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ABSOLUTE MEASUREMENTS OF THE ^{235}U AND ^{238}U FISSION CROSS-SECTIONS IN THE ^{252}Cf FISSION NEUTRON SPECTRUM

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ABSTRACT

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Absolute measurements have been made of the fission cross-sections of ^{235}U and ^{238}U in the ^{252}Cf fission neutron spectrum, using the method of coincidences between fission events in a target made of the isotope studied, and the associated fission fragments of californium. A detailed description of the measurement method is provided, covering the reasons for choosing the geometrical conditions of the experiment, the calculations made for that purpose, the preparation of targets, the detection apparatus and sources of errors. ^{235}U and ^{238}U fission cross-sections in the ^{252}Cf fission neutron spectrum are calculated from differential fission cross-section measurements.

INTRODUCTION

At the present time, the growth of nuclear power - and especially the development of fast neutron reactors - has meant that the requirements of accuracy of fission cross-section measurements of different heavy elements are considerably more stringent. Absolute fission cross-section measurements of heavy elements are used in reactor calculations and also as reference data for relative fission cross-section measurements.

The main difficulty arising when measuring neutron fission cross-sections is the accuracy with which the neutron flux is determined. In addition, it is possible that the energy spectrum of neutrons will be distorted by scattering and moderation in structural materials. This factor is also extremely important, since fission cross-sections are sensitive to the energies of the neutrons causing fission.

In our absolute measurements of the ^{235}U and ^{238}U fission cross-sections averaged over the ^{252}Cf fission spectrum we used the method of coincidences between fission events in the sample being measured and in ^{252}Cf. For this purpose we employed the results of sufficiently accurate measurements of $\bar{\nu}$ for ^{252}Cf from Ref. [1], the influence of scattered neutrons being practically excluded (provided that the resolving time of the coincidence circuit is sufficiently short). Moreover, weighing of the ^{252}Cf fission source becomes unnecessary and emphasis can be placed on the following basic requirements:

1. Ensuring that the geometrical conditions for the experiment are very precise and are convenient for calculation;
2. Constructing a device with a minimum amount of scattering material near the ^{252}Cf source and the target of fissionable material;
3. Precise weighing of the targets of the fissionable element and ensuring that the thickness of the fissionable element foil is even over the whole target area;
4. Achieving maximum fission fragment detection efficiency;
5. Protecting the device from electrical interference, since if the first four conditions are satisfied the coincidence counting rate will be, at best, a few events per minute.

THE GEOMETRICAL CONDITIONS FOR THE EXPERIMENT

The most convenient experimental geometry for the calculations would be one in which a ^{252}Cf point source irradiated a target in the form of a segment of a spherical surface, onto the outside of which a layer (foil) of the isotope being studied had been deposited, with the ^{252}Cf source being placed in the centre of this sphere. This type of geometry, however, imposes conditions on the technique for deposition and weighing of the isotope studied which are difficult to meet. Hence the basic geometry chosen was one in which a "point" source of ^{252}Cf irradiates with neutrons a flat isotope target. This arrangement of source and target permits the geometrical conditions of the experiment to be calculated fairly accurately (Fig. 1).

Preliminary estimated have shown that in order to achieve the accuracy required for determining the fission cross-sections (1.5-2%), the geometrical conditions will have to be very stringent, as follows:

(a) The distance between the fissionable isotope foil and the ^{252}Cf source, as well as the radius of the fissionable foil, must be known to within a few microns; since in real conditions the ^{252}Cf neutron fission source will still have finite dimensions, the foils of californium and fissionable material must be parallel; and the purity with which the backing surfaces are manufactured must be of the 12th class;

(b) The thickness of the ^{252}Cf source backings and the isotope target should not exceed 400 μm;

(c) We need to know the distribution law for the density of the fissionable material over the target; if the material is fairly evenly distributed (with an unevenness of less than 1%) calculation of the geometrical conditions will be considerably easier.

In our preliminary measurements of fission cross-sections [3] the ^{252}Cf source and the isotope target were placed with their backings touching each other. As a result there was a high scattering probability for the neutrons moving from the fission source to the edges of the isotope target.

