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PROPOSED DETERMINATION OF NUCLEAR FUEL BURNUP
BASED ON THE RATIO OF TWO STABLE FISSION PRODUCTS
OF THE SAME ELEMENT

William J. Maack

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OF THE SAME ELEMENT

By

William J. Maeck

PHILLIPS
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ABSTRACT

A new method is proposed for the determination of nuclear fuel burnup based on the ratio of selected pairs of two fission product nuclides of the same element. The ratios Kr^{83}/Kr^{84} , Xe^{131}/Xe^{132} , and Nd^{143}/Nd^{144} , which change with irradiation are useful as fission monitors. With this technique, many problems associated with the conventional fission product analysis for burnup are eliminated.

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PROPOSED DETERMINATION OF NUCLEAR FUEL BURNUP BASED ON THE RATIO OF TWO STABLE FISSION PRODUCTS OF THE SAME NUCLID

by

William J. Maeck

I. INTRODUCTION

Presently, the determination of nuclear fuel burnup by fission product analysis is based on the quantitative determination of a selected fission product nuclide. The number of fissions is calculated from the determined total atom abundance of that nuclide divided by its fission yield. The accuracy of this method depends on several nuclear properties of the nuclide as well as several chemical properties.

One of the necessary nuclear properties is that the number of atoms of the nuclide present at the end of irradiation is directly proportional to the number of fissions. In the following, simplified, production scheme of several fission product nuclides of common Z, the abundance of the A nuclide is directly proportional to fissions only if the capture cross sections of the A-1 and the A nuclide are insignificant. The effect of the neutron capture cross section upon

the production of some selected A nuclide is shown in the next figure. If the cross section of A-1 is large, the relative production of A increases with fissions. Conversely, if the capture cross section of A to form A+1 is large, the relative production of A decreases with fissions.

Another nuclear property of major importance is the effect of neutron energy on fission yields. The wide variations in the fission yields of U^{233} , U^{235} , and Pu^{239} for thermal neutrons and fission neutrons are shown in Fig. 1, 2, and 3. These values were obtained from a recent compilation (10) of estimated total chain fission yields and are estimated to be reliable to ± 10 percent.

Even if a nuclide is used whose production is directly proportional to fissions, that is $\sigma_C^A - 1$ and σ_C^A are zero, chemical factors must be considered. The sample must completely dissolve to give a true homogeneous solution and the analysis for the abundance of A must be quantitative. These requirements are becoming more difficult to meet as fuel element technology advances. This is especially true for the corrosion resistant metals and the refractory or ceramic-clad materials. Even the storage of dissolved fission product solutions introduces problems of wall adsorption and plating (3,8).

In general, recent advances and current explorations in burnup analysis have been restricted to refinements in the measurement techniques for the fission product analysis rather than in new concepts. While recent advances in mass-spectrometric isotope-dilution analysis for stable fission products are certainly valuable, effort also should be devoted to new concepts which might avoid some of the current difficulties.

In this report, a new concept of burnup analysis is proposed which is based on abundance ratios of selected fission product nuclides of the same element.

II. PROPOSED CONCEPT FOR DETERMINATION OF BURNUP BASED ON RATIOS OF FISSION PRODUCT NUCLIDES

A. Basic Premise

For the same simplified model of fission product production shown on page 1, where the nuclides A-1, A, and A+1 are of the same element, assume that the fissionable source is monoisotopic, the flux is isotropic, and the flux level is constant during the irradiation period. Now consider the case where the capture cross section of A is large and that of A+1 is small. (This is the very

condition that is unsuitable for the conventional determination of burnup which is based on the quantitative measurement of a single fission product nuclide. It is unsuitable because, as the irradiation proceeds, some A nuclides capture neutrons to form A+1 nuclides. This results in the abundance of A+1 being higher and that of A being lower than that calculated from direct chain fission yields.) Rather than considering the individual behavior of these nuclides, consider the ratio A+1/A. The result, as shown to the right, is an exponential function with the ratio increasing relative to irradiation time and hence to fissions.

If this concept of ratio measurement is useful for burnup analysis, several problems associated with the conventional chemical burnup determination are circumvented.

1. An absolute atom abundance measurement is not required; only a ratio is determined.
2. Volume measurements, difficult to do with highly radioactive samples, are not required.
3. Complete sample dissolution, while desirable, is not mandatory because only the isotopic composition of a selected fission product element is measured.

B. Evaluation and Computer Data

In evaluating this proposal, five pairs of unshielded fission product nuclides were found for which the thermal capture cross section of the A nuclide is greater than 100 barns and that of A+1 is small. These are:

1. $\text{Kr}^{83} \xrightarrow[220]{\sigma} \text{Kr}^{84} \xrightarrow[1]{\sigma}$
2. $\text{Cd}^{113} \xrightarrow[27,000]{\sigma} \text{Cd}^{114} \xrightarrow[1.2]{\sigma}$
3. $\text{Xe}^{131} \xrightarrow[120]{\sigma} \text{Xe}^{132} \xrightarrow[5]{\sigma}$
4. $\text{Nd}^{143} \xrightarrow[330]{\sigma} \text{Nd}^{144} \xrightarrow[5]{\sigma}$
5. $\text{Sm}^{149} \xrightarrow[41,500]{\sigma} \text{Sm}^{150} \xrightarrow[?]{\sigma}$

Of these, the isotopes of krypton, xenon, and neodymium appear useful as fission monitors. Cadmium is in a portion of the mass yield curve for U^{235} , which is strongly dependent on neutron energy (Figure 2). Samarium-149, with its high capture cross section, may prove useful as a monitor for fuels of low burnup; however, the reaction rate of Sm^{149} , n_{th} , Sm^{150} is strongly energy dependent due to its sharp, thermal cross section cutoff.

