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## Practical Consideration of Nuclear Fuel Spiking for Proliferation Deterrence

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PRACTICAL CONSIDERATION OF NUCLEAR FUEL SPIKING  
FOR PROLIFERATION DETERRENCE

J. E. Selle, P. Angelini, R. H. Rainey,  
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## CONTENTS

ABSTRACT . . . . .	1
INTRODUCTION . . . . .	2
SPIKANT SELECTION . . . . .	2
Fission Product Spikants . . . . .	10
Potential Spikants Produced by Other Production Methods . . . . .	11
Dose Rates Available from Fission Product Spikants $^{106}\text{Ru}$ and $^{144}\text{Ce}$ . . . . .	14
Determination of the Amount of $^{59}\text{Co}$ Required to Produce $^{60}\text{Co}$ . . . . .	17
REPROCESSING . . . . .	18
Fission Product Buildup . . . . .	19
Duplex Spikant Reprocessing . . . . .	20
EFFECTS OF SPIKANTS ON FUEL PROPERTIES . . . . .	23
Melting Point . . . . .	23
Thermal Properties . . . . .	25
Mechanical Properties . . . . .	26
Sinterability . . . . .	27
Methods of Adding Spikants to Fuels . . . . .	28
SUMMARY AND CONCLUSIONS . . . . .	29
ACKNOWLEDGMENTS . . . . .	30
REFERENCES . . . . .	30
APPENDIX . . . . .	34



PRACTICAL CONSIDERATION OF NUCLEAR FUEL SPIKING  
FOR PROLIFERATION DETERRENCE

J. E. Selle, P. Angelini, R. H. Rainey,\*  
J. I. Federer, and A. R. Olsen

ABSTRACT

The use of a gamma active radionuclide with nuclear fuel has been proposed as a way to inhibit unauthorized diversion of the fuel and thus provide proliferation deterrence. Proposed dose rate ranges have varied from small additions to increase detectability of diverted material up to large additions to provide lethal doses in a relatively short exposure time. The purpose of this report is to examine some of the practical aspects of incorporating spikants into nuclear fuel in an attempt to identify any adverse consequences of their use.

Selection of potential spikants was made by the application of some somewhat arbitrary radiation criteria to 64 candidate spikants followed by an analysis of the chemical and physical state of each potential spikant. As a result of this analysis the list of candidates was narrowed to  $^{60}\text{Co}$ ,  $^{106}\text{Ru}$ , and  $^{144}\text{Ce}$ . Following this, we investigated the practical aspects of the use of these three spikants in nuclear fuel. Among the subjects considered are: dose rates available from fuel elements, fission product buildup, chemical behavior of spikants during reprocessing, and possible effects of spikants on refabrication and on the fuel properties.

Neither of the fission product spikants  $^{106}\text{Ru}$  or  $^{144}\text{Ce}$  is present in sufficient quantity to produce the maximum radiation dose rate level considered. Nonradioactive nuclides of ruthenium and cerium dilute the radioactive nuclides to 2-4% of the total element in the fission products two years after removal from the reactor. Recycling ruthenium and cerium will result in dilution of the radionuclides even further by a buildup of stable isotopes of each of these elements. Approximately 50% of the fission product ruthenium and 3-5% of the cerium can be co-processed with the fuel, while cobalt cannot be co-processed at all.

No single radionuclide was found to be preferred in all stages of reprocessing and refabrication. In order to provide deterrence in all stages of reprocessing and refabrication, a duplex spiking process appears necessary, in which two different spikants,  $^{106}\text{Ru}$  and  $^{60}\text{Co}$ , are used in different portions of reprocessing. The use of nominal amounts of ruthenium or cobalt as spikants is not expected to adversely affect fuel performance.

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## INTRODUCTION

The use of a gamma active radionuclide with nuclear fuel has been proposed as a way to inhibit unauthorized diversion of the fuel and thus deter proliferation. Proposed dose rate ranges have varied from small additions to increase detectability of diverted material up to large additions to provide lethal doses in a relatively short exposure time.

Earlier work<sup>1</sup> provided a detailed discussion of the process of spikant selection based on radiation characteristics and possible chemical and physical states. The results of this phase of the selection process are summarized in the present work and have been extended to consider the practical aspects of incorporating the spikants into the fuel cycle. These aspects include spikant production, dose rates available from spent fuel, spikant behavior in the reprocessing, conversion, and refabrication portions of the fuel cycle, and possible effects on fuel performance.

Selection of a dose rate that is high enough to discourage either terrorist activity or national proliferation is beyond the scope of this paper as are possible legal conflicts, such as might develop between the deliberate addition of spikants and the current U.S. Federal law and regulation, which now requires that worker and public exposure risk be held as low as reasonably achievable. Practical considerations of the economics of remote fabrication and other fuel cycle components are not discussed.

## SPIKANT SELECTION

Selection of spikants for deterrence is largely a subjective process, which involves the application of rather arbitrary radiation criteria to available radionuclides. Details of the selection procedure are described in detail in ref. 1 and will not be repeated here.

We analyzed 64 radionuclides to produce a list of primary spikant candidates. Pertinent data on these nuclides and the criteria for rejecting most of them as potential spikants are given in the Appendix.

Considerations of half-life, gamma ray energy, gamma ray intensity, and production method were used to develop a list of primary spikant candidates given in Table 1. Nuclides were included in Table 1 on the basis of useful radiation characteristics and reasonable production rates only, and no consideration was given to physical properties such as melting point and boiling point or to thermodynamic properties. Included in this table is the concentration of the nuclide required for an LWR fuel element ( $\sim 530$  kg heavy metal to produce 27,000 R/h at 0.30 m (1 ft) two years after fabrication. This dose rate will provide a 50% probability of death after 1 min of exposure. These values are for the radionuclide only and do not include other isotopes of the same element. Therefore, actual elemental concentrations will be much higher.

All discussions concerning the concentrations of radioactive spikants are based on the criterion of a radiation dose rate of 27,000 R/h at 0.30 m (1 ft) from a fuel element containing 530 kg  $UO_2$ . The use of other criteria result in different spikant concentrations needed to produce the stipulated radiation dose rate level. This is shown in Table 2, which gives the concentrations of  $^{60}Co$ ,  $^{106}Ru$ , and  $^{144}Ce$  required to produce 1000 R/h at various distances and fuel quantities. The table shows that the dose rate used in this work, 27,000 R/h at 0.30 m (1 ft) from an LWR fuel element containing 530 kg fuel, is not unreasonable compared with other possible selection criteria. In fact the criterion used throughout this report requires only about one-sixth the spikant required to produce 1000 R/hr at 1.0 m from a 5-kg spherical source.

Thermodynamic data<sup>2</sup> are summarized in Fig. 1,<sup>2,3</sup> which gives the free energy of formation for the oxides of the potential spikants and their decay products listed in Table 3. Superimposed on this figure are data for the oxygen potential of various uranium-plutonium fuels. Oxides of rhodium ( $Rh_2O_3$ ), praseodymium ( $Pr_2O_3$ ), and iridium ( $Ir_2O_3$ )

Table 1. Primary Spikant Candidates

Nuclide	Half-Life	Decay Product	Concentration for 27,000 R/h <sup>a</sup> at t = 2 Years (%)	Production Method <sup>b</sup>	Chemical Form	Remarks
<sup>46</sup> Sc	83.9 d	<sup>46</sup> Ti	0.0374	<sup>45</sup> Sc(100%) (n,γ) σ = 23 b	Oxide	
<sup>60</sup> Co	5.26 y	<sup>60</sup> Ni	0.0030	<sup>59</sup> Co(100%) (n,γ) σ = 37 b	Element	
<sup>65</sup> Zn	245 d	<sup>65</sup> Cu	0.0108	<sup>64</sup> Zn(48.9%) (n,γ) σ = 0.46 b	Element	Low σ (n,γ); low bp
<sup>106</sup> Ru	367 d	<sup>106</sup> Pd	0.0343 <sup>c</sup>	Fission product	Element	
<sup>110m</sup> Ag	253 d	<sup>110</sup> Cd	0.0037	<sup>109</sup> Ag(48.6%) (n,γ)	Element	Low intensity; low bp (Cd)
<sup>124</sup> Sb	60 d	<sup>124</sup> Te	0.88	<sup>123</sup> Sb(42.8%) (n,γ) σ = 3.3 b	Element	Short half-life; low bp (Sb,Te)
<sup>144</sup> Ce	284 d	<sup>144</sup> Nd	0.422 <sup>d</sup>	Fission Product	Oxide	Low intensity
<sup>194</sup> Os	60 y	<sup>194</sup> Pt	0.84	<sup>192</sup> Os(41%) (n,γ) σ = 1 b <sup>193</sup> Os(31%) (n,γ) σ = 200 b	Element	Low intensity

<sup>a</sup>For lower dose rates, the required concentration will decrease proportionally.

<sup>b</sup>Cross sections are in barns; 1 b = 1 × 10<sup>-28</sup> m<sup>2</sup>.

<sup>c</sup>Total ruthenium equals 50 times <sup>106</sup>Ru 2 years after removal from reactor.

<sup>d</sup>Total cerium equals 37 times <sup>144</sup>Ce 2 years after removal from reactor.

Table 2. Nuclide Concentrations Required to Produce Various Dose Rates at Various Distances and Fuel Quantities

Nuclide	Concentration for Each Condition, %			
	27,000 R/h at 0.30 m Fuel Element, 530 kg	1000 R/h at 1.00 m 5 kg Spherical	1000 R/h at 0.30 m 1 kg Spherical	1000 R/h at 0.30 m 5 kg Spherical
$^{60}\text{Co}$	0.0030	0.0195	0.0098	0.0022
$^{106}\text{Ru}$	0.0343	0.22	0.1114	0.0251
$^{144}\text{Ce}$	0.422	2.715	1.372	0.3087

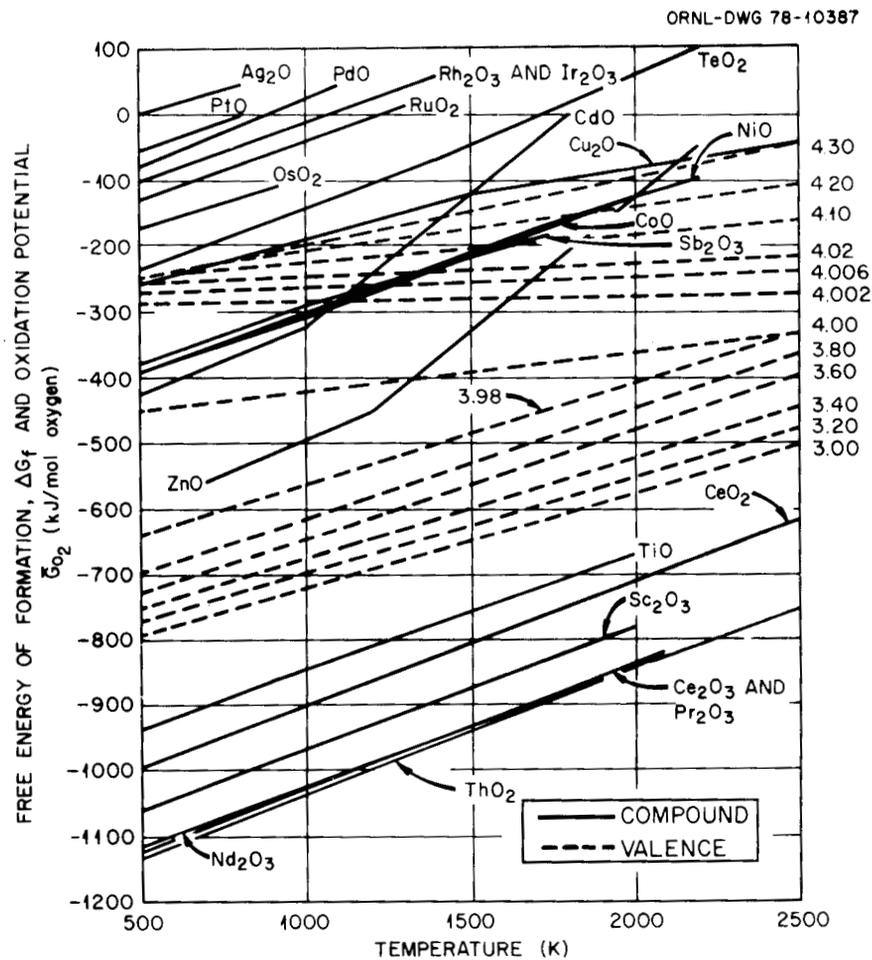


Fig. 1. Thermodynamic Data for Potential Spikants and Decay Products.

