Imaging with Neutrons: The Other Penetrating Radiation

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ABSTRACT

Neutron radiography is a well-known imaging technique among those working at nuclear research facilities. However, the move from laboratory to industry has been hampered by the generally large neutron flux requirements and by the relatively small number of nuclear research reactors. The development of imaging techniques that require lower total neutron exposure than traditional methods, coupled with improvements to non-reactor neutron sources suggests that broader application of neutron radiology may be imminent.

Keywords: neutron, imaging, radioscopy, radiography, attenuation, contrast, sensitivity, resolution, gadolinium, scintillation

1. INTRODUCTION

Neutron radiography has existed as a testing technique and as a research tool for over 60 years and has grown in use and application throughout that time. Furthermore, the general location of nuclear research reactors at universities and national laboratories has ensured the availability of researchers with widely varied backgrounds and interests. An active international community sees approximately 50-100 papers published each year. The majority of publications are in refereed conference proceedings and journals. There are two on-going series' of international technical meetings (The World Conferences on Neutron Radiography and the International Topical Meetings on Neutron Radiography). Standards are being actively developed at the national and international levels that support the commercial application of neutron radiography. A society has been formed among neutron radiographers worldwide. The technology is ready for widespread application with a substantial worldwide capability for the production of neutron radiographs and with a reasonably large and organized body of professionals to support it.

Nevertheless, commercial application of neutron imaging has generally lagged the technical developments in the field, with the result that there are remarkable developments that are not being exploited commercially--yet. This lag results from the generally small number of locations at which this non-destructive testing technique and research tool can be practiced. There are few sites in the world where organizations are using their own neutron radiographic equipment to inspect parts of their own manufacture. Application at the industrial scale has been largely limited to the examinations of small, high-value parts that can be shipped to one of the reactor sites where commercial neutron radiography is conducted or to one of several military sites. Broader application than this requires either a proliferation of service organizations or of in-house inspection systems, or of both. Either of these requires a cost-effective neutron source.

Requirements upon neutron sources are being lessened as imaging technology improves. When high-energy neutrons are used, neutron-source costs and complexity drop dramatically, although the scope of application changes substantially as well. The availability of high-yield neutron sources based upon proton/deuteron accelerators and metal (Li, or Be) targets may encourage the proliferation of facilities that will move neutron imaging ahead of other non-destructive testing techniques and into the commercial mainstream.

Imaging with neutrons involves one of: radiography, where images are produced on film; radioscopy, where digital images are prepared; and tomography, where third-dimensional data are calculated and presented; and radiology, which we will consider to be the general term. The developments foreseen in neutron radiology derive from research being conducted in areas such as:

- Light-emitting scintillation screens for thermal neutrons,
- Proton recoil scintillators for fast-neutron imaging
- RFQ (radio-frequency quadrupole proton accelerators),
- Neutron beam guides, and
- Accelerator targets (lithium and/or beryllium).

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Additionally, this technique is benefiting from the explosion of more generally known technologies, including:

High-quantum-efficiency CCD (charge-coupled device) cameras,
Laser-stimulated-readout imaging plates,
Inexpensive computing power, for:
  Computed tomography,
  Monte Carlo design simulation,
  Image manipulation (contrast-stretching, background subtraction, etc.), and
  Shielding calculations.
Writable and Re-writable compact discs,
Large hard-disc devices and inexpensive memory, and
High-speed data transmission.

Neutron imaging, with a small number of noteworthy exceptions, has largely remained a solution in search of problems. It has been generally perceived as expensive and inaccessible. This paper introduces the reader to the available neutron sources, to the contrast available from neutrons of various energies (including a comparison with photons) and to the imaging techniques that are being employed. Discussed in the paper are the implications upon application development and feasibility of the existing technology. Areas of potential application growth are explored

2. NEUTRON SOURCES

Neutrons have generally been most readily available in quantity at nuclear research reactors. However, the number of research reactors has been steadily declining worldwide for some years. Other sources of neutrons do exist, and may have useful special applications. This section describes, in very general terms, the neutron sources that are available today, and indicates where these sources might find application.

2.1 Nuclear Reactors

Nuclear reactors represent an abundant source of thermal neutrons. Neutron flux at the entrance of a neutron collimator can be on the order of $10^{14}$ n/cm$^2$·s. After collimation with a pinhole or divergent collimator, typical neutron flux at the image plane is generally in the $10^6$ - $10^7$ n/cm$^2$·s range. This provides a good balance of depth of field (through collimation ratios on the order of 100) and speed, with exposure times to high-resolution X-ray film of several minutes to several tens of minutes.

