HIGH BURNUP FUEL ISSUES

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One of the major current challenges to nuclear energy lies in its competitiveness. To stay competitive the industry needs to reduce maintenance and fuel cycle costs, while enhancing safety features. Extended burnup is one of the methods applied to meet these objectives. However, there are a number of potential fuel failure causes related to increased burnup, as follows: 1) *Corrosion* of zirconium alloy cladding and the water chemistry parameters that enhance corrosion; 2) *Dimensional changes* of zirconium alloy components, 3) Stresses that challenge zirconium alloy *ductility* and the effect of hydrogen (H) pickup and redistribution as it affects ductility, 4) Fuel rod internal pressure, 5) Pellet-cladding interactions (*PCI*) and 6) pellet-cladding mechanical interactions (*PCMI*). This paper discusses current and potential failure mechanisms of these failure mechanisms.

KEYWORDS : High Burnup, Zirconium Alloys, Nuclear Fuel, Failure Mechanisms, Normal Operation, LOCA, RIA, Corrosion, PCI, PCMI, Dimensional Changes

1. INTRODUCTION

One of the major present challenges to nuclear energy lies in its competitiveness. To stay competitive the industry needs to reduce maintenance and fuel cycle costs, while enhancing safety features. Extended burnup is one of the methods applied to meet these objectives. There are a number of issues that need resolution to be able to successfully implement extended burnup.

While the nuclear design of fuel can apparently be extended well beyond our current burnup limits, albeit at some economic cost, the physical performance of materials and components can have a finite end as burnup increases and no amount of analytical work will extend their life.

The performance of the critical fuel components is the result of a complex interaction of a large number of variables that challenge the evaluation of the mechanisms in progress and the prediction of their behaviour at extended and more severe conditions. The technologies involved include just about every aspect of materials science imaginable: properties of materials, metallurgy, structural mechanics, coolant chemistry, physical chemistry, and their basic mechanisms just to mention a few examples. In addition, exposure to radiation changes all of the physical properties and processes: the properties of the structural materials and of the coolant change, transformations in structure and composition occur in all the materials (true alchemy!), and these processes occur in a non-homogeneous and non-equilibrium manner throughout the core.

A study of the materials' performance is difficult even outside the reactor's radiation field and provides limited data. Test reactors offer a good tool for evaluating a limited number of variables and mechanisms and have provided some valuable data, however, the operation and use of these reactors is expensive. The final performance evaluation is in the power reactor itself since it provides all the variables of importance; however, the lack of instrumentation, the inability to control testing time, as well as the difficulty of separating variables makes interpretation of ongoing processes difficult. The final evaluation of new materials and fuels for high burnups progresses necessarily through the stages mentioned: ex-reactor testing, test reactor evaluation of samples, power reactor evaluation of samples or full fuel assemblies.

The degree of success achieved in fuel performance to date has been remarkable considering the lengthy evaluation process required and the tough conditions the fuel assembly is exposed to in service.

The subsections to follow summarize the major parameters that influence the potential burnup limitations and the current and potential fixes that can extend the limits.

2. INCENTIVES FOR GOING TO HIGH BURNUPS

The list below represents the incentives that existed in the early days of the nuclear industry for operating fuel to high burnups. Most of the incentives are still valid however, the value of and the emphasis on each one is slowly changing with time. The incentives are:

- Economics --- lower fuel cycle costs,
- Capability for longer cycles --- increased capacity factors, decreased radiation doses. The economic gains due to longer cycles facilitated by extended burnups have been taken advantage of to a large degree by the current cycle lengths and burnup levels.
- Improved resource utilization --- decreased amount of uranium, Separative Work Units (SWU) and fuel assemblies per unit energy produced,
- Increased margin to storage capacity limits. However, the inability to send fuel for reprocessing or to a permanent storage site has caused a spent fuel assembly log-jam in the spent fuel pools and effectively eliminated this high burnup incentive,
- Eventual decreased offsite shipping and storage costs. However, the significantly increased time required for high burnup fuel to decrease its decay heat in a spent fuel pool before it can be loaded into an intermediate dry storage cask and the unknown schedule for shipping the fuel from the dry cask to a permanent storage site prevents a reliable estimate for the capacity and cost required for the intermediate wet and dry storage facilities.