To further evaluate the basic proposal, the buildup and burn-out of the nuclides of krypton, xenon, and neodymium as a function of depletion in thermal fission were calculated by a computer program (5). The input data are given in Figures 4-6. The cross section values were obtained from the Nuklidkarte (2) and the thermal fission yields of U^{235} and Pu^{239} from Katcoff (7). Yields for the thermal fission of U^{233} were taken from Ferguson and O'Kelly (4).

The machine calculations were made at thermal fluxes of 5×10^{12} and 1×10^{14} n-cm⁻²-sec⁻¹ for xenon and krypton. For neodymium, flux levels of 1×10^{13} and 1×10^{14} n-cm⁻²-sec⁻¹ were used. Exposure levels to 1×10^{23} atoms of fuel were selected to give fissionable isotopic depletions ranging from 1.5% to 90%. The output data are given in Tables 1-12.

The ratios of the fission product nuclide pairs, $\text{Kr}^{84}/\text{Kr}^{83}$, $\text{Xe}^{132}/\text{Xe}^{131}$, and $\text{Nd}^{144}/\text{Nd}^{143}$, are given in Tables 1-12 as a function of depletion for thermally fissioned U^{233} , U^{235} , and Pu^{239} , and also in Figures 7 and 8 for thermally fissioned U^{235} . The values for Xe^{131} include the holdup in the decay of the 8-day I^{131} isotope and those of neodymium include the Nd^{144} formed from decay of the 285-day Ce^{144} isotope and the Nd^{143} formed from 13.7-day Pr^{143} . The calculation for the Nd^{144} abundance is at a time of 285-days ($t_{1/2}$ of Ce^{144}) which is a reasonable cooling period for highly irradiated fuel.

A data compilation of percent change in the selected ratios as a function of depletion is also given in Tables 1-12. These data are plotted in Figures 9 and 10 for U^{235} . This calculation is based on instantaneous fission yield values obtained as either a direct ratio of the fission yields or as an extrapolation of the ratios to zero depletion.

C. Sensitivity and Measurement

As expected, the sensitivity of the ratio of the various nuclide pairs is dependent upon the cross section of the capturing nuclides. Thus, the change in the ratios as a function of depletion is $\text{Nd}^{144}/\text{Nd}^{143} > \text{Kr}^{84}/\text{Kr}^{83} > \text{Xe}^{132}/\text{Xe}^{131}$. The abnormally large change in the $\text{Nd}^{144}/\text{Nd}^{143}$ ratio is due to the 285-day holdup of Ce^{144} .

The nuclide pairs selected for evaluation consist of stable nuclides which can be mass spectrometrically determined, circumventing errors associated with radiochemically determined nuclides such as half-life and decay schemes. Based on recent literature data (1), new ratio-measuring analytical mass spectrometers are capable of a relative accuracy of 0.01% for ratio range of 1:1 to 1:100 when determining gases. At a 10% depletion level of U^{235} , the ratio of $\text{Kr}^{84}/\text{Kr}^{83}$ increases approximately 3% from zero depletion; however, a 0.01% measurement error should result in less than 1% relative error in the depletion value.

D. Flux Effect

The effect of neutron flux (fission rate) has been evaluated by programing the production and burnout of xenon, krypton, and neodymium fission product nuclides at two different flux levels. The output data are tabulated in Tables 1-12. The $\text{Kr}^{84}/\text{Kr}^{83}$ and $\text{Xe}^{132}/\text{Xe}^{131}$ ratios are essentially independent of flux level as shown in Figure 11.

The $\text{Nd}^{144}/\text{Nd}^{143}$ ratio is significantly flux dependent (Figure 12). This effect is primarily due to the holdup by 285-day Ce in the 144 mass chain.

Another isotope pair with a large flux dependency is $\text{Xe}^{136}/\text{Xe}^{134}$. This particular ratio, however, is constant with U^{235} depletion at a specific flux level (Figure 12) because neither isotope has a significant capture cross section. The dependency of the $\text{Xe}^{136}/\text{Xe}^{134}$ ratio on the thermal flux level is shown in Figure 13. In addition to the data obtained from the calculations in this program, additional data points are included which were calculated from the work of Weber (11) for fission gas production as a function of flux. The close agreement of the two sets of data is shown in Figure 13. The value of the $\text{Xe}^{136}/\text{Xe}^{134}$ ratio reaches a maximum and then decreases only at flux levels higher than those normally encountered in power reactor operation.

Because the $\text{Xe}^{136}/\text{Xe}^{134}$ ratio is independent of depletion for a specific fuel and is flux level dependent, it is worth considering for use as a monitor for average flux level. Serious consideration, however, must be given to the production of Xe^{136} from Xe^{135} because the sharp cutoff of the high thermal cross section for Xe^{135} could affect the ratio in regions of shifting neutron spectra.

E. Spectrum Effect

There appear to be no strong resonance peaks for Xe^{131} , Xe^{132} , Kr^{83} , Kr^{84} , Nd^{143} , or Nd^{144} in the thermal or near thermal neutron energy region (6). The first strong resonance peak for krypton is 520 barns at 28eV, but as yet is unassociated with a specific isotope. For xenon, the first strong resonance peak is 10,000 barns for Xe^{131} at 14eV. The principal effect of this resonance peak in a faster than thermal reactor would be to increase the sensitivity of the $\text{Xe}^{132}/\text{Xe}^{131}$ ratio with respect to depletion. The situation is similar with neodymium. The first strong resonance for Nd^{143} is 400 barns at 56eV. Generally, there should be no strong spectrum effects upon any of these ratios for well moderated thermal reactors. For epithermal reactors, the problem of resonance capture and spectrum effect is present with this proposed method of determining nuclear fuel burnup, as it is with other currently used methods. Experimental data are required to establish the magnitude of the problems.

An additional consideration would be to search for a pair of stable fission product nuclides which might conceivably serve to describe flux hardness based on ratio changes. One nuclide of the pair should have a high resonance and a low thermal cross section and the other an insignificant resonance cross section.

