Resonance Enhancement in the Accelerator Transmutation of 1.3-day ^{232}Pa and 2.1-day ^{238}Np

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Abstract. The suggestion that the transmutation of actinide waste into fission products might best be done with thermalized spallation neutrons and odd-odd target materials such as ^{238}Np has been studied. During the 1993 LAMPF/PSR cycle, we measured the fission cross section of 1.3-day ^{232}Pa and 2.1-day ^{238}Np from 0.01 eV to 40 keV at the LANSCE facility, and have carried out a preliminary resonance analysis of the observed structure and of the thermal region, with a 1/v representation above a few eV. In the present study, we calculate the reaction rates of these two species and ^{247}Cm in a "resonance reactor," an accelerator-driven assembly whose slowing-down properties are well known. Our model is a 1.8 m^3 block of lead with a helium-cooled tungsten target in the center, i.e, the Rensselaer Intense Neutron Source (RINS). We include the effects of adding moderator outside an idealized lead slowing-down assembly, giving resonance enhancement factors for ^{232}Pa and ^{238}Np, and present parameters for the accelerator required to drive such an assembly to accomplish actinide burnup of these species.

INTRODUCTION

The suggestion was first made by Bowman et al [1] that the transmutation of actinide waste into fission products might be optimized by burning odd-odd target materials such as ^{238}Np and ^{242}Am in an assembly driven by a spallation neutron source. Because of the expected large fission widths of the odd-odd target isotopes, fission of these species would be the dominant reaction. It was also conjectured that the close spacings of the resonances could lead to a cross-section shape over the thermal Maxwellian that could be significantly different from 1/v, and that low-lying resonance structure could lead to a substantial enhancement of the burnup properties. In order to assess such effects, a measurement of the resonance fission cross section of these short-lived odd-odd targets is required. Of interest also, in connection with the Th-^{233}U fuel cycle, is the fission cross section of 1.3-day ^{232}Pa in the resonance region.

During the 1993 cycle of the Los Alamos Clinton P. Anderson Meson Physics Facility (LAMPF), we measured the fission cross sections of ^{232}Pa and ^{238}Np on a 5-m flight path at the Los Alamos Manuel J. Lujan Neutron Scattering Center (LANSCE). The samples were prepared by irradiating thick metal targets of ^{232}Th or ^{238}U with 19-MeV deuterons at the Los Alamos Ion Beam Facility (IBF), followed by radiochemical separation of the proactinium or neptunium produced by this irradiation. This material was then deposited onto a thin titanium backing that was inserted as one of the electrodes into a parallel plate fission chamber. The chamber was then transported to LANSCE facility, positioned in the neutron beam line, and the fission-fragment pulses were recorded over the next few days as a function of fission-fragment pulse height and neutron time of flight. The development of the technique, and a preliminary reduction and analysis of the data obtained, were described at the recent International Conference on Nuclear Data for Science and Technology. [2]

As part of the present study, we have carried out a final data reduction and a more elaborate analysis of the data from 0.01 eV to 40 keV, with a 1/v extrapolation to 100 keV. This parameterization was then used to calculate the reaction rates of these two species, and of another material of interest, ^{247}Cm, in an accelerator-driven assembly whose slowing-down properties are similar to those of the Rensselaer Intense Neutron Source (RINS), a 1.8 m³ cube of pure lead with a helium-cooled tungsten target in the center. [3] We calculated the neutron flux in a similar-sized cylindrical lead assembly, and studied the effects of adding a moderator blanket on the outside. We found that the resonance enhancement factors in this latter assembly, with a D$_2$O blanket, can be significant. However, the accelerator required to drive such an assembly in order to accomplish efficient burnup of these short-lived actinides may be beyond present technology.

