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by

H. K. Fauske, D. H. Cho, M. Epstein,
M. A. Grolmes, and R. E. Henry

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GENERIC CONSIDERATIONS OF LMFBR HYPOTHETICAL ACCIDENT ENERGETICS

H. K. Fauske, D. H. Cho, M. Epstein, M. A. Grolmes, and R. E. Henry
Argonne National Laboratory
Argonne, Illinois 60439

F. J. Martin and A. Padilla
Hanford Engineering Development Laboratory
Richland, Washington 99352

ABSTRACT

This paper provides a preliminary assessment of generic accident energetics issues associated with alternatives relative to the reference (U,Pu) oxide fuel in liquid metal fast breeder reactors. The alternatives considered include thorium- and uranium-based oxide, carbide and metal fuel types. This assessment is made within the context of low probability, but potentially large consequence accidents, e.g., core-disruptive accidents.

INTRODUCTION

Alternatives to the reference uranium-plutonium oxide fueled reactor and "conventional" aqueous reprocessing are currently being proposed and evaluated in the U. S. (NASAP)* and international (INFCE)** activities with the purpose of minimizing the risk of nuclear weapons proliferation and materials diversion. Thorium-based fuels have been characterized as alternative fuels¹ and are being evaluated in reactor systems²⁻⁴ because of a presumed higher level of resistance to proliferation and diversion of the denatured uranium-thorium cycle.⁵ It has also been noted⁶ that other fuel types, such as metal fuel, as well as oxide and carbide may also be compatible with one or more alternate reprocessing technologies. Without commenting on the relative merits of such alternatives or likely outcome of various ongoing assessments, it seems prudent to also consider the

*Nonproliferation Alternative Systems Assessment Program.

**International Nuclear Fuel Cycle Evaluation.

safety implications of the various fast reactor fuel candidates which might be a valuable adjunct to the proliferation and diversion resistance evaluations. It is not likely that safety considerations will be dominant in current evaluations, focused on nonproliferation; however, safety considerations are necessary and inevitable in the longer run.

It should be clear that a significant risk from fast reactors, as for thermal reactors, becomes only of concern in the case of a core disruptive accident (CDA). It follows that the key issue in the assessment of safety of fast reactors besides accident prevention is the potential for generating highly energetic events during a postulated CDA. While much attention has been given to detailed mechanistic calculations of CDAs using accident analysis computer codes (SAS,⁷ MELT,⁸ SIMMER,⁹ FUSS¹⁰), these codes do not readily lend themselves to an assessment of safety characteristics of the alternative fuel candidates. This is because the available codes have usually been developed to specifically handle the reference oxide fuel and/or they are still largely in a developmental stage. Hence, in this preliminary evaluation it was deemed necessary to rely upon physical principles in assessing safety implications of the alternative fuels.¹¹

Following this approach we find that significant differences in accident energetic characteristics are much more dependent on whether the fuel is in the oxide, carbide, or metal form, than on whether the fuel is ^{233}U or ^{239}Pu or the fertile material is ^{238}U or thorium with the exception of the sodium void effect. A discussion of these differences is given below.

KEY ENERGETICS ISSUES AND DESIRABLE FUEL CHARACTERISTICS

Relative to accident energetics in the LMFBR system, two generic conditions (independent of core design) are generally considered: 1) energetic recriticality (fuel collapse)¹² and 2) energetic fuel-coolant interactions (vapor explosion).¹³ However, the most important concern over potential energetics is the condition

that these phenomena may be coupled, i.e., some initial separation of the core which results in a localized vapor explosion which subsequently drives the fuel back into the core region. This situation is generally referred to as a pressure driven recriticality.¹⁴

The concern over energetic recriticality events is further amplified, since the levels of energetics (damage potential) resulting from hydrodynamic disassembly may be rather sensitive to small variations in ramp rate (current mechanistic estimates indicate the damage potential varies approximately as the square of the ramp rate through prompt critical).¹⁵ Since large uncertainties in the ramp rate of the magnitude that may lead to breach of current containment concepts are likely to remain high, it follows that it is desirable to be able to eliminate energetic hydrodynamic disassembly conditions altogether. Therefore, from a safety point of view, the following desirable fuel characteristics emerge:

- *Upon fuel disruption, the system should display dispersive* characteristics capable of driving the fuel to a permanent subcritical state, i.e., preclude gross fuel compaction and energetic gravity-driven recriticalities.*
- *Beyond fuel melting or conditions required for displaying dispersive characteristics, the system should not satisfy conditions which may lead to explosive** vapor formation, i.e., prevent overall energetic fuel-coolant thermal interactions and the energetic pressure-driven recriticalities.*

*Dispersive implies the presence of a volatile material (sodium or steel) which is able to do work by receiving heat from the disrupted fuel. This effect will provide extended or long-term dispersal as compared to possible short-term dispersal by fission-gas release.

**Explosive characteristics imply significant energy transfer between fuel and coolant resulting in vaporization on a small time scale relative to the acoustic relief time of the system.

In addition to the two generic conditions, the design-dependent condition of positive sodium-void worth in LMFBRs must also be considered.* This problem is of little or no concern for ^{233}U -based fuel,** but with a large homogeneous core of ^{239}Pu -based fuel the net sodium-void reactivity may be 5 to 8 dollars. Following a postulated unprotected loss-of-flow accident, fuel failure conditions may be reached due to sodium voiding in high power subassemblies, with the reactor near prompt critical and a substantial fraction of the core still not voided. In this condition it is theoretically possible that fuel pin failures in both voided and unvoided channels, may lead to superprompt burst conditions at high ramp rates due to fuel densification near the midplane and/or accelerated sodium voiding. It is also theoretically possible that fuel dispersal may override further reactivity additions due to sodium voiding. The problem here is that the level of energetics becomes sensitive to mechanistic details of competing material motions on a millisecond time scale in a highly disrupted geometry. Hence, for the ^{239}Pu -based fuel, the following additional desirable safety characteristics emerge:

- *Subjected to unprotected loss-of-flow or transient overpower conditions, the fuel should display early disruption[†] and dispersive characteristics to cancel sodium-void reactivity well before reaching prompt-critical condition.*

A preliminary evaluation of the availability of these desirable safety characteristics relative to fuel type is indicated in Table I. *If sodium-bond vaporization can be demonstrated as an effective means for early and extended fuel*

*In addition to possible design options based upon limited fuel removal, this problem may be largely eliminated through heterogeneous designs, but with the penalty of increasing the fuel inventory and thus increasing the doubling times.

**For ^{233}U -based fuel the fuel capture-to-fission ratio is relatively insensitive to energy-spectrum variations, while this effect is significant for ^{239}Pu fuel.

[†]Early disruption implies a relatively short time between incipient sodium voiding and fuel disruption in lead subassemblies, since the sodium boiling temperature represents the upper limit of operation prior to ultimate fuel disruption for all fuel types.

Table I. Relationship between Desirable Safety Characteristics and Fuel Type for Unprotected Transients (LOF and TOP)

Safety Characteristic	Oxide	Carbide	Metal
<p>Extended fuel dispersal following fuel disruption</p> <p><i>Concern: Gravity Driven Recriticality</i></p>	<p>Favorable;* by clad vaporization. Steel boiling point \approx fuel melting point in LMFBRs.</p>	<p>Less favorable; steel boiling point well above fuel melting point. Na-bond not available since steel melting point well below fuel melting point. May be provided by fission gas release.</p>	<p>Favorable;** by Na-bond vaporization since the (U,Pu) fuel melting point well below clad melting point. Less favorable for thorium-based metal fuel, since fuel melting point is above cladding melting point.</p>
<p>Lack of explosive fuel-coolant interaction following extended fuel dispersal</p> <p><i>Concern: Pressure Driven Recriticality</i></p>	<p>Favorable;† interface temperature well below spontaneous nucleation for Na.</p>	<p>Less favorable; interface temperature sometimes exceeds spontaneous nucleation for Na.</p>	<p>Favorable; if Na-bond fuel dispersal is effective. Fuel melting temperature slightly above Na-boiling temperature.</p>
<p>Early fuel disruption and dispersal</p> <p><i>Concern: Sodium Void Effect and Fuel-Coolant Interaction</i></p>	<p>Less favorable; 4-5 s^{††} between fuel pin dryout and disruption.</p>	<p>Less favorable; 3-4 s^{††} between fuel pin dryout and disruption.</p>	<p>Favorable; \approx 1.0 s^{††} between fuel pin dryout and disruption for (U,Pu) fuel. Less favorable for thorium-based fuel.</p>

