BNFL ASSESSMENT OF METHODS OF ATTAINING HIGH BURNUP MOX FUEL

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Abstract

It is clear that in order to maintain competitiveness with UO_2 fuel, the burnups achievable in MOX fuel must be enhanced beyond the levels attainable today. There are two aspects which require attention when studying methods of increased burnups - cladding integrity and fuel performance.

Current irradiation experience indicates that one of the main performance issues for MOX fuel is fission gas retention. MOX, with its lower thermal conductivity, runs at higher temperatures than UO_2 fuel; this can result in enhanced fission gas release. This paper explores methods of effectively reducing gas release and thereby improving MOX burnup potential.

1. INTRODUCTION

In opting to displace a fraction of its UO_2 fuel requirements with MOX fuel, an utility would expect the MOX fuel to attain the same discharge burnups as its UO_2 fuel. If the MOX fuel were to be discharged at a lower burnup, this would imply an increased fuel procurement requirement and hence increased fuel cycle costs. Thus, the very minimum requirement for MOX burnup is that it should at least match that of the coresident UO_2 fuel. There are, however, significant differences in the underlying economic drivers.

For normal UO_2 fuel, the fuel cycle economics generally improves with increasing burnups, but has a tendency to level out or even deteriorate a little beyond 55 GWd/t. This levelling results from a balance between the increasing uranium ore and enrichment costs needed to attain high burnups and the extra electrical generation from each fuel assembly during its lifetime; the two effects are roughly compensating at burnups of 55 GWd/t or more. Therefore, for UO_2 fuel, there is little incentive from the point of view of fuel cycle economics to increase burnups much beyond these levels, although there may be other important drivers, such as minimising spent fuel quantities that may influence an utility's decision to push for higher burnups.

The burnup dependence of the fuel cycle economics is quite different for the MOX component of a core, because fuel fabrication is the main cost component and it does not vary significantly with burnup. Thus the total cost of a MOX fuel assembly is essentially constant with burnup, whereas the electricity it generates increases proportionally with burnup. Simplistically, the fuel cycle cost of the MOX component is therefore inversely proportional

to burnup, which gives an incentive to increase burnups beyond that where the UO_2 levels out. In practice, discounting effects somewhat offset the improvement with burnup (since the electricity is generated over an extended period of time its discounted value is lower than it would be if it was generated instantaneously), but there still remains a significant benefit to be gained.

This paper briefly reviews some of the technical issues and possible solutions associated with extending MOX discharge burnups. While the focus is on fuel technology, it is recognised that any developments in this area must be matched by equivalent developments in cladding performance, particularly with regard to corrosion resistance; a detailed discussion of this area however lies beyond the scope of this paper.

2. FACTORS INHIBITING HIGH BURNUP

For all fuel types, including conventional UO_2 fuels, a number of technical factors combine to inhibit the trend toward the target of higher discharge burnup. Principal amongst these are (a) thermal performance, (b) accelerating fission gas release, and (c) increasing cladding waterside corrosion. For a given discharge burnup, all are somewhat more limiting for MOX than UO_2 because of differences in power histories.

MOX tends to have a flatter reactivity variation with burnup than UO_2 because of the lower burnout rates of the fissile nuclides in the lower thermal flux and because of the effect of Pu-240 acting as a fertile material by being converted to Pu-241 by neutron captures. An important effect of the different reactivity characteristics is that MOX assemblies, all other things equal, tend to run at a higher rating than equivalent UO_2 assemblies late in their lives. Both clad corrosion and fission gas release are affected particularly by high ratings late in life and this may lead to the MOX fuel being more limited in both respects at any given burnup.

With MOX, various inherent characteristics of the fuel tend to enhance the factors which inhibit high burnup. For example, the lower thermal conductivity, smaller grain sizes and higher late-in-life powers associated with MOX fuel all exaggerate the problems of fission gas release, clad corrosion and reduced thermal margins. The challenge for the fuel vendor is the development of advanced MOX fuels capable of overcoming these difficulties and achieving the levels of burnup which optimise fuel cycle economics.

3. THERMAL PERFORMANCE

As burnup progresses both the thermal conductivity and melting point of fuel are reduced. Although the rate of degradation is not significantly different with MOX fuel compared with UO_2 fuel, the initial level is in each case lower. Fission gas release can also become significant at high burnup, leading to a reduction of fuel-clad gap conductance. For the reasons discussed below, gas release tends to be more pronounced in MOX fuels, enhancing this reduction. The combined effect of these factors on the high burnup thermal performance of MOX fuel is two-fold. Firstly, steady state temperature levels are increased, particularly since the late-in-life rating level in MOX assemblies can be high. These higher temperatures can lead to further fission gas release and swelling. Secondly, the margin to fuel melting under fault and accident conditions is decreased.