In order to reduce the proportion of these neutrons we have to place the backings with the californium and isotope at a certain distance from each other. This distance must of course be increased within reasonable limits in such a way that the counting efficiency in the channel which is detecting

fission of the isotopes studied is good enough for collection of the statistics required. For this, the accuracy with which the distances between the backing surfaces is fixed must lie within 10-15 μm and they must be parallel to within 20-30'.

We designed a device for fixing the position of the source and target which, for the sake of simplicity, will be called the "assembly" from here on (Fig. 2). Two versions of the upper part of the assembly were made by which it was possible to measure with two different distances between the backings. A number of fission cross-sections were measured with this assembly. However, an evaluation of the number of fission events in the isotope studied caused by neutrons scattered in the material of the assembly showed that the figure amounted to some 2-3% of the total number of fission events; furthermore, the accuracy with which this correction could be calculated was insufficient for the overall measurement accuracy of 1.5-2%. For this reason we constructed another assembly containing almost two orders of magnitude less structural material than its predecessor. This assembly is shown in Fig. 3. In this case, the accuracy with which the position of source and target was fixed was almost the same as before ($\pm$ 15 μm).

After numerous trials, we developed a technique for making the backings from 1X18HIOT stainless steel. Blanks cut out of steel sheeting were worked on both sides with a grinding machine and the surfaces were then brought up to the required degree of workmanship (∇ 12) by hand. During this finishing process the plane-parallelism of the backings was regularly checked with a Type 201 profilograph-profilometer. The thickness of the backings was measured with an optimeter and the surface finish with an interferometer. The sides of the backings were maintained plane-parallel to within $\pm$ 4 μm. Given a backing thickness of 30 μm, the correction for neutron scattering is 3%.

PREPARATION AND WEIGHING OF THE TARGETS

In order to prepare targets of fissionable material with high uniformity of the foil, we designed and constructed a device by means of high-frequency plasma technique for spraying material onto a rotating backing while it was cooling. One of the advantages of this technique is that losses of material during spraying are small. Secondly, in its final form this method can be used to produce isotope foils up to 1 mg/cm^2 thick with a foil thickness which is uniform to within 1%.

The isotope targets obtained by high-frequency spraying were visually completely uniform; the colour of the thin isotope layer was the same over the whole target area (since the foils are sufficiently thin, the presence of an interference pattern would mean that the foil had been deposited unevenly over the radius or the azimuth). With a view to quantitative determination of the degree of unevenness of the targets produced we used a technique for scanning them with a surface-barrier semi-conductor detector closed off by a thick diaphragm with an aperture 0.5-2 mm in diameter. The measurements showed that the targets we produced have a non-uniformity of less than 1%.

The targets were weighed by measuring their alpha activity in 2π geometry in a mesh ionization pulse chamber (^{238}U) and a semi-conductor detector in a small solid angle (^{235}U).

In our measurements we tried to use isotopes of high separation purity. Although the isotopes investigated were supplied with guaranteed specifications, in all cases we performed isotopic analyses with an alpha spectrometer containing a silicon surface-barrier detector and AI-1024 and NTA 512B analysers in a chamber with a small solid angle. The energy resolution of the spectrometer was 27 keV. In cases in which the sample studied contained impurity isotopes indistinguishable from each other by the alpha spectrometric method, mass spectrometric analysis was used (for ^{235}U). The results of the isotopic analyses we performed enabled the accuracy with which the basic isotope of the target was weighed to be increased and a correct estimate to be made of the contribution to fission events from associated isotopes.

The ^{252}Cf fission neutron source was prepared by thermoscattering of californium. The diameter of the ^{252}Cf foil was 3.0 $\pm$ 0.2 mm.

APPARATUS

Figure 4 shows a block diagram of the device we used. The fission fragment detector for the isotope studied and ^{252}Cf was a double ionization chamber with depths of 1 mm for californium fission fragments, and 4 mm for fission fragments of the isotope studied.

In order to increase its time-resolving ability, the chambers operated in pulsed current mode. By this means it was possible to bring the pulse front up to 8-10 ns with a total pulse duration of some 80 ns for the chamber with the isotope studied, and 20 ns for the chamber with californium. Pulses from both halves of the chamber were amplified by fast current amplifiers and passed to integral discriminators and coincidence circuits.