F. Use as Differentiating Indicators

In the operation of low-enrichment, uranium-fueled reactors, it is often desirable to know the fraction of the total fissions resulting from the various fissioning sources. Generally, 90% or more of the fission result from the thermal fission of U^{235} and Pu^{239} . The balance is due to the fast fission of U^{238} and Pu^{240} . The thermal Pu^{241} fission fraction is small.

One proposed method (9) of differentiating the two major sources of fission is to observe the changes in the fission yields of those nuclides whose yields vary as a function of the fissioning isotopes. Changes in the fission yields as a function of the fissioning nuclide are shown in Figures 14, 15, and 16.

One ratio which has been suggested to differentiate U^{235} and Pu^{239} fissioning is mass number 90 to 107. The thermal fission yields of this ratio vary from 30 for pure U^{235} fission to 0.65 for Pu^{239} fission. While this ratio is indeed large, the yield of mass 107 is highly energy dependent (Figure 2). This must be considered for reactors whose neutron spectra are variable. A second consideration is that an absolute measurement of the mass 90 and mass 107 isotopes must be made.

Examination of Figure 14, which gives the fission yield ratios of the thermal fissioning of U^{235} with respect to the thermal fission of Pu^{239} , indicates that the yield ratios of the xenon isotopes are relatively independent of the fissioning source while a significant difference is observed in the region of the krypton isotopes.

The isotopes Xe^{134} and Kr^{86} , which have small capture cross sections, give a ratio which is independent of depletion and varies with the fissioning nuclides (Figure 17). For the thermal fission of U^{235} , the Xe^{134}/Kr^{86} ratio has a value of 3.99 compared to 9.7 for the thermal fission of Pu^{239} . Thus, for a low-enrichment fuel, ratio value between 3.99 and 9.7 should describe the fraction of fissions occurring from the two primary fission sources. To obtain this ratio, absolute amounts of Xe^{134} and Kr^{86} must be determined in the sample analyzed.

A better ratio for use as a differentiating monitor is Nd^{148}/Nd^{150} (Figure 18). The ratio value for the thermal fission of U^{235} is 2.55, while that for the thermal fission of Pu^{239} is 1.71. This ratio is independent of burnup and does not require a quantitative determination of two elements.

A computer program is being composed for a low-enrichment fuel to calculate the changes in the $\text{Nd}^{148}/\text{Nd}^{150}$ ratio with respect to the fraction of fissions occurring in each fissioning nuclide.

III. EXPERIMENTAL

To experimentally evaluate the usefulness of fission product isotopic ratios for the determination of burnup, an extensive irradiation program has been initiated in the Materials Testing Reactor.

A series of 36 capsules containing known amounts of U^{233} , U^{235} , Pu^{239} , and Pu^{241} as oxides was prepared. The oxides were weighed into high-purity aluminum capsules which were sealed by welding. The capsules are being irradiated for varying periods of time to give burnups ranging from approximately 8 to 60%. After cooling for 3 to 4 months, the capsules will be dissolved under controlled conditions. The krypton and xenon will be quantitatively collected and analyzed for total abundance and isotopic ratios. A combination carrier-spike of Nd^{142} will be added to the dissolution flask prior to dissolution. Fissions will be calculated by the loss of the heavy elements based on mass spectrometric isotope dilution technique.

The first series of capsules was discharged from the reactor in April, 1964. The final series will be discharged in January, 1965. The capsules are in the A-28 SE position of the MTR which has a thermal flux of 6×10^{15} n-cm²-sec⁻¹ and a cadmium ratio for cobalt of approximately 15.

IV. RECOMMENDATIONS

Based on the machine computations, used to describe the usefulness of fission product isotope pairs as fission monitors, an extensive irradiation program is recommended. Standard burnup samples should be placed in several different reactors of varying flux environments to adequately evaluate the proposal. In addition to the thermal MTR irradiations, in progress in this laboratory, samples should be placed in a fast reactor such as EBR II as well as in several of the power reactors in which a high fraction of the flux is resonance. This approach would facilitate the evaluation of such variables as flux and spectrum effects.

Until such data are available, it is recommended that each reactor should be calibrated with a set of standards.

V. ACKNOWLEDGEMENT

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Table 1. Buildup and Burnout of Krypton and Xenon
 Fuel - 1×10^{23} atoms U^{235} Flux - 5×10^{12} n-cm⁻²sec⁻¹

ATOMS PRESENT

IRRADIATION TIME	0.1 yr	0.5 yr	1.0 yr	3.0 yr	5.0 yr	10.0 yr
% DEPLETION	1.08	5.28	10.28	27.78	41.86	66.20
Nuclide						
Kr ⁸³	4.940 E18 ^a	2.400 E19	4.633 E19	1.207 E20	1.749 E20	2.487 E20
Kr ⁸⁴	9.105 E18	4.472 E19	8.749 E19	2.409 E20	3.700 E20	6.129 E20
Kr ⁸⁶	1.837 E19	8.991 E19	1.751 E20	4.732 E20	7.133 E20	1.129 E21
Kr ⁸⁴ /Kr ⁸⁶ ratio	0.496	0.497	0.500	0.509	0.519	0.543
Kr ⁸⁴ /Kr ⁸³ ratio	1.843	1.863	1.889	1.997	2.116	2.464
% change in Kr ⁸⁴ /Kr ⁸³ ratio ($t_0 = 1.838$)	0.273	1.36	2.77	8.65	15.12	34.06
Xe ¹³¹	1.962 E19	1.288 E20	2.581 E20	6.984 E20	1.035 E21	1.547 E21
I ¹³¹	0.856 E19	0.086 E20	0.081 E20	0.065 E20	0.005 E21	0.003 E21
Total - 131	2.818 E19	1.374 E20	2.662 E20	7.049 E20	1.040 E21	1.550 E21
Total - 132	4.277 E19	2.097 E20	4.097 E20	1.121 E20	1.711 E21	2.795 E21
Xe ¹³⁴	7.334 E19	3.589 E20	6.987 E20	1.887 E21	2.843 E21	4.488 E21
Xe ¹³⁶	8.101 E19	3.972 E20	7.736 E20	2.091 E21	3.151 E21	4.983 E21
Xe ¹³⁶ /Xe ¹³⁴ ratio	1.105	1.107	1.107	1.108	1.108	1.110
Xe ¹³² /Xe ¹³¹ ratio	1.518	1.526	1.551	1.590	1.645	1.803
% change in Xe ¹³² /Xe ¹³¹ ratio ($t_0 = 1.516$)	0.132	0.660	2.31	4.88	8.51	18.9