Nuclear reactors can also be fitted with cold sources, filters and guide tubes to provide neutrons in the sub-thermal or cold energy range (~1 meV). A cold neutron beam from a well-designed guide tube provides a substantial benefit to the neutron imaging specialist by almost entirely eliminating unwanted radiations from the beam.

There have been several first-class research reactors built in the past few years (JRR-3 in Japan and Hanaro in Korea, for example), with excellent neutron beam facilities. The scientific community in Canada, together with Atomic Energy of Canada, Limited are planning a new state-of-the-art research reactor with truly spectacular neutron-beam capabilities including: a large cold source, a well-conceived guide hall, and both cold- and thermal-neutron guides. These developments bode well for the continuing development of advanced neutron imaging techniques.

A few reactors have been used for neutron imaging that have particularly high fast neutron flux relative to the thermal neutron flux. Initial work on fast neutron imaging was conducted at one such reactor. Most research reactors can generate epi-thermal neutron beams (1 eV - 10 keV) if suitable filters are installed. These epi-thermal beams can be tuned in energy to some extent to optimize the sensitivity to elements with resonance absorptions at particular energies. This applies equally to analytical techniques as to imaging.

Nuclear research reactors are not commonplace items. The high cost of building a reactor plus the enormous perceived administrative load to manage licensing, coupled with the magnitude of the assumed liability and the decommissioning and waste disposal costs make them unattractive, generally, in the private sector.
2.2 D-T tubes

These neutron sources\(^5\) are based upon deuteron beams impinging upon tritiated targets with relatively low accelerating potential and fairly high current, with the resultant yield of 14 MeV fusion neutrons at a rate of up to \(\sim 10^{11}\) n/s. These 14 MeV neutrons are very penetrating in and of themselves and can be used directly for fast-neutron imaging. However, the high-energy neutrons that are produced can, in principle, be moderated to a lower temperature to allow epi-thermal, thermal or perhaps even cold neutron imaging. In practice though, neutrons of such high energy require large moderators and yield relatively low thermal neutron fluxes. The relative portability of systems based upon D-T tubes of this sort makes in-field application quite possible, particularly where the contrast required can be obtained with unmoderated or under-moderated (i.e., fast or epi-thermal) neutrons. In these cases, the small size of the moderator assembly yields a small, bright source of neutrons, permitting high intensity beams at the image plane at acceptably large collimation ratios.

D-T tubes are being used largely for thermal neutron radioscopy, since exposure times to film would be excessive.

2.3 Accelerators

Several technologies have been developed for accelerating beams of protons or deuterons. Cyclotrons, both conventional and super-conducting have been developed and are commercially available that can produce >100 mA or more of protons with energies in the 12 MeV range\(^3\). Such cyclotrons can be used for generating collimated thermal neutron beams with fluxes on the order of \(10^6\) n/cm\(^2\)-s when used with a suitably-designed beryllium or lithium target.

Similarly, radio-frequency quadrupole (RFQ) accelerators, coupled with drift-tube linear accelerators, can be used for the generation of neutrons through the interaction of the accelerated protons or deuterons with Be or Li target material. As with the cyclotrons, there are commercially-available units that can generate neutrons of sufficient intensity for industrial-scale neutron imaging. The generation of \(3 \times 10^{13}\) n/s is possible with such systems\(^4\), with the result that thermal neutron flux at the image plane of a well-designed system may be as high as \(10^9\) n/cm\(^2\)-s.

These accelerators could well be used for epi-thermal or fast neutron imaging as well, but are unlikely to generate sufficiently high flux for cold-neutron imaging.

Neither of these accelerator types is particularly portable, and neither is inexpensive. In either case though, commercial acceptability of accelerators is higher than for research reactors of similar capability.

2.4 Isotopic sources

While several types of neutron source based upon neutron emission from radioisotopes exist, all are of extremely low intensity when compared to either accelerators or nuclear reactors. Such sources have found limited application--primarily where maximal portability is required. Neutron flux tends to be low, background radiation tends to be relatively high, there is a significant waste-disposal problem and the cost is high--both for the initial purchase and for the replacement after decay of the source itself.

3. CONTRAST

The contrast that is obtained in neutron radiography is generally quite different from that obtained when X-rays or gamma rays are used. However, there is no simple rule of thumb that describes adequately the relationships in attenuation between X-rays and neutrons. This is complicated further when it is recognized that neutrons of different energies behave in very different ways in any given material. The change in attenuation mechanisms affects the types of applications that are sensible with neutrons of any particular energy. This section describes the categories of neutron energy and the characteristics of neutrons in those energy ranges.

