High Burn-up Structure in Nuclear Fuel: Impact on Fuel Behavior

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When UO_2 and $(\text{U,Pu})\text{O}_2$ fuels locally reach high burn-up, a major change in the microstructure takes place. The initial grains are replaced by thousands of much smaller grains, fission gases form micrometric bubbles and metallic fission products form precipitates. This occurs typically at the rim of the pellets and in heterogeneous MOX fuel Pu rich agglomerates. The high burn-up at the rim of the pellets is due to a high capture of epithermal neutrons by ^{238}U leading locally to a higher concentration of fissile Pu than in the rest of the pellet. In the heterogeneous MOX fuels, this rim effect is also active, but most of the high burn-up structure (HBS) formation is linked to the high local concentration of fissile Pu in the Pu agglomerates. This Pu distribution leads to sharp borders between HBS and non-HBS areas (Fig.1).

Fig. 1: Sharp limit between HBS and non-HBS areas in a 55 GWd/t_{HM} MOX MIMAS fuel (from [1]).

In these MOX fuels, the HBS forming at various radial positions, and not only at the rim, it was shown that the size of the new grains, of the bubbles and of the precipitates increase with the irradiation local temperatures. Other parameters have been shown to have an influence on the HBS initiation threshold, such as the irradiation density rate, the fuel composition with an effect of the Pu presence, but also of the Gd concentration in poisoned fuels, some of the studied additives, like Cr, and, maybe some of the impurities. However, not all the differences in the UO_2 HBS rim extent measured by different teams on various fuels have been explained [2-3]. The effect of impurities may be the main reason for these differences, but it has not been documented enough yet. It has been shown recently, with examinations of a UO_2 fuel in which ^{235}U was heterogeneously distributed, that a high Pu concentration is not mandatory for HBS formation [4].

Several changes in the fuel behavior occur concomitantly with the HBS formation. An increase of the fission gas release and an increase in the fuel swelling rate are measured [3]. It was shown by indirect and direct approaches that HBS formation was not the main contributor to the increase of fission gas release at high burn-up [1, 5-6]. Indeed, studying the Kr/Xe ratio or some isotopic ratio in the gases collected during rod puncturing and using the differences in the gas productions as a function of the radial position, it was shown that the HBS areas were not the main source of the released gases. SIMS measurements of the local retention confirmed this trend. Nonetheless, the formation of a strong bonding between the fuel pellet periphery and the inner surface of the cladding, with the formation of the inner zirconia layer induces tensile stresses in the fuel during power

decrease periods. This leads to radial cracking of the rim and, consequently, to some fission gas release. The amount of gases released by this mechanism never was evaluated. HBS formation does participate to fuel swelling increase, but it is not the only phenomenon involved, other areas showing also gas bubble formation. It must be noted that all porosity observed is not new, a local decrease of the large fabrication pores being measured. The pressure in the HBS bubbles has been evaluated. At 650 K, it is about 78 MPa, and decreases with the increasing burn-up. This corresponds to an average atomic volume of $\sim 150 \text{ \AA}^3$, i.e. more than three times the UO_2 trivacancy volume, hence the increase in the swelling [1].

Impact of HBS on the fuel behavior during ramp on high burn-up fuels is unclear. The generalized fuel to cladding contact prior to the ramp is certainly the major parameter. HBS may play a role, but no direct evidence of that was found, though, prior to the ramps, high strain of the HBS can be seen in the large fabrication pores decrease as well as on free surface HBS major swelling. In loss of coolant accident (LOCA) type conditions, out of pile heating tests on fuel sections as well on specially designed experimental discs have shown, like during in-pile LOCA tests, the fragmentation of HBS [7-8]. The areas prone to fragmentation are the HBS areas but also the high precipitation areas [9]. The particular power history of the heterogeneous UO_2 , with an increase in the linear powers at high burn-up, led to a generalized fission gas release of the HBS spot bubbles. In addition, a generalized opening of grain boundaries is evidenced by Cs departures and deposits [4].

It could therefore be interesting to design a fuel that would be particularly resistant to HBS formation and to gas precipitation into large bubbles at high temperature.

A large grain UO_2 fabricated by NFD (Japan), with a long oxidizing sintering, was shown to be locally highly resistant both to bubble formation at high temperature and to HBS [10]. Expanding this to the whole fuel is an appealing idea.

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HIGH BURN-UP STRUCTURE IN NUCLEAR FUEL: IMPACT ON FUEL BEHAVIOR

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73 GWd/t_U

why at the rim?

Pu diametral profile in UO₂ fuel at 50 GWd/t_U

↗ Pu production

→ ↗ density fissiles atoms

→ ↗ local power

→ ↗ local burnup and dpa

> 1000 dpa

HBS or rim effect

Capture of n by ²³⁸U

→ Pu build-up in UO₂

Resonance peaks for epithermal neutrons

→ much higher Pu build-up at pellet periphery (x 2 to 3)

In a heterogeneous MOX fuel

Very high BU spots → HBS formation, not only at the rim

(JNM 2008)

In a heterogeneous MOX fuel

(JNM 2008)

grain, bubble and metallic precipitates size increase with the irradiation temperatures

Periphery

centre

EPMA Mo maps

influence on the HBS formation

- fission density rate
- fuel composition (Pu, Gd)
- dopants (Cr)
- impurities?

Cr_2O_3 doped UO_2 62 GWd/t_U at 500 μm from pellet's periphery

(D. Baron NET 2009)

High Pu is not mandatory for HBS

Heterogeneous UO_2 case in Halden reactor

SIMS ^{235}U map

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Concomitant phenomena

■ increase in the fission gas release rate

rod puncturing

(NET 2009 + OECD proc cad 2000)

Increase in the fission gas release rate

HBS not the major contributor :

- analyses of the released gas compositions
- direct retention measurements (SIMS +EPMA)
- HBS fully transformed discs Halden IFA 649 for NFIR

(OECD proc cad 2000)

+ (Itagaki 1996)
+ (Lemoine 2009 –MOX)

(JNM 2008)

(JNM 2014)

NFIR discs

Standard UO_2 ,
103 GWd/t_U
Average fission gas
release close to 2.9%
of the produced gases
for 100% HBS discs

Increase in the fission gas release rate

Yet, cracks in the rim → gas release

(NET 2009)

Internal zirconia
formation → strong
mechanical link with the
clad

Power decrease periods
→ fuel cracking

83 GWd/t_U

■ increase in the fuel swelling

UO₂ Fuel swelling in a PWR

(NET 2009)

rod mean diameter in span 5. M5® Claddings
(Zirconia formation corrected)

■ HBS only partly responsible for swelling increase

83 GWd/t_U

+ Gas content in the HBS bubbles :

In lost of coolant situations

72 GWd/t_U

NFIR
Fully HBS discs
ramped at
1200°C
fragmentation

SEM images : Fuel broken into small fragments
(JNM 2014)

Fragments surface similar to
intentionally broken HBS

High BU + heterogeneous + ramp

Power increase on the high BU heterogeneous UO_2

— EPMA Xe

• SIMS Total

■ SIMS Base Line

(JNM 2013)

a)

b)

c)

d)

e)

Can HBS be avoided in UO_2 fuels?

MINOS NFIR

Large grain UO_2
fabricated without
dopant by NFD
(Japan)

At 96 GWd/t_{HM}
Locally, very high resistance to HBS formation

(JNM 2015)

- A research started a few decades ago
- Still progressing
- With good hopes on
  - o the new SEM, + EBSD+FIB+STEM+EDS
  - o the coming TEM
  - o MARS beam line at SOLEIL