^aEquivalent to 4.940×10^{18}

Table 2. Buildup and Burnout of Krypton and Xenon
 Fuel - 1×10^{23} atoms U^{235} Flux - 1×10^{14} n-cm⁻²sec⁻¹

ATCMS PRESENT

IRRADIATION TIME	0.1 yr	0.5 yr	1.0 yr
% DEPLETION	19.5	66.2	88.6
Nuclide			
Kr ⁸³	8.631 E19 ^a	2.487 E20	2.599 E20
Kr ⁸⁴	1.676 E20	6.129 E20	8.928 E20
Kr ⁸⁶	3.322 E20	1.128 E21	1.513 E21
Kr ⁸⁴ /Kr ⁸⁶ ratio	0.505	0.543	0.590
Kr ⁸⁴ /Kr ⁸³ ratio	1.942	2.464	3.435
% change in Kr ⁸⁴ /Kr ⁸³ ratio ($t_0 = 1.838$)	5.66	34.0	86.9
Xe ¹³¹	3.554 E20	1.495 E21	1.797 E21
I ¹³¹	1.470 E20	6.515 E19	2.202 E19
Total - 131	5.024 E20	1.560 E21	1.819 E21
Total - 132	7.798 E20	2.785 E21	3.981 E21
Xe ¹³⁴	1.336 E21	4.530 E21	1.604 E21
Xe ¹³⁶	2.023 E21	6.867 E21	9.187 E21
Xe ¹³⁶ /Xe ¹³⁴ ratio	1.514	1.516	5.727
Xe ¹³² /Xe ¹³¹ ratio	1.552	1.785	2.189
% change in Xe ¹³² /Xe ¹³¹ ratio ($t_0 = 1.516$)	2.37	17.7	44.4

^aEquivalent to 8.631×10^{19}

Table 3. Buildup and Burnout of Neodymium
 Fuel - 1×10^{23} atoms U^{235} Flux - 1×10^{13} n-cm⁻²sec⁻¹

ATOMS PRESENT						
IRRADIATION TIME	0.1 yr	0.5 yr	1.0 yr	3.0 yr	5.0 yr	10.0 yr
% DEPLETION	2.15	10.28	19.50	47.84	66.20	88.58
Nuclide						
Pr ¹⁴³	4.944 E19 ^a	5.399 E19	4.843 E19	3.139 E19	2.034 E19	6.874 E18
Nd ¹⁴³	5.905 E19	4.555 E20	8.927 E20	2.026 E21	2.488 E21	2.331 E21
Total - ¹⁴³	1.085 E20	5.095 E20	9.411 E20	2.057 E21	2.508 E21	2.338 E21
Ce ¹⁴⁴	9.702 E19	3.906 E20	6.014 E20	6.905 E20	4.987 E20	1.743 E20
Nd ¹⁴⁴	4.707 E18	1.067 E20	3.680 E20	1.935 E21	3.466 E21	6.111 E21
Total - ¹⁴⁴ , 285 d. after shutdown	5.322 E19	3.020 E20	6.687 E20	2.280 E21	3.716 E21	6.198 E21
Nd ¹⁴⁵	7.177 E19	3.427 E20	6.477 E20	1.563 E21	2.127 E21	2.718 E21
Nd ¹⁴⁶	5.547 E19	2.667 E20	5.084 E20	1.275 E21	1.806 E21	2.573 E21
Nd ¹⁴⁸	3.086 E19	1.477 E20	2.802 E20	6.866 E20	9.484 E20	1.263 E21
Nd ¹⁵⁰	1.209 E19	5.792 E19	1.098 E20	2.693 E20	3.724 E20	4.975 E20
Nd ¹⁴⁶ /Nd ¹⁴³ ratio 285 d. after shut- down	0.511	0.523	0.540	0.620	0.720	1.100
% change in Nd ¹⁴⁶ /Nd ¹⁴³ ratio ($t_0 = 0.510$)	0.20	2.55	5.88	21.6	41.2	116
Nd ¹⁴⁶ /Nd ¹⁴⁵ ratio	0.773	0.778	0.785	0.816	0.849	0.947
Nd ¹⁴⁸ /Nd ¹⁵⁰ ratio	2.553	2.550	2.552	2.557	2.547	2.539
Nd ¹⁴⁴ /Nd ¹⁴³ ratio 285 d. after shut- down	0.491	0.593	0.711	1.108	1.482	2.651
% change in Nd ¹⁴⁴ /Nd ¹⁴³ ratio ($t_0 = 0.465$)	5.59	27.5	52.9	138	219	470

^aEquivalent to 4.944×10^{19}

Table 4. Buildup and Burnout of Neodymium
 Fuel - 1×10^{23} atoms U^{235} Flux - 1×10^{14} n-cm⁻²sec⁻¹