3.1 Thermal neutrons
Most worldwide work in neutron radiology has been done using thermal neutrons (i.e., neutrons with energies of about 0.023 eV). This has been the case because of the ready availability of high-intensity thermal neutron beams at nuclear research reactors and because of the large variations among the neutron scattering and absorption cross sections of different elements. Figure 1 shows the attenuation by matter of photons with energies of 100 keV to 10 MeV and by thermal neutrons. The reduction in neutron flux as a beam passes through a material is described by

$$\Phi = \Phi_0 e^{-\mu t}.$$  \hspace{1cm} (1)

Where $\Phi$ is the resultant neutron flux, $\Phi_0$ is the incident neutron flux, $\mu$ is the linear attenuation coefficient and $t$ is the thickness of the material. The overall range of attenuation coefficients for thermal neutrons covers nearly four orders of magnitude compared to only three for 100 keV photons and to just over two orders for 1 MeV and 10 MeV photons. Figure 1 also shows that the periodicity that is evident in the photon attenuation data is entirely absent in the neutron data. Consequently, greater contrast may often be obtained between two materials that are of similar atomic number with thermal neutrons than would be possible with photons.

Additionally, it is important to note that in some cases the relatively low attenuation coefficients for thermal neutrons for some materials permits greater penetration than would be possible with photons.

The ability to obtain contrast from hydrogenous materials is an important factor for the application of neutron radiography. Figure 2 shows a neutron radiographic image (prepared on AGFA D3 sc film from a vapour-deposited Gd screen after an irradiation of 11 minutes at Beamport 2 of the McMaster Nuclear Reactor). This image demonstrates very clearly the ability of neutrons to image even relatively thin sections of biological materials. Application of this ability to image hydrogen-containing materials is found in:

The examination of nuclear fuel cladding for zirconium hydride,
The examination of airframe sections for water, and for hydrated aluminum corrosion products, and
The examination of nuclear fuel for water ingress.

Gadolinium absorbs neutrons much more effectively than any material absorbs photons, as is clearly evident in Figure 1, where the highest value of the attenuation coefficient shown is for gadolinium. Hence, Gd is a typical material for use as the basis of a thermal-neutron-radiographic contrast agent. Several methods exist for adding Gd to a material in order to render it more opaque to neutrons and to thereby make it more visible on a neutron radiograph. The technique of contrast enhancement has been well developed for the inspection of investment-cast jet-engine turbine blades. These blades are typically cast to near net shape with a ceramic core to leave air passages for cooling of the finished blade. Removal of the ceramic core is problematic, and contrast-enhanced neutron radiography has been the most effective method yet developed for positively identifying the presence of residual core material. Figure 3 shows a typical jet-engine turbine blade with a very large amount of residual core material contained within it. In this case, the ceramic core was manufactured without Gd and a Gd salt was added to the ceramic as part of the inspection process.

3.2 Fast neutrons

Figure 4 shows the attenuation of neutrons of thermal energy and energies of 100 keV, 1 MeV and 10 MeV for several materials. When compared with thermal neutrons, one sees that there is very little variation in attenuation among the elements when neutron energies are in the MeV range. Most materials have cross sections in the 1-10 barn range for neutrons with MeV energies. Consequently, there is not much contrast available to allow different elements to be discerned. However, the number of nuclei per unit volume does vary from element to element, and penetration of fast neutrons through most materials is quite good. Therefore, it is possible, and perhaps sensible, to image thick objects where significant density changes are being sought. For example, cracks in thick (10-30 cm) objects can be identified using fast neutrons.
In general, nuclear research reactors are not very good sources of fast neutrons, since research reactors usually contain an abundance of moderating material. So, most of the available neutrons are well thermalized. A typical measure of the relative abundance of thermal neutrons and fast neutrons is the cadmium ratio. This measure is the ratio of thermal neutron intensity to fast neutron intensity and is obtained by obtaining two measurements of the activation of gold foils—one with the bare foil and the other with a foil covered with cadmium. Cadmium has a very high absorption cross section for thermal neutrons but this drops off quite quickly at energies only slightly higher than thermal. So, the gold foil that is irradiated without a cadmium cover responds to all of the neutrons while the gold foil behind the cadmium cover is exposed primarily to neutrons with energies higher than thermal.

Comparison of fast neutrons with thermal neutrons

![Comparison of fast neutrons with thermal neutrons](image)

Figure 4  Attenuation by matter of thermal and fast neutrons

Obtaining fast neutrons is relatively easy from smaller neutron sources, such as D-T tubes, radioisotopic sources and accelerators. These sources provide relatively small sources of reasonably high intensity fast neutrons, with average neutron energies in the range from 100 keV to 14 MeV, depending upon the source. Were these neutrons to be moderated to thermal energies there would be losses of intensity to absorption by the moderating material and the source would be made generally more diffuse. Using the fast neutrons directly would result in a smaller, more intense source of neutrons.

