Earth-Science Reviews 129 (2014) 120–135

Contents lists available at ScienceDirect

Earth-Science Reviews

journal homepage: www.elsevier.com/locate/earscirev

Neutron imaging of hydrogen-rich fluids in geomaterials and engineered porous media: A review

E. Perfect [a,*], C.-L. Cheng [a,b], M. Kang [a,c], H.Z. Bilheux [c], J.M. Lamanna [d], M.J. Gragg [e], D.M. Wright [b]

[a] Department of Earth and Planetary Sciences, University of Tennessee – Knoxville, Knoxville, TN 37996, United States
[b] Environmental Sciences Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831, United States
[c] Chemical and Engineering Materials Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831, United States
[d] Department of Mechanical, Aerospace and Biomedical Engineering, University of Tennessee – Knoxville, Knoxville, TN 37996, United States
[e] Altamont Environmental, Inc., Asheville, NC 28801, United States

ARTICLE INFO

Article history:
Received 15 July 2013
Accepted 25 November 2013
Available online 12 December 2013

Keywords:
Neutron imaging
Porous media
Radiography
Tomography
Water
Hydrocarbons

ABSTRACT

Recent advances in visualization technologies are providing new discoveries as well as answering old questions with respect to the phase structure and flow of hydrogen-rich fluids, such as water and oil, within porous media. Magnetic resonance and x-ray imaging are sometimes employed in this context, but are subject to significant limitations. In contrast, neutrons are ideally suited for imaging hydrogen-rich fluids in abiotic non-hydrogenous porous media because they are strongly attenuated by hydrogen and can "see" through the solid matrix in a non-destructive fashion. This review paper provides an overview of the general principles behind the use of neutrons to image hydrogen-rich fluids in both 2-dimensions (radiography) and 3-dimensions (tomography). Engineering standards for the neutron imaging method are examined. The main body of the paper consists of a comprehensive review of the diverse scientific literature on neutron imaging of static and dynamic experiments involving variably-saturated geomaterials (rocks and soils) and engineered porous media (bricks and ceramics, concrete, fuel cells, heat pipes, and porous glass). Finally some emerging areas that offer promising opportunities for future research are discussed.

Contents

1. Introduction

As imaging technologies continue to improve our ability to visualize the phase structure and flow of hydrogen-rich fluids at the pore scale, the resulting high resolution data sets provide opportunities for

* Corresponding author. Tel.: +1 865 974 6017.
E-mail address: eperfect@utk.edu (E. Perfect).

0012-8252/$ – see front matter © 2013 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.earscirev.2013.11.012

evaluating existing models, and developing new theoretical frameworks. While magnetic resonance imaging (MRI) and x-ray imaging continue to be employed in this context, both techniques are subject to significant limitations. For example, x-ray imaging relies on the use of tracers to differentiate between air and water in variably-saturated porous media (Basavaraj and Gupta, 2004), while MRI is limited by the range of pore sizes that can be visualized (Chen et al., 2003) and the presence of iron in the solid matrix (Hall et al., 1997). In contrast, neutrons are ideally suited for this application because of their strong attenuation by hydrogen in water and oil, and their relative insensitivity to both the gas phase in pores and solid constituents, such as silica and iron.

Brenizer (2013) has reviewed the history of neutron imaging from its conception to the present day. The neutron itself was discovered by James Chadwick in 1932. Only 3 yr later Hartmut Kallmann and Ernst Kuhn in Berlin, Germany began to make radiographic images of objects using neutrons. However, little progress was made on neutron imaging until the 1950's when technical improvements in the film employed opened up the field to practical applications. Neutron imaging is based on measuring the transmitted intensity of neutrons through an object, either in two dimensions (radiography) or three dimensions (tomography).

In the geosciences neutrons were initially used to measure the water content of soil. Research based on the thermalization of "fast" neutrons released from a source probe inserted into an access tube commenced in the 1950's following the seminal paper by Gardner and Kirkham (1952). It was not until the 1970's, however, that neutron imaging was first applied to natural and engineered porous media (Reijonen and Pihlajavaara, 1972; Subraman and Burkhart, 1972; Wilson et al., 1975; Lewis and Krinitzsky, 1976). Although there have been previous reviews of this topic with respect to applications in earth science, material science, and engineering (Lehmann et al., 2004; Wilding et al., 2005; Winkler, 2006; Banhart et al., 2010; Hess et al., 2011; Kardjilov et al., 2011), none of these focused specifically on neutron imaging of hydrogen-rich fluids in variably-saturated porous media.

This review paper provides an overview of the general principles behind the use of neutron radiography and tomography. The standards for neutron imaging are also examined. The main body of the paper consists of a comprehensive review of the diverse scientific literature on neutron imaging of static and dynamic experiments involving hydrogen-rich fluids in variably-saturated abiotic porous media. We consider the following natural and engineered materials: bricks, ceramics, concrete, fuel cells, heat pipes, porous glass, rocks, and soils. The focus is on nano-, micro- and meso-scale porous systems in which capillary forces dominate over gravity. Research on macro-porous materials such as aircraft wings with an internal honeycomb structure (Hungler et al., 2009), or biological materials such as plant roots and wood xylem tissue (e.g., Nakanishi and Matsubayashi, 1997), is beyond the scope of this review and will not be covered. Finally, some new developments in neutron imaging that offer exciting opportunities for future research will be discussed.