ATOMS PRESENT			
IFRADIATION TIME	0.1 yr	0.5 yr	1.0 yr
% DEPLETION	19.50	66.20	88.58
Nuclide			
Pr ¹⁴³	4.322 E20 ^a	2.242 E20	7.578 E19
Nd ¹⁴³	5.267 E20	2.371 E21	2.349 E21
Total - ¹⁴³	9.589 E20	2.595 E21	2.425 E21
Ce ¹⁴⁴	8.803 E20	2.428 E21	2.378 E21
Nd ¹⁴⁴	7.084 E19	1.457 E21	3.847 E21
Total - 144, 285 d. after shutdown			
Nd ¹⁴⁵	6.473 E20	2.120 E21	2.694 E21
Nd ¹⁴⁶	5.084 E20	1.806 E21	2.571 E21
Nd ¹⁴⁸	2.802 E20	9.484 E20	1.263 E21
Nd ¹⁵⁰	1.098 E20	3.724 E20	4.975 E20
Nd ¹⁴⁵ /Nd ¹⁴³ 285 d. after shutdown	0.530	0.696	1.060
% change in Nd ¹⁴⁶ /Nd ¹⁴³ ratio ($t_0 = 0.505$)	4.95	37.8	110
Nd ¹⁴⁶ /Nd ¹⁴⁵ ratio	0.785	0.852	0.954
Nd ¹⁴⁸ /Nd ¹⁵⁰ ratio	2.552	2.547	2.539
Nd ¹⁴⁴ /Nd ¹⁴³ 285 d. after shutdown	0.533	1.029	2.077
% change in Nd ¹⁴⁴ /Nd ¹⁴³ ratio ($t_0 = 0.365$)	46.0	182	469

^aEquivalent to 4.322×10^{20}

Table 5. Buildup and Burnout of Krypton and Xenon
 Fuel - 10^{23} atoms Pu^{239} Flux - 5×10^{12} n-cm⁻²sec⁻¹

ATOMS PRESENT

IRRADIATION TIME	0.1 yr	0.5 yr	1.0 yr	3.0 yr	5.0 yr	10.0 yr
% DEPLETION	1.59	7.70	14.80	38.16	55.11	79.85
Nuclide						
Kr^{83}	3.378 E18 ^a	1.625 E19	3.097 E19	7.685 E19	1.065 E20	1.374 E20
Kr^{84}	5.490 E18	2.671 E19	5.164 E19	1.361 E20	2.010 E20	3.082 E20
Kr^{86}	8.869 E18	4.296 E19	8.262 E19	2.131 E20	3.078 E20	4.463 E20
$\text{Kr}^{84}/\text{Kr}^{86}$ ratio	0.619	0.622	0.625	0.639	0.653	0.691
$\text{Kr}^{84}/\text{Kr}^{83}$ ratio	1.625	1.644	1.667	1.771	1.887	2.243
% change in $\text{Kr}^{84}/\text{Kr}^{83}$ ratio ($t_0 = 1.621$)	0.247	1.42	2.84	9.25	16.4	38.4
Xe^{131}	3.070 E19	1.996 E20	3.949 E20	1.019 E21	1.445 E21	1.968 E21
I^{131}	1.388 E19	1.312 E19	1.211 E19	8.792 E18	6.382 E18	2.865 E18
Total - 131	4.458 E19	2.127 E20	4.070 E20	1.028 E21	1.451 E21	1.971 E21
Total - 132	6.141 E19	2.982 E20	5.754 E20	1.504 E21	2.204 E21	3.319 E21
Xe^{134}	8.721 E19	4.224 E20	8.122 E20	2.093 E21	3.021 E21	4.369 E21
Xe^{136}	1.094 E20	5.313 E20	1.022 E21	2.635 E21	3.805 E21	5.513 E21
$\text{Xe}^{136}/\text{Xe}^{134}$ ratio	1.254	1.258	1.258	1.259	1.259	1.262
$\text{Xe}^{132}/\text{Xe}^{131}$ ratio	1.377	1.402	1.414	1.463	1.519	1.684
% change in $\text{Xe}^{132}/\text{Xe}^{131}$ ratio ($t_0 = 1.375$)	0.14	1.96	2.84	6.40	10.5	22.5

^aEquivalent to 3.378×10^{18}

Table 6. Buildup and Burnout of Krypton and Xenon
 Fuel - 10^{23} atoms Pu^{239} Flux - 1×10^{14} n-cm⁻²sec⁻¹

ATOMS PRESENT

IRRADIATION TIME	5 day	10 day	20 day	50 day	80 day	150 day
% DEPLETION	4.29	8.40	16.10	35.53	50.45	73.20
Nuclide						
Kr^{83}	9.100 E18 ^a	1.772 E19	3.362 E19	7.191 E19	9.878 E19	1.315 E20
Kr^{84}	1.486 E19	2.917 E19	5.623 E19	1.263 E20	1.828 E20	2.769 E20
Kr^{86}	2.396 E19	4.690 E19	8.987 E19	1.983 E20	2.818 E20	4.091 E20
$\text{Kr}^{84}/\text{Kr}^{86}$ ratio	0.620	0.622	0.626	0.637	0.649	0.677
$\text{Kr}^{84}/\text{Kr}^{83}$ ratio	1.633	1.646	1.673	1.756	1.851	2.106
% change in $\text{Kr}^{84}/\text{Kr}^{83}$ ratio ($t_0 = 1.621$)	0.74	1.54	3.21	3.33	14.2	29.9
Xe^{131}	2.352 E19	9.532 E19	2.595 E20	7.876 E20	1.206 E21	1.786 E21
I^{131}	9.853 E19	1.659 E20	2.103 E20	1.960 E20	1.532 E20	8.303 E19
Total - 131	1.220 E20	2.612 E20	4.698 E20	9.836 E20	1.359 E21	1.869 E21
Total - 132	1.702 E20	3.655 E20	6.620 E20	1.449 E21	2.024 E21	3.003 E21
Xe^{134}	2.443 E20	5.207 E20	9.423 E20	2.007 E21	2.824 E21	4.066 E21
Xe^{136}	4.252 E20	9.137 E20	1.651 E21	3.513 E21	4.943 E21	7.123 E21
$\text{Xe}^{136}/\text{Xe}^{134}$ ratio	1.740	1.755	1.752	1.750	1.750	1.752
$\text{Xe}^{132}/\text{Xe}^{131}$ ratio	1.395	1.399	1.409	1.473	1.489	1.607
% change in $\text{Xe}^{132}/\text{Xe}^{131}$ ratio ($t_0 = 1.390$)	0.36	0.65	1.37	5.97	7.12	15.6

