Nuclear Data for Science and Technology(1988 MITO), 995- 998, Copyright © 1988 JAERI.

NUCLEAR-PHYSICAL CHARACTERISTICS OF NEPTUNIUM ISOTOPES.

S.S.Kovalenko,Yu.A.Selitsky,V.B.Funshtein,S.V.Khlebnikov,V.A.Yakovlev

V.G.Khlopin Radium Institute, Leningrad 197022, USSR

Abstract: A survey is given of the work carried out at the Radium Institute on studying the nuclear-physical properties of neptunium that influence the work of nuclear power installations. There are presented the measured cross-sections of the reaction ^{237}Np(n,2n), giving rise to the short-lived isomer ^{236}Np(s), at E_n= 7+15 MeV and on ^{252}Cf spontaneous fission neutrons, the cross-sections of ^{237}Np, ^{236}Np(s) and the long-lived isomer ^{236}Np(l), by thermal neutrons and the respective resonance fission integrals, the cross-sections of ^{236}Np(l) fission by neutrons in the energy range E_n = 0.0253 eV + 20 keV, the partial half-life of ^{235}Np in relation to alpha radiation. New data are given on the cross-sections of reactions ^{238}U(p,xn)Np at E_p = 7+30 MeV applied for production of neptunium isotopes and plutonium-236.

(neptunium, isomer, neutrons, reactions, fission,cross-section)

Introduction

A series of works studying nuclear-physical properties of neptunium that influence the work of power installations have been carried out at the Radium Institute. A closed fuel cycle with broad reproduction of nuclear fuel is a characteristic feature in exploitation of fast-neutron nuclear reactors. The closed fuel cycle includes storage, transportation and reprocessing of spent fuel assemblies. In the nuclear reactor fuel there is ^{236}Np arising as a result of the ^{237}Np(n,2n) reaction in two isomeric states: the short-lived ^{236}Np(s) with the half-life $T_{1/2}$ = 22.5±0.4 h and spin I = 1^- /1/, and the long-lived ^{236}Np(l) with $T_{1/2}$ = 1.55×10^5 years and I = 6^- /2/. The decay chain of ^{236}Np including ^{236}Pu and ^{232}U causes formation of radioactive gases and nuclides with hard gamma radiation, which hinders regeneration of plutonium and uranium fuel. To solve the problem of calculating the accumulation of ^{236}Pu and 232U in nuclear fuel it was necessary to measure the cross-sections of the ^{237}Np(n,2n) reaction and the cross-sections of ^{236}Np and ^{237}Np fission by thermal and resonance neutrons.

A practical interest to studying the ^{236}Np(l) fission also arises due to this nuclide's having a great (~3000 b /3-7/) cross-section of fission by thermal neutrons, σ_f^{th}, and being accumulated in sufficient amounts it could be used as an energy source.

Besides, the great value of σ_f^{th} for ^{236}Np(l) can cause systematic error in measuring σ_f^{th}(^{237}Np) if neptunium has even a slight (10^{-5}+ 10^{-6}atom/atom) admixture of ^{236}Np(l). Therefore it was necessary to check if the previously published values of σ_f^{th}(^{237}Np) hadn't been distorted by the probable presence of ^{236}Np(l) admixture in the used preparations.

From the scientific view-point the study of ^{236}Np isomers enables to find out: 1) how nuclei with the same nucleonic composition differing only by the spin manifest themselves in fission; 2) the cause of great values of the cross-sections of odd-even nuclei's fission by thermal neutrons; 3) the energy dependence of the isomeric ratio - the ratio of the cross-sections of ^{236}Np(s) and ^{236}Np(l) formation.

Production of ^{236}Np.

