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METHODS FOR DETERMINING BURNUP IN ENRICHED URANIUM-235 FUEL IRRADIATED BY FAST REACTOR NEUTRONS

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16. Abstract

Methods of determining burnup of enriched uranium-235 in fast neutron irradiations are discussed. Fission yields of those fission products that vary less than 5 percent in a thermal or a fast neutron environment are listed. The potential burnup monitors are discussed with respect to their diffusion behavior during high-temperature irradiations, ease of mass spectrometer analysis, and adverse effects of neutron capture reactions. Recommendations are made as to the most reliable methods.

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SUMMARY

Methods of determining burnup of uranium-235 irradiated in a fast neutron flux are discussed. A compilation of current uranium-235 fission yield data is included. Only isotopes whose fission yields do not vary much with neutron energy are considered.

Selection of a suitable burnup monitor is determined by the duration of the irradiation, temperature, chemical form of the fuel matrix and cladding, and presence or absence of rare earth binder.

Neodymium-148 is suggested as the most suitable burnup monitor if there has been no loss of fuel during the irradiation and no rare earth binder has been used.

The recommendations in the report are limited to conditions of enrichment and burnup in which contributions from uranium-238 fast fissions and plutonium fissions are insignificant.

INTRODUCTION

Accurate determination of burnup or atom percent fission is important in evaluating nuclear fuels and in correlating irradiation damage with the extent of irradiation. There are many considerations in the choice of a method of determining burnup. Fast irradiations impose a particular constraint because many fission yields and the capture to fission cross section ratio of uranium-235 (U^{235}) vary significantly with neutron energy, and the neutron energy spectrum is not always carefully defined. For those methods suitable for fast as well as thermal burnup determination, this report discusses all of the important considerations in selection of a method. A compilation of the latest U^{235} fission yield data found in the literature is included. Recommendations for fast neutron burnup determination are presented.

This report is confined to a burnup range in enriched uranium where U^{238} and plutonium isotope contributions may be ignored. For example, in fuel enriched to 93 percent U^{235} the fission product contributions of U^{238} and plutonium (Pu) are less than in fuel of low enrichment for a given amount of burnup. Alternatively, one can irradiate to higher burnups using 93 percent enriched U^{235} without considering the fission contributions of these other heavy metals. The experimenter must select an enrichment consistent with his desired operating conditions. The required accuracy of burnup analysis will dictate whether contributions from U^{238} and Pu may be ignored.

This report first lists the properties of an ideal fast burnup monitor. This is followed by a brief discussion of a fast reactor spectrum. Possible burnup methods are described. Data on individual fission products are tabulated. Because of the aforementioned constraints imposed by neutron spectrum variation, data are arbitrarily restricted to fission products whose thermal fission yield is within 5 percent (relative) of the fast yield. Descriptions of spectrophotometric methods for the analysis of several elements or groups of elements are included.

A discussion of flux wires, fission products distribution by gamma scanning, fuel cladding dosimetry, and other techniques which can be correlated to burnup is not included.

BURNUP BY FAST NEUTRONS

Properties of the Ideal Fast Fission Burnup Monitor

The "ideal" fission product monitor should have the following properties:

(1) Be refractory in both the elemental and oxide state
(2) Have no gaseous or volatile precursors
(3) Be stable or have a long half-life
(4) Be simple to separate from matrix, fuel, and cladding by standard chemical methods
(5) If radioactive, have a simple and established decay scheme and a gamma spectrum that may be readily resolved from other isotopes of the same element
(6) If to be used for isotope dilution, have a shielded isotope of the same element that permits monitoring for natural contamination and another stable or long-lived isotope to be used as a diluent
(7) Have a low capture cross section to thermal neutrons, although this is less significant in a fast neutron irradiation
(8) Have no significant production of the isotope by thermal neutron capture of another isotope

(9) Have the same fission yield for all neutron energies and different fissioning species

Except for property (9) these properties are all desirable characteristics of a burnup monitor in thermal irradiations.

Energy Distribution of Fast Reactor Neutrons

The neutron energy distribution of virgin neutrons emitted from fissioning U^{235} is called a fission spectrum. The fission spectrum peaks at about 600 keV and has a mean energy of about 2 MeV. However, in any fast reactor there is some downward shift in the energy of the virgin neutron spectrum. Most of the fast fission yields listed in this report were obtained from irradiations in EBR I (ref. 1). The neutron spectrum in EBR I was reported (ref. 2) as peaking at 100 keV and having a mean energy of 400 keV. This was discussed with Dr. J. E. Rein (of Idaho Nuclear, Idaho Falls, Idaho) in August 1969. He said the EBR I numbers were obtained from Argonne physicists but no actual measurements were made. He believes, however, that their irradiations were in the hardest neutron spectrum that could be achieved in an existing fast reactor at steady-state operation. The spectrum was harder in EBR I than in EBR II.

