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Neutron radiography and tomography applied to fuel degradation during ramp tests and loss of coolant accident tests in a research reactor

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ABSTRACT

Neutron radiography (NR) is performed at the Institute for Energy Technology (IFE) in Norway since the late 1970s. The application of the non-destructive method was to acquire post-irradiation examination (PIE) data (e.g. fuel integrity and hydrogen up-take in cladding) from safety and integrity tests of nuclear fuels performed under the Organization for Economic Co-operation and Development (OECD) Halden Reactor Project (HRP). The method was later applied under re-fabrication and instrumentation operations of experimental nuclear fuel rods prior to testing in Halden Boiling Water Reactor (HBWR), and for a variety of PIE projects, e.g. reactor power ramp testing, PCI failure detection and fuel degradation experiments. Neutron radiography has also proved to be a very useful tool for examination of nuclear fuels irradiated in the Loss-of-Coolant Accident (LOCA) experimental series initiated in the early 2000s. Neutron tomography data is acquired while an increased international focus arose on fuel fragmentation, fuel relocation and fuel dispersal processes that occur during the LOCA events for high burn-up nuclear fuels. Hydrogen up-take of the fuel cladding, fuel pellet-clad bonding condition, fuel fragmentation, particle size distributions, and other features obtained from neutron tomography data are quite relevant for reactor core safety impact study of LOCA events simulated in the HBWR. Neutron tomography studies of LOCA tested fuel were done in cooperation with the SCK·CEN institute in Mol, Belgium, and the University of Antwerp in Belgium. It's interesting to observe that the image reconstruction results obtained from the SART method are quite good regarding the relatively few sample rotations utilized under acquisition of neutron radiography projections in the tomography studies of the LOCA examination.

1. Introduction

Norway was among the first countries to put a nuclear research reactor in operation. The Halden Reactor Project (HRP) was affiliated to OECD NEA and the first agreement was signed in 1958. Customers in the HRP are nuclear utilities, vendors, licensing authorities and R&D centers. The project contributes to improve the safety at nuclear plants around the world through investigation of fuels and irradiated reactor materials. The neutron radiography facility at the IFE JEEPII (Joint European Energy Pile) heavy water research reactor facility located at Kjeller, Oslo region, was constructed and built during 1972–1974.

Neutron radiography is a powerful non-destructive material examination technique (Lehmann et al., 2003; Jenssen and Oberländer, 2002). The high attenuation of neutrons in hydrogenous materials, with a high penetration for heavy metals, makes neutron radiography a complementary technique to X-ray imaging. Unlike X-rays and γ-rays, neutron interaction is characterized by nuclear rather than electronics of the medium through which it passes. The development of neutron radiographic relevance had to wait for the advent of sufficiently intense neutron beams which became available with the development of research reactors in the 1950s. Today, also spallation initiated neutron sources (SINQ) are utilized in neutron radiography with excellent examination results, e.g. at PSI in Switzerland (Lehmann et al., 2011).

Neutron radiography of irradiated fuel rods is extensively used for image acquisition and qualitative analysis of cladding with respect to hydrogen up-take, secondary fuel degradation experiments and reactor power ramp experiments performed under the

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HRP. Pellet cladding interaction (PCI) failures and clad hydriding are well documented from neutron radiographs. Some examples from ordinary neutron radiography are discussed in the paper. Neutron radiography is applied on experimental boron carbide (B_4C) control rods and under re-fabrication of high burn-up fuel rods which are modified with new instrumentation for further testing in the HBWR. Design basic (DB) LOCA experiments are performed for many years in the HBWR (Oberländer et al., 2008; Kolstad et al., 2011). Neutron radiography is a very useful tool utilized under re-fabrication of irradiated fuel rods and interpretation of examination results, e.g. fuel fragmentation, relocation and disposal under LOCA examination.

Tomography utilizing neutrons has developed over the last 10–15 years as an interesting expansion of the traditional two-dimensional neutron radiography. The meaning of computerized tomography (CT) is the reconstruction of a function from its line or plane integrals, irrespective of the field where this technique is applied. Neutron tomography provides three-dimensional spatially resolved images which normally display the attenuation coefficient distribution over the sample volume. The reconstruction of image cross-sections is based on a set of radiographs (projections) obtained at different equidistant angles spread 180° around the sample. The reconstruction of the sample attenuation coefficient acquired from a fuel rod irradiated in the Halden Reactor in a particular LOCA test is demonstrated with the Simultaneous Algebraic Reconstruction Technique (SART) using the open source ASTRA toolbox developed at the Antwerp University (Andersen and Kak, 1984). Also, another tomography method based on Chebyshev moments, which is very promising, is presented in the paper. The motivation for applying CT in neutron radiography in examination of irradiated fuel rods is to obtain fuel rod macrographs without cutting the rod for destructive metallography and ceramography examinations.