^aEquivalent to 9.100×10^{18}

Table 7. Buildup and Burnout of Neodymium
 Fuel - 10^{23} atoms Pu²³⁹ Flux - 1×10^{13} n-cm⁻²sec⁻¹

ATOMS. PRESENT						
IRRADIATION TIME	0.1 yr	0.5 yr	1.0 yr	3.0 yr	5.0 yr	10.0 yr
% DEPLETION	3.15	14.80	27.41	61.76	79.85	95.94
Nuclide						
Pr ¹⁴³	4.708 E19 ^a	4.941 E19	4.213 E19	2.219 E19	1.169 E19	2.356 E18
Nd ¹⁴³	5.642 E19	4.268 E20	8.159 E20	1.688 E21	1.911 E21	1.528 E21
Total - ¹⁴³	1.035 E20	4.762 E20	8.580 E20	1.710 E21	1.923 E21	1.530 E21
Ce ¹⁴⁴	8.539 E19	3.364 E20	5.023 E20	5.000 E20	3.032 E20	6.468 E19
Nd ¹⁴⁴	4.170 E18	9.361 E19	3.177 E20	1.567 E21	2.653 E21	4.240 E21
Total - ¹⁴⁴ , 285 d. after shutdown	4.687 E19	2.618 E20	5.689 E20	1.817 E21	2.804 E21	4.272 E21
Nd ¹⁴⁵	7.103 E19	3.324 E20	6.129 E20	1.358 E21	1.723 E21	1.965 E21
Nd ¹⁴⁶	5.880 E19	2.785 E20	5.183 E20	1.193 E21	1.579 E21	2.024 E21
Nd ¹⁴⁸	3.929 E19	1.844 E20	3.413 E20	7.679 E20	9.910 E20	1.184 E21
Nd ¹⁵⁰	2.294 E19	1.077 E20	1.994 E20	4.490 E20	5.798 E20	6.955 E20
Nd ¹⁴⁶ /Nd ¹⁴³ 285 d. after shutdown	0.568	0.585	0.604	0.698	0.819	1.323
% change in Nd ¹⁴⁶ /Nd ¹⁴³ ratio ($t_0 = 0.560$)	1.43	3.12	7.86	24.6	46.2	136
Nd ¹⁴⁸ /Nd ¹⁵⁰ ratio	1.713	1.712	1.712	1.710	1.709	1.702
Nd ¹⁴⁴ /Nd ¹⁴³ 285 d. after shutdown	0.453	0.550	0.663	1.063	1.458	2.792
% change in Nd ¹⁴⁴ /Nd ¹⁴³ ratio ($t_0 = 0.422$)	7.35	30.6	57.1	152	246	562

^aEquivalent to 4.708×10^{19}

Table 8. Buildup and Burnout of Neodymium
 Fuel - 10^{23} atoms Pu²³⁹ Flux - 1×10^{14} n-cm⁻²sec⁻¹

ATOMS PRESENT			
IRRADIATION TIME	0.1 yr	0.5 yr	1.0 yr
% DEPLETION	27.41	79.85	95.94
Nuclide			
Pr ¹⁴³	3.884 E2C ^a	1.367 E20	2.755 E19
Nd ¹⁴³	4.864 E2C	1.855 E21	1.561 E21
Total - ¹⁴³	8.748 E2C	2.092 E21	1.588 E21
Ce ¹⁴⁴	7.406 E20	1.723 E21	1.452 E21
Nd ¹⁴⁴	6.285 E19	1.170 E21	2.813 E21
Total - ¹⁴⁴ , 285 d. after shutdown	4.331 E20	2.031 E21	3.539 E21
Nd ¹⁴⁵	6.129 E20	1.717 E21	1.947 E21
Nd ¹⁴⁶	5.183 E20	1.579 E21	2.023 E21
Nd ¹⁴⁸	3.413 E20	9.910 E20	1.204 E21
Nd ¹⁵⁰	1.994 E20	5.798 E20	7.181 E20
Nd ¹⁴⁶ /Nd ¹⁴³ 285 d. after shutdown	0.592	0.755	1.274
% change in Nd ¹⁴⁶ /Nd ¹⁴³ ratio ($t_0 = 0.565$)	4.78	33.6	125
Nd ¹⁴⁸ /Nd ¹⁵⁰ ratio	1.712	1.709	1.677
Nd ¹⁴⁴ /Nd ¹⁴³ 285 d. after shutdown	0.495	0.971	2.229
% change in Nd ¹⁴⁴ /Nd ¹⁴³ ratio ($t_0 = 0.310$)	59.7	216	619

^aEquivalent to 3.884×10^{20}

Table 9. Buildup and Burnout of Krypton and Xenon
 Fuel - 1×10^{23} atoms U^{235} Flux - 5×10^{12} n-cm⁻²sec⁻¹

