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BACKGROUND CONDITIONS FOR NEUTRINO EXPERIMENTS WITH THE ARTIFICIAL ^{147}Pm SOURCE

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BACKGROUND CONDITIONS FOR NEUTRINO EXPERIMENTS WITH THE ARTIFICIAL ^{147}Pm SOURCE: Preprint ITEP 2-97/

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In this article the background conditions of the experiments were conducted using the ^{147}Pm artificial antineutrino source are considered. Neutron and γ-background of this antineutrino source is defined by the impurities of long lived radionuclides (^{106}Ru, ^{144}Ce,· ^{152,154}Eu, ^{241}Am, ^{242}Cm and isotopes of others TPE). It is demonstrated that with the passive shield (25 cm of W) and neutron shield (15 cm of water with dissolved boron acid) the 5 MCi activity promethium source can be used in ν,e- scattering experiments to investigate of electromagnetic properties of neutrino.

Fig. - 2, ref. - 16.

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In a number of papers [1-5] the use of K-capture or β-decay isotopes of high activity for creation of the (anti)neutrino source aiming at the study of neutrino properties in ν,e-scattering reaction have been proposed. Expected rate of the useful events is $\lesssim$ 0.1 $day^{-1} \cdot keV^{-1} \cdot kg^{-1}$, and the most attractive area of detected energy ranges from ~ 3 keV to 30 keV. Therefore, these experiments would require low-threshold, low background big mass (over 100 kg) detectors placed in deep underground laboratories to eliminate the cosmic ray background [3-6]. Requirements of this kind are imposed on the detectors for registering Dark Matter (see detector of DAMA collaboration in ref. [4,7]).

Optimal geometry for these experiments would be if the detector is placed around the neutrino source ("4π-geometry") or other where the detector can view the source from a maximum angle. This condition stipulates minimum possible dimensions for the source. As those dimensions are almost entirely defined by the thickness of passive source shielding [3] it is important to evaluate the permissible level of long lived γ- and neutron emitting radioisotope contaminants in the material of the source.

Recovery of a suitable long lived isotope from the spent nuclear power plant (NPP) fuel fission products seems to be the promising way to obtain the working isotope for the artificial neutrino source (ANS) [3-6]. In this paper we will discuss the ANS of ~ 5 MCi activity based upon the long lived isotope of ^{147}Pm ($t_{1/2}$=2.62 y), the fission product produced with high yield of $2.7 \cdot 10^{15}$ Bk/ton of the spent WWER-440 composite fuel after 3 year cooling time. To create the ANS of the above said intensity about 70 ton of the spent NPP fuel should be reprocessed and about 5 kg of ^{147}Pm isotope recovered. The said quantity of ^{147}Pm in the Pm_2O_3 oxide form will occupy the volume of not more than 1 L.

1.1. Sources of γ-background.

The γ-flux (E_γ < 3 MeV) in the deep underground laboratory is ~ 1 $cm^{-2}s^{-1}$ [8] and the interaction of that much emission with a ~ 1 kg detector will yield the background count of ~10^6 day^{-1}. Hence, for experiments in ν,e-scattering the γ-flux must be reduced by a factor of over 10^7. Accordingly, the ANS's γ-flux must be suppressed to have the value of one on the ANS surface < 10^{-6} $cm^{-2}s^{-1}$ (with regard to the fact that the possibility of low energy yield one-fold due to Compton scattering interaction of a γ-ray in the big detector is known to be less than 10^{-3}).

Sources of γ-ray in the ANS in question are the following (see Tables 1 and 2):

- $^{146,148,148m}Pm$ isotopes;
- the long lived fission products of ^{60}Co, ^{137}Cs, $^{106}Ru(^{106}Rh)$, $^{144}Ce(^{144}Pr)$, $^{152,154}Eu$, etc.;
- γ-ray accompanying the spontaneous fission of plutonium and transplutonium elements (TPE), namely, Am, Cm, Cf.

Taking into account relatively short half life of the working isotope and the cooling time of spent fuel assemblies before shipment from NPP (> 3 y) the recovery of promethium isotopes is desirable to be performed immediately upon the delivery of spent fuel to the reprocessing plant, and duration time of the entire recovery cycle should not exceed 6 months. From the analysis of isotope composition of promethium and rare earth elements (REE, see Table 1 and 2) it follows that the γ-radiation of long lived ^{146}Pm isotope (E=0.747MeV and E=0.736 MeV with overall yield of 56%) represents the most dangerous and, moreover, irremovable by means of chemical decontamination procedures, high energy γ-ray source of the ANS under discussion.

