Chapter 9
Neutron Imaging for Nuclear Power Industry

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9.1 Introduction

The ever increasing demand for electric energy owing to industrialization and growing economy necessitates expansion in nuclear power production throughout the world. To increase the safety and improve the efficiency of nuclear power, research and development in this field are constantly progressing. The next-generation reactors are focussed on better fuel utilization, proliferation resistance and enhanced safety. This requires better materials that can sustain these temperatures and increased radiation damage. The components related to operating reactors also require periodic examinations to ensure safe working conditions. Apart from that, the issue of waste disposal in a safe manner is also a challenge. Neutrons being unique probes offer assistance in a wide range of problems associated with reactors. With increased flux at modern neutron sources together with advanced sample environments, it becomes possible to conduct studies of reaction kinetics at reactor operating temperatures. The data generated using neutrons facilitate benchmarking simulations and modelling of crystal structure evolution and thermo-mechanical treatment. Development of advanced neutron detection methods widened the scope of neutron imaging in nuclear industry especially for nuclear materials, where heavy elements (e.g. uranium) are to be imaged together with light elements (e.g. hydrogen, oxygen).

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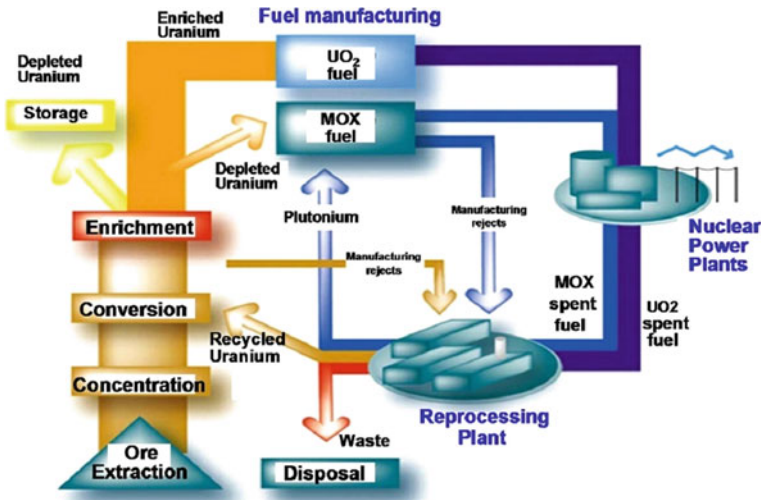e-mail: shefali@barc.gov.in

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Fig. 9.1 Schematic of nuclear fuel cycle [1]

The nuclear fuel cycle begins with the preparation of fuel, the service period, i.e. the time during which the fuel is being used in the reactor for electricity generation and ends with the management of the spent fuel which can either be reprocessed and reused or diposed off.

Figure 9.1 shows a schematic of a general nuclear fuel cycle. Various stages involved from ore extraction to its disposal have been shown. Nondestructive testing (NDT) is mandatory at every step of nuclear fuel cycle to maintain the safe working and operation of the plants and facilities. Several techniques like radiography (neutron and X-ray), ultrasonic testing, eddy current and magnetic testing have been routinely used for NDT. Of these X-ray and neutron radiography are being explored in the nuclear field providing complementary information. Both these techniques form an indispensable part of NDT studies. Neutron imaging finds immense applications in the nuclear industry; ranging from testing of new fuels and reactor materials to investigation of irradiated failed components to find out the root cause of damage. In the subsequent subsections, we discuss the application of neutron imaging for hydrogen embrittlement studies, postirradiation investigations and testing of new fuels.

9.1.1 Neutron Imaging for Fuel Enrichment Studies

U-235 is the only nuclide existing in nature (in any appreciable amount) which is fissile with thermal neutrons (OECD Nuclear Energy Agency, 2003). Natural uranium has 99.284% U-238 isotope, with the U-235 isotope constituting only about 0.711%. Uranium enrichment is required for light water reactors (LWRs) and for

other reactors. Neutron radiography can be used for monitoring of isotopic enrichment in fuel pellets loaded into a fuel pin [2] through attenuation comparison. Frajtag [2] conducted neutron radiography studies on fresh fuel pellets in a fuel pin. With neutron radiography, varying degrees of enrichment also could be studied.

Plutonium recycling in existing thermal reactors requires design of fuel assemblies containing pellets of different enrichments at different positions. A final check of the loading pattern is required to ensure the quality assurance of these fuels. In 1983, Gosh et al. [3] used neutron radiography to monitor plutonium enrichment in mixed oxide fuel pellets inside fuel pins.

Tremsin et al. [4] used neutron resonance absorption imaging and transmission Bragg edge imaging for fuel pellets study at LANSCE pulsed moderated neutron source of Los Alamos National Laboratory. The significant difference in the absorption cross sections of U-235 and U-238 isotopes was used, and very accurate nondestructive spatial mapping of the enrichment level of fuel pellets was carried out. The distribution of other isotopes in the spent fuel elements (Nd, Gd, Pu, etc.) was also demonstrated. This information can then be used for the investigation of fuel burn up rates for fuel elements placed at different rod positions in the reactor core. Tremsin concluded that the contrast between the pellets of different densities depends strongly on the range of neutron energies used. The more thermal part of the beam spectrum reveals the pellet with the lowest density as an object with the highest transmission whilst the coldest part of the neutron spectrum shows the opposite (Ref. Fig. 9.2).

9.1.2 Quality Assurance of Fabricated Fuels

As a result of the increasing demand for nuclear power, there has been an emphasis on the reliability of nuclear fuel performance for safety considerations. Fuel assemblies must withstand the operational requirements of the reactor and the irradiated fuels need to be stored in water basins for prolonged periods before reprocessing or disposal. One of the keys to consistent and reliable fuel production is the quality assurance of fresh fuels. Neutrons being transparent to metals can be used to image voids, inclusions, cracks or any other defects in freshly fabricated fuels. One of the first applications of neutron radiography was nuclear fuel characterization in the early 1960s using the film technique. Since then, neutron radiography has frequently been used as a quality control measure for fresh fuels. The fuel pin of PHWR type reactors contains natural uranium dioxide in the pellet form encased in a zircaloy cladding tube and sealed at both ends by welding of end plugs. Figure 9.3 shows the neutron radiography studies conducted on a fresh PHWR fuel pin. The radiography image shows some inclusion in the cladding material. For a detailed examination, tomography studies were conducted. The reconstructed image as well as a slice showing the flaw is shown. The study was carried out at neutron imaging beamline at Dhruva [5].

