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Fuel (UO_2) R&D: Needs, Approaches, and Results

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Abstract – *Various R&D needs related to fuel pellet material (UO_2) are being addressed through international collaborations. One proven approach in this area has been nearly isothermal irradiation of samples of UO_2 variants in a test reactor, followed by post-irradiation examinations (PIE) and measurements. The results and data are subsequently utilized in fuel performance and design considerations of commercial LWRs. Since the 1989, five such irradiation campaigns have been completed in the Halden test reactor in specially designed instrumented fuel assemblies (IFAs). Although there were variations in the detailed designs of each of these IFAs, the basic approach of isothermal irradiations under controlled conditions has remained the same. These IFAs have yielded valuable in-pile fuel behavior data as well as pre-characterized high burn-up PIE samples. Several completed and on-going projects will be described in the context of these IFA irradiations and the R&D needs they are addressing. The paper includes experimental details, fuel temperature calculations, key observations, and relevant key PIE results obtained from the samples from these IFA irradiations in collaborative programs such as HBRP, NFIR and HPG. Finally, an outlook is provided on future fuels R&D needs.*

I. INTRODUCTION

The 'standard product' UO_2 fuel pellet has basically remained unchanged for many years and provides excellent performance in all but the most extreme reactor operations. However, the requirement to prolong fuel residence in commercial reactors, thus increasing discharge burn-ups, has led to a need for a detailed understanding and measurements of properties of the fuel, especially at high burn-ups. The fuel properties of prime concern are (a) densification and swelling; (b) formation of high burn-up structure (HBS) at the pellet rim; (c) thermal conductivity; (d) creep rate and mechanical properties; and (e) fission gas release (FGR) during steady-state and transient conditions. These properties are indispensable inputs to various fuel performance and design codes, which are used to design and ensure operational margins of LWR fuel in service.

A minimum size of irradiated fuel sample is required for such properties characterizations in a hot cell lab. For example, thermal conductivity measurement by laser flash technique requires a flat sample of ~ 1 mm^2 size. Depending on the fuel property to be measured or characterized, the sample itself must be free of strong gradients in irradiation temperature and burn-up, not only to limit sample fragmentation during testing, but also to obtain unambiguous data. These attributes are not met if the fuel sample were to be retrieved directly from a discharged commercial fuel rod. Therefore the approach to addressing the needs has been to pre-irradiate fuel samples in a test reactor under controlled quasi isothermal conditions in a well-defined power history, followed by measurements of the properties in the hot cell lab. International collaborative programs HBRP (High Burn-up Rim Project), NFIR (Nuclear Fuel Industry Research) and HPG (Halden Project Group) have taken this approach.

Another approach is to infer the properties from instrumented test rods irradiated in a test reactor. This approach is most successfully implemented in Halden test reactor under the auspices of HPG. Both approaches are complementary and underscore important differences in fuel behavior as observed in the presence of neutron flux (in-pile) versus that from as-irradiated fuel samples by out-of-pile PIE techniques when the neutron flux is absent.

A newly developing approach of computer simulations coupled with accelerator irradiations could complement the test-reactor irradiation and PIE approach referred to above. This is the focus of a Japanese research program called NXO (New Cross Over), which will be briefly described in Section IV.

In this paper we mainly focus on the first approach, where the fuel sample (i.e., a specific fuel variant of UO_2) is typically in the form of a thin circular disc. The irradiation is done under nearly isothermal condition in especially designed test rodlets.

II. DISC IRRADIATION EXPERIMENTS

The industry has completed several separate disc irradiation campaigns over the last few years, of which five were irradiated in the Halden boiling water reactor (see Table I). Here, the irradiation rig in each case was a uniquely designed instrumented fuel assemblies (IFA) to meet certain specific objectives. The basic design principle was irradiation of a stack of fuel discs ~1 mm thick sandwiched between Mo discs. The latter were used to conduct the heat away and maintain a desired uniform temperature across the fuel disc. This is shown schematically in Fig. 1. The fuel disc temperature varies with Mo disc thickness, the radial gap, and the fill gas. The fuel discs were enriched to ~20- 26 w/o ^{235}U for rapid burn-up accumulation of ~20-30 MWd/kgU per year.

