Behavior of Transmuter Fuels of Accelerator Driven Systems under Severe Accident Conditions


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For exploiting the generic features of accelerator driven systems (ADS) in multi-strata fuel cycle strategies, innovative fuels have to be developed. These so-called dedicated fuels should allow a maximization of incineration/transmutation rates. They are characterized by a high MA content and possibly by the lack of classical fertile materials as U238 or Th232. Generally, these fuels will be subjected to the conditions of a fast neutron spectrum ADS. A wide variety of fuel concepts is considered, such as various combinations of chemical state, fuel state and fuel form are possible. The chemical state can be a metal, nitride or oxide, the fuel state can be a solid solution or a composite, the fuel form can be a pellet or a (coated) particle. European R&D for ADS fuel will mainly concentrate on oxide fuel forms such as inert matrix mixed oxide or composites in which the oxide actinide phase is mixed with an oxide or a metal matrix.

At the start of the advanced fuel development the safety behavior of these fuels is rather unknown. However, the safety considerations are important for a successful implementation of these fuels. The safety assessment has to address the whole spectrum from normal operation to the behavior under low probability events which could lead to core melting and destruction. First analyses of these dedicated fuels reveal significant differences compared to the classical (U,Pu)O2 fast reactor fuels in the high temperature range. This may have a significant impact on accident scenarios, especially for severe accident conditions. The impact of these significant changes on the safety of a waste burning ADS is investigated and first results are presented. Requirements for modelling in codes and necessary experiments are outlined. A successful development of these fuels represents a cornerstone of the ADS transmuter program.

KEYWORDS: Accelerator Driven System, Transmuter, Dedicated Fuel, Safety, Core Disruptive Accidents

I. Introduction

Fuels in ADS with the mission of transmutation of minor actinides (MAs) and plutonium (Pu) are called ‘dedicated’ fuels, as their composition, their chemical state and fuel form are optimised 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 and 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 fuels1, extending the past experience of MOX fuel. The omission of uranium from the fuel has a significant impact on the fuel properties. A solid solution dedicated fuel will 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. In addition, actinide redistribution during irradiation (e.g. AmO2), increased cladding corrosion, higher fission gas release and pressure build-up due to formation of helium (resulting from alpha-decay) have to be taken into account. Helium release from the fuel during normal operation will lead to an increased pin-pressure. The increased He content will have a decisive influence on pin failure mechanisms under transient conditions.

To cope with the fore-mentioned deteriorated thermal-physical conditions, composites like CERCER or CERMET or solid solution containing an inert component will be the choice for transmuter fuels. The feasibility and fabricability of such composite fuels is presently under investigation (see e.g. Ref. 2). To help selecting the most promising fuel candidate, the feasibility studies on these fuels also comprise safety investigations. These safety analyses are closely related to the basic design of an ADS for MA incineration, possessing reasonable safety parameters. In a second step the transient behavior of such fuels has to be assessed up to the point of
accident conditions. From different analyses it is known that such accelerator driven transmuters (ADTs) operated with innovative fuels may possess deteriorated safety parameters, which have to be coped with the in-built subcriticality. The hypothetical severe accident regime, which also covers core melting conditions, fuel rearrangement scenarios and therefore the potential for the elimination of the subcriticality, has therefore to be investigated and the possible consequences have to be assessed. As will be outlined in the following chapters, the new dedicated fuels show some new features and phenomena under design extension conditions (DEC), which have not been encountered in the classical (U,Pu)O₂ fast reactor fuels. The safety conditions for these fuels has therefore to be re-assessed. An extensive experimental program ranging from irradiation to safety related experiments will be necessary for a final judgement on these innovative fuels.

II. Dedicated Fuels and Their Thermal-Physical Properties

For the assessment of dedicated fuels various thermal-physical data have to be generated over the temperature range required by the application of the defence-in-depth strategy. Currently these data are rather incomplete, especially for the high temperature range. The safety analysis codes like SIMMER-III require a rather complete set of equation of state and thermal-physical data up to the critical point. For (U,Pu)O₂ a rather complete data base up to 10 000 K has been generated over the years. For the MA containing fuels basically all data above the melting transition are missing. It is one of the important tasks and future research issues to provide these safety relevant high temperature data also for the dedicated fuels.