2. Source, principle and techniques utilized in neutron radiography

Two neutron radiography (Harms and Wyman, 1986) techniques are utilized at the Institute for Energy Technology under examination of irradiated experimental rods with UO_2 or MOX fuels, zircaloy cladding and instrumentation devices. Both techniques allow a collimated thermal neutron beam to be used and enable neutron radiography of very active samples. The traditional method uses an activation transfer technique, utilizing a dysprosium foil and X-ray film. The other method uses a special solid nuclear track-etch recorder, designed for detecting ionizing particle tracks. Solid nuclear track-etch recorders have numerous applications, e.g. dosimetry of neutrons and of heavier ionizing particles such as alpha particles, protons, fission fragments, spallation fragments, heavy ions and very heavy ions such as cosmic rays, etc. In every instance the thickness of the cellulose–nitrate film should be in accordance with the mass and energy of the particle type to be detected in order to obtain the optimum level of energy transfer. The track-etch recorder consists of a 100 μm thick plastic film of lightly rose-tinted cellulose nitrate, coated on both sides with a neutron to α-particle converter material. The film is digitized prior to neutron radiograph analysis. A spatial resolution of 40–50 μm is achievable for neutron radiography performed in the JEEPII reactor for both methods applied on fuel rods.

2.1. Neutron source

The primary source of neutrons was obtained from one of the radial channels of the JEEP II reactor at the Institute for Energy Technology, Kjeller. The sample or fuel rod was irradiated by a collimated thermal neutron beam of height 220 mm and width limited to 30 mm. The thermal neutron flux in the beam channel is $\sim 10^7\ \mathrm{cm^{-2}\,s^{-1}}$, and the length between graphite collimator and activation screen is ∼3000 mm.

2.2. Principle

The principle of image formation in neutron radiography is based on attenuation of a collimated neutron beam transmitted through a sample. The degree of attenuation is measured by detecting the intensity I of the neutron beam transmitted through the sample or fuel rod with unaltered direction and it is given by Beer–Lambert attenuation law

$$I = I_0 e^{-\int \mu(x,y,E)\mathrm{d}s} = I_0 e^{-\int \Sigma_t \mathrm{d}s} \tag{1}$$

The integration must be performed along the neutron beam trajectories. I_0 is the intensity of the incoming beam and Σ_t is the total macroscopic cross section of the material, i.e. $\Sigma_t = \sum(n\sigma_a)/V + \sum(n\sigma_s)/V.\Sigma$ is the sum of all the different isotopes in the volume V and n is the number of atoms of one kind with microscopic absorption cross-section and microscopic scattering cross-section given by σ_a and σ_s, respectively. The Radon transform gives:

$$g(X,\alpha) = \int \mu(x,y,E)\mathrm{d}s = -\ln\left(\frac{I}{I_0}\right) \tag{2}$$

X and α represent the spatial coordinate in the projected images (neutron radiographs) and the sample rotation angle (anti-clockwise), respectively. The x,y and E represent the spatial coordinates in a non-rotating system and the neutron energy, respectively.

2.3. Dysprosium foil and X-ray film technique

The method relies on the build-up of radioactivity in the foil produced by neutron absorption. In this way an activation image is formed in the foil. The neutrons captured by the dysprosium foil will generate radioactivity that subsequently decays with a convenient half-life,

$$^{164}_{66}\mathrm{Dy} + ^{1}_{0}\mathrm{n} \rightarrow ^{165}_{66}\mathrm{Dy} \xrightarrow[2.3\mathrm{h}]{\beta^-;\gamma} ^{165}_{67}\mathrm{Ho} \tag{1}$$

The pattern of radioactivity is transferred to the X-ray film by simply placing the activated metal foil in close contact to the X-ray film. The X-ray film is then irradiated (β and γ radiation) for several hours and thereafter developed using a standard photographic technique. The β-particles radiated from the foil surface are the main source for exposing the X-ray film. The β-particles take short and straight paths on their way to the X-ray film and so give good resolution. The γ-radiation will blur the X-ray film and it's therefore undesirable.

2.4. Cellulose nitrate film technique with track-etch recorder

The film consists of a 100 μm thick plastic layer substrate of lightly rose-tinted cellulose nitrate material. The film is primarily intended for recording the emission from α-particle sources (below 4 MeV). The film is therefore coated with a neutron to α-particle converter material for neutron radiography utilization. The record thus obtained is a neutron radiographic image formed by means of the converter material. Transmitted neutrons will reach the vacuum deposited lithium tetra borate layer or converter screen that