Neutron imaging can be used for studying any inclusions in fuel assemblies. Figure 9.4 shows cadmium and wax inclusions in PHWR and BWR fuel pin [6].
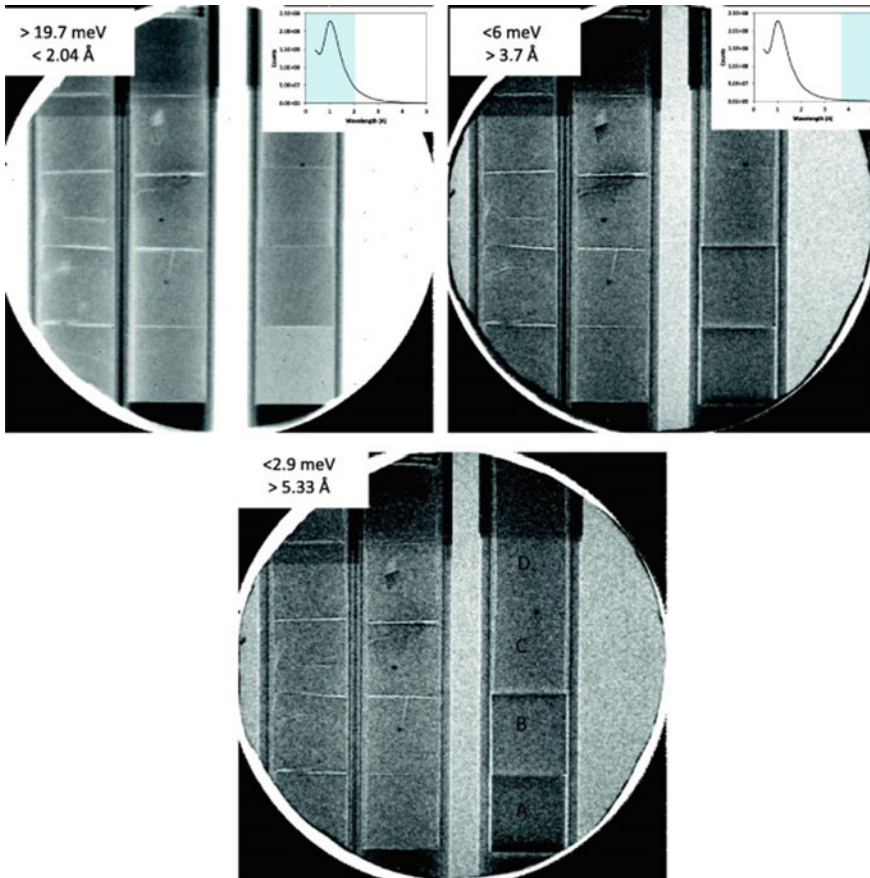
Fig. 9.2 Thermal neutron transmission radiographs of fuel pellets. Different energy ranges highlight the lowest(>19.7 meV) and highest(<6 meV) density object as per the transmission [4]. {Reproduced with permission from Elsevier Copyright Clearance Centre}

The images have been taken by the transfer method. The zirconium hydride formed is also visible owing to the difference in the cross section between zirconium and zirconium hydride. A typical experimental nuclear fuel pin consists of a thin-walled cladding tube filled with MOX(uranium, plutonium and thorium oxides) fuel pellets and hermetically sealed at both ends. The fuel pins are subjected to irradiation testing in research rectors and thus serve the purpose of studying fuel performance. Enriched uranium-aluminium alloy fuel sandwiched between aluminium-clad plates and roll-bonded into fuel plates are used as a core in small research reactors employed for physics research and applications like neutron radiography [7]. To reduce the centre temperature of the fuel pellet during irradiation and thereby reduce the pellet clad mechanical interaction, annular pellets (pellets with a central void) or pellets with

Fig. 9.3 Neuton radiograph
of fresh PHWR fuel pin
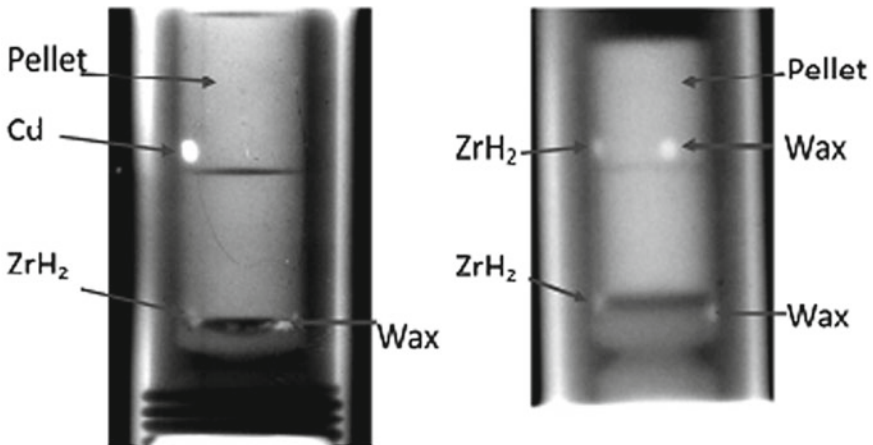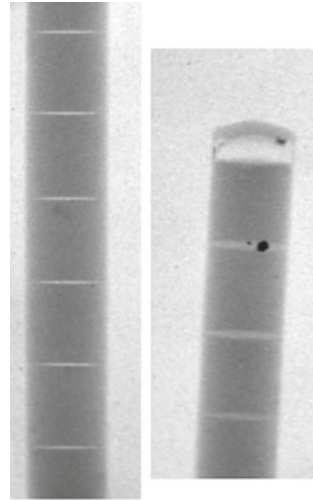showing fuel pellets and an
inclusion

Fig. 9.4 Neutron radiography conducted on fresh PHWR and BWR [6]

depleted UO2 central core are being considered in BWR fuel pins. The deep penetration of neutrons in dense and high Z material is utilized to check the integrity of fuel pellets and also identify the anuular pellets. Water ingress inside a fuel pin because of a breach in the clad can be seen in the neutron radiograph (Ref. Fig. 9.5).