TABLE I

Fuel Disc Irradiations at Halden

IFA# (Program)	Time period	Fuel Temperature (°C)	Max burn-up (MWd/kg)
IFA-569 (NFIR)	1989-1991	600-800	77
IFA-563 (HPG)	1991-1996	900-1200-1400	36
IFA-601 (HBRP)	1993-1996	500-700-900-1200	103
IFA-649 (NFIR)	2001-2006	550-700	103
IFA-655 (HPG)	2001-2006	500-700	125

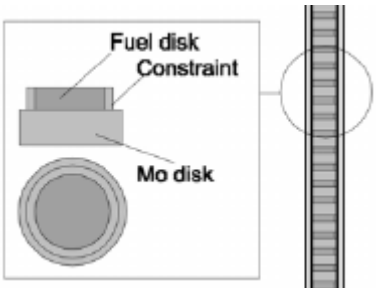

Fig. 1. Basic concept of stacked disc irradiation at Halden

The test rodlets contained a number of fuel variants. They were instrumented somewhat differently in each IFA, as summarized in Table II. The instrumentation included, neutron flux detectors (ND), fuel thermocouples (TF), rod pressure transducers (PF), stack elongation transducer (EF), and gas flow lines (GF), coupled with external γ-spectroscopy to measure in-pile FGR.

The fuel temperatures are dependent on the geometrical information given in Table II. They can be estimated from simplified heat conduction principles (Fig. 2). More detailed finite element computations have also been performed in some cases. A typical example is shown in Fig. 3. These FEM calculations were performed with the QuickField code. The examples in Fig. 3 is for discharge temperature of a 96 MWd/kgU UO_2 fuel disc in the IFA-649 irradiation. The figure shows radial temperature distribution at the disc surface (top plot) and the disc center (bottom plot). In this case from disc center to disc outer radius the temperature difference radially is less than 40°C, whereas axially (along the fuel disc axial center line) the difference is about 110°C (side plot).

TABLE II

Fuel variants, instrumentation, and some design features of the test rodlets

IFA# (Program)	Fuel variants	Instrumentation	Disc diameter x thickness (mm)		Diametral gap (μm)
			Fuel	Mo	
IFA-569 (NFIR)	UO_2, Gd-doped	NDs, TFs and GFs	10 x 1	10.32 x 3	350
IFA-563 (HPG)	UO_2, Gd-doped	NDs, TFs and GFs	9.9 x 1	10.35 x 2	300
IFA-601 (HBRP)	UO_2, Gd-, Ce-, Mg-doped	NDs, TFs and GFs	5 x 1	(7.76-8.26) x (1.2-3.0)	100-200-350-600
IFA-649 (NFIR)	UO_2, Gd-, Cr- doped, MOX, soft-pellets, and ZrB_2 coated	NDs, TFs and PFs	8.2 x 1	(10.37-10.57) x (1.7-2.3)	100-150-200-250-300
IFA-655 (HPG)	UO_2 and MOX	NDs, TFs, PFs, EFs, and GFs	8.2 x 1	10.55 x1 10.40 x 1	100 and 250

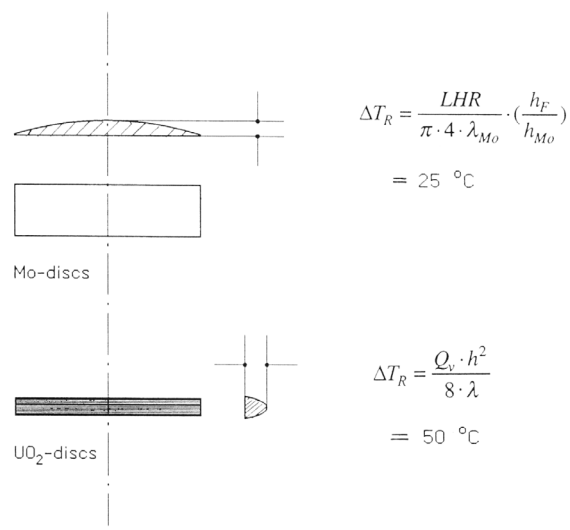

Fig. 2. Simplified estimation of temperature gradients in disc irradiation experiments

