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Physical and mechanical characterization of irradiated uranium dioxide with a broad burnup range and different dopants using acoustic microscopy

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ABSTRACT

Scanning acoustic microscopy is a non-destructive technique that allows determining the local material elastic properties by measuring the velocity of acoustic waves propagating in matter. High frequency acoustic waves are generated by a piezoelectric transducer, focused and then detected by the same transducer after having interacted with the sample. This technique has been employed in the past to assess different types of irradiated nuclear fuel and unirradiated chemical analogues of UO2 and it has enabled to relate the Rayleigh wave velocity of propagation with the Young's modulus and the density of the material. In the present study, thanks to new measurements on irradiated fuel and to analysis of data from the open literature, the variation of the density with burnup is determined up to ~ 100 GWd·t^{-1}M. The porosity is then determined taking account of the irradiated fuel matrix swelling. Finally, an expression is proposed describing Young's modulus as a function of burnup, that can be used in fuel performance calculation.

1. Introduction

During its life in reactor the nuclear fuel is subjected to accumulation of radiation damage and fission products (FPs) and experiences significant micro-structural changes. Typically, every fission produces around 100 000 displacements in the crystal lattice and two fission products which can be classified, according to Kleykamp [1], in four categories: 1) fission gases and other volatile fission products e.g. Kr, Xe, Br, I; 2) metallic precipitates e.g. Mo,Tc, Ru, Rh, Pd, Ag, Cd, In, Sn, Sb, Te; 3) oxide precipitates e.g. Rb, Cs, Ba, Zr, Nb, Mo, Te; 4) fission products dissolved as oxides in the matrix e.g. Sr, Zr, Nb, and the rare earths Y, La, Ce, Pr, Nd, Pm, Sm. Some of the FPs can be present in different forms and with different degrees of volatility, depending on the local chemical conditions; for instance oxygen potential can drive transitions of non-volatile FPs between metallic and oxide precipitates affecting the speciation of other FPs, such as Ba or Cs which might form ternary compounds with Mo, Zr or U [2] [3]. Moreover, oxygen potential may strongly impact the volatility of compounds formed by semi-volatiles fission products such as Mo, Rh, Ba, Pd, Tc. A small but not completely negligible contribution to the damage is derived from the radioactive decay of fission products or higher actinides produced by neutron capture. In particular, alpha decays cause displacements in the lattice and accumulation of He that may precipitate in bubbles [4]. The most notable structural modification which typically affects the periphery of the Light Water Reactor (LWR) fuel pellet, when its average burnup reaches about 50 GWd·t^{-1}M, is the formation of the High Burnup Structure (HBS). HBS is characterised by the subdivision of the original grains into grains of 0.1–0.3 μm and the formation of a high concentration of micron-sized pores containing the fission gas [5].

At the beginning of the irradiation the LWR fuel experiences a densification, but around 10–15 GWd·t^{-1}M this phenomenon

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saturates and the nuclear fuel volume increases with further increase of the burnup. This is due to the accumulation of insoluble solid fission products and noble fission gases. Both processes are responsible for the matrix swelling, which is considered to be between 0.8% and 1% per 10 GWd·t^{-1}M up to a burnup of 50 GWd·t^{-1}M [6–9]. Spino et al. [7] concluded that the matrix swelling is about 1% per 10 GWd·t^{-1}M up to 60 GWd·t^{-1}M, but at greater burnup it decreases (0.32% per 10 GWd·t^{-1}M) and becomes dependent only upon solid fission products precipitation. The initial densification causes a fraction of the fabrication porosity to disappear, whilst the production of extended defects such as voids contributes to the generation of new pores in the irradiated fuel. Fission gas atoms precipitate into bubbles in correspondence with defects location, or in some pores remaining after the initial densification. Intragranular and intergranular fission gas bubbles grow in size due to the absorption of the fission gas continuously generated or because of other growth mechanisms, reaching sizes $\geq 1\mu m$. The increase of the porosity enhances the total fuel swelling. The global effect of the swelling is a density decrease and this contributes to the reduction of Young's modulus observed when the burnup increases [10–13]. The determination of the fuel elastic modulus is necessary to predict the in-reactor fuel mechanical behaviour, as it is related to the tangential or hoop stress and to other relevant processes e.g. the cracking pattern during a power ramp [14]. Moreover, the increase of fuel volume filling the initial pellet-cladding gap can result in chemical and mechanical interactions between fuel and cladding [15] that may affect the cladding lifetime and impact fuel safety.