From the fission product yield for ^{146}Pm (~$5 \cdot 10^{-9}$ activity ratio towards ^{147}Pm) minimum thickness of the passive shielding made of tungsten, the most effective of the readily available materials, is 20-22 cm. However, a number of data (see references in [3, 11]) point out the relative activity of ^{146}Pm to be 2-3 order of magnitude higher than conventional value, and if so, the necessary thickness of passive shielding for the promethium source of ~5 MCi activity appears to be 25 cm of tungsten.

With all the above said, the radiation "purity" of the source will be defined by the quality of the procedure used for chemical recovery (decontamination) of promethium from fission products, primarily from closely related REE isotopes of cerium and europium (see Table 2). The technology available in the Russian Federation allows to obtain the purity of promethium ~10^{-10} g/g [3,12]. High yield of dangerous long lived isotopes of ^{90}Sr, ^{106}Ru, ^{134,137}Cs, ^{144}Ce, and ^{152,154}Eu as compared to the yield for working ^{147}Pm isotope is also worth to mention. So far as γ-ray accompanying the spontaneous fission of Pu and TPE is concerned, the total activity of the ANS material under the conditions given will be negligible.

Considering the attenuation factor for tungsten shielding of d=25 cm, ^{147}Pm purification factor of 10^{-10} g/g, relative content of hazardous isotopes of Sr, Ce, Ru, Cs and others, as well as the yield for high energy γ-ray of the isotopes in question (see Table 1) we can conclude the thickness of tungsten calculated to suppress the γ-radiation of ^{146}Pm to be sufficient to suppress γ-radiation of all the other radioactive isotope contaminants. It should be noted that to reduce the γ-radiation of tungsten stipulated by the impurities of natural emitters from U and Th series the additional ~ 5 cm thick shielding made of MOb commercial brand copper will be required.

Summarizing the above said, in the most unfavorable case the thickness of shielding against the γ-radiation of the contaminants in the ^{147}Pm based ANS will not exceed 30 cm.

2. Neutron induced background.

The neutron flux in the deep underground laboratory is ~ 10^{-6} $cm^{-2}s^{-1}$ [8], and the expected rate of background events caused by the elastic neutron scattering on the nuclei of the detector substance will be ~ 20 $kg^{-1}\cdot day^{-1}$. Therefore, in low background experiments the neutron background is reduced, as a rule, by neutron shielding (and by discrimination of the neutron events according to the pulse shape) by 2 - 3 order of magnitude down to < 10^{-8} $cm^{-2}s^{-1}$. The neutron flux of ANS should be suppressed to the same level.

Neutron sources in the ANS material are the following (see Table 3):

- spontaneous fission of plutonium and transplutonim elements (TPE) - Am, Cm, Cf;

- (α,n)-reaction on the isotopes of light elements in the ANS material composition (heavy oxygen isotopes of $^{17,18}O$ for Pm_2O_3);
- (γ,n)-reaction on deuterium (if water will be used as the cooling agent and/or moderator).

Let's consider the neutron background channels in more detail, assuming the ~ 10^{-10} g/g purity of the ANS material and its activity being ~5 MCi.

2.1. Spontaneous fission of Pu and TPE.

In Table 3 [13,14] the characteristics for Pu and TPE isotopes are given. From the Table 3 data it follows that the most hazardous neutron sources are even isotopes of curium ($^{242,244,246}Cm$), plutonium ($^{238,242}Pu$), and californium ($^{250,252}Cf$).

However, despite the high yield of Pu in the fission products (~ 9 kg/ton) due to the separation of plutonium starting from the very first stage of spent fuel reprocessing, by the time of promethium recovery the plutonium admixture will be practically defined by the background in the chromatographic purification column. If we take 10^{-10} g/g as the assumption for that background, the ^{240}Pu isotope with the rate of spontaneous fission in the ANS of ~20 day^{-1} will be the most hazardous among the plutonium isotopes.

Californium isotopes possess very low absolute ($2.2 \cdot 10^{-6}$ and $4.0 \cdot 10^{-8}$) and relative ($3 \cdot 10^{-8}$ and $6 \cdot 10^{-10}$ atoms of Cf per atom of Pm) yield and thus may be excluded from consideration.

Summarizing the above, ^{244}Cm isotope appears to be the main source of neutrons from spontaneous fission due to high absolute and relative yield of it. Decay rate of the isotope will amount ~10^5 day^{-1}.