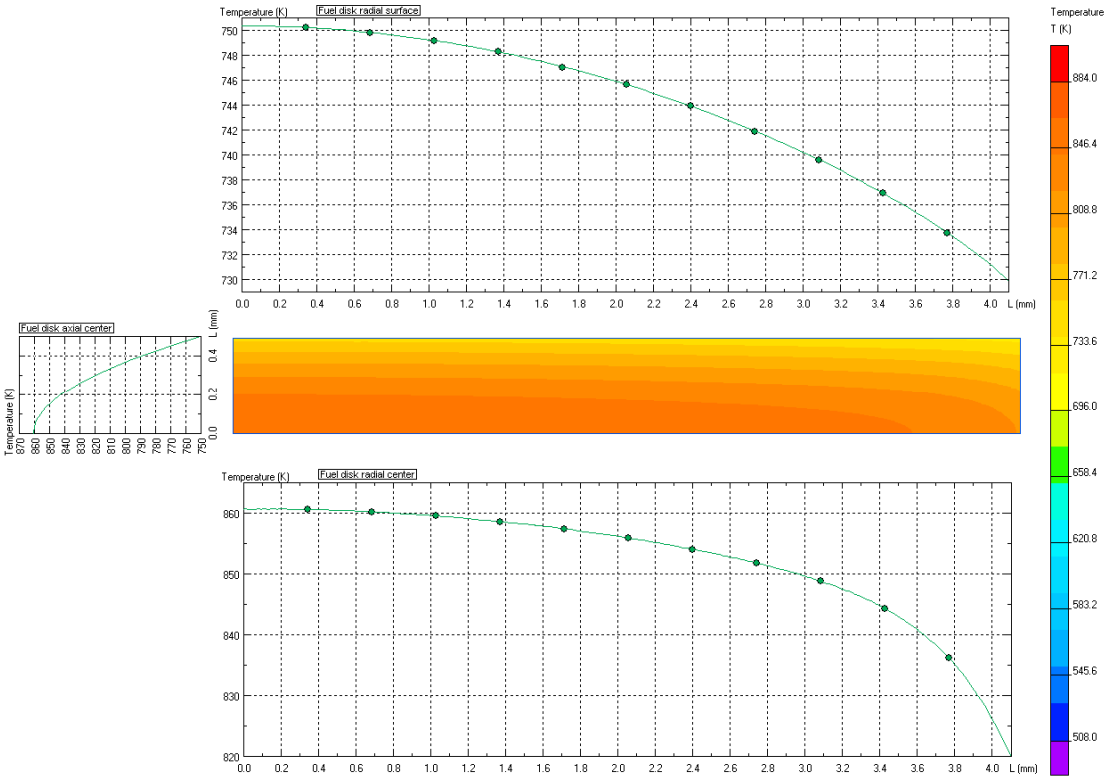

Fig. 3. Example of the detailed FEM calculations of temperature distribution in a UO_2 fuel disc irradiated in IFA-649 at a burn-up of 96 MWd/kgU. Initial disc diameter of disc was 8.2 mm and a diametral gap of 100 μm.

EPMA measurements were performed to examine uniformity of the burn-up along the disc radius. Fig. 4 shows the radial distributions of neodymium along a radial direction of a disc from IFA-601. The fission product Nd is a good burn-up indicator because it is immobile in UO_2. The Nd profile in the figure exhibits only a shallow gradient indicating that the burn-up does not vary significantly over the disc radius.

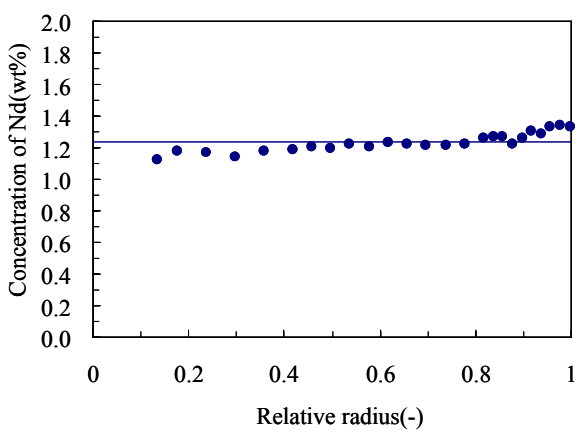

Fig. 4. Uniformity of burn-up distribution in IFA-601 disc, as measured by radial profile of Nd by EPMA [Ref 1]

III. KEY OBSERVATIONS AND RESULTS

III.A. IFA-569

This was the first disc irradiation experiment to be initiated at Halden. The IFA contained 6 UO_2 rods and 3 (U, Gd)O_2 rods. The rig was equipped with 8 TFs to directly measure the irradiation temperatures and 4 NDs to obtain detailed information on rod powers. Typical measured temperatures are shown in Fig. 5. Two of the rods were equipped with GFs to perform gas sweep experiments at the end of the irradiation. These results are shown in Fig. 6. The data show an evidence of grain boundary bubble interlinkage for Gd fuel at a burn-up of ~40 MWd/kg Oxide and ~1200°C and that for standard UO_2 at 1150°C at a burn-up of ~50 MWd/kg Oxide.