Table 3. Melting and Boiling Points of Various Spikant Elements and Their Decay Products in Elemental and Oxide Form and Probable State

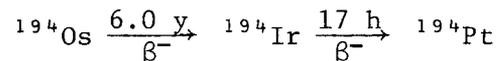
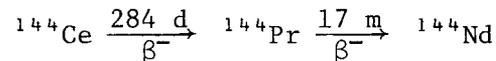
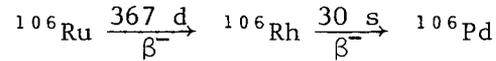
Element	Element Property, °C		Probable Form <sup>a</sup>	Oxide	Oxide Property, <sup>b</sup> °C	
	Melting Point	Boiling Point			Melting Point	Boiling Point
<u>Spikants</u>						
Sc	1539	2730	Sc <sub>2</sub> O <sub>3</sub>	Sc <sub>2</sub> O <sub>3</sub>	(2227)	<sup>c</sup>
Co	1495	2900	Metal	CoO	1805	(2627)
Zn	420	906	Metal	ZnO	1975	
Ru	2500	4900	Metal	RuO <sub>2</sub>	1127 <sup>c</sup>	
				RuO <sub>4</sub>	27	<sup>c</sup>
Rh	1966	4500	Metal	Rh <sub>2</sub> O <sub>3</sub>	1115 <sup>c</sup>	
Ag	961	2210	Metal	Ag <sub>2</sub> O	187 <sup>c</sup>	
Sb	630	1380	Metal	Sb <sub>2</sub> O <sub>3</sub>	655	1425
Ce	804	3470	CeO <sub>2</sub>	Ce <sub>2</sub> O <sub>3</sub>	1687	(3227)
				CeO <sub>2</sub>	2727	
Pr	919	3020	Pr <sub>2</sub> O <sub>3</sub>	Pr <sub>2</sub> O <sub>3</sub>	(1927)	
				PrO <sub>2</sub>	427 <sup>c</sup>	
Os	~2700	~4900	Metal	OsO <sub>2</sub>	650 <sup>c</sup>	
Ir	2454	5500	Metal	Ir <sub>2</sub> O <sub>3</sub>	(1177)	(1977)
<u>Decay Products</u>						
Ti	1668	3260	TiO	TiO	1737 <sup>c</sup>	
				Ti <sub>2</sub> O <sub>3</sub>	2127	3027
				Ti <sub>3</sub> O <sub>5</sub>	(2177)	(3327)
Ni	1435	2730	Metal	NiO	1957	<sup>c</sup>
Cu	1083	2593	Metal	Cu <sub>2</sub> O	1230	<sup>c</sup>
Pd	1552	3980	Metal	PdO	877 <sup>c</sup>	
Cd	321	795		CdO	<sup>c</sup>	
Te	450	990	Metal	TeO <sub>2</sub>	733	<sup>c</sup>
			vapor			
Nd	1019	3180	Nd <sub>2</sub> O <sub>3</sub>	Nd <sub>2</sub> O <sub>3</sub>	2272	
Pt	1769	4530	Metal	PtO	507 <sup>c</sup>	
				PtO <sub>2</sub>	450	477 <sup>c</sup>

<sup>a</sup> Assumes fuel oxygen-to-metal ratio of 1.998 or less.

<sup>b</sup> Values in parentheses are estimated.

<sup>c</sup> Decomposes.

are included in Fig. 1 because these elements would be present as transients in the spikants  $^{106}\text{Ru}$ ,  $^{144}\text{Ce}$ , and  $^{194}\text{Os}$  with the following decay schemes:



Although rhodium, praseodymium, and iridium should be present only in small amounts, they are included to provide a complete analysis of all possible constituents in fuel-spikant systems.

In the presence of  $(\text{U,Pu})\text{O}_{2-x}$ , the compounds  $\text{TiO}$ ,  $\text{Sc}_2\text{O}_3$ ,  $\text{Ce}_2\text{O}_3$ ,  $\text{CeO}_2$ ,  $\text{Pr}_2\text{O}_3$ , and  $\text{Nd}_2\text{O}_3$  are stable, while all the other oxides are unstable. For  $(\text{U,Pu})\text{O}_{2.0}$ ,  $\text{ZnO}$  is stable below about 1300 K, while above this temperature zinc is present in the elemental (vaporized) form. As the cation valence of the fuel increases,  $\text{CoO}$ ,  $\text{NiO}$ ,  $\text{Sb}_2\text{O}_3$ , and  $\text{CdO}$  become stable below about 1100 K, for a valence of 4.002. As the fuel valence increases, the temperature of stability also increases. Thus, as uranium and plutonium are fissioned and oxygen is released from the fuel, the oxygen-to-metal ratio will increase, and certain oxides can become stable at lower temperatures. However, the temperature range of stability is too low to be useful for stabilizing volatile elements such as zinc and antimony.

Melting and boiling point data<sup>2,4</sup> for the elements under consideration and their oxides are summarized in Table 3. Also given in Table 3 is the probable form of each spikant and its decay product, based on the information given in Fig. 1.

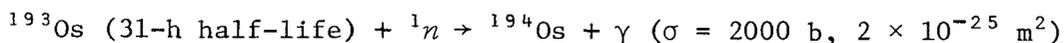
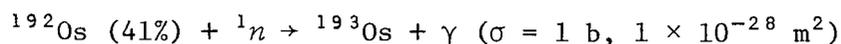
Spikants that would be present as oxides are  $^{46}\text{Sc}$  and  $^{144}\text{Ce}(\text{Pr})$ . The decay products of these nuclides are titanium and neodymium, respectively, which would also be present as oxides. Cerium and neodymium should be soluble in the fuel and thus should be indistinguishable from the fuel matrix. Scandium and titanium may not be completely soluble and may appear as second-phase oxides.

The potential spikant  $^{65}\text{Zn}$  would volatilize at temperatures above  $906^\circ\text{C}$  and migrate to colder portions of the fuel pin. At temperatures between  $420$  and  $906^\circ\text{C}$  zinc is in the liquid state, in which it has been shown<sup>5</sup> to be very corrosive. This makes zinc undesirable as a spikant. Decay products cadmium and tellurium vaporize at  $765$  and  $990^\circ\text{C}$ , respectively, and would also migrate to cooler portions of the fuel pin, causing severe corrosion problems. Thus, the spikants  $^{110\text{m}}\text{Ag}$  and  $^{124}\text{Sb}$  are also considered to be undesirable. Antimony itself is volatile above  $1380^\circ\text{C}$ .

The remaining spikants  $^{60}\text{Co}$ ,  $^{106}\text{Ru}$ , and  $^{184}\text{Os}$  would also be present in the elemental form and would form a second phase in the grain boundaries of the columnar grain region or in the central void. In later stages of burnup these elements would form a homogeneous alloy with fission products such as rhodium, palladium, and molybdenum. The decay products nickel, palladium, and platinum would behave similarly.

The analysis is summarized in Table 4, which lists the potential problems of the various decay schemes. Even though scandium should exist as the oxide ( $\text{Sc}_2\text{O}_3$ ), the half-life of  $^{46}\text{Sc}$  is rather short (83.9 d), which decreases the value of this nuclide as a spikant.

Because the intensity of the gamma radiation provided by  $^{194}\text{Os}$  is quite low, 0.84% of the nuclide is required to provide the stipulated radiation level. This nuclide can be produced by the following reactions:



The efficiency of these reactions for producing  $^{194}\text{Os}$  can be expected to be low since two ( $n, \gamma$ ) reactions are required, which not only raises the cost of production but results in a low nuclide concentration. Even if the  $^{194}\text{Os}$  isotopic concentration would be as high as 10% the total osmium content of the fuel matrix would be at least 8%. This is considered excessive, particularly when it would be present as

Table 4. Summary Analysis of Primary Spikant Candidates

Nuclide	Decay Product	Probable Form	Potential Problems
Sc		Oxide	Short half-life
	Ti	Oxide	
Co		Metal	
	Ni	Metal	
Zn		Metal	Volatile, corrosive liquid
	Cu	Metal	
Ru(Rh)		Metal	
	Pd	Metal	
Ag		Metal	
	Cd	Metal	Volatile, corrosive liquid
Sb		Metal	Volatile, short half-life
	Te	Metal	Volatile, corrosive liquid
Ce(Pr)		Oxide	
	Nd	Oxide	
Os(Ir)		Metal	Low-intensity gammas
	Pt	Metal	

the metal, dispersed in the fuel matrix, probably at the grain boundaries. For this reason  $^{194}\text{Os}$  can be eliminated from further consideration for the stipulated radiation level.

Consideration of these factors narrows the list of primary candidates to  $^{60}\text{Co}$ ,  $^{106}\text{Ru}$ , and  $^{144}\text{Ce}$ . Of these, only cerium would be present as the oxide, soluble in the fuel. Cobalt and ruthenium would be present in the elemental form, as separate phase metallic particles dispersed in the fuel matrix. Cerium and ruthenium have the advantage that they are present as fission products in the spent fuel, so that it would not be necessary to set up a separate facility to produce the nuclides by irradiation. However, reprocessing schemes would have to be altered to separate the cerium and ruthenium from other fission products.

Phase relationships between stoichiometric  $\text{PuO}_2$ ,  $\text{ThO}_2$  and  $\text{UO}_2$  show complete solid solubility in the binary systems.<sup>3,6-8</sup> This should extend to the ternary system as well, although no ternary diagram was found in the literature. Large deviations from stoichiometry occur in both the Pu-O and U-O systems, with the single phase still maintained.

However, these deviations occur in opposite directions, namely,  $\text{PuO}_{2-x}$ <sup>9-11</sup> and  $\text{UO}_{2+x}$ .<sup>12</sup> A wide range of single-phase solid solution exists<sup>13,14</sup> between  $\text{PuO}_{2-x}$  and  $\text{UO}_{2+x}$ .

Slight substoichiometry is possible<sup>15</sup> in the Th-O system at temperatures above 1500°C. However, substoichiometry has not been reported in the Th-U-O system. Deviations from stoichiometry in this system are toward hyperstoichiometric compositions,  $\text{MO}_{2+x}$ . No data were found on the  $\text{PuO}_{2-x}$ - $\text{ThO}_2$  system.

Table 5 summarizes the phase relationships for the various spikants and decay products with  $\text{PuO}_2$ ,  $\text{ThO}_2$  and  $\text{UO}_2$ . Extensive solubility is found between fuel constituents and the rare earth oxides  $\text{CeO}_2$ ,  $\text{PrO}_x$ , and  $\text{Nd}_2\text{O}_3$ . No solubility data were reported for cobalt, nickel, ruthenium or their oxides.

Table 5. Summary of Phase Relationships Between Potential Spikants or Decay Products and Fuel Components

Spikant	Ref.	Observation
$\text{CeO}_2$	16	Soluble in all proportions with $\text{PuO}_2$
	17-19	Soluble in all proportions with $\text{ThO}_2$
	20-22	Soluble in all proportions with $\text{UO}_2$
$\text{PrO}_x$	22	Complete solubility with $\text{ThO}_2$
$\text{CoO}$	24	$\text{CoU}_2\text{O}_6$ formed with $\text{UO}_2$
$\text{Nd}_2\text{O}_3$	25	Soluble in $\text{ThO}_2$ : about 35 mol % at 1400°C, <40 mol % at 2200°C
	26-29	Extensive solubility in $\text{UO}_2$
$\text{NiO}$	30	Compounds formed with $\text{UO}_2$ ; no solubility data reported
$\text{RuO}_2$		No information available

#### Fission Product Spikants

Potential spikants derivable from fission products are compiled in Table 6. Of the nuclides in this table,  $^{95}\text{Nb}$ ,  $^{95}\text{Zr}$ , and  $^{103}\text{Ru}$  can be eliminated from further consideration on the basis of their short half-lives. The nuclide  $^{125}\text{Sb}$  is undesirable because of the high vapor pressures of both antimony and the decay product tellurium, and  $^{137}\text{Cs}$

Table 6. Summary of All Potential Fission Product Spikants

Radionuclide	Decay Product	Half-Life	Concentration for 27,000 R/h After 2 Years (%)	Criteria for Possible
$^{85}\text{Kr}$	$^{85}\text{Rb}$	10.76 y	0.0825	Gaseous; low gamma energy
$^{95}\text{Nb}$	$^{95}\text{Mo}$	35 d	370	Short half-life; low gamma energy
$^{95}\text{Zr} \downarrow$ $^{95}\text{Nb}$	$^{95}\text{Mo}$	65 d 35 d	0.59	Short half-life; low gamma energy
$^{103}\text{Ru}$	$^{103}\text{Rh}$	39.6 d	135	Short half-life; low gamma energy
$^{106}\text{Ru} \downarrow$ $^{106}\text{Rh}$	$^{106}\text{Pd}$	367 d 30 s	0.0343	Low gamma intensity
$^{125}\text{Sb}$	$^{125}\text{Te}$	2.7 y	0.0220	Low gamma energy; high vapor pressure (Sb and Te)
$^{137}\text{Cs}$	$^{137}\text{Ba}$	30.0 y	0.125	Low gamma energy; low boiling point (Cs) corrosive
$^{144}\text{Ce} \downarrow$ $^{144}\text{Pr}$	$^{144}\text{Nd}$	284 d 17 m	0.422	Low gamma intensity

is undesirable because of its low boiling point and corrosiveness. The inert gas nuclide  $^{85}\text{Kr}$  cannot be incorporated into the fuel, so it is not useful.