*Some in-pile demonstration exists.

**No demonstration exists.

†Based on extensive analysis and experiments, significant explosive energetic fuel-coolant interactions are precluded with essential certainty for the UO₂-PuO₂ system.

††Equivalent time at full power.

‡²³⁹Pu-based fuels.

dispersal, the (U,Pu) metal fuel would appear to have the potential to satisfy all the proposed desirable safety characteristics with the oxide fuel a close runner-up.

The less favorable position of carbide fuels as viewed in this context could be improved by one or more possible ways. We note that compared to oxide fuels, little active safety attention has been given to carbide fuels. It should not be precluded at this time that further development will successfully establish mechanistic means for accommodating these characteristics. Alternately, an equivalent safety basis might also be accomplished by more attention to design features which preclude whole-core accidents.

Further specific details of accident energetics potentials for the various fuel types are given below relative to unprotected and protected accidents.

ACCIDENT ENERGETICS

Prime candidates for satisfying all the desirable safety characteristics would appear to be a fuel element (fuel, bond, and cladding) which becomes dispersive and which does not display explosive characteristics upon contacting the coolant. These considerations lead to safety concerns for the carbide fuels because of less favorable behavior relative to timely fuel dispersal and increased potential for explosive fuel-coolant interactions as compared with the oxide types (uranium- as well as thorium-based).

For oxide fuels, the cladding boiling point is roughly equal to (uranium-plutonium) or somewhat lower (thorium) than the melting point of the fuel suggesting that steel vapor from clad boiling can provide an effective dispersal mechanism (see Table II). Explosive sodium vapor formation is not likely to be involved for oxide-fuel/sodium systems (see Fig. 1). U-Pu metal fuels which are bonded with and have become logged with sodium have a potentially similar dispersive mechanism through the fact that sodium bond would begin boiling very nearly

Table II. Temperature Characteristics for Oxide, Carbide, and Metal Fuels

Material	Oxide, °C	Carbide, °C	Metal, °C
Na Boiling Temperature	~ 950	~ 950	~ 950
Na-bond Boiling Temperature	-	~1150 (fresh fuel pin)	~1150 (fresh fuel - can be regulated by fuel pin fill pressure)
Clad Melting Temperature	~1400	~1400	~1400
Fuel Melting Temperature	~2800	~2400	~1150
Clad Boiling Temperature	~2800	~2800	
Fuel Boiling Temperature	~3300	~4500	