DATA REDUCTION AND ANALYSIS

We found that production of the odd-odd samples ^{232}Pa and ^{238}Np by the (d,2n) reaction with ten to fifteen μA of 19-MeV deuterons on thick samples of ^{232}Th and ^{238}U, respectively, gives a few tens of nanograms of the isotopes of interest, which is an adequate amount to permit us to carry out the measurement. There is a competing reaction, the (d,n), which has about four times the integrated cross section, and produces four times as much ^{233}Pa and ^{239}Np as ^{232}Pa and ^{238}Np. The presence of these isotopes is not a problem with fresh samples, as they are subthreshold fissioners, and their fission contribution is small compared to the statistical spread of the data and can be neglected. However, these isotopes are also reasonably short-lived, and their decay products are fissile; ^{233}Pa decays into ^{233}U with a 27.0-day half life, and ^{239}Np into ^{239}Pu with a 2.35-day half life. While the presence of these fissile decay products causes a serious background problem, in another sense it is an enormous advantage: we can normalize the ^{232}Pa fission cross section, through the radiochemically measured protactinium isotopic ratios and the half lives, to well-known resonances in ^{233}U, and the ^{238}Np fission cross section to resonances in ^{239}Pu. The results are summarized in Table 1; in both cases, the thermal (2200 m/s) fission cross sections are found to be substantially higher than those reported in the Mughabghab compilation. [4]

Table 1. Contents of the Fission Chambers[a] and Normalization Results.

Fission Chamber Number	Contents	Amount of Isotope of Interest[a]	Amount of Other Isotope[a]	Normalizing Resonances Used (eV)	2200 m/s Cross Section
1	Protactinium	33 ng ^{232}Pa	137 ng ^{233}Pa	1.8 + 2.3 ^{233}U	1500±100 b
2	Neptunium	5.9 ng ^{238}Np	33 ng ^{239}Np	0.296 ^{239}Pu	2600±200 b
3	Neptunium	3.2 ng ^{238}Np	14.8 ng ^{239}Np	0.296 ^{239}Pu	--

[a]At the time of the final radiochemical separation from uranium or plutonium.

The actual content of the isotope of interest in the fission chambers during the course of the experiment ranged from about 90 % at the beginning of data taking to between 10 and 30 % at the end. In order to take account of the decay of the isotope of interest and the growing in of the fissile decay-product isotopes, we automatically read out the data to disk every hour. Because of the wide

variability of the neutron beam intensity, we found it necessary to process each of these short runs, and to average the results, reduced to fission cross section for the isotope of interest and weighted according to the product of the average amount of sample material present during that hour and the number of counts recorded by a [235]U foil mounted in the same chamber. It is important to emphasize that the [235]U fission monitor served only as a beam-intensity and flux-shape monitor; the data normalization being done through the [233]U or [239]Pu fission cross sections as described above.

By far, the largest source of background was the fission of the decay products. We found that for the [238]Np data, an adequate correction for [238]Pu and [239]Pu fission could be achieved by using a broadened ENDF/B-VI fission cross section. For the [232]Pa data, the representation in ENDF/B-VI for [232]U and [233]U did not give satisfactory results. Some of the [232]U resonances were overcorrected, e.g., the one at 6 eV, while others were undercorrected. We ended up using a tabulated fission cross section for [233]U, that of Moore et al [5], which had been measured with very similar resolution conditions. For [232]U, we fitted the present data to obtain a new set of neutron widths, with the resonance energies and fission widths listed in BNL-325. The background-corrected data are shown in Figs. 1 and 2 for [232]Pa and [238]Np, respectively.
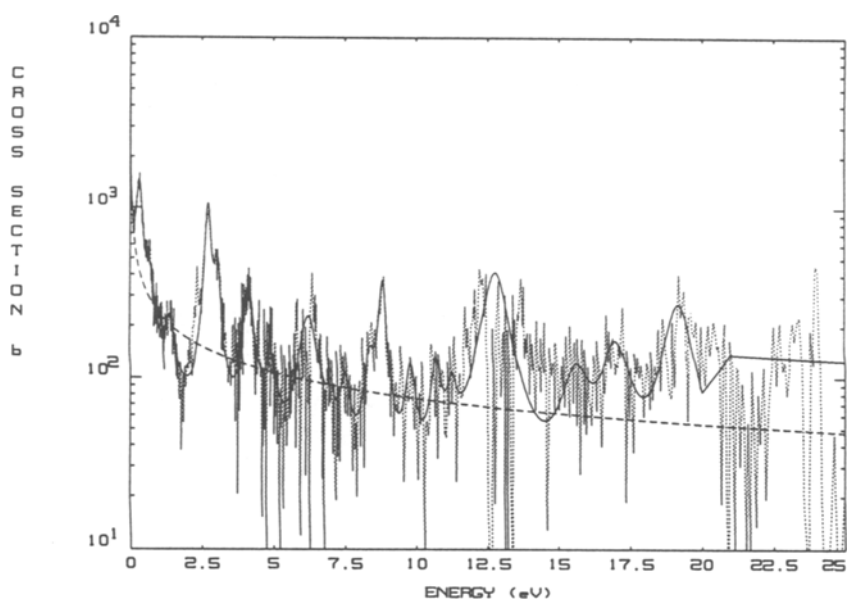