Discharge burn-ups of 28, 41, 67 and 80 MWd/kgHM were achieved. The irradiated rods were shipped to Windscale hot cell lab to perform thermal diffusivity (TD) measurements. The TD results have been published previously [2]. The data base has been utilized to develop a thermal conductivity model that has been implemented in fuel performance codes. In addition, density measurements and out-of pile annealing test for FGR were also performed.

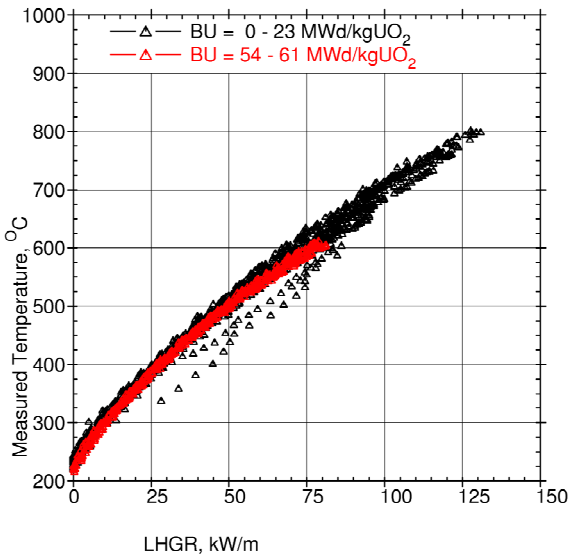

Fig. 5. In-pile temperature measurements in IFA-569

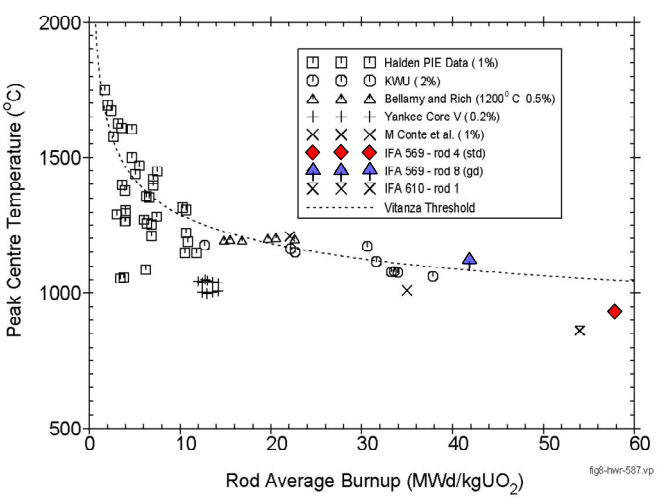

Fig. 6. Fission gas bubble grain boundary inter-linkage observed in IFA-569 compared to other published data and Vitanza threshold

PIE of IFA-569 discs revealed an important phenomenon of thermal diffusivity (TD) "recovery". The as-irradiated disc samples exhibited a rather low value of TD initially. When heated above irradiation temperature, the initially low values of the measured TD kept increasing with temperature. The recovery was completed at ~1200°C. The phenomenon can be explained in terms of annealing of irradiation damage and nucleation and growth of precipitates (particularly fission gas bubbles). Fission products, whether in solid solution or as precipitates, are principally responsible for the conductivity degradation with burn-up, since they markedly impede the phonon movement within the fuel matrix. The point defects, which were continuously created and annealed in-pile, do also contribute toward the recovery during out of pile testing of as-irradiated fuel material.

The TD recovery is not just limited to out-of-pile anneals only. The same basic principles apply to the in-pile conditions as well, with one major exception: Radiation damage (defects) and fission atoms are created and re-solved simultaneously under in-pile conditions, whereas only annealing occurs in out-of-pile samples. The lack of source term to radiation damage and fission atoms should not alter the fundamental processes involved in recovery phenomenon. Our studies have concluded the following:

a. The recovery at lower temperatures is mainly due to radiation damage annealing whereas it is due to fission atoms annealing at higher temperatures;

b. The kinetics of radiation damage recovery appears to be fairly rapid (of the order of few minutes) out-of-pile, but may be significant in-pile where the damage is created and annealed simultaneously.

The kinetics of recovery phenomenon has not been fully exploited by rigorous modeling at a point defects and phonon scale. However, some attempts have been made (e.g., [3]).

