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EVALUATION OF FUEL RELEASE RATE
AND MECHANISM TESTS UNDER
RBCB CONDITIONS

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INTRODUCTION

The following memorandum has been prepared to fulfill milestone B2 of SG006/BH872 (Advanced Oxide Fuel). This task includes theoretical evaluation of fuel/fission product release behavior from failed LMFBR fuel elements as well as an on-going experimental investigation of the mechanism of oxide fuel dispersal into flowing liquid sodium. The primary objectives of this work are to develop a fuel source term that can be used in predictive models for primary heat transfer system contamination and to understand the separate influences of important system variables (such as flow rate, oxygen impurity level) on this source term. The present report is written in two parts: the first, in condensed form, is an updated evaluation of fuel (U,Pu) and fission product release data, and the second describes the current status of supporting experimental work at General Electric's Vallecitos Laboratory.

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1. FUEL AND FISSION PRODUCT RELEASE RATES AND RELEASE FRACTION

1.1 Data Sources

In this evaluation two previous assessments of fuel and fission product release are considered^(1,2) together with actual release data from the following GE breached fuel pin tests: B9D⁽³⁾, B9A⁽⁴⁾, F2U⁽⁵⁾ and F11A^(6,7). For estimates of fuel release rates from breached mixed-oxide pins some recent French-German results from SILOE reactor loop tests⁽⁸⁾ have also been utilized. The two previous assessments were included in the current PSAR update for CRBR, chapter 11⁽¹⁾, and a report on improvements in the maintainability and inspection of LMFBR primary heat transfer systems⁽²⁾. A recent EPRI report has also reviewed the same data sources in some detail⁽⁹⁾, however, no new recommendations were offered.

1.2 Recommended Fuel/Fission Product Release Values

We have estimated fuel and fission product release rates and release fractions from a defected fuel pin having a significant cladding breach (exposed fuel area > 0.5 mm²). In this new assessment, the so-called "nonvolatile" fission products (i.e., the lanthanides, Sr, Ba, Zr, Nb, Ru, Mo, etc.) are treated as two separate classes - one (Class 4), showing limited volatility, the other (Class 5), behaving indistinguishably from fuel. In this latter case, transport in the coolant is primarily as (fuel) microparticles of 1-10 μ m diameter. These microparticles may originate by spallation or complete detachment of fuel grains (larger fuel particles may also enter the coolant but these are expected to 'sink' rather than 'swim'). True dissolution of fuel and nonvolatile fission products into liquid sodium is small⁽¹⁰⁾ and is not expected to contribute significantly to fuel/fission product transport.

Fission products have been grouped into five escape classes:

Class 1: Noble gases (Kr,Xe) and ¹³⁶Cs

Class 2: Other Cs isotopes (¹³⁴Cs,¹³⁷Cs)

Class 3: Volatiles - I, Sb, Te

Class 4: Semi-volatiles - Sr, Ba($^{140}\text{Ba}/\text{La}$), ^{141}Ce , $^{95}\text{Nb}/\text{Zr}$, (Ru?)

Class 5: Non-volatile fission products and fuel.

Once the cladding has breached, isotopes in Classes 1, 2 and 3 are released rapidly into the sodium coolant, whereas isotopes in Classes 4 and 5 are released more slowly at a continuous rate that is proportional to the exposed surface area of the fuel:

$$\begin{aligned}\text{Rate constant (Class 4)} &\approx 5 \times 10^{15} \text{ atoms/cm}^2\text{sec} \\ \text{Rate constant (Class 5)} &\approx 1 \times 10^{15} \text{ atoms/cm}^2\text{sec}.\end{aligned}$$

The Class 4 rate constant is based upon the (reference) Class 5 fuel release rate constant taking into account the higher apparent mobility/volatility of Sr, Ba, Ce, La, etc., resulting from their rare gas and alkali metal precursors. The reference (Class 5) rate constant of 1×10^{15} atoms/cm² is the highest value computed from measured fuel releases from pins such as B9D, F11A and Siloe S-4 (note the actual value computed for F11A was somewhat lower, $\sim 2 \times 10^{13}$ atoms/cm²sec).