Fig. 1. The fission cross section of [232]Pa from 0.01 to 25 eV. The solid line shows a single-level broadened representation that preserves the area. The dashed line is a 1/v curve that passes through the 2200 m/s value.

The resonance structure shown in Fig. 1 was parameterized by a single-level broadened representation of the data below 20 eV that preserves the area. Above 20 eV, we have chosen to use a 1/v curve that preserves the area below 220 eV, which is the upper limit of the known resonance parameters for [232]U. Above 220 eV, for purposes of this study, we use the same 1/v curve to 100 keV and above.

Below 8 eV, the resonance structure shown in Fig. 2 is also parameterized by a single-level broadened representation that preserves the area. Above 8 eV, we have chosen to use a 1/v curve that preserves the area in the region below 100 eV. For purposes of this study, we use an extrapolation of this 1/v curve for [238]Np as a representation to 100 keV and above. The dashed line

in both Figs. 1 and 2 is a 1/v curve that passes through the measured fission cross section at the thermal (2200 m/s) value. We define resonance enhancement as the ratio of the reaction rate calculated with the resonances included to that calculated from the 1/v curve only.
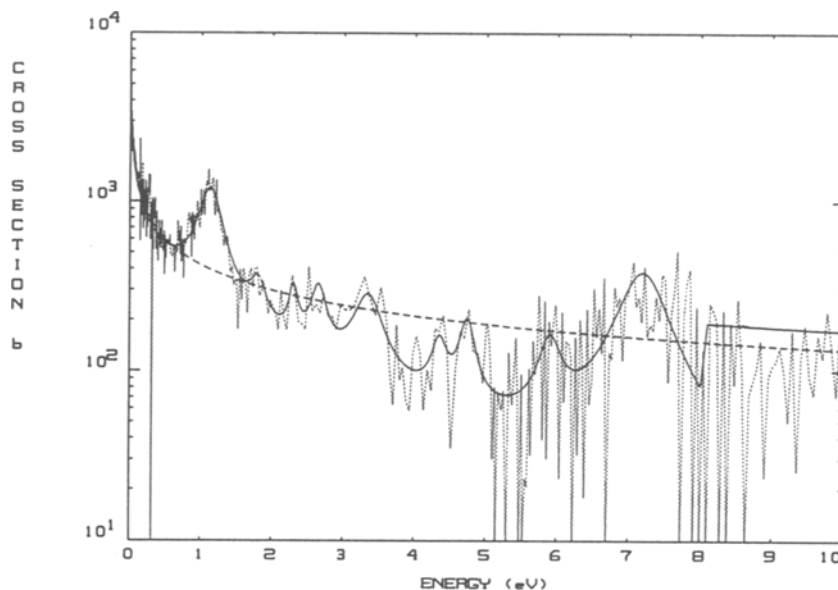

Fig. 2. The fission cross section of ^{238}Np from 0.01 to 10 eV. The solid line shows a single-level broadened representation that preserves the area. The dashed line is a 1/v curve that passes through the 2200 m/s value.