Added incentives in European countries (and in the future in Japan) favoured decreased number of fuel assemblies for reprocessing and refabrication due to,

- High back end cost of reprocessing,
- Reality of Pu recycle and its high fabrication cost.

The economic incentives for extending burnup levels will most likely disappear at batch average burnups in the range of 60 to 70 GWD/MT. Economic analyses that represent all costs at conditions prevailing today have not been published and those tend to flatten out at these burnups. Unaccounted costs and uncertainties in the back-end costs will if anything increase fuel costs at burnup levels above these. Since the increase or decrease of fuel costs at these burnup levels are very sensitive to the input, the break-even point for extended burnup requires a plant specific analysis.

In the opinion of this author, based on this and other factors discussed, extension of burnup to levels that require >5% enrichment are highly unlikely.

The reduction in margins to nuclear, thermal and safety analysis limits poses challenges to fuel management methods in order to maintain the desirable as well as the licensing margins. Modified fuel designs and fuel management methods have succeeded to meet the design and licensing limits with 4.95% as well as 5.95% enriched fuel. The major modifications have been the increased amount of burnable absorbers to hold down the increased reactivity, more sophisticated reactivity zoning and nuclear calculations for their accommodation. Other modifications include optimization of the H/U ratios, improved spacer designs and optimization of fuel management methods. Detailed studies of designs >5% enrichment may reach nuclear or thermal limits that will be difficult to maintain by design modifications.

2.1 Potential Failure Mechanism at High Burnups

It appears that increased burnup may reduce the margins towards the following failure modes during normal operation and anticipated operational occurrences:

- Corrosion
- PCI/PCMI
- Dimensional changes

and the corresponding failure modes during Loss of Coolant Accident, LOCA and Reactivity Initiated Accident, RIA:

- "brittle" fuel rods failure during LOCA resulting in "non-coolable fuel geometry"
- fuel dispersal during RIA

The effect of increasing burnup on the above failure modes are discussed in the sequel.

2.1.1 Corrosion

<u>Increased burnup</u> will increase the degree of zirconium alloy material corrosion since higher burnup also <u>means in general longer residence time</u> in the reactor. This holds true for all zirconium alloy material fuel components, Figure 1. However, the development of new PWR alloys have significantly reduced corrosion rates compared to that of Zry-4 and thus increased the margins towards corrosion failures. The improved corrosion performance of new materials will however most likely be used to increase the corrosion duty (higher power, longer residence time and use of different water chemistries) of the fuel thus decreasing the margin to corrosion failures. It is the belief of the authors that corrosion will always limit the burnup.

To reach higher burnups, the enrichment of the fuel must increase and therefore, the fuel rod power over its lifetime will increase. This situation tends to <u>increase the fuel clad temperature</u> that for PWRs will increase the corrosion rate (since the corrosion rate is much less dependent on temperature in BWRs, similar effect will not be seen in BWRs).

Also, higher enrichment fuels in PWRs will require an <u>increase in the LiOH coolant concentration</u>. This since reactivity control in PWRs is to a large extent controlled by the boron concentration in the coolant, and increased fuel reactivity will require an increase in the boron coolant concentration that in turn will require an increase in the LiOH coolant concentration to maintain the optimum pH. The tendency for increased fuel rod power with increased burnup may result in <u>increased tendency for</u>

