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Applications of neutron radiography for the nuclear power industry

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Abstract

The World Conference on Neutron Radiography (WCNR) and International Topical Meeting on Neutron Radiography (ITMNR) series have been running over 35 years. The most recent event, ITMNR-8, focused on industrial applications and was the first time this series was hosted in China. In China, more than twenty new nuclear power plants are under construction and plans have been announced to increase the nuclear capacity by a factor of three within fifteen years. There are additional prospects in many other nations. Neutron tests were vital during previous developments of materials and components for nuclear power applications, as reported in the WCNR and ITMNR conference series. For example a majority of the 140 papers in the Proceedings of the First WCNR are for the benefit of the nuclear power industry. Many of those techniques are being utilized and advanced to the present time. Neutron radiography of irradiated nuclear fuel provides more comprehensive information about the internal condition of irradiated nuclear fuel than any other non-destructive technique to date. Applications include examination of nuclear waste, nuclear fuels, cladding, control elements, and other critical components. In this paper, applications of neutron radiography techniques developed and applied internationally for the nuclear power industry since the earliest years are reviewed, and the question is asked whether neutron test techniques, in general, can be of value in development of the present and future generations of nuclear power plants world-wide.

Peer-review under responsibility of the organizing committee of ITMNR-8.

Keywords: neutron radiography, post-irradiation examination, nuclear fuel

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1. Introduction

Understanding the behavior of nuclear fuels during irradiation and under transient conditions contributes to development of safer and more-efficient nuclear reactors. Neutron radiography (NR) of irradiated nuclear fuel and other critical components provides more comprehensive information about the internal condition of irradiated nuclear fuel than any other non-destructive technique to date. This international conference series, including the World Conference on Neutron Radiography (WCNR) and International Topical Meeting on Neutron Radiography (ITMNR), has been running over 35 years. Examinations using NR were vital during previous developments of materials and components for nuclear power applications. The value of neutron radiography in development of reactor materials and components was evident in the significant number of publications on the topic in the 1970s and 1980s. For example, a majority of the 140 papers in the Proceedings of the First WCNR (Barton and von der Hardt, 1981) are for the commercial nuclear power industry. Included in those proceedings are reviews of the diverse techniques being applied in Europe (Domanus and von der Hardt, 1981), Japan (Kobayashi et al., 1981), the United States (DeVolpi, 1981), and at many other centers. However, the number of publications in the WCNR and ITMNR conference series on nuclear power applications waned through the 1990s, with only a modest resurgence since the mid-2000s.

There are a number of review papers discussing the application of neutron examination techniques to nuclear applications (Ross, 1977; Ashraf and Kahn, 1992; Vogel, 2013; Parker and Joyce, 2015; International Atomic Energy Agency, 2015; de Beer, 2015) and NR in general (Brenizer, 2013). In this paper, the techniques developed and applied internationally for the nuclear power industry since the earliest years are reviewed briefly, and particular emphasis is given to evaluate trends and lessons learned. Drawing from these perspectives, the question is asked to what extent neutron examination techniques will continue being of value in development of the present and future generations of nuclear power plants worldwide. Neutron radiography plays an important role in development of safer and more efficient nuclear power plants by providing valuable information for development of new fuels, cladding, and other reactor materials, which is discussed in the following section.

2. Nuclear Applications of Neutron Examinations

The earliest neutron radiographs were produced in 1935 by Kallmann, with only modest progress through the 1950s, and only four NR programs worldwide in 1964 (Brenizer, 2013). A facility specifically built for NR at the 200 kW Juggernaut reactor provided the first application of NR to study irradiated nuclear fuel (Berger and Beck, 1963). Fifteen years later, by 1977, there were over sixteen facilities in Europe dedicated to NR for irradiated nuclear fuel, and a review paper cites 154 references to related work worldwide (Ross, 1977).