The melting points of minor actinide oxides are much lower than for ordinary fast reactor fuel with an average Pu enrichment of ~20%. For AmO₂ in Ref. 1 the melting temperature is given at 2448 K, the melting temperature of (Puₐₐ Amₐₐ)O₂ is estimated at 2630 K (Ref 2). The lower thermal conductivity both of the Pb/Bi coolant and of the MA compounds leads to restrictions on the linear rating during normal operation conditions. In addition, the lack of a significant Doppler³ contributes to a higher vulnerability to overpower accidents. A high fuel thermal conductivity is therefore essential and CERMET and specially tailored CERCER fuels become of main interest. In addition it has to be taken into account that during irradiation the thermal conductivities will decrease further.

Analyses³ show that the dedicated fuel should contain a significant share of non-degraded plutonium, with a share of approximately 40%. Fabricability, neutronics and safety considerations define limits for the matrix materials, with a range of 30-40% for highly neutron absorbing and 50-60% for low absorbing materials. Details of a wide range of investigations will be given in Ref. 9. In Tab 1 various potential matrix materials are given and show the large difference of thermal conductivities¹⁰.

<table>
<thead>
<tr>
<th>Fuel</th>
<th>Matrix</th>
<th>Melting Temp. [K]</th>
<th>Thermal Conductivity (Wm⁻¹K⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CERCER</td>
<td>ZrO₂</td>
<td>2983</td>
<td>2.3</td>
</tr>
<tr>
<td>CERCER</td>
<td>MgO</td>
<td>3100</td>
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<tr>
<td>CERMET</td>
<td>Cr</td>
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<tr>
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<td>Mo</td>
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<tr>
<td>CERMET</td>
<td>W</td>
<td>3695</td>
<td>115.0</td>
</tr>
</tbody>
</table>

Tab 1 Melting temperatures and thermal conductivities for several matrix materials (thermal conductivities given at 1100 K)

III. Safety Analyses with Core Melting Conditions

The safety objectives common to all approaches for future nuclear plants, including accelerator driven transmuters (ADT) are to protect individuals, society and the environment from harm, to ensure that in all operational states radiation exposure within the installation is kept below prescribed limits and as low as reasonably achievable and to take all reasonably practicable measures to prevent accidents in nuclear installations and to mitigate their consequences should they occur.

These safety objectives are achieved through the application of the defense-in-depth strategy. The application of these fundamental safety principles to the ADT concept requires that attention is paid to the special safety features of these concepts, which are a combination of characteristics of liquid-metal fast reactors (LMFRs) with the important feature of sub-criticality and the connection of the core with an external neutron source. In the defense-in-depth strategy attention is also given to design extension conditions (DEC), which include scenarios and phenomena with core destruction and melting. The current paper mainly deals with the safety issues of the innovative fuels under severe accident conditions. The pre-transient conditions of the fuel might be strongly influenced by the beam stability in accelerator driven systems. However, given the time-frame for the ADT development beam stability should no more be a problem and its potential influence is not discussed here.
Severe accident conditions in an ADS are analyzed in a similar way as in critical reactors. Within the EU 5th FP PDS-XADS, a paramount list of transients and accidents is analyzed currently for an 80 MWth ADS with ordinary fast reactor fuel. Unprotected accidents (beam-on) as the ULOF, UTOP, ULOHS, blockage accidents and transient over-current (UTOC) accidents are studied. The small-size ADS with ordinary fuel shows a remarkable un-vulnerability against severe transients. For a larger ADS with MA fuel, safety problems could be identified and special preventive and mitigative measures have to be taken to provide an adequate level of safety. As examples for initiators with the potential for severe transients, including pin destruction and melting, a UTOP scenario, with a reactivity addition approaching criticality and an unprotected (beam-on) blockage accident are shown in the following. Due to the lack of the prompt negative Doppler effect in an ADS with dedicated fuel, any transient could proceed rather unhampered, once the subcriticality level is eliminated. Thermal expansion could substitute the missing Doppler to some extent. In the case of insufficient prompt feedback a core disassembly would have to be expected, as material motion would then be the only relevant negative reactivity feedback. In Fig. 1 a UTOP accident in a (U,Pu)O₂ fuelled ADS is shown for demonstration. The pin failure under flowing Pb/Bi leads to an effective fuel sweep-out in this SIMMER-III simulation, and the ADS is neutronically shut-down. The effects of fuel sweep-out, the impact of the spacer grids on blockage formation etc. need further investigations and have to be fully understood for a sound assessment of an ADS under severe UTOP conditions.