Release fractions, including estimated 2σ uncertainties, are summarized in Table 1. For Classes 4 and 5, release fractions were computed for a hypothetical breached fuel pin residing in the reactor core for 60 days with a constant exposed fuel area of 1 cm². Distributions of the semi-volatile and non-volatile fission products between coolant, cold trap and PHTS inner surfaces were estimated primarily from the B9D⁽³⁾ and F11A/XY-2 results⁽⁷⁾.

1.3 Notes on Recommended Release Values

(a) The values in Table 1 for the fractions of material retained and released are, within a factor of 2, in agreement with values in chapter 11, PSAR/CRBR, for Classes 1-3. If a time interval of two to six months is assumed as a reasonable maximum residence time for a single breached pin, then agreement also exists for Classes 4 and 5. It is felt that the assumed

Table 1
Estimated Material Balance for a Breached Fuel Pin

Material/ Class	Fraction of Material- Retained	Material- Released	Fraction of Material -		"Plate Out"	Cover Gas
			Coolant	Cold Trap		
1 (Kr, Xe, ^{136}Cs)	~ 0	~ 1	$0.95(^{136}\text{Cs})$	$0.05(^{136}\text{Cs})$	~ 0	~ 1 (Kr, Xe)
2 (^{134}Cs , ^{137}Cs)	~ 0.5 -0.45 +0.45	~ 0.5 +0.45 -0.45	$0.2(^{134}\text{Cs})$ $0.1(^{137}\text{Cs})$	$0.3(^{134}\text{Cs})$ $0.4(^{137}\text{Cs})$	~ 0 ~ 0	0
3 (Volatile)	~ 0.8 -0.20 +0.15	~ 0.2 +0.20 -0.15	$0.02(^{125}\text{Sb}, \text{Te})$ $0.18(\text{I}, ^{127}\text{Sb})$	$0.18(^{125}\text{Sb})$ $0.02(\text{I}, ^{127}\text{Sb}, \text{Te})$	$0.16(\text{Te})$	0
4* (Semi-Volatile)	0.985 -0.035 +0.012	0.015 +0.035 -0.012	$\sim 10\%(\text{Sr}, \text{Ba})$ < 1%($^{95}\text{Nb}/\text{Zr}$, $^{140}\text{Ba}/\text{La}$, ^{141}Ce)	$\sim 10\%(\text{Sr}, \text{Ba})$ < 1%($^{95}\text{Nb}/\text{Zr}$, $^{140}\text{Ba}/\text{La}$, ^{141}Ce)	$\sim 80\%(\text{Sr}, \text{Ba})$ $\sim 99\%(^{95}\text{Nb}/\text{Zr}$, $^{140}\text{Ba}/\text{La}$, $^{141}\text{Ce})$	0
5* (Fuel & 'Fixed' Fission Products)	0.997 -0.006 +0.0025	0.003 +0.006 -0.0025	< 1%	$\sim 1\%^{**}$	$\sim 98\%^{**}$	0

*Exposed Area of Fuel = 1 cm^2 ;
Maximum residence time of breached pin = 60 days

**The distribution of U, Pu fuel and 'fixed' fission products between the cold trap and primary circuit inner surfaces will depend on many factors (e.g., size of particulates, flow rate, loop design) and hence may vary from system to system.

release fraction of 0.1 for plutonium given in Section 4.2 of GEFR-00405 is unrealistically high. It is also considered unrealistic to treat the fuel and the non-volatile (fixed), fission products as separate entities, as implied by the recommendation of different release fractions.