Neutron radiography unlike X-ray can be used for discriminating nearby elements. This unique capability can be utilized to identify UO_2 and PuO_2 -UO_2 mixed oxide fuel pellets inside sealed experimental fuel pins. Whilst mixing of PuO_2 with UO_2 for making MOX pellets, there are chances of PuO_2 agglomerates being present as
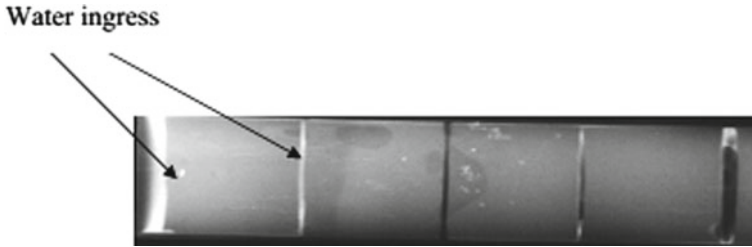
Fig. 9.5 Water ingress inside a PHWR type fuel pin because of a breach in the clad [7]
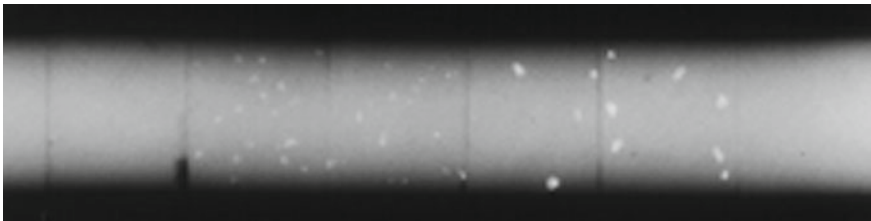

Fig. 9.6 Neutron radiography of plutonium oxide agglomerates inside MOX fuel pellets [7]

inclusions in the MOX matrix. Neutron radiography can be used to check homogeneity of this mixing. Figure 9.6 reveals PuO_2 agglomerates in MOX matrix in the neutron radiograph.

9.1.3 Testing of New Materials for Reactors

The future of nuclear energy depends not only on the availability of nuclear fuels but also on the development of advanced structural and functional materials. These materials are to be used in extreme environments like high temperature/pressure, intense neutron irradiation, presence of magnetic/electric fields, corrosive agents and all that in combination with complex loading states and cyclic operation histories. Further, developments in advanced materials for the existing thermal reactors may improve safety margins, reduce the frequency of component replacement and, ensure higher burn ups thereby improving fuel cycle economics and safety. For advanced future reactors (e.g. fast reactors, high-temperature reactors, fusion reactors, accelerated driven systems), the development of advanced materials is crucial due to the extreme environment and operating conditions associated with these innovative systems [8]. Nuclear technologies rely on the availability of advanced fuel and structural materials. These structural materials should be able to resist extreme conditions like high neutron, proton and gamma fluxes and fluencies, high temperatures, or corrosive and abrasive media and combinations of them. In particular, new concepts for nuclear energy production like fusion or next-generation fission reactors require materials

suitable for the application at temperatures up to 1000 °C under strong irradiation resulting in atomic displacements up to 120 displacement per atom (dpa). The very high safety standards valid for nuclear installations involve excluding material failure under all real operating or hypothetical incident accident conditions. To predict the material behaviour even at the end of the usage, the structural processes occurring must be understood. These processes can result in the degradation of the mechanical properties. Examples are thermal ageing, irradiation and hydrogen embrittlement, formation or relaxation of residual stresses, corrosion and erosion or material fatigue. In the development phase of new reactor concepts, the material behaviour during operation and transient conditions has to be understood, often by extrapolation of the known behaviour.

For irradiated materials where the background may be high because of the activation of the materials, neutron studies can be conducted. SINQ is the Swiss neutron source at the Paul Scherrer Institute (PSI) for research purposes, based on spallation in a lead target irradiated by 590 meV proton beam. The target rod consists of lead in a zircaloy tube. Since some of the lower central rods are regularly heated by the proton beam to beyond the melting temperature of lead (327 °C) with the corresponding volume expansion, the tubes have a lead filling of only 90%. Lehmann et al. [9] and the group conducted studies to investigate the targets (virgin and irradited) to be used in SINQ at NEURAP beamline. Figure 9.7 shows radiography for a virgin rod (top) and of an irradiated rod (middle and bottom). This way, an empty horizontal gap is formed in the top region of each rod. Studies were also conducted to see the effect of irradiation on target rods and it was observed that spallation products are formed and also because of cyclic melting and solidification, redistribution of lead also takes place.
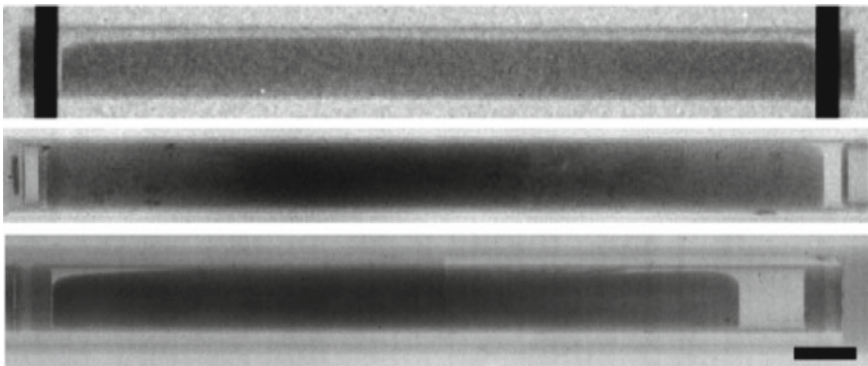

Fig. 9.7 Neutron radiography of a virgin SINQ target rod (**top**); Neutron radiography of a target rod after long irradiation (**middle**); Neutron radiography depicting the change in Pb distribution due to irradiation during operation (**bottom**) [9]