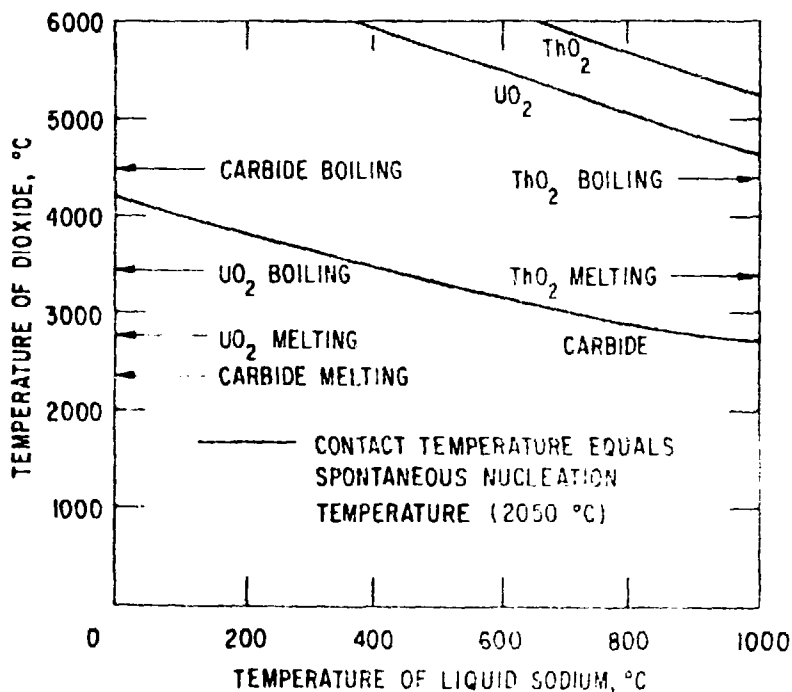


Fig. 1. Illustration of Necessary Temperatures for Oxide and Carbide Fuels to Cause Spontaneous Nucleation (Explosive Boiling) upon Contact with Liquid Sodium.

coincident with initiation of fuel disruption¹⁶ (i.e., at the metal fuel melting temperature which is below the cladding melting temperature - see Table II).

In this context it would appear important that the cladding material remains intact so as to prevent escape of the sodium bond prior to fuel melting. The

latter condition would appear to be assured since the rates of cladding penetration upon exceeding the eutectic temperature for the metal-clad alloy is relatively slow compared to the time scale for unprotected transients of interest (see Table III). If this fuel dispersal by sodium bond vaporization can be

Table III. Cladding Penetration Rates above the Eutectic Temperature for (U,Pu) Metal (10 w/o Zr). (Inferred from Results Obtained on the U-5 wt% Fs/SA 304 Stainless-steel System).¹⁶

Temperature, °C	Rate, mils/min
~ 800	~0.1 + eutectic temperature
~ 900	~0.5 + Na boiling temperature
~1020	~3.0
~1100	~8.0
~1150	10 mils/s + fuel melting temperature

demonstrated, the U,Pu metal fuel appears at least as favorable as the U,Pu oxide fuel with respect to desirable safety characteristics. Some less likely phenomena (e.g., extensive fuel slumping) would be required to raise the fuel temperature sufficiently to achieve an explosive fuel-coolant interaction. The dispersive potential appears less favorable for thorium-based metal fuels since the cladding melts at a lower temperature than does the fuel which would allow sodium bond to escape prior to fuel disruption; the safety concerns then become similar to those noted for carbide fuels.

Monotonic fuel dispersal characteristics of carbide fuels appears less favorable since the clad boiling point is well above the carbide melting point (see Table II), and thus the prospects for an early termination of the accident via fuel removal are less likely than for the oxide and metal fuel cases. However, the relatively high fission-gas retention for this fuel type may lead to extended fuel dispersal as compared to oxide fuel.¹⁷ At temperatures near its boiling point where extended carbide fuel dispersal becomes possible, a strong argument

on the basis of physical principles can no longer be used to eliminate the concern for explosive formation of sodium vapor. In the case of the carbide-sodium system, spontaneous nucleation is satisfied upon contact (see Fig. 1). However, while considerable experimental support is available,¹⁸ we note that the spontaneous nucleation criterion as a guide for assessing the possibility of an explosive energetic fuel-coolant interaction is not universally accepted.¹⁹ In particular, the potential for a pressure-driven recriticality event would appear to be more difficult to rule out for the carbide fuel. Therefore, in comparison with the oxide fuel, significantly more detailed mechanistic descriptions of core-disruptive accident phenomena are likely to be required to assess accident mitigation and consequences.

