

Spontaneous Fission Neutron Spectrum of $\text{Cf}^{252}\dagger$ A. B. SMITH AND P. R. FIELDS, *Argonne National Laboratory, Lemont, Illinois*

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The spontaneous fission neutron spectrum of Cf^{252} from 0.2 to 7.0 Mev has been measured. Time-of-flight techniques were employed to determine the lower energy portion of the spectrum while proton recoils in emulsions were used to study the higher energy neutrons. The measured neutron spectrum is, within the experimental accuracy, described by the empirical relation $N(E) \propto \exp[-0.88E(\text{Mev})] \sinh[2.0E(\text{Mev})]^\ddagger$, where $N(E)$ is the number of neutrons of energy E per unit energy interval. The experimental results are compared with the theoretically determined Cf^{252} fission neutron spectrum.

INTRODUCTION

KNOWLEDGE of the fission neutron spectrum is fundamental to most applications of a fission process. Despite this fact only the spectrum of U^{235} has been measured in detail.¹ The Pu^{239} , U^{233} , and Cf^{252} fission neutron spectra have been studied²⁻⁴ to varying degrees. In none of these measurements are the data reliable in the low-energy neutron range, and none of these measurements shows a clear maximum.

With such limited information available it was decided to carry out a careful measurement of the spontaneous fission neutron spectrum of Cf^{252} [$t_{1/2}$ (fission) = 66 years)]. This experiment allows accurate comparison of the spontaneous Cf^{252} fission spectrum with the spectrum of the neutron induced fission of U^{235} . These two spectra can be related to the existing theory^{5,6} and some preliminary conclusions formed regarding the systematics of fission neutron emission. From the practical point of view, it appears that Cf^{252} will soon be available in amounts large enough to form sizable fission neutron sources. A well-known fission neutron spectrum from such sources could be of considerable aid in critical studies.

EXPERIMENTAL PROCEDURE

The neutron distribution in the energy range 2-7 Mev was determined with proton recoil emulsions. From 200 kev to 3 Mev the neutron spectrum was measured with time-of-flight techniques.⁷ In using the latter method we have assumed that the "prompt"

neutrons are emitted within times $< 10^{-11}$ second after fission. This assumption has been verified experimentally.^{8,9} The Cf^{252} fission source, emitting 1.2×10^5 neutrons per minute, was volatilized onto a thin aluminum planchet and mounted within a gas scintillation cell.¹⁰ In the time-of-flight work this cell is used as the time marker for the fission event. The cell is of a type that has been used successfully at this laboratory for some time. The unit has a very fast rise time (in the order of 10^{-9} sec) and sufficient resolution to enable one to clearly distinguish the fission events from the alpha activity of the sample. For the measurements the bias of the counter was so chosen that the fission detection efficiency was 100%.

After leaving the fission source the neutrons traverse a flight path of 80 cm before striking the neutron detector. This detector consisted of a $1\frac{1}{8} \times 1\frac{1}{2}$ in. piece of Pilot B plastic scintillator¹¹ mounted on a RCA 6342 photomultiplier tube. During some of the runs this neutron detector was covered with $\frac{1}{4}$ in. of lead. However, this shielding was found to be unnecessary and was dispensed with throughout most of the work. Periodically the background was determined by inserting a hydrogenous scattering cone between the source and the detector. The experimental measurements were conducted in large rooms in order to reduce scattering effects to a minimum.

The time it takes a fission neutron to traverse the 80-cm flight path is between 30-150 μsec . This time interval was measured in two ways. In the first method the signal from both detectors was displayed on a single trace of a Tektronix oscilloscope¹² and photographed. The film was later projected and the spacing between "pips" on the trace measured. A parallel, wide-band circuit was arranged to trigger the oscilloscope sweep only for pairs of pulses coincident within the time interval of interest. The oscilloscope's sweep speed was calibrated against a crystal standard oscillator. The

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¹ L. Cranberg *et al.*, Phys. Rev. **103**, 662 (1956).

² N. Nereson *et al.*, Los Alamos Scientific Laboratory Report LA-1078 (unpublished).

³ K. Henry and M. Haydon, Applied Nuclear Physics Division Annual Report, September 10, 1956, Oak Ridge National Laboratory, ORNL-2081 (unpublished).

⁴ E. H. Hjalmar *et al.*, Arkiv Fysik **10**, 357 (1956).

⁵ R. B. Leachman and C. S. Kazek, Jr., Phys. Rev. **105**, 1511 (1957).

⁶ R. B. Leachman, *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1955* (United Nations, New York, 1956), Vol. 2, Paper P/592.

⁷ L. Cranberg, *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1955* (United Nations, New York, 1956), Vol. 2, Paper P/577.

⁸ J. S. Fraser, Phys. Rev. **88**, 536 (1952).

⁹ Smith, Friedman, and Fields, Phys. Rev. **102**, 813 (1956).

¹⁰ C. Egger and C. Huddleston, Nucleonics **14**, No. 4 (1956).

¹¹ Pilot Chemical Company, Waltham, Massachusetts.