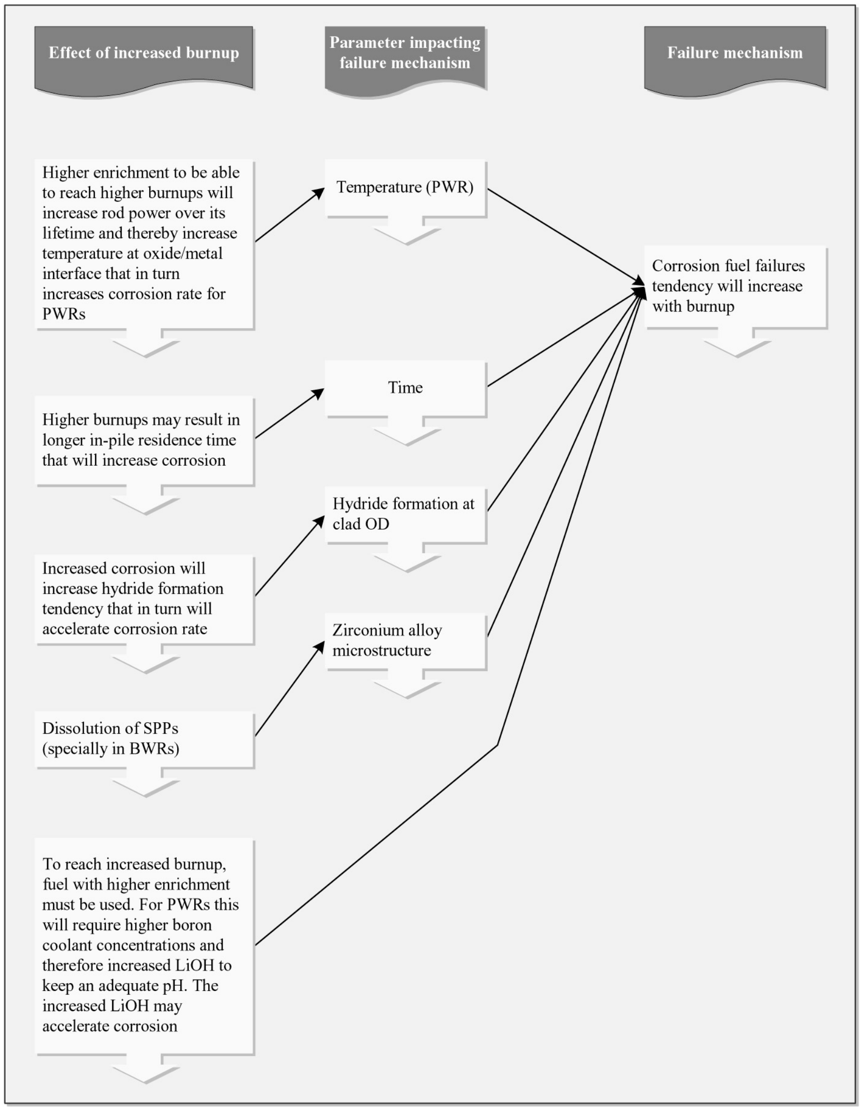

Fig. 1. The Effect of Increased Burnup on Corrosion Related Material Failures. A Downward Pointed Arrow Indicate that this Parameter will Make the Situation Worse for the Cladding by Going to Higher Burnups While a Upward Pointed Arrow Indicate that this Parameter will Improve Performance by Going to Higher Burnups

subcooled boiling in the hottest channels and may together with the increased LiOH coolant content tend to deteriorate the protectiveness of the zirconium oxide layer with accelerated corrosion as a result.

Both in BWRs and PWRs there is a tendency for SPP dissolution with increasing fast fluence (that corresponds to increased burnup). The larger SPPs in Zircaloys for PWR application results in a slower dissolution rate compared to that of Zircaloys for BWRs. When the SPPs have dissolved there is a larger risk that corrosion rate accelerates provided that the coolant chemistry is "aggressive" (what "aggressive" means we do not yet know).