ATOMS PRESENT						
IRRADIATION TIME	0.1 yr	0.5 yr	1.0 yr	3.0 yr	5.0 yr	10.0 yr
% DEPLETION	0.91	4.45	8.71	23.92	36.60	59.80
Nuclide						
Kr ⁸³	9.625 E18 ^a	4.693 E19	9.096 E19	2.409 E20	3.547 E20	5.232 E20
Kr ⁸⁴	1.609 E19	7.931 E19	1.559 E20	4.370 E20	6.824 E20	1.171 E21
Kr ⁸⁶	2.695 E19	1.323 E20	2.588 E20	7.110 E20	1.088 E20	1.779 E21
Kr ⁸⁴ /Kr ⁸⁶ ratio	0.597	0.599	0.602	0.615	0.627	0.658
Kr ⁸⁴ /Kr ⁸³ ratio	1.672	1.690	1.714	1.814	1.924	2.238
% change in Kr ⁸⁴ /Kr ⁸³ ratio ($t_0 = 1.668$)	0.24	1.32	2.76	8.75	15.3	34.2
Xe ¹³¹	1.943 E19	1.280 E20	2.576 E20	7.086 E20	1.066 E21	1.651 E21
I ¹³¹	8.486 E18	8.555 E18	8.174 E18	6.812 E18	5.677 E18	3.599 E18
Total - 131	2.792 E19	1.365 E20	2.658 E20	7.154 E20	1.072 E21	1.655 E21
Total - 132	3.825 E19	1.883 E20	3.695 E20	1.029 E21	1.596 E21	2.700 E21
Xe ¹³⁴	4.905 E19	2.409 E20	4.709 E20	1.293 E21	1.977 E21	3.225 E21
Xe ¹³⁶	7.396 E19	3.639 E20	7.117 E20	1.955 E21	2.991 E21	4.886 E21
Xe ¹³⁶ /Xe ¹³⁴ ratio	1.508	1.510	1.511	1.512	1.513	1.515
Xe ¹³² /Xe ¹³¹ ratio	1.370	1.379	1.390	1.438	1.489	1.631
% change in Xe ¹³² /Xe ¹³¹ ratio ($t_0 = 1.366$)	0.29	0.95	1.76	5.27	9.00	19.4

^aEquivalent to 9.625×10^{18}

Table 10. Buildup and Burnout of Krypton and Xenon
 Fuel - 1×10^{23} atoms U^{235} Flux - 1×10^{14} n-cm⁻²sec⁻¹

ATOMS PRESENT

IRRADIATION TIME	5 day	10 day	20 day	50 day	80 day	150 day
% DEPLETION	2.47	4.87	9.51	22.10	32.94	52.72
Nuclide						
Kr ⁸³	2.608 E19 ^a	5.127 E19	9.909 E19	2.236 E20	3.231 E20	4.789 E20
Kr ⁸⁴	4.380 E19	8.677 E19	1.703 E20	4.026 E20	6.103 E20	1.015 E21
Kr ⁸⁶	7.325 E19	1.447 E20	2.824 E20	6.567 E20	9.791 E20	1.569 E21
Kr ⁸⁴ /Kr ⁸⁶ ratio	0.598	0.600	0.603	0.613	0.623	0.647
Kr ⁸⁴ /Kr ⁸³ ratio	1.679	1.692	1.719	1.801	1.889	2.119
% change in Kr ⁸⁴ /Kr ⁸³ ratio ($t_0 = 1.668$)	0.66	1.44	3.06	7.97	13.3	27.0
IRRADIATION TIME	5.13 day	11.3 day	21.3 day	51.3 day	81.3 day	151.3 day
% DEPLETION	2.53	5.49	10.09	22.60	33.37	53.03
Nuclide						
Xe ¹³¹	1.495 E19	6.108 E19	1.687 E20	5.370 E20	8.632 E20	1.418 E21
I ¹³¹	6.283 E19	1.073 E20	1.397 E20	1.437 E20	1.255 E20	8.859 E19
Total - 131	7.778 E19	1.684 E20	3.084 E20	6.807 E20	9.887 E20	1.506 E21
Total - 132	1.067 E20	2.318 E20	4.274 E20	9.670 E20	1.444 E21	2.356 E21
Xe ¹³⁴	1.371 E20	2.982 E20	5.496 E20	1.233 E21	1.821 E21	2.893 E21
Xe ¹³⁶	2.801 E20	6.088 E20	1.120 E21	2.511 E21	3.708 E21	5.892 E21
Xe ¹³⁶ /Xe ¹³⁴ ratio	2.043	2.042	2.038	2.036	2.036	2.037
Xe ¹³² /Xe ¹³¹ ratio	1.372	1.376	1.386	1.421	1.461	1.564
% change in Xe ¹³² /Xe ¹³¹ ratio ($t_0 = 1.366$)	0.44	0.73	1.46	4.03	6.95	14.5

^aEquivalent to 2.608×10^{19}

Table 11. Buildup and Burnout of Neodymium
 Fuel - 1×10^{23} atoms U^{235} Flux - 1×10^{13} n-cm⁻²sec⁻¹

ATOMS PRESENT

IRRADIATION TIME	0.1 yr	0.5 yr	1.0 yr	3.0 yr	5.0 yr	10.0 yr
% DEPLETION	1.81	8.71	16.66	42.12	59.80	83.40
Nuclide						
Pr ¹⁴³	4.494 E19 ^a	4.976 E19	4.544 E19	3.154 E19	2.191 E19	8.808 E18
Nd ¹⁴³	5.366 E19	4.166 E20	8.232 E20	1.929 E21	2.443 E21	2.447 E21
Total - ¹⁴³	9.850 E19	4.664 E20	8.686 E20	1.961 E21	2.465 E21	2.456 E21
Ce ¹⁴⁴	7.282 E19	2.954 E20	4.591 E20	5.541 E20	4.248 E20	1.758 E20
Nd ¹⁴⁴	3.580 E18	8.228 E19	2.866 E20	1.563 E21	2.895 E21	5.448 E21
Total - ¹⁴⁴ 285 d. after shutdown	3.999 E19	2.293 E20	5.163 E20	1.840 E21	3.107 E21	5.536 E21
Nd ¹⁴⁵	5.724 E19	2.752 E20	5.245 E20	1.305 E21	1.823 E21	2.446 E21
Nd ¹⁴⁶	4.346 E19	2.105 E20	4.046 E20	1.045 E21	1.519 E21	2.264 E21
Nd ¹⁴⁸	2.213 E19	1.067 E20	2.040 E20	5.148 E20	7.297 E20	1.019 E21
Nd ¹⁵⁰	9.244 E18	4.459 E19	8.528 E19	2.155 E20	3.057 E20	4.280 E20
Nd ¹⁴⁶ /Nd ¹⁴³ 285 d. after shutdown	0.441	0.451	0.466	0.533	0.616	0.922
% change in Nd ¹⁴⁶ /Nd ¹⁴³ ratio ($t_0 = .439$)	0.46	2.73	6.15	21.4	40.3	110
Nd ¹⁴⁸ /Nd ¹⁵⁰ ratio	2.394	2.393	2.392	2.389	2.387	2.381
Nd ¹⁴⁴ /Nd ¹⁴³ 285 d. after shutdown	0.406	0.492	0.594	0.938	1.260	2.218
% change in Nd ¹⁴⁴ /Nd ¹⁴³ ratio ($t_0 = 0.383$)	6.01	28.4	55.1	145	229	479