However, generating useful images from an intense source of fast neutrons still requires some ingenuity, as such neutrons are not very easy to collimate. This is because there are no strong absorbers of fast neutrons with which to build apertures or collimator walls. Nevertheless, fast-neutron imaging systems can be effectively built and used.

Figure 5 shows a comparison of linear attenuation coefficients for fast neutrons and for photons. It is evident that fast neutrons can penetrate some materials far more readily than can 10 MeV photons. As a consequence of this, there will be some inspections that are ideally suited to fast-neutron use. Furthermore, fast-neutron systems may be able to compete head-to-head with high-energy photon systems even in areas where the traditional systems are already well established.
Figure 5  Linear attenuation coefficients for fast neutrons and for photons

3.3  Cold neutrons

While cold neutrons (~1 meV) penetrate materials less than do thermal neutrons, the differential attenuation between two materials may increase, depending upon the precise energies and materials involved. This can result in substantially-improved contrast between two materials. For example, neutrons with energies below the Bragg cutoff will be attenuated less than those above, so where a crystalline material is being examined it may be possible to exploit this feature. An example is the inspection of ferrous materials for hydrogenous material--where greater contrast is available with cold neutrons than with thermal neutrons--even though penetration through the bulk material is less.

Since cold neutrons are readily detected, it is possible to build high-resolution detectors that function with high efficiency. Consequently, excellent statistics are available in the obtained data so semi-quantitative or quantitative determinations can be made.

3.4  Resonance (Epi-thermal) neutrons

In the energy region between thermal energies (0.023 eV) to perhaps 10 keV is a regime where many nuclides have resonance absorptions. Within a small energy range, the absorption cross-section for a particular nuclide may change by several orders of magnitude. Exploitation of such a resonance can yield tremendous contrast--particularly if the nuclide involved has an increased absorption in the energy region of interest and the bulk material has a low cross section. With sufficient care in the filtration of the neutron beam to limit the neutron energy to the region of interest, and with sufficient knowledge of the sample structure, it is possible to design very sensitive instruments. Since resonance absorptions will be accompanied by the emission of characteristic gamma rays, this technique can be extended to provide a very sensitive quantitative analysis.

4.  IMAGING TECHNIQUES

4.1  Direct method using film
The generation of neutron-radiographic images on high-resolution X-ray film using the internal-conversion electrons from metallic foils of gadolinium is the most common and most familiar technique. Materials other than gadolinium can be used, but the combination of high cross section, prompt decay, and the release of low-energy internal-conversion electrons does make gadolinium an ideal material. Two types of conversion screen are used. Metallic foils of Gd with a suitable thickness of 25 µm are commercially available in widths of up to 15 cm. Thinner foils yield increased exposure times while thicker foils may yield slightly-reduced resolution with no significant improvement in exposure time. Vapour-deposited Gd conversion screens have also been available. These are generally obtained as 25 µm thick coatings on a 3.4 mm thick aluminum substrate with a corrosion-protection film of sapphire about 1 nm thick. These vapour-deposited conversion screens are available in nearly arbitrary sizes up to at least 35 cm X 43 cm.

The direct method of neutron radiography requires neutron fluences on the order of 1-2 X 10^9 n/cm²⋅s, depending upon the optical density required on the film, and upon the details of the film and the development technique.

Single-coated films are generally used for direct-method imaging with X-ray film even though shorter exposure times would be realized with the more common double-coated films. Typically, only the slowest X-ray films are available in single-coated versions, so selecting a faster film even when resolution requirements would permit such a change cannot readily reduce the exposure times.

During the preparation of a direct-method neutron radiograph, the X-ray film and conversion screen are placed in intimate contact, usually in a vacuum cassette, so that maximal effect is obtained upon the film emulsion from the internal-conversion electrons released by the conversion screen. The emulsion also responds to gamma rays—including those in the beam itself, those generated by interaction of the beam with the sample and fixtures and those released by the conversion screen. The gamma-ray contribution to the image is something that the neutron radiographer generally wants to minimize since such contributions tend to reduce the desired contrast (increase the signal-noise ratio). Use of a double-emulsion film would essentially double the effective contribution of gamma rays to the darkening of the film, without increasing the contribution from the internal-conversion electrons, which cannot penetrate the film base to reach the second emulsion.