Both in PWRs and BWRs it appears that hydrides at the metal/oxide interface may accelerate corrosion rate, thus with increased burnup, corrosion produced hydrogen absorbed in the zirconium alloy material will increase. This hydrogen may eventually precipitate out as hydrides and as such may accelerate the corrosion rate. Again, the development of new corrosion resistance PWR alloys will also reduce hydrogen pickup (that is the product of corrosion rate and hydrogen pickup fraction) reducing the tendency for hydride driven corrosion acceleration late in life.

There are also some other important water chemistry changes that are not driven by the objective to reach higher burnups but for reasons such as reducing cracking tendencies (NMCA with HWC, HWC, and Zn-injection), and to limit radiation dose (Zn-injection, increased pH in PWRs). In most cases these water chemistry changes results in a more aggressive corrosion environment that may limit fuel burnup.

2.1.2 Hydrides

As mentioned above, increased burnup results in more corrosion produced hydrogen that will be picked up by all zirconium alloy materials such as fuel outer channels (in BWRs), grid sheets, water rods (in BWRs), guide tubes (in PWRs) and fuel rods. Hydrogen in excess of about 100 – 150 wtppm will precipitate out as zirconium hydrides that may embrittle the material to various extent dependent upon not only the hydride concentration but also how the hydrides are distributed and oriented in the material. The newly developed corrosion resistance PWR alloys will pick up less hydrogen and therefore decrease the hydride embrittlement effect, discussed below. Generally the following can be said:

- increased fraction of hydrides will reduce ductility and fracture toughness. It is important to keep in mind that the embrittlement effect of the hydrides is very temperature dependant. This embrittlement effect is far less at 350 ºC than that at e.g. 100ºC.
- nonuniform distribution of hydrides reduces ductility and fracture toughness more than uniformly distributed hydrides (at a constant hydrogen content), e.g., fuel rods with a hydride rim at the clad outer diameter show lower ductility compared to a fuel rod with the same concentration of hydrides but uniformly distributed in the clad thickness. Non-uniform hydride distribution is only found in components subjected to a heat flux. This effect is driven by the thermal gradient and consequently, this effect is only seen in fuel rods where hydrogen in soluble form tends to locate at areas with lower temperatures, such as at the clad OD. With increased surface heat flux, that may be a consequency of the higher reactivity fuel to reach high burnups, the tendency for hydride rim formation will be stronger since the clad thickness thermal gradient will be steeper. Also, oxide spallation (tendency increases with oxide thickness) tends to increase formation of hydride blisters at the clad OD that significantly may decrease ductility and fracture toughness.

