Neutron Separation Energies and Pairing Energies for Heavy Nuclei*

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A six-inch double-focusing mass spectrometer has been employed to determine 32 mass differences between neighboring isotopes of even-Z elements in the region $Z=64$ to 82. Neutron separation energies and neutron pairing energies are easily calculated from the experimental results. These calculated values are sufficiently precise to conclude that lack of uniformity in neutron separation energy and pairing energy systematics appears in regions where other nuclear properties exhibit changes in their systematics. Neutron separation energy data have also been employed for the isotopic assignments of several nuclear reactions.

INTRODUCTION

THIS paper presents the results of some mass-doublet measurements in the rare-earth and heavy-mass region performed with a six-inch double-focusing mass spectrometer. These measurements were undertaken to study neutron binding-energy systematics for $A>150$. Thirty-two mass differences between neighboring, even Z, isotopes in the region $Z=64$ to 82 have been determined. These doublets can be measured with sufficient precision to make them valuable in the determination of neutron separation energies and pairing energies and also in the assignment and verification of nuclear reaction Q values.

PROCEDURE

The general procedure of mass measurement has been discussed previously and has not been modified for the present determinations.[1-3] The mass spectrometer employed has the property that the mass of the ion collected is directly proportional to the resistance of a voltage divider which determines the electric fields in the instrument. Thus the mass difference, ΔM, measured in terms of a difference in resistance ΔR is calculated by means of Eq. (1)[3]:

$$\Delta M=\left(\frac{M_L}{R}\right)\Delta R \quad \text{or} \quad \Delta M=\left(\frac{M_H}{R+\Delta R}\right)\Delta R. \quad (1)$$

The masses M_L and M_H refer respectively to the masses of the lighter and the heavier constituents of the doublet, and R is a fixed, accurately known resistance. In a typical run, twenty determinations of ΔR are recorded. Four or more such runs, taken generally on different days and under different experimental conditions, are averaged to obtain the final result for a mass difference.

In the region $A>150$, increasingly large C^{13} satellite corrections[2] must be applied to hydrocarbon doublets because of resolution limitations of the mass spectrometer employed. Because of inherent inaccuracies in the calculation of these corrections, doublets containing hydrocarbon comparison ions were not employed. Instead, mass doublets composed of two isotopes of a particular element differing by one mass number were measured. Doublets such as these, called isotopic mass units, have been employed previously,[2] but largely as a consistency test for the hydrocarbon doublet data. Because of the good agreement of the previous isotopic mass-unit measurements with the corresponding mass differences derived from hydrocarbon doublet data, we feel that we may rely on the isotopic mass-unit determinations alone in the present region where hydrocarbon doublet results with small C^{13} satellite corrections could not be obtained. In general, doublets containing hydrocarbon comparison ions are preferred because an actual atomic mass is determined. However, because of the emphasis of this paper on neutron separation and pairing energies, we feel that the isotopic mass-unit measurements can supply satisfactory results.

RESULTS

Table I lists the doublets measured and the mass differences obtained. The use of Eq. (1) for the computation of ΔM requires an approximate value for either M_L or M_H. The assumption of the masses of the secondary standards $C^{12}=12.0038156\pm4$ and $H^1=1.0081451\pm2$[4] combined with the doublets listed in the review article by Duckworth *et al.*[5] provides us with mass values accurate enough for our purpose. High accuracy for these masses is not required. For example, a change of 10 mMU in M_L or M_H would not lead to a change in the value of these doublets that is more than the quoted error. Previous mass results for the element ytterbium appear to be incorrect. For this reason the required ytterbium masses were estimated from average binding energy per nucleon systematics for this region.

The general procedure of error analysis for the

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[1] National Bureau of Standards Circular 522, 1953 (unpublished).
[2] W. H. Johnson, Jr., and A. O. Nier, Phys. Rev. **105**, 1014 (1957).
[3] Quisenberry, Scolman, and Nier, Phys. Rev. **102**, 1071 (1956).
[4] Quisenberry, Giese, and Benson, Phys. Rev. **107**, 1664 (1957), this issue. Each error listed refers to the last significant figure of the particular result.
[5] Duckworth, Hogg, and Pennington, Revs. Modern Phys. **26**, 463 (1954).

TABLE I. Measured mass differences.