Fast fission yields obtained from references 1 and 3 are listed in reference 3 as fission spectrum yields. Although the term "fission spectrum yields" is not entirely accurate, it is widely used. Certain special terms used in this report are defined in the glossary.

Discussion of Possible Burnup Methods

Depletion of uranium. - The most direct way to measure burnup is to determine the loss of uranium. In order to obtain accuracy, an analysis of uranium content requires high burnup and analysis of the whole fueled specimen.

Change in isotopic composition of fuel. - The most reliable method of measuring burnup in thermal irradiations has been the mass spectrometric determination of the change of U^{236} to U^{235} ratio. Burnup or atom percent fission is calculated from the relation

$$F_5 = \frac{N_5^0\left(R_{6/5} - R_{6/5}^0\right)}{R_{6/5} + \alpha_5(1 + R_{6/5})} \tag{1}$$

where

F_5	atom percent fission attributable to U^{235}
N_5^0	initial atom percent abundance of U^{235}
$R_{6/5}$, $R_{6/5}^0$	postirradiation and preirradiation ratios of U^{236} to U^{235}
α_5	capture to fission cross-section ratio in U^{235}

In some high-temperature irradiations this has been the only burnup procedure possible because of loss of fission products and fuel from the specimen.

This method actually measures the number of capture reactions occurring in U^{235} atoms. Calculation of atom percent fission by isotopic change is dependent on knowing the ratio of the number of captures to the number of fissions. This ratio, $alpha_5$, varies with neutron energy. If the neutron spectrum is not always accurately known, an uncertainty in the value of $alpha_5$ results. Despite this added uncertainty, one may have to resort to isotopic measurements for some high-temperature irradiations because there may be migration or loss of fission products or the associated fuel. Then additional irradiations that match the nuclear environment at low temperature would enable one to calculate $alpha_5$ for the high-temperature irradiation environment. $Alpha_5$ is calculated from low-temperature irradiations by using fission product measurements together with the change in the 236 to 235 isotopic ratio of the associated fuel. This value of $alpha_5$ from the low-temperature irradiation may then be used to determine burnup for the high-temperature irradiation by the isotopic change method.

Fission product monitors. -

Stable fission product monitors: In another mass spectrometric burnup method currently used for thermal irradiations the concentration of neodymium-148 (Nd^{148}), a stable isotope, is determined by isotope dilution with Nd^{150}. Neodymium-148 has many properties that make it desirable as a burnup monitor. It has a low activation cross section, and sensitivity by thermal emission mass spectrometer analysis is high. Neodymium is refractory and does not migrate in most fuels at high temperatures. Nd^{142}, a shielded isotope, allows monitoring for natural neodymium contamination.

Disadvantages of neodymium include problems in ion exchange purification in some fuels. George Buzzelli (of Gulf General Atomics, La Jolla, California) has stated that he has observed a tendency for neodymium to migrate in carbide fuels at high temperatures (unpublished data). Other stable isotopes can be determined by isotope dilution providing there is another isotope of the same element that may be used as a diluent. Further discussion is given in the section Selection of a Burnup Monitor.

Radiochemical determination of burnup from individual isotopes: The concentration of a radioactive fission product is used in another category of methods. A number of radioactive fission products have been useful thermal monitors under the right circum-

stances: 12.8-day barium-140 (Ba^{140}), 65-day zirconium-95 (Zr^{95}), 284-day cerium-144 (Ce^{144}), 29-year strontium-90 (Sr^{90}), and 30-year cesium-137 (Cs^{137}). Barium-140 can only be used for very short irradiations, Zr^{95} and Ce^{144} for progressively longer irradiations. Cesium and, to a lesser extent, strontium are volatile and migrate at higher temperatures. If atoms of the fission product migrate to or from the region analyzed, the calculated burnup will be in error.