The study of ^{236}Np properties is possible only in the case if there is a sufficient amount of purified substance. ^{236}Np may be produced by many nuclear reactions, such as ^{235}U(d,n),^{235}U(α,p2n), ^{236}U(d,2n),^{238}U(p,3n),^{238}U(d,4n), ^{237}Np(γ,n) ^{237}Np(n,2n),^{237}Np(d,p2n), ^{237}Np(d,t). The reactions with ^{237}Np don't enable to reach the necessary purity of neptunium-236 because of impossibility to separate ^{236}Np chemically from the material of the target. ^{236}Np(l) was produced by the ^{238}U(p,3n) reaction having the greates of all the reactions with uranium yield (~0.3 ng/μA·h at E_p = 30 MeV for a thick metallic uranium target). It was necessary to get ^{236}Np(s) with a small impurity of the short-lived neptunium isotope ^{238}Np ($T_{1/2}$= 2.1days) that has a great cross-section σ_f^{th}. Accumulation of ^{236}Np(s) with negligibly small ($\lesssim$ 0.5%) impurity of ^{238}Np was done by the reaction ^{236}U(d,2n) at E_d = 12.6 MeV.

The number of ^{236}Np(l) nuclei was determined by two methods: on the base of mass-spectrometric analysis of the isotopic ratio ^{236}Np(l)/^{237}Np with measuring the alpha activity of ^{237}Np, and by measuring the gamma activity of ^{236}Np(l) (E_γ = 158 and 160 keV). The amount of the ^{236}Np(s) formed was determined by the alpha radiation of the ^{236}Pu daughter nucleus. For determination of the yield of ^{236}Np(l) in chemical separation the tracer ^{235}Np was used, accumulated in the reaction ^{238}U(p,4n).

The number of ^{235}Np nuclei was measured by its gamma and alpha activity. Because of considerable discrepancy of the values of partial times of ^{235}Np alpha decay (about twice) in literature, its value was defined more exactly and turned out to be $(4.2 \pm 0.2) \times 10^4$ years /8/.

In fig.1a the cross-sections of neptunium isotopes' formation during ^{238}U irradiation with protons /9/ are shown. The solid and the dash curves represent the results of calculation within the framework of the statistical model by the modified variant of the STAPIF program with account of the contribution of pre-equilibrium processes. A good agreement between the experimental data and the theoretical calculations attracts attention.

Fig.1b presents new data on the isomeric ratio $R = \sigma(s)/\sigma(l)$ in the reaction $^{238}U(p,3n)^{236}Np$. The decrease of R with the growth of the proton energy is qualitatively comprehensive, because with an increase of E_p the introduced angular momentum I grows and the formation of the high-spin state of $^{236}Np(l)$ becomes more probable.

Cross-sections of the reaction $^{237}Np(n,2n)$

The cross-section values $\sigma_{n,2n}(s)$ of the reaction $^{237}Np(n,2n)^{236}Np(s)$ are important for calculating the accumulation of ^{236}Pu and ^{232}U in the reactor fuel. Measurement of these cross-sections is hindered by the small number of $^{236}Np(s)$ nuclei formed. Earlier the