This leaves only  $^{106}\text{Ru}$  and  $^{144}\text{Ce}$  as potential candidates, which substantiates their inclusion in Table 1.

#### Potential Spikants Produced by Other Production Methods

Potential spikants listed in Tables 1 and 6 are those that can be produced by  $(n, \gamma)$  reactions or are present as fission products. Nuclides produced by  $(n, 2n)$  reactions or by charged particle ( $\alpha$ -particle or proton) interaction with specific isotopes were not included in these tables because of the inefficiency of production and resulting high cost

of these radionuclides. Radionuclides produced by these reactions would usually have the advantage that they can be chemically separated from the target nuclei so that buildup of stable nuclides may not be a problem.

If we can assume that the cost of production of spikants is not a limiting factor, the radionuclides listed in Table 7 have potential for use as spikants. Arguments presented earlier on the vapor pressures of either spikants or decay products eliminate  $^{54}\text{Mn}$ ,  $^{68}\text{Ge}$ , and  $^{194}\text{Hg}$ . The half-life of  $^{88}\text{Y}$  of 108 d is too short to be of use as a spikant, and the half-life of  $^{184\text{m}}\text{Re}$  of 169 d is marginal.

The rare earth radionuclides  $^{143}\text{Pm}$ ,  $^{144}\text{Pm}$ ,  $^{146}\text{Pm}$ , and  $^{150}\text{Eu}$  are trivalent elements and as such would be extremely difficult to separate from fission product rare earths, many of which act as neutron absorbers. This objection actually applies only to  $^{146}\text{Pm}$  and  $^{150}\text{Eu}$ , since their half-lives are long enough (5.5 and 6.2 years, respectively) that they could be recycled. If they would be recycled, the  $^{146}\text{Pm}$  would be diluted to about 58% of the total element in the fission products and the  $^{150}\text{Eu}$  would be diluted to about 38% of the total element in the fission products. In addition, the europium fission products contain many nuclides with very high cross sections for thermal neutrons, so this option is even less desirable. The cross section for thermal neutrons for  $^{146}\text{Pm}$  is  $8.4 \text{ E-}25 \text{ m}^2$ , which is high enough to interfere with the fission process. Recycling of  $^{143}\text{Pm}$  and  $^{144}\text{Pm}$  is not practical because of their relatively short half-lives of 267 and 401 d, respectively. Assuming a 4.4-year cycle (2.4 years in reactor + 2 years cool down and recycle), most of these nuclides will decay out. However, separation from the rare earth target material will be extremely difficult so that dilution of the radionuclide with the target material is likely.

This leaves  $^{102}\text{Rh}$ ,  $^{102\text{m}}\text{Rh}$ , and  $^{172}\text{Hf}$  as possible candidate spikants. The longer half-lives of  $^{102\text{m}}\text{Rh}$  and  $^{172}\text{Hf}$  of 2.1 and 5.0 years, respectively make them attractive. After the first recycle  $^{102}\text{Rh}$  and  $^{102\text{m}}\text{Rh}$  would be diluted by rhodium fission products to about 1.8% of the total rhodium in the fuel. Thus, in order to incorporate 0.0034%  $^{102\text{m}}\text{Rh}$  into the fuel, 0.19% Rh would have to be added to the fuel. This is not considered excessive. However, subsequent recycling will increase the

Table 7. Summary of Potential Spikants Produced by (n,2n) or Charged Particle Reactions

Radionuclide	Decay Product	Half-Life	Concentration for 27,000 R/h After 2 Years (%)	Possible Production Methods <sup>a</sup>	Criteria for Possible Rejection
<sup>54</sup> Mn	<sup>54</sup> Cr	303 d	0.0049	<sup>55</sup> Mn(100%) (n,2n)	Low gamma energy; high vapor pressure (Mn)
<sup>68</sup> Ge ↓ <sup>68</sup> Ga	<sup>68</sup> Zn	275 d 68 m	0.0057	<sup>66</sup> Zn(28%) (α,2n)	Low gamma intensity; high vapor pressure (Zn)
<sup>88</sup> Y	<sup>88</sup> Sr	108 d	0.0204	<sup>89</sup> Y(100%) (n,2n) <sup>88</sup> Sr(83%) (p,n)	Short half-life
<sup>102</sup> Rh	<sup>102</sup> Ru	206 d	0.022	<sup>103</sup> Rh(100%) (n,2n) <sup>102</sup> Pd(0.95%) (n,p) <sup>102</sup> Ru(32%) (p,n)	Low gamma intensity
<sup>102m</sup> Rh	<sup>102</sup> Ru	2.1 y	0.0034	<sup>102</sup> Ru(32%) (p,n)	
<sup>143</sup> Pm	<sup>143</sup> Nd	267 d	0.034	<sup>141</sup> Pr(100%) (α,2n) <sup>144</sup> Nd(24%) (p,2n)	Low gamma intensity
<sup>144</sup> Pm	<sup>144</sup> Nd	401 d	0.0061	<sup>144</sup> Sm(3.2%) (n,p) <sup>141</sup> Pr(100%) (α,n) <sup>144</sup> Nd(24%) (p,n)	Low gamma intensity
<sup>146</sup> Pm	<sup>146</sup> Nd	5.5 y	0.0219	<sup>146</sup> Nd(17%) (p,n)	Low gamma energy
<sup>150</sup> Eu	<sup>150</sup> Sm	6.2 y	0.0176	<sup>151</sup> Eu(48%) (n,2n) <sup>150</sup> Sm(7.5%) (p,n)	Low gamma intensity
<sup>172</sup> Hf ↓ <sup>172</sup> Lu	<sup>172</sup> Yb	5 y 6.7 d	0.0167	<sup>175</sup> Lu(97%) (p,4n)	
<sup>184m</sup> Re ↓ <sup>184</sup> Re	<sup>184</sup> W	169 d 35 d	0.050	<sup>185</sup> Re(37.1%) (n,2n) <sup>184</sup> W(31%) (p,n)	Low gamma energy
<sup>194</sup> Hg ↓ <sup>194</sup> Au	<sup>194</sup> Pt	~1.3 y 39.5 h	0.045	<sup>197</sup> Au(100%) (p,4n)	Low gamma intensity; high vapor pressure (Hg)

<sup>a</sup>Target nuclide, its isotopic abundance, and production reaction.

amount required, because of buildup of stable isotopes of rhodium. Hafnium should be present in the fuel as the oxide, and in the quantity required, 0.0167%, should be soluble in the fuel. Thus, it would not constitute an additional phase in the system and its effect on the properties of the fuel should be minimal.

#### Dose Rates Available from Fission Product Spikants $^{106}\text{Ru}$ and $^{144}\text{Ce}$

The nuclide concentrations required to produce 27,000 R/h at 0.305 m for 530 kg of fuel after 2 years, given in Table 1, assume that the isotope is 100% pure. In reality this is never the case since other isotopes of the same element are present and decrease the isotopic concentration of the spikant. Figures 2 and 3 summarize the isotopic concentrations of the fission products  $^{106}\text{Ru}$  and  $^{144}\text{Ce}$ , respectively, as a function of time since removal from the reactor for four different reactor systems. These concentrations were obtained from the ORIGEN Code<sup>3</sup> under the conditions summarized in Table 8. One year after removal from the reactor, none of the systems indicate isotopic concentrations of either  $^{106}\text{Ru}$  or  $^{144}\text{Ce}$  greater than 6.5%. Two years after removal from the reactor the isotopic concentrations are less than 3%. For a PWR this means that to satisfy the criterion of 27,000 R/h 2 years after fuel refabrication (total of 4 years after removal from the reactor), the cerium concentration would have to be 15.6% and the ruthenium concentration about 1.7%. Neither of these values is attainable from fission products for the stipulated radiation level.

Radiation levels attainable as a function of time after removal from the reactor are summarized in Fig. 4. This figure assumes that 100% of each fission product is processed with the fuel (100% recovery of either  $^{106}\text{Ru}$  or  $^{144}\text{Ce}$ ). The maximum dose rate attainable from 530 kg of fuel would be about 2000 R/h for  $^{144}\text{Ce}$  and about 13,000 R/h for  $^{106}\text{Ru}$  from a PWR and about 27,000 R/h for  $^{106}\text{Ru}$  from a LMFBF fuel element.

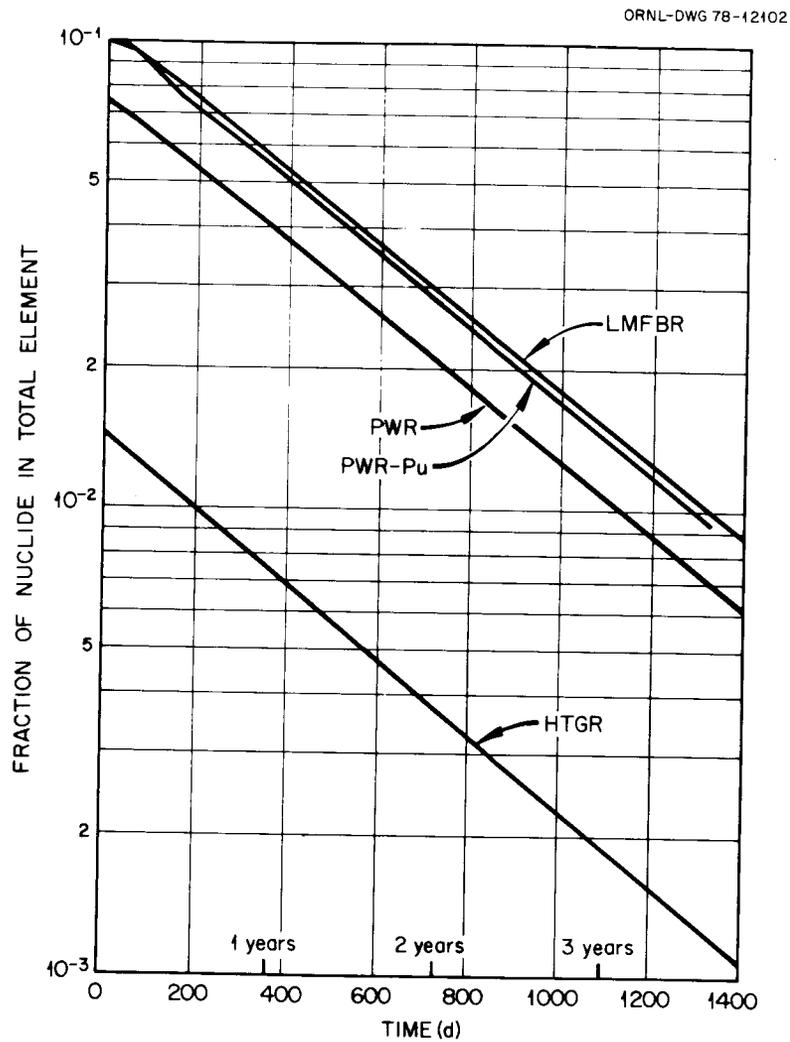


Fig. 2. Fraction of  $^{106}\text{Ru}$  in Total Element as a Function of Time After Removal from Reactor.