To improve reliability the number of coincidences of ^{252}Cf fission fragments with fission fragments of the element studied was recorded three times. Each coincidence circuit was a conventional fast-slow coincidence circuit with τ = 20 ns. In addition to these channels, a channel for time analysis of coincidences over the time range 0-100 ns was introduced. For this analysis we used a VAK-100 time-amplitude converter operating on the "start-stop" principle, and an AI-1024 analyser. The start signal was the signal from the chamber with the isotope being studied, while the stop signal was the one from the californium chamber after an initial time lag of 60 ns. When processing the results, there was reciprocal monitoring of the operation of the time analysis channel and the coincidence circuits.

Determination of the slope in the plateau of the chamber counting characteristic by integral discriminators [3] was duplicated by an amplitude analysis of the pulse spectrum of the isotope studied using an AI-1024 multichannel analyser. The amplitude analysis made possible a more accurate extrapolation of the pulse spectrum from fragments in the low-energy region.

FISSION CROSS-SECTION CALCULATION

The ^{235}U and ^{238}U fission cross-sections in spontaneous ^{252}Cf fission neutrons were found from the expression

$$\sigma_f = \frac{2\pi A R^2 N_f}{N_0 P \bar{\nu} G K_\tau N_{Cf}}, \quad (1)$$

where σ_1 is the fission cross-section of the isotope studied;

A is the atomic weight of the isotope studied;

R is the radius of the active foil target;

N_f is the number of fission events for the isotope studied;

N_o is the Avogadro number;

P is the weight of the fissionable material in the target;

$\bar{\nu}$ is the average number of prompt neutrons per spontaneous ^{252}Cf fission event;

G is the geometrical factor;

K_τ is the correction for counting errors in the ^{252}Cf fission fragment counting channel;

N_{Cf} is the number of ^{252}Cf fission events.

For the calculations based on Eq. (1) a correction was introduced for fission of impurities in the target studied. For this, use was made of the data from the isotopic analysis performed. The value of K_τ was determined from the expression

$$K_\tau = \frac{1}{1 - \tau N_{Cf}},$$

where τ was taken to be equal to the resolving time of the coincidence circuit (20 ns).

For the conditions of our experiment we obtained the following expression describing the geometrical factor (see Annex):

$$G = I_1 + I_2 + I_3, \quad (2)$$

where I_1 is the basic term for fission by neutrons which have not been scattered in the backing material when the ^{252}Cf source is a "point" source;

I_2 is the correction term for the finite dimensions of the source;

I_3 is the correction term for fission events caused by neutrons scattered in the backing material.

ANALYSIS OF EXPERIMENTAL ERRORS

Table 1 shows relative errors in all values in Eq. (1). The value $\bar{\nu}$ for ^{252}Cf after subtraction of delayed neutrons [2] was taken from Ref. [1]. The error in determination of the geometrical factor G was found by numerical calculation on a computer by variation of the parameters in Eq. (2). For example, the parameter H (the distance between the foil of fissionable isotope and the ^{252}Cf source) in the equation for G was known to within $\pm$ 15 μm (as measured by the optical method). The corresponding values for the parameter H - the minimum (H-ΔH) and maximum (H+ΔH) - were fed into the computer, the values of G were calculated with them, and then the relative errors were determined (with respect to the basic value of G corresponding to the basic value of H). Next, the maximum was chosen from the errors obtained since the dependence of G on the parameters included in it is not linear. All the parameters in the equation for the geometrical factor were treated in this way. The total relative error in the factor G was found by quadratic addition of the errors occurring in variation of all the parameters in Eq. (2).

When weighing the ^{235}U in the device with a small solid angle, all the necessary dimensions and distances were measured by the optical method to within a few microns. The error in calculation of the solid angle was in this case 0.3%. Experimental verification with a ^{241}Am standard calibrated by the alpha-X-ray coincidence method to within 0.1% confirmed the correctness of the solid angle calculation.

When measuring the alpha activity of ^{238}U in 2π geometry, we introduced corrections for the absorption of alpha particles in the isotope foil and for scattering of alpha particles from the backing, with the corresponding errors being taken into account [4].