2.2. (α, n)-reaction of heavy oxygen isotopes.

If light elements and α-decaying isotopes are present in the ANS material the fast neutrons may be born in (α, n)-reaction. This is the case when the promethium oxide Pm_2O_3 is used for material in the ANS, and decays of ^{147}Sm (the product of ^{147}Pm decay), uranium, plutonium, and TPE isotopes are the source of α-particles.

In several years α-activity of the ANS material will be ~ 10^6 s^{-1} on account of the accumulation of long lived ^{147}Sm isotope ($T_{1/2}$ - $1.06 \cdot 10^{11}$y, E_α=2.23 MeV). However, due to the low α-particle energy the yield of (α, n)-reaction on heavy

oxygen isotopes is low, it follows that the neutron intensity on this channel will be negligible (< 1 day^{-1}).

Neutron yield for α-decay of Pu and TPE isotopes will be defined by the half life of those, energy of α-particles, and the fission product yield of the isotope given relative to the total yield of the chemical element in question (see Table 3).

Taking as the basis the above criteria and the results of simple calculations one can conclude the α-decay of ^{244}Cm and ^{241}Pu isotopes to be the primary source of neutrons in (α, n)-reaction on heavy oxygen isotopes (10^4 and 10^3 neutrons per day, respectively).

2.3. (γ,n)-reaction as a neutron source.

There are only two isotopes, namely, deuterium and ^{7}Be capable to produce neutrons in (γ,n)-reaction of their nuclei with high energy γ-ray emitted as the result of decay of long lived fission γ-emitters, namely, ^{106}Rh(^{106}Ru) and ^{144}Ce(^{144}Pr). Deuterium is encountered in water composition at the natural ratio of 0.015% of hydrogen content. Water may be used as a cooling agent and/or efficient fast neutron moderator in the ANS neutron shielding.

Threshold of (γ, n)- reaction on deuterium is 2.23 MeV, and the cross section is about 1 mbarn [14]. When water is used as a moderator it should be noted that it is most efficient for moderation of fast neutrons of <1 MeV energy. Therefore, it's reasonable to place water inside the tungsten shielding in such a way that the thickness of tungsten layer between promethium oxide and water being sufficient to convert the fast neutrons into the energy range below 1 MeV.

Estimates based upon the values for content of hazardous γ-emitting contaminants in the ANS material along with the data on the capture of neutrons by the tungsten layer show the application of water to be of no hazard from the standpoint of neutrons born in (γ,n)-reaction.

3. Evaluation of neutron shielding for the ANS

Passive shielding from γ-ray made of 25 cm of tungsten plus 5 cm of copper actually does not attenuate the fast neutron flux, and its value on the surface of the ANS is equal to $6 \cdot 10^{-5}$ cm^{-2}s^{-1} (see Fig.1). So, about 3 order of magnitude reduction of the neutron flux is necessary.

That demands either 2-3 order of magnitude improvement in purity of Pm preparation with regard to Am and Cm or taking special action to suppress the neutron flux by insertion of light elements into the ANS shielding as the moderator. Compounds of boron (lithium) are the best choice for efficient capture of moderated neutrons for they do not form high energy γ-ray as the result of the capture.

To optimize the parameters of neutron shielding the calculation have been made using multi group program in the 26 group approach for the BNAB-26 system of constants [15]. Water was used for the moderator, and boron for capture material in the form of boric acid solution. A 10 cm thick NaI(Tl) scintillator was used as the detector assumption for calculations, and the "ANS - detector" assembly was surrounded with passive shielding of 5 cm thick copper and 20 cm thick lead layers. ANS neutron shielding of 15 cm thick was placed between tungsten and copper layers.

According to the calculations, the total neutron flux on the detector surface is reduced down to $\sim 2 \cdot 10^{-8}$ $cm^{-2}s^{-1}$, and attenuation factor amounts ~500, geometric factor included (see Fig.2).

Conclusions

As the production of high purity ^{147}Pm isotope had been developed and full scale implemented in the former USSR now there is the capability to recover it from the NPP spent fuel within a short time span aiming at the creation of the artificial neutrino source of up to 5-6 MCi activity.