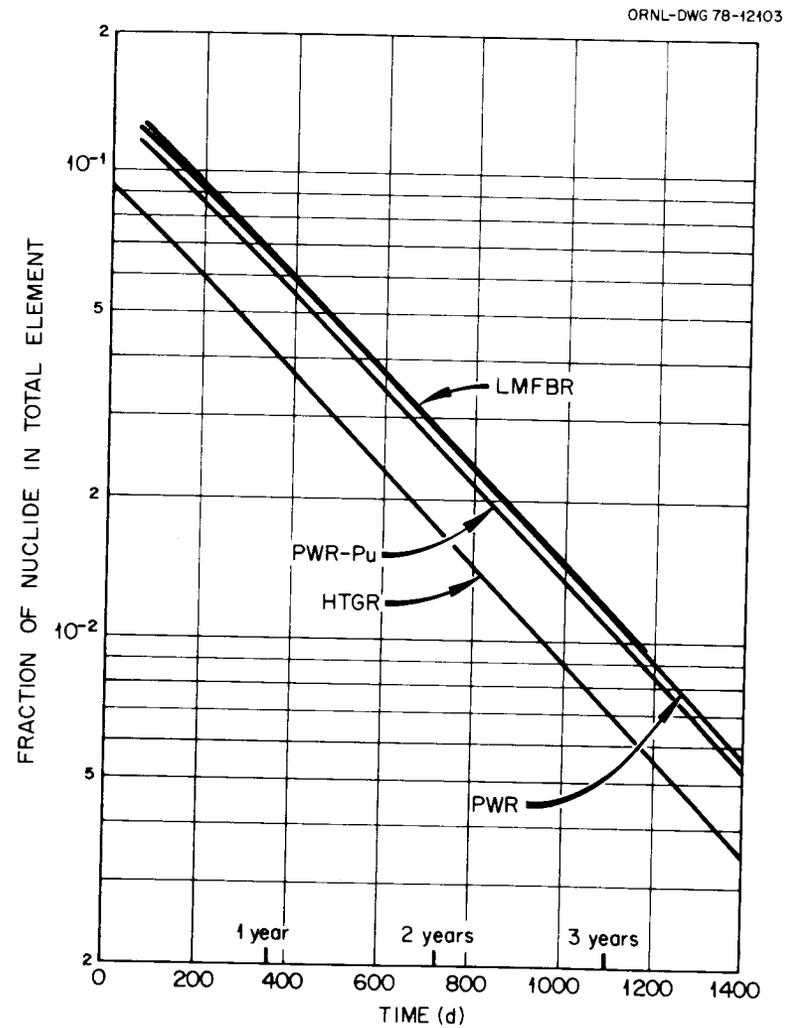


Fig. 3. Fraction of  $^{144}\text{Ce}$  in Total Element as a Function of Time After Removal from Reactor.

Table 8. Conditions Used for ORIGEN Calculations of Fission Product Concentration

Reactor	Flux ( $n/m^2 s$ )	Power (MW/tonne)	Burnup (MWd/tonne)
PWR-U	$3.89 \times 10^{17}$	37.50	33,000
PWR-Pu	$2.06 \times 10^{17}$	37.50	33,000
HTGR	$8.04 \times 10^{17}$	64.57	94,271
LMFBR	$2.49 \times 10^{19}$	49.28	37,118

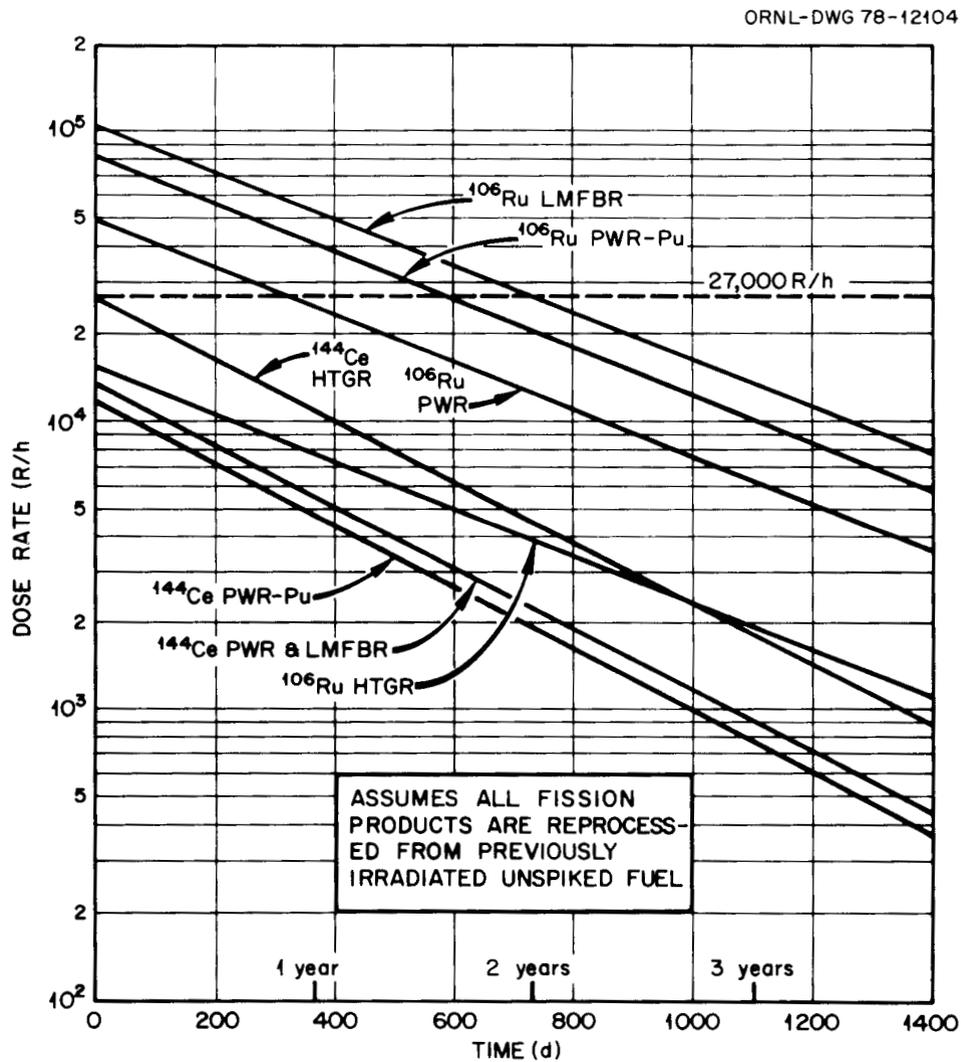


Fig. 4. Dose Rate as a Function of Time After Removal from the Reactor.

### Determination of the Amount of $^{59}\text{Co}$ Required to Produce $^{60}\text{Co}$

Some radionuclides are produced by neutron irradiation of specific isotopes. For example,  $^{60}\text{Co}$  is produced by the irradiation of  $^{59}\text{Co}$  in a reactor and is available commercially. However,  $^{60}\text{Co}$  could be produced in situ by the introduction of  $^{59}\text{Co}$  into the fuel.

We calculated the amount of  $^{59}\text{Co}$  necessary to produce enough  $^{60}\text{Co}$  to provide 27,000 R/h in PWRs and LMFBRs. These calculations used the conditions given in Table 9. Losses due to neutron capture by  $^{60}\text{Co}$  were ignored. If one assumes that no  $^{60}\text{Co}$  is present at the beginning of the first irradiation cycle in a PWR, 293 ppm  $^{59}\text{Co}$  by weight (referred to the oxide) will produce 39 ppm  $^{60}\text{Co}$ , which will decay to the required 30 ppm by the time the fuel is reprocessed, refabricated, and inserted back into the reactor. A similar calculation for the LMFBR shows that 1.64%  $^{59}\text{Co}$  would be required.

Table 9. Conditions Assumed in  $^{59}\text{Co}$  Calculations

	PWR	LMFBR
Time in reactor, years	2.41	2.0
Cool-down time, years	0.5	0.5
Reprocess, refabricate, years	1.5	1.5
Flux, $\text{n/m}^2 \text{ s}$	$3.8 \text{ E} + 17$	$2.5 \text{ E} + 19$
$^{59}\text{Co}$ cross section, $\text{m}^2$	$3.7 \text{ E} - 29$	$1.2 \text{ E} - 30$

Under equilibrium conditions, a minimum of 30 ppm  $^{60}\text{Co}$  is maintained in the fuel along with enough  $^{59}\text{Co}$  to replace  $^{60}\text{Co}$  lost by decay during irradiation, cool-down, reprocessing, and refabrication. For the PWR, the amount of  $^{59}\text{Co}$  was found to be 132 ppm, and for the LMFBR, the amount is 0.69%. This large difference is due to the very low cross section of  $^{59}\text{Co}$  for fast neutrons compared with that for thermal neutrons. The concentration of  $^{60}\text{Co}$  in the total amount of cobalt present is therefore 18.5% for the PWR case and 0.043% for the LMFBR.

The large amount of  $^{59}\text{Co}$  needed to produce the required amount of  $^{60}\text{Co}$  in an LMFBR suggests that a more practical approach would be to produce  $^{60}\text{Co}$  in thermal reactors for use in an LMFBR.

At the present time, co-processing of cobalt with the fuel is not possible, nor can it be separated from the fission product stream. Therefore the  $^{59}\text{Co}$  cannot be intimately mixed with the fuel as part of the fuel matrix. Instead, wafers or shims could be strategically placed within the fuel element and physically separated during recycle.

### REPROCESSING

Any proposed fuel spikant system must consider potential reprocessing problems resulting from the introduction of spikants into the fuel. Questions that need to be addressed include:

1. the chemistry of fuel spikant systems as related to the solvent extraction steps,
2. expected yields of fission product spikants, and
3. modifications of the process flowsheets.

Each of the three spikants — Ru, Ce, and Co — will be briefly reviewed with respect to its effect on the fuel reprocessing.

Ruthenium — The chemistry of ruthenium in the solvent extraction system is complicated, and the paths through the fuel reprocessing system depend on many parameters. When fuel is dissolved for recovery, the dissolver solution contains at least four chemical forms of ruthenium.<sup>32</sup> The solvent extraction partition coefficients of these species vary by at least 5 orders of magnitude. One of these has a higher distribution coefficient than any other fission product. The equilibrium between the various species, the rate of conversion from one species to another, and the extraction by the solvent depend on the acidity, temperature, TBP concentration, and the presence of other ions (such as nitrite and bisulfite) in the solution.

In view of this complexity, any process for the extraction of ruthenium with the fuel will require careful control. Because of this, considerable development would be required to develop a reliable process.

At the present time it is anticipated that with 2.5 *M* acid solution 10–20% of the fission product ruthenium could be processed with the fuel. Decreasing the acid content to about 1 *M* should increase this amount to about 50%. The total amount of ruthenium recovered can be increased further by reprocessing the extracted stream and physically mixing the recovered ruthenium with the fuel later in the process, after conversion from the nitrate to the oxide.

Cerium — Extraction of cerium in a form free from other rare earth neutron poisons is a difficult task and, at the present time, it is felt that a process could be designed to extract 3–5% of the available cerium with the fuel. This can be increased by conversion of Ce(III) in the fission product stream to Ce(IV), which extracts more readily, and adding this material to the fuel later in the reprocessing step. Separation depends strongly on the oxidation potential of the solution.<sup>33</sup>

Cobalt — Cobalt forms complexes with the result that it does not extract with the fuel constituents. Therefore cobalt would have to be produced separately from the fuel and fed into the product stream. This addition can be made either as the nitrate to the nitrate products or as the oxide to the converted product.

While there appears to be no doubt that reprocessing of spikants to some degree is possible, it also appears probable that at least a portion of the spikants will have to be added back into the recovered fuel later in the reprocessing stage. This means that portions of the reprocessing facility will not contain the prescribed radiation levels and, as a consequence, the fuel will be more susceptible to diversion from these locations. This problem may possibly be overcome by a duplex process involving two or more different spikants. This will be discussed in a later section.

#### Fission Product Buildup

Fission products have limited usefulness as spikants for diversion. The advantage of fission products is that they are produced in the fuel as a direct result of the fission process so that special reactors are

not necessary for their production. If the useful nuclides such as  $^{106}\text{Ru}$  and  $^{144}\text{Ce}$  were the only isotopes of these elements, there would be no problem. However, other isotopes of ruthenium and cerium are also produced as fission products, resulting in the dilution of the radio-nuclides by stable nuclides of the same elements.

At present, no reasonable method is known for separating isotopes of ruthenium and cerium. Therefore, each time the fission product spikants are reprocessed back into a reactor, the amount of stable isotopes of ruthenium and cerium will increase by a constant amount. As a result, the amount of total element required to produce a dose rate level of 27,000 R/h increases with each cycle. This is shown in Fig. 5. This figure shows that after 6 cycles, the ruthenium concentration would have to be about 10% to produce the specified radiation level. The cerium concentration would have to be about 106%, an impossible level. Figure 5 assumes that enough spikant nuclide is produced to provide 27,000 R/h. However, this is not the case, and Fig. 6 shows that ruthenium can provide about 13,000 R/h and cerium can provide about 2000 R/h if all the element is reprocessed.