We further note that for (U,Pu) metal fuel, the low melting temperature and favorable dispersive characteristics provided by the sodium bond at least partially offset the fact that the sodium void coefficient tends to be more positive than for oxide and carbide fuel. However, if in some way a large positive reactivity ramp rate could be introduced before the fuel was molten, the possibility would exist for significant conversion of fuel thermal energy to work by the sodium bond. The reactivity ramp rates likely to be introduced by sodium voiding are not large enough for this to be a problem; some less likely phenomenon would be required. It is also only for very high ramp rates that the lower Doppler coefficient for the metal fuel would become a problem. Energy release in the fuel for more probable ramp rates would not be much larger for the lower Doppler coefficient of the metal fuel.

In regard to the sodium void effect in general, we note that the void coefficient is much smaller in ²³³U fueled systems which largely eliminates the incentive for heterogeneous cores. This is particularly true for the metal fuel where

the ^{233}U -Th system hardly has a central positive sodium void region and it is clearly negative for the bulk of the core.*

Finally, in the case of a postulated loss-of-heat-sink accident,** oxide and carbide fuel would experience disruption only after the coolant level drops below the core, since sodium boiling would appear capable of removing decay-heat power levels.²⁰ It therefore follows that the mechanical damage potential from a loss-of-heat-sink accident is small even if an energetic recriticality event is postulated, since there is no liquid sodium to transmit the kinetic energy to the reactor vessel head components. This favorable safety characteristic may not be available for the metal fuels because the low-melting eutectic temperature between the fuel and clad ($\sim 800^\circ\text{C}$) is such that fuel disruption may occur in the presence of coolant. However, in this case continued sodium vaporization may provide fuel dispersal, thereby preventing energetic recriticalities.

CONCLUDING REMARKS

- The apparent dispersive nature of oxide fuel and the likely absence of recompaction forces (inherent physical characteristics) may be used to explore relatively simple design features (such as lead subassemblies with upper structures removed to allow early and sufficient fuel removal prior to whole-core voiding to adequately compensate for the sodium void worth) so as to provide a possible alternative to the heterogeneous core concept in eliminating the longstanding concern about the problem of the positive sodium void coefficient. The feasibility of introducing such design features must also be considered relative to generating potential adverse effects associated with normal functional requirements.

*It is noted that on the basis of breeding performance ^{233}U -Th cores are less attractive than ^{238}U -Pu for oxide, carbide and metal.

**For this accident class, which by definition leads to whole-core involvement, the core meltdown process occurs at low power (decay heat) and is therefore treated separately from unprotected accidents.

- *If extended fuel dispersal by sodium bond vaporization can be demonstrated, the (U,Pu) metal fuel appears at least as favorable as the oxide fuels relative to the generic accident conditions (recriticality and fuel-coolant interaction). Some less likely phenomenon would be required like extensive fuel slumping to raise the fuel temperature sufficiently to achieve an explosive fuel-coolant thermal interaction condition. On the other hand, for the thorium-based metal fuel, the dispersive potential appears less favorable. In this case the safety concerns becomes similar to that noted for the carbide fuels.*

- *For (U,Pu) metal fuel, the low melting temperature and apparent favorable dispersive characteristics at least partially offset the fact that the sodium void coefficient tends to be more positive than for oxide and carbide. However, if in some way a large positive reactivity ramp rate could be introduced before the fuel was molten, the possibility would exist for significant conversion of fuel thermal energy to work by the sodium bond. It is also only for very high ramp rates that the lower Doppler coefficient for the metal fuel may become a problem. In regard to the sodium void effect in general, the void coefficient is much smaller in ^{233}U fueled systems which largely eliminates the incentive for heterogeneous cores.*

- *There are safety concerns with the carbide fuels, because of less favorable characteristics relative to timely fuel dispersal and benign fuel-coolant interactions, as compared with the oxide and metal fuel types. In particular, the potential for a pressure-driven recriticality event would appear to be more difficult to rule out for this fuel type. It would seem most appropriate for carbide fuel types, that consideration be given to designs which assure early accident termination either by engineered inherently safe shutdown systems or lead subassemblies of a different fuel composition such as metal which may be shown to be inherently dispersive early in the accident sequence.**

*It is with pleasure that we note and acknowledge that this possibility was suggested in the course of discussions with Dr. E. P. Wigner.

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