surface of the iron sphere – track length averaged flux (tally 104) in the spherical layer (cell 52) of 0.5 cm thickness located at 1.55 cm above outer sphere radius R. For this the spherical surfaces 51 (Si(Li) front plane) and 52 (its rare plane) has to be settled by user at radii R + 1.55 ± 0.25 cm. Additional segmentation of this tally by cylinder 53 of radius 0.059 cm is also possible, if user wishes to model absolutely exact the sensitive volume of the Si(Li) detector (our check has shown the acceptable agreement with the spectrum averaged over the whole spherical cell 52).

If the user wishes to compute neutron leakage spectra then he has to address to the point detector tallies 5 and 15 (proton recoil gas proportional detectors D1, D2 and D3) at distance 108 cm or tally 4 (^{3}He-Si detector) close to iron sphere surface, for more details see [7, 8]. It is worse to note that in the case of the thickest (minimum leakage) KFK Fe sphere of diameter 40 cm the 15 cm long PVC shadow bar was shown to be slightly transparent for the fast neutrons [34]. Accounting for such shadow bar in our MCNP simulations results to (1 – 8)% decreasing of C/E for neutron leakage in interval (0.05 – 5.0) MeV.

The results of the MCNP calculations, i.e. the neutron and γ-rays tallies, are the particle spectral yields in whole solid angle 4π. The normalization is per one total neutron from spontaneous fission of ^{252}Cf, i.e. per ν(tot) = 3.7676 neutrons.

The MCNP perturbation card allows estimation of the sensitivities to the nuclear reaction cross sections and to other parameters of the benchmark. The input deck contains an example of perturbation cards to compute sensitivities to the partial neutron cross sections with a help of 10% variation of the iron shell density.

Other details are available in the MCNP input deck as self-explaining comments.

5. Sample validation of ENDF/B-VIII.0 against the γ-ray spectra from KFK (and IPPE) iron spheres with a Cf-source

^{252}Cf encapsulated source. The calculated absolute energy distribution of γ-rays from the bare KFK Cf-source is compared with experimental one in Fig. 5.1 (the "KFK fit" designates the fit and energy extrapolation used by KFK experiment authors for source representation in their discrete ordinate transport calculation [4]). Underestimation of our calculations below 0.5 MeV and above 2 MeV is in line with a warning given by the authors of the KFK experiment: "**quantitative interpretation of the measurements will be restricted to the energy region between 0.5 and 2 MeV**" [5]. This tendency of too high yield of γ-rays above 2 MeV is also confirmed, as seen in Fig. 5.1, by the γ-ray spectrum measured independently at IPPE up to 9.1 MeV [11].

The energy spectrum of γ-rays created by (n,xγ) reactions on materials of the matrix containing californium and two capsules was also calculated. It is seen that primary gammas from spontaneous fission of ^{252}Cf dominate by one-two orders of magnitude over the secondary ones created by neutrons in the Cf-source structure materials.