Selection of a burnup monitor: There is no fission product monitor, stable or unstable, that is ideal for all circumstances. Choice of suitable burnup monitors must be made on an individual basis considering the length of irradiation, temperature of irradiation, matrix and cladding materials dissolved with the fuel, and neutron energy spectrum. If one could define the neutron spectrum in an irradiation and if one knew the correct fission yield to use for an isotope produced in that spectrum, one would not be concerned by a change in fission yield with neutron energy. But the uncertainties in knowing the spectrum and the fission yield make it desirable to select fission products having yields that vary only slightly with neutron energy. For this investigation fission products whose relative yield changes more than 5 percent have been arbitrarily excluded. Also excluded are yields less than 1 percent and unstable isotopes whose half-lives are less than 60 days. Fission yields are taken from references 1 and 3. A list of the isotopes to be considered is found in table I. Table II summarizes the properties of elements and isotopes pertinent to burnup determination by mass spectrometry. Table II is based on the list of properties specified in the section Properties of the Ideal Fast Fission Burnup Monitor for the "ideal" fission burnup monitor. More discussion on each isotope is given in the appendix.

Analyses of chemical species. - Instead of analyzing individual fission product isotopes, one may determine all of the isotopes of an element by spectrophotometric analysis. Sensitivities are not generally as good as with mass spectrometric or radiochemical analysis, but this can be compensated by a higher summation on fission yield.

Spectrophotometric analysis of zirconium: A method for determining burnup by the spectrophotometric analysis of fission product zirconium complexed with Arseneazo III has been reported. Satisfactory results have been reported by George Buzzelli (of Gulf General Atomics) in the analysis of carbide fuels, and the method has been proposed to the ASTM Committee E-10 Sub I (Burnup of Nuclear Fuels). The procedure is now being evaluated by several member laboratories including the Plum Brook Reactor Facility.

A comparison of zirconium fission yields in U^{235} irradiated with thermal and fission spectrum neutrons is shown in table III.

Zirconium cannot be used as a monitor in zircaloy clad fuels.

Spectrophotometric determination of molybdenum: Molybdenum complexed with thiocyanate or as phosphomolybdate can also be analyzed spectrophotometrically. Methods can be found in standard quantitative analysis textbooks. As mentioned in the

appendix, molybdenum escapes from uranium dioxide (UO_2) in high-temperature irradiations. Molybdenum (Mo) has been useful in carbide fuels and may also be of value in nitride fuels. Comparative yields of the molybdenum fraction are shown in table IV.

Spectrophotometric determination of technetium: A spectrophotometric analysis of the thiocyanate complex of technetium (Tc) has been used by Dr. R. Larsen (of Argonne National Laboratory) and has been proposed as an ASTM method. The tentative method is currently being evaluated by ASTM Committee E-10. There is only one isotope, Tc^{99}, so the fractional fission yield is lower than zirconium or molybdenum. For equivalent sensitivity higher burnup is required. A 22-barn thermal cross section means a correction may be required in long irradiations. In a fast flux the burnup of Tc^{99} would be less than in a thermal flux but uncertainties in the capture cross section make corrections more difficult to evaluate.

Technetium is the daughter of 67-hour Mo^{99} so loss of molybdenum from oxide fuels must be considered.

Rare earth analyses. - Two burnup methods have been developed at Argonne National Laboratory that involve quantitative determination of the total rare earth fraction (including lanthanum (La)). This fraction represents nearly a 50 percent fission yield. The summation of the yields is also virtually independent of neutron energy. This makes a rare earth analysis attractive for irradiations in poorly defined neutron spectra. Transmutation by either radioactive decay or neutron capture is not a problem as the product is always another rare earth. A negligible addition to the La^{139} contribution comes from Ba^{138} captures. The cross section of Ba^{138} is only 0.4 barn. The comparative fission yields are shown in table V.

A spectrophotometric titration analysis of fission product rare earths is described in reference 4.

An X-ray analysis of fission product rare earths is described in reference 5.

GENERAL RECOMMENDATIONS

It should be emphasized that this report is concerned with moderate burnup of enriched U^{235} fuel where contributions of U^{238}, the plutonium isotopes, and other fissionable species may be ignored.

The choice of a burnup monitor used in fast neutron irradiations will be dictated by the conditions of the irradiation: extent and uniformity of the irradiation; temperature; chemical form of the fuel, matrix, and cladding; and presence of rare earth binders.

Because of the advantages mentioned in the appendix, Nd^{148} is the best all-around candidate in irradiations where the fuel holds together. It is necessary to determine the amount of associated fuel when a fission product is used. If some fuel is lost during an

irradiation, one cannot assume a proportionate loss of the fission product. If rare earth binders are used, neodymium will be limited to conditions in which the natural contamination contribution is low enough that it can be corrected by monitoring the 142 mass peak without loss of precision.

