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Journal of Nuclear Materials 320 (2003) 147-155



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# Safety aspects of oxide fuels for transmutation and utilization in accelerator driven systems

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#### Abstract

General safety aspects of fuels under development for accelerator driven systems (ADS) are reviewed and discussed. These fuels should allow a maximization of transmutation and incineration rates, which excludes fertile  $UO_2$  as a component or matrix. The accumulated knowledge on data, phenomena and scenarios of fast reactors with  $(U,Pu)O_2$  oxide fuels and sodium cooling serves as background for this review. For future ADS both the reactor system itself, the fuel and the coolant are innovative compared to traditional critical fast reactors. For the fuel, these boundary conditions lead to many open questions, starting from basic thermal physical, thermal mechanical and irradiation data to the behavior under transient conditions. The choice of fuel naturally has a significant impact on whole core behavior and safety too, including the influence on related neutronics parameters, on failure propagation and disruption behavior under accident conditions. Key safety issues are discussed and a first assessment of phenomena and scenarios is given. Areas of research and technology in which further work is required to resolve important safety issues are highlighted. © 2003 Elsevier Science B.V. All rights reserved.

# 1. Introduction

Fuel behavior and fuel failure are a central concern for the economic operation of any reactor. This will also hold for reactors dedicated to transmute and incinerate nuclear waste. Extensive irradiation and test programs, combining experimental techniques and theoretical analyses have to be established. They should provide the necessary data on fuel life, operating limits and fuel behavior under transient and accident conditions. In the current paper the latter aspect is specifically treated. Innovative fuel types are required to exploit the full potential of accelerator driven systems (ADS) in multistrata fuel cycle strategies [1]. Generally, these ADS fuels will be subjected to the conditions of a fast neutron spectrum. So-called 'dedicated fuels' should allow the maximization of incineration and transmutation rates and are characterized by a high minor actinide (MA) content and the lack of the classical fertile materials as U238 or Th232. These requirements have a severe impact on fuel and plant safety [2–4]. To cope with deteriorated safety parameters of these fuels, the subcriticality of the ADS is an important pre-requisite for their utilization. In other transmutation strategies, mainly with critical reactors, the development of MA targets, with low MA content, inserted into the core or at the core periphery is investigated. Conventional fuel can thus be utilized [5]. This development is not discussed here.

The safety potential of a new type of fuel should be assessed early in the development of such an ADS, since safety evaluations can have a significant impact on the conceptual design of the reactor and the plant. The early safety studies should also help to pre-select potential candidates of fuel, thereby saving resources and helping to focus on necessary research programs. Covered under the fuel safety aspect are fuel failure mechanisms and phenomena, which are normally not encountered under reactor operation and might involve degraded geometry. Typical examples of safety related experimental series for such conditions have been performed in the CABRI

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[6] and TREAT [7] facilities. A wealth of information has been gained mainly for fast  $(U,Pu)O_2$ , metal, and carbide reactor fuels with sodium as coolant. Based on the results of the tests, extrapolation to full-scale reactor conditions could be performed with sophisticated analytical techniques and codes.

The choice of an ADS as a transmuter may involve three important changes compared to the already built and operated fast reactors with sodium cooling. The system is a subcritical one, driven by an external source, a new innovative fuel and heavy liquid metal (HLM) coolant as Pb or Pb/Bi may be utilized. Gas cooling could be another choice, but there does not exist operational experience with a fast reactor system too. The three mentioned changes influence each other and have to be seen in one context. The intended innovations have a significant impact on all safety aspects of the system. Time scales for power changes in an ADS could be significantly shorter compared to neutronically critical systems [8,9], requiring some shock-resistance of the fuel. Furthermore, since the HLM coolant has a much higher boiling point than sodium, the accident scenarios with voiding and pin disruption investigated in the CABRI and TREAT tests, cannot be directly transferred to the new conditions. The high density of the HLM coolants will also lead to buoyancy effects, not observed with sodium coolant.

The safety behavior of most transmuter fuels is unknown and only some general tendencies in relation to the classical  $(U,Pu)O_2$  fast reactor fuels can be deduced. Since the properties of these dedicated fuels significantly deviate from those of classical fast reactor fuels, it is important, both to reassess the list of known safety issues and topics and to identify, if specific and unique new issues exist. A reasonable starting point for safety investigations is the reexamination of common safety issues.

Safety considerations are important for a successful implementation of these fuels, finally demonstrating reasonable behavior for the whole spectrum from normal operation to low probability events which could lead to core destruction. The successful development of these fuels represents a corner-stone of the ADS program.