9.1.4 Neutron Radiography for Nuclear Waste Disposal

Nuclear power is said to be the solution of the energy crisis around the world. It can produce a large amount of energy from relatively small amount of fuel and also the waste generated in the whole process is small. The problem lies in the fact that almost all the waste generated is radioactive and thus requires careful management. Radioactive waste is generated not only in the nuclear fuel cycle but in many other sectors like agriculture, research, minerals exploration, medicine, etc., which use radioisotopes or radioactive particles. The only respite is that the level of hazard of all radioactive waste decreases with time. So the storage of radioactive waste is a crucial aspect of its management cycle. The waste must be stored such that it can be retrieved whenever required, and at the same time, it is completely isolated from the environment. The materials used for storing and transporting nuclear or radioactive waste must be fool-proof to ensure no leakage of radioactivity as it can be hazardous. Nondestructive testing of the casks and packing materials ensures their integrity. Nuclear materials are usually transported in packages which should ensure stringent safety standards like impact resistance, thermal shock, etc. Aluminium foam plays a major role as cushioning material in transport and prevents damage from shock or impact. The advantage of such faoam structure is that it can have the strength of the metal but with comparatively less weight. Neutron radiography allows us to image the entire bulk and calculate pore size pore fraction which can be used for modelling the structure. Figure 9.8 shows the 3D tomogram generated at Dhruva neutron imaging beamline [5].

The nuclear or radioactive waste which cannot be reprocessed immediately or which has to be dumped in radiological repositories needs to be packed in drums.
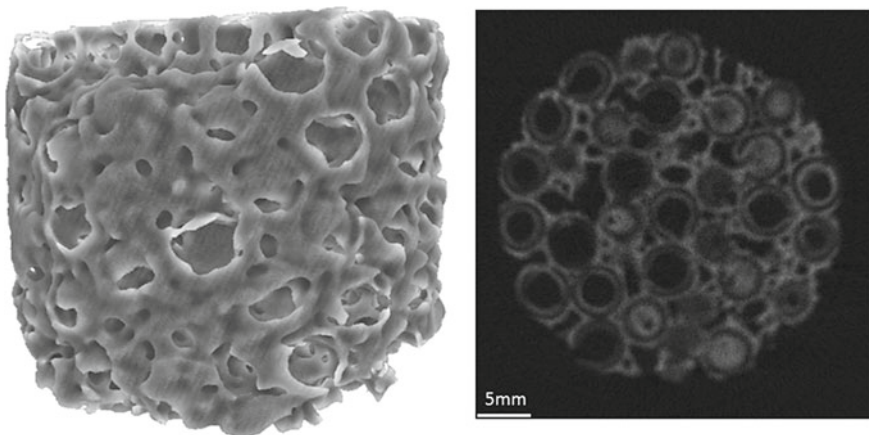

Fig. 9.8 3D reconstruction of aluminium foam used as cushion for nuclear material transport packages. The right picture shows a slice from the reconstructed tomogram. Pore size and density can be calculated from the images [5]. {Reproduced with permission from Elsevier Copyright Clearance Centre}

There are many active and passive methods that are being routinely used for drum scanning. They include segmented gamma ray scanning [10], gamma-transmission measurements (radiography and tomography using radioactive sources [11] or accelerator [12]), neutron emission counting (with time correlation analysis to distinguish between neutrons originating from spontaneous fission or (alpha, n) reactions, respectively, [13]) and neutron interrogation techniques (to induce fission events) [14]. Recently, neutron radiography studies have also been attempted for nondestructive analysis of the radioactive waste packages. Fission neutron radiography of 200 L waste drums was demonstrated first time at the NECTAR facility by Bucheral and coworkers [15]. Figure 9.9 shows the tomographic investigations of 200 L and 280 L mock-up waste drums using fission neutrons and cobalt-60 source.

Cementitious material is the most common medium used for safely encasing and disposing of low and intermediate-level radioactive waste. It acts as a barrier to prevent water from contacting the radionuclides and as a cover that prevents the migration of radionuclides. Both these processes depend basically on limiting water transport through the cementitious layer, thereby defining the performance of the cementitious waste form. Neutrons can be an excellent probe when studying water transport especially in high Z materials like cement. Mcglinn [16] studied water transmission through two types one unleached and the other leached for 8 years. Results for the unleached sample are shown in Fig. 9.10.
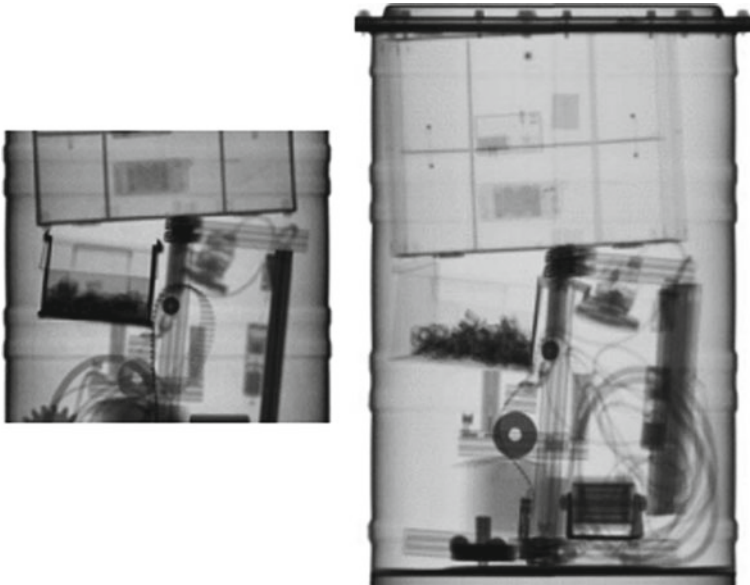