If some fuel is lost, then the determination of the change in the U^{236} to U^{235} ratio is recommended. Representative preirradiation specimens are needed. Additional irradiations may be needed to establish the value of $alpha_5$, the U^{235} capture to fission cross-section ratio. Specimens so irradiated would have to be encapsulated in a manner that would insure no loss of fission products. The burnup value obtained from fission product analysis could be substituted in equation (1) to determine the value of $alpha_5$ in this environment.

The Ce^{142} isotope dilution method should make a good backup monitor. The uncertainties in fission yield are less than with neodymium, but the short-lived xenon and cesium precursors might cause losses at extreme temperatures. A rare earth binder will cause the same problems as with the neodymium determinations.

Cerium-144 is also recommended for irradiations that are short compared to its 284-day half-life.

The zirconium spectrophotometric method may be of value in analyzing samples that have no dissolution problems. Success has been reported with carbide fuels, and perhaps nitride fuels will be as appropriate.

The use of the total rare earth fraction is attractive because the yield is virtually independent of neutron energy.

For lower temperatures Cs^{137} is still a good choice. With a germanium-lithium (Ge-Li) detector, Cs^{137} can now be analyzed in gross aliquots of dissolver solution.

The limitation to isotopes having a fission yield spread of less than 5 percent is arbitrary. There will be experiments in which less precise burnup values are needed. Then some isotope not recommended in this report may be acceptable.

Lewis Research Center,
National Aeronautics and Space Administration,
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120-27.

APPENDIX - DISCUSSION OF PROPERTIES OF ELEMENTS AND ISOTOPES PERTINENT TO FAST REACTOR BURNUP DETERMINATION BY MASS SPECTROMETRY

Since the majority of Plum Brook Reactor fueled experiments are related to space power systems, behavior of isotopes at high temperatures is important. In some cases even though the stable isotope in the main chain is refractory there may be noble gas or volatile precursors. If the half-life of the precursor is sufficiently long and if the yield relative to mass chain yield is high, migration from the fission area may occur before transmutation to the stable form occurs. In low-temperature irradiations fission monitor selection is less critical. The isotopes discussed in this appendix are considered in elemental groups since some of the properties are the same. The ionization potentials listed are an index of sensitivity of mass spectrometer analysis; the lower the better.

Rubidium

Rubidium-87 (Rb^{87}) sensitivity for mass spectrometer analysis is excellent. The rubidium ionization potential is 4.2 volts. Rubidium-85 could be used as a spike. However, there is no shielded isotope to monitor for natural contamination. Rubidium-87 has a 12-barn thermal cross section. Rubidium melts at 38.5° C and boils at 700° C. The 76-minute krypton precursor has the same yield as its rubidium daughter; thus, this isotope cannot be used at elevated temperatures. Rubidium is not recommended because of the thermal properties.

Strontium

Strontium-88 (Sr^{88}) sensitivity is very good. The strontium ionization potential is 5.7 volts. Strontium-86 and strontium-87 are both shielded isotopes, although some Sr^{86} is produced by neutron capture in Rb^{85}. In addition, some Sr^{88} is produced from Rb^{87}. Strontium melts at 774° C and boils at 1042° C. However, the oxide melts at 2470° C and boils at 3000° C. The worst feature for high-temperature operation is the 2.8-hour krypton precursor (relative yield, 90 percent). Strontium is not recommended because of the krypton precursor.

Yttrium

Yttrium-89 (Y^{89}) is the only stable or long-lived isotope; thus, it cannot be analyzed by isotope dilution mass spectrometry. The metal melts at 1490° C and boils at 2500° C. The oxide melts at 2420° C. The capture cross section is only 1.2 barns. While Y^{89} cannot be analyzed mass spectrometrically, a colorimetric procedure could be developed. Yttrium is not recommended at present.

Zirconium

Zirconium has four isotopes that could be analyzed by isotope dilution mass spectrometry. Zirconium-90, the only shielded isotope, is only partially so by holdup as the 29-year Sr^{90} precursor.

Zirconium sensitivity on the mass spectrometer is fair; the ionization potential is 6.8 volts; however, there is some zirconium impurity in rhenium filaments.

The most attractive aspect of zirconium is its thermodynamic properties. The melting point of zirconium is 1868° C, and the boiling point is greater than 2900° C. The oxide melts at 2710° C; no value was found for the boiling point. Zirconium is the least likely to migrate at high temperatures of any of the fission products.