## 2. Safety assessment for new fuels in ADS

For a complete safety assessment of an ADS with dedicated fuel, one has also to investigate the whole spectrum of events from normal operation to severe core disruptive accidents. For normal operation, economical aspects play a significant role. A high availability of the plant would result in the desired high transmutation rates. For economics and radiological reasons (fuel cycle) high burnup levels have to be achieved. During ADS operation, deviations from steady state power generation may occur related e.g. to beam losses and beam trips, power ramping, power/flow mismatches and coolant flow disturbances causing clad temperature raise. Those transients can lead to a fuel element lifetime shortening and fuel pin failures. Fuel failure limits and operational margins versus reactor protective, instrumentation surveillance and shut-down systems have to be determined.

As the fast spectrum ADS core is not in its neutronically most reactive configuration, the assessment of severe accidents will play a similar role in the safety assessment as for critical fast reactors. The subcriticality is not a decisive issue here, as the intrinsic fuel material worth is much higher [2-4] and fuel rearrangement could eliminate this subcriticality. The safety assessment will therefore also include unprotected accidents (corresponding to a failure of beam tripping) with core melting and core disruption. This class of accidents is judged as hypothetical, because of the very low probability of occurrence. These accidents are nevertheless investigated because of their damage potential. In addition, these investigations provide information on the very intrinsic behavior of a reactor under fault accident conditions, they help to identify and exclude cliff-edge effects and they provide information for containment design. The ultimate goal of these efforts is to exclude very severe accident scenarios by design. As for an ADS, we leave the safe ground of experience in many ways, it is especially advisable to look carefully into the safety issues of severe accidents. As the subcriticality is traded-in for the generally deteriorated safety parameters of dedicated fuels/cores, the subcriticality might not be counted as an additional safety asset [4].

In the current paper the focus is not on operational safety but more oriented towards transients and accidents and their relation to the new fuels. Special emphasis is put on the assessment of the propagation and escalation potential from local disturbances to subassembly scale or even core involvement.

#### 3. Potential safety problems of dedicated cores

The safety issues of dedicated fuel could be related to the impact on the fuel pin itself, the behavior of the subassembly and the impact on the whole core behavior. For the 'pin-level' thermal physical data, irradiation behavior, etc., for the subassembly and core-level additionally neutronics aspects become of more importance.

#### 3.1. The impact of new fuels on fuel pin behavior

Fuels specifically designed for transmutation of minor actinides (MAs) and/or plutonium are called 'dedicated' fuels, as their composition, their chemical state and fuel form are optimized for this special purpose. At present, a wide variety of concepts is considered for dedicated fuels where various combinations of chemical state, fuel state and fuel form are possible. The chemical state can be a metal, nitride or oxide (carbides and hydrides are discussed too), the fuel state can be a solid solution or a composite (a ceramic-ceramic CERCER, a ceramic-metal CERMET or a metal-metal METMET), the fuel form can be a pellet or a (coated) particle. In Europe main activities will concentrate on oxide fuels [8], extending the past experience of MOX fuel. The absence uranium in the fuel has a significant impact on the fuel properties. The solid solution dedicated fuels generally have a lower melting point than U-based oxide fuel, and the thermal conductivity will be lower too. This will result in a smaller margin to melting. Because of changes in properties of the actinide elements and compounds along the series, also potential differences in actinide redistribution during irradiation (e.g. AmO<sub>2</sub>), increased clad corrosion, higher fission gas release and pressure built-up due to formation of relatively large amounts of helium (resulting from alpha-decay) deserve closer investigation. The drawback of the limited thermal margin can be mitigated by the use of composite fuels, in which the matrix is a good thermal conductor. An extensive experimental program ranging from determination of basic quantities as thermal physical properties, from irradiation to safety related experiments will be necessary for a final assessment on these innovative fuels. As will be noted later, thermal physical properties might have a significant impact on the whole range of accident phenomena and scenarios.

Within the European 5th Framework Programme 'FUTURE', first investigations and analyses of uranium-free dedicated fuels containing americium are under way [10]. Fuels to be studied are  $(Pu_xAm_y)O_{2-k}$  and  $(Pu_xAm_yZr_2)O_{2-k}$  fuels. The fuels for transmutation may also contain Np and Cm. Besides the homogeneous fuels, composite fuels with MgO, steel or tungsten are under discussion. One should note that the MAs may form solid solutions with ZrO<sub>2</sub>, but not with MgO or steel. Describing melting conditions of composite fuels under accident conditions will therefore become more complex [11]. First investigations showed that for operational and safety reasons the MA content (in relation to Pu) might have to be limited to about 40-45% [10]. Many of the properties of these advanced fuels are presently unknown and have to be estimated. Thermal properties for MA compounds, as thermal conductivities, thermal expansion (an important inherent safety feature), specific heats, etc. are unknown. The more this holds for the equation of state data as vapor pressures or liquid compressibilities. For safety analyses in the severe accident range one may need data up to 6000 K and more. For standard fast reactor fuel these high temperature data are available [12,13]. There it becomes obvious that for the dedicated transmuter fuels described above large 'white areas' exist. In Table 1 some typical data of oxide fuels under consideration in comparison with standard fast reactor (U,Pu)O<sub>2</sub> fuel is given. The table should give some impression of the range and tendencies of the thermal physical data in relation to standard fast reactor fuel.