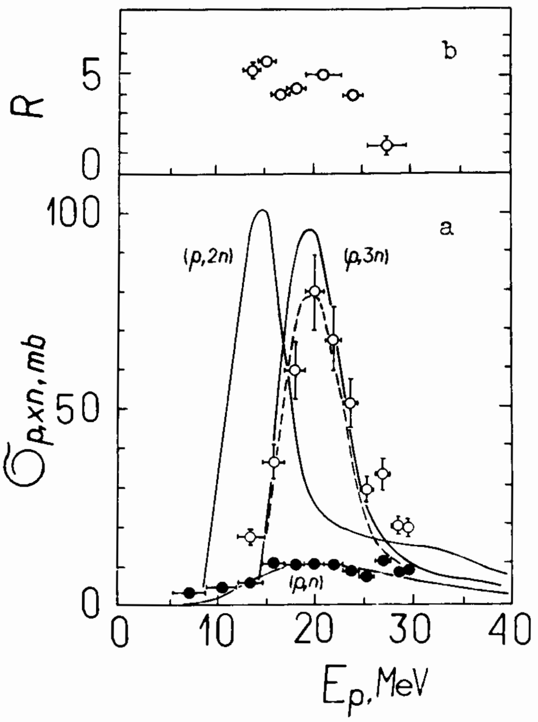

Fig.1: a - Experimental cross-sections of the reactions $^{238}U(p,n)^{238}Np$ (●) and $^{238}U(p,3n)^{236}Np(s)$ (○) and the calculated cross-sections of the reactions $^{238}U(p,xn)^{239-x}Np$, $x=1\div3$ (continuous curves) and $^{238}U(p,3n)^{236}Np(s)$ (dashed curve);

b - Isomer ratio $R = s/l$ of the reaction $^{238}U(p,3n)^{236}Np$.

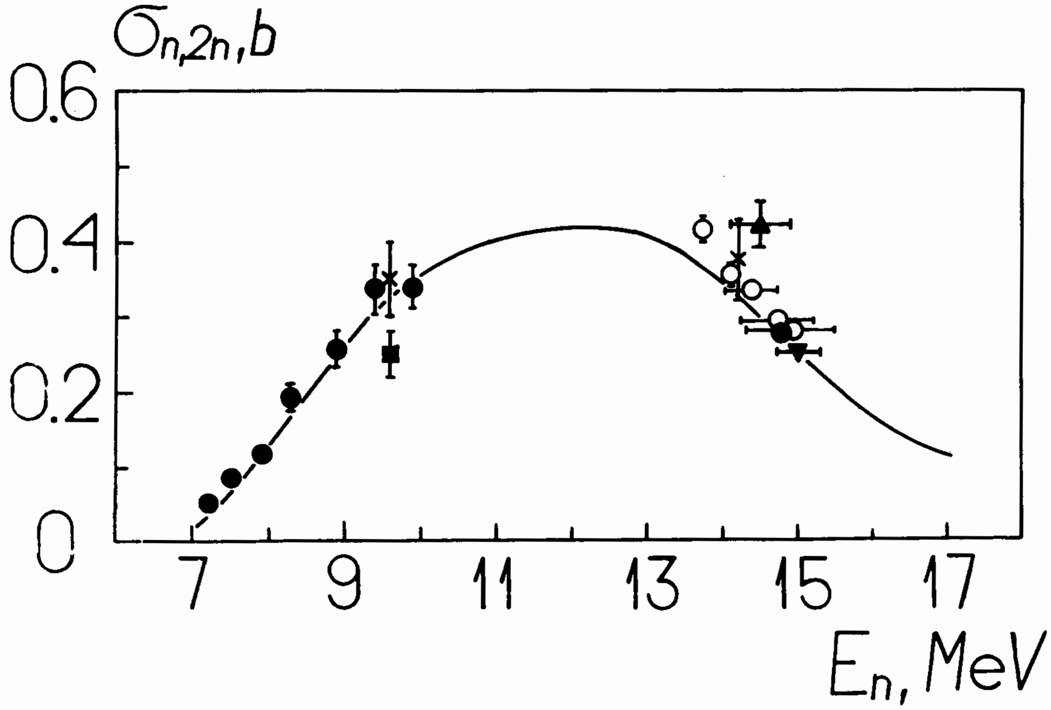

Fig.2. Experimental cross-sections of the reaction $^{237}Np(n,2n)^{236}Np(s)$ (▲ - /10/, ○ - /11/, ▼ - /12/, x - /13/, ■ - /14/, ● - /15-18/) and the calculated dependence of this reaction's cross-section from work /19/ (continuous curve).