The cross section of Zr^{92} is 0.2 barn, Zr^{93} is <4 barns, Zr^{94} is 0.08 barn, and Zr^{96} is 0.05 barn.

Since Zr^{93} does not occur in natural zirconium, it should make a good monitor. There is a 3-second krypton precursor. Zirconium-96 has the smallest spread in fission yield and no krypton precursor.

Zirconium-91 is the recommended spike.

These isotopes have possibilities, but the spectrophotometric method mentioned in the section Spectrophotometric analysis of zirconium should be faster and easier. The spectrophotometric method is currently being evaluated by the burnup committee of ASTM.

Molybdenum

There are four molybdenum isotopes to be considered. Molybdenum melts at 2622° C, and the boiling point is 4800° C; however, the oxide is quite volatile, melting at 791° C and boiling at 1200° C. Thus, the isotopes of this element can be used in high-temperature irradiations if the fission products remain in the metallic state but should not be used if oxidation is possible.

(Not everyone agrees that the loss of molybdenum in oxide fuels is caused by formation of volatile molybdenum trioxide (MoO_3). There has been evidence at Argonne National Laboratory and Dounreay of precipitation and formation of a refractory alloy of molybdenum, ruthenium, and other noble metal fission products.)

Molybdenum has an ionization potential of 7.1 volts. The element still can be analyzed by thermal emission, but sensitivity is poor. Molybdenum is an impurity in the rhenium filament.

Molybdenum-92, molybdenum-94, and molybdenum-96 are shielded isotopes. Some Mo^{96} is produced from Mo^{95} capture. Molybdenum-92 or molybdenum-94 would be a suitable spike. The cross section of Mo^{95} is 14 barns, which could limit its usefulness in long irradiations. The cross section of Mo^{97} is 2 barns, Mo^{98} is 0.5 barn, and Mo^{100} is 0.1 barn. Because of the fission yield spreads, Mo^{100} would be the preferred isotope.

Because of the poor sensitivity, mass spectrometric analysis is not recommended. The volatility of MoO_3 or the precipitation of metallic molybdenum precludes the use of molybdenum in oxide fuels at elevated temperature. In carbide and nitride fuels the molybdenum can be used.

Technetium

The only long-lived isotope of technetium is 2.14×10^5 year Tc^{99}. Technetium cannot be analyzed by isotope dilution mass spectrometry. A colorimetric analysis is referred to in the section Spectrophotometric determination of technetium. The melting point is 2400° C and the boiling point is greater than 3000° C. Technetium is not recommended in oxide fuels but has been of value in carbide fuels.

Tellurium

Tellurium (Te) melts at 452° C and boils at 1390° C. The oxide sublimes at 450° C. This eliminates tellurium from serious consideration as a burnup monitor except at low temperatures. This is unfortunate as the fission yields of Te^{130} are the same for both thermal and fission spectrum neutrons.

Tellurium has an ionization potential of 9.0 volts and could not be analyzed by thermal emission.

Tellurium is not recommended for burnup analysis.

Cesium

Cesium is quite volatile, melting at 28.5° C and boiling at 670° C. Migration and escape can be severe in high-temperature irradiations.

Cesium-133 is the only stable isotope. Cesium-137 could be used as a spike, but there is no shielded isotope and cesium is a common contaminant. Cesium has an ionization potential of 3.89 volts and sensitivity on the mass spectrometer is excellent. The thermal neutron capture cross section is 31 barns. Volatile precursors include 20-hour iodine (I) and 5.3-day xenon (Xe). This isotope is not recommended.

Cesium-137 is the classical burnup monitor used by the radiochemist. The long half-life, simple decay scheme, and chemical properties have made it attractive in low-temperature applications. There is an interference in the gamma spectrum from Cs^{134} in long irradiations. High resolution Ge-Li detectors have reduced that interference to a considerable degree. Refinements in the ASTM Committee E-10 Sub 1 tentative standard (ASTM E-320) have improved the precision. The Cs^{137} radiochemical method has now been submitted for consideration as an ASTM standard.

The capture cross section is 0.1 barn. Volatile precursors include 6.3-second I and 3.8-minute Xe. Relative yields of the precursors are 43 and 86 percent.

Cesium-137 can still be a useful fission monitor, even at elevated temperatures, if the entire irradiated specimen can be analyzed and there has been no rupture of the cladding.

