Chapter 12
Behavior of Solid Fission Products in Oxide Fuel Elements

Why are fission products so important?

1.) They influence the availability of oxygen

2.) Their volume is different

3.) Fission gases like Xe and Kr escape increasing pressure in fuel

4.) Solid FP can change the thermal conductivity and melting point

5.) Gaseous FP can change the thermal conductivity of gas in gap.

Since target burnups are ≈ 10%, we can have as much as 20% in FP in the fuel

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12.1 Elemental Yield of Fission Products

From nuclear physics;

$$(cy)_{ZM}^{A} = \sum_{Z'}^{Z} (iy)_{Z'M}^{A}$$

Cumulative Yield **Independent Yield**

$$ {}_{52}Te^{135} \xrightarrow[(<30s)]{} {}_{53}I^{135} \xrightarrow[6.7hr]{} {}_{54}Xe^{135} \xrightarrow[9.2hr]{} {}_{55}Cs^{135} \xrightarrow[2,600,000y]{} {}_{56}Ba^{135} $$

I	Xe	Cs	Ba
6.1	6.3	6.41	Stable 6.41

Chain Yield $Y_A = (cy)_{{}_Z M^A}$

Three Things effect the final distribution;

1.) **Fissile isotope (note Pu shift)**
2.) **Neutron energy**
3.) **Neutron flux (burnout)**

See figures, 12.2 and 3.16

Fission Product yield

Elemental Yield $$ Y_i = \left[\frac{1}{\dot{F}\, t_{irr}} \right] \sum_A N_{i,A} $$

Fission Rate (for $\dot{F}$)

Concentration of element i resulting from the decay chain at mass # A (for $N_{i,A}$)

Note also;

$$ \sum Y_i = 2 $$

One is not usually interested in the isotopes, but rather the elements. See Olander for a calculation of

$$Y_{Cs} = y_{133} + y_{135} + y_{137} \quad \text{(figure 12.1)}$$

$$N_{Cs^{133}} = y_{133} \dot{F} t_{irr} \left(1 - \frac{\{1 - \exp[-\lambda_{Xe} t_{irr}]\}}{\lambda_{Xe} t_{irr}} \right)$$

Since $t_{1/2}$ = 5.3 d for Xe_{133}

$N_{Cs^{133}} \approx Y_{133} \dot{F} t_{irr}$ for times > 5 days

See table 12.1

Importance of Various FP's

Zr + Nb • Zr can combine with O_2 -->ZrO_2

• Zr, Nb can take C away from UC

Y+ rare earths • High crossections, oxides

Ba + Sr • Sr radioactivity, zirconates

Mo.Ru,Tc • Volatile oxide, oxygen buffer

Rh,Pd • Reprocessing

Cs +Rb • Volatile, corrosive

I + Te • I is volatile, corrosive

Xe +Kr • Gases, pressure on cladding, thermal conductivity of gap

Fast ^{235}U	Fast (Mixed Oxide)	
1.) Y + Rare Earths	Y + Rare Earths	(0.493)
2.) Zr + Nb	Noble metals	(0.456)
3.) Noble Metals	Xe + Kr	(0.25)
4.) Xe + Kr	Zr + Nb	(0.219)
5.) Mo	Cs + Rb	(0.209)
6.) Cs + Rb	Mo	(0.206)
7.) Ba + Sr	Ba + Sr	(0.109)
8.) I + Te	I + Te	(0.07)

12.3
Chemical State of Fission Products in Oxide Fuels

Big question is, Do the FP's form Oxides?

$$\frac{2}{n}(FP) + O_2(g) = \frac{2}{n}(FP)O_n$$

Valence of FP Cation = 2n

Key is the oxygen partial pressure

The oxygen partial pressure at which the element and oxide coexist comes from the Law of Mass Action

$$\Pi_{O_2} = \exp\left(\frac{\Delta G^{\circ}_{FP}}{RT}\right)$$

where ΔG°_{FP} = Free energy of formation of FP oxide at T, per mole of oxygen

If ΔG is positive or only slightly negative, it will take a high Π_{O_2} from the fuel to cause an oxide to form.

If ΔG is strongly negative, then even small oxygen partial pressure from the fuel will form oxide

The oxides from $UO_{2\pm x}$ have oxygen potentials ranging from;

-170 kJ/mole (Hyper)

to - 670 kJ/mole (Hypo) **[Figure 11.12]**

Figure 12.6
Figure 12.7

If the free energy of formation of the oxide is more negative than the fuel, then the oxide will form

If the free energy of formation of the oxide is less negative than the fuel, then the elemental form will remain.

Note the case for Mo:

The ΔG_{FP} is close to stoichiometric fuel

Hypo - Mo Metal
Hyper- MoO_2

Mo Can Buffer Oxygen Content

see Table 12.3

How Do FP Oxides Actually Fit into the Oxide?

When oxidation state = 4, no problem!

When sesquioxides form, M_2O_3, and charge is +3, then electric neutrality is required;

1.) Produce oxygen vacancy (fig. 12.8b)

$$\frac{1}{2}O^{=} \text{ ¾ ¾} \rightarrow \frac{1}{2}O_2(g) + e^{-} +$$

2.) U atoms oxidize to U^{+5} or U^{+6} (fig. 12.8c)

U^{+4}----> $U^{+5} + e^{-}$

U^{+4}----> $U^{+6} + 2e^{-}$