For the transition to weights the alpha half-lives of ^{235}U and ^{238}U from Ref. [5] were used:

$$T_{\alpha\frac{1}{2}}(^{235}U) = (7.0381 \pm 0.0048) \times 10^{8} \text{ a};$$

$$T_{\alpha\frac{1}{2}}(^{238}U) = (4.4683 \pm 0.0029) \times 10^{9} \text{ a}.$$

When determining the error in the number of fission events of the isotope studied, we considered, in addition to the statistical error, the error in extrapolation to zero fragment energy. This operation was performed in respect of integral counting characteristics (linear extrapolation) and the amplitude spectrum

of fragments. Corrections were made for absorption of fission fragments in the foil of the isotope studied [4] in the form t/2R, where t is the thickness of the isotope foil in mg/cm^2 and R is the full path of the fragment in the foil (U_3O_8).

FINAL RESULTS AND DISCUSSION

The counting time for the number of fission events needed to achieve the same accuracy as we did in measuring one cross-section was from 240 to 400 h of round-the-clock measurements. Table 2 gives measurements of fission cross-sections and data obtained by other authors [6].

To compare our results with differential cross-section measurements, we made numerical calculations of the ^{235}U and ^{238}U fission cross-sections in the ^{252}Cf fission neutron spectrum. For the calculations, we used the data compiled on differential ^{235}U and ^{238}U fission cross-sections from Ref. [7]. The spectrum of ^{252}Cf fission neutrons was approximated by the Maxwellian distribution $n(E) = \sqrt{E}\exp-\frac{E}{T}$ with the parameter T = 1406 keV.

The fission cross-sections of ^{235}U and ^{238}U were calculated with the help of a computer. The values obtained are in good agreement with our experimental results. Here it should be borne in mind that the accuracy of the compiled differential cross-sections now available is lower than that of our own experimental fission cross-sections averaged over the fission spectrum. The calculations have also shown that the fission cross-section of ^{235}U hardly depends at all on the parameter T of the Maxwellian distribution when it varies by $\pm$10%.

The authors are grateful to B.M. Aleksandrov for preparing the ^{252}Cf sources and to K.G. Pochtarev for his extensive computer calculations.

A N N E X

CALCULATION OF THE GEOMETRICAL FACTOR G

The Monte Carlo method ought to have been used for calculating the geometrical factor G, but in order to achieve the required accuracy in calculation of this factor (to within 1%) the number of neutron "histories" counted for neutrons yielded by the californium source would have had to be so large that a very long computer time would have been needed.

We therefore adopted the method of approximated semi-analytical calculation of the geometrical factor. In so doing a number of very important assumptions were made:

1. The cross-section σ_{sf} used in subsequent calculations for the scattering of neutrons in the backing material was obtained by averaging partial scattering cross-sections over the fission neutron spectrum. Elastic scattering processes make the main contribution to this process, since inelastic scattering and the (n,p), (n,γ) etc. reactions play a fundamental role only for neutrons of the fission spectrum of which proportion in the spectrum itself is very small;

2. σ_{ss} - the scattering cross-section of neutrons scattered previously - was also obtained by averaging. When taking this process (double scattering) into account it was assumed that the doubly scattered neutrons simply "drop out of the game";

3. For the calculations it was assumed that the angular distribution of scattered neutrons is isotropic in the laboratory system. This assumption was verified by a calculation in which highly "exotic" forms of angular distribution of scattered neutrons were tested, without any significant effect on the final results.

The expressions for I_1, I_2 and I_3 are given below. With the assumptions described above the derivation of these equations is entirely valid. However, we do not show all calculations, but merely outline their derivations.

EXPRESSION FOR I_1

$$I_1 = \int_0^{\theta_0} \exp - \frac{n \sigma_{sf} (H_1 + H_2)}{\cos \theta} \operatorname{tg} \theta \, d\theta \,, \qquad (3)$$

where n is the number of nuclei in 1 cm^3 of backing material;

σ_{sf} is the scattering cross-section of ^{252}Cf fission neutrons in the backing material;

H_1 is the thickness of the ^{252}Cf backing;

H_2 is the thickness of the backing of the isotope studied;

$\theta_o = \text{arctg}\,\frac{R}{H}$, where R is the target radius of the isotope studied and H is the distance between the californium foil and the foil of the isotope studied.

