



Post irradiation examination of nuclear fuel: Toward a complete analysis



Claude Degueldre ^{a, c}, Johannes Bertsch ^{a, *}, Matthias Martin ^b

^a LNM, NES, PSI, 5232 Villigen, Switzerland

^b AHL, NES, PSI, 5232 Villigen, Switzerland

^c Engineering Department, Lancaster University, Lancaster LA1 4YW, UK

ARTICLE INFO

Article history:

Received 19 February 2015

Received in revised form

19 February 2016

Accepted 31 March 2016

Keywords:

Irradiated nuclear fuel

Post irradiation analysis

Fission products

Actinides

ABSTRACT

Nuclear fuel material changes arise over the whole fuel life-cycle, during operation, after unloading and during storage. For a good characterization and assessment of the fuel comprehensive post irradiation examination (PIE) is essential. PIE has to take the complementarities of various methods into account and the accessibility of sample material, i.e. whether an examination can be done in non-destructive manner, or whether destructive preparation steps are needed. Besides materials questions also fuel system issues need to be addressed, e.g. the interaction between pellet and cladding. Further, a PIE campaign has to be composed in a sequential and cost effective way.

Today, for a complete irradiated fuel characterization a suite of elemental and isotopic analyses is needed, together with chemical speciation and structure determination. This requires the use of advanced spectroscopic techniques allowing a resolution giving access to burn-up related questions. Thus, besides typical hot-laboratory instrumentation also synchrotron radiation is used more and more. Further, differentiation of the analytical methods between those with a more passive character, using the activity of the fuel itself, or with a more interactive character using excitation e.g. by micro-beam is useful. In practice, this study presents specific examples comparing analyses of non-irradiated with irradiated fuel in the fuel pellet center and the periphery:

- Fuel morphology and porosity.
- Intermetallics particles as temperature indicator.
- Cesium volatility, precipitation and distribution in irradiated fuel.
- Fission gas ratio and density in aggregates.
- Challenges in actinide speciation and reactivity in irradiated fuel.

The discussion focuses on the potential of the techniques and their limitation (interference, detection limit, applicability) for fuel analysis. It also discusses and compares pore, fission gas aggregate and sub-grain densities as well as other phase occurrence.

It may be concluded that in dependency on the scientific question a specific combination of isotopic, chemical, physical or structure related methods is necessary, composed in a sequential and incremental way. In future amendments are needed for targeted sample preparation, micro-tomography and testing on a microscopic level.

© 2016 Published by Elsevier Ltd.

1. Introduction

Fuel vendors and nuclear power plants seek for an increase of electricity output per fuel unit while the fuel safety must not be

compromised or shall be improved. In a long co-operation between fuel vendors and power plants in Switzerland and with the approval of the authorities an increase of burn-up limit could be reached. The Paul Scherrer Institute (PSI) with its Hot-Laboratory (AHL) and the Laboratory for Nuclear Materials (LNM) have been largely involved in concomitant Post-Irradiation Examination (PIE) campaigns and in scientifically discussing and clarifying related materials questions e.g. Degueldre et al. (2011a). Today, PIE is a suite of methods, complementary and organized in an effective and

* Corresponding author.

E-mail addresses: claude.degueldre@unige.ch, c.degueldre@lancaster.ac.uk (C. Degueldre), johannes.bertsch@psi.ch (J. Bertsch).

economical way. Advanced analytical tools such as synchrotron radiation are more often applied.

The scientific questions behind PIE and irradiated fuel concern the changes of the fuel with burn-up: How is the fuel performance changed? What is the impact on the safety under normal and transient conditions? And how can the degradations be amended by measures, e.g. by appropriate fuel operation, by adjusted fuel fabrication or by mixing additives into the fuel? To approach these questions the fuel needs to be best characterized with respect to its chemistry, isotopic composition and physical properties as well as morphology and structure e.g. see Degueldre et al. (2011a). It is not sufficient to investigate the properties of a fuel pellet or rod from an integral point of view. The fuel does not remain homogeneous with burn-up, the properties change in the pellet with radius and along the fuel rod axis. By consequence, a challenge is a representative characterization, and – when applicable – taking 3D effects into account.

The scope of nuclear fuel PIE increases. Traditional PIE comprise macroscopic visual inspection, γ -radiography and microscopic characterization with elemental and isotopic analysis of fission products and fuel additives e.g. ceramography, Electron Probe Micro-Analysis (EPMA) and Secondary Ion Mass Spectroscopy (SIMS), respectively. A specific focus has been on fission gas creation, accumulation and distribution.