Doublet	ΔM amu[a]
$Gd^{155}-Gd^{154}$	1.002 15
$Gd^{156}-Gd^{155}$	0.999 90
$Gd^{157}-Gd^{156}$	1.002 20
$Gd^{158}-Gd^{157}$	1.000 53
$Dy^{161}-Dy^{160}$	1.002 10
$Dy^{162}-Dy^{161}$	1.000 21
$Dy^{163}-Dy^{162}$	1.002 26
$Dy^{164}-Dy^{163}$	1.000 80
$Er^{167}-Er^{166}$	1.002 06
$Er^{168}-Er^{167}$	1.000 65
$Yb^{171}-Yb^{170}$	1.001 88
$Yb^{172}-Yb^{171}$	1.000 40
$Yb^{173}-Yb^{172}$	1.002 17
$Yb^{174}-Yb^{173}$	1.000 97
$Hf^{177}-Hf^{176}$	1.002 25
$Hf^{178}-Hf^{177}$	1.000 88
$Hf^{179}-Hf^{178}$	1.002 36
$Hf^{180}-Hf^{179}$	1.001 13
$W^{183}-W^{182}$	1.002 23
$W^{184}-W^{183}$	1.000 99
$Os^{187}-Os^{186}$	1.002 14
$Os^{188}-Os^{187}$	1.000 33
$Os^{189}-Os^{188}$	1.002 55
$Os^{190}-Os^{189}$	1.000 52
$Pt^{195}-Pt^{194}$	1.002 45
$Pt^{196}-Pt^{195}$	1.000 49
$Hg^{199}-Hg^{198}$	1.001 82
$Hg^{200}-Hg^{199}$	1.000 31
$Hg^{201}-Hg^{200}$	1.002 26
$Hg^{202}-Hg^{201}$	1.000 64
$Pb^{207}-Pb^{206}$	1.001 74
$Pb^{208}-Pb^{207}$	1.001 06

[a] The error in each measured mass difference is ±0.06 mMU, i.e., ±6 for the last significant figure. This error is to be considered as a standard error. We estimate that the limit of error is about three times the standard error.

present work is similar to that used previously in this laboratory. However, use of the isotopic mass-unit technique necessitated consideration of several contributing errors that were previously negligible. For the present doublets, the statistical standard error calculated for the set of runs is not the predominant source of error. Errors arising from resistance-calibration uncertainties, in the case of wide doublets such as these, contribute most to the final doublet error. We have assumed errors in our resistor calibration of 50 parts per million in R and 30 parts per million in ΔR. The final error in a mass difference has been taken to be the square root of the sum of the squares of the individual contributing errors. The quoted errors are to be considered as standard errors. We estimate that the limit of error is about three times the standard error.

A verification of the reliability of doublet results can be made by measuring some doublet having a known mass difference. In this region, hydrocarbons differing by one hydrogen mass and selected to have the smallest possible C^{13} corrections were the most convenient doublets for this verification. Although the C^{13} correction for these selected doublets was small, it was taken into consideration in the calculation of the final doublet result and its error. The final average of these results is 1.008156±70 amu which because of the magnitude of the error can be considered to be a satisfactory comparison with the accepted mass difference 1.0081451 ±2 amu.

Several of the present mass differences can be compared with corresponding mass differences derived from nuclear-reaction Q values. Table II indicates the comparisons that may be made. In most cases, there is good agreement between the two independent methods of measurement. This fact, together with the good agreement found for the hydrogen mass-unit measurements, appears to indicate that there is no significant systematic error in the present results.

NEUTRON BINDING ENERGIES

The region $N=82$ to 126 shows considerable promise for the study of the systematics of various nuclear properties. The "distance" between the shell closures is greater than any other in the naturally occuring elements. In addition, it contains the region of large static nuclear deformation. Many nuclear properties in this

TABLE II. A comparison of the present mass differences with similar mass differences obtained from nuclear-reaction Q values. The particular type of reaction employed is indicated after the nuclear reaction result.