Neutron radiography of irradiated nuclear fuel and other critical components provides more comprehensive information about the internal condition of irradiated nuclear fuel than any other non-destructive technique to date. Neutron examination techniques have been applied to study fuel swelling and cracking (Chichester and Harp, 2016; Sun et al., 2016), pellet-clad interactions, hydridization of Zircaloy cladding (Yasuda et al., 2002; Grosse et al., 2011; Agrawal et al., 2012), cladding microstructural properties (Kruželová et al., 2012), fuel pellet fragmentation during transient conditions (Richards et al., 1982; Grosse et al., 2012; Jenssen et al., 2014), central void formation in fuel, burn-up and enrichment variations, pellet-clad and pellet-pellet gaps (Notea et al., 1983; Sim et al., 2013), evidence of fuel melting, and migration of material (Rice et al., 2015). Neutron radiography also provides post-irradiation analysis of highly-radioactive lead target rods from proton-accelerator neutron sources (Vontobel et al., 2006). Many of the same radioactive material inspection techniques developed in the early years of NR are still being utilized and advanced to the present time (Jenssen and Oberländer, 2002; Vontobel et al., 2006; Craft et al., 2015). Applications of neutron examination techniques to nuclear waste containers (Hausladen et al., 2007; McGlinn et al., 2010; Bücherl et al., 2017) will become increasingly important as nuclear waste storage concerns continue to grow. Additionally, NR enables analysis of two-phase flow experiments that provide insights into the coolant behavior of boiling water reactors and provides data for validation of computational models (Takenaka et al., 1996).

Neutron scattering applications to nuclear materials is probably one of the most promising fields of neutron examination techniques for nuclear materials in the future, as is indicated by the more than 200 references in recent reviews (Vogel, 2013; International Atomic Energy Agency, 2015). Radiation scattering techniques provide insights into materials on the microstructural scale, providing material scientists with important information for engineering materials. Additionally, the non-destructive nature of neutron diffraction enables in-situ testing, which provides unique examination capabilities for researchers.

3. Methods for Imaging in Neutron Radiography of Radioactive Materials

From the 1990's advances in computer processors, memory storage, and imaging sensors initiated rapid expansion of neutron imaging capabilities (Brenizer, 2013). The development of CCD-based low-light cameras in combination with neutron-sensitive scintillator screens, fast data processing and memory storage has enabled real-time NR systems that have become ubiquitous within the worldwide neutron radiography community. These systems are relatively simple and inexpensive to develop, requiring only a light-tight enclosure, a scintillator screen, a low-light CCD camera, a lens and mirror, and local radiation shielding. The combination of a narrow-pulse neutron source and ultrafast time-resolved neutron detection, now possible using micro-channel plate (MCP) detectors, has enabled examination of neutron resonances using time of flight methods (Tremsin et al., 2013).

While new digital NR systems continue to develop, methods for highly-radioactive objects have remained relatively stagnant. Some facilities still use activation foils in combination with film, the same method used in the earliest days of neutron radiography. The track-etch imaging using nitrocellulose is also available (Markgraf, 1990; Jenssen et al. 2014). The major development for NR of highly-radioactive objects in recent decades is the use of photostimulable phosphor image plates, which are also sensitive to the beta decay radiation emitted from activated foils. Most facilities performing these examinations today still use the transfer method, but in combination with image plates rather than film (Wei et al., 2014; Craft et al., 2017). The most advanced of any of these techniques is the use of image plates with dysprosium oxide doped directly into the phosphor (Vontobel et al., 2006). Unfortunately, these image plates were not produced on a production scale and are not commercially available. Future development and production of neutron-sensitive image plates doped with delayed-decaying isotopes (e.g. dysprosium and indium) is one potential area for improvement of radiographic imaging techniques for highly-radioactive objects.