Barium

Barium melts at 725° C and boils at 1140° C. The oxide melts at 1923° C and boils at 2000° C. Barium-138, the only unshielded isotope, has 17-minute xenon and 32-minute cesium precursors. The relative yields are 26 and 74 percent of the terminal mass chain yield.

Sensitivity on the mass spectrometer is excellent. The ionization potential is 5.2 volts. Barium is an impurity in rhenium filaments. However, shielded isotopes permit corrections.

This isotope is not too promising, primarily because of the 17-minute xenon precursor.

Cerium

Cerium melts at 804° C and boils at 3257° C. The oxide melts at 2800° C. No data are available on the boiling point of the oxide. Loss of cerium isotopes should be

negligible. George Buzzelli (of Gulf General Atomics) observed some loss of cerium (and neodymium) in carbide fuels at very high temperatures. He assumes the cerium and neodymium exist as the more refractory oxide in UO_2 but as the metal in the carbide fuel. Although the boiling point of the metal is high, the vapor pressure is also somewhat high. Cerium has an ionization potential of 5.6 volts. Sensitivity is excellent on the mass spectrometer.

There are two shielded isotopes, Ce^{136} and Ce^{138}. Either one could be used as a spike.

Cerium-140 has 1.5-second I^{140}, 13.7-second Xe^{140}, and 65-second Cs^{140} as precursors. The relative yield for xenon is 37 percent and for cesium is 82 percent. The capture cross section is 0.6 barn.

This isotope is not recommended, as the properties of Ce^{142} are much more attractive.

Cerium-142 has a 1.2-second xenon and 2.3-second cesium precursor. The relative yield of the xenon is only 8.9 percent. The yield for cesium is 48 percent.

The thermal cross section of Ce^{142} is 1 barn.

Although the volatile precursors may introduce problems in high-temperature irradiations, the half-lives of the precursors are short and the relative yield of the xenon isobar is low. Cerium-142 is attractive because of the small change in yields when irradiated in the two neutron environments.

This method cannot be used in fuels containing a rare earth binder.

Separation of the cerium should be simple since it elutes from the resin right after neodymium (see the section of this appendix entitled Neodymium). Cerium-136 is the recommended spike. Cerium-142 is recommended.

Cerium-144 has 8.8-second xenon and 1-second cesium precursors, but the relative yields are only 0.2 and 14 percent of the cumulative yield of the mass chain.

The isotope has a 284-day half-life so that its use is limited to irradiations of moderate length in a uniform flux.

The thermal cross section is 1 barn.

Because of interference using a sodium iodide detector with a Ce^{141} gamma, Ce^{144} has in the past been beta-counted. Absorbers were used to eliminate all but the 2.99 MeV beta of praseodymium-144 (Pr^{144}). With a high resolution germanium (lithium drifted) detector the Ce^{144} gamma, 0.134 MeV, can be used for analysis of an aliquot of dissolver solution.

Cerium-144, along with Nd^{148}, is a favored burnup detector at the Dounreay fast reactor facility (ref. 6).

Cerium-144 is recommended in irradiations where uncertainties introduced by radiation history are negligible.

Praseodymium

Praseodymium has a melting point of 950° C and a boiling point of 3212° C. We have found no thermodynamic data on the oxide.

There is only one stable isotope, Pr^{141}, and no long-lived isotopes that could be used for isotope dilution.

The cross section of Pr^{141} is 19 barns. Praseodymium is not recommended, as there is no sensitive method of analysis.

Neodymium

Neodymium melts at 1018° C and boils at 3127° C. The oxide melts at 2272° C; no information was found for the boiling point.

Neodymium has an ionization potential of 5.51 volts, so the sensitivity for mass spectrometric analysis is excellent.

Neodymium-143 has a 1-second xenon and a 1.6-second cesium precursor. The relative yields are 3 and 28 percent. A 330-barn cross section limits the usefulness of this isotope as a burnup monitor.

Neodymium-144 has the same volatile precursors as cerium (see the section Cerium) and a 19-barn cross section. This isotope is not recommended because of its formation from Nd^{143} and because of the long holdup as cerium.

Neodymium-145, as well as all higher mass chains, has no volatile precursor but has a 50-barn cross section. It is not recommended.

Neodymium-146 has only a 2-barn cross section but will also be produced from Nd^{145}. It is not recommended.

Neodymium-148 has a 4-barn cross section. This is the most versatile burnup monitor. The thermal fission yield is almost the same for Pu^{239} as for U^{235}. That does not concern us in this investigation but it is the primary reason much development has taken place in the analysis.