However, today the possibility of speciation and structural analysis by advanced spectroscopic techniques e.g. synchrotron radiation provides a resolution inherent to changes in the fuel related to increasing burn-up or high burn-up structure, see e.g. Degueldre et al. (2010).

The paper is organized as follows: in the next chapter the methodological approach is described considering also the strong radioactivity of irradiated fuel. In a further chapter the methods are presented focusing and applying techniques operated at PSI. Advantages and drawbacks of techniques combination are analyzed with emphasis on the performance and safety relevant issues addressed by the fuel vendors, the operators and the regulators. Results are subsequently discussed and combined to build a more comprehensive picture. Finally, the conclusions comprise the learned and future needs.

2. Methodological approach

PIE begins with visual inspection of the fuel element or of the fuel rod in the reactor pool or in the hot-cell prior to further analyses using selected techniques according to the required analytical needs.

The analytical techniques can be discriminated between passive ones, taking profit of the activity of the irradiated fuel, and those with interactive character. Based on this classification Table 1 lists possible techniques discussed in this work. After description of the techniques, their advantages and drawbacks as well as their combination are applied and discussed later. Techniques using phonon, positrons, muons or antineutrinos are not included because they focus on very specific properties. The techniques presented in Table 1 would complete 'traditional' PIE's on the fuel rods such as: dimension measurements, corrosion layer thickness determination, rod puncturing for gases analysis ... Table 1 also indicates the analytical information (isotope, element, species or morphology) provided by the applied techniques.

For all these techniques their sensitivity, detection limits in term of amount or concentration and in term of lateral resolution or volume of the investigated sample are relevant. The sensitivity, κ , and detection limit, DL , of the concentration C of isotope, element or species must be discussed at theoretical and experimental level.

From the experimental side these concentration and amount limits are given by:

$$C_{DL} = 3 \cdot \sigma \cdot \kappa \quad (1)$$

where C_{DL} is the concentration at the detection limit and σ (au) is the standard deviation of the limiting noise signal; the factor 3 is suggested in the definition of detection limit by IUPAC e.g. Curie (1999).

By consequence, the minimum detectable mass, $m_{\min,DL}$ of, e.g. an element in a matrix is dependent on the concentration limit C_{DL} and the minimal mass of the sample, m_{\min} , which just still yields a usable signal.

$$m_{\min,DL} = m_{\min} \cdot C_{DL} \quad (2)$$

From the theoretical side, a detection limit may be evaluated from the physical-chemical process and from the performance of the analytical unit.

For all analyses, sample volume, mass or amount, the flux of reagent, the size of the analyzed part of the sample and the acquisition time or time of analysis are key parameters linked to the detection limit. The nature and origin of the sample dictate the size of the sample. However, the size of the sample is also coupled with the analytical technique for which time and detection limits are key parameters for its application. The detection limit is a function of the number of atoms, a function of the volume of the sample, sub-sample excitation conditions, and the acquisition

Table 1

Analytical techniques, including excitation (if any) and detection for isotope, element, or species characterization.

		Detection			
		Photon	Electron	Neutron	Ion
Excitation	none/passive	γ -tomography ⁱ , γ -spectroscopy ^j	β -spectroscopy ^j	n-radiography ⁱ , n-tomography ^j	
	Photon	optical microscopy ^m , XRF ^e , XAS ^e , XRD ^m , γ -tomography ^m			laser ablation – ICPMS ^{i a}
	Electron	EDS ^e , EPMA ^m , WDS ^e	SEM ^m		Mass spectrometry ⁱ
	Neutron Ion			n-radiography ^m	ICPMS ⁱ , SIMS ^{i b}

XRF – X-ray fluorescence, XAS – X-ray absorption spectroscopy, XRD – X-ray diffraction, EDS – energy dispersive spectroscopy, SEM – scanning electron microscopy, EPMA – electron probe microanalysis, WDS – wave dispersive spectroscopy, ICPMS – inductively coupled plasma mass spectrometry, SIMS – secondary ion mass spectrometry. Note: methods dealing with phonons, muons, neutrinos are not included in this study.

Analytical result about: isotope (ⁱ), element (^e), speciation (^s), morphology (^m).

^a Material evaporation by laser and subsequent excitation by plasma ionization.

^b Excitation by collision cascade ionization and sputtering.

Download English Version:

<https://daneshyari.com/en/article/8084887>

Download Persian Version:

<https://daneshyari.com/article/8084887>

[Daneshyari.com](https://daneshyari.com)