4. Facilities for Neutron Radiography of Radioactive Materials

Handling radioactive materials requires special infrastructure and personnel support that are not available at all neutron beamline facilities. Facilities intended for evaluation of radioactive objects are specifically designed to accommodate such materials. However, retrofitting an existing beamline facility to accommodate radioactive materials is possible. Based on extensive literature review, there are three approaches for neutron beam examination of radioactive specimens, including:

1) A cask transfer system that couples to a radiation-shielded bunker also allows a radioactive sample to be introduced into the beam. This is by far the most common approach. (e.g. Mayer, 1972; Korneev et al., 1983; Lehmann et al., 2003; Sim et al., 2004; Singh, 2011; Wei et al., 2013; Jennsen et al., 2014)
2) A neutron beamline located adjacent to a hot-cell facility to allow radioactive samples to be introduced into a radiation-shielded neutron beam bunker. (e.g. Manoharan, 2012; Craft et al., 2015)
3) A neutron beamline facility located underwater at a pool-type reactor. The water provides the necessary radiation shielding. (Barton and Perves, 1966; Farny and Houelle, 1971; Parrat et al., 2013)

There are many NR facilities reported in WCNR-1 (Barton and von der Hardt, 1981) designed to examine irradiated nuclear fuel, most of which used a shielded cask to transfer the specimens. There is a striking contrast between the large number of NR facilities available for radioactive nuclear fuel evaluations prior to 1990 and the relative small number available in 2017. For example, in 1990, there were at least sixteen NR facilities in Europe dedicated to nuclear fuel materials science. These were eleven cask type NR facilities and five pool type NR facilities as follows: Mol, Belgium (2); Geestacht, Germany (2); Riso, Denmark (1); Cadarache, Grenoble, Saclay and Valduc, France (6); Harwell, UK (1); Cassaclia, Italy (1); Petten, Netherlands (2) and Kjeller, Norway (1). (Barton and von de Hardt 1981; Barton et al. 1987; Domanus 1992). Of the facilities reported on in WCNR-1 designed to examine irradiated nuclear fuel, the facilities that continue to perform such examinations today are located at research centers that produce and examine irradiated fuel specimens as part of fuel development efforts. These centers are few in number, which partially explains the limited use of NR for this application. The facilities presently available (2017) for NR of radioactive materials include:

1) China Mianyang Research Reactor (CMRR) located in Mianyang, China – Two neutron beamlines, one thermal and one cold, with a shielded block that will couple to transfer cask. (Sun et al., 2016)
2) China Advanced Research Reactor (CARR) located near Beijing, China – A thermal neutron beamline with a shielded block that couples to a transfer cask. (Wei et al., 2013)
3) Idaho National Laboratory, USA – two beamline facilities at the Neutron Radiography Reactor (NRAD), one located beneath a hot-cell and one that couples to a transfer cask. (Craft et al., 2015)
4) SINQ at Paul-Scherrer Institute, Switzerland – A thermal neutron beamline facility, NEUTRA, with a shielded block that couples to a transfer cask. (Lehmann et al., 2003)
5) Institute for Energy Technology, Kjeller, Norway – A thermal neutron beamline at the Joint European Energy Pile heavy water research reactor facility that couples to a transfer cask. (Jennsen et al., 2014)
6) Korea Atomic Energy Research Institute (KAERI), South Korea – A thermal neutron beam at the High Flux Advanced Neutron Application Reactor (HANARO) that couples to a cask. (Sim et al., 2004, 2013)
7) Indira Gandhi Centre for Atomic Research (IGCAR), India – A thermal neutron beamline at KAMINI (Kalpakkam Mini reactor) located beneath a hot-cell. (Manoharan, 2012)
8) Bhabha Atomic Research Centre, India – A thermal neutron beamline at the CIRUS reactor with a shielded facility that couples to a transfer cask. (Singh, 2011)

It is interesting to note that countries typically have one major center where the majority, though not all, of the national research and development of nuclear materials occurs. This is likely the practical economic outcome of the significant infrastructure and personal required for such activities. Additionally, the complexities of shipping irradiated nuclear fuel between facilities require significant lead time and administrative involvement. The money required for shipping irradiated fuel could be the most significant budget item of a fuel development program, and would be better spent science than shipping. Thus, it is generally more attractive to develop the examination capabilities where the irradiated fuel is located than to transport the fuel to the capability.