In targeting possible solutions to the problem of high temperatures, two basic strategies can be considered:

- Changes in fuel rod design
- Changes affecting fuel physical properties

The former category includes the use of smaller diameter rods and also the use of annular pellets. Both of these options are already available with existing technology and to some extent are already partially proven. Both could be brought into use with relative ease, although each would result in some degree of fabrication cost penalty. The advantage of hollow fuel is also doubtful under circumstances where gas diffusion can be sufficiently rapid to induce swelling of the unrestrained central portions of the annular pellet.

In the latter category come developments which could lead to an improvement of fuel thermal conductivity. (Other developments aimed specifically at improving fission gas retention also provide an indirect benefit, but these are discussed separately below.) One way to increase the thermal conductivity of fuel is to reduce its porosity content; two options are available for this. The population of fine (sub-micron and micron sized pores) is determined by sintering conditions. Various techniques are available, including longer sintering times, modified sintering atmospheres, and the use of active powders and dopants, which could reduce or largely eliminate the residual fine porosity. The benefits of this, however, are unlikely to be significant, since this population of pores is in any case quickly eliminated by irradiation.

A potentially more fruitful approach to producing high density fuel is via the elimination of the more stable larger pore population. In many fuel manufacturing routes the characteristics of the large pore population are controlled by the use of pore forming agents, in BNFL's case this being CONPOR. Complete elimination of the pore former can be used to produce fuels up to 98% dense. UO_2 fuel of this density has been irradiated without problems for many years in the UK's gas-cooled reactors, although use in MOX fuel and at high burnup remains untested. Moreover, the role of the large pores in accommodating fuel swelling and creep during normal and off normal operation, while not fully understood and quantified, may be significant. The complete elimination of the large pore population may therefore be too radical a step, and more value may come from research into adaptation of pore forming agents to produce fuels with a higher density but which still demonstrate satisfactory swelling accommodation.

The other major option for increasing the thermal conductivity of fuel is via the introduction of a second, higher conductivity, material. This could be an oxide, for example some recent UO_2 experiments have studied beryllium oxide (BeO)[1]. This has the highest conductivity of any oxide, a high melting point, and low neutron absorption, although its toxicity presents handling difficulties. By sintering above the eutectic temperature of UO_2 -BeO, BeO can be precipitated almost continuously along the grain boundaries, yielding conductivity improvements of the order of 25%. The other candidate for a high conductivity second material is a metal, in other words producing a cermet fuel. Cermets have been studied extensively in the past, with molybdenum generally considered to be the most promising metal. Recent work with UO_2 has shown that it is possible to fabricate pellets in which high density fuel agglomerates are enclosed within a continuous, interconnected metal matrix using as little as 5% molybdenum content.

The in-reactor performance of such advanced fuels is largely untested. The impact on MOX fuel manufacturing routes is also unclear. Nevertheless, should significant improvement of fuel thermal conductivity be deemed imperative for the accomplishment of high MOX fuel burnups, then some promising development paths would appear to be available.

Further, more radical development possibilities could combine both pellet design changes and materials property changes. In particular, the use of duplex pellets would seem to offer potential advantages, although again with some likely cost penalty. Duplex fuel designs have been used in a number of trial irradiations in the past, but have been limited to studies of the effects of dual enrichments. In principle, the two pellet regions could be designed not only with different enrichments, but also with different microstructural characteristics. The optimum microstructure for fuel positioned in the hotter central regions of a pellet are not the same as those for fuel near the pellet periphery. This additional degree of freedom therefore offers scope for development of fuels with improved thermal performance, as well as other advantageous, characteristics.

4. FISSION GAS RELEASE

Fission product release, and associated gas bubble swelling, is potentially burnup limiting in all fuel types. With MOX fuel, the inherently lower thermal conductivity, higher late-in-life powers, generally smaller grain sizes and, possibly, inhomogeneity of the pellet structure would all point to higher gas release levels compared with standard fuel. Mitigating the effects of these four factors is therefore important to the achievement of high MOX burnup. The first two of these factors have already been discussed elsewhere in this paper, the latter two are considered below. While the focus here is on potential solutions to the underlying problems, it is clear that other more straightforward design changes, such as introducing larger or cooler plenum volumes, or using annular fuel, can also alleviate the problems of gas release and might also have a role to play in any high burnup MOX fuel design.

The present generation of MOX fuels have mean grain sizes that are typically a factor of two to three times smaller than present UO_2 fuels. Although, to first order, grain size has no effect on pre-interlinkage gas release rates, nor on the incubation burnup necessary to achieve interlinkage, there is a strong inverse correlation between gas release and grain size in the post-interlinkage, thermal release regime that is most significant, and potentially life limiting, at high burnup. Increasing fuel grain size is therefore a key to unlocking the high burnup potential of MOX fuel.