#### Duplex Spikant Reprocessing

From the discussion on reprocessing, it appears that ruthenium is the only potential spikant that can provide high dose rates during most of the reprocessing steps. Cobalt complexes out during the separation exchange; only 3-5% of the cerium is carried along with the product. Since about 50% of the ruthenium can be coprocessed with the product, a higher level of diversion resistance is provided. However, during conversion to the oxide or sintering the ruthenium can oxidize and vaporize out of the fuel. This would leave the fuel vulnerable to diversion. It can therefore be concluded that none of the potential spikants can individually render the fuel completely diversion-resistant during reprocessing, conversion, and refabrication. This is summarized in Table 10, which gives the relative diversion resistance of the fuel for each potential spikant after the separation exchange and after the conversion

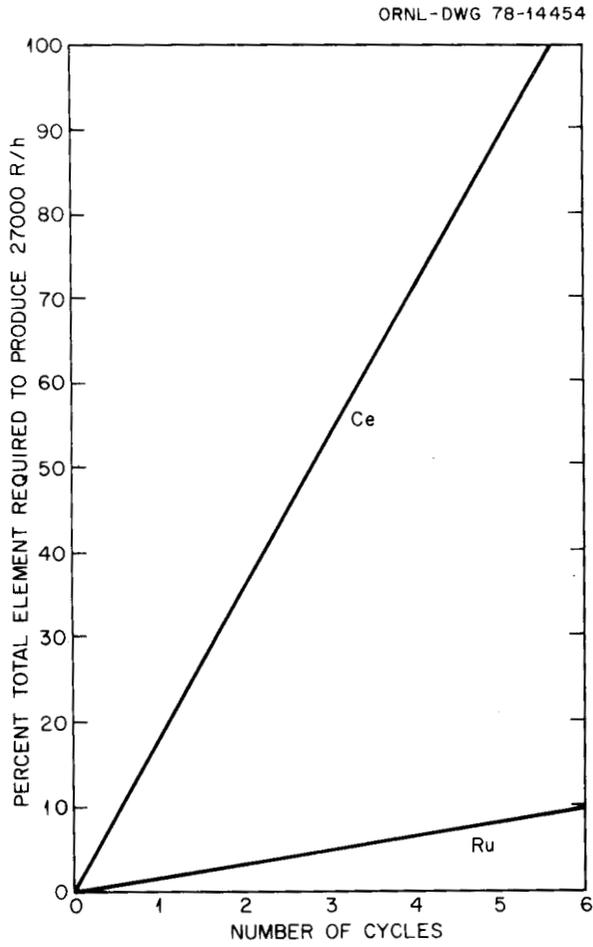


Fig. 5. Total Amount of Ruthenium and Cerium Required to Produce 27,000 R/h After Recycling.

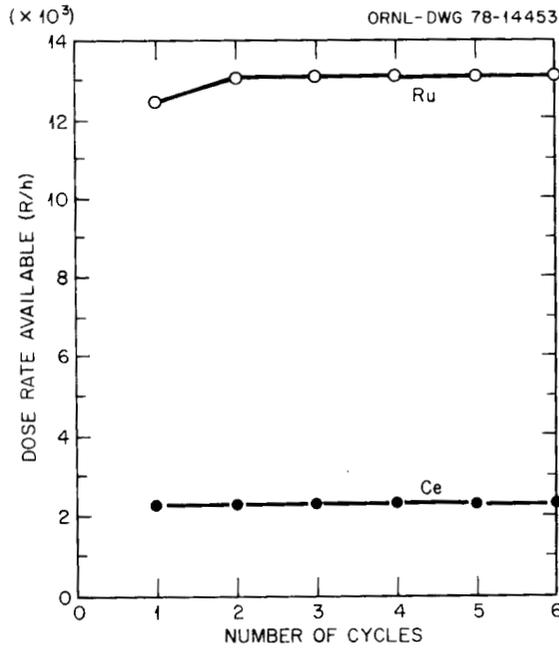


Fig. 6. Dose Rates Available from  $^{106}\text{Ru}$  and  $^{144}\text{Ce}$  After Recycling.

Table 10. Diversion Resistance of Nuclear Fuel  
by Potential Spikants During Reprocessing  
and Refabrication

Radionuclide	After Separation Exchange	After conversion and Refabrication <sup>α</sup>
<sup>60</sup> Co	low	high
<sup>106</sup> Ru	high	low
<sup>144</sup> Ce	low	intermediate

<sup>α</sup>Assumes <sup>60</sup>Co and <sup>144</sup>Ce are separated from the fission product stream and added back into the product stream before conversion.

and refabrication steps. These ratings assume that the <sup>60</sup>Co and <sup>144</sup>Ce are separated from the fission product stream and added back into the product stream before conversion.

This problem can be overcome by a "duplex" spiking procedure in which both <sup>106</sup>Ru and <sup>60</sup>Co are used in different stages of reprocessing to ensure continuous diversion resistance. In this process, <sup>59</sup>Co is incorporated into the fuel or fuel element to produce <sup>60</sup>Co during irradiation. About 300 ppm of <sup>59</sup>Co would be required in a PWR for this purpose. About 50% of the ruthenium can be reprocessed with the fuel while the cobalt is separated from the fuel in the reprocessing head-end steps or during the separation exchange process. The cobalt separated from the head-end step can be reprocessed and added to the fuel as the nitrate. During conversion from the nitrate to the oxide (calcining), the ruthenium might volatilize as the oxide and be condensed on a cold finger and fed into the fission product stream. It might, in fact, be desirable to deliberately volatilize the ruthenium at this step to prevent the buildup of stable isotopes of ruthenium in subsequent reactor cycles. The cobalt should remain with the fuel during calcining and sintering, thereby maintaining a high dose rate level. During the period of time from the separation exchange column to the point the cobalt is added to the product stream, the dose rate depends on the available <sup>106</sup>Ru, which

should produce about 5000 R/h at 0.30 m from the surface of an LWR fuel element. A partial functional flow diagram showing this scheme is given in Fig. 7.

Diversion resistance by spiking becomes increasingly more difficult as the required dose rate increases. This is shown in Fig. 8, which indicates the spikants that can be used to produce various dose rates while providing limited or maximum diversion resistance. Limited diversion resistance is defined as the case in which there is at least one point during reprocessing where the radiation dose rate is inadequate to provide diversion resistance. Maximum diversion resistance is defined as a case in which an appreciable radiation dose rate ( $\sim 5000$  R/h) is present at all times during reprocessing.

This figure shows that based on a dose rate criterion of 27,000 R/h at 0.30 m from the surface of an LWR fuel element,  $^{60}\text{Co}$  provides only limited diversion resistance. The amount of  $^{106}\text{Ru}$  and  $^{144}\text{Ce}$  in the fission products is inadequate to provide this dose rate level. Since cobalt complexes out of the product stream during the separation exchange, only limited diversion resistance is obtained from  $^{60}\text{Co}$ . For a specified dose rate of  $\sim 10,000$  R/h, both  $^{60}\text{Co}$  and  $^{106}\text{Ru}$  can provide limited diversion resistance. Cerium is useful only for limited diversion resistance at dose rate levels of about 1000 R/h.

## EFFECTS OF SPIKANTS ON FUEL PROPERTIES

### Melting Point

Since the metallic spikants  $^{60}\text{Co}$ ( $^{60}\text{Ni}$ ) and  $^{106}\text{Ru}$ ( $^{106}\text{Pd}$ ) are insoluble in the fuel, no effect on melting point of the fuel would be expected, and no evidence to the contrary has been found. Even if these spikants were added to the fuel as oxides, the oxides would decompose or be reduced to metal at relatively low temperatures ( $<1000^\circ\text{C}$ ).

The rare earth oxides, which should be representative of cerium, form solid solutions with  $\text{UO}_2$ , and probably with  $\text{ThO}_2$  and  $\text{PuO}_2$  also. A phase diagram of the  $\text{UO}_2\text{-Nd}_2\text{O}_3$  system<sup>29</sup> indicates that addition of

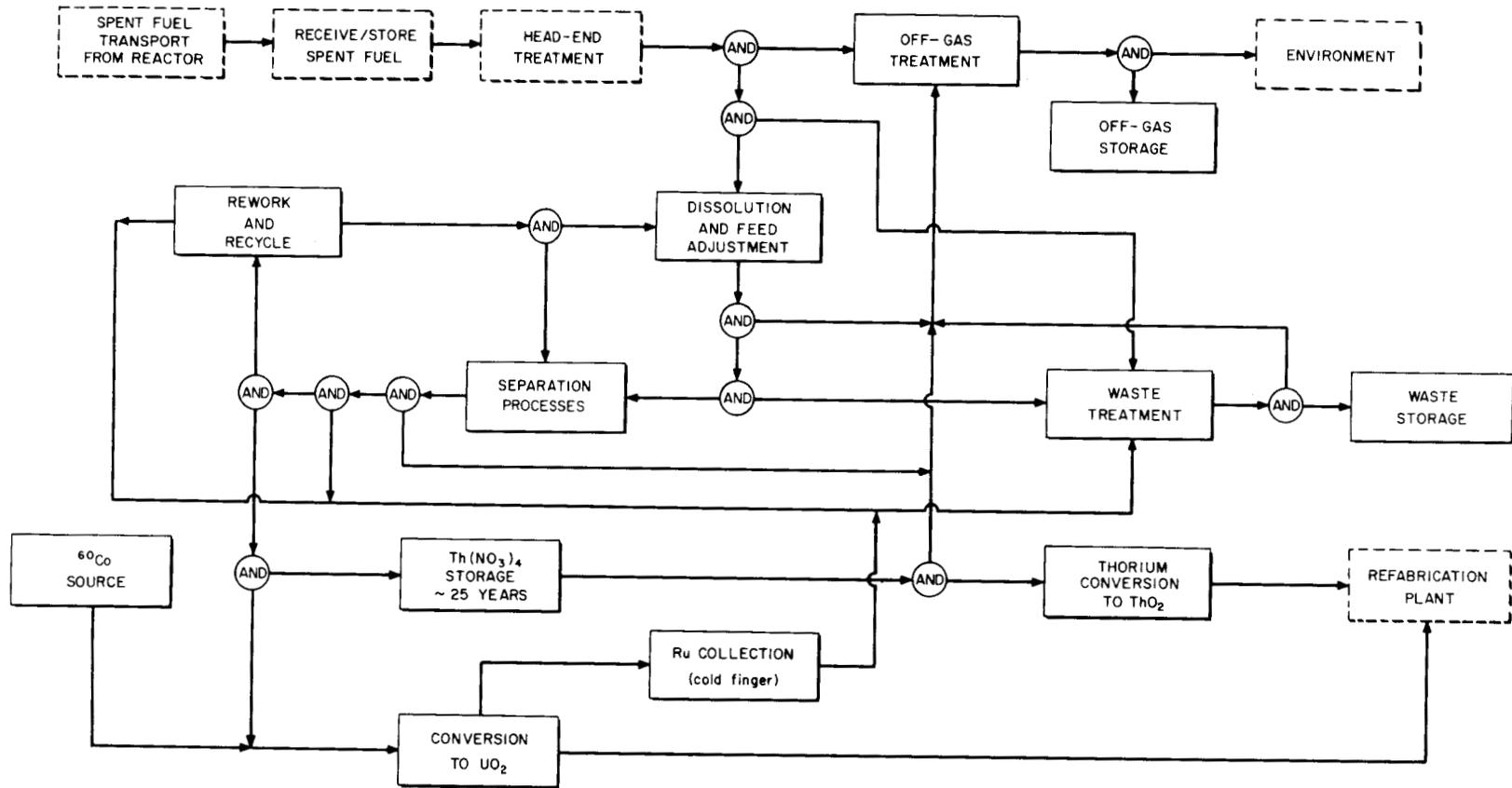


Fig. 7. Partial Functional Flow Diagram of Duplex Spiking Process.

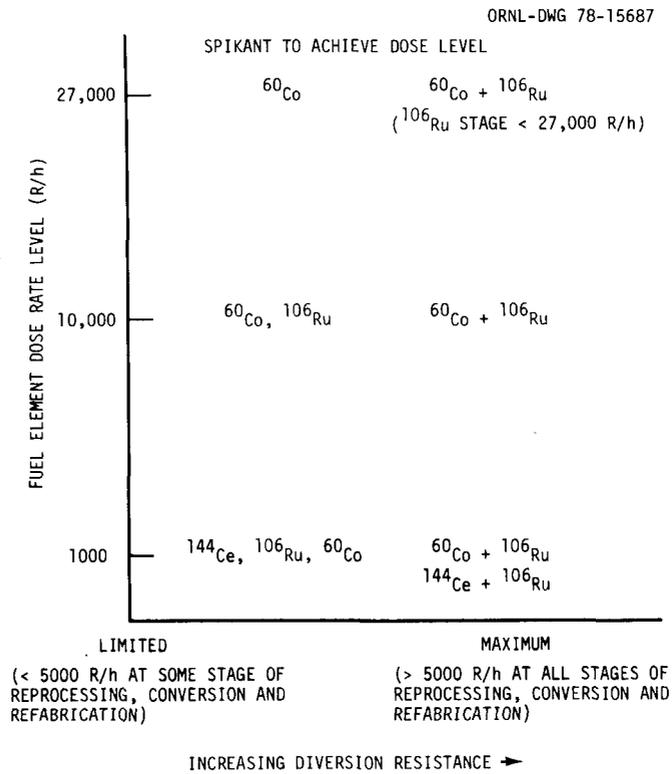


Fig. 8. Useful Spikants at Various Dose Rate Levels and Desired Diversion Resistance.