^aEquivalent to 4.494×10^{19}

Table 12. Buildup and Burnout of Neodymium
 Fuel - 1×10^{23} atoms U^{235} Flux - 1×10^{14} n-cm⁻²sec⁻¹

ATOMS PRESENT			
IRRADIATION TIME	0.1 yr	0.5 yr	1.0 yr
% DEPLETION	19.7	66.2	38.6
Nuclide			
P ¹⁴³	4.011 E20 ^a	2.370 E20	9.529 E19
Nd ¹⁴³	4.840 E20	2.313 E21	2.451 E21
Total - ¹⁴³	8.851 E20	2.550 E21	2.546 E21
Ce ¹⁴⁴	6.709 E20	1.968 E21	2.054 E21
Nd ¹⁴⁴	5.841 E19	1.273 E21	3.501 E21
Total - ¹⁴⁴ 285 d. after shutdown	3.938 E20	2.257 E21	4.528 E21
Nd ¹⁴⁵	5.241 E20	1.817 E21	2.426 E21
Nd ¹⁴⁶	4.046 E20	1.519 E21	2.262 E21
Nd ¹⁴⁸	2.040 E20	7.297 E20	1.019 E21
Nd ¹⁵⁰	8.529 E19	3.057 E20	4.280 E20
Nd ¹⁴⁶ /Nd ¹⁴³ 285 d. after shutdown	0.457	0.596	0.888
% change in Nd ¹⁴⁶ /Nd ¹⁴³ ratio ($t_0 = 0.422$)	8.29	41.2	110
Nd ¹⁴⁸ /Nd ¹⁵⁰ ratio	2.392	2.387	2.381
Nd ¹⁴⁴ /Nd ¹⁴³ 285 d. after shutdown	0.445	0.885	1.778
% change in Nd ¹⁴⁴ /Nd ¹⁴³ ratio ($t_0 = 0.292$)	52.4	203	509

^aEquivalent to 4.011×10^{20}

Figure 1. Ratio of the Fission Yields of Fast Fission to Thermal Fission for U^{233}

Figure 2. Ratio of the Fission Yields of Fast Fission to Thermal Fission for U^{235} .

Figure 3. Ratio of the Fission Yields of Fast Fission to Thermal Fission for Pu^{239}

Cross sections are in barns for thermal neutron capture

<u>FISSION YIELDS</u>			
<u>PATH NO.</u>	<u>U^{233}</u>	<u>U^{235}</u>	<u>Pu^{239}</u>
1	1.17	0.544	0.29
2	1.95	1.00	0.47
3	0.58	0.293	0.127
4	3.27	2.02	0.76

Figure 4. Computer Scheme for the Reactor Production of Krypton Isotopes CPP-S-3170

Cross sections are in barns for thermal neutron capture

PERCENT FISSION YIELDS

<u>PATH NO.</u>	<u>U²³³</u>	<u>U²³⁵</u>	<u>Pu²³⁹</u>
1	3.39	2.93	3.78
2	4.64	4.38	5.26
3	5.78	6.62	6.91
4	5.95	8.06	7.47
5	5.8	6.3	7.02
6	6.63	6.46	6.63

CPP-S-3171

Figure 5. Computer Scheme for the Reactor Production of Xenon Isotopes

Cross sections are in barns for thermal neutron capture

PERCENT FISSION YIELDS

PATH NO.	U ²³³	U ²³⁵	Pu ²³⁹
1	5.99	6.03	4.57
2	4.61	5.62	3.93
3	3.47	3.98	3.13
4	2.63	3.07	2.60
5	1.34	1.71	1.73
6	0.56	0.67	1.01

CPP-S-3172

Figure 6. Computer Scheme for the Production of Neodymium Isotopes

Figure 7. Ratio of Inert Gas Isotopes Produced in U^{235} Thermal Fission as a Function of U^{235} Depletion

Figure 8. Ratio of Neodymium Isotopes Produced in U^{235} Thermal Fission as a Function of U^{235} Depletion

Figure 9. Percent Change in Inert Gas Isotope Ratio as a Function of Depletion

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Figure 10. Percent Change in Neodymium Isotope Ratio as a Function of Depletion

Figure 11. Effect of Neutron Flux Level on Isotope Ratios

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Figure 12. Effect of Neutron Flux Level on Isotope Ratios

Figure 13. Machine Computation of the Isotope Ratio Xe^{136}/Xe^{134} as a Function of the Flux Level for the Thermal Fission of U^{235}

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Figure 14. Ratio of the Fission Yields for the Thermal Fission of Pu^{239} and U^{235}

Figure 15. Ratio of the Fission Yields for the Fast Fission of Pu^{239} and U^{235}

Figure 16. Ratio of the Fission Yields for the Fast Fission of Pu^{239} and U^{238}

Figure 17. Isotopic Ratio of $\text{Xe}^{134}/\text{Kr}^{86}$
as a Function of Depletion for the Thermal Fission of U^{235} and Pu^{239}

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Figure 18. Isotopic Ratio of $\text{Nd}^{148}/\text{Nd}^{150}$
as a Function of Depletion for the Thermal Fission of U^{235} and Pu^{239}

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