In the case of high purity promethium oxide the decay of ^{146}Pm isotope will be responsible for the most part of the ANS γ-radiation. In the most unfavorable case (see Chapter 2.1.) ^{146}Pm impurity will be $\sim 10^{-6}$ g/g and its γ-radiation will define the total thickness of the passive shielding as 25 cm of tungsten to get the γ-flux of $\sim 10^{-9}$ $cm^{-2}s^{-1}$. Antineutrino flux at the outer ANS surface will be $\sim 2 \cdot 10^{13}$ $cm^{-2}s^{-1}$ in the case.

Neutron emission of the ANS material will be defined by the spontaneous fission of $^{244,246}Cm$ isotope contaminants ($\sim 10^5$ day^{-1}) and by the (α, n)-reaction on heavy oxygen isotopes accompanying the α-decay of ^{244}Cm ($\sim 10^4$ day^{-1}) and ^{241}Pu ($\sim 10^3$ day^{-1}). Hence, to reduce the neutron background it is necessary to provide the purity of promethium oxide of $\sim 10^{-13}$ g/g with regard to curium and

$\sim 10^{-12}$ g/g with regard to plutonium (the estimation is done for the detector of calorimetric type, i.e. no special rejection of background events was used). Another way to solve the problem is to apply the 15 cm thick neutron shielding in the ANS (e.g., water with dissolved boron compound). However this measure will increase the ANS shielding dimensions, which, in its turn will reduce the antineutrino flux density approximately by a factor of 2. Further reduction of the background from the fast neutron scattering on the detector substance nuclei can be provided by pulse shape discrimination of such events (in [16] the attenuation factor ranged from 10 to 40 depending on the energy range).

Note. Due to the low probability ($<10^{-3}$) for one-fold Compton scattering of γ-ray in big size detector with the small transfer of 3 keV - 30 keV energy to electrons this energy range is preferable for use as the working one. In the case the requirements imposed on the level of high energy γ-emitting impurities may be lowered by 3 orders of magnitude. It's also worth to mention that the effect of possible abnormal magnetic momentum of neutrino is mostly revealed in the above energy range.

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Table 1.
Long lived isotopes of Pm, isolated from spent reactor fuel
(cooling time - 3 years)

Isotope, $T_{1/2}$	Specific activity, Ci/g	Isotope yield per 1 ton of spent fuel, Bq/ton	Isotope activity per 1 MCi of ^{147}Pm	Energy of γ-ray, (yield, %)
^{146}Pm (5.53 y)	443	$1.32 \cdot 10^{7}$	5 mCi	0.747 MeV (33.9 %) 0.735 MeV (22.9 %) ... 0.454 MeV (65.1 %) ...
^{147}Pm (2.62 y)	927	$2.69 \cdot 10^{15}$	1 MCi	0.197 MeV ($3.4 \cdot 10^{-7}$ %)
^{148m}Pm (41.3 d)	$2.14 \cdot 10^{7}$	$1.2 \cdot 10^{7}$	4.4 mCi	... 1.014 MeV (20.2 %) 0.915 MeV (17.1 %) 0.725 MeV 32.7 %) ... 0.630 MeV (88.6 %)
^{148}Pm (5.37 d)	$1.64 \cdot 10^{5}$	$6.35 \cdot 10^{5}$	0.24 mCi	2.28 MeV (0.044 %) 1.73 MeV (0.038 %)

Table 2.

Long lived isotopes, the products of fission of nuclear fuel.
(cooling time - 3 years, the level of impurities < 10^{-7}%)

Isotope ($T_{1/2}$)	Activity per 1 ton of spent nuclear fuel, Bq (Specific activity, Ci/g)	γ-radiation, energy, (yield, %)	Remarks
^{90}Sr (^{90}Y) (28.8 y)	$2.4 \cdot 10^{15}$ (139.4 Ci/g)	bremsstruhlung E < 2.28 MeV	^{90}Sr/^{88}Sr=1.45
^{106}Ru(^{106}Rh) (1.02 y)	$2.1 \cdot 10^{15}$ ($3.3 \cdot 10^{3}$ Ci/g)	3.40 MeV ($1.26 \cdot 0^{-5}$%) ... 3.05 MeV (0.0003%) ... 2.36 MeV (0.0224%) ... 1.56MeV (0.158%) ... 1.05 MeV (1.53%) ...	^{106}Ru/$^{100-103}$Ru < 10^{-2}
^{144}Ce-(^{144}Pr) (284.3 d)	$2.64 \cdot 10^{15}$ ($3.18 \cdot 10^{3}$ Ci/g)	0.133 MeV (19.2%)	Yield of cerium per 1 ton of spent fuel: m(Ce)=2 кg/ton ^{144}Ce/Ce~0.01
^{144}Pr (17.28 min)	$2.64 \cdot 10^{15}$ ($7.55 \cdot 10^{7}$ Ci/g)	2.65 MeV (0.00019 %) ... 2.186 MeV (0.70 %) 1.56 MeV (0.00023 %) 1.489 MeV (0.272 %) ...	(0.03 mCi)
^{152}Eu (12.4 y)	$2.3 \cdot 10^{11}$ (176.4 Ci/g)	... 1.77 MeV (0.087 %) ... 1.408 MeV (20.8 %) ... 0.964 MeV (14.5 %) ...	Yield of Eu per ton of spent fuel: ^{153}Eu=100 g/ton ^{152}Eu/^{153}Eu $=4 \cdot 10^{-4}$ (0.07 mCi)