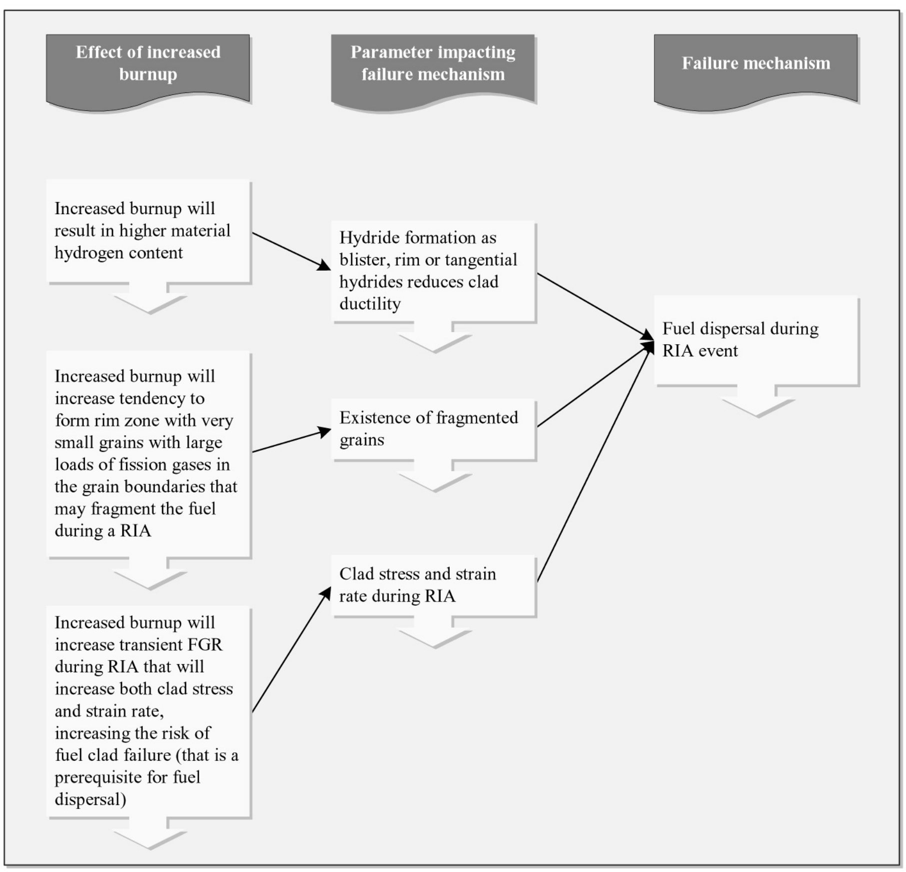

Fig. 2. Effect of Increased Burnup on RIA Fuel Performance

- the fuel cladding will become more embrittled, by formation of hydrides that are oriented perpendicular to the major tensile stress direction. It is e.g. well-known that radially oriented hydrides reduce fuel rod ductility more than tangentially oriented hydrides in a situation where the major stress direction is in the clad hoop direction such as e.g. ramping. Specifically for RXA cladding materials, increased hydride concentration in the cladding increases the tendency to form radial instead of tangential hydrides. SRA material show much less tendency to form radial hydrides, most likely due to that a preferential site for hydride precipitation is the grain boundaries and there are more radial grain boundaries in RXA than in SRA material. <u>Thus, at least for RXA material, increased burnup, i.e., higher hydride concentration increases the fraction of radial hydrides</u>.

The embrittlement effect of hydrides may facilitate fuel component failure such as:

- fuel outer channels and grids during seismic loading
- fuel rods during transport container drop (accident conditions)
- all zirconium alloy material components during impact loading during fuel handling, e.g., if a fuel assembly hits the pool wall during outage handling operation
- fuel rod during PCMI loading either during a class I or II transients or during a RIA event, Figure 2. During class I and II operation the licensing criterion is that the fuel rod must not fail during the transient while limited amount of fuel failures are accepted during RIA. However, a prerequisite for the non-acceptable RIA event of fuel dispersal is that the fuel rod has failed because otherwise the fuel fragments will be contained in the fuel rod. Therefore, one may argue that brittle failure of fuel cladding during RIA may facilitate fuel dispersal.
- Fuel rods during LOCA quenching or post-LOCA events, Figure 3.

Other effects of hydrogen are the following:

- The potential impact of hydrogen in solution and hydrides on creep performance during the LOCA event. Data clearly show that hydrogen/hydrides impacts thermal creep rate of unirradiated materials. It remains to be seen if there is similar impact on irradiated material. If this is the case, the fuel vendor must correctly model this impact in their codes to ensure that the LOCA fuel design criteria are met.
- Both hydrogen in solution and hydrides will expand the material and therefore contribute to dimensional changes (elongation, bowing) of components (e.g. fuel rods, guide tubes, fuel outer channels, grids) that may limit burnup. It should also be pointed out that there may be an interaction between hydrides and irradiation growth that increases growth rate.
- Hydrides at the zirconium oxide/metal interface will increase corrosion rate, see previous section.
- Since hydrogen/hydrides seems to be involved in secondary degradation of failed fuel, increased hydrogen