#### 3.2. The impact of new fuels on core parameters

For demonstrating the impact of new fuels on core safety parameters, we refer to some generic analyses for a heavy metal cooled ADS with 'dedicated' oxide fuel [2-4]. In these analyses a dedicated fuel with a high MA load has been chosen. The background strategy scenario of this fuel is a double strata concept with a first stratum global reactor fleet of UOX-PWR, MOX-PWR and LMFRs, delivering MAs (and Pu, if needed for operational purposes) into a second stratum ADS fleet for final incineration [1]. The transuranic (TRU) fuel used for the analyses was a mix of 25% Pu and 75% MAs in a ZrO<sub>2</sub> matrix. The ADS had a thermal power of 1200 MW with a subcriticality level of  $k_{\rm eff} = 0.98$ . The neutronic calculations for this specific core gave a Doppler constant of about -100 pcm, a  $\beta_{eff}$  of  $\sim 150$  pcm and a very short neutron generation time of  $\Lambda \sim 2.0 \times 10^{-7}$  s. These data already show the deterioration of the kinetics parameters in a dedicated core. A considerable positive core void has been calculated with about 5030 pcm and the steel worth amounts to about 2240 pcm. Note that steel melting will advance any HLM coolant boiling, and

Table 1 Thermal physical data of some oxide fuels

1 2				
	$(U_{0.8}Pu_{0.2})O_2$	$(Pu_{0.8}Am_{0.2})O_2$	$(Pu_{0.32}Am_{0.08}Zr_{0.6})O_{2-k}$	MgO matrix
Melting point (K)	3023	[2630]	[2770]	3250
Thermal conductivity ( $W m^{-1} K^{-1}$ )	2.3	[1.6]	[1.6]	6.5
Specific heat $(kJ kg^{-1} K^{-1})$	0.64	[0.36]	[0.50]	1.36
Density $(kg m^{-3})$	11 080	11 510	[7960]	3580
Boiling point (K)	3811	_	_	_

The data in brackets have been estimated from known data on comparable systems, the thermal conductivity and specific heat data refer to 1737 K.

steel relocation may introduce positive reactivity. The available reactivity potentials from steel relocation and HLM voiding could eliminate the subcriticality margin built into this core. The coolant voiding problem is relieved by the high boiling point of Pb or Pb/Bi. However, core voiding could be triggered by breached pins and a massive (He) gas blow-down from the fission gas plenum. The production of He by the Am and Cm decay is a common feature of dedicated fuels with high MA content and for potential shut-down periods in a transmuter one would have to take into account the still ongoing helium production by decay. The criticality potential of the fuel is large, with about 60 available critical masses. Any local subassembly melting with subsequent fuel compaction processes could drive the core towards criticality.

For transients under design basis conditions, the ADS with a dedicated core seems to behave in a stable way [4]. Under these conditions, the source-dynamics dominates all other processes. The safety problems are more related to severe transients and accidents, where the safety parameters (void, Doppler, critical masses) exert their maximum influence. Entering these severe transient or accident regimes, the following issues might cause problems in ADS dedicated cores:

- the high intrinsic core material reactivity worths;
- mechanisms that could lead to a core material rearrangement and eliminate the built-in subcriticality level;
- the lack of sufficient fast negative feedback and the deteriorated kinetics parameters.

According to the safety analyses in [2–4], a type of cliff-edge effect could exist for dedicated cores with the implication of severe power transients. The lack of an impeding, prompt negative reactivity feedback, as e.g. the Doppler effect, together with deteriorated kinetics parameters, could lead to a drastic increase of the potential for accident energetics. In [2–4] a number of design and safety measures are proposed to prevent and mitigate these severe accident potentials. One important measure is the limitation of the MA quantities in the fuel.

We want to emphasize that many design proposals for improving e.g. the void, the Doppler coefficient, the neutron generation time, etc., relying on the introduction of moderators (e.g. hydrides) or specific geometric choices, as the p/d ratio, fail, when they should play their role during accident initiation and progression. Known moderators have very low dissociation temperatures, dissociation and separation processes may even introduce additional reactivity. Optimized p/d ratios loose their meaning under core-melt conditions. The fore-mentioned measures may play their role in accident prevention, but for guaranteeing a non-energetic accident behavior under core-melt conditions and preventing cliff-edge effects, other type of safety measures are necessary [4]. The safety strategy and the installed measures should assure a non-energetic accident evolution for early and late accident phases, even with whole core disruption.