In Eq. (3) the term $\exp - \frac{n\sigma_{sf}(H_1 + H_2)}{\cos\theta}$ relates to the scattering of neutrons in the backing material which enter the backing at an angle θ, thereby causing the scattered neutrons to drop out of the general flux. The multiplier $tg\theta$ is explained by the fact that, for a neutron entering the foil of fissionable material with thickness t at an angle θ, the foil thickness penetrated will be $\frac{t}{\cos\theta}$, and then if we take into account units of the solid angle $\sin\theta d\theta$ (integration over the angle ϕ has already been performed) we get $tg\theta d\theta$.

EXPRESSION FOR I_2

$$I_2 = \frac{2}{\pi a_0^2}\int_0^{a_0} a\,da\left\{\int_{\theta_1}^{\theta_2}\exp - \frac{n\sigma_{sf}(H_1+H_2)}{\cos\theta}\,tg\theta\,\arccos\frac{ctg\theta_1 ctg\theta_2 - ctg^2\theta}{ctg\theta(ctg\theta_1 - ctg\theta_2)}d\theta - \right.$$
$$\left. - \pi\int_{\theta_1}^{\theta_0}\exp - \frac{n\sigma_{sf}(H_1+H_2)}{\cos\theta}\,tg\theta\,d\theta\right\}. \qquad (4)$$

In this equation the notation is the same as in Eq. (3). In addition, a_o (the radius of the ^{252}Cf source) and $\theta_1 = \text{arctg}\,\frac{R-a}{H}$ and $\theta_2 = \text{arctg}\,\frac{R+a}{H}$ are introduced.

Equation (4) is obtained in a way essentially the same as in the previous case. However, when taking into account the "non-point" nature of the source, integration over the angle ϕ causes the term $\arccos\{f(\theta)\}$, which depends on the angle θ, to appear. The detailed calculations have been omitted here as being of little interest.

The limits of integration θ_1 and θ_2 in the first range are also due to asymmetry in integration over the angle ϕ (in the case in which the fission neutron point source is moved over a distance a from the centre of the californium backing).

The second integral (with limits from θ_1 to θ_o) appeared because the first integral (with limits from θ_1 to θ_2), which was later integrated over a, includes a part which has already been taken into account in I_1. It is easy to see that this part is the second integral.

EXPRESSION FOR I_3

$$I_3 = \int_0^{H_1} dx \left\{ \int_0^{\theta_x} \exp - \frac{n\sigma_{sf}x}{\cos\theta} \operatorname{tg}\theta d\theta \int_0^{\theta_{H-x}} \exp - \frac{n\sigma_{ss}(H_1+H_2-x)}{\cos\theta} \operatorname{tg}\theta d\theta \right\} +$$

$$+ \int_{H-H_2}^{H} dx \left\{ \int_0^{\theta_x} \exp - \frac{n\sigma_{sf}(x-H+H_1+H_2)}{\cos\theta} \operatorname{tg}\theta d\theta \int_0^{\theta_{H-x}} \exp - \frac{n\sigma_{ss}(H-x)}{\cos\theta} \operatorname{tg}\theta d\theta \right\}, \quad (5)$$

where $\theta_x = \operatorname{arctg} \frac{R}{x}$ and $\theta_{H-x} = \operatorname{arctg} \frac{R}{H-x}$.

The first term is obtained from fission of the isotope studied by neutrons which have been scattered in the californium backing. The first variable in this term is, so to speak, the "intensity" of the neutron source which has arisen as a result of their scattering over a distance x from the plane of the backing.

The second variable is similar to I_1 but in it σ_{sf} is replaced by σ_{ss}. It relates to double scattering of neutrons, which causes an attenuation of the flux from the above-mentioned "source".

The second variable in Eq. (5) is similar to the first but relates to fission of neutrons scattered in a backing on to which the isotope being studied has been deposited.