$\sigma_{n,2n}(s)$ were determined only in the region E_n = 14÷15 MeV /10-12/(excluding the only measurement at E_n = 9.6 MeV /13,14/), where in the experiment great neutron fluxes are attainable. First we measured the $\sigma_{n,2n}(s)$ at E_n = 14.8 MeV, using both the traditional method of determining the number of ^{236}Np(s) nuclei by the alpha radiation of the daughter ^{236}Pu, and the developed new methods by means of registration of ^{236}U gamma radiation in decay of ^{236}Np(s) (E_γ = 642 and 688 keV) /15,16/. Then a series of experiment was carried out for determination of $\sigma_{n,2n}(s)$ in the range E_n = = 7÷10 MeV, where the yield of ^{236}Np(s) for the reactor neutron spectrum is maximum /17,18/. The neutron flux was determined by means of the reactions $^{27}Al(n,\alpha)$, $^{238}U(n,f)$ and $^{238}U(n,2n)$. The results of these and earlier /10-14/ experiments are presented in fig.2. The experimental values of $\sigma_{n,2n}(s)$ are well described by the results of the recently fulfilled calculations of these cross-sections /19/(the curve in fig.2).

The value of the cross-section $\sigma_{n,2n}(s)$ was measured for ^{252}Cf spontaneous fission neutrons /20/. The neutron flux was determined by means of activation detectors (^{27}Al, ^{197}Au, ^{48}Ti, ^{58}Ni), that have threshold energies close in value to the threshold of the $^{237}Np(n,2n)$ reaction. The value of $\sigma_{n,2n}(s)$ for ^{252}Cf spontaneous fission neutrons appeared to be 4.7±0.5 mb. A calculation of the cross-section averaged over the spectrum of ^{252}Cf fission neutrons was done as a check of agreement of the measured cross-sections on monoenergetic neutrons and the integral cross-section. A numerical integration indicated a value of 3.5±0.6 mb which agrees with the integral experiment.

Cross-sections of neptunium nuclei neutron fission

The cross-sections of ^{236}Np(l), ^{236}Np(s) and ^{237}Np fission by thermal neutrons (with a Maxwell spectrum), σ_f^{th}, and their resonance integrals of fission, I_f, were measured on the WWR-M reactor of the Leningrad Institute of Nuclear Physics. The measurements were done by the relative method using ^{235}U and ^{239}Pu. During measurement of the resonance integrals the thermal neutrons were absorbed by a cadmium screen 1 mm thick. The fission rate of the short-lived isomer ^{236}Np(s) decreased with time in accord with its known half-life $T_{1/2}$ = 22.5 h.

In table 1 there are the results of those experiments. The given in table 1 value of σ_f^{th} for ^{237}Np was calculated taking account of the Westcott multiplier for ^{237}Np /25/ and corresponds to the neutron energy 0.0253 eV.

Table 1. Cross-sections of thermal neutron fission and the resonance integrals of neptunium nuclei fission.

Nuclide	σ_f^{th}, b	I_f, b	Literature
^{236}Np(l)	2760±170	1030±100	/21,22/
^{236}Np(s)	2740±140	690±360	/22,23/
^{237}Np	0.020±0.001	4.70±0.23	/24/

The experiments for determination of $\sigma_f^{th}(^{237}Np)$ and $I_f(^{237}Np)$ were carried out twice: before and after a tenfold enrichment of neptunium with respect to the mass A = 237 on an electromagnetic mass-separator. The results of those experiments proved to be equal within the limits of the errors (±5%). The coincidence of the results of measuring $\sigma_f^{th}(^{237}Np)$ with one another and with the previously known values /26,27/shows that these values are not distorted by a possible presence of the ^{236}Np(l) admixture in the applied preparations.

The ^{236}Np(l) fission cross-section in the neutron energy range from 0.0253 eV to 0.3 eV was measured with a monochromator with a graphite crystal on the WWR-M reactor of the Leningrad Institute of Nuclear Physics, and in the range E_n = 0.1 eV ÷ 20 keV, by means of a lead slowing down spectrometer on the linac "Fakel" at the I.V.Kurchatov Institute of Atomic Energy (fig.3 /28/).