## 4. Categorization of safety issues for transmuter fuels

As a starting point and orientation for the safety assessment, phenomena and scenarios related to  $(U,Pu)O_2$  fast reactor oxide fuel are considered. If the general classes of safety problems stay the same for the new innovative transmuter fuels, is yet unknown. Even if the general classes would remain the same, the phenomenology, the individual sequence of events and the potential consequences might change. This requires analytical work to be performed. As a consequence, the known phenomena and scenarios from standard oxide fuel have to be reassessed and reexamined and possible new and unique safety features of the innovative dedicated fuel have to be identified. A sound fuel characterization under steady state and transient conditions has to be developed and failure criteria have to be derived for various accident scenarios.

In [14] the lines of defense approach (LOD) is utilized to classify safety issues, which can be followed for transmuter fuel too [3]. In the first line one is concerned with normal operation and prevention of any malfunction, proceeding via limitation of core damage up to containment of accidents and their consequences in the primary system, which corresponds to a whole core or plant involvement. Finally the attenuation of radiological products completes the lines.

A listing of safety issues is presented in Table 2, roughly classified along the LOD [15,16] with the emphasis on severe transients with the potential of core melting and disruption. The table starts with issues related to the limitation of core damage and accident escalation, then proceeds to core and containment issues. Safety issues refer to problem areas which have to be investigated and resolved to assure the overall safety of the plant.

Based on Table 2 some general safety trends can be deduced, however most of the details in phenomenology and scenarios are unknown. For the new fuels there seem to exist common features and characteristics with  $(U,Pu)O_2$ , but also marked differences can be noted. The changes in thermal physical properties, e.g. thermal conductivities (compared to classical FR fuel) could significantly influence accident scenarios. One has to discriminate between the solid solution fuels and the composite fuels, because of their largely different characteristics. Higher thermal conductivities in composite fuels might lead to less restructuring, crack-healing Table 2

Key safety issues related	to core damage li	imitation, primary	system, containment,	, and attenuation of ra	diological consequences

Safety issue	Problem	Faults/phenomena
Limitation of core damage and f		<ul> <li>Cladding defects</li> <li>Fuel-pin loading error</li> <li>Blockage formation</li> <li>Fuel stability</li> <li>Fission gas and He behavior + kinetics/release fractions/release mode</li> <li>Fault propagation and speed</li> <li>Fuel release</li> <li>Fuel-coolant compatibility</li> <li>Detection</li> <li>Coolability</li> <li>Long term failed fuel operation limitian</li> </ul>
Subassembly-to-subassembly propagation	Can thermal or mechanical failure of subassembly and duct walls lead to attack of adjacent subassem- blies and propagation?	<ul> <li>Propagation and speed</li> <li>Fuel stability at melting</li> <li>Blockages</li> <li>Energetic events</li> <li>Detection</li> <li>Coolability</li> </ul>
Limit of core damage from whole core accident initiators	What is core damage resulting from whole core accident initiators and what route does core-melt accidents take, non-energetic or energetic?	<ul> <li>Early accident termination by fuel squirting, fuel sweep out/relocation</li> <li>Fuel stability at melting</li> <li>Power excursions and mechanical co disassembly</li> <li>Non-energetic thermal meltdown</li> <li>In-place coolability</li> </ul>
Primary system/containment/atte Accident energetics: voiding	enuation of radiological consequences Can voiding processes introduce enough reactivity to eliminate subcriticality or lead to power increase with pin failures in non-voided regions and autocatalytic effects?	<ul> <li>Voiding by fisgas/He blow-down</li> <li>Local voiding or clad relocation triggering over-power transients</li> <li>Autocatalytic processes</li> <li>In-pin compactive fuel motions</li> </ul>
Accident energetics: clad relocation	Can clad removal introduce enough reactivity to eliminate subcriticality or lead to power increases with pin failures and autocatalytic effects?	<ul> <li>Clad relocation with its impact on later fuel motion</li> <li>Fission gas/He effects</li> <li>Clad relocation triggering over-pow transient</li> <li>Blockage formation</li> </ul>
Accident energetics: recriticality	Can fuel compaction lead to criticality and prompt critical states?	<ul> <li>Fuel dispersal potential</li> <li>Fuel compaction potential (Fission gas + He pressures) versus fuel squirting + relocation</li> <li>Fuel stability and stability of matrix</li> <li>Fuel melting and separation of components</li> <li>Fuel melting versus steel boiling</li> <li>Molten versus granulated fuel</li> <li>Coherency of fuel motion (power profiles)</li> <li>Liquid fuel compressibility under compaction</li> <li>Viscosity of fuel rubble configuratio</li> <li>Fuel release from core (blockage</li> </ul>