¹² Model 517, Tektronix Inc., Portland, Oregon.

time resolution of this method, measured as the full width of the prompt-gamma peak at half-maximum, is 5–7 μsec . In all, more than 20 000 traces were measured.

The preceding technique is satisfactory but tedious. For this reason a time to pulse-height converter was constructed. This unit linearly transformed time intervals in the range 0–120 μsec into voltage pulses which were sorted in a 256-channel pulse-height analyzer. Upon using the same criteria as above, the time resolution of this system was 2–3 μsec . Because of its ease of operation and its accuracy, this unit was used for most of the experimental measurements. All of the time-of-flight data were corrected for the energy dependence of the neutron detector's efficiency. This efficiency was determined by comparing the response of the scintillator to the response of a flat "long" counter in a monoenergetic neutron beam from the $\text{Li}(p,n)$ reaction.

For the proton recoil method, Ilford C-2, 400-micron emulsions were exposed to the Cf^{252} source in such a manner that the neutrons entered the emulsions at an angle of 5° – 10° with the emulsion surface. Furthermore the 1 in. \times 3 in. emulsion plates were arranged so that the neutrons made in the region scanned an angle of 10° or less with the longitudinal plate axis. The tracks were measured in swaths 7.5 mm long, starting 5 mm from the leading edge of the plates and extending no more than 2 mm from the longitudinal axis. The plates were processed by the temperature development meth-

od¹³ and treated with wood resin to reduce shrinkage. A Bausch & Lomb research microscope, fitted with a Leitz G.F. 10 \times eyepiece and a 53 \times Leitz oil-immersion objective, was used in the measurements. About 1400 tracks were measured, the work being divided equally between two scanners. Good observer agreement was obtained. The tracks accepted for measurement fell within a square prism whose axis lay along the longitudinal plate axis and whose half-angle was 20° . Only those tracks having, in the unprocessed emulsion, a projected length along the prism axis of 15 microns or more were measured. Both ends of a track had to terminate at least two microns from the emulsion surfaces. Corrections for the probability of escape were

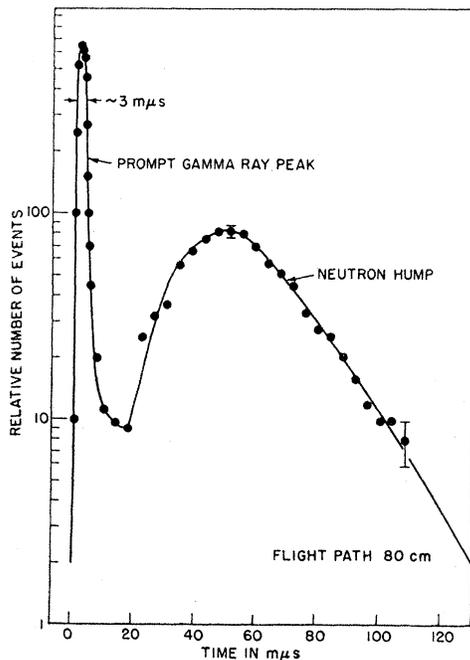


FIG. 1. The distribution in time of Cf^{252} fission neutrons as measured over a flight path of 80 cm.

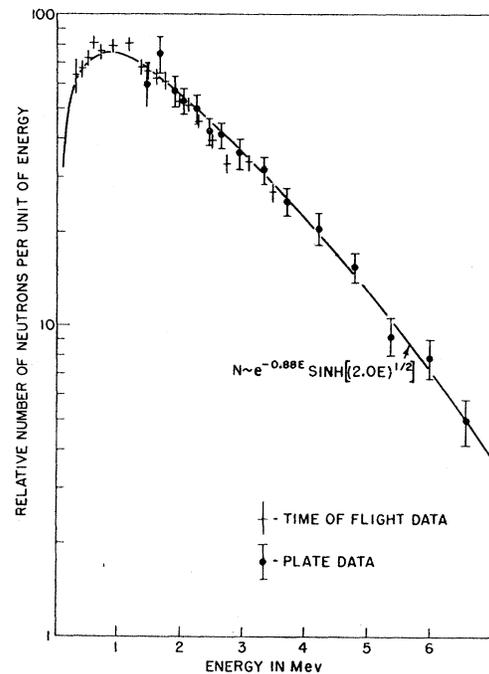


FIG. 2. Experimentally determined energy spectrum of Cf^{252} fission neutrons.

made using the empirical factors obtained at the Los Alamos Laboratory.¹⁴ The tracks were grouped into 0.2-Mev intervals according to the average value of the $\cos^2\theta$, where θ is the neutron-proton angle in the laboratory system.

RESULTS AND CONCLUSIONS

Eight spectral measurements were carried out with the time-of-flight techniques. A typical experimental curve is shown in Fig. 1. The prompt gamma-ray peak is clearly defined followed by the broad neutron "hump." Figure 2 shows one of the time-of-flight measurements converted to the energy scale and cor-

¹³ Dilworth, Occhialini, and Payne, *Nature* 162, 102 (1948).