$\text{Nd}_2\text{O}_3$  to  $\text{UO}_2$  lowers the liquidus temperature of the solid solutions about  $14^\circ\text{C}/\text{mol } \%$   $\text{Nd}_2\text{O}_3$  over the range 0 to 25 mol %  $\text{Nd}_2\text{O}_3$ . Liquidus or solidus data for the  $\text{UO}_2\text{-CeO}_2$  system have not been found.

#### Thermal Properties

Murabayashi<sup>18</sup> used x-ray diffraction and ceramography to verify solid solution in  $(\text{Th,Ce})\text{O}_2$  containing up to 12 mol %  $\text{CeO}_2$ . The thermal conductivity at  $25^\circ\text{C}$  varied from 9.7 to 4.2 W/m k as the  $\text{CeO}_2$  content increased from 1 to 12 mol %. This agrees with the general contention that the thermal conductivity of a ceramic solid solution is lower than that of the individual components.

X-ray diffraction showed complete solid solution in  $\text{UO}_2\text{-Gd}_2\text{O}_3$  mixtures used for several measurements. The thermal expansion coefficient increased from about  $1.05 \times 10^{-5}$  to  $1.17 \times 10^{-5}/^\circ\text{C}$  as the  $\text{Gd}_2\text{O}_3$  content increased from 0 to 24 mol %.

Beisswenger et al.<sup>34</sup> annealed UO-CeO<sub>2</sub> pellets containing up to 19.4 mol % CeO<sub>2</sub> in a temperature gradient of 1400 to 2200°C. The CeO<sub>2</sub> migrated to the higher temperature regions by a thermal diffusion process. The following quantities were measured at 19.4 mol % CeO<sub>2</sub> and 2050°C: diffusion coefficient,  $D = 5.4 \times 10^{-12}$  m<sup>2</sup>/s; thermal diffusion coefficient  $D' = 1.6 \times 10^{-14}$  m<sup>2</sup>/s·K; activation energy,  $Q = 460$  kJ/mol. Thermal migration, however, was not observed<sup>35</sup> in UO<sub>2</sub>-Gd<sub>2</sub>O<sub>3</sub> solid solutions containing up to 24 mol % Gd<sub>2</sub>O<sub>3</sub> as the result of annealing at a maximum temperature of 2100°C with a thermal gradient of 1350°C/cm.

### Mechanical Properties

No information was found on the effects of cerium, ruthenium, or cobalt on the mechanical properties of uranium, plutonium, or thorium oxides. In order to provide a background of possible effects, the results of studies of other elements on fuel properties are included in this section.

Christie and Williams<sup>36</sup> conducted creep-in-bending tests on hypo- and hyperstoichiometric UO<sub>2</sub> containing additions of Y<sub>2</sub>O<sub>3</sub>. The UO<sub>2</sub>-Y<sub>2</sub>O<sub>3</sub> mixtures were prepared by coprecipitation from nitrate solution, calcining in air at 700°C, then reducing in H<sub>2</sub> at 700°C. The oxygen-to-uranium ratios of the powders were varied by heating in air at 140°C. The material was pressed, then cut into rectangular specimens. Nonstoichiometric specimens were sintered at 1400°C in argon for 7.2 ks (2 h), while stoichiometric specimens were prepared from nonstoichiometric ones by sintering at 1400°C in H<sub>2</sub> for 3.6 ks (1 h). Creep tests were done at 940°C at a maximum fiber stress of 35 MPa, in argon for nonstoichiometric specimens and in H<sub>2</sub> for stoichiometric ones. Nonstoichiometric specimens (O/U  $\approx$  2.27) containing approximately 0.5 and 1.0 mol % Y<sub>2</sub>O<sub>3</sub> deformed substantially less than specimens containing no Y<sub>2</sub>O<sub>3</sub>, and stoichiometric specimens containing Y<sub>2</sub>O<sub>3</sub> deformed hardly at all. If grain boundary flow contributes appreciably to overall deformation, the lower creep rates in specimens containing Y<sub>2</sub>O<sub>3</sub> might be related to the larger grain size, which was five to ten times larger in specimens containing Y<sub>2</sub>O<sub>3</sub> than in pure UO<sub>2</sub>.

Armstrong and Irvine<sup>37</sup> studied creep in specimens of  $\text{UO}_2\text{-ZrO}_2$ ,  $\text{UO}_2\text{-Y}_2\text{O}_3$ ,  $\text{UO}_2\text{-CaO}$ ,  $\text{UO}_2\text{SiO}_2$ . Ceramographic examination indicated that specimens containing either  $\text{ZrO}_2$ ,  $\text{Y}_2\text{O}_3$ , or  $\text{CaO}$  were single-phase solid solutions, whereas  $\text{SiO}_2$  appeared in grain boundaries and within grains. Creep-in-bending tests were conducted at 1200 to 1450°C at stresses of 14 to 112 MPa. The steady-state creep rate at 1400°C (typical results) was similar for  $\text{UO}_2\text{-CaO}$  and  $\text{UO}_2\text{-Y}_2\text{O}_3$  specimens. In both cases the creep rate decreased with increasing  $\text{CaO}$  or  $\text{Y}_2\text{O}_3$  over the range 0.1 to 1.0 mol %. An even larger decrease in creep rate was caused by  $\text{ZrO}_2$ , which could be a stand-in for  $\text{HfO}_2$ . The opposite effect was caused by  $\text{SiO}_2$  additions, which resulted in a high creep rate relative to pure  $\text{UO}_2$ . Intergranular cracks formed in many specimens during creep, and the crack depth measured from the specimen surface was greater in specimens containing  $\text{CaO}$ ,  $\text{Y}_2\text{O}_3$ , and  $\text{SiO}_2$  than in  $\text{UO}_2$  control specimens.

Fission products that remain as metallic precipitates may enhance or retard creep rates depending on their physical state or location.<sup>38</sup>

#### Sinterability

In one study<sup>36</sup> separate additions of 0.1 to 1.0 mol %  $\text{ZrO}_2$ ,  $\text{Y}_2\text{O}_3$ ,  $\text{CaO}$ , or  $\text{SiO}_2$  were dry blended with  $\text{UO}_2$ , pressed into bars, then sintered at 1700 to 1750°C in  $\text{H}_2$ . The densities of these materials exceeded 98% theoretical density. Mixtures of  $\text{UO}_2$  and  $\text{Gd}_2\text{O}_3$  were coprecipitated with  $\text{NH}_4\text{OH}$  from nitric acid solutions.<sup>34</sup> The precipitates were calcined, then reduced in  $\text{H}_2$ . The powder was pelletized and sintered at 1600°C in  $\text{H}_2$  for 7.2 ks. Sintered densities were 94 to 96% of theoretical density (t.d.) for pellets containing up to 24 mol %  $\text{Gd}_2\text{O}_3$ .

Littlechild, however, observed that additions of  $\text{Gd}_2\text{O}_3$  to  $\text{UO}_2$  reduced sinterability.<sup>39</sup> The powders were blended in varying proportions, sieved through 60 mesh, mixed again, pressed into pellets, and sintered at 1620°C in  $\text{H}_2$ . The addition of  $\text{Gd}_2\text{O}_3$  necessitated long sintering times to attain high densities. The density decreased from 96 to 89% t.d. as the  $\text{Gd}_2\text{O}_3$  content increased from 0 to 30 mol % for a

sintering time of 5.4 ks at 1620°C. Over the same composition range the density exceeded 96% t.d. for a sintering time of 36 ks, and exceeded 97% t.d. for 270 ks. The grain size decreased from about 25  $\mu\text{m}$  to 5  $\mu\text{m}$  as the  $\text{Gd}_2\text{O}_3$  content increased from 0 to 30 mol %.

The effects of the metallic additions Cr, Fe, Mo, Ni, and Si on the sintering behavior of  $\text{UO}_2$ -20 wt %  $\text{PuO}_2$  were studied.<sup>40</sup> The metals were separately added to the  $\text{UO}_2$ - $\text{PuO}_2$  powder at levels of 250, 1000, 2000, and 5000 ppm by weight. Only Cr and Si caused a decrease in the densities of sintered pellets, and then only at the 2000 and 5000 ppm levels. The densities of pellets containing Fe, Mo, and Ni were essentially constant over the range 250 to 5000 ppm added metal.

#### Methods of Adding Spikants to Fuels

Solid solutions of oxides have been prepared both by mechanical blending of the components and by coprecipitation from nitrate solutions. Armstrong and Irvine<sup>36</sup> prepared  $\text{UO}_2$  containing 0.1 to 1.0 mol % of either  $\text{CaO}$ ,  $\text{Y}_2\text{O}_3$ , or  $\text{ZrO}_2$  by mechanical blending followed by pressing and sintering at 1750°C. Ceramographic examination showed that the materials were single phase. Mechanical blends of  $\text{UO}_2$  and  $\text{Gd}_2\text{O}_3$ , however, sometimes resulted in incomplete solid solution formation after sintering.<sup>35,39</sup> Wada,<sup>35</sup> for example, reports that x-ray diffraction revealed incomplete solution even after sintering at 2000°C for 7.2 ks (2 h). When the same material was prepared by coprecipitation from nitrate solution, solid solutions were obtained after sintering at 1600°C for 7.2 ks (2 h). Solutions containing 0.5 and 1.0 mol %  $\text{Y}_2\text{O}_3$  in  $\text{UO}_2$  consisted of a single phase following coprecipitation and sintering at 1400°C for 3.6 ks (1 h).<sup>36</sup>

The metals Cr, Fe, Mo, Ni, and Si were added to  $\text{UO}_2$ -20 wt %  $\text{PuO}_2$  by mechanical blending.<sup>40</sup> The degree of dispersion, however, was not discussed.

## SUMMARY AND CONCLUSIONS

Practical considerations concerning the potential effects of spikants on fuel performance and the technology associated with obtaining appropriate spikants and maintaining their presence with the fuel constituents throughout the fuel cycle were evaluated. A list of 64 radionuclides with favorable characteristics has been reduced to four promising possibilities. These are the fission products  $^{106}\text{Ru}$  and  $^{144}\text{Ce}$  and the deliberately manufactured radionuclides  $^{60}\text{Co}$  and  $^{172}\text{Hf}$ . However, the anticipated high cost of producing  $^{172}\text{Hf}$  essentially eliminates it as a candidate.

The evaluation of the use of  $^{60}\text{Co}$ ,  $^{160}\text{Ru}$ , and  $^{144}\text{Ce}$  can be summarized as follows.

1. Cerium in the fission products from a PWR fuel element (530 kg fuel) will provide a maximum of about 2000 R/h at 0.30 m (1 ft) after 2 years, while ruthenium will provide about 13,000 R/h.
2. Approximately 50% of the fission product ruthenium and 3–5% of the cerium could be coprocessed with the fuel.
3. Nonradioactive nuclides of cerium and ruthenium dilute the radioactive nuclides to 2–4% of the total element in the fission products 2 years after removal from the reactor.
4. Recycling cerium and ruthenium will dilute the radionuclides by a buildup of stable isotopes of each of these elements. This buildup will increase each time the fuel is irradiated. The worst case is for cerium, where, after six reprocessing cycles, total replacement of the fuel with  $\text{CeO}_2$  will not provide 27,000 R/h at 0.30 m (1 ft). For ruthenium, after six reprocessing cycles, the required concentration will rise to 10% total element to achieve the same dose rate.
5. Cobalt-60 can readily be manufactured in a thermal reactor and less readily in a fast reactor. Only 0.003% (30 ppm) by weight of the fuel is required to produce the dose rate of 27,000 R/h at 0.30 m.
6. Cobalt cannot be coprocessed with the fuel materials but will be lost in the early stages of reprocessing.
7. In order to provide maximum diversion deterrence, a duplex spikant reprocessing scheme may be necessary. In it about 50% of the

fission product ruthenium is coprocessed with the fuel, and  $^{60}\text{Co}$  is added to the fuel before the conversion step. During the conversion and sintering steps, the ruthenium may volatilize away while the cobalt should remain with the fuel to provide the necessary diversion resistance.

8. The use of nominal amounts of ruthenium or cobalt as spikants is not expected to adversely affect fuel performance.

Much of the information discussed in this report is predictive and will require experimental verification before final selection of a spikant or spikants can be made. Among the areas in which initial experimental work is being planned are: stability of ruthenium and cobalt during conversion and refabrication, verification of the amount of cerium and ruthenium that can be coprocessed with the fuel, separation of cobalt from the fission products, effect of spikants on sintering and on the mechanical properties of the fuel, and the effect of the various spikants on the oxygen potential of the fuel.

