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J. SPINO
CEC Institute for Transuranium Elements,
Karlsruhe, Germany

Abstract

This paper presents a review of different behaviours of fuel elements in relation to fission gases. The influence of the fission gas level and its location regarding different fuel structures is commented. The gas release process at high burnup is described and some topics that need to be further investigated are suggested.

The behaviour of the fission gases in water reactor fuels has been a matter of concern since more than 30 years, and even today questions arise especially when burnups above 4-5 irradiation cycles are envisaged. In that case, more pronounced effects on fuel behaviour are expected, either if these gases are released to the rod free-volume, or if they remain trapped in the fuel matrix. In the first case, besides the drop of the gap-conductance due to the low thermal conductivities of Xe and Kr, an excessive pressure build-up can occur at high release rates, leading to undesired cladding lift-off and gap reopening effects at end of life. On the other hand, occluded gases in the fuel voids induce matrix swelling, enhancing the pellet-cladding mechanical interactions and the risks of cladding failures by stress corrosion cracking. Given the above set of interrelated effects, the design challenge is to achieve the highest possible retention of the fission gases in the fuel matrix, compatible with the lowest fuel swelling and the lowest probability of cladding failures.

In different international meetings on water reactor fuels performance during the last years it has been shown that up to burnups of around 40 GWd/tHM and under normal reactor operation conditions, the integral fission gas release for PWR fuels can be maintained under the acceptable level of about 1 % of the inventory [1-2]. In this respect, it is interesting to note that with the introduction of improved fuel element designs and with appropriated operational measures, both allowing a more homogeneous power distribution in the fuel rods, a reduced integral gas release can be achieved not only under steady state operation [1-2], but also under load following conditions [2]. In the burnup range described (i.e. < 40 GWd/tHM), if temperature effects are moderate, the integral gas release can be almost entirely described by athermal processes, namely knock-out and recoil, showing a slight dependence with burnup and the specific area of the fuel [2-3].

Differently to the above, on surpassing burnups of about 40 GWd/tHM a steady increase of the gas release rate towards values of several percents at about 70 GWd/tHM has been reported by different fuel vendors and utilities [1-5]. In this burnup range, also the onset and growth of the so-called 'rim structure', i.e. the porous and submicron grained structure formed at the pellet edge due to the fissioning of converted Pu$^{239}$, has been observed [3-6]. In this region, almost the totality of Xe is depleted from the fuel matrix and contained in pressurized pores [6-7]. In some appreciations, venting of these pores through cold creep and microcracking has been postulated as a possible cause for the overall release enhancement at high burnups [8-9]. However, analysis of the gas retained at different radial positions demonstrated that the majority of the gas which is released at these burnups stems from the central regions of the pellets [10]. Accordingly, the widest consensus today is that under normal irradiation conditions the depleted gas at the fuel periphery would remain trapped in the pores, being the main contribution of the rim zone to elevate the central fuel temperatures due to the additional thermal barrier created in the gap [4-6,10-12]. The thermomechanical behaviour of this zone under severe transient conditions (i.e. RIA) is still not completely understood.
An apparent change of the gas release processes at high burnups becomes evident from the plot of the centerline temperatures for 1% integral release versus the peak pellet burnup, when it is compared with the well established Vitanza or Halden threshold [10,13,14]. The Vitanza threshold, denoting the fuel centerline temperatures at which saturation of grain boundaries is achieved, shows an asymptotic trend to temperatures of 1000-1100 °C at burnups above 30 GWd/tHM [13]. On the contrary, the derived values for high burnup fuels indicate continuous decrease of these temperatures up to values of about 600 °C at 55 GWd/tHM [10,13]. This trend splitting may be due to two reasons, namely to an enhanced intragranular diffusion at high burnups promoting saturation of the grain boundaries at lower temperatures, or to the underestimation of the centerline temperatures due to overvaluation of the fuel thermal conductivity [13]. Due to the experimental evidences of a high temperature regime even after the 4th cycle, the second explanation appears to be the most plausible one [13]. In that case, doing the proper corrections to the thermal conductivity, the Vitanza threshold appears to be still applicable at extended burnups.

Considering the mitigation of gas release through fuel material improvement, practically the unique measure explored up to now has been the grain size enlargement. Larger grain sizes would retard gas atom diffusion to the grain boundaries, and would show also smaller swelling [15]. Various types of large grained fuels have been tested in-pile, namely undoped urania [16], UC doped with metal oxides (NbO2, TiO2, Cr2O3, La2O3) [16-18] and UC doped with Al and Ti-silicates [16]. Under steady state operation, both gas release and swelling were improved in all cases. However, under simulated power transient conditions after a burnup of ~25 GWd/tHM, only undoped and silicate doped large grained fuels showed better behaviour than standard UC. Contrarily, the NbO2 and TiO2 doped fuels showed a high gas release under transient heating [16]. Since doping of large grained UC is necessary to counteract its otherwise worse creep and PCI behaviour, the above results suggest a more preferable use of silicates as plasticizers (grain boundary sliding) than of dissolved niobia and titania. However, in-reactor ramp testing of silicate doped fuels is needed to prove their actual effectiveness as PCI remedy, as it was the case of niobia, in spite of the experienced large bubble swelling and increased cladding ridging [19].