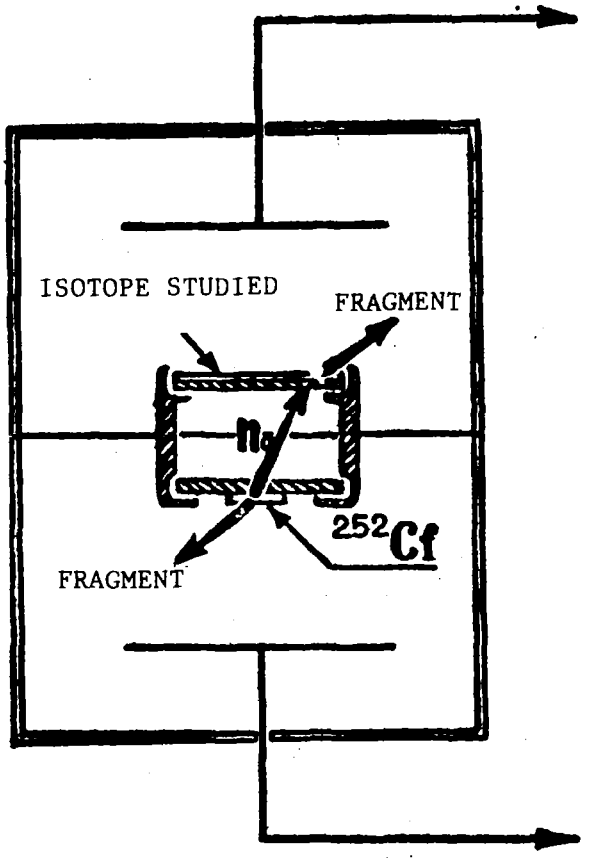

Fig. 1. Layout of experiment

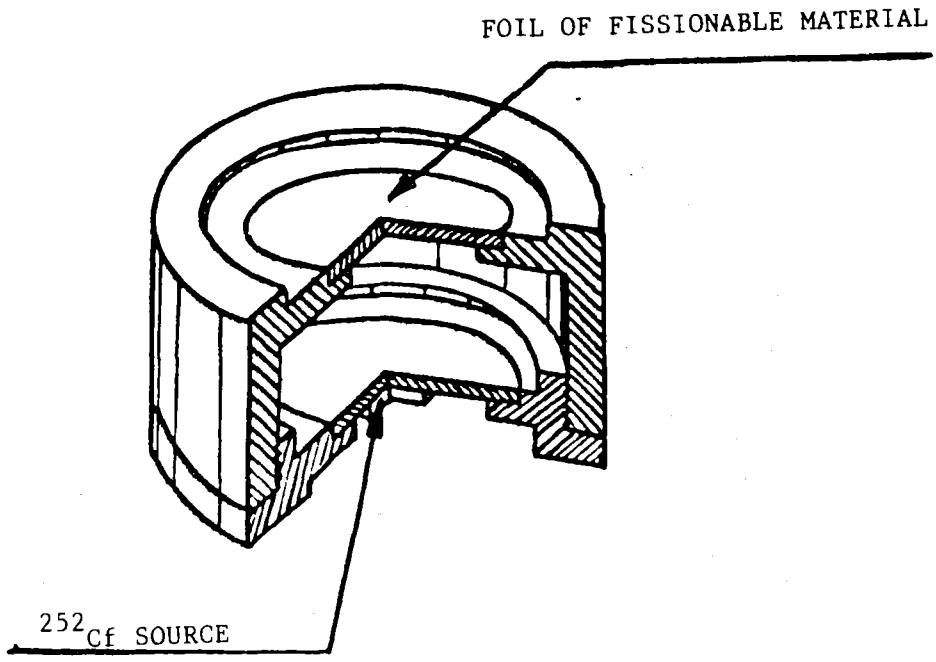

Fig. 2. First version of "assembly" containing ^{252}Cf source and target of fissionable isotope.