Table 2 (continue).

^{154}Eu (8.5 y)	$2.51 \cdot 10^{14}$ (264 Ci/g)	1.89 MeV (0.0006%) ... 1.59 MeV (1.86 %) ... 1.275 MeV (35.5 %) ...	^{154}Eu/^{153}Eu =0.25 (7 mCi)
^{158}Tb (150 y)	$6.98 \cdot 10^{6}$ (15 Ci/g)	1.19 MeV (1.71 %) 1.11 MeV (2.06 %) 0.962MeV (19.8 %) 0.944 MeV (43 %)	(15 μCi)
^{160}Tb (72.3 d)	$5.53 \cdot 10^{8}$ ($1.13 \cdot 10^{4}$ Ci/g)	1.31 MeV (2.92 %) ... 1.27 MeV (7.49 %) ... 1.18 MeV (15.2%) ..	^{160}Tb/^{158}Tb ~ ~ 0.1

Table 3
Isotopes of plutonium and transplutonium elements (TPE)
[13,14]

Isotope	Half-life (α - decay), years	Half-life (spontaneous fission), years	Yield of isotope, g/ton, cooling time of spent nuclear fuel - 3 years
Plutonium			
^{238}Pu	87.34	$4.77 \cdot 10^{10}$	75
^{239}Pu	$2.41 \cdot 10^{4}$	$7.8 \cdot 10^{15}$	$5.5 \cdot 10^{3}$
^{240}Pu	$6.57 \cdot 10^{3}$	$1.15 \cdot 10^{11}$	$1.98 \cdot 10^{3}$
^{241}Pu	14.4	$>3 \cdot 10^{15}$	$1.25 \cdot 10^{3}$
^{242}Pu	$3.75 \cdot 10^{5}$	$6.84 \cdot 10^{10}$	$3.7 \cdot 10^{2}$
Americium			
^{241}Am	433	$\sim 10^{14}$	240
^{242m}Am	152	$8.8 \cdot 10^{11}$	0.26
^{243}Am	$7.37 \cdot 10^{3}$	$3.35 \cdot 10^{13}$	70
Curium			
^{242}Cm	162.8 days	$\sim 6.1 \cdot 10^{6}$	$\sim 4 \cdot 10^{-3}$
^{243}Cm	28.5	$5.37 \cdot 10^{11}$	~ 0.15
^{244}Cm	18.11	$1.37 \cdot 10^{7}$	21.3
^{245}Cm	$8.5 \cdot 10^{3}$	$1.39 \cdot 10^{12}$	~ 1
^{246}Cm	$4.7 \cdot 10^{3}$	$1.8 \cdot 10^{7}$	~ 0.1
^{248}Cm	$3.4 \cdot 10^{5}$	$4.1 \cdot 10^{6}$	$1.7 \cdot 10^{-5}$
Californium			
^{250}Cf	13.1	$1.7 \cdot 10^{4}$	$2.2 \cdot 10^{-6}$
^{252}Cf	2.638	85.38	$4.0 \cdot 10^{-8}$