During the last twenty years, the variation of Young's modulus as a function of burnup has been investigated on irradiated UO_2 and MOX samples in addition to chemical analogues of irradiated UO_2 [16–18]. Laux et al. [12] observed a decrease of Young's modulus as a function of burnup, with a reduction of 25% between 0 and 100 GWd·t^{-1}M. In particular, they have demonstrated [12] that assuming a constant Poisson's ratio ($\nu = 0.3$) only one side (typically a polished surface) of the fuel specimen can be used to calculate Young's modulus from the Rayleigh wave velocity, when knowing the density of the specimen. Thus, a relation between the density of the irradiated fuel and the Rayleigh wave velocity has been determined by Laux et al. in Ref. [18] enabling the determination of the fuel Young's modulus without prior measurements of the density.

In this paper, a new relation describing Young's modulus as a function of burnup is proposed. The empirical approach used takes account of the relations between density and acoustic properties as well as the variation of density as a function of the burnup. By separating the matrix swelling and porosity increase effects in the burnup range considered (up to 100 GWd·t^{-1}M), the approach used in a previous work [19] is confirmed and justified and the local determination of porosity is made possible. The steps followed are summarised below:

Step 1. Verification of the existing relations between Rayleigh wave velocity, density and Young's modulus for uranium dioxide.

Step 2. Investigation of the relation between density, corrected for the average porosity, and burnup using the experimental results from irradiated fuel samples and data available in the open literature. This allows an estimation of the matrix swelling in the 0–100 GWd·t^{-1}M burnup range as well as calculation of the local porosity.

Step 3. Examination of the variation of the bulk density as a function of burnup and combination with the findings of step 1.

Step 4. Definition of a new empirical relation describing the variation of Young's modulus in the burnup range 0–100 GWd·t^{-1}M, taking into account also dopants concentration.

2. Materials and methods

2.1. Samples

Table 1 lists the samples investigated by acoustic microscopy during approximately twenty years of experimental campaigns. In this work, a new measurement campaign has been performed on commercial fuels irradiated in Pressurised Water Reactors (PWR) [19] [20] and on special irradiation discs irradiated in the Halden reactor and belonging to the Nuclear Fuel Industry Research (NFIR) program [21].

As required to perform this type of characterization, the specimens (cross sections cut from the LWR fuel rod, or special irradiation discs) were mounted on microscopy holders. Prior to the measurements, each sample underwent grinding and polishing by means of diamonds pastes with particle sizes from 12 μm to 1 μm, in order to prepare the surface for analysis.

2.2. Measurement technique: high frequency acoustic microscopy

The above-mentioned samples were characterised using non-destructive high frequency acoustic microscopy based on the measurement of the Rayleigh wave velocity. A dedicated setup was installed in a hot cell and adapted to the assessment of irradiated nuclear material. Rayleigh waves are a particular type of surface waves, slower than the bulk transverse and longitudinal modes, which penetrate the material to a depth comparable with their wavelength. A high frequency acoustic microscope is generally composed of a piezoelectric transducer placed on one flat side of a high purity silica cylinder which constitutes the acoustic lens. The lens lower extremity is provided with a small spherical diopter, etched and well polished, whose role is to focus the incident waves onto the specimen [32,33]. The piezoelectric transducer converts the incoming electrical signal into plane acoustic waves, which are transmitted to the sample through a coupling liquid. The incident waves are focused on the sample and then are reflected back and detected by the same piezoelectric transducer. The coupling liquid must ensure the transmission of the acoustic waves with negligible attenuation and must be chosen taking into account the acoustic impedance of the material under investigation [34]. For uranium dioxide a suitable coupling liquid is methanol. The amplitude of the reflected waves is converted to a grey scale; therefore by scanning mechanically the sample along a plane parallel to the sample surface it is possible to obtain an acoustic image. The resolution of the technique depends on the frequency applied: higher frequencies give better spatial resolution. Fig. 1-(I) shows the surface acoustic image of one of the NFIR specimens. The frequency applied is 140 MHz; the spatial resolution of the image is about 7 μm. A sample of an acoustic image acquired at 60 MHz is shown in Fig. 1-(II), where the spatial resolution is slightly less than 15 μm. An interesting and advantageous feature of the acoustic imaging in comparison to optical methods is the possibility to focus below the surface obtaining a sub-surface image, with a limitation in depth dependent on the applied frequency. To determine the elastic properties of materials, the acoustic microscope is used in *Acoustic signature mode* [32]. By gradually defocusing, achieved by approaching the acoustic sensor to the specimen, an interference is generated between the reflected normal waves and the Rayleigh waves. The piezoelectric transducer thus receives a pseudo-periodical signal whose period is related to the Rayleigh wave velocity. If Δz is the spatial period of the signal (m), v_R the Rayleigh