Worldwide demand for nuclear science education, training, research, technology development, and reactor services decreased through the 1990s and early 2000s (Dodd, 2002). The impact of fewer sources is somewhat countered by the advent of advanced digital radiography systems in the 1990's and subsequent increase in the range of applications, as discussed in Section 3.

5. Progression of the Nuclear Power Industry

The vitality of NR for nuclear industrial applications is inherently coupled to the nuclear power industry worldwide. In the 30 year period from 1945 to 1975, predictions of economical and safe nuclear power generation were evident from advisors to politicians (USA President Eisenhower, Atoms for Peace international convention, Geneva, Switzerland, 1956) and advisors to industry (e.g. USA Westinghouse, PWR; General Electric, BWR light water reactors; UK, Magnox and AGR graphite reactors; Canada, CANDU heavy water reactors; France, Phoenix fast breeder reactor). To support these efforts, many governments financed or otherwise supported operation of research reactors at laboratories and universities. A review published by International Atomic Energy Agency in 1977 provides over 150 references to projects specifically on NR for inspection of nuclear fuels (Ross, 1977). In contrast, during the 30 year period 1975-2005, programs to develop and utilize nuclear power were reduced in many countries due in large part to disenchantment following major nuclear accidents, including Three Mile Island in 1979 and Chernobyl in 1986. France is one notable exception where nearly 80% of its electrical power generation is produced by nuclear power. The United States (U.S.) Nuclear Regulatory Commission (NRC) approved more than ten construction permits per year on average between 1967 and 1978 but ceased completely following the Three Mile Island incident, and no new reactor permits were issued between 1979 and 2012.

There has been much renewed interest in nuclear power in recent years due in part to the need to reduce carbon pollution. While nuclear power has declined in some countries, others are actively increasing their nuclear energy capacity. Germany, for example, obtained around 25% of its electricity from 17 nuclear reactors in 2011, but has since begun phasing out nuclear energy, now only producing 16% of its electricity from eight remaining reactors (World Nuclear Association, 2016), with plans for complete phase-out of nuclear power in Germany by 2022 (Renn and Marshall, 2016). In sharp contrast, China, now a major fraction of the world’s economy, is notable for their ambitious plans to rely increasingly on nuclear reactors, with 34 nuclear power reactors currently in operation and over 20 additional reactors under construction (World Nuclear Association, 2016). Furthermore, China has become largely self-sufficient in reactor design and construction, as well as other aspects of the fuel cycle, and is making full use of western technology while adapting and improving it (World Nuclear Association, 2016).

The future of nuclear power in the USA is more uncertain. Fracking is making natural gas plants more economical to operate compared to other power sources in the USA, which is putting many nuclear plants in the USA at risk of premature retirement due to economics. However, there are some reasons for optimism for the future of the nuclear power industry in the USA. For example, the Watts Bar Unit 2 nuclear power plant officially entered commercial operation in October of 2016 and represents the first new nuclear generation in the USA in 20 years (Tennessee Valley Authority, 2016). The USA has 100 operating reactors as of October 2016 according to the U.S. Nuclear Regulatory Commission, with 34 boiling water reactors (BWRs) and 66 pressurized water reactors (PWRs). The U.S. Department of Energy (DOE) projects that the annual power generating capacity of nuclear power in the USA will remain constant at ~790 billion kWh through 2040 but that USA electricity demand will rise 22% by 2040 (Energy Information Administration, 2016). That means the USA will need hundreds of new power plants to provide carbon-free electricity and continued economic growth. Maintaining nuclear energy's current 20% share of electrical power generation in the USA would require building one new reactor every year starting in 2016, or 20-25 new units by 2040 based on DOE forecasts. However, this pace of construction is not currently manifest, as there are only four nuclear plants currently under construction in the USA.