High furnace temperature and/or long sintering times are the most orthodox means to fabricate large grained material. However, very high temperatures and/or very long times are likely to be required to achieve significant improvements in the grain size of MOX fuel. Alternative sintering techniques are therefore likely to be necessary. One such approach is the use of sintering atmospheres with higher oxygen potential produced using either CO_2 or water vapour in hydrogen. A second approach involves the use of active powders. This technology has been studied in the context of high burnup UO_2 fuels [2], but application to mixed oxides may be less successful due to differences between the UO_2 and PuO_2 powders. A third approach involves seeding. Seed crystals of UO_2 (from recycled sintered scrap material), introduced at the powder blending stage, grow preferentially during sintering, leading to a

large grained material. A separate BNFL paper at this meeting describes the technique in more detail [3]. All of these approaches may offer some potential for achieving large grain size MOX fuel.

The use of dopants is an alternative approach to producing large grained material. Many dopants, including niobia, chromia, titania and magnesia, have been shown to readily produce large grain UO_2 fuels. However, the irradiation experience shows that the benefits of large grains are at least partially offset by the associated increase in fission product diffusion rates, resulting in only modest benefits in gas retention. Work on UO_2 with this class of dopants is still being pursued, e.g. [4], although within BNFL [5] the focus is on achieving improved PCI resistance (which is less of an issue with MOX fuel) rather than fission gas retention. A small amount of work has been published on doped MOX fuels, but this remains a fruitful area of research.

A second class of fuel dopants are those which form a liquid phase during sintering, leading to enhanced grain boundary diffusion. Such dopants are capable of producing large grain fuel but without the drawback of enhanced matrix diffusivity. A recent study has confirmed the satisfactory performance of alumino-silicate doped UO_2 in irradiation trials to 60 MWd/kgHM burnup. Application of this technology to MOX fuel may be worth pursuing.

It is well established that mixed oxide fuels with poor plutonium homogeneity lead to a marked enhancement of fission gas release for a broad range of irradiation conditions. On the other hand more uniform dispersal of the plutonium in solid solution can lead to a small reduction of thermal conductivity. BNFL's Short Binderless Route (SBR) produces a MOX fuel microstructure which meets the optimum for current fuel duties. BNFL is involved in the development of fuels targeted at higher burnup duties where fission product retention is paramount. Adaptation of the SBR to meet these requirements is considered to be relatively straightforward.

5. OTHER CONSIDERATIONS

Fuel Cladding

Current cladding technology is only demonstrated for burnups up to approximately 60 GWd/t. For higher burnups new cladding types will be required, irrespective of whether the fuel is UO_2 or MOX. The difference in fission gas release, although significant, is modest enough that relatively small changes in fuel design (such as a larger plenum volume or optimised grain size) are sufficient to accommodate it.

Power Histories

One possible approach to improving the high burnup potential of MOX fuel involves optimisation of core design schemes to give lower ratings in MOX assemblies late during their lifetime, while compensating early in life with higher ratings. What is important here is not so much the average rating that a MOX assembly is subjected to, but that of the peak rating locations. This is because the fission gas release is nonlinearly dependent on rating and the peak locations therefore make the biggest contribution to the overall plenum inventory. There is the possibility of optimising fuel shuffling patterns with respect to power histories, using

automated loading pattern search methods. Although the field of loading pattern optimisation is a very active research area at the moment, little work has been carried out to optimise in this respect; normally loading patterns are optimised with respect to single parameters such as reactivity coefficients, cycle length or feed enrichment. Optimising on power histories is a more difficult task, but one which is now within reach of the latest optimisation codes.

The use of burnable poisons has a limited role to play in achieving high MOX burnups. They are essential to hold down the excess reactivity early in life of the fuel, however they have little role beyond the first cycle of irradiation, as they are designed to be completely burnt out by then. Nevertheless, without burnable poisons, high burnup fuel management schemes would be impractical to achieve.

6. CONCLUSIONS

It is believed that BNFL's SBR MOX has the capability of exceeding the current burnup constraints on MOX fuels; however, the current generation of fuels have not been optimised for very high burnup operation, > 60 GWd/t. Assuming fuel developments can be matched by appropriate advances in cladding technology, a number of avenues are available which can be pursued in the quest for higher burnup, many of which may, in principle, be easily incorporated in the SBR. These range from straightforward design changes to more radical developments:

- Smaller diameter rods, larger and cooler plenum volumes and annular pellets;
- Core design optimisation with respect to power histories
- Fuel microstructures specifically optimised for high burnup duty with respect to plutonium homogeneity and pore and grain size distributions
- Use of dopants
- Use of duplex pellet designs
- Use of oxide or metal additives to increase thermal conductivity.

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