As one can see from fig.3, the energy dependence of the ^{236}Np(l) neutron fission cross-section differs insig-

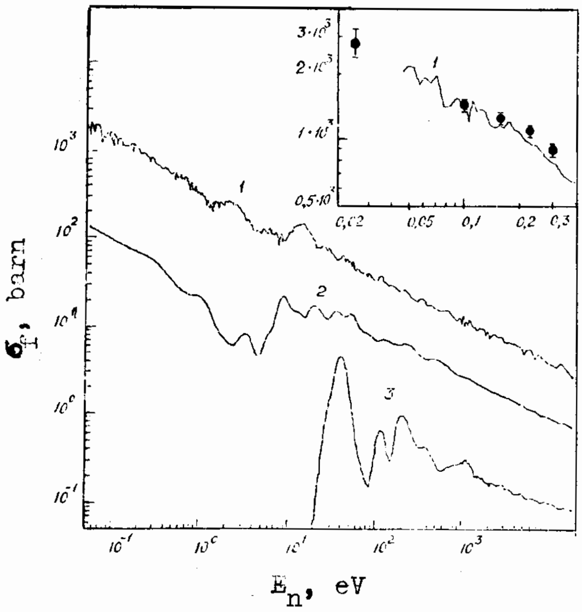

Fig.3. Fission cross-section of ^{236}Np(l) (curve 1). Curves 2 and 3 are ^{235}U and ^{237}Np fission cross-sections multiplied by coefficient which accounts their concentrations in the target.

nificantly from the 1/v-dependence in the whole energy range E_n = 0.0253 eV ÷ 20 keV. The resonance integral of ^{236}Np(l) fission, calculated on the base of these data, proved to be 1040±60 b, which well agrees with its experimental value (table 1). The σ_f value for ^{236}Np(l) at E_n= 0.0253 eV, σ_f=2770±260b, coincides with the value σ_f^{th} = 2760±170b (table 1) measured for the Maxwell neutron energy spectrum (T=330 K).

As noted above, the thermal neutron fission cross-sections of odd-odd nuclei are greater than those of even-odd nuclei. In fig.4 the known from literature values are presented of odd-odd and even-odd target nuclei thermal neutron fission cross-sections, including the data for ^{236}Np(l) and ^{236}Np(s) obtained by us. As seen from fig.4, the nuclides are divided into two groups by a straight line, above which are the odd-odd nuclei fission cross-sections (above twice, on the average). The increased density of the compound nucleus's levels is one of the factors that may cause great cross-sections of the odd-odd nuclei fission. In work /29/ there was found no correlation between the σ_f^{th} and the density of the levels of the compound nucleus in analysis of the whole collection of the odd-odd nuclei. Such an analysis becomes more correct if to consider pairs of isomers of the odd-odd nuclei having the same nucleon composition. The cross-section of the isomers of ^{242}Am, ^{244}Am, ^{254}Es, correlate with the spin dependence of the level's density, but the σ_f^{th} for ^{236}Np(s) and ^{236}Np(l), measured by us, don't prove it. The relationship of σ_f^{th} for ^{236}Np(s) and ^{236}Np(l) seems to be determined mainly by such a casual element as the degree of closeness of the compound nucleus's resonance to the thermal energy of neutrons. Another factor that can also cause great values of σ_f^{th} and I_f is an increased density of the transition states on the fission barrier and, hence, a great number of open fission channels of the formed odd-even compound nucleus. On the base of analysis of the ^{236}Np(l) fission cross-section in the resonance region of the neutron energy, averaged values were obtained of the total (0.5±0.2 eV), the fissioning (0.45±0.16 eV) resonance widths and the number of the open fission channels N_f=21±8 (by an order of magnitude greater than the N_f for even-odd target nuclei).