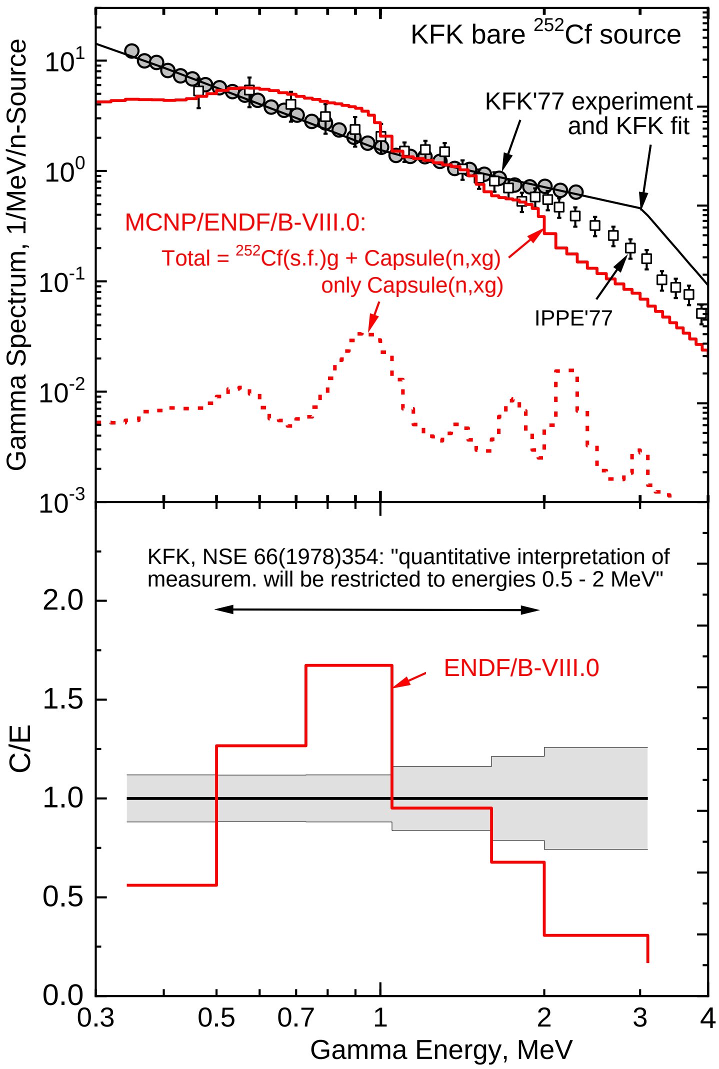


Fig. 5.1. γ-ray spectra from the bare Cf source (top): symbols - measurements at KFK and IPPE (for comparison); curves – KFK fit/extrapolation (black curve) and calculations by MCNP with ENDF/B-VIII.0: total (red solid curve) and contribution from (n,xγ) reactions on matrix and two capsules (red dashed curve). C/E ratios (bottom): integrated over the selected energy intervals - red histogram, grey area – experimental uncertainties.

The leakage γ-ray energy spectra were calculated by MCNP with cross section data from ENDF/B-VIII.0 [17] for three KFK iron spheres of outer diameters 25, 30 and 35 cm. The results are displayed and compared with measured data in Figs. 5.2 and 5.3. The spectra of the γ-rays originated in the bare Cf-source (i.e., primary gammas from ^{252}Cf(s.f.) and secondary ones from the neutron reaction on source structure materials) and transmitted through the Fe sphere are separately plotted. It is seen that their contribution decreases from ≈ (50 – 70)% to ≈ (20 – 30)% when the iron shell thickness increases from 12.5 to 17.5 cm. It means that in the case of the rather thick iron layer (≥ 20 cm) the Fe(n,xγ) reactions will make the dominant contribution to the transmitted γ-rays, whereas the direct γ-rays from the bare ^{252}Cf source will be substantially attenuated.

The validation of the evaluated transport data is usually performed in terms of Calculation over Experiment (C/E) ratios. The bottom parts of Figs. 5.2 and 5.3 plot the C/E ratios after integration of the experimental and calculated γ-ray leakage spectra over rather broad energy intervals. The integration boundaries were selected to capture the specific structures (such as peaks, change of the energy trend, etc.) observed in the experimental energy distributions.