Mass difference	Mass difference in amu[a] Present result	Nuclear reaction result[b]
$Gd^{158}-Gd^{157}$	1.000 53±6	1.002 2 ± 4 (n,γ)[c]
$W^{184}-W^{183}$	1.000 99±6	1.001 02 ± 2 (n,γ)
$Pt^{195}-Pt^{194}$	1.002 45±6	1.002 43 ± 9 (γ,n)
		1.002 4 ± 2 (d,p)
$Pt^{196}-Pt^{195}$	1.000 49±6	1.000 480±13 (n,γ)
		1.000 43 ±20 (d,p)
		1.000 18 ±20 (γ,n)
$Hg^{200}-Hg^{199}$	1.000 31±6	1.001 4 ± 4 (n,γ)[d]
		1.000 36 ± 3 (n,γ)[e]
$Hg^{201}-Hg^{200}$	1.002 26±6	1.002 09 ±20 (γ,n)
$Pb^{207}-Pb^{206}$	1.001 74±6	1.001 754± 8 (n,γ)
		1.001 79 ± 2 (d,p)
		1.001 56 ± 7 (γ,n)
		1.001 81 ± 5 (d,t)
$Pb^{208}-Pb^{207}$	1.001 06±6	1.001 060± 8 (n,γ)
		1.001 08 ± 3 (d,p)
		1.000 96 ± 9 (γ,n)[f]
		1.001 08 ± 5 (d,t)

[a] Each error listed refers to the last significant figure of the particular result.

[b] Unless otherwise noted, nuclear-reaction Q values are obtained from the review article, D. M. Van Patter and W. Whaling, Revs. Modern Phys. 26, 402 (1954).

[c] This reaction appears to be misassigned. A discussion of this is given in the section on Nuclear Reaction Assignments.

[d] This reaction appears to be misassigned or incorrect.

[e] Adyasevich, Groshev, and Demidov, *Proceedings of the Conference of the Academy of Sciences of the U.S.S.R. on the Peaceful Uses of Atomic Energy, Moscow, July, 1955* (Akademiia Nauk, U.S.S.R., Moscow, 1955) [translated by the Consultants Bureau, New York, (1955)], Phys. Math. Sci., p. 270.

[f] This result is calculated from a weighted average of the individual results listed in Van Patter and Whaling.

area exhibit systematic behavior that is predicted from calculations based on the collective model.

The present experimental mass differences may be employed to determine a number of neutron separation energies, S_n,[6] and pairing energies, P_n, for even-Z nuclei in this region. A neutron mass of 1.0089860±10 amu[7] was employed in these calculations. The results of these calculations are given in Table III. The pairing energy associated with the "last" pair of neutrons for the nucleus with Z protons and N neutrons is given by Eq. (2).

$$\begin{aligned} P_n(Z,N) &= S_n(Z,N) - S_n(Z,N-1) \qquad N \text{ even} \\ &= [M(Z,N-1) - M(Z,N-2)] \\ &\quad - [M(Z,N) - M(Z,N-1)]. \end{aligned} \tag{2}$$

TABLE III. Neutron separation energies and neutron pairing energies determined from the present mass-difference results.

Isotope	N	S_n mMU[a]	P_n mMU[b]
$_{64}Gd^{155}$	91	6.84	
Gd^{156}	92	9.09	2.25
Gd^{157}	93	6.79	
Gd^{158}	94	8.46	1.67
$_{66}Dy^{161}$	95	6.89	
Dy^{162}	96	8.78	1.89
Dy^{163}	97	6.73	
Dy^{164}	98	8.19	1.46
$_{68}Er^{167}$	99	6.93	
Er^{168}	100	8.34	1.41
$_{70}Yb^{171}$	101	7.11	
Yb^{172}	102	8.59	1.48
Yb^{173}	103	6.82	
Yb^{174}	104	8.02	1.20
$_{72}Hf^{177}$	105	6.74	
Hf^{178}	106	8.11	1.37
Hf^{179}	107	6.63	
Hf^{180}	108	7.86	1.23
$_{74}W^{183}$	109	6.76	
W^{184}	110	8.00	1.24
$_{76}Os^{187}$	111	6.85	
Os^{188}	112	8.66	1.81
Os^{189}	113	6.44	
Os^{190}	114	8.47	2.03
$_{78}Pt^{195}$	117	6.54	
Pt^{196}	118	8.50	1.96
$_{80}Hg^{199}$	119	7.17	
Hg^{200}	120	8.68	1.51
Hg^{201}	121	6.73	
Hg^{202}	122	8.35	1.62
$_{82}Pb^{207}$	125	7.25	
Pb^{208}	126	7.93	0.68

[a] The error associated with the S_n calculations is ±0.06 mMU.
[b] The error associated with the P_n calculations is ±0.03 mMU.

[6] In conformity with growing practice, we will use the term neutron separation energy, S_n, in place of the term binding energy of the last neutron, B_n, which has been previously employed in publications from this laboratory.