• Fuel release from core (blockage versus relocation potential) (*continued on next page*)

Table 2 (continued)

Safety issue	Problem	Faults/phenomena
		<ul><li>Pressure driven recriticalities</li><li>Tuning of power excursions</li><li>Boosting of power excursions</li></ul>
Accident energetics: disassembly	Can accident paths lead to mechanical disassembly and how does energetic excursion behave?	<ul> <li>Doppler feedback</li> <li>Kinetics parameters</li> <li>Vapor pressures</li> <li>Thermal fuel expansion</li> <li>Inert gas pressures</li> <li>HLM coolant inertia</li> </ul>
Accident energetics: fuel coolant interactions (FCI)	Can thermal reactions between fuel and coolant play a role for accident energetics?	<ul><li>Mechanical loadings</li><li>Pressure driven processes (recriticalities)</li></ul>
System mechanical response	Can accident scenarios challenge the primary and containment system. What is appropriate design basis and what is the expected response?	<ul> <li>ADS topology</li> <li>Impact of HLM coolant</li> <li>Fuel/HLM heat transfer</li> <li>Fuel/HLM vessel loads + reaction with concrete</li> </ul>
Post accident heat removal	How is post accident heat removal impacted?	<ul> <li>Decay heat load</li> <li>Fuel redistribution in vessel</li> <li>Partially intact pin structures</li> <li>Core debris behavior and fuel bed heights</li> <li>Fuel-HLM compatibility</li> <li>Coolability</li> <li>Criticality</li> </ul>
Radiological consequences	What is the radiological source term?	<ul><li>High MA content</li><li>HLM interaction</li></ul>

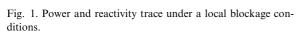
and higher fission gas retention. Fission gas induced fuel swelling coupled with low fuel creep rates at operating temperatures might lead to higher cladding loads. With its impact on the fission gas release the dispersive potential of these innovative fuels, a mechanism to shutdown nuclear excursions is strongly determined. For  $(U,Pu)O_2$  fast reactor fuel the thermal conductivity also has a pronounced influence on the Doppler effect, which is of lesser importance for dedicated fuels with their already small Doppler. The influence of fuel conductivity on thermal reactions with the coolant (FCI) and pressure build-up is a minor issue in the case of HLM cooling. The conductivity will also play a role in post accident heat removal, if under core disruptive conditions fuel is released from the core and settles in the vessel.

For all fuels the high temperature stability is of concern. The segregation of e.g.  $AmO_2$  would cause problems both in solid solution and composite fuels. The marked difference in the melting and evaporation behavior of solid solution and composite fuels, where the individual phases of the latter fuels will melt and evaporate at different temperatures, will complicate any accident analysis. This could mean a significant increase of the recriticality potential, if the fissile component would no longer be 'diluted', could move separately from its matrix and compact. The dispersive potential of fuel/steel mixtures, known from FR oxide fuel, because of the proximity of the fuel melting and steel boiling point (<sup>fuel</sup> $T_{melt} \sim {}^{steel}T_{boil}$ ), will be diminished for the solid solution and composite fuels, both due to the higher distances of the relevant phase transition temperatures, but also by the differences in scenarios (compared to sodium cooled reactors), with steel melting advancing coolant boiling.

# 4.1. Limitation of core damage and prevention of escalation

Solid solution fuel with reduced thermal conductivity and lower melting points will also experience lower power-to-melt ratios and might reveal a tendency for pin-to-pin failure propagation. Axial location and type of failure together with internal and external fuel motion will influence the further scenario. Gas release from breached pins could interrupt cooling. Molten fuel, ejected from the pin could granulate and trigger local blockage formation. Severe FCIs can however be excluded. In the case, a high conductivity matrix exists, rapid pin-to-pin propagation can be excluded, as molten fuel release is not likely and molten fuel release would not lead to an energetic reaction with the HLM coolant. Slow blockage build-up and propagation is an issue which has to be investigated carefully for the different types of fuel. HLM coolants are prone to the formation of additional thermal resistance at the fuel rod surfaces as consequence of a separation of impurities from the coolant flow and their adhesion at the hot surfaces [17]. PbO oxides transported by the flow might be accumulated on the clad walls. An additional aspect is the utilization of grids due to the usually larger p/d ratios in HLM flows. From experiments it is well known that spacer grids tend to 'collect' debris and favor horizontally spreading blockages [18]. Detection of such passive blockages is difficult. Some very preliminary calculations show indeed some propagation potential under blockage conditions. Total blockages of the inner subassembly ring (RZ geometry) have been assumed in some scoping calculations with the SIMMER-III code [19]. These calculations were mainly performed to test the operability of the code and to obtain possible trends of the accident evolution under such extreme conditions. SIMMER-III is well suited to calculate blockage formation and propagation processes, however, due to the total lack of experimental information the results have to be regarded with caution. To describe the SIMMER code family, SIMMER-III is a two-dimensional (RZ,XY), three-velocity-field, multi-phase, multi-component, Eulerian, fluid-dynamics code coupled with a structure model (fuel pins, etc.) and a space-, time- and energy-dependent neutron dynamics model. The transient time-dependent neutron flux distribution is calculated with the improved quasistatic method based on a transport theory solution for the space-dependent part. Besides the SIMMER-III code, the 3D SIMMER-IV code (XYZ,RZO) is under development and operation.