(b) The final four columns of the table are developed from values given in both chapter 11, PSAR/CRBR⁽¹⁾, and Section 4.2, GEFR-00405⁽²⁾, and in general these values agree within the large uncertainties that exist. There are two noticeable inconsistencies, however. The first of these is in the PSAR/CRBR Chapter 11. Tables 11.1-7 and 11.1-9 show, respectively, the plutonium "soluble" with 95% dispersed in the sodium coolant and also the cold trap very effectively (100%) removing plutonium from the coolant. The second inconsistency is that of treating release of the fuel and the residual fission products separately (both PSAR/CRBR and GEFR-00405). As mentioned earlier it is much more reasonable to treat the fuel-residual fission product mixture as discrete microparticulates. For this reason a single set of values is proposed for all members of Class 5.

(c) 'Drop-out' of large fuel fragments ($w > 5$ mg, $d > 1$ mm, say) is not considered to contribute directly to fuel release/transport values because of the unlikelihood that such large particles will be entrained in the flowing coolant. More correctly, the corresponding fuel release would be calculated from the area of the fragment and the appropriate rate constant.

(d) Some of the metallic radionuclides in Class 4 are expected to exhibit a downstream effect as a result of having short-lived alkali metal and/or rare gas precursors. In fact, this behavior has been observed for $^{95}\text{Nb/Zr}$, $^{140}\text{Ba/La}$ and ^{141}Ce in the recent XY-2 deposition sampler test⁽⁷⁾. There are a few inconsistencies, however. Ruthenium (^{103}Ru and ^{106}Ru) has a high release fraction^(3,7) yet does not appear to exhibit a downstream effect. On the other hand, ^{141}Ce exhibits a downstream effect but, at least in the XY-2 test, did not show a release fraction significantly different from ^{144}Ce and fuel.

1.4 Estimate of Plutonium Buildup in CDS/LDP Primary Coolant

An estimate of the Pu buildup in the PHTS of the Large Development Plant (CDS/LDP) was made using the Class 5 release rate ($1 \times 10^{15} \text{ U}_{1-x}\text{Pu}_x \text{ atoms/cm}^2\text{sec}$) and certain assumptions about the number of allowable breached pins, their residence times, and areas of exposed fuel, viz:

Pin failure level = 0.1%

No. of failed pins = 35

Exposed fuel surface/pin = 1 cm^2

Fraction of Pu in the $\text{U}_{1-x}\text{Pu}_x\text{O}_2$ fuel = 0.15

Amount of Na coolant = $2.38 \times 10^9 \text{ g}$.

Average breached pin residence time in reactor core = $5.184 \times 10^6 \text{ sec}$ (60 days)

Therefore, for the averaged Pu concentration in the coolant

$$[\text{Pu}] = \frac{1 \times 10^{15} \times 1 \times 5.184 \times 10^6 \times 0.15 \times 35 \times 239}{6.023 \times 10^{23} \times 2.38 \times 10^9}$$
$$= 4.5 \times 10^{-9} \text{ gPu/gNa, or, 4.5 wppb.}$$

At coolant temperatures of 600°C ($\sim 1100^\circ\text{F}$) or less, the major portion ($>90\%$) of the plutonium will still be associated with MO_2 microparticulates; very little true dissolution will occur as the solubility of PuO_2 at this temperature is $\text{ca. } 0.2 \text{ ppb}^{(10)}$. It should be noted that this calculated Pu level, which is an upper limit because it does not take account of deposition around the primary coolant circuit, is lower than either of the maximum allowable operating levels stated for CDS/LDP (10 ppb)⁽²⁾ or CRBR (100 ppb)⁽⁶⁾. Also, the calculated quantity of Pu released into the coolant is consistent with the fuel release fraction given in Table 1.