Fig. 9.9 Radiography of a mock-up waste drum using fission neutrons (**left**) and 60Co source (**right**) [15]. {Reproduced with permission from Elsevier Copyright Clearance Centre}
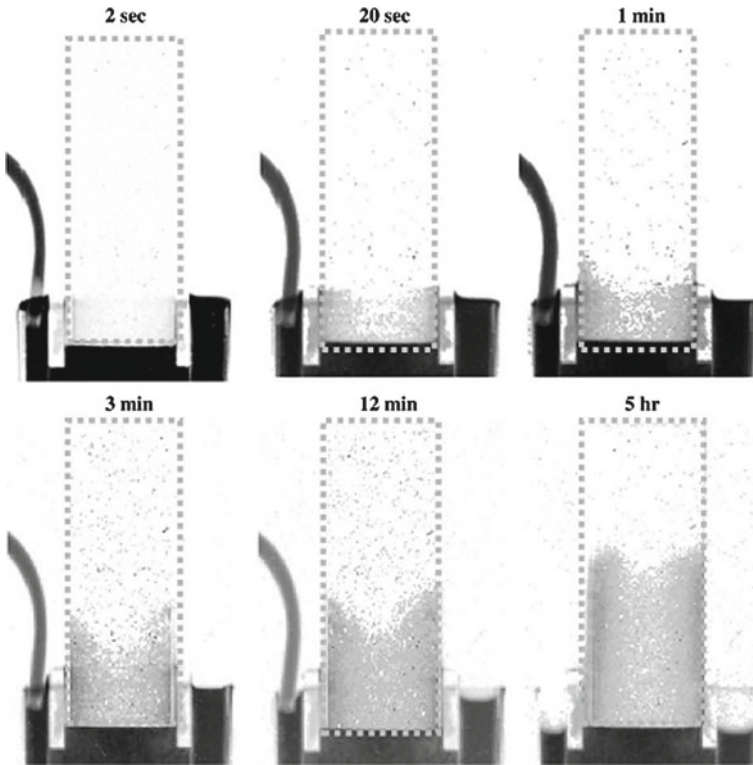
Fig. 9.10 Neutron radiographs taken at differents instant of time to show the movement of water front [16]. {Reproduced with permission from Elsevier Copyright Clearance Centre}

9.1.5 Postirradiation Studies

For the nuclear fuel development programme information about the fuel performance at various stages—pre-irradiated, interim and postirradiated is extremely important. Postirradiation examinations provide valuable information like fuel cracking, swelling, melting, fissile material redistribution, etc. Postirradiated samples cannot be examined using conventional techniques because of their inherent high radiation. However, neutron radiography can help qualitative as well as quantitative information about the fuel and associated structural materials. This feedback helps in improving fuel design. Neutron imaging plays a valuable role in postirradiation examination as it is a nondestructive method and can be used even for highly radioactive samples. Indirect neutron radiography used for such samples is a two-step process involving building radioactivity on a neutron-absorbing screen followed by exposing a film in contact with this screen to generate the image of the test sample.

One of the most common forms of degradation found in metals is hydrogen embrittlement. It is a metal's loss of ductility and reduction of load-bearing capability due to the absorption of hydrogen atoms or molecules by the metal resulting in reduced strength of the material. When talking about the nuclear power industry, the first and foremost material that comes into mind is zirconium. Zirconium alloys are frequently used at nuclear structural materials for water reactors because they low thermal neutron capture cross section, excellent mechanical properties along with possess good resistance to corrosion in high-temperature water [17, 18]. They, however, like other metals are susceptible to hydrogen induced damage: hydrogen embrittlement [19, 20]. It could be manifested in two major forms, namely delayed hydride cracking and hydride blisters. Therefore hydrogen estimation is, necessary to predict the life of in-service components. Though there are several techniques available for the same, because of hydrogen atom's high absorption cross for thermal neutrons, neutron radiography has the advantage of being a nondestructive technique providing bulk information within a very short duration. Once calibrated using standard samples of known hydrogen content, neutron radiography can estimate the exact amount of hydrogen in any sample within experimental errors. Figure 9.11 shows the attenuation coefficient as a function of neutron wavelength. The utility of neutron radiography for studying hydrides is quite evident because of the huge difference between zirconium and its hydride [21].

Figure 9.12 shows the tomography studies on zirconium alloy coupons precharged with a known amount of hydrogen carried out at CIRUS imaging beamline, BARC by Ashish et al. [22]. Calibrating the setup with a known amount of hydrogen
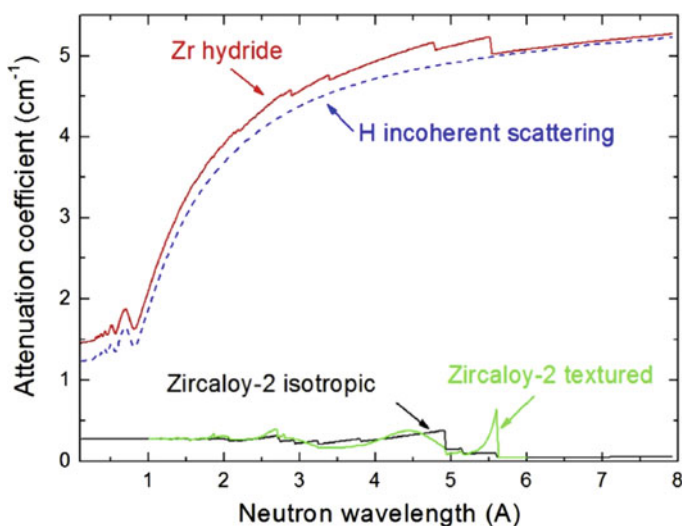

Fig. 9.11 Neutron attenuation vs neuton wavelength graph for zirconium and its hydride. The difference in mean attenuation coefficient of zirconium alloy versus hydride is quite evident [21]. {Reproduced with permission from Elsevier Copyright Clearance Centre}
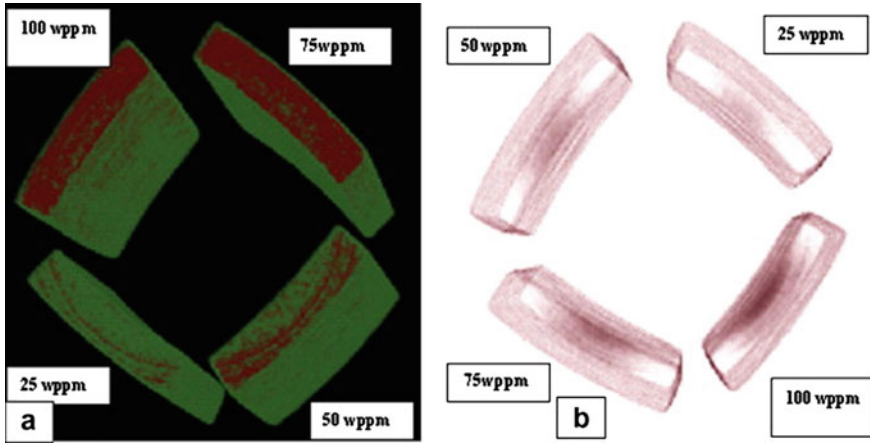
Fig. 9.12 Neutron tomography results on zirconium alloy pre-charged coupons. The green part is the base matrix and red indicates the amount of hydrogen present [22]. {Reproduced with permission from Elsevier Copyright Clearance Centre}

pre-charged coupons, it was also attempted to measure the hydrogen content of an unknown sample.