A SIMPLE ACCELERATOR-DRIVEN ASSEMBLY

The experience gained in the calculation of the measured neutron slowing-down spectrum in the RINS, the Rensselaer Intense Neutron Source, suggests that, if the accelerator-driven assembly we choose in this study of resonance enhancement is not too different from the RINS, then the same calculational technique should give a reasonable estimate of the neutron slowing-down properties. The RINS consists of a 1.8-m cube of pure lead, driven by a helium-cooled tantalum electron-bremsstrahlung target in the center. The flux has been measured many times: every fission cross-section measurement in this assembly requires a flux measurement. The flux has also been successfully calculated, with the MCNP code [6], a study that gave new insight into the resolution properties of the RINS.

The present study began with a simple cylindrical scaled-down copy of the RINS: a 1.5-m D and 1.5-m high solid cylinder of pure lead, again we calculated the slowing-down properties as if it were to be driven with an electron-bremsstrahlung neutron source. The resulting calculated flux shape, as shown in Fig. 3, was a bit disappointing in the thermal region; we modified it by adding a 75-cm thick D_2O blanket. The geometry and dimensions of this simple array, for the MCNP calculations, are described in Table 2. Finally, we tested, in a very crude way, whether this calculation might be appropriate for a proton-driven spallation source, by introducing a 19-MeV point source of neutrons instead of the electron-bremsstrahlung source. The results were a factor of 2 larger than those in Fig. 3, which is reasonable agreement for such a study as this.
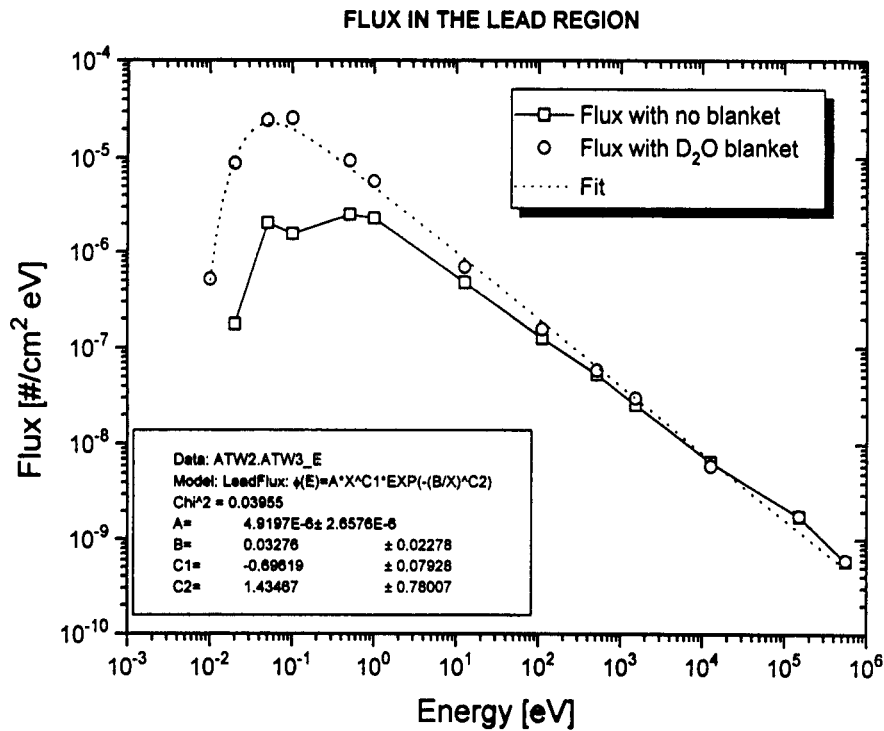
Fig. 3. Calculated neutron flux, driven by one neutron per unit time, for the pure lead assembly with D_2O moderator described in Table 2.

Table 2. Geometry and Dimensions of the Lead Cylinder and D_2O Blanket for MCNP.