After a reactivity loss by fuel redistribution, the damage propagates to neighboring subassemblies causing a reactivity and power increase. The calculations displayed in Fig. 1 may indicate that investigations have to be performed to clarify this safety issue.

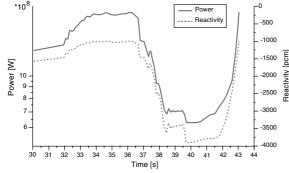


Mechanical subassembly-to-subassembly propagation by energetic fuel coolant interactions (FCIs) can be excluded under HLM conditions. For composite fuels, the thermal physical properties will support the nondispersiveness of material configurations, leading to the formation of low viscosity fuel rubble with further tendency for blockage formation and plugging. Again, stability of the fuel matrix and processes of blockage formation with matrix material are unknown but of essential importance for the course of accident scenarios. Possible eutectic formations have to be carefully monitored. An important point is the timely detection of blockages under HLM conditions. Analyzing severe unprotected accidents it becomes obvious that under the 'beam-on' assumption, the neutronic effects of fuel squirting /6/, fuel dispersal and sweep-out, i.e. the introduction of negative reactivity and its impact on the power level, is by far not so effective as in a critical reactor. This 'power stability' could definitely be a trigger for propagation. In the case that sufficient positive reactivity is added and criticality is approached, not only the power level goes up, but by the change of the fundamental shape factor, the peripheral core regions may see higher power densities. The high He production/ content, either in the fuel or in the fission gas plenum is an important safety topic. Depending on the 'location' and 'timing' the gas release, it could either act on the fuel in a dispersive or in a compactive way. The unprotected subassembly propagation accident (USAP) thus needs special attention, also including the aspect of timely detection.

For whole core accident initiators as the unprotected over power (UTOP) and the unprotected over current (UTOC), a class of reactivity and beam (neutron source) disturbances will exist which will not lead to whole core damage. Fuel squirting/sweep out or fuel rubble removal might lead to conditions of an in-place coolability of the partially destroyed-reactor configuration. With the HLM coolants and their high boiling points some coolability potential can be expected. Accidents as the unprotected loss of flow (ULOF) and the unprotected loss of heat sink (ULOHS) will probably always involve the whole core (if no special measures as a third shut-down level [4] has been installed). Thus as a general tendency one could deduce that these whole core initiators have the potential to lead to a whole core involvement. Again, the high helium production will play a role here, with its impact on voiding processes and e.g. fuel compaction processes through a 'gun-barrel' effect, pushing the remaining fuel pellets or fuel particles into the core.

# 4.2. Primary system/containment/attenuation of radiological consequences

Voiding by boiling processes is a minor issue in HLM cooled reactors, but voiding by fission gas and He