The other simulation assumes a blockage in the central ring of subassemblies around the target device. Again clad melting and subsequent fuel redistribution leads to a reduction of reactivity and power. The power and reactivity development of this blockage accident simulation is given in Fig. 2. In Fig. 3 the fuel particle distribution after uncladding of the pins is displayed and shows the expanding damage in the central core area. The particles are swept away from the high importance regions and the reactivity is reduced as a consequence.

In all cases no severe power excursion is observed but a benign shut-down of the ADS by fuel sweep-out is observed. It should however be noted that these are very first simulations of an ADS under heavy liquid metal cooling conditions, and that they do not include the complication of a new innovative fuel. The uncertainty of calculations with the new fuels will be even higher.

IV. Procedure for Safety Assessment and Identification of Safety Issues
For the discussion of the DEC behavior of advanced transmuter fuels the lines of defense approach (LOD) has been applied in Ref 16,
which allowed the identification and classification of open safety issues. The first line of LOD is concerned with normal operation and prevention of any malfunction. The issues to be discussed comprise mainly:

- Pin-to-pin Failure Propagation
- Subassembly to Subassembly Propagation
- Limit of Core Damage from Whole Core Accident Initiators

Under DEC conditions the LOD covers the limitation of core damage up to the issue of containment of accidents and their consequences in the primary system, which corresponds to a whole core or plant involvement. The following safety issues were covered in Ref 16.

- Accident Energetics: Voiding
- Accident Energetics: Clad Relocation
- Accident Energetics: Recriticality
- Accident Energetics: Disassembly
- Accident Energetics: Fuel Coolant Interactions (FCI)
- System Mechanical Response
- Post Accident Heat Removal
- Radiological Consequences

This procedure led to the identification of specific safety issues to be more closely investigated for these dedicated fuels.

V. Safety Issues for Dedicated Fuels

The following assessment mainly reflects the conditions of an ADT with Pb/Bi cooling. Similar work has to be performed for gas cooled ADS options too. Based on the safety issues given in Chapter IV important phenomena and accident scenarios are deduced, their possible impact is discussed and necessary research areas are identified:

1) Stability of envisaged fuel composites CERMET or CERCER at high temperature and under melting conditions:
A key question is the potential of the separation of components under high temperature conditions. The impact of ‘multiple’ melting points has to be analyzed. As can be observed from Fig. 4, the melting points of e.g. AmO₂ and most of the matrix materials matrix differ significantly. In the case of the separation of the fissile and the underlying matrix, subsequent layering and fuel compaction, a recriticality concern exists. Further question are related to the matrix stability after fuel separation, matrix disruption and subsequent liquid/particle agglomerations. The viscosity of these mixtures defines the velocity of any fuel motion and the blockage versus fuel relocation potential. Due to the coupling of neutronics and thermal-physical/mechanical/thermal-hydraulic fuel behavior, any change of the fuel/matrix ratio opens up specific accident scenarios.

2) Dispersiveness of fuel configurations:
In a general sense the dispersiveness of core material mixtures is important for the achievement of non-compact fuel configurations, implying neutronically a low reactivity and the absence of recriticality phenomena. Different mechanisms such as boiling and intrinsic pressure built-up could prevent compacted fuel configurations. For ordinary (Pu,U)O₂ oxide fast reactor fuel a key safety issue and dispersal mechanism was the close proximity of the fuel melting point (Tₘₐₐₜ) with the steel boiling point (Tₜₜₜ). It could even be claimed that in a general sense the safety philosophy for FRs was built on this principle. For the new dedicated fuels with a much lower melting point, or ‘multiple’ melting point, this condition does not hold true any longer. As can be observed in Fig. 4, the heat generating constituents AmO₂ or (Pu,Am)O₂ have melting points way below the steel boiling point. The impact of this new situation has to be investigated and the potential contribution of the Pb/Bi coolant to the dispersal potential should be analysed. Pb/Bi could serve as a ‘dispersive substitute’ for steel. The dispersive potential of steel may strongly depend on the topology and dynamics of the fuel/steel mixture, where local steel vapour blanketing can also hamper the heat transfer. Out-of-pile experiments as BALL TRAP will provide further insight into these issues. Additionally, to obtain a better insight into these phenomena, an in-pile experimental series as SCARABEE-N would be required. In Fig. 4 the melting points of various fuel and matrix materials are compared with the boiling point of steel and coolant.