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APPENDIX  
COMPLETE LIST OF POTENTIAL FUEL SPIKANTS

Radionuclide	Half-Life	Cross Section Spikant-Barns (Decay Product)	Principal Gammas		Concentration for 27,000 R After 2 Years (ppm)	Possible Production Methods	Criterion for Rejection
			Energy (keV)	Inten- sity (%)			
<sup>22</sup> Na	2.60 y	<sup>22</sup> Na; $\sigma = 2.9$ ( <sup>22</sup> Ne; $\sigma = 0.048$ )	511 1275	180 100	7.8	<sup>23</sup> Na(100%) ( <i>n, 2n</i> )	Production method
<sup>42</sup> Ar <sup>42</sup> K	33 y 12.4 h	( <sup>42</sup> Ca; $\sigma = 0.65$ )	<sup>42</sup> K 1524	18	1018	<sup>40</sup> Ar( <i>n, <math>\gamma</math></i> ) <sup>41</sup> Ar( <i>n, <math>\gamma</math></i> ) (99.6%) $\sigma_1 = 0.61$ b $\sigma_2 = 0.5$ b	Gaseous element
<sup>46</sup> Sc	83.9 d	<sup>46</sup> Sc; $\sigma = 8$ ( <sup>46</sup> Ti; $\sigma = 0.6$ )	889 1120	100 100	374	<sup>45</sup> Sc(100%) ( <i>n, <math>\gamma</math></i> ); $\sigma = 23$ b	
<sup>44</sup> Ti <sup>44</sup> Sc	48 y 3.9 h	( <sup>44</sup> Ca; $\sigma = 1.0$ )	<sup>44</sup> Sc 511 1159	188 100	230	<sup>45</sup> Sc(100%) ( <i>p, 2n</i> )	Production method
<sup>54</sup> Mn	303 d	<sup>54</sup> Mn; $\sigma < 10$ ( <sup>54</sup> Cr; $\sigma = 0.36$ )	835	100	49	<sup>55</sup> Mn(100%) ( <i>n, 2n</i> ) <sup>54</sup> Fe(5.8%) ( <i>n, p</i> ) <sup>54</sup> Cr(2.4%) ( <i>p, n</i> )	Low <i>E</i> Production method
<sup>56</sup> Co	77 d	( <sup>56</sup> Fe; $\sigma = 2.63$ )	511 847 1040 1240 1760 2020 2600 3260	40 100 15 66 15 11 17 13	480	<sup>56</sup> Fe(92%) ( <i>p, n</i> )	Production method
<sup>58</sup> Co	71.3 d	<sup>58</sup> Co; $\sigma = 1880$ ( <sup>58</sup> Fe; $\sigma = 1.15$ )	511 810	30 99	2400	<sup>59</sup> Co(100%) ( <i>n, 2n</i> ) <sup>58</sup> Ni(68%) ( <i>n, p</i> ) <sup>55</sup> Mn(100%) ( $\alpha, n$ )	Short half-life; low <i>E</i> ; production method
<sup>59</sup> Fe	45 d	( <sup>59</sup> Co; $\sigma = 37.2$ )	1095 1292	56 44	86,000	<sup>58</sup> Fe(0.31%) ( <i>n, <math>\gamma</math></i> ); $\sigma = 1.1$ b <sup>59</sup> Co(100%) ( <i>n, <math>\gamma</math></i> )	Short half-life Production method
<sup>60</sup> Co	5.26 y	<sup>60</sup> Co; $\sigma = 2.0$ ( <sup>60</sup> Ni; $\sigma = 2.8$ )	1173 1332	100 100	30	<sup>59</sup> Co(100%) ( <i>n, <math>\gamma</math></i> ); $\sigma = 37$ b	
<sup>65</sup> Zn	245 d	( <sup>65</sup> Cu; $\sigma = 2.17$ )	511 1115	3.4 49	108	<sup>64</sup> Zn(48.9%) ( <i>n, <math>\gamma</math></i> ); $\sigma = 0.46$ b	
<sup>68</sup> Ge <sup>68</sup> Ga	275 d 68 m	( <sup>68</sup> Zn; $\sigma = 0.072$ )	<sup>68</sup> Ga 511 1078	176 3.5	57	<sup>66</sup> Zn(28%) ( $\alpha, 2n$ )	Low <i>I</i> ; production method
<sup>83</sup> Rb	83 d	( <sup>83</sup> Kr; $\sigma = 200$ )	530 790	93 0.9	2700	<sup>84</sup> Sr(0.56%) ( <i>n, 2n</i> ) <sup>83</sup> Sr(Ec) <sup>81</sup> Br(49%) ( $\alpha, 2n$ ) <sup>85</sup> Rb(72%) ( <i>p, 3n</i> ) <sup>83</sup> Sr(Ec)	Low <i>E</i> Short half-life; production method

APPENDIX (continued)

Radionuclide	Half-Life	Cross Section Spikant-Barns (Decay Product)	Principal Gammas		Concentration for 27,000 R After 2 Years (ppm)	Possible Production Methods	Criterion for Rejection
			Energy (keV)	Inten- sity (%)			
<sup>84</sup> Rb	33d	( <sup>84</sup> Kr; σ = 0.13)	511 800 1010 1900	42 74 0.5 0.8	6.3 × 10 <sup>6</sup>	<sup>85</sup> Rb(72%) (n,2n) <sup>84</sup> Sr(0.56%) (n,p)	Short half-life; low I; production method
<sup>82</sup> Sr ↓ <sup>82</sup> Rb	25 d 1.3 m	( <sup>82</sup> Kr; σ = 45)	<sup>82</sup> Rb 511 777	192 9	5.5 × 10 <sup>8</sup>	<sup>85</sup> Rb(72%) (p,4n)	Short half-life; low E; production method
<sup>85</sup> Sr	64 d	( <sup>85</sup> Rb; σ = 0.46)	514	100	13,100	<sup>84</sup> Sr(0.56%) (n,γ); σ = 1.4 b <sup>86</sup> Sr(9.9%) (n,2n) <sup>85</sup> Rb(72%) (p,n)	Low E
<sup>85</sup> Kr	10.76 y	<sup>85</sup> Kr; σ = 1.66 ( <sup>85</sup> Rb; σ = 0.46)	514	41	825	<sup>84</sup> Kr(56.9%) (n,γ); σ = 0.06 b Fission product	Gaseous; low E Low σ
<sup>88</sup> Y	108 d	( <sup>88</sup> Sr; σ = 0.0058)	898 1836	91 100	204	<sup>89</sup> Y(100%) (n,2n) <sup>88</sup> Sr(83%) (p,n)	Production method
<sup>88</sup> Zr ↓ <sup>88</sup> Y	85 d 108 d	( <sup>88</sup> Sr; σ = 0.0058)	<sup>88</sup> Zr 394 <sup>88</sup> Y 898 1836	97 91 100	175	<sup>89</sup> Y(100%) (p,2n)	Production method
<sup>91m</sup> Nb	62 d	( <sup>91</sup> Zr; σ = 1.6)	1210	3	270,000	<sup>89</sup> Y(100%) (α,2n) <sup>91</sup> Zr(11%) (p,n)	Low I; short half-life; production method
<sup>95</sup> Nb	35 d	<sup>95</sup> Nb, σ < 7 ( <sup>95</sup> Mo; σ = 14.5)	765	100	3.7 × 10 <sup>6</sup>	Fission product <sup>95</sup> Mo(15.7%) (n,p) <sup>96</sup> Zr(2.8%) (p,2n)	Short half-life; low E
<sup>95</sup> Zr ↓ <sup>95</sup> Nb	65 d 35 d	( <sup>95</sup> Mo; σ = 14.5)	Zr 756 Nb 765	49 100	5,900	<sup>94</sup> Zr(17.4%) (n,γ); σ = 0.03 b fission product <sup>96</sup> Zr(2.8%) (n,2n)	Low E; short half- life; low σ
<sup>95m</sup> Tc ↓ <sup>95</sup> Tc	61 d 20 h	( <sup>95</sup> Mo; σ = 14.5)	<sup>95m</sup> Tc 584 780 823 838 1042 <sup>95</sup> Tc 768	36 12 9 27 4 ≈3	16,300	<sup>95</sup> Mo(16%) (p,n)	Short half-life; low I; production method
<sup>103</sup> Ru	39.6 d	( <sup>103</sup> Rh; σ = 150)	497 610	88 6	1.35 × 10 <sup>6</sup>	<sup>102</sup> Ru(31.6%) (n,γ); σ = 1.4 b fission product <sup>104</sup> Ru(18.9%) (n,2n) <sup>103</sup> Rh(100%) (n,p)	Low E; short half-life

APPENDIX (continued)

Radionuclide	Half-Life	Cross Section Spikant-Barns (Decay Product)	Principal Gammas		Concentration for 27,000 R After 2 Years (ppm)	Possible Production Methods	Criterion for Rejection	
			Energy (keV)	Inten- sity (%)				
$^{102}\text{Rh}$	206 d	$(^{102}\text{Ru}; \sigma = 1.3)$	475	57	220	$^{103}\text{Rh}(100\%) (n, 2p)$ $^{102}\text{Pd}(0.95\%) (n, p)$ $^{102}\text{Ru}(32\%) (p, n)$	Low <i>I</i> ; production method	
			511	25				
			628	4				
			1103	3				
			1307	0.5				
			1570	0.2				
$^{105}\text{Ag}$	40 d	— $(^{105}\text{Pd}; \sigma = 14)$	443	10	$6.0 \times 10^6$	$^{103}\text{Rh}(100\%) (\alpha, 2n)$ $^{105}\text{Pd}(22\%) (p, n)$	Short half-life; low <i>I</i> ; production method	
			620–680	12				
			1088	2				
$^{106}\text{Ru} \downarrow$ $^{106}\text{Rh}$	367 d 30 s	$^{106}\text{Ru}; \sigma = 0.146$ $(^{106}\text{Pd}; \sigma = 0.013)$	$^{106}\text{Rh}$	512	21	343	Fission product	Low <i>I</i>
				622	11			
				1050	1.5			
				1130	0.5			
				1550	0.2			
$^{102\text{m}}\text{Rh}$	2.1 y	$(^{102}\text{Ru}; \sigma = 1.3)$	418	13	34	$^{102}\text{Ru}(32\%) (p, n)$	Production method	
			475	95				
			632	54				
			698	41				
			768	30				
			1050	41				
			1110	22				
			$^{108\text{m}}\text{Ag}$	≈100 y				$(^{108}\text{Cd}; \sigma = 1.1)$
614	90							
722	90							
$^{110\text{m}}\text{Ag}$	253 d	$^{110}\text{Ag}; \sigma = 82$ $(^{110}\text{Cd}; \sigma = 11)$	$^{110}\text{Ag}$	658	96	37	$^{109}\text{Ag}(48.7\%) (n, \gamma); \sigma = 3 \text{ b}$	
				680	16			
				706	19			
				764	23			
				818	8			
				885	71			
				937	32			
				1384	21			
				1505	11			
				$^{114\text{m}}\text{In}$	50.0 d			
724	3.5							
1299	0.16							

APPENDIX (continued)

Radionuclide	Half-Life	Cross Section Spikant-Barns (Decay Product)	Principal Gammas		Concentration for 27,000 R After 2 Years (ppm)	Possible Production Methods	Criterion for Rejection
			Energy (keV)	Inten- sity (%)			
<sup>115m</sup> Cd	43 d	(115In; σ = 202)	485	0.31	1.0 × 10 <sup>7</sup>	114Cd(28.9%) (n, γ); σ = 0.14 b 116Cd(7.6%) (n, 2n) 115In(95.8%) (n, p)	Short half-life Low I; low σ
			935	1.9			
			1290	0.9			
<sup>121m</sup> Te + <sup>121</sup> Te	154 d 17 d	(121Sb; σ = 0.055)	1100	3	445	120Te(0.089%) (n, γ); σ = 0.14 b 122Te(2.5%) (n, 2n) 121Sb(57%) (p, n)	Low I Production method
			508	16			
			573	72			
<sup>124</sup> Sb	60 d	124Sb; σ = 6.5 (124Te; σ = 6.8)	603	97	8800	Sb(42.8%) (n, γ); σ = 3.3 b	Short half-life
			644	7			
			720	14			
			967	2.4			
			1048	2.4			
			1310	3			
			1370	5			
			1450	2			
			1692	50			
			2088	7			
<sup>125</sup> Sb	2.7 y	(125Te; σ = 1.55)	427	31	220	124Sn(6.0%) (n, γ) 125mSn 125mSn(β <sup>-</sup> ) (9.1 m); σ = 0.1 b 125Te(7.0%) (n, p) Fission product (low yield)	Low E; low σ
			463	10			
			599	24			
			634	11			
			660	3			
<sup>134</sup> Cs	2.05 y	134Cs; σ = 140 (134Ba; σ = 0.158)	570	23	57	133Cs(100%) (n, γ); σ = 30 b	Low I
			605	98			
			796	99			
			1038	1.0			
			1168	1.9			
			1365	3.4			
<sup>137</sup> Cs	30.0 y	137Cs; σ = 0.11 (137Ba; σ = 5.1)	662	85	1250	Fission product	Low E
<sup>143</sup> Pm	0.73 y	(143Nd; σ = 325)	742	47	340	141Pr(100%) (α, 2n) 144Nd(24%) (p, 2n)	Low E; production method
<sup>144</sup> Pm	1.1 y	(144Nd; σ = 3.6)	474	45	61	144Sm(3.2%) (n, p) 141Pr(100%) (α, n) 144Nd(24%) (p, n)	Low E; production method
			615	99			
			695	99			
<sup>144</sup> Ce + <sup>144</sup> Pr	284 d 17 m	144Ce; σ = 1.0 (144Nd; σ = 3.6)	695	1.5	4220	Fission product	Low I
			1487	0.29			
			2186	0.7			