III.B. IFA-563

This IFA contained 23 UO_2 rods and 1 (U, Gd)O_2 rod. The rig was equipped with 12 TFs and 8 NDs. Three rods were equipped with GFs. The design of IFA-563 was similar to that of IFA-569. But the main purpose in this case was to provide data on the effect of grain size on in-pile FGR. Accordingly, the UO_2 fuel had three different grain sizes 4, 8 (standard) and 16 μm.

White and Turnbull [4] have analyzed several in-pile gas sweep experiments, including IFA-563 and -569, in an attempt to deduce gas diffusion coefficients. Because of the ability to flush with different gases, thereby producing different fuel temperatures for the same power, the start-up of IFA-563 offered a unique opportunity to obtain data on the fission gas diffusion coefficient. For this reason, the start-up was in the form of a staircase power ramp with each period at part power held long enough to obtain data on released fission gas with both helium and argon sweep gases.

For low fuel temperatures, the release of radioactive fission gases is probably controlled by the third term in the diffusion coefficient proposed by Turnbull et al. [5, 6]:

$$D = D_1(T) + D_2(F,T) + D_3(F) \quad (1)$$

where D is the diffusion coefficient as a function of fission density F (fissions/m^3/s) and temperature T (K). As defined, the third term D_3 is assumed to be athermal, i.e., dependent on local power but independent of temperature. However, analysis of the start-up ramp suggested that even at low temperatures there was a weak temperature dependence as illustrated in Fig. 7. The upper diagram shows the experimentally estimated diffusion coefficient plotted against reciprocal temperature alongside the diffusion coefficient of Turnbull et al [5, 6]. The lower diagram shows the same data plotted alongside a revised diffusion coefficient with a weak temperature dependence for D_3, equivalent to an activation energy of 8000 kcals/mole.

The revised formulation for this term is:

$$D_{3,revised} = 3x10^{-34} \cdot F \cdot \exp(-4000/T) \qquad (2)$$

As can be seen from the figure, it provides a much better fit to the data.

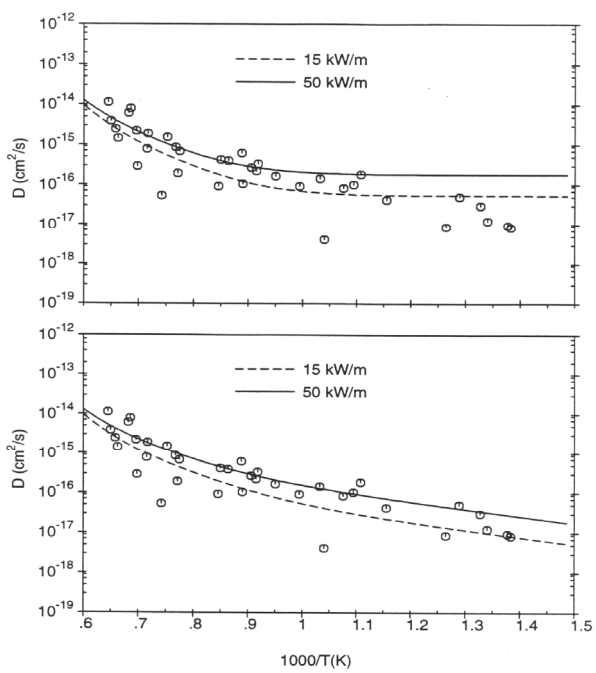

Fig. 7. Start of life data from IFA-563 showing the departure from low temperature athermal diffusivity. This behavior was suspected from other Halden data but required isothermal irradiation IFA-563 for confirmation [Ref 4]

III.C. IFA-601

The HBRP was designed to obtain fuel samples at four different target burn-ups, each irradiated at four different temperatures (see Table III). This was accomplished in IFA-601. Further details are given in Ref 1.

TABLE III

Fuel variants, instrumentation, and some design features of the test rodlets

Rod #	Fuel Temperature (°C)	Max burn-up (MWd/kg U)
1	600	55, 70, 85, 100
2	800	55, 70, 85, 100
3	1000	55, 70, 85, 100
4	1250	55, 70, 85, 100

The object was to generate a wide matrix of PIE samples, with unique burn-up and irradiation temperature pedigrees, from which the threshold conditions for formation of HBS can be determined. Through a very detailed advanced PIE work, this goal was successfully achieved [1, 7, 8]. The burn-up threshold for HBS initiation was found to be between 55 and 65 GWd/tM and the temperature threshold below 1100°C (see Fig. 8). Below this irradiation temperature, and above 65 GWd/tM, all samples were fully restructured, while the HBS initiation could be detected very locally at ~ 55 GWd/tM.

