SAVANNAH RIVER LABORATORY
TECHNICAL DIVISION

DPST-AFCT-77-154

DISTRIBUTION

J. D. SPENCER, SRL
R. E. NAYLOR, WILM.
L. H. MEYER
C. H. ICE, SRL
J. M. BOSWELL
H. J. GROH
D. L. McINTOSH
J. W. WADE
G. F. MERZ
P. L. ROGGENKAMP
W. E. GRAVES
M. L. HYDER
F. R. FIELD
L. A. HEINRICH
H. D. HARMON
J. B. MELLEN
J. T. RATLIFF
F. D. R. KING
J. P. CHURCH
R. L. REED
TIS FILE

MEMORANDUM

DECEMBER 7, 1977

TO: J. D. SPENCER

FROM: T. C. GORRELL TCG

CHOICE OF SPIKING MATERIAL FOR RECYCLED LWR FUEL

INTRODUCTION AND SUMMARY

The feasibility of using certain highly radioactive nuclides to spike recycle LWR fuel was investigated. The purpose of spiking is to minimize the likelihood of diversion of the fuel. The amounts of spikeant required and the reactivity effects on subsequent reactor operation were evaluated for a single fuel rod of MOX fuel.

For spent fuel cooling times of six months or greater, only three groups of fission products are attractive, viz., Zr, Ru and Cs. Table 3 lists dose rates calculated for a single fuel rod containing the fission products recycled from a similar spent fuel rod, at three cooling times. Radiation dose rates of several hundred R/hr at 3 feet can be achieved.

The reactivity effects of individual fission products are small, and can be compensated for by very small increases in the Pu content of MOX fuel. Use of the gross fission product inventory as a spikeant would result in significant reactivity losses if the rare earth fraction were not extracted.

^{60}Co is also an attractive spikeant candidate. Specific loadings of only a few mg/ft would be required. The ^{59}Co could be irradiated in separate rods in a LWR fuel bundle, or could be obtained elsewhere, e.g.,SRP.

DISCUSSION

The spiking of nuclear materials is generally defined as the addition of radioactive nuclides to recycled fuel material for protective purposes. The intent of spiking is to enhance detection and to make difficult the theft of significant quantities of fissile material. There are many considerations in implementing the spiking concept such as choice of spikeant, shielding required for workers, additional capital costs, and penalties to reactor operation. Only two parameters were investigated in the study reported here:

1. The amounts of several fission products and cobalt-60 required to achieve a given radiation dose.

2. The reactivity effects of these nuclides.

The fuel material spiked was a mixed Pu/U oxide containing 4.5% total Pu. The assay is given in Table 1. The effects of a given spikeant are insensitive to the fuel assay.

The radiation dosage required was arbitrarily set at 100 R/hr at 3 feet from a single fuel rod having an OD of 0.325 inch. This radiation level is considered to be sufficiently large to deter casual theft of the rod, and can be readily achieved by the addition of available quantities of several spikeants.

Required Nuclide Concentrations

Several candidate spikeants were selected for study. The nuclides chosen together make up greater than 90% of the gamma activity in the spent fuel fission product inventory at cooling times in excess of six months. Dose rate calculations were made by R. L. Reed of RPD using ANISN, a one-dimensional S_n transport theory code. The SRL 22-group cross section set was used. Dose conversion factors were obtained from Reference 1. Properties of the spikeants are given in Table 2. The final column in Table 2 lists the concentration of each nuclide required to provide the 100 R/hr dose from one fuel rod, independent of the other nuclides. In those cases where more than one nuclide is listed at a given mass number, the total radiation from the set is equal to 100 R/hr. The nuclide contents in each mass number set are in secular equilibrium, which fixes their ratios at constant values independent of cooling time.

The fission product contents given in Reference 2 (from an ORIGEN calculation) were used to obtain available spikeant concentrations as a function of spent fuel cooling time. The MOX fuel exposure was 33,000 MWD/MT. Dose values are shown in Table 3, at cooling times of 0.5, 1.0 and 1.5 years. It was assumed that spikeant material from only one spent fuel rod would be available for use in a new rod.

Few of the candidate spikeants have practical application for fuel cooling times in excess of six months; the short half life nuclides have decayed to very low concentrations. The ^{95}Zr set could be used if recovered from spent fuel before one year. The ^{106}Ru set or the total Cs set could be used with up to two years'cooling. If the total fission product inventory were used as a spikeant, the fraction of the total required for 100 R/hr is 8%,13%, and 18% at 0.5, 1.0 and 1.5 year cooling times, respectively.

^{60}Co was also considered as a spikeant material. The ^{60}Co concentration in the fuel rod required is 2.6×10^{-6} atoms/barn-cm for a dose rate of 100 R/hr at 3 ft. The specific loading is 4.2 mg/ft (or 5 curies/ft) in the 0.325 inch rod. A significant amount of ^{59}Co will also be present. A $^{59}Co/^{60}Co$ ratio of 16 was calculated for ^{59}Co irradiated in an LWR fuel assembly for three years, then decayed 1 year. The reactivity effects of ^{59}Co are discussed in the next section.