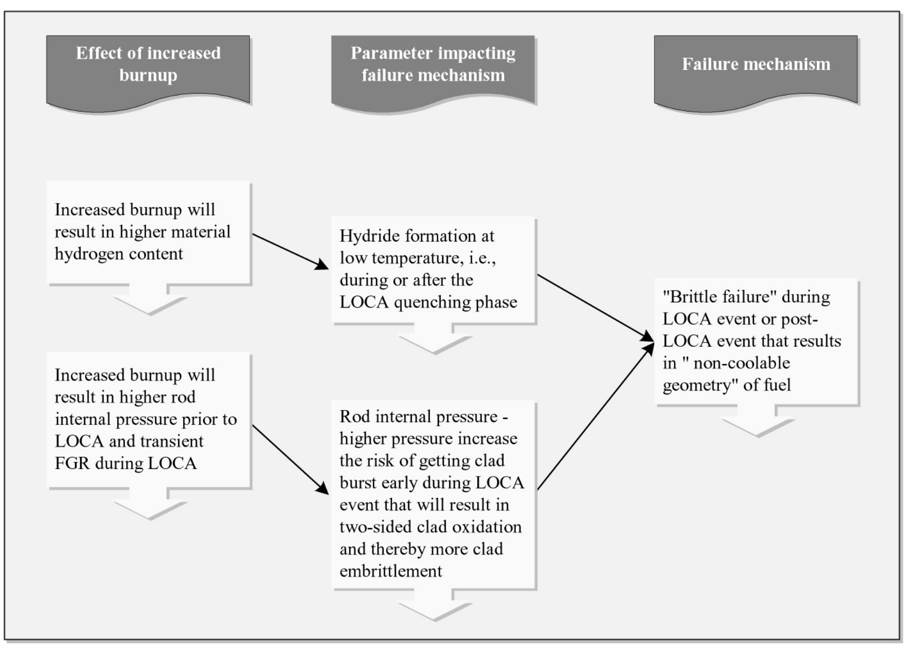

Fig. 3. LOCA Impact of Increased Burnup

/hydride concentration in the fuel cladding with increased burnup may increase the risk of formation of secondary damage resulting in fuel washout with increased burnup.

2.1.3 Dimensional Changes

In addition to hydrogen/hydrides in the zirconium alloy material also irradiation growth and irradiation creep contributes to dimensional changes of fuel components that may limit fuel burnup, Figure 4.

2.1.4 Pellet

If we now turn to the fuel pellet, increased burnup will result in:

- More fission products produced
- Increased Fission Gas Release, FGR
- Increased Transient Fission Gas Release, TFGR
- Formation of a rim zone at the pellet periphery at fuel pellet average burnup of about 50 MWd/kgU
- Fuel-clad bonding at fuel pellet average burnup of about 50 MWd/kgU
- Increased swelling
- Increased transient fission gas swelling

The increased fuel rod internal pressure with burnup (due to more fission products produced and more FGR) may facilitate excessive clad embrittlement during the LOCA clad oxidation phase. The reason being that, clad burst during the LOCA oxidation phase will occur earlier with increased rod internal pressure, thus resulting in two -sided oxidation (and thereby accelerated embrittlement) of the fuel cladding. The increased rod internal pressure may also increase the tendency for fuel clad ballooning and fuel relocation during the LOCA event. The relocation of the fuel into the ballooned area may increase the fuel clad temperature at this location thereby increasing the clad embrittlement effect.

The increased TFGR with burnup will increase the clad strain rate and thereby embrittle the material during a class II transient and a RIA event PCMI loading. For all materials, higher strain rate promotes brittle failures since there is less time for dislocation slip and twinning that are the deformation modes during plastic deformation.