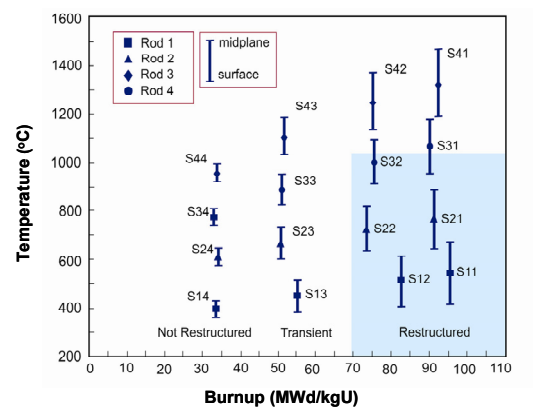

Fig. 8. Threshold for fuel restructuring [Ref 1]

The HBS in LWR rod is also known as rim structure. Ref [9] reported rim width versus average pellet burn-up obtained from PIE of commercial rod fuel pellet material. The data from Ref 9 are reproduced here in Fig. 9. In comparing the onset of HBS (or rim structure formation) from the data in Figs. 8 and 9, certain important differences must be noted. First, the initial fuel enrichment is higher in all disc irradiation experiments compared to that in a typical LWR rod. In IFA-601, it was 26.5 wt% ^{235}U, which led to a fission density about ~ 4 times higher than in LWR fuel. Second, the disc samples in IFA-601 were not constrained, unlike the condition prevailing in the rim region of a LWR fuel rod. Third, the roughly uniform burn-up in the disc samples must be contrasted against relatively steep radial burn-up gradient in the fuel rod and the fact that the local burn-up determination in the LWR

pellet is inherently less accurate. Finally, irradiation temperatures are different in two cases. In HBS (rim) region of a commercial fuel rod, the rim temperature can only be estimated by codes, while temperature control was attained by design in IFA-601. Given these differences, the HBS threshold from IFA-601 appears to be a conservative value [9].

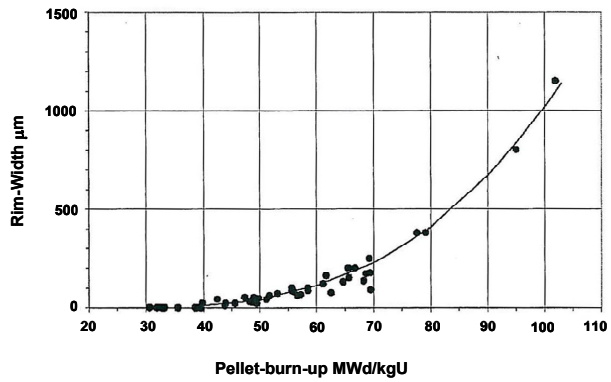

Fig. 9. Rim width (µm) vs pellet burn-up [MWd/kgU] in LWR rod [Ref 9]

Extensive measurements [7] of materials properties were made such as lattice parameter, microstructure, density, and thermal diffusivity [10]. The TD results were found to be consistent between NFIR (IFA-569) and HBRP (IFA-601) samples, as shown in Fig. 10. The high burn-up density data from the two disc irradiation campaigns have also been compared [11].

From the microstructure studies (a large number of SEM and TEM characterizations), a better understanding of the phenomena involved in the fuel restructuring processes has been achieved: bubbles and sub-grains sizes, interactions between sub-micron bubbles and dislocations, role of precipitates [8]. In addition, mechanical properties [12] have been pursued. Further details on this are covered in section IV.

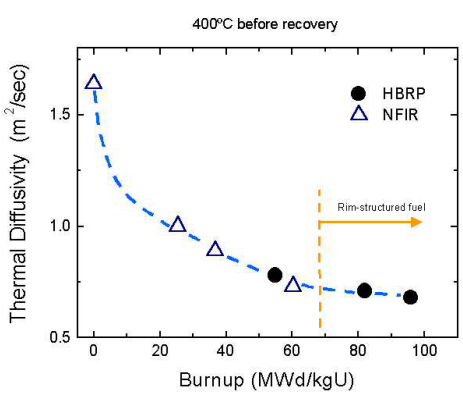

Fig. 10. Rim width (µm) vs. pellet burn-up [MWd/kgU] in LWR rod [Ref 9]

III.D. IFA-649

This irradiation rig has irradiated 24 rodlets in five Halden cycle loadings. A number of fuel variants (see Table II) have achieved burn-ups ranging form 50 to105 MWd/kgU. The fuel variants also included two distinct types of MOX fuel. In addition, two irradiation temperatures (550 and 700°C) and two types of specimen restraints (Mo ring around the disc and hydrostatic pressure) were implemented. There were no gas flow lines. The irradiated fuel specimens have been shipped to two European hot cell labs for advanced PIE work that have been initiated. The objectives of PIE is to (a) perform FGR release studies at normal and high external pressures coupled with advanced electron microscopy to determine local gas distribution and quantification (LGDQ) and (b) measure high burn-up properties, including extending the industry data base of thermal diffusivity, specific heat and fuel mechanical properties measurements.