Much work is needed to achieve reactor designs with assured safety and dramatically improved economics, especially considering recent examples of high costs for the reactor projects in Finland, France, and Britain (Berthélemy and Rangel, 2015). While enthusiasm for nuclear energy may be waning in some countries, it is surging in others. Ever-increasing demand for energy in developing economies and concern for climate change indicate that nuclear energy will continue to play a major role in worldwide energy production for generations to come.

6. Publications from the Worldwide Community

The vitality of the NR community is reflected in its publications, and this section summarizes the progression of these publications. Figure 1 shows the number of publications published in some of the WCNR and ITMNR conference proceedings along with the number of those that apply to the nuclear power industry. As shown in figure 1 at the First World Conference on Neutron Radiography in 1981 very large proportion (45%) of the 140 papers presented were on nuclear power industry applications. By the Tenth World Conference (WCNR-10) in 2014 only 8% of the papers related to the nuclear power industry.

Note however that most of the reports in the conference series relate to progress in techniques not actual studies for materials evaluation. The proprietary nature of most fuel development programs means that actual studies performed with NR would be written only in internal confidential reports and not published in open sources. In addition to the WCNR and ITMNR conference series, there are other sources that show how NR was important to the nuclear industry prior to 1990, including:

1) Conference proceedings *Radiography with Neutrons* contains 28 papers of which four focus on nuclear industry applications. (Hawkesworth, 1975)

2) Conference proceedings *Neutron Radiography and Gaging* contains 24 papers of which three focus on nuclear industry applications. (Berger, 1976)
3) A compilation of the *International Neutron Radiography Newsletter* 1964-1977 contains over one thousand references on NR of which 105 are on NR applied to the nuclear power industry. (Barton, 1977)
4) A review article *Neutron Radiographic Inspection of Nuclear Fuels* is 25 pages with 154 references all related to NR for the nuclear industry. (Ross, 1977)
5) A book *Neutron Radiography Handbook* is 169 pages of which 27 focus on applications of NR for the nuclear power industry. (von der Hardt and Rottger, 1981)
6) A book *Handbook of Materials Testing Reactors and Associated Hot Laboratories* in the European Community is 234 pages. (von der Hardt and Rottger, 1977)
7) A volume containing 158 neutron radiographs on film that illustrate defects in fuel and cladding and can be removed for comparative viewing. (Domanus, 1984)
8) A later book *Practical Neutron Radiography* has 270 pages with 158 references (Domanus, 1992). The table 21.2 on page 242 provides data on fifteen NR facilities in the political group known then as the European Communities. Facilities in Norway and Switzerland would be in addition to this.
9) For early work in the USA, searches should include the *Journal of Nuclear Materials* and journals of the American Nuclear Society (ANS) and the American Society for Nondestructive Testing (ASNT).