ASTM Committee E-10 Sub 1 has perfected the analysis of Nd^{148} for burnup determinations. The method is now an ASTM standard and can be found in reference 7.

Neodymium-148 is recommended as the primary monitor in those irradiations that do not suffer loss of fuel and/or fission products in an unpredictable manner. There has been some loss in carbide fuels at very high temperatures.

This method cannot be used in fuels containing a rare earth binder.

GLOSSARY

Alpha$_5$, α_5 - The ratio of the neutron capture cross section to fission cross section of uranium-235.

Burnup - Atom percent fission or the number of fissions occurring for 100 initial heavy metal atoms of atomic mass greater than 225.

Fast flux - fission spectrum flux - Fast flux refers to a neutron flux with an energy distribution typical of that which exists in a fast reactor. Fission spectrum flux is discussed in the section Fission Product Monitors.

Fission yield - The number of atoms of a particular isotope that are produced for each 100 fissions.

Hard neutrons - Fast neutrons.

Isobars - Nuclides of differing atomic number but the same atomic weight.

Shielded isotope - In a mass chain, fission products nearly always decay by beta emission until a stable isobar is formed. The isobar with the next higher atomic number cannot be formed and is called a shielded isotope.

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TABLE I. - URANIUM-235 FISSION YIELDS OF

SELECTED FISSION PRODUCTS

Isotope	Fission yield				Relative difference in fission yield, percent	Half-life, yr
	Thermal neutrons, percent	Reference	Fast neutrons, percent	Reference		
Rb^{87}	2.54	1	2.66	1	4.6	4.7×10^{10}
Sr^{88}	3.61	1	3.63	1	.55	Stable
Y^{89}	4.76	3	5.00	3	4.9	Stable
Zr^{92}	5.95	1	5.8	3	2.6	Stable
Zr^{93}	6.34	1	6.1	3	3.9	9.5×10^{5}
Zr^{94}	6.41	1	6.2	3	3.3	Stable
Mo^{95}	6.45	1	6.47	1	.3	Stable
Zr^{96}	6.23	1	6.1	3	2.7	Stable
Mo^{97}	5.86	1	6.13	1	4.5	Stable
Mo^{98}	5.77	1	6.04	1	4.6	Stable
Tc^{99}	6.14	1	5.9	3	4.0	2.14×10^{5}
Mo^{100}	6.24	1	6.35	1	1.7	Stable
Te^{130}	2.1	3	2.1	3	----	Stable
Cs^{133}	6.73	1	6.69	1	.6	Stable
Cs^{137}	6.28	1	6.20	1	1.3	29.2
Ba^{138}	6.80	1	6.60	1	3.0	Stable
Ce^{140}	6.31	1	6.21	1	1.6	Stable
Pr^{141}	6.1	3	6.3	3	3.2	Stable
Ce^{142}	5.88	1	5.82	1	1.0	Stable
Nd^{143}	5.90	1	5.80	1	1.7	Stable
Ce^{144}	5.40	3	5.15	3	4.7	284-day
Nd^{144}	5.42	1	5.26	1	3.0	Stable
Nd^{145}	3.86	1	3.85	1	.3	Stable
Nd^{146}	2.95	1	3.00	1	1.7	Stable
Nd^{148}	1.67	3	1.73	3	4.7	Stable

TABLE II. - SUMMARY OF PROPERTIES OF ISOTOPES PERTINENT TO BURNUP[a]