¹⁴ L. Rosen, *Nucleonics* 11, No. 7, 32 (1953).

rected for the neutron detection efficiency of the plastic scintillator. Also shown in Fig. 2 are the results of the proton recoil emulsion measurements normalized to the time-of-flight data. The fission neutron spectrum of Cf^{252} is qualitatively like that of U^{235} . It is well known that the latter is described by the empirical expression,^{15,1}

$$N(E) \propto e^{-bE} \sinh[(cE)^{\frac{1}{2}}],$$

where $N(E)$ is the number of neutrons of energy E per unit energy (E measured in Mev), b is 1.036/MeV, and c is 2.29/MeV. The same empirical expression was fitted to the Cf^{252} data from this experiment. Excellent agreement with the measured values was obtained (see Fig. 2) with $b = (0.88 \pm 0.05)/\text{MeV}$ and $c = (2.0 \pm 0.2)/\text{MeV}$. This empirical distribution for Cf^{252} is compared in Fig. 3 with the theoretical calculations of Leachman⁹ and the experimentally determined fission neutron spectrum of U^{235} .¹ From Fig. 3 and from a comparison of the respective constants in the above empirical expression, it is evident that the Cf^{252} fission neutron spectrum is more energetic than that of U^{235} . Leachman's theoretical spectrum is in qualitative agreement with experiment, but quantitatively lacking in lower energy neutrons. The results of Hjalmar *et al.*⁴ are compatible with the present work over the limited energy range of their measurement.

We have attempted to interpret our results on the basis of the continuum model of the nucleus,¹⁶⁻¹⁸ realizing that such an approach is a first approximation only. Initially we assume that the fission fragments of Cf^{252} emit, on the average, two neutrons, per fission per fragment¹⁹ and after emission still retain enough energy to be described as being in a continuum of energy states. Under these conditions the neutron emission can be treated as a double "boiloff."¹⁸ In addition to the continuum premise the following are assumed to be true:

1. Neutron emission occurs from the fragments after fission.
2. Neutron emission is isotropic in the fragment space.
3. All fission occurs from the most probable mode.
4. The light and heavy fragments are equally excited.
5. Considering all fission processes, the ratio of the neutron emission from the light to that from the heavy fragment is a linear function of the most probable fission mass ratio.

By using continuum theory and the known kinetic energies of Cf^{252} fission fragments, the fission neutron

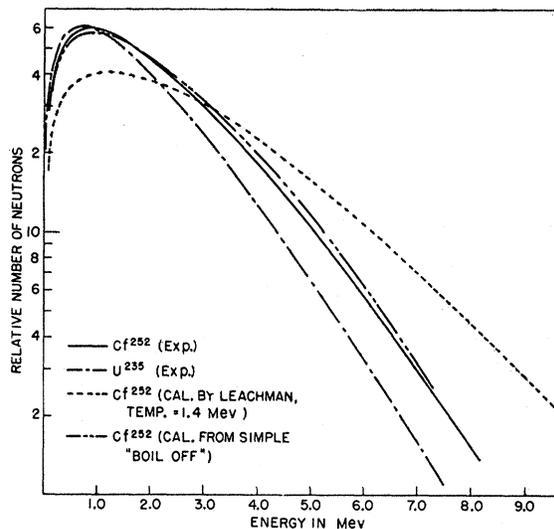


FIG. 3. The fission neutron spectrum of Cf^{252} compared with theory and with the experimentally determined U^{235} fission neutron spectrum.

spectrum was calculated for a wide range of fragment excitation energies, temperatures, and neutron binding energies. The best fit with experiment was obtained with a nuclear temperature of 1.0 Mev, a fragment excitation energy of 10 Mev, and a neutron binding energy of 4.5 Mev. As evident from Fig. 3, the agreement with experimental data is remarkably good in view of the relatively coarse assumptions employed. A similar interpretation of the U^{235} spectrum by Fraser¹⁷ also gives good agreement with experiment although the lower value of ν in the case of uranium ($\nu \sim 1.2$ neutrons per fragment per fission) makes the double neutron emission concept less valid than for Cf^{252} .

The greater average fission neutron energy of Cf^{252} , as compared to that of U^{235} , is partly attributable to the higher kinetic energy of the fission fragments.⁹ Also the excitation energies of the fission fragments from Cf^{252} are probably somewhat greater than those for U^{235} as evidenced by the higher value of ν for Cf. This greater excitation energy would lead to more energetic neutron emission. Before a detailed interpretation of the phenomena can be made, much more information about the level structure and mass of the neutron rich fission fragments must be available. Until then only an empirical approach is possible. This experiment shows that the fission neutron spectrum of Cf^{252} is, for most practical applications, essentially identical to that of U^{235} .

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¹⁵ B. Watt, Phys. Rev. **87**, 1037 (1952).

¹⁶ V. Weisskopf, Phys. Rev. **52**, 295 (1937).

¹⁷ J. S. Fraser, Phys. Rev. **88**, 536 (1952).

¹⁸ B. T. Feld *et al.*, U. S. Atomic Energy Commission Report, NYO-636 (unpublished).

¹⁹ D. A. Hicks *et al.*, Phys. Rev. **97**, 564 (1955).