4.2 Indirect method using film

The indirect method of neutron radiography usually employs materials such with reasonably high cross sections, reasonably low energy emitted radiations and reasonably short decay half-lives, coupled with X-ray film. The technique is to irradiate the conversion screen with the sample, and to later expose a film to the decay of the conversion screen. So, we find the most common materials used for indirect-method neutron radiography are dysprosium (Dy) and indium (In). The methods for both materials have been well developed by the nuclear industry. Indirect-method neutron radiography is very useful for the examination of radioactive materials since the conversion screen is activated only by neutrons and the method is therefore completely insensitive to other radiations. The examination of irradiated nuclear is often conducted using the indirect method.

Neutron fluence requirements for the indirect method tend to be higher than for the direct method--largely because the materials used have lower absorption cross-sections. Furthermore, substantial time is required after neutron exposure for the decay of the conversion screen to expose the film. So, where a direct-method image might require a five-minute exposure at a particular facility, an indirect-method image may not be available for 8 hours. Clearly, when speed is of importance and where the beam composition and sample conditions permit, the direct method would generally be chosen.

4.3 Track-etch imaging

Track-etch imaging employs the damage caused to a plastic film (e.g., nitrocellulose) by heavy particles. Typically, a lithium or boron material is coated on a plastic film or placed against it, so that the alpha particles emitted by these materials after neutron absorption can create damage tracks in the plastic. After exposure, the plastic is etched with an appropriate agent (e.g., KOH) under suitable conditions for the material, the reagent and the exposure. Typical conditions would be 3 M KOH at 50°C for 20 minutes, although there is quite a lot of variation possible.

The etchant attacks the damaged plastic more readily than it does the virgin material, so etch pits are formed at the damage tracks. After etching a cleaning the film, it can be viewed either with oblique lighting or with a microscope after coating with
a thin layer of gold. This technique is capable of very high resolution imaging, and yields a largely linear response to irradiation, so results can be excellent. It is also insensitive to gamma rays so is consequently often used for nuclear fuel examinations.

A variation of the track-etch technique employs boron or lithium already contained in the material being examined to generate an autoradiograph. In this case, a polished specimen of the material is mounted to a suitable track-etch film and exposed to neutrons. Activation of the boron or lithium in the specimen will result in an eventual image that shows the distribution of the boron or lithium. This has been found useful in the development of boron-containing steels for example.

4.4 Scintillators and CCD cameras

Scintillators employing either $^{6}\text{LiF-ZnS}$ or gadolinium oxysulphide scintillators have been widely used for neutron imaging. Substantial reductions in required exposure time can be obtained, albeit with a reduction in potential resolution as well. There have been developments in scintillators, so we now find that the output light spectrum can be matched reasonably well with the response of particular cameras.

CCD (charge-coupled device) camera technology has been progressing at an enormous rate. The availability of scientific grade cameras with excellent quantum efficiency and an ability to operate with low illumination levels is making neutron radioscopy (images generated electronically rather than on film) much more widespread than previously. Numerous workers report excellent results, using a very wide range of cameras and scintillators.

4.5 Imaging plates

The imaging plate technology that has been developed for the medical field is being adapted for and adopted by the non-destructive testing community. While the initial capital costs associated with obtaining imaging plates and the involved readout equipment is quite high, there are several advantages to offset the initial high costs. Most importantly, there are few recurring costs with imaging done in this way. There is no need to purchase chemicals for film processing, or to dispose of those chemicals later. Neither is it necessary to purchase film. Furthermore, the images are immediately available in digital form, and can be transmitted to customer premises immediately or processed in whatever manner is appropriate.

4.6 Computed tomography

Increases in computing capability, coupled with steady improvements in digital image capture techniques are making the possibility of commercial-scale computed thermal neutron tomography more likely. This technique offers the same advantages to the neutron radiographer as does X-ray computed tomography to the X-radiographer. Not all of the work that has been done in developing algorithms for computed tomography with photons applies when neutrons are used, because the details of the interactions of neutrons with matter differ markedly from those when photons are used. Nevertheless, neutron tomography will build upon the body of tomographic work conducted with photons.

5. DISCUSSION AND CONCLUSIONS

Neutron imaging is a well-developed technology, with several companies actively involved in the provision of inspection services and/or in the design and delivery of equipment. The explosion of electronic technologies is of great benefit to neutron radiologists, since it is becoming increasingly possible to generate, manipulate, store and transmit images digitally, with lower neutron flux requirements than needed for film. The relatively recent development of fast-neutron imaging techniques may yield many new applications where the penetrating ability of these particles is beneficial. While it remains unlikely that neutron radiology will ever dominate the non-destructive imaging field, it does hold the promise of gaining a mainstream foothold.

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