[7] The neutron mass employed was determined from the mass of H^1 (see reference 4) together with the $n-H^1$ mass difference from A. H. Wapstra, Physica **21**, 367 (1955).

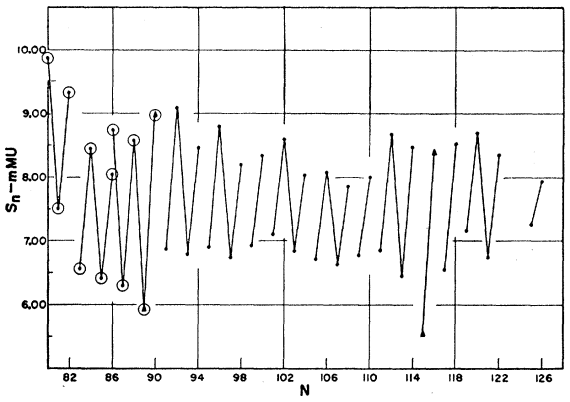

FIG. 1. Neutron separation energies S_n plotted as a function of the neutron number N. Data from a given element are connected by a solid line. The circled results from $N=80$ to 90 are taken from reference 2. A result indicated by a triangle is calculated by using nuclear-reaction Q values or β-decay energies.

Because the pairing energy is found by taking the difference between two experimental mass differences, determined under the same conditions, the principal source of error in an individual mass-difference determination, that due to resistance-calibration errors, is largely eliminated. Instead, the predominant error in the final result is just that due to the statistical errors. An error of ±0.03 mMU has been assigned to these experimental neutron pairing energies.

Neutron separation energy, S_n, is plotted as a function of the neutron number in Fig. 1. For completeness we have included values from $N=80$ to 90 taken from the previous paper[2] and also several neutron separation energies, derived from nuclear-reaction Q values. While the present results provide interesting data, the fact that odd-Z elements are not included does provide some limitation. To conform with the limitation of the present results, we have chosen from the previous investigation only stable, even-Z results for the region $N=80$ to 90. This limitation obscures to some extent the decrease in neutron separation energy so clearly shown in the previous, more complete study.[2] Nevertheless, several general conclusions may be reached from the results illustrated in Fig. 1. It is interesting to observe that the values of S_n remain in general rather constant in the region $N=82$ to 126. However, there are several minor departures from uniformity that should be pointed out. There appears to be an increase in the neutron separation energy of odd-N neutrons beyond $N=90$. For even-N neutron separation energies, there is a more or less general decline from $N=90$ to 110 followed by a discontinuity between $N=110$ and 112, with the values at $N=112$ and beyond somewhat larger than the values immediately below $N=112$. Because no other S_n values were available, the values at $N=115$ and 116 were calculated from nuclear-reaction Q values[8,9] for iridium,

[8] H. Kubitschek and S. M. Dancoff, Phys. Rev. **76**, 531 (1949).
[9] Sher, Halpern, and Mann, Phys. Rev. **84**, 387 (1951).

an odd-Z element. The S_n value at $N=115$ is considerably smaller than the neighboring values. This large departure cannot be attributed to the fact that the nucleus has odd-Z.

With the present data, it is possible to make a more complete study of the relation between neutron pairing energy and other nuclear properties in the region $N=82$ to 126. Many nuclear properties exhibit lack of uniformity in this region. The change from a vibrational to a rotational type of energy level spectrum takes place between $N=88$ and 90.[10] Also anomalies in the isotope shift[11] and electric quadrupole measurements[12] indicate a change in nuclear structure in the neighborhood of $N=90$. In a recent review article, Way *et al.*[13] have discussed several of these properties of nuclei. One of the properties that they investigate is the ratio of the energy of the second excited state to the energy of the first excited state. This property has previously been studied by Scharff-Goldhaber and Weneser[14] who found that the ratio is quite constant from $N=36$ to 90 at a value of about 2.2 but changes to about 3.3 above $N=90$. Way *et al.* have extended this analysis to show that the value of 3.3 is maintained from $N=90$ to 112 and decreases to about 2.2 again for $N=114$ to 134. For $N=136$ and higher, the value is again about 3.3. They state that a value of 3.3 is in agreement with that expected for rotational states. The prediction of a rotational-state region from $N=90$ to about 112 is also indicated in the study of systematics of the energy of the first 2+ state in even-even nuclei by Alder *et al.*[15] Their results show that excited state energies for the osmium nuclei at $N=110$, 112, and 114 are only slightly larger than the predicted maximum for a rotational state. The value for osmium at $N=116$ is considerably larger than the other three values. The experimental values for platinum at $N=114$, 116, 118, and 120 are much larger than the predicted maximum for rotational states.