Cell Number	Description	Height	Radius (Thickness)
1	Lead Cylinder	150 cm	75 cm
2	Flux Tally Region	20 to 40 cm	(20 cm)
3	D_2O Moderator	150 cm	(75 cm)

As is shown in Fig.3, the fit to the calculated flux with the D_2O blanket is given by

$$\Theta\,(\,E\,) = (\,4.9197\ e\text{-}6\,)\,E\,{**}\,(\,\text{-}0.69619\ \exp\,(\,\text{-}\,(\,0.03276\,/\,E)\,{**}\,1.43467\,)\,), \tag{1}$$

where the flux has the units of neutrons/cm²-eV per unit time, and is driven by a neutron production rate of 1 neutron per unit time. The reaction rate is given by $\int \sigma_f\,(E) \cdot \Theta\,(E)\,dE$. We define the resonance enhancement factor as the ratio of the above quantity to the integral $\int (C\,/\,\sqrt{E}) \cdot \Theta\,(E)\,dE$, where the constant C is chosen to be equal to σ_f (0.0253 eV) $\cdot \sqrt{0.0253}$. The energy dependence of the calculated reaction rates in the simple assembly of Table 2, with resonances included and for the 1/v dependence of the cross sections for [232]Pa, [238]Np, and [247]Cm, another actinide of interest to ATW, are listed in Table 3. The resonance enhancement factors are the ratios of the pairs of entries in the bottom line of Table 3, 1.84 for [232]Pa, 1.13 for [238]Np, and 8.07 for [247]Cm in this assembly. Even though the resonances in [247]Cm have a very large relative effect on the reaction rate, the value of this quantity is still only about half that of the odd-odd isotopes.

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Table 3. Calculated Reaction Rates for ^{232}Pa, ^{238}Np, and ^{247}Cm in the Assembly of Table 2.

Energy range (eV)	^{232}Pa with resonances	^{232}Pa as 1/v	^{238}Np with resonances	^{238}Np as 1/v	^{247}Cm with resonances	^{247}Cm as 1/v
0 to 0.01	4.8 e-6	4.8 e-6	8.3 e-6	8.3 e-6	5.8 e-7	5.8 e-7
0.01 to 0.1	0.00192	0.00179	0.00308	0.00313	0.00021	0.00022
0.1 to 1	0.00622	0.00334	0.00581	0.00582	0.00041	0.00041
1 to 10	0.00309	0.00218	0.00466	0.00380	0.00547	0.00050
10 to 100	0.00338	0.00138	0.00314	0.00243	0.00139	0.00027
10^2 to 10^3	0.00232	0.00088	0.00200	0.00154	0.00081	0.00017
10^3 to 10^4	0.00151	0.00057	0.00130	0.00100	0.00084	0.00007
10^4 to 10^5	0.00094	0.00036	0.00081	0.00062	0.00075	0.00004
$> 10^{5}$ [a]	(0.00110)	0.00022	(0.00095)	0.00037	(0.00087)	0.00002
0 to 10^5	0.01934	0.01053	0.02081	0.01835	0.01033	0.00128

[a] Numbers in parentheses are estimates, based on an electron-bremsstrahlung neutron spectrum.

ACCELERATOR REQUIREMENTS

The fluxes and reaction rates above were calculated for a neutron production rate of one neutron per unit time. The next step is to calculate the Bateman equation to see if the assumed assembly can be driven in such a way as to accomplish burning of actinides by fission of ^{238}Np or ^{232}Pa. The differential equation for the production and removal of, e.g., ^{238}Np is given by

$$dN_{238} = ((N_{237} \cdot \sigma_\gamma^{237} \cdot \Theta) - \lambda_{238} - (\sigma_f^{238} + \sigma_\gamma^{238}) \cdot \Theta) \, dt, \qquad (2)$$