For high burnup fuel, the development of the fuel rim

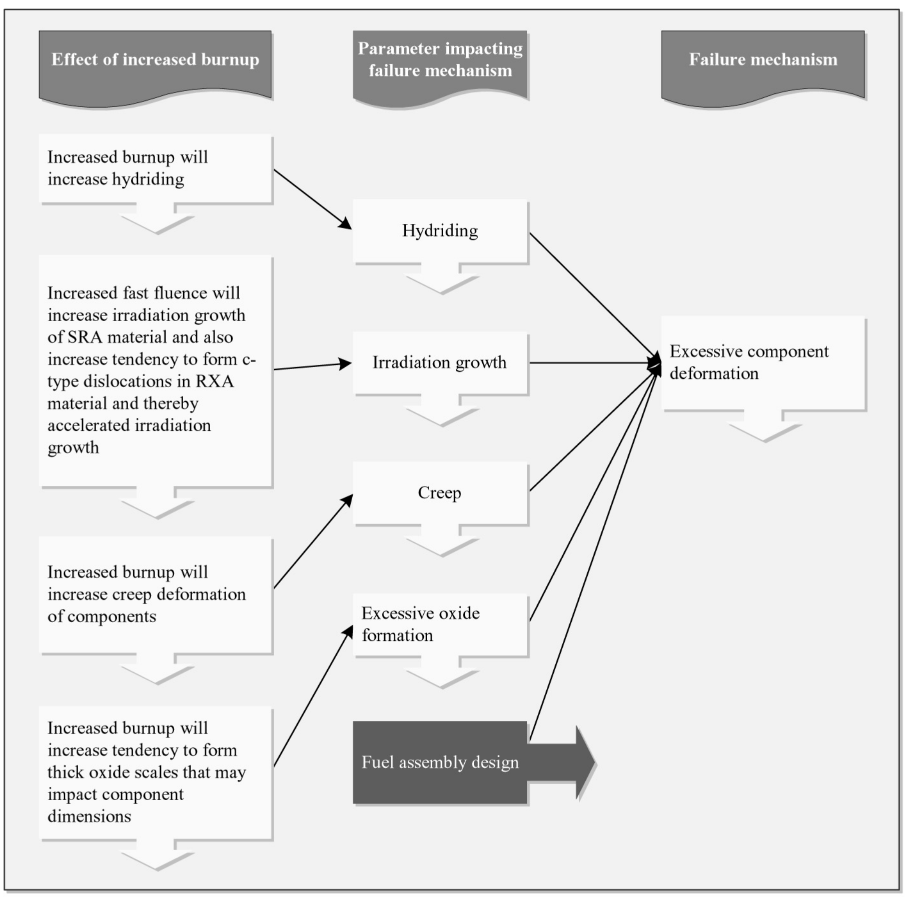

Fig. 4. Increased Burnup Effect on Dimensional Instability

zone, that consists of very small grains and large loads of fission gases in the grain boundaries, may increase the risk of fuel dispersal (provided that the fuel cladding fails) during the RIA event. For high burnup materials, most of the fissile material is in the fuel pellet periphery and consequently this zone will experience the largest temperature increase during the RIA event. The fission gases in the grain boundaries will expand when it is heated, during the RIA pulse, tending to separate the grains. If the temperature excursion at the clad periphery is more moderate, such as e.g. during a class II transient, the TFGR may be negligible and the large porosity in the rim zone may actually decrease the PCMI/PCI loading thus increasing the failure threshold.

It is not clear if and in such a case how the fuel clad bonding will impact tendency to fuel failures.

With increased burnup, more fission products such as Cd and Cs are produced and transient fission gas swelling will result in increased cladding strains during a power transient. Thus the PCI threshold will most likely decrease with increased burnup.

Figure 5 shows the potential impact of increasing burnup on PCI/PCMI failure tendency. It is believed that the failure tendency may increase with increased burnup due to the embrittlement of the cladding. However, at the same time one has to keep in mind that the reactivity of the fuel decreases with burnup and thereby there is a decrease in failure tendency with increased burnup. For example, if a control rod in a BWR is pulled adjacent to a fresh fuel assembly the power increase will be much higher compared to a similar situation but adjacent to an old assembly with low reactivity.