Figure 2 illustrates neutron pairing energy calculated from the data appearing in Fig. 1 and plotted as a function of the neutron number. The P_n value at $N=116$, derived from nuclear reaction data for iridium, is the only odd-Z neutron pairing energy in the diagram. Pairing energy results in a lower mass region[16] suggest that one would expect an even-Z value at $N=116$ to be slightly larger than the present odd-Z value. The previously reported[2] increase in the neutron pairing energy in the neighborhood of $N=90$ is shown in Fig. 2. In addition, another maximum at $N=116$ is indicated

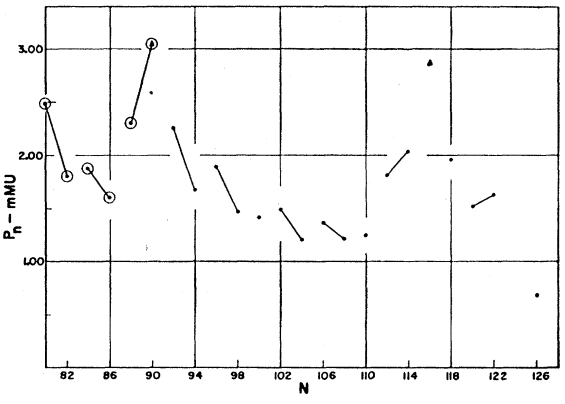

FIG. 2. Neutron pairing energy P_n plotted as a function of the neutron number N. Data from a given element are connected by a solid line. The circled results from $N=80$ to 90 are taken from reference 2. A result indicated by a triangle is calculated by using nuclear-reaction Q values or β-decay energies.

by the new results. The actual maximum at $N=116$ is the odd-Z value for iridium calculated from nuclear Q-value results. The neutron pairing energy value at $N=116$ is not required, however, for the conclusion that the neutron pairing energy becomes large in this area. Values at $N=112$, 114, and 118 from our measurements are also considerably higher than neighboring values. From a consideration of these results together with other nuclear properties we conclude that the neutron pairing energy is large where there is a transition from a nuclear model characterized by vibrational energy states to one characterized by rotational energy states. As neutrons are added and the shell is filled, we reach a point where the model changes from a rotational type to a vibrational type. Here the neutron pairing energy is again large. Because neutron separation energy and pairing energy are related quantities, it is clear that lack of uniformity near $N=90$ and 116 will also appear in the neutron separation energy results, as is observed in Fig. 1.

In order to have a better understanding of the nature of the maximum in the neutron pairing energy in the region near $N=116$, one must have considerably more data. A direct determination of stable atomic masses together with active atomic masses that could be calculated from reaction Q values and β-decay energies would provide a much more complete set of data. Confirmation of the low S_n value at $N=115$ would also be very valuable. It is doubtful, however, that further data will change our general conclusions about neutron pairing energy systematics in this region.

NUCLEAR REACTION ASSIGNMENTS

The present measured mass differences offer an independent verification of a number of nuclear-reaction Q values. Furthermore, in several cases we have been able to assign a target nucleus, with reasonable certainty, to reactions where the target nucleus was previously unassigned or assigned incorrectly.

[10] N. P. Heydenberg and G. M. Temmer, Phys. Rev. **100**, 150 (1955).
[11] P. Brix and H. Kopfermann, Phys. Rev. **85**, 1050 (1952).
[12] P. Brix, Z. Physik **132**, 579 (1952).
[13] Way, Kundu, McGinnis, and van Lieshout, *Annual Review of Nuclear Science* (Annual Reviews, Inc., Stanford, 1956), Vol. 6, p. 129.
[14] G. Scharff-Goldhaber and J. Weneser, Phys. Rev. **98**, 212 (1955).
[15] Alder, Bohr, Huus, Mottelson, and Winther, Revs. Modern Phys. **28**, 432 (1956).
[16] Quisenberry, Scolman, and Nier, Phys. Rev. **104**, 461 (1956).