Isotope	Ionization potential, V	Spike	Shielded isotope	Method of analysis			High-temperature behavior		Activation loss or production, b	Volatile precursors	Recommendations
				Mass spectrometric	Radiochemical	Other	Metal	Oxide			
Rb^{87}	4.2	85	None	X	--	--	Poor	Poor	[b]-12	Yes	No
Sr^{88}	5.7	86, 87	86, 87	X	--	--	Fair	Fair	12	Yes	No
Y^{89}	(c)	None	None	--	--	--	Good	Good	-1.2	Yes	No
Zr^{92}	6.8	91	90	--	--	X	Excellent[d]	Excellent	-.2	Yes	Isotopic analysis not recommended
Zr^{93}	6.8	91	90	---	--	X	Excellent	Excellent	-< 4	Yes	
Zr^{94}	6.8	91	90	--	--	X	Excellent	Excellent	-.08	Yes	Element
Zr^{96}	6.8	91	90	X	--	X	Excellent	Excellent	-.05	No	(e)
Mo^{95}	7.1	92, 94	92, 94, 96	--	--	X	Excellent	Poor	-14	No	Isotopic analysis not recommended
Mo^{97}	7.1	92, 94	92, 94, 96	--	--	X	Excellent	Poor	-2	No	Element not recommended[e]
Mo^{98}	7.1	92, 94	92, 94, 96	--	--	X	Excellent	Poor	-.5	No	
Mo^{100}	7.1	92, 94	92, 94, 96	--	--	X	Excellent	Poor	-.2	No	
Tc^{99}	(c)	None	None	--	--	X	Good	Poor	-22	(e)	No[e]
Te^{130}	9.0	122, 124	122, 124	--	--	--	Poor	Very poor	-.2	No	No
Cs^{133}	3.89	137	None	X	--	--	Poor	Poor	-54	Yes	No
Cs^{137}	3.89	133	None	--	X	--	Poor	Poor	-.1	Yes	Low temperature
Ba^{138}	5.2	132	134	--	--	--	Good	Good	-.4	Yes	No
Ce^{140}	5.6	136, 138	136, 138	--	--	X	Good	Good	-.6	Yes	No
Ce^{142}	5.6	136, 138	136, 138	X	--	X	Good	Good	-1.5	(e)	Yes
Ce^{144}	5.6	136, 138	136, 138	--	X	X	Good	Good	-1	No	(e)
Pr^{141}	(c)	None	None	--	--	X	Good	Good	-19	Yes	Yes
Nd^{143}	5.5	150	142	--	--	X	Good	Good	-330	Yes	No
Nd^{144}	5.5	150	142	--	--	X	Good	Good	-5+330	No	No
Nd^{145}	5.5	150	142	--	--	X	Good	Good	-50	No	No
Nd^{146}	5.5	150	142	--	--	X	Good	Good	-.2	No	No
Nd^{148}	5.5	150	142	X	--	X	Good	Good	-4	No	Yes[e]

[a]For details see appendix.

[b]Negative value indicates loss from activation; positive value indicates production from other isotope (cross section in barns).

[c]Immaterial, as it cannot be analyzed by isotope dilution.

[d]Excellent means no significant migration of the isotope at high temperatures.

[e]See appendix.

TABLE III. - COMPARATIVE URANIUM-235

YIELDS OF ZIRCONIUM

Isotope	Thermal, percent	Reference	Fast, percent	Reference
Zr^{91}	5.90	1	5.5	3
Zr^{92}	5.95	1	5.8	3
Zr^{93}	6.34	1	6.1	3
Zr^{94}	6.41	1	6.2	3
Zr^{96}	6.23	1	6.4	3
	Total 30.83		Total 30.0[a]	

[a]Relative difference in fission yield, 2.73 percent.

TABLE IV. - COMPARATIVE URANIUM-235

YIELDS OF MOLYBDENUM

Isotope	Thermal, percent	Reference	Fast, percent	Reference
Mo^{95}	6.45	1	6.47	1
Mo^{97}	5.86	1	6.13	1
Mo^{98}	5.77	1	6.04	1
Mo^{100}	6.24	1	6.35	1
	Total 24.32		Total 24.99[a]	

[a]Relative difference in fission yield, 2.72 percent.

TABLE V. - URANIUM-235 FISSION YIELDS

OF TOTAL RARE EARTH FRACTIONS

Isotope	Thermal, percent	Reference	Fast, percent	Reference
La^{139}	6.48	3	6.21	3
Ce^{140}	6.31	1	6.21	1
Pr^{141}	6.1	3	6.3	3
Ce^{142}	5.88	1	5.82	1
Nd^{143}	5.90	1	5.80	1
Nd^{144}	5.42	1	5.26	1
Nd^{145}	3.86	1	3.85	1
Nd^{146}	2.95	1	3.00	1
Sm^{147}	2.12	1	2.3	3
Nd^{148}	1.69	1	1.75	1
Sm^{149}	1.00	1	1.09	1
Nd^{150}	.64	1	.83	1
Eu^{151}	.41	1	.44	1
Sm^{152}	.21	1	.31	1
Eu^{153}	.16	3	.21	3
Sm^{154}	.06	1	.10	1
Gd^{155}	.03	3	.07	3
Gd^{156}	.01	3	.03	3
Gd^{157}	.01	3	.02	3
Gd^{158}	.00	3	.01	3
	Total 49.24		Total 49.61[a]	

[a]Relative difference in fission yield, 0.75 percent.

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