III.E. IFA-655

Unlike IFA-649, IFA-655 was equipped with gas flow lines and stack elongation transducers. This allowed studying of the behavior and properties of HBS fuel by in-pile measurements of stable and radioactive FGR and fuel swelling. The UO_2 and MOX fuel variants in IFA-655 were identical to those of IFA-649. An additional purpose of IFA-655 irradiations was to produce HBS discs to be used in transient testing at Halden. The latter are presently in progress.

The classical method of in-pile gas sweep coupled with external monitoring of radioactive isotopes was followed throughout the whole IFA-655 irradiation. This provided data on release to birth (R/B) and surface to volume ratios (S/V) for various fuel variants (see Fig. 11).

It can be seen that for all rods, apart from rod 8 and 12, irradiation at low temperature (≤700°C) had no significant increase in gas release even when the sample passed through the burn-up regime of HBS formation. This is an important point with implications for FGR modeling at high burn-up, where some models assume, erroneously, that release increases locally due to HBS formation.

The large step in derived R/B and S/V values for two rods— 8 and 12 are due unintentional temperature excursions to around 1000°C. These results can be compared and shown to be in accord with the high burn-up data points in Figure 6.

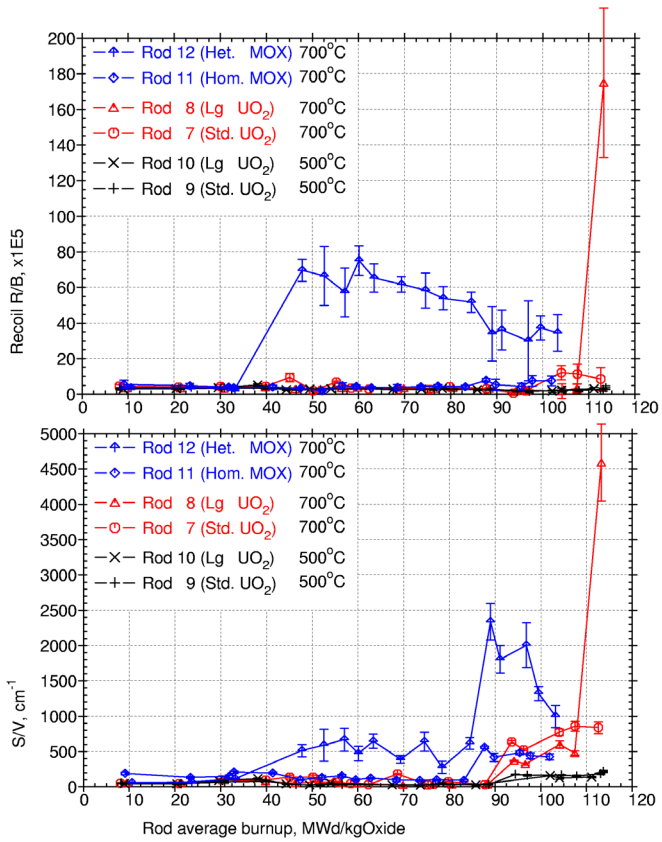

Fig. 11 Release to birth (R/B) and surface to volume ratios (S/V) from gas flow measurements in IFA-655

IV. FUEL R&D NEEDS AND OUTLOOK

The five disc irradiation campaigns highlighted in this paper have shown that this approach to fuel R&D has a proven record of success, yielding insights into fuel behavior in-pile, e.g., through gas sweep experiments, but more importantly, from out-of pile characterizations and measurements.

The fuel samples from IFA-649 are still being utilized in further work in NFIR. Since FGR is still not clearly understood and has proven difficult to model in engineering fuel performance codes, extensive out-of pile annealing studies with concurrent FGR and detailed PIE are required. Thus the studies on IFA-649 fuel variants have been aimed at getting quantitative data on (a) High burn-up fuel during the course of a LOCA-type transient; (b) effect of MOX heterogeneity; and (c) The effect of fuel additives and microstructure.