Table II contains a comparison of a number of the present mass differences with similar mass differences derived from nuclear reactions. The $Gd^{157}(n,\gamma)Gd^{158}$ reaction, measured by Kubitschek and Dancoff[8] and used to determine the $Gd^{158}-Gd^{157}$ mass difference appears to be misassigned. One can compare their measured energy of 6.4±0.4 Mev with several predicted energies that can be determined from the present mass differences. These predicted Q values are listed in Table IV. The reaction energy of 6.4±0.4 Mev can be assigned to either $Gd^{154}(n,\gamma)Gd^{155}$ or $Gd^{156}(n,\gamma)Gd^{157}$ rather than to $Gd^{157}(n,\gamma)Gd^{158}$.

In the $Pt^{196}-Pt^{195}$ comparison in Table II, one observes that the agreement found between the measured mass difference and the mass differences calculated from the (n,γ) and the (d,p) reaction energies is very good. The (γ,n) reaction energy predicts a smaller mass difference. However, when one considers the error assigned to this reaction energy, this is really a minor disagreement. For the mass difference $Pb^{207}-Pb^{206}$ our results agree well with those calculated from the (n,γ), (d,p), and (d,t) reaction energies but disagrees with the mass difference derived from the (γ,n) reaction. The Q value for this reaction is a weighted average of four different measurements. The disagreement is somewhat reduced when one compares our value with the individual (γ,n) measurements having correspondingly larger errors.

Sher *et al.*[9] measured two thresholds for the (γ,n) reaction on wolfram. Threshold energies of 6.25±0.30 and 7.15±0.30 Mev were obtained. Table IV lists the two Q values that can be predicted from the present wolfram mass differences. The 6.25-Mev threshold should clearly be assigned to the reaction $W^{183}(\gamma,n)W^{182}$. The wolfram isotope at mass number 183 is the only odd-A, stable isotope. It is unlikely that the neutron separation energy for any stable, even-A wolfram isotope would be as low as 6.25 Mev. Comparison of the 7.15±0.30 Mev threshold with the values for wolfram reaction energies in Table IV suggests that this threshold energy possibly could be assigned to the $W^{184}(\gamma,n)W^{183}$ reaction. When one considers that in general the neutron separation energy in this region and therefore the γ-ray threshold decreases with an increase in the neutron number, it would seem more reasonable to assign the 7.15-Mev threshold to the $W^{186}(\gamma,n)W^{185}$ reaction.

A number of de-excitation γ-ray energies from the (n,γ) reaction for platinum and wolfram have been reported by Kinsey and Bartholomew.[17] We confirm their assignment of the 7.42±0.02 Mev γ ray to the $W^{183}(n,\gamma)W^{184}$ reaction; see Table IV. They assign the γ ray with an energy of 6.182±0.008 Mev to the reaction $W^{182}(n,\gamma)W^{183}$. Our measurements predict a value of 6.29±0.06 Mev for the ground state γ ray. This suggests that the 6.182-Mev γ ray may not be the ground-state transition. For platinum, they assign the 7.920 ±0.012 Mev γ ray to the $Pt^{195}(n,\gamma)Pt^{196}$ reaction. Our predicted energy for this reaction from Table IV, 7.91±0.06 Mev, confirms their assignment of this energy to the ground state reaction. They indicate that the γ ray with energy 6.07±0.04 Mev probably could be assigned to the ground-state transition in the reaction $Pt^{194}(n,\gamma)Pt^{195}$. The ground-state γ-ray energy, predicted from the present results, is 6.09±0.06 Mev. This excellent agreement suggests that their assignment is correct.

TABLE IV. Some selected nuclear-reaction Q values predicted from the present experimental mass differences.

Nuclear reaction	Predicted Q value Mev
$Gd^{154}(n,\gamma)Gd^{155}$	6.37±0.06
$Gd^{155}(n,\gamma)Gd^{156}$	8.45±0.06
$Gd^{156}(n,\gamma)Gd^{157}$	6.39±0.06
$Gd^{157}(n,\gamma)Gd^{158}$	7.88±0.06
$W^{183}(\gamma,n)W^{182}$ or $W^{182}(n,\gamma)W^{183}$	6.29±0.06
$W^{184}(\gamma,n)W^{183}$ or $W^{183}(n,\gamma)W^{184}$	7.45±0.06
$Pt^{194}(n,\gamma)Pt^{195}$	6.09±0.06
$Pt^{195}(n,\gamma)Pt^{196}$	7.91±0.06

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[17] B. B. Kinsey and G. A. Bartholomew, Can. J. Phys. 31, 1051 (1953).