Reactivity Effects of Spikeants

As expected, the addition of spikeants resulted in only small decreases in lattice reactivity. Calculations were made using the GLASS code to determine the increase in Pu content of the fuel necessary to compensate for the reactivity losses. For small reactivity changes, the zero-exposure k_∞ value was increased 0.1% Δk by a 1.2% increase in total Pu content (the Pu fraction in Table 1 was increased from 4.546% in the base case to 4.600% to increase k_∞ 0.1%). For the same fuel exposure, this extra Pu would not be burned, but rather would be carried along throughout the process as a slightly higher Pu inventory.

Multigroup reactivity calculations were made using the GLASS code to determine the effect of adding ^{141}Ce and ^{144}Ce to the fuel rod in the concentrations necessary for the 100 R/hr dose. The Δk_∞ calculated was -0.005% for each, which would be compensated for by an increase in total Pu content of only 0.06%. The nuclide with thelargest cross section -^{134}Cs, is not represented in the multigroup library so was not evaluated specifically, but would be expected to have a poisoning effect about four times that of the Ce isotopes. Again, the necessary increase in Pu content is small - about 0.24%.

If ^{60}Co is used as the spikeant, and the $^{59}Co/^{60}Co$ ratio is 16 as discussed earlier, the Δk_∞ is -0.19%. The total Pu content of the fuel must be increased by 2.3%.

A much larger reactivity loss would be experienced if fractions of the entire fission product inventory were used as a spikeant. The poisoning effect would exceed 1% in k_∞ if the wastes were a year old or more. The losses could be reduced significantly by removing the rare earth fission products, and utilizing primarily the Cs and Ru fractions

References

1. DPST-70-233,*"ANISN-SRL"*, H. K. Clark and H. E. Hootman,January 22,1970.

2. DPST-AFCT-77-152, *"^{14}C Production in LWR Fuel"*, J.P. Church,Nov.9,1977.

Table 1

Content of Mixed Oxide Fuel Material

Nuclide	Content, g/MT	
^{234}U	267	
^{235}U	6,583	
^{238}U	947,692	
^{238}Pu	1,008	4.546%
^{239}Pu	22,403	
^{240}Pu	11,603	
^{241}Pu	6,652	
^{242}Pu	3,792	
Total	10^6 g	

Table 2

Properties and Required Contents of Spikeant Candidates

Nuclide	$T_{1/2}$	Capture σ2200, cm^{-2}	Content for 100 R/hr @ 3 ft. Atom Number Density*	Grams/ft
^{91}Y	58.6 d	1.4	7.2×10^{-5}	1.8×10^{-1}
^{95}Zr	65.5 d	0.5	2.1×10^{-7}	5.4×10^{-4}
^{95m}Nb	3.6 d	-	2.5×10^{-10}	6.4×10^{-7}
^{95}Nb	35.1 d	1.5	2.2×10^{-7}	5.6×10^{-4}
^{103}Ru	39.6 d	7.7	2.6×10^{-7}	7.2×10^{-4}
^{103m}Rh	56 m	-	2.6×10^{-10}	7.2×10^{-7}
^{106}Ru	1.01 y	0.15	1.5×10^{-6}	4.3×10^{-3}
^{106}Rh	30 s	-	1.5×10^{-12}	4.3×10^{-9}
^{125}Sb	2.73 y	1.0	3.4×10^{-5}	1.15×10^{-1}
^{125m}Te	58 d	-	9.0×10^{-7}	3.0×10^{-3}
^{134}Cs**	2.06 y	140.	2.0×10^{-6}	7.2×10^{-3}
^{137}Cs	30.1 y	0.1	8.0×10^{-5}	3.0×10^{-1}
^{137m}Ba	2.55 m	-	1.2×10^{-11}	4.4×10^{-8}
^{141}Ce	32.5 d	29.	2.5×10^{-6}	9.5×10^{-3}
^{144}Ce	284 d	1.0	4.2×10^{-5}	1.6×10^{-1}
^{144}Pr	17.3 m	-	1.8×10^{-9}	7.0×10^{-6}
^{60}Co	5.27 y	-	2.6×10^{-6}	4.2×10^{-3}

*Values given are atomic number densities in units of atoms/barn-cm. 1 atom/barn-cm = 10^{24} atoms/cm^3. Content of ^{238}U = 2.15×10^{-2} atoms/b-cm in the MOX fuel.

**^{134}Cs is not a direct-yield fission product; it is formed by neutron capture in ^{133}Cs. Its content must be calculated by codes such as CINDER or ORIGEN.

Table 3

Spikeant Dose Rates - Fission Products from One Fuel Rod

Nuclide	Dose Rate, R/hr @ 3 ft*		
	0.5 year cool	1.0 year cool	1.5 year cool
^{91}Y	0	0	0
^{95}Zr	230	35	5
^{95m}Nb			
^{95}Nb			
^{103}Ru	35	0	0
^{103m}Rh			
^{106}Ru	630	450	315
^{106}Rh			
^{125}Sb	0	0	0
^{125m}Te			
^{134}Cs	230	195	165
^{137}Cs	65	65	65
^{137m}Ba			
^{141}Ce	0	0	0
^{144}Ce	15	10	5
^{144}Pr			
Total	1205	755	555
Fraction Required for 100 R/hr	8%	13%	18%

*Fission products from one spent fuel rod were placed in one new fuel rod, at the three cooling times.