where N_{237} is the number of ^{237}Np atoms present at time t, σ_γ^{237} is the capture cross section of ^{237}Np (i.e, the production cross section for ^{238}Np), Θ is the neutron flux, λ_{238} is the decay constant and σ_f^{238} and σ_γ^{238} are the fission and capture cross sections of ^{238}Np. In order to convert actinides to fission products in an efficient way through ^{238}Np, two conditions must obtain. First, the ^{238}Np fission cross must be much larger than the capture cross section, and second, the product of the fission-plus-capture cross section and the flux must be much larger than the decay constant. The closed-form solution of this equation is well-known; Fig. 4 shows a family of curves giving the number of atoms of ^{238}Np as a function of time for five decades of flux level ranging from 2·e13 to 2·e17 n/cm²-s in the accelerator-driven assembly of the previous section. In Fig. 4, Curves A and B show the characteristic behavior when the flux level is too low. Curve E, at the highest flux level, shows a different behavior: the amount of ^{238}Np peaks and is reduced rapidly as the ^{237}Np disappears.

Can we burn ^{238}Np in an efficient way? The answer appears to be "No", at least not with the simple assembly we have chosen to calculate. Let us suppose we drive such an assembly with 1A of 800 MeV protons, and that we have solved the problem of dissipating the 800MW of heat produced. If we use the usual number of 22 neutrons produced in liquid lead by each proton [7], then the production rate is 22/(1.6 e-19) n/s, which, multiplied by the appropriate entry in the bottom line of Table 3, gives a value of 2.9 e-6 per second for the fission reaction rate $\sigma_f \cdot \Theta$. The decay constant λ_{238} for a 2.117-day half life is 3.8 e-6 per second, implying that in this case actinide burning is about 40% efficient. We have seen that a factor of two increase in the flux may be

expected if we use a more realistic neutron driving spectrum; this increases the actinide burning to 60%. Other factors of two may be possible; we need a factor of 10 to 100.
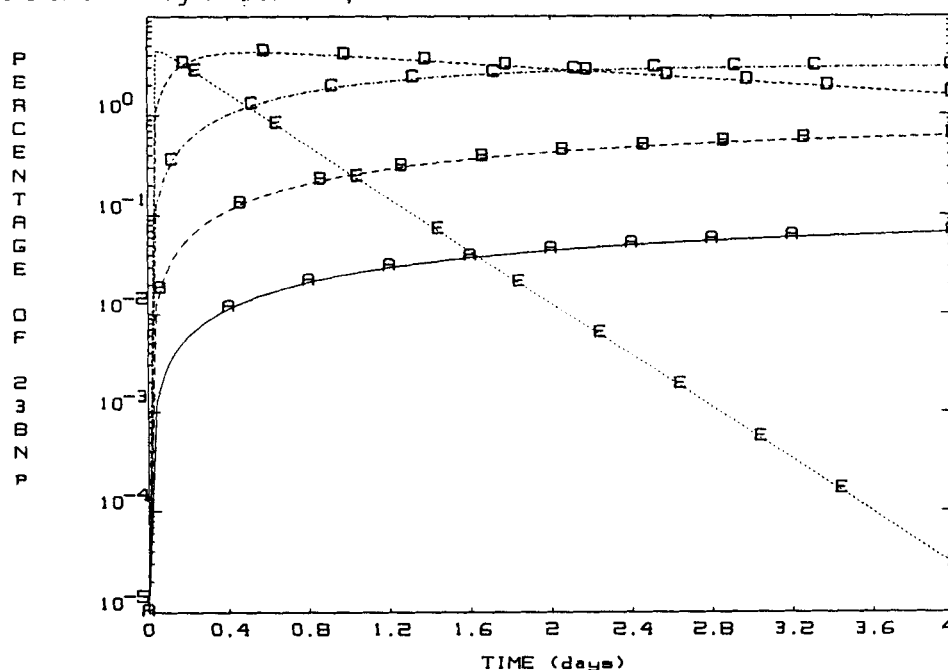

Fig. 4. The number of atoms of ^{238}Np, relative to the number of ^{237}Np atoms present at t = 0, as a function of time in days, for five decades of neutron flux ranging from 2·e13 (Curve A) to 2·e17 n/cm^2-sec (Curve E).

ACKNOWLEDGEMENTS

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