2. LABORATORY STUDY OF FUEL DISPERSAL IN SODIUM

2.1 Results from Capsule Experiments

Valuable insight into the separation of true solubility of oxide fuel in sodium from microspallation/mechanical dispersal effects was obtained earlier from results of some filter capsule experiments⁽¹⁰⁾. As well as providing estimates of the solubility limits of UO_2 and PuO_{2-x} in liquid Na over the temperature range 575-810°C this work showed that the nature of the dispersed fuel microparticulates ($d \leq 10 \mu\text{m}$) was very dependent on the physical state of the original mixed oxide fuel. That is, the finer microparticulates were composed of individual fuel grains whose sizes and compositions reflected the degree of U-Pu intermixing during fuel pellet sintering. From a physically blended fuel, for example, the smallest microparticles ($< 5 \mu\text{m}$) were Pu-rich whereas the larger microparticles ($5 \mu\text{m} < d < 10 \mu\text{m}$) were U-rich. For truly homogeneous fuel pellets (e.g., coprecipitation route) the composition of the dispersed microparticulates would be close to that of the original fuel for all size ranges. The true molecular solubilities of UO_2 and PuO_{2-x} in liquid Na with low oxygen content ($0 < 6.5 \text{ppm}$) were estimated to be

$$C_{\text{UO}_2} (\mu\text{g/gNa}) \leq 2.3 \times 10^4 \exp (-2.75 \times 10^4 / \text{RT})$$

and

$$C_{\text{PuO}_{2-x}} (\mu\text{g/gNa}) \leq 2.5 \times 10^8 \exp (-5.01 \times 10^4 / \text{RT})$$

where R is the gas constant ($= 1.987 \text{ cal/mol K}$).

2.2 Small-Scale Flowing Sodium Test Facility

A small, versatile flowing sodium test system was designed and built to investigate several important aspects of the sodium-fuel interactions that occur during run-beyond-cladding-breach operation of LMFBR fuel or blanket elements. For example, a logical extension of our previous characterization

work on fuel dispersal mechanisms⁽¹⁰⁾ was to perform experiments in which liquid sodium flows over the exposed fuel surface to determine whether flow rate (and oxygen level) influences the ratio of dissolved fuel to dispersed fuel.

The small-scale flowing sodium test facility (SFSTF), which is shown schematically in Figure 1, was built as an integral part of a high purity incot atmosphere glove box to facilitate the handling of test specimens. The total sodium inventory is ~0.8 kg and flow rates of up to 0.5 gal/min through the test section are possible. The test section has been designed to operate at ~700°C, and higher temperatures are attainable at low flow rates. Oxygen concentration is controlled by hot and/or cold trapping, and a continuous absolute measure is provided by a theoretical electrochemical sensor (oxygen meter). The sampling unit, which operates 200 to 300 degrees cooler than the high temperature test section, contains a removable holder for a variety of stainless steel filters. Photographs of the SFSTF at various stages of construction are shown in Figure 2. The main components of the (unmodified) loop system are illustrated in Figure 2a: (a) high temperature test section, (b) hot trap, (c) electrochemical oxygen meter, (d) sampling section, (e) cold trap, (f) dump tank (not shown), (g) electro-magnetic pump, (h) flow meter, (i) 5 KW heater, and (j) inert atmosphere glove box.

Initial evaluation of system performance was carried out towards the end of 1980. These test runs demonstrated that the main components were functioning satisfactorily, however, it became obvious that the oxygen meter would have to be repositioned in a bypass if desired test temperatures (>700°C) were to be attained. This modification is presently in progress and it is illustrated by the broken line in Figure 1.

In preparation for the initial fuel dispersal experiments some development work has been performed to establish the conditions necessary to successfully spike the outer layers of dense UO_2 pellets with 284-day ^{144}Ce . This

Figure 1 Schematic for Small-Scale Flowing Sodium Test Facility

(a)

(b)

Figure 2. Photographs of Small-Scale Flowing Sodium Test Facility before (a) and after (b) installation of trace heaters, insulation, etc.

is being attempted by applying small quantities of high specific activity $\text{Ce}(\text{NO}_3)_3$ aqueous solution to the surfaces of green UO_2 pellets prior to sintering. Rapid drying, followed by sintering in hydrogen at 1680°C is expected to diffuse small quantities of ^{144}Ce into the surface UO_2 grains, thereby providing a spike for subsequent grain identification during and after dispersal experiments. The purpose of the initial dispersal experiments will be to determine the extent of UO_2 dispersal as a function of Na flow rate, oxygen impurity level, and initial stoichiometry (O:U) of the urania specimens.

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