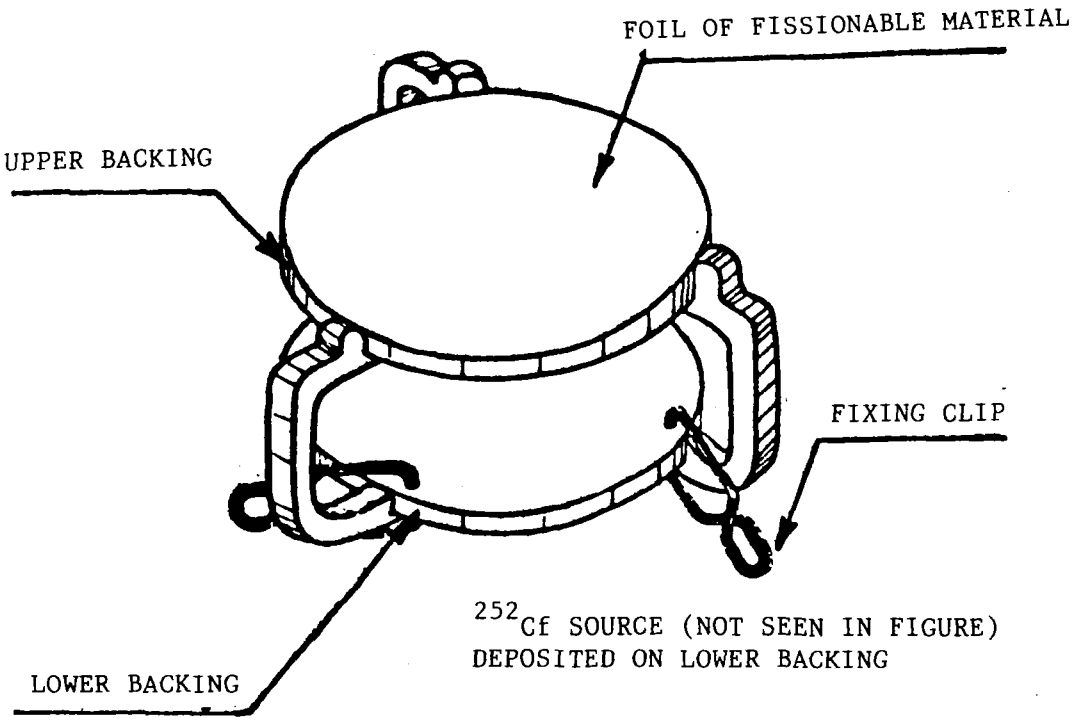

Fig. 3. Final verison of "assembly" containing ^{252}Cf source and target of fissionable isotope.