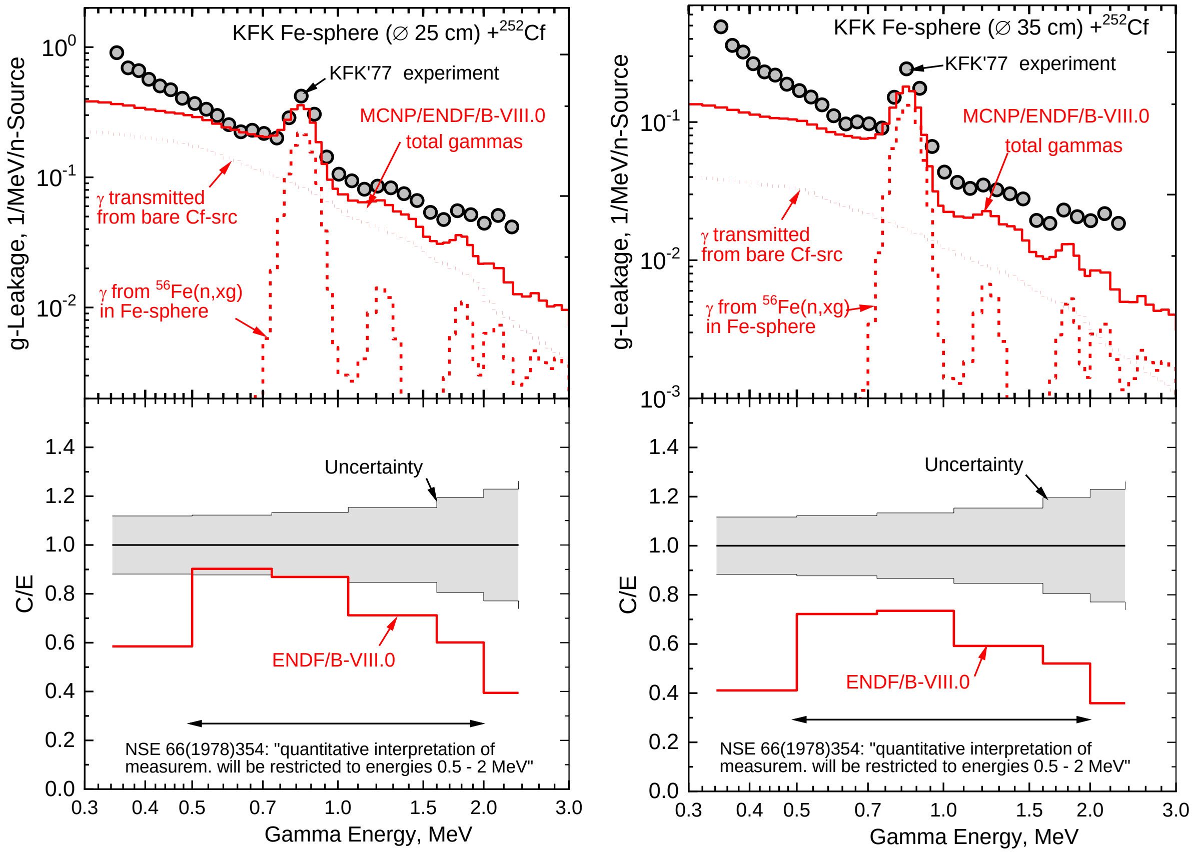


Fig. 5.2. Comparison of the spectra of γ-rays leaking from Fe spheres of diameter 25 and 35 cm fed by the ^{252}Cf source in the center. Symbols – KFK measurements. Curves – MCNP transport calculations with ENDF/B-VIII.0: primary γ-rays born in the bare Cf source and transmitted through Fe sphere (pointed curve), secondary γ-rays produced by the Fe(n,xγ) reaction in iron sphere (dashed), and total leakage (solid). Bottom parts plot the corresponding C/E ratios integrated in the energy intervals selected to capture the visible γ-ray spectra features.

As a conclusion from the KFK benchmark exercise we may conclude that ENDF/B-VIII.0 underestimates the γ-ray leakage by (20 – 40)% in the energy interval 0.5 – 2.0 MeV, which was recommended by the KFK experiment authors for the quantitative validation.

Since the iron spheres of the outer diameter 30 cm was also measured at IPPE, the cross-comparison of the validation conclusions derived from two independent experiments has interest. The results shown in Fig. 5.3 (right half-part) demonstrate the comparable (30 – 40)% underestimation when the ENDF/B-VIII.0 cross section data are used for n-γ transport in iron. It is worthwhile to notice that authors of IPPE experiment have also highlighted such underestimation of γ-ray yield [14].