Figure 9.13 shows the neutron radiography images of the failed fuel element. Studies were carried out to understand the reason for rupture. Hydrogen embrittlement in metals particularly zirconium alloys manifests as blistering and delayed hydride cracking [23–25]. These two processes can occur even if the total hydrogen content in the pressure tube is less than the critical value. This occurs because the hydrogen which enters the zirconium matrix is highly mobile and is quick to respond to any concentration, stress or temperature gradients.
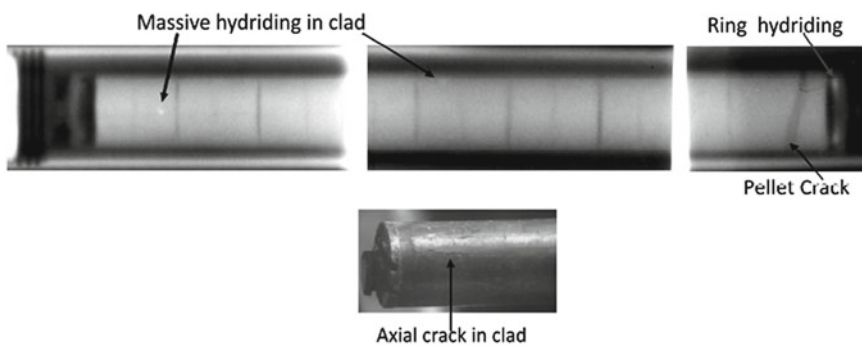

Fig. 9.13 Radiograph showing metal hydriding in fuel. The metal hydride has attenuation cross section 16 times the matrix hence is clearly visible in the radiography [7]

Hydrogen diffusion studies in various zirconium alloys have been attempted by Grosse and Buitrago [21] using neutron radiography. Shukla [26] conducted systematic studies for measuring diffusivity parameters for Zr-2.5%Nb alloy used in Indian PHWRs as pressure tube material. Figure 9.14 shows the neutron radiography and Fig. 9.15 shows hydrogen depth profiles obtained.

Hydrogen present in the zirconium pressure tube as a result of downside corrosion migrates towards regions of higher stress and gets precipitated making that specific
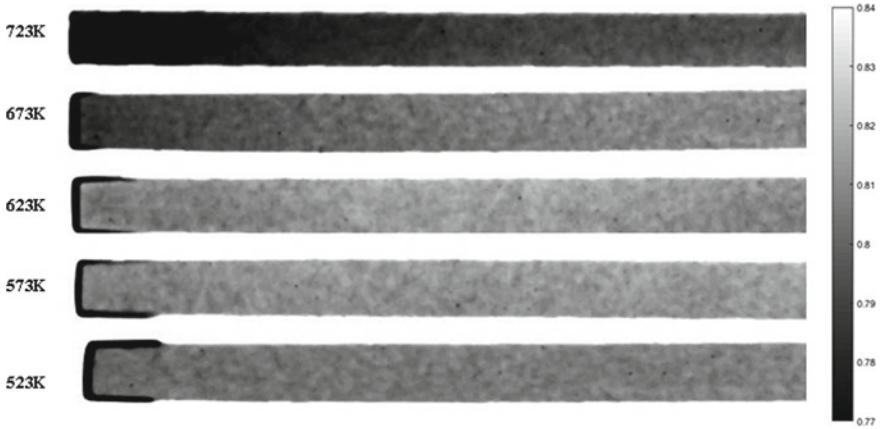

Fig. 9.14 Radiograph showing depth profile of hydrogen in samples. The hydride layer on one extreme end denotes the source of hydrogen [26]. {Reproduced with permission from Elsevier Copyright Clearance Centre}
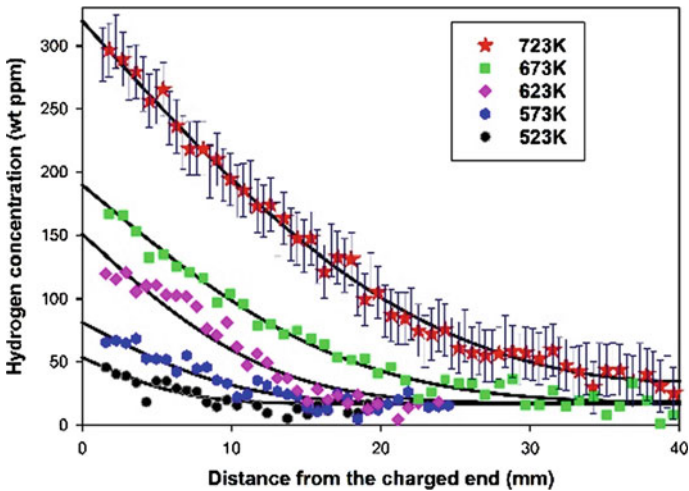