![Fig. 4 Melting point of various fuels and matrix materials versus boiling points of steel and Pb/Bi](image-url)
3) Behavior of un-clad pin stubs:
Under the conditions of the high boiling point of Pb/Bi, clad melting will proceed in time any coolant boiling, e.g. under ULOF conditions. Clad melting will leave behind partly un-clad fuel stacks. The potential blockage formation of steel freezing melting will leave behind partly un-clad fuel stacks.

4) Fuel/coolant interaction/redistribution/post accident heat removal:
‘Classical’ FCI processes with pressure build-up, will play a less significant role under Pb/Bi conditions, compared to sodium cooled reactors (despite the high heat conductivity of the composite fuels). However, un-clad fuel or molten fuel could partly go into solution with the coolant and the densities of fuel and coolant are similar. These effects could lead to a complex redistribution of fuel in the vessel and the primary system after a core disruption. Depending on the time-scale of these processes, the fuel could be diluted and redistributed leading to a reduction of the core reactivity and the elimination of a recriticality concern. The processes may however severely influence the post accident heat removal.

5) Fission gas and He behavior:
MA fuels will contain He as an additional pressure source. The amount, location, and kinetics of the combined fission gas and He release has to be investigated. The presence of pressure sources could lead to both dispersive (fuel dispersal) and compactive effects (in-pin fuel compaction, pin-stub motion by plenum gas).

6) Fuel condition as function of burn-up:
Impact of burn-up (transmutation) on fuel conditions has to be understood. The transmutation process will strongly shift the relation of the isotopes in the fuel and will have an impact on the thermal-physical quantities.

7) Fuel pin failure modes:
The fuel pin behavior and potential failure modes under various transient conditions and accident scenarios have to be investigated. The impact of burn-up (transmutation) on failure modes has to be understood.

8) Disassembly processes:
Due to the neutronic ‘features’, as a reduced Doppler feedback and the smaller neutron generation time, more severe excursions could take place than in cores with (U,Pu)O$_2$ fuel. In addition, a disassembly process is strongly determined by 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.

9) Post Accident Heat Removal:

The PAHR problem is 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. The potential of fuel solubility and the close density ratio of fuel and coolant have to be taken into account in the overall decay heat removal strategy. A partial solubility of fuel in the coolant would have a major impact on decay heat removal, as redistribution of heat sources may interfere with planned natural convection strategies.

10) Radiological Consequences:
The impact of fuel is mainly related to the large amounts of Pu, Np, Am and Cm in the core, which has to be reflected in the containment design.

These 10 points comprise a first list of safety issues which have to be tackled in detail for a better understanding of innovative dedicated fuels under severe accident conditions.

VI. Conclusion
For the new ADT fuels the knowledge base has to be significantly expanded compared to the ordinary oxide fast reactor fuel under sodium cooling conditions. Based on the present assessment it can be concluded, that for ADTs with dedicated fuels the key phenomena, the specific sequence of events, the accident scenarios and the potential consequences could change significantly compared to those for (U,Pu)O$_2$ fuelled, sodium cooled fast reactors. Stability questions of the fuel, its behavior in relation to a heavy coolant with high boiling point, the recriticality phenomenon and issues related to the PAHR phase deserve special attention.

In a first step a rather complete set of high temperature thermal-physical data has to be generated. Without this knowledge base any safety analysis will be plagued by very large uncertainties. A reliable fuel characterization for steady state and
transient conditions has to be developed, and failure criteria have to be derived for the different accident conditions and scenarios. The following major items and their impact on accident scenarios have to be investigated and assessed:

- thermal physical properties over the required temperature range in dependence on burn-up and transmutation processes
- stability of the fuel and melting separation of components
- 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 and propagation potential
- fuel/cooler compatibility and reactions
- fuel behavior and redistribution processes under the condition of Pb/Bi cooling

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 modelling of the new fuels, covering new phenomena and scenarios. 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.

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