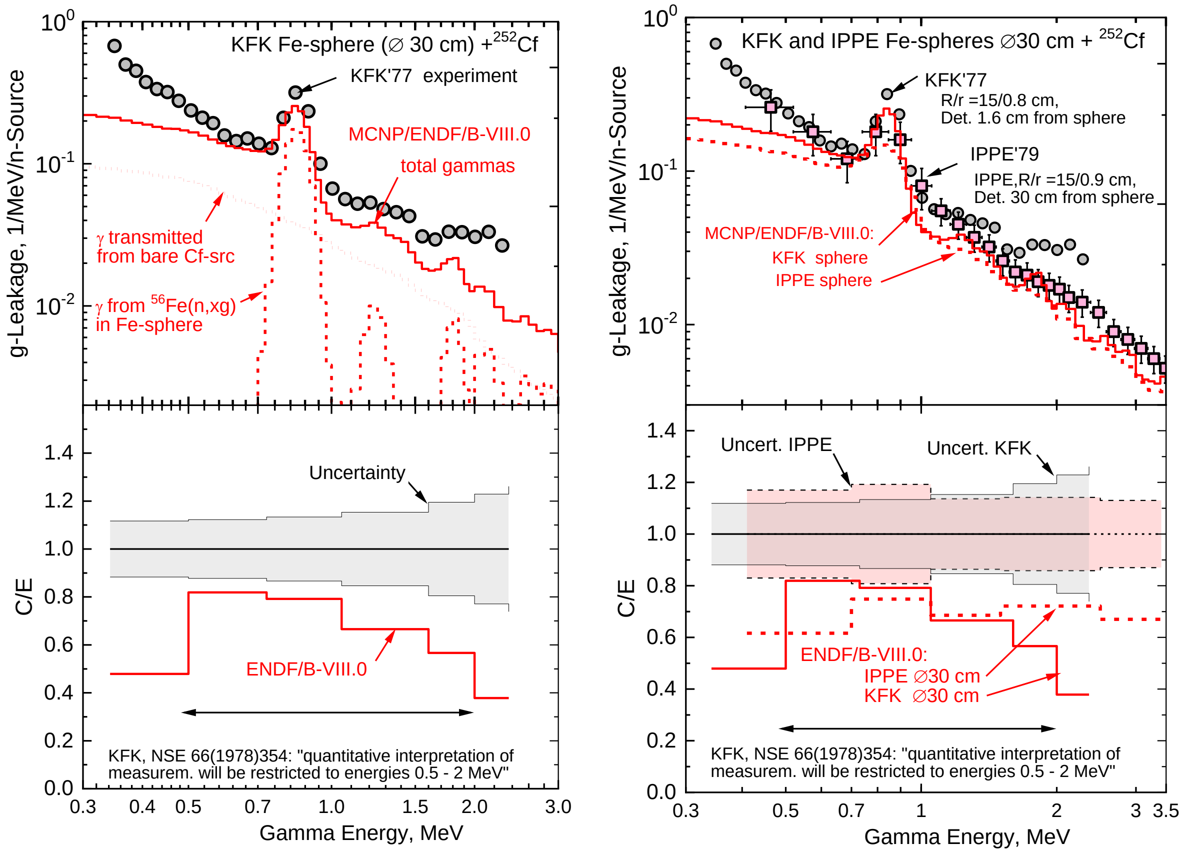


Fig. 5.3. The spectra of γ-rays leaking from the Ø30 cm Fe sphere fed by the ^{252}Cf source (left) and comparison with IPPE (right). Symbols – KFK and IPPE measurements. Curves – MCNP transport calculations with ENDF/B-VIII.0. Bottom parts plot corresponding C/E ratios integrated in the energy intervals selected to capture the γ-ray spectra features.

Fig. 5.4 depicts the calculated sensitivity of the γ-ray leakage spectra from the KFK iron sphere of Ø30 cm with a Cf-source to the variation of the neutron elastic, inelastic and capture cross sections on the materials of the sphere in the several energy intervals. Sensitivities were computed by MCNP employing the perturbation cards. The energy intervals were selected to capture the energy regions below the first excited level in ^{56}Fe, i.e. 0.847 MeV, and above.

It is observed that the γ-rays leakage increases when elastic scattering increases, however and only when neutron energy exceeds 0.85 MeV – threshold for reaction ^{56}Fe(n,n'γ). The inelastic collisions act practically similar but in the opposite direction. Sensitivity to the capture and to (n,2n) cross sections are several hundred times lower than to elastic and inelastic. This points to the small fraction of the thermal and high energy (threshold for ^{56}Fe(n,2nγ) is 11.399 MeV) neutrons in the system.