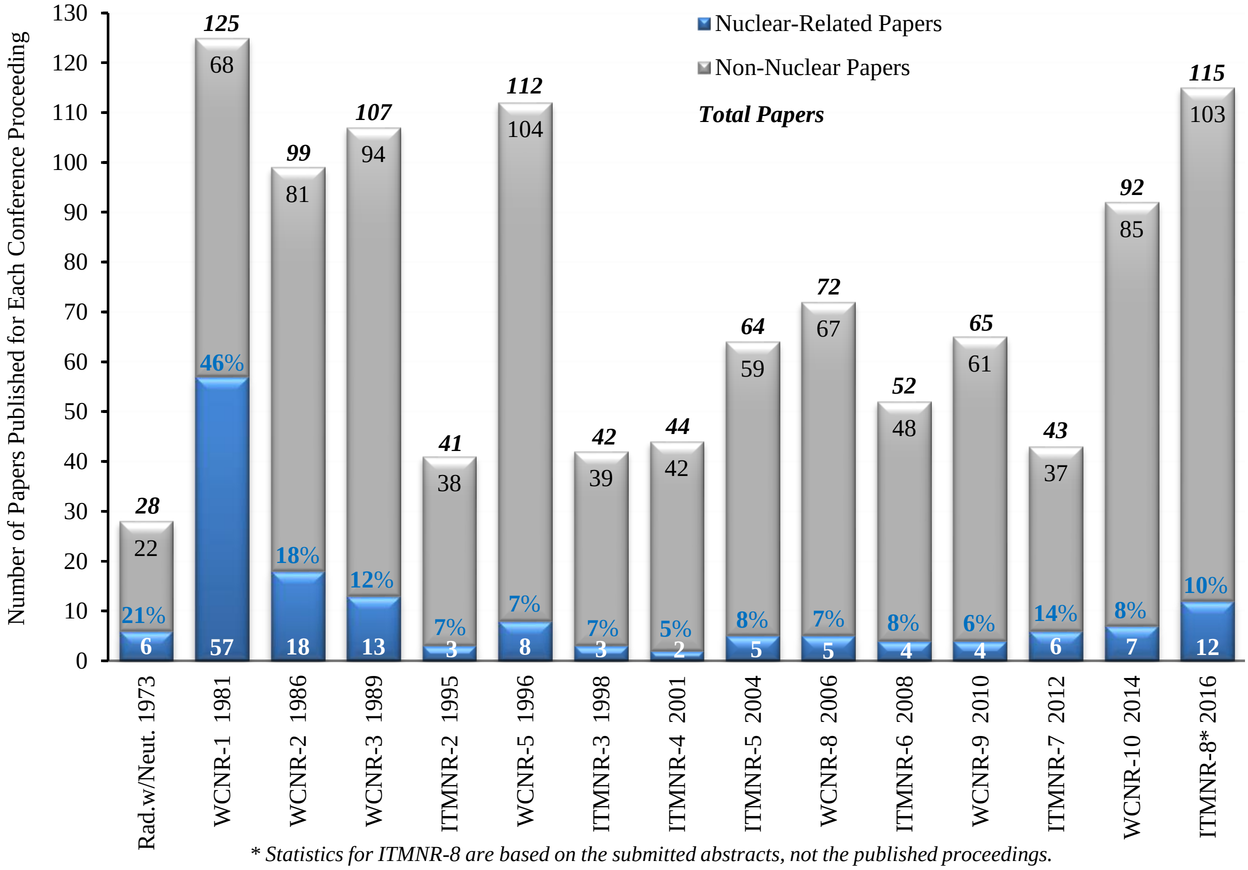

Fig. 1. Number of publications for the WCNR and ITMNR conference series highlighting those related to industrial nuclear power applications.

7. Summary

The vitality of NR for nuclear applications has closely followed that of the nuclear power industry, as discussed in Section 5 and reflected in Figure 1. Examinations of nuclear materials using NR continued through the 1990s, but the number of publications in the WCNR/ITMNR series dwindled, which can be attributed to the downturn in the nuclear power industry. This downturn, compounded by the stagnation of applicable imaging techniques and the limited number and geographic isolation of facilities, as described in Section 4, led to further-diminished involvement of researchers in the conference series and worldwide community. There is a modest resurgence of publications on nuclear power applications in recent years, which could be attributed to increasing enthusiasm for carbon-free nuclear power to address climate change while still allowing developing societies to grow.

Overall, the future looks promising for nuclear power applications of neutron beamline examinations. There is a seemingly continuous development of various advanced and novel imaging systems that initiated in the 1990's and continues to the present day. Accompanying the development of new neutron beam examination techniques has been the discovery of new areas for application of these techniques. Additional advances since the 1990's in neutron beam intensity and quality, neutron detection efficiency, dynamic resolution, and spatial resolution have made neutron examination techniques useful for new areas of pure science and applications, which is reflected in the corresponding increase in the number of publications based on neutron beam examination techniques. Lessons recorded in earlier conferences in this series and the references cited support the case that neutron radiography for nuclear power applications will continue to play important roles in the foreseeable future.

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