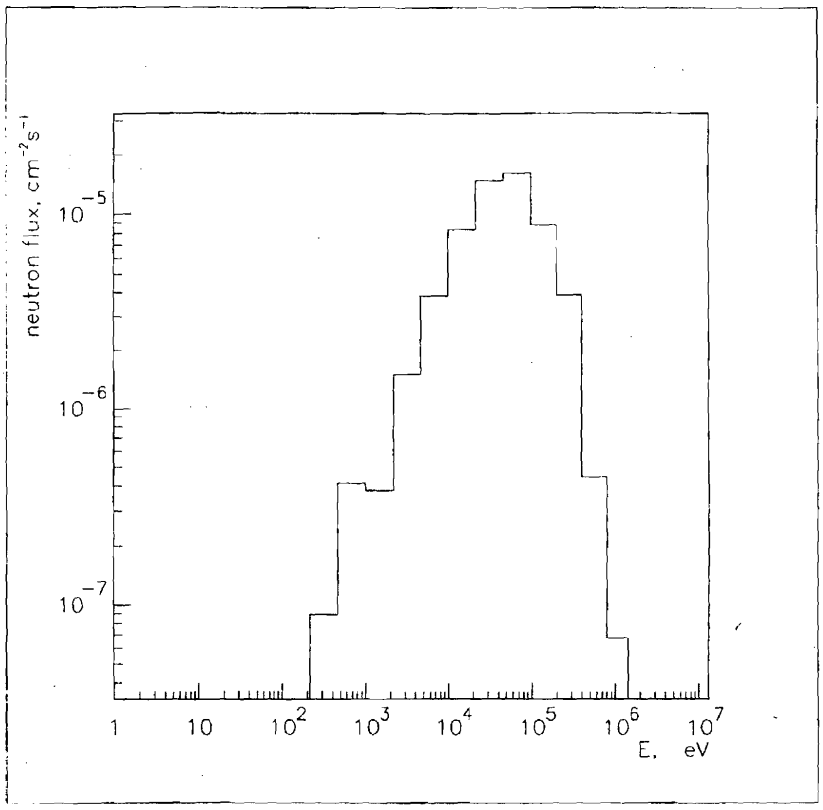

Рис.1. Спектр нейтронов на поверхности ИИН. Защита: W (25 см) + Cu (5 см).

Fig.1. Energy neutron spectrum on the surface of the neutrino source. Passive shielding - 25 cm of W + 5 cm of Cu).

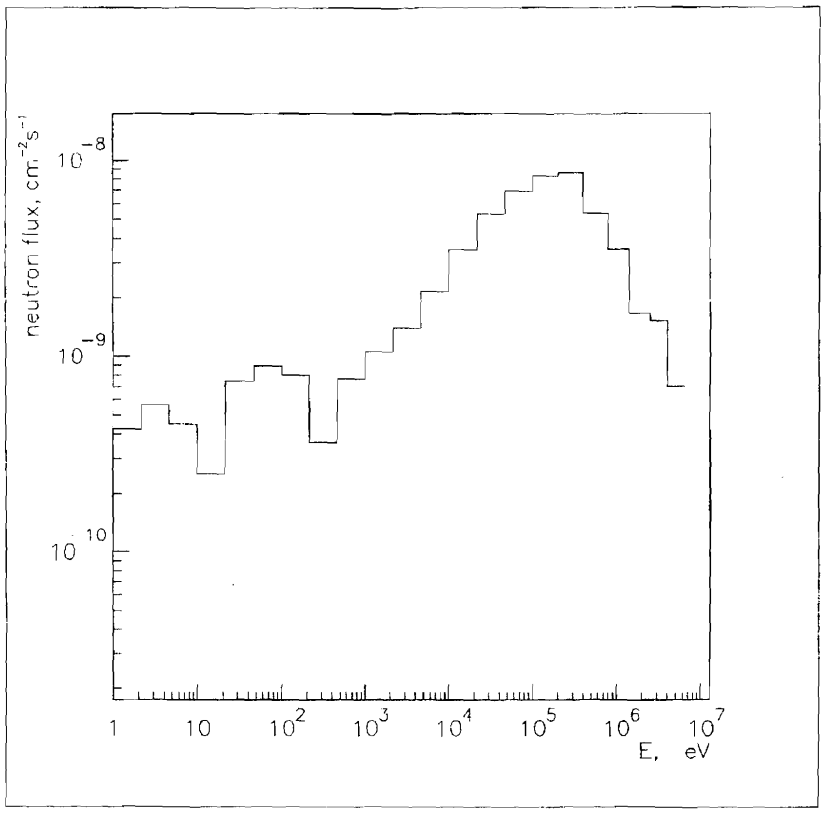

Рис.2. Спектр нейтронов на поверхности ИИН. Защита: W (25 см) + вода с растворенной борной кислотой (15 см) + Cu (5 см).

Fig.2. Energy neutron spectrum on the surface of neutrino source. Passive shielding 25 cm of W + 15 cm of water contained boric acid + 5 cm of Cu.

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