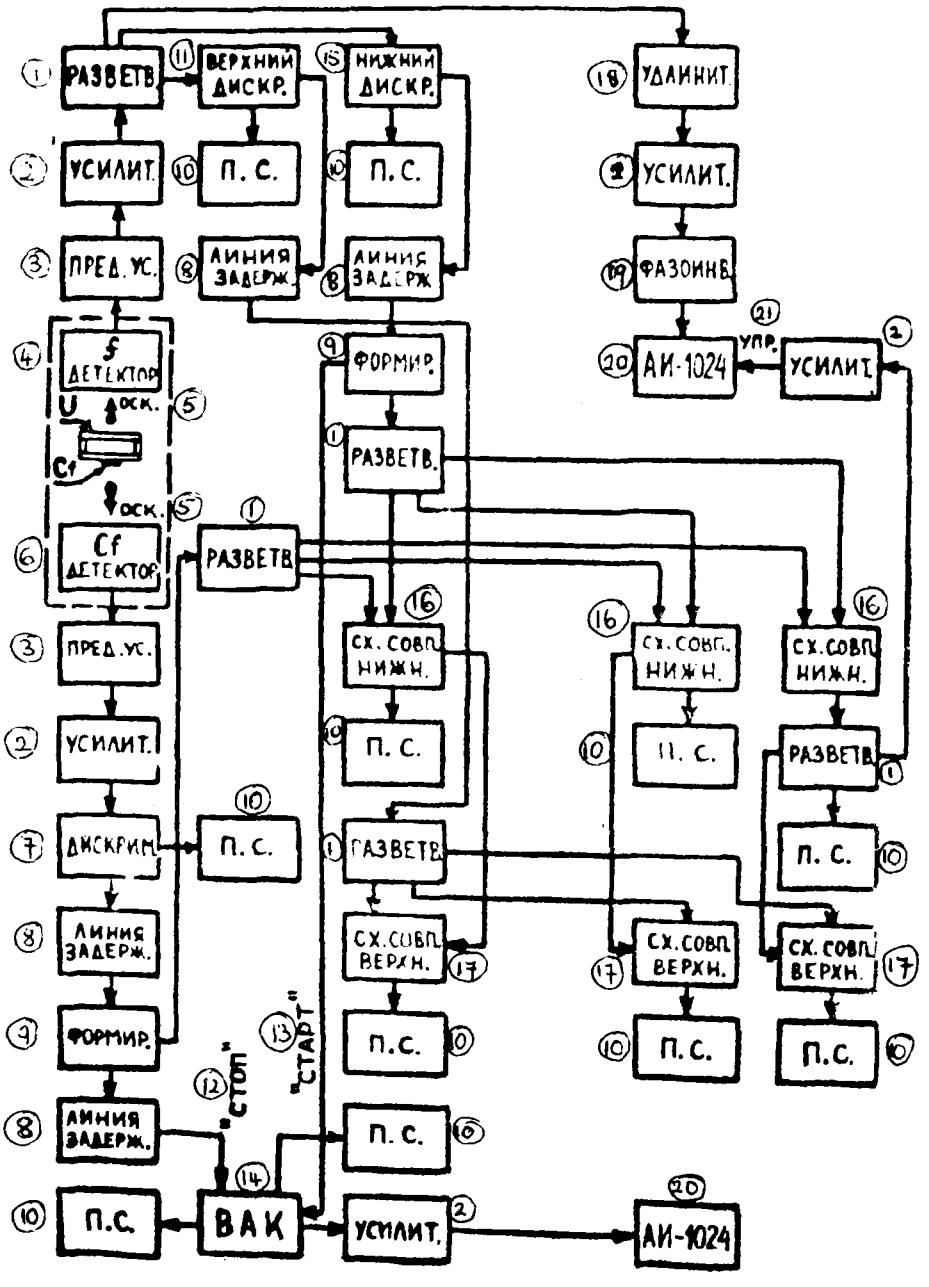

Fig. 4. Block diagram of electronic part of device for measuring fission cross-sections in the ^{252}Cf fission neutron spectrum

Key: 1. Branch; 2. Amplifier; 3. Preamplifier; 4. f detector; 5. Fragment; 6. Cf detector; 7. Discriminator; 8. Delay line; 9. Shaper; 10. PS [type of single-channel detector]; 11. Upper discriminator; 12. Stop; 13. Start; 14. VAK; 15. Lower discriminator; 16. Lower coincidence circuit; 17. Upper coincidence circuit; 18. Extension; 19. Phase inverter; 20. AI-1024; 21. Controlling

TABLE 1

Relative errors in parameters determining the ^{235}U and ^{238}U fission cross-sections, %

Error sources		^{235}U	^{238}U
$\bar{\nu}$ (^{252}Cf)		0.35	0.35
Geometrical factor G		0.71	0.71
Weighing in chamber with small solid angle	Statistical error	0.35	
	Proportion of isotope studied	0.41	
	Solid angle	0.3	
	Half-life	0.068	
Weighing in chamber with 2π geometry	Statistical error	-	0.55
	Proportion of isotope studied*/	-	0.30
	Extrapolation of α-particle spectrum to zero energy	-	0.51
	Correction for absorption in foil and scattering	-	0.30
Determination of number of fission events in the isotope studied	Statistical error	0.8	1.11
	Contribution of impurity fission	0.3	0.14
	Extrapolation of fission fragment spectrum to zero energy	0.40	0.37
	Correction for absorption in foil	0.3	0.25
ERROR IN CROSS-SECTION DETERMINATION		1.44	1.68

*/ The error in determination of the proportion of ^{238}U includes the error in measurement of the half-life of ^{238}U.

TABLE 2

^{235}U and ^{238}U fission cross-sections in the ^{252}Cf fission neutron spectrum, mb

Nucleus	Present paper		Ref.[6]
	Experiment	Theory	
^{235}U	1266 ± 19	1281	1207 ± 52
^{238}U	347 ± 6	352	324 ± 14

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