Similarly, there is continuing need for high burn-up fuel properties data. In particular:

A. Onset of HBS formation and its dependence on grain size, additives, temperature and/or hydrostatic restraint;

B. Determination of density and changes in porosity; the effect of hydrostatic restraint;

C. Thermal conductivity as a function of burn-up; the effect of grain size and fuel variants; the implication of HBS formation;

D. Specific heat measurements; and

E. Evolution of the mechanical properties including thermal creep measurements; the mechanisms involved in visco-plastic deformation behavior

With respect to item E above, creep properties of the pellet have been used as an argument to relax start-up and reconditioning constraints. During LHGR changes, even without pellet-clad contact, a high creep rate in the fuel could relax the stresses in the pellet and avoid early fuel cracking. The latter phenomenon directly impacts PCI-SCC modeling of the fuel rods. Therefore a better knowledge of fuel creep, especially for irradiated fuel, will certainly be helpful.

Generally, creep formulation is based on two major terms. A term due to thermal creep and another one due to irradiation creep. Compressive tests and three points bending tests on non-irradiated fuel materials have allowed developing thermal creep models. Often a multiplicative term on the thermal creep, proportional to dF/dt (fission rate per unit volume), is used to represent the acceleration of creep due to the irradiation. In practice, the thermal creep is predominant above 1000°C (mainly activated during power ramps), while the irradiation creep is a long term creep predominant under 1000°C (base irradiation).

In this context, the concept of fuel disc irradiation is planned also for a new experiment on fuel creep within the HPG. The objective is to measure the irradiation-induced creep rate of UO_2 fuel and other fuel variants as a function of varying applied compressive stress at a fixed fission rate. Fuel variants (in the form of 1 mm thick discs) will be subjected to compressive load by means of a bellows loading unit with the load applied through external gas lines through bellows. An LVDT type elongation detector will measure the overall length change of the fuel disc stack from the creep Fuel disc temperatures can be varied and controlled by means of an external gas line to the fuel rods, giving the possibility of changing the fill gas composition (and thus irradiation temperature).

Many observations on high burn-up fuel suggest a drastic evolution of the fuel mechanical properties [12]. It is also important to elucidate the thermal-mechanical behavior of the fuel with the objective of improving the engineering design of the fuel rods, especially after a strong pellet-cladding mechanical contact has been established. This knowledge can be incorporated into the fuel modeling codes, where the aim is to numerically

simulate the interaction between a fractured ceramic cylinder (nuclear fuel pellet) and a metallic tube (Zircaloy cladding). While the evolution of the cladding mechanical properties with neutron fluence is relatively well understood through a large number of mechanical testing, this is not true for fuel pellet material. For nuclear fuel pellet, the evolution of the mechanical properties with burn-up is challenging because of the steep radial gradient in the properties as a result of the irradiation, but also because of the pellet cracking and micro-cracking. The remaining questions in this area are about the evolution with burn and temperature of the: (a) elastic properties; (b) thermal creep behavior, where the possibility for the grain boundaries sliding mechanisms may become predominant; and (c) irradiation creep.

For the two first items, out of pile characterization must be conducted in hot cells on well characterized irradiated samples. The fuel disc irradiation concept providing reasonably sized samples with quasi-homogeneous properties are pertinent for such characterization.

Even with the extensive PIE in hot cell labs described in this paper, a first principle based understanding of irradiation induced effects in ceramic oxide fuel materials is lacking. These include detailed evolution of the valency changes of cations in non-stoichiometric UO_2 and mechanism of the defect ordering of oxygen sub-lattice. In this context, the principal approach of the NXO program is to perform simulation of underlying complex processes as a whole. Its applications could range from a qualitative mechanistic study to a quantitative material property evaluation.

As an example, the grain subdivision of the HBS formation can be studied by focusing on the principal processes using accelerator irradiation, with confirmatory comparison to be made with TEM observations on IFA-601 specimens.

Although irradiation and PIE are currently key requirements in licensing, they are time consuming and often expensive. For design of advance new materials and to understand the effects of fuel additives, for example, NXO's fundamental approach can provide valuable insights, ultimately accelerating the development of new advanced material and by providing supporting data base for their licensing.

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ACRONYMS

EPMA	Electron probe microanalysis
LWR	Light water reactor
LOCA	Loss of coolant accident
MOX	Mixed oxide
SEM	Scanning electron microscopy
TEM	Transmission electron microscopy