Fig. 9.15 Hydrogen concentration change with depth data is fitted to obtain diffusivity values [26]. {Reproduced with permission from Elsevier Copyright Clearance Centre}
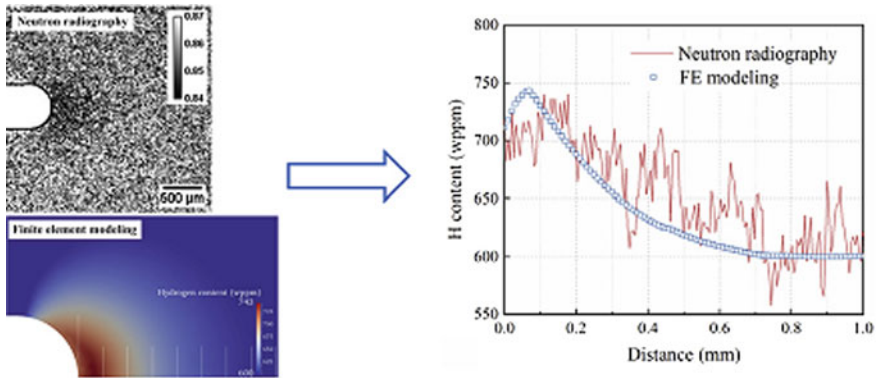
Fig. 9.16 High-resolution radiography showing the hydrogen concentration near a notch [27].
{Reproduced with permission from Elsevier Copyright Clearance Centre}

area brittle. This phenomenon is known as delayed hydride cracking (DHC) and is the
major cause of failure of pressure tubes. Studies have been carried out to understand
the kinetics of hydrogen movement under stress gradients. Gong [27] carried out
hydrogen diffusion studies in Zircaloy nuclear fuel cladding materials under stress
using high-resolution (10 μm) neutron radiography. The results obtained were in
agreement with finite element modelling. Figure 9.16 shows the high-resolution
neutron radiography results and the FEM results for comparison.

Whenever a temperature gradient occurs, the hydrogen migration leads to the
formation of blisters. Agarwal [22] studied blistering in a pressure tube spool and
PHWR fuel pin. Figure 9.17 shows the data which neutron tomography studies have
obtained.

Hydrogen-assisted cracking in electrochemically hydrogen-charged technical iron
and supermartensitic steel was studied using neutron tomography. Griesche [28]
carried out blister-related studies on technical iron and developed a 3D model.
Hydrogen accumalations around cracks and hydrogen gas trapped in cracks could
be visualized through neutron tomography. Figure 9.18 shows the reconstructed 3D
model of the hydrogen-charged technical iron sample.

Figure 9.19 shows the hydrogen-assisted cracking results obtained in super
martensitic steel. Dog bone shaped samples pre-charged with hydrogen were exposed
to constant tensile load with subsequent sample rupture. It was observed that the
tensile sample ruptured at a load of 2/3 of the theoretical tensile strength value for
non-hydrogenous material. An apparent degradation in the mechanical property had
occurred and was proved by this experiment.

Nuclear fission has long been explored and widely used in reactors worldwide.
The efficiency of nuclear fission is low compared to the energy produced if we opt for
nuclear fusion. Also the problem of spent fuel storage and disposal can be tackled.
The fission–fusion hybrid energy system may be a practical approach in attaining
sustainable development of nuclear energy. Metallic fuel used in subcritical blankets
can be promising candidates for a fusion-fission hybrid energy system. U–Zr alloy
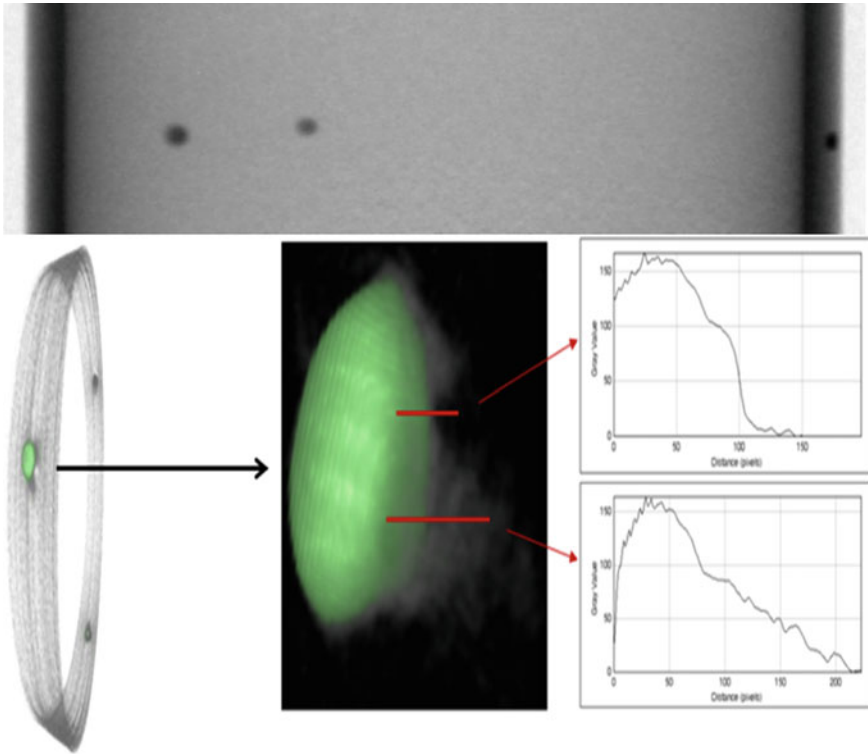
Fig. 9.17 Hydride blister in PHWR pressure tube [22]. {Reproduced with permission from Elsevier Copyright Clearance Centre}