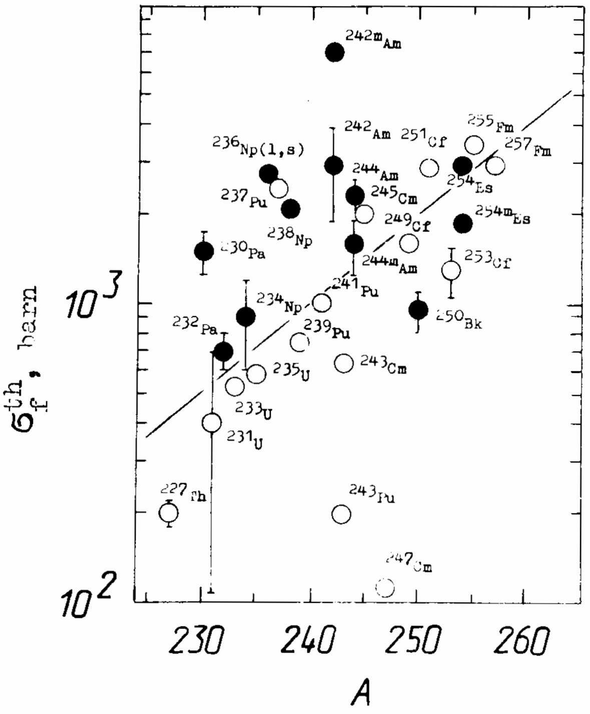

Fig.4. Cross-sections of the thermal neutron fission of even-odd (○) and odd-odd (●) nuclei.

References.

1.C.M.Lederer et al :Nucl.Phys.A135,36(1969)
2.M.Lindner et al: J.Inorg.Nucl.Chem. 43, 2071 (1981)
3.M.H.Studier et al:Phys.Rev.97,88 (1955)
4.A.Jaffey : ANL-6600, 125 (1961)
5.C.M.Lederer et al.: Tables of isotopes, N.-Y. (1978)
6.J.E.Gindler et al.:J.Inorg.Nucl.Chem. 43, 445 (1981)
7.M.Lindner et al:Report DOE Nucl.Data Committee,82 (1983)
8.V.A.Ageev et al:Proc.XXXVI Conf.Nucl. Spectr.Nucl.Struct.(Apr.1986,Kharkov, USSR),141 (1986)
9.V.A.Ageev et al:Yad.Fiz.46,760 (1987)
10.J.L.Perkin et al:Nucl.En.A/B14,69(1961)
11.J.H.Landrum et al:Phys.Rev.C8,1938 (1973)
12.K.Lindeke et al:Phys.Rev.C12,1507(1975)
13.T.Nishi et al.:NEANDC(J)42L,20
14.T.Nishi et al.:Rep.Stand.Discrep.Subcommitt.INDC (19 June 1980)
15.E.A.Gromova et al.:Sov.At.Energy 54, 116(1983)
16.E.A.Gromova et al.:Proc.Eur.Top.Conf. Neutron Induced Reactions (1982,Smolenice,Czechosl.),Bratislava10,403(1982)
17.N.V.Kornilov et al:Sov.At.Energy 58 , 131 (1985)
18.N.V.Kornilov et al:Proc.3th Advis.Group Meet.Trans.Is.Nucl.Data(1984,Upsala, Sweden)IAEA-336,305 (1985)
19.A.V.Ignatyuk et al:At.En.63,110(1987)
20.E.A.Gromova et al:Sov.At.Energy60, 95 (1986)
21.B.N.Belyaev et al:Sov.At.Energy 60, 177 (1986)
22.Yu.A.Selitsky et al:Proc.4th Int.Symp. Neutron Induced Reactions (1985,Smolenice,Czechosl.),Bratislava,3845 (1985)
23.E.A.Gromova et al.:Sov.At.Energy 56, 230 (1984)
24.V.V.Kozharin et al:Sov.At.Energy 60, 498 (1986)
25.E.Grintakis et al:Rad.Acta22,128(1975)
26.A.Ghiorso et al:The Transuranium Elements,2,1639 (1949)
27.C.Wagemans et al:Nucl.Phys.A285,32 (1977)
28.G.V.Valskiy et al: Proc.Int.Conf.Neutr. Phys.(1987,Kiev, USSR)
29.H.Diamond et al:J.Inorg.Nucl.Chem.30, 2553 (1968)