Less explored up to now has been the possibility of increasing the matrix gas retention by pinning of gas bubbles at finely dispersed precipitates. Hindering the bubble growth through anchorage at preferential sites may result not only in less swelling [20], but also in a better thermal conductivity through a smaller amount of large matrix voids [21]. Avoidance of fuel hyperstoichiometry is also a point which must be attended at high burnups in order to impede gas release enhancement [22] and thermal conductivity deterioration [21]. From the point of view of pellet design, use of the alternative anular pellets, diminishing gas release and PCI effects [23-24], might be considered.

Utilization of gadolinia as burnable neutron absorber is at present a standard design feature in PWRs and BWRs to achieve more flexible core management [25]. Up to 10 wt. % Gd2O3 in UC fuels has been normally used, with no detrimental effects in the fission gas release and swelling behaviour being reported [26]. However, due to the lower thermal conductivity of the gadolinia bearing fuels, higher operation temperatures and subsequently larger gas release of these rods are expected [16].

Burning of Pu as mixed oxide (MOX) fuel in water reactors is today an alternative not only for spent fuel recycling but also for the dispositioning of weapons fissile material. MOX fuels behave similarly to UC fuels [27-30], with even superior resistance to PCI failures [29], but with the drawback of an apparently higher gas release [28]. However, this is due to the higher power rating of the MOX fuels, since when plotted as function of the linear power the integral gas release of MOX is similar to that of UC [28,30]. Since in heterogeneous MOX fuels most of the fission gases are produced within the dispersed Pu-rich agglomerates, gas release to the rod free volume may occur only if enough channels (grain boundary porosity) are available in the matrix [31]. With increasing
burnup the amount of Xe migrated into the matrix increases significantly, being released after a certain time according to thermally activated processes [32]. In this sense, it is noteworthy that the onset of gas release for solid and hollow MOX fuels has been appropriately predicted by application of the Vitanza threshold for UO$_2$ [30].

*From the above exposed it appears that the fission gas release behaviour is in general terms well understood and controlled up to average burnups of about 40 GWd/tM, whereas for extending burnups beyond this level some further investigations seem to be needed in topics like*

- Diffusion coefficient of Xe in heavily irradiated UO$_2$ matrix with large amounts of lattice defects and high fission products concentrations (precipitated and dissolved);

- Elucidation of the true capability of the rim zone to retain fission gases within its closed porosity. Thermal-mechanical stability of the rim structure under rapid power transients (RIA);

- Intrgranular fission gas retention at preferential anchorage sites (i.e. finely dispersed precipitates);

- Gas retention enhancement vs. worsening of mechanical properties in large grained fuels. Matrix dissolved vs. grain boundary segregated plasticizers;

- Heterogeneous vs. homogeneous Pu-distribution in MOX fuels.

*FIG. 1. PWR fission gas release as function of the rod average burnup (Ref. 2).*
FIG. 2. Integral fission gas release from PWR rods under steady state and load following conditions (Ref. 1).

FIG. 3. Threshold temperature for 1% integral gas release as function of the peak pellet burnup (Refs. 5 and 13).
FIG. 4. Fission gas release of standard and non-additive large grained $UO_2$ fuels, (BWR, type 9x9) Ref. 16).

FIG. 5. Burst release rate vs. grain size in post-irradiation annealing for doped and updoped $UO_2$ fuels (Ref. 16).
FIG. 6. Bubble swelling vs. grain size in post-irradiation annealing for doped and undoped UO$_2$ fuels. (Ref. 16)

FIG. 7. SEM micrographs of post-irradiation annealed fuel samples.  
(a) Standard UO$_2$  
(b) Undoped large grained UO$_2$  
(c) TiO$_2$-doped UO$_2$  
(d) Bentonite-doped UO$_2$
FIG. 8. Fission gas release of PWR - MOX and UO₂ fuels as function of the rod average burnup (Ref. 32).

FIG. 9. Fission gas release of PWR - MOX and UO₂ fuels as function of the linear hear rate (LHR) (Ref. 30).
FIG. 10. EMPA diamteral scan of Xe in the UO$_2$ matrix of MOX fuels after the first, second and third irradiation cycles (Ref. 32).
REFERENCES


[12] KJAER-PEDERSEN, N., "Rim effect observations from the third Risø fission gas project", ibid p. 111.