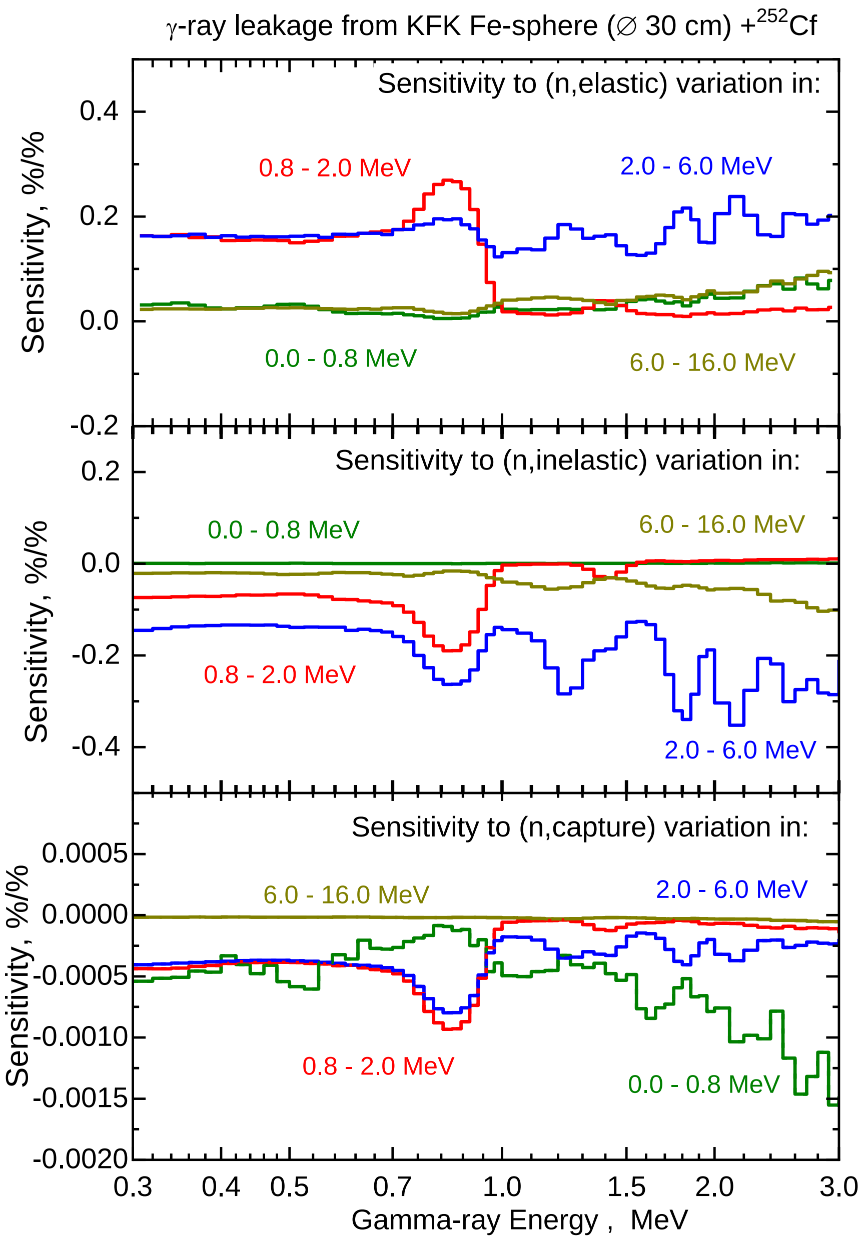


Fig. 5.4. Energy sensitivity of the γ-ray leaking from the KFK iron sphere of Ø30 cm with a ^{252}Cf source to the variation of the neutron elastic (top), inelastic (middle) and capture (bottom) cross sections on the materials of the sphere in the indicated energy intervals. Calculations are done by MCNP with ENDF/B-VIII.0.

Summary

The new entry for the SINBAD database was created. It comprises the KFK measurements of the γ-ray leakage spectra from three iron spheres of the outer diameter 25, 30 and 35 cm with spontaneously fissioning ^{252}Cf source in the center. The experiment was performed in 1977 and was an extension of the pervious benchmark where the neutron leakage spectra were measured. The latter is already available in the SINBAD database.

Following the SINBAD requirements, the new entry includes the short (abstract) and more detailed descriptions of facility, methods and final numerical results with uncertainties which are necessary to validate the evaluated nuclear data in this benchmark. To facilitate this task the MCNP input deck was created and included in this new SINBAD entry.

The specific (and new) feature of the proposed input deck is the modelling of ^{252}Cf(s.f.) as a source of neutrons and γ-rays, both from prompt and delayed emission. It allows to get gamma and neutron responses in one MCNP run. The modelling of ^{252}Cf(s.f.) as a source of neutrons is straightforward since the prompt neutron spectrum is a neutron standard and contribution of the delayed neutrons is relatively small.

The challenge in the ^{252}Cf(s.f.) source modelling is the energy distributions and multiplicities of the γ-rays. Fortunately the prompt gamma emission was already measured in dozens of experiments and also theoretically studied. This information was used in present work to assess the prompt γ-ray spectrum and multiplicity. The situation is turned to be much worse for the delayed γ-ray emission: we managed to find and use the relevant data produced only back in 1965. For the nuclear data validation and another californium applications it would be useful to establish the reference prompt and delayed γ-ray energy spectra and their multiplicities.

Assembled MCNP input deck was used for sampling radiation transport calculations with ENDF/B-VIII.0 neutron reaction cross section data. It was shown that this library underestimate the γ-ray yield within 0.5 – 2.0 MeV by 20 - 40%. The comparable underestimation was found in the similar IPPE benchmark carried out a few years later than KFK.

It turns out that KFK and IPPE has measured the γ-ray leakage spectra from iron sphere of the same outer diameter 30 cm. It gave us a unique option to compare directly (also and more precisely through C/E analysis) two independent measurements and confirm the agreement between them. This valuable fact increases the reliability of conclusions derived from validation analysis of both benchmarks.

Acknowledgment

One of the authors (S.S.) acknowledges Prof. S.-H. Jiang for providing the numerical data obtained in the KFK iron γ-ray leakage experiment, important details about measurements and review of this report.

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