release from damaged pins might represent a potential mechanism. Due to the high boiling point of the coolant clad relocation will proceed any boiling process, could set the fuel stack free and add reactivity by clad motion and later compaction of fuel rubble. Again blockage formation by steel freezing may take place. Due to the close density ratio of the fuel (depending also on the porosity) with the coolant, 'fuel compaction' may not necessarily take place only in the downward direction. The recriticality issue is still a concern, as sufficient 'critical' masses may be available under fuel compactive conditions. Most probably the fuel will not exist in molten form, but as rubble with high viscosity, consequently lower compactive velocities and lower coherence of any material motion has to be expected. Under these conditions no severe excursions and core disassemblies would be encountered initially. Under-cooled rubble configurations will finally melt and molten fuel motions might become possible in confined, blocked core areas. The dispersiveness of these configurations may be lower than for normal FR oxide cores because of the arguments given already above on dispersiveness of innovative fuels. The fuel compressibility under single phase conditions and the equation of state plays a significant role under fuel compactive motion conditions, as the energy release in a recriticality is the lower, the more rapid and the higher single phase pressures in the liquid can build up. The stability of the fuel is a major concern in the recriticality phenomenology. In the case the fuel could separate from its matrix, or parts of the compound could disintegrate, recriticalities could more easily become possible. An important feature of the standard (U,Pu)O<sub>2</sub> fuel is the non-separation of Pu and U under melting conditions. One can imagine the increase of the recriticality potential if the PuO<sub>2</sub> would melt and become mobile before UO2. The phenomenon of tuning [22], where coherency of compactive fuel motions (and consequently increased reactivity ramp rates) is enforced via the neutronics and thermal hydraulic coupling, may be more significant in an ADS, with a strong external source and reduced reactivity feedback. The phenomenon of boosting [22] may be worth investigating, where mobile fuel in the core peripheries is heated up and axially expands into the high fuel worth and source importance regions. A further essential point is the exclusion of pressure driven recriticality scenarios via FCIs. Though the concern in sodium cooled reactors on this issue could be relieved over the years with numerous experiments and analytical work [20,21], the potential was still existent. Due to the combination of the innovative fuel with the HLM coolant this concern can be discarded. No energetic FCI's have to be expected for innovative fuels reacting with Pb or Pb/Bi. Any severe power excursion and disassembly process will probably be more severe than in cores with  $(U,Pu)O_2$  fuel, if the Doppler feedback and the neutron generation time cannot be significantly improved in dedicated fuel cores. But in addition the disassembly is a strong function of the vapor pressure, the gas (He) content of the fuel and the confinement of the fuel masses. The heavy coolant could serve as a hampering buffer with its large inertia and could favor an additional energy accumulation before dispersal. Under the conditions of a late core-melt configuration it may be well assumed that the beam has been shut-off in the meanwhile and that the negative reactivity feedback of the core disassembly may become fully effective. If the beam is still assumed to be 'on' at least the source importance would have been reduced in the distorted core configurations. Concerning the system mechanical response, the fuel may not have the decisive influence. Thinking of very severe scenarios, the coolant may play a more dominant role. If under core disassembly conditions with fuel vaporization, the surrounding coolant above the core is accelerated and hits the upper lid of the reactor, the scenario with HLM shows a different characteristics compared to sodium [23]. The high density, low compressibility and low HLM vapor pressures lead to a shock impact characteristics, whereas with sodium coolant, lower, but persisting pressures will load the vessel structures [23]. The question of post accident heat removal is again closely related to the fuel and is an example that severe accident safety issues may have a significant impact on the design of the plant: Firstly, the higher decay heat of the MA fuel has to be taken into account; secondly, the close density ratio of the fuel and HLM coolant make the distribution of any mobile (disrupted, granulated) fuel within the vessel with its internal structures a difficult task to predict and determine. This has to be taken into account in the overall decay heat strategy. A partial solubility of fuel in the coolant would have a major additional impact on decay heat removal, as redistribution of heat sources my interfere with natural convection cooling strategies. Finally, the thermal physical properties will determine coolable rubble bed heights, hot spots and criticality conditions of the fuel. Referring to the radiological consequences and the fuel impact, one has to remember that the fuel contains large amounts of Pu, Np, Am and Cm which has to be taken into account in a containment design. This area again opens a wide field for research.

## 5. Conclusions

Based on this first assessment it can be concluded, that similar classes of safety problems as for classical fast reactor oxide fuels can be identified for the new innovative transmuter fuels. However the importance of the individual safety classes, the key phenomenology, the specific sequence of events, the accident scenarios and the potential consequences could change significantly. There do exist several important areas and phenomena where the ADS, representing a new system with specific time scales and dynamics, with a new innovative fuel and new coolants, may differ from the critical fast reactor system with  $(U,Pu)O_2$  fuel and sodium cooling. Stability questions of the fuel and its behavior in relation to a heavy coolant with high boiling point need special attention. The recriticality phenomenon and issues related to the PAHR phase deserve special attention. The following major items and their impact on accident scenarios have to be investigated and assessed:

- thermal expansion of fuel;
- stability of the fuel;
- melting separation of components;
- high temperature behavior;
- dispersiveness of the fuel/steel/coolant system;
- content, distribution and kinetics of fission gases and He;
- fuel and clad failure conditions and behavior under various transient scenarios;
- blockage formation;
- propagation potential;
- fuel/coolant compatibility and reactions;
- fuel redistribution in HLM pools.

Though the above issues were deduced for oxide dedicated fuels, similar concerns hold for metal and nitride fuels with high MA load. To investigate these issues a broad technological program has to be launched. This must include investigation of (1) the fundamental material properties up to the high temperature range, (2) the irradiation behavior, (3) the interaction/ reaction between fuel, clad and coolant, (4) the fuel and fuel pin behavior under transient conditions. A sound fuel characterization for steady state and transient conditions has to be developed and failure criteria have to be derived for accident scenarios. A sufficient experimental capability, both out-of-pile and in-pile, has to be provided to tackle these issues. In addition the analytical capability (integrated accident codes) has to be significantly extended, including the modeling of the new fuel, the simulation and description of the new phenomenology and scenarios. A basic problem and obstacle is the obvious incoherence of time scales in experimental and analytical efforts. In summary, significant efforts will be necessary to assess the relevant safety issues when transmuter fuel is introduced and replaces the well known traditional oxide fast reactor fuel.