3. SUMMARY

This paper has reviewed the different potential fuel failure causes related to increased burnup. The limits to material and component performance as a function of extended burnup are in approximate decreasing order of potential, current challenge:

- *Corrosion* of zirconium alloy cladding and the water chemistry parameters that enhance corrosion,
- *Dimensional changes* of zirconium alloy components,

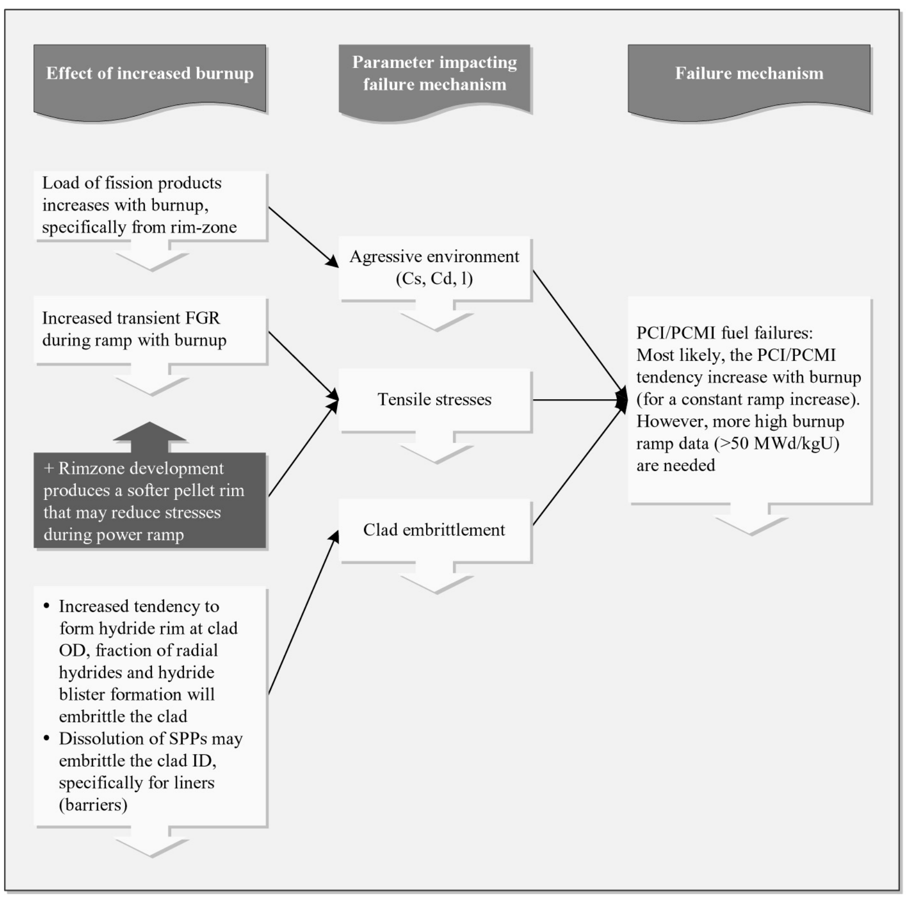

Fig. 5. Schematic View of the Effect of Increased Burnup on the PCI/PCMI Failure Tendency

- Stresses that challenge zirconium alloy *ductility* and the effect of hydrogen (H) pickup and redistribution as it affects ductility,
- Fuel rod internal pressure,
- Pellet-cladding interactions *(PCI)* and pellet-cladding mechanical interactions *(PCMI)*,

The list has not changed significantly in over a decade, [1]. The only items above that have posed limits to extending burnups have been *corrosion* and *dimensional changes* in both BWRs and PWRs and *PCI* in BWRs. Improved materials and operating procedures have been able to exceed all of these limits and have not reached new limits within current operating strategies.

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