2. Neutron imaging

Neutron imaging beamlines have traditionally been installed at reactor-based facilities, although a few are associated with spallation sources. Table 1 lists the most well-known existing neutron imaging facilities, along with their beamline parameters. Many of these have been in operation for decades. Over time, two main factors have ensured a rapid increase in neutron imaging capabilities and applications: (1) higher neutron fluxes at some facilities, and (2) advances in digital imaging. As a result, thermal neutron fluxes can be as high as 10^8 n cm^{-2} s^{-1}, while the use of charge-coupled device (CCD) cameras allows for 2-dimensional (2D) real-time radiographs with spatial resolutions of up to ~15 μm (Table 1).

2.1. Neutron transmission radiography

Neutron transmission radiography (NTR) is a non-destructive, non-invasive 2-dimensional (2D) imaging technique based on the attenuation (absorption and scattering) of a neutron beam as it passes through a sample, as illustrated in Fig. 1. The resulting "flat" image is a map of the neutron attenuation within the sample under investigation. Neutrons interact with the nucleus of the atom rather than with its electron cloud. The interaction forces between neutrons and nuclei are not correlated with the atomic number of the element, but instead depend upon the particular isotope of the element (Anderson et al., 2009; Strobl et al., 2009). For example, neutrons are highly sensitive to light isotopes such as ^{1}H, ^{6}Li, ^{10}B, and rather insensitive to heavier isotopes such as ^{82}Pb.

For a monochromatic (single wavelength) beam traversing a homogeneous sample, the measured intensity, I, is given by the Lambert–Beer law (Anderson et al., 2009):

$$I = I_0 e^{-\mu\tau} \tag{1}$$

where I_0 is the incident beam intensity, μ is the attenuation coefficient in cm^{-1} and τ is the sample thickness. In the case of a polychromatic neutron beam going through a heterogeneous sample comprised of n elements, Eq. (1) becomes:

$$I(\lambda) = \int_{\lambda_{min}}^{\lambda_{max}} I_0(\lambda) e^{\left[-\sum_{i=1}^{n}(\tau_i \mu_i(\lambda)\right]} d\lambda \tag{2}$$

where λ is the neutron wavelength, τ_i is the thickness of element i, and $\mu_i(\lambda) = \frac{\sigma_i(\lambda) m \rho N_A}{M}$ is the linear attenuation coefficient of element i, where $\sigma_i(\lambda)$ is the microscopic cross section of element i, m is the number of moles of a molecule, ρ is the density, M is the molecular weight, and N_A is the Avogadro constant.

Both absorption and scattering influence the level of contrast in a 2D image. Imaging a thick sample with a polychromatic neutron beam can result in artifacts due to beam hardening (Hassanein, 2006). As the beam passes through the sample, its mean energy increases (i.e., it becomes "harder") because the lower-energy neutrons are preferentially absorbed, leaving behind only the higher energy neutrons.

As indicated by Eq. (2), the contrast mechanism strongly depends upon the radiation source, i.e. the range of neutron wavelengths available at the beamline. Using the different neutron wavelengths at pulsed spallation sources it is possible to obtain multiple radiographs of the same sample, each with very different contrasts (a kind of "multispectral" imaging known as time-of-flight imaging).

The following worked example illustrates the impact of two different wavelengths on neutron transmission. First order approximations of the attenuation coefficients for water (H_2O) in thermal (1.54 Å) and cold (9 Å) monochromatic neutron beams can be calculated based on Eq. (3):

$$\mu_{H_2O} = \sigma_{(H)} \times 2 \times \rho_{(H_2O)} \times N_A / M_{(H_2O)} + \sigma_{(O)} \times \rho_{(H_2O)} \times N_A / M_{(H_2O)} \tag{3}$$

where $\sigma_{(H)} = 82$ barn and $\sigma_{(O)} = 4$ barn at 1.54 Å (National Institute of Standards and Technology, 2013), $\sigma_{(H)} = 110$ barn and $\sigma_{(O)} = 6$ barn at 9 Å (Brookhaven National Laboratory, 2013), $\rho_{(H_2O)} = 1$ g cm^{-3}, $N_A = 6.022 \times 10^{-23}$ mol^{-1}, and $M_{(H_2O)} = 18.02$ g mol^{-1}. The resulting values for μ_{H_2O} are 5.62 cm^{-1} and 7.55 cm^{-1} for thermal and cold neutrons, respectively. Using these values in Eq. (1) gives the neutron transmission (I/I_0) as a function of water thickness. Fig. 2 shows the transmission curves for water in thermal and cold monochromatic neutron beams assuming no scattering effect. For any given water thickness, attenuation of the cold neutron beam is greater than with the thermal neutron beam.

Secondary scattered neutrons, as well as background from the environment, can also cause artifacts in the levels of contrast in radiographic images (Hassanein, 2006). The errors produced by scattering and background are often much larger than those due to beam hardening