#### References

 M. Salvatores, I. Slessarev, G. Ritter, P. Fougeras, A. Tchistiakov, G. Youinou, A. Zaetta, Nucl. Instrum. and Meth. A 414 (1998) 5.

- [2] W. Maschek, A. Rineiski, K. Morita, M. Flad, in: GLOBAL 2001, Paris, France, 9–13 September 2001.
- [3] W. Maschek, A. Rineiski, S. Wang, M. Flad, K. Morita, in: International Conference on Nuclear Applications for the New Millennium, AccApp'01 & ADTTA'01, Reno, USA, 11–15 November 2001.
- [4] W. Maschek, A. Rineiski, M. Flad, K. Morita, P. Coste, Nucl. Technol. 141 (2003) 2.
- [5] A. Vasile, G. Rimpault, J. Tommasi, C. De Saint Jean, M. Delpech, K. Hesketh, H. Beaumont, T. Newton, P. Smith, W. Maschek, D. Haas, Ch. De Raedt, G. Vanbenepe, J.C. Lefevre, in: International Conference on Back-End of the Fuel cycle: From Research to Solutions, GLOBAL 2001, Paris, France, 9–13 September 2001.
- [6] I. Sato, U. Imke, W. Pfrang, J. Papin, M. Berne, in: Proceedings of the International Topical Meeting on Sodium Cooled Fast Reactor Safety, Obninsk, Russia, 3– 7 October 1994.
- [7] A.E. Klickman, D.H. Thompson, W.A. Ragland, A.E. Wright, R.G. Palm, R.J. Page, in: Proceedings of the LMFBR Safety Topical Meeting, Lyon, France, 1982.
- [8] D.C. Wade, in: OECD/NEA 6th Information Exchange Meeting on Actinide and Fission Product P & T, Madrid, Spain, 2000.
- [9] R.J.M. Konings (Ed.), EUR 19928 EN, Institute for Transuranium Elements, 2001.
- [10] FUTURE, FUels for Transmutation of TransURanium Elements, Contract FIKI-CT-2001-00148, 5th Framework Programme EU, 2001.
- [11] M.A. Mignanelli, R. Thetford, in: Proceedings on Advanced Reactors with Innovative Fuels, Chester, UK, 2001.
- [12] W. Breitung, K.O. Reil, Nucl. Sci. Eng. 101 (1989) 26.
- [13] W. Breitung, K.O. Reil, Nucl. Sci. Eng. 105 (1990) 205.
- [14] J.D. Griffith, J. Graham, P. Greebbler, R.T. Lancet, in: International Meeting on Fast Reactor Safety and Related Physics, Chicago, USA, 1976.
- [15] D. Rose, L.W. Deitrich, M.A. Grolmes, D.R. Ferguson, in: International Meeting on Fast Reactor Safety and Related Physics, CONF-761001, Vol. 3, Chicago, USA, 5–8 October 1976.
- [16] M.A. Grolmes, in: Topical Meeting Proceedings Advanced LMFBR Fuels, Tucson, USA, 10–13 October 1977.
- [17] A.A. Sedov, IAEA TECDOC-1157, LMFR Core Thermohydraulics: Status and Prospects, IAEA, Vienna, Austria, June 2000.
- [18] C.v. Minden, G.F. Schultheiss, GKSS 82/E/50, Forschungszentrum Geesthacht, 1982.
- [19] S.A. Kondo, H. Yamano, T. Suzuki, Y. Tobita, S. Fujita, X. Cao, K. Kamayama, K. Morita, E.A. Fischer, D.J. Brear, N. Shirakawa, M. Mizuno, S. Hosono, T. Kondo, W. Maschek, E. Kiefhaber, G. Buckel, A. Rineiski, M. Flad, P. Coste, S. Pigny, J. Louvet, T. Cadiou, JNC TN9400 2001-002, Japan Nuclear Cycle Development Institute, 2000.
- [20] H.K. Fauske, Nucl. Safety 550 (17) (1976) 5.
- [21] K. Konisji, M. Isozaki, S. Imahort, A. Furutani, SA. Kondo, D.J. Brear, IWGFR/89, O-arai, Japan, 1994.
- [22] T.G. Theofanous, C.R. Bell, NUREG/CR-3224, 1984.
- [23] W. Maschek, A. Rineiski, K. Morita, G. Mühling, M. Flad, R. Konings, in: IAEA TCM Meeting, Argonne, USA, 2000.