which is used as blanket material might become the most potential material for metallic fuel as it can withstand high temperature, high pressure and radioactive environment. Nondestructive testing, however, is required to ensure reliability. Since the irradiated U–Zr alloy fuel is highly radioactive routine methods of inspection cannot be used. Indirect neutron radiography is used for the same. Yong Sun [29] and group carried out neutron radiography studies to obtain the inner information of irradiated U–10Zr alloy. They studied the swelling behaviour of irradiated U–10Zr alloy fuel at various burn ups using indirect neutron radiography. It was demonstrated that the swelling rate increases with an increase in burn up. Figure 9.20 shows the neutron radiography result for a particular sample, the metallic alloy enclosed in an aluminium powder shell is shown. Swelling is quite evident at position A as shown in Fig. 9.20. Figure 9.20b shows that the grey value on line A is different from B and C indicating some leakage, and uranium has flown out into the aluminium powder shell.
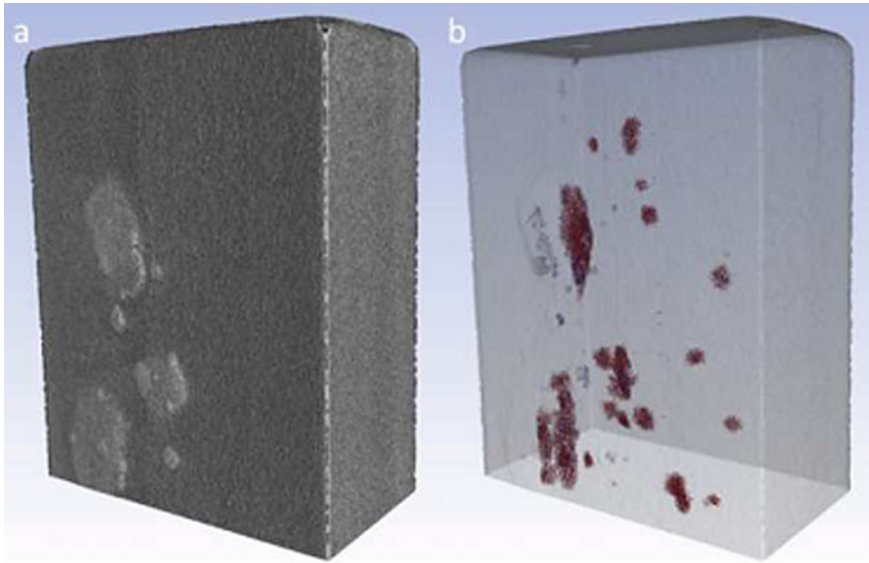
Fig. 9.18 Inclined view on the reconstructed 3D model of a hydrogen-charged iron sample. The rendered surface with blisters is shown, the cracks are shown in blue and the trapped hydrogen in red [28]. {Reproduced with permission from Elsevier Copyright Clearance Centre}
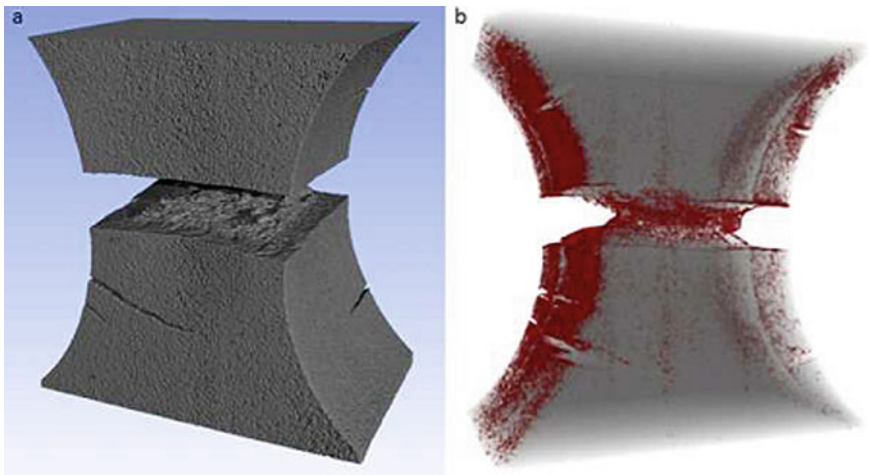

Fig. 9.19 Inclined view on the reconstructed 3D model of a part of a hydrogen-charged super martensitic tensile sample. **a** and **b** depict the rendered surface and the hydrogen distribution in red, respectively [28]. {Reproduced with permission from Elsevier Copyright Clearance Centre}
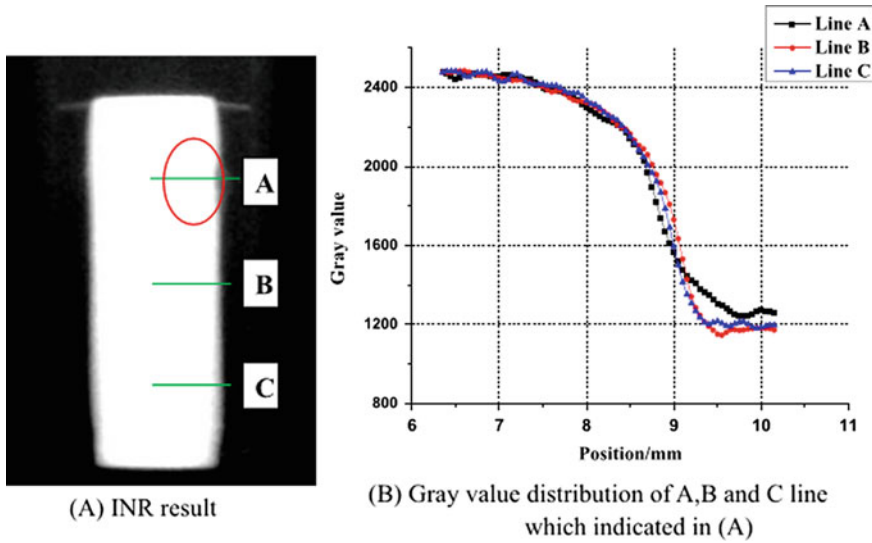
(A) INR result

(B) Gray value distribution of A,B and C line which indicated in (A)

Fig. 9.20 Indirect neutron radiography (INR) result and grey value distribution of A B C line indicated in (left figure) of sample [29]. {Reproduced with permission from Elsevier Copyright Clearance Centre}

9.2 Summary

This chapter deals with one of the most important applications of neutron imaging: the nuclear industry. Neutron radiography and tomography are routinely used to investigate of fresh and irradiated fuels. It also serves the purpose of testing new fuel compositions and designs. Indirect neutron radiography provides the answers especially for irradiated samples where most of the techniques fail to provide information because of the high gamma background. The materials used for reactor structure, fuel and coolant clad need to withstand extreme conditions. Neutron imaging allows the testing and development of such materials. Hydrogen embrittlement is a major issue for all structural materials particularly zirconium and iron. Being sensitive to hydrogen, neutrons can be used to evaluate hydrogen content in different metals as well as to study its diffusion parameters under stress temperature and concentration gradients. Neutron imaging, thus, finds numerous applications linked to the nuclear power industry. This chapter attempts to give a glimpse of it.

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