APPENDIX (continued)

Radionuclide	Half-Life	Cross Section Spikant-Barns (Decay Product)	Principal Gammas		Concentration for 27,000 R After 2 Years (ppm)	Possible Production Methods	Criterion for Rejection		
			Energy (keV)	Inten- sity (%)					
$^{146}\text{Pm}$	5.5 y	$^{146}\text{Pm}$ ; $\sigma = 8400$ ( $^{146}\text{Nd}$ ; $\sigma = 1.4$ )	453 750	65 65	219	$^{146}\text{Nd}(17\%) (p, n)$	Low $E$ ; production method		
$^{148m}\text{Pm}$	42 d	$^{148}\text{Pm}$ ; $\sigma = 22,000$ ( $^{148}\text{Sm}$ ; $\sigma = 2.7$ )	$^{148m}\text{Pm}$ 413 551	17 95	$2.7 \times 10^5$	$\left\{ \begin{array}{l} ^{146}\text{Nd}(17\%) (n, \gamma) \\ ^{147}\text{Nd}; \sigma = 2 \text{ b} \\ ^{147}\text{Nd}(11 \text{ d})(\beta^-) \quad ^{147}\text{Pm} \\ ^{147}\text{Pm}(2.6 \text{ y})(n, \gamma) \quad ^{148m}\text{Pm} \\ ^{148}\text{Sm}(11.3\%) (n, p) \\ ^{151}\text{Eu}(47.8\%) (n, \alpha) \\ ^{148}\text{Nd}(5.7\%) (p, n) \end{array} \right.$	Short half-life; low $I$		
$^{148}\text{Pm}$	5.4 d		630 727 916 1015 551 914 1465	87 36 21 20 1.9 1.1 1.6					
$^{148}\text{Eu}$	54 d	( $^{148}\text{Sm}$ ; $\sigma = 2.7$ )	413 551 620 720 872 917 967 1033 1166 1345 1620	18 120 90 18 7 5 5 7 5 8 11	22,000			$^{148}\text{Sm}(11\%) (p, n)$	Short half-life; production method
$^{150}\text{Eu}$	6.2 y	( $^{150}\text{Sm}$ ; $\sigma = 102$ )	439 584 740 1049 1248 1347	86 60 21 9 5 4	176			$^{151}\text{Eu}(48\%) (n, 2n)$ $^{150}\text{Sm}(7.5\%) (p, n)$	Low $I$ ; production method
$^{146}\text{Gd} \downarrow$ $^{146}\text{Eu}$	50 d 4.6 d	( $^{146}\text{Sm}$ ; $\sigma = ?$ )	$^{146}\text{Eu}$ 511 634 666 710 749 900 1058 1160 1298 1408 1535	7 77 12 13 100 8 7 6 6 5 8	44,500			$^{144}\text{Sm}(3.2\%) (\alpha, 2n)$	Short half-life; production method

APPENDIX (continued)

Radionuclide	Half-Life	Cross Section Spikant-Barns (Decay Product)	Principal Gammas		Concentration for 27,000 R After 2 Years (ppm)	Possible Production Methods	Criterion for Rejection
			Energy (keV)	Inten- sity (%)			
<sup>152</sup> Eu	14 y	<sup>152</sup> Eu; σ = 2300 ( <sup>152</sup> Sm; σ = 206)	779	14	497	<sup>151</sup> Eu(48%) (n, γ); σ = 5300 b	High cross section
			965	15			
			1087	12			
			1113	14			
			1408	22			
<sup>154</sup> Eu	7.8 y	<sup>154</sup> Eu; σ = 1500 ( <sup>152</sup> Gd; σ = 85)	593	6	226	<sup>153</sup> Eu(52.2%) (n, γ); σ = 480 b	High cross section
			724	21			
			759	5			
			876	12			
			1000	31			
			1278	37			
<sup>158</sup> Tb	1200 y	<sup>158</sup> Gd; σ = 2.5)	782	10	42,200	<sup>159</sup> Tb(100%) (n, 2n) ( <sup>156</sup> Dy(0.052%) (n, γ) <sup>157</sup> Dy { <sup>157</sup> Dy[EC] <sup>157</sup> Tb <sup>157</sup> Tb(150 y) (n, γ) <sup>158</sup> Gd(25%) (p, n)	Long half-life; low I; production method
			950	69			
			1110	2.2			
			1190	1.8			
<sup>160</sup> Tb	72.1 d	<sup>160</sup> Tb; σ = 525 ( <sup>160</sup> Dy; σ = 61)	879	31	7,440	<sup>159</sup> Tb(100%) (n, γ); σ = 46 b	Low I; short half- life
			966	31			
			1178	15			
			1272	7			
<sup>166m</sup> Ho	1200 y	<sup>166</sup> Er; σ = 15)	529.8	10	32,000	<sup>165</sup> Ho(100%) (n, γ); σ ≈ 1b	Long half-life; low I
			711.7	59			
			810.3	63			
			950.9	3.0			
			1241.4	1.0			
			1400.7 + 1427.1	1.1			
<sup>168</sup> Tm	93 d	<sup>168</sup> Er; σ = 1.95)	448	27	1,330	<sup>169</sup> Tm(100%) (n, 2n) <sup>168</sup> Er(27%) (p, n)	Low I; production method
			630	14			
			730	40			
			820	88			
			917	4			
			1280	3			
<sup>173</sup> Lu	1.37 y	<sup>173</sup> Tb; σ = 19)	637	1.5	11,300	<sup>173</sup> Yb(16%) (p, n)	Low E; production method
<sup>174m</sup> Lu↓ <sup>174</sup> Lu	140 d	<sup>174</sup> Yb; σ = 65)	1240	9	3,940	<sup>175</sup> Lu(97%) (n, 2n) <sup>174</sup> Yb(32%) (p, n)	Low I; production method
	3.6 y						

APPENDIX (continued)

Radionuclide	Half-Life	Cross Section Spikant-Barns (Decay Product)	Principal Gammas		Concentration for 27,000 R After 2 Years (ppm)	Possible Production Methods	Criterion for Rejection
			Energy (keV)	Inten- sity (%)			
<sup>172</sup> Hf ↓ <sup>172</sup> Lu	5 y 6.7 d	( <sup>172</sup> Yb; σ = 1.3)	<sup>172</sup> Lu 810 900 1090	21 45 60	167	<sup>175</sup> Lu(97%) (p,4n)	Production method
<sup>182</sup> Ta	115.1 d	<sup>182</sup> Ta; σ = 8200 ( <sup>182</sup> W; σ = 20.7)	1122 1189 1222 1231	34 16 27 13	800	<sup>181</sup> Ta(100%) (n,γ); σ = 21 b	High cross section
<sup>184m</sup> Re ↓ <sup>184</sup> Re	169 d 38 d	( <sup>184</sup> W; σ = 1.8)	<sup>184</sup> Re 793 896 904	≈27 ≈12 ≈28	500	<sup>185</sup> Re(37.1) (n,2n) <sup>184</sup> W(31%) (p,n)	Low E; production method
<sup>188</sup> W ↓ <sup>188</sup> Re	69.4 d 17 h	<2 ( <sup>188</sup> Os; σ = 4.3)	<sup>188</sup> Re 633 829 932	0.9 0.3 0.4	7.4 × 10 <sup>5</sup>	<sup>186</sup> W(28.4%) (n,γ) <sup>187</sup> W; σ = 40 b <sup>187</sup> W(24 h) (n,γ); σ ≈ 90 b	Low E; short half-life
<sup>185</sup> Os	93.6 d	( <sup>185</sup> Re; σ = 112)	646 875	80 14	2,800	<sup>184</sup> Os(0.018%) (n,γ); σ < 200 b <sup>186</sup> Os(1.6%) (n,2n) <sup>185</sup> Re(37%) (p,n)	Low E; production method
<sup>194</sup> Hg ↓ <sup>194</sup> Au	~1.3 y 39.5 h	( <sup>194</sup> Au; σ = 2)	<sup>194</sup> Au 1469 1596 1887 2044	3 3 4 4	445	<sup>197</sup> Au(100%) (p,4n)	Production method; low I
<sup>192m</sup> Ir ↓ <sup>192</sup> Ir	≈650 y 74 d	<sup>192</sup> Ir; σ = 1100 ( <sup>192</sup> Pt; σ < 14)	<sup>192</sup> Ir 468 589 604 612	49 4 9 6	55,000	<sup>191</sup> Ir(38.5%) (n,γ); σ = 0.4 b <sup>193</sup> Ir(61.5%) (n,2n)	Low E; long half-life
<sup>192</sup> Ir	74.2 d	<sup>192</sup> Ir; σ = 1100 ( <sup>192</sup> Pt; σ < 14)	468 589 604 612	49 4 9 5	15,700	<sup>191</sup> Ir(38.5%) (n,γ); σ = 100 b	Low E; short half-life
<sup>194</sup> Os ↓ <sup>194</sup> Ir	6.0 y 17 h	( <sup>194</sup> Pt; σ = 1.2)	<sup>192</sup> Ir 640 939 1160 1480 1700	1.0 0.4 0.8 0.6 0.2	8,400	<sup>192</sup> Os(41%) (n,γ) <sup>193</sup> Os; σ = 1 b <sup>193</sup> Os(31 h) (n,γ); σ = 200 b	Low I
<sup>207</sup> Bi	30 y	( <sup>207</sup> Pb; σ = 0.7)	570 1063 1771	98 77 9	749	<sup>209</sup> Bi(100%) (α,2n) <sup>211</sup> At <sup>211</sup> At(7.2 h) (41% σ) <sup>207</sup> Pb(22%) (p,n)	Production method

APPENDIX (continued)

Radionuclide	Half-Life	Cross Section Spikant-Barns (Decay Product)	Principal Gammas		Concentration for 27,000 R After 2 Years (ppm)	Possible Production Methods	Criterion for Rejection
			Energy (keV)	Inten- sity (%)			
$^{209}\text{Po}$	103 y	( $^{209}\text{Bi}$ ; $\sigma = 0.034$ )	910	0.5	$8.2 \times 10^5$	$^{209}\text{Bi}(100\%)(p, n)$	Low E; low I; production method
$^{232}\text{U}$	74 y	$^{232}\text{U}$ ; $\sigma = 73.1$  ( $^{228}\text{Th}$ ; $\sigma = 123$ )	$^{212}\text{Bi}$ 727	7	2800	$\left\{ \begin{array}{l} ^{230}\text{Th}(n, \gamma) ^{231}\text{Th}(25 \text{ h}) \\ (\beta^-) ^{231}\text{Pa} \\ ^{231}\text{Pa}(32,500 \text{ y})(n, \gamma) ^{232}\text{Pa} \\ ^{232}\text{Pa}(1.3 \text{ d})(\beta^-) ^{232}\text{U}; \\ \sigma_1 = 23 \text{ b}; \sigma_2 = 200 \text{ b} \\ ^{232}\text{Th}(100\%)(n, 2n) ^{231}\text{Th} \\ ^{231}\text{Th}(25 \text{ h})(\beta^-) ^{231}\text{Pa} \\ \sigma = 1.5 \text{ b}; E = 8-13 \text{ MeV} \\ ^{231}\text{Pa}(n, \gamma)(\beta^-) ^{232}\text{U}; \\ \sigma = 200 \text{ b} \\ ^{235}\text{U}(\gamma, 3n) \end{array} \right.$	Long half-life
			785	1.1			
			1620	1.8			
			$^{208}\text{Tl}$ 511	8			
			583	31			
			860	4			
2614	36						



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