Quantitative Crack Analysis Using Neutron Radiography with Gadolinium Contrast Enhancement Agents

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INTRODUCTION

Neutron radiography is a valuable tool for non-destructive examination of irradiated nuclear fuel; however, the ability to detect fine cracks in cladding materials is generally limited by the resolution of the imaging technique. Infiltrating fine cracks with a contrast agent can enhance the ability to determine and quantify cracks through neutron radiography. Establishing a documented standard for the quantitative analysis of crack structures using neutron radiography will provide a valuable tool for examining irradiated materials, particularly advanced cladding materials, higher burnup fuels, and accident tolerant claddings. This project tested three gadolinium infiltrant solutions containing varying amounts of methanol and ammonium lauryl sulfate to quantify the effect of lowering the surface tension of the infiltrant solution on the ability of the infiltrant to penetrate small cracks. Scanning and digitally processing the resulting radiographic images provided a quantitative measurement of the crack density, extent of infiltrant penetration, and the effectiveness of washing to remove the residual gadolinium.

BACKGROUND

Neutron radiography is an established technique for investigating the internal structure of materials containing high atomic number elements, which significantly attenuate x-rays and gamma rays, but (in most cases) not neutrons. Neutrons are also preferable for imaging radioactive objects, like used nuclear reactor fuel, due to the large amount of imaging noise that results from the gamma rays emitted by the irradiated object. Neutron imaging techniques, such as foil-transfer neutron radiography, that are not sensitive to gamma rays can provide clear images of highly radioactive objects.

Image contrast is a primary limitation to crack detection through neutron radiography. When cracks in an object are filled with air, the cracks cannot be easily distinguished from the surrounding material, as the difference between neutron attenuation by air and by non-absorbing materials is generally small. There are two solutions to this problem, increase the imaging time to increase the image contrast, or fill the cracks with a highly-attenuating contrast agent. Since the contrast agent will have a much larger attenuation coefficient than the surrounding material, the imaging time will not need to be extended to resolve the cracks in the material.

A contrast agent for neutron radiography must provide a large cross section for neutrons, and should be readily prepared into a solution. Gadolinium(III) nitrate meets both requirements, as the gadolinium provides the largest thermal neutron cross-section of any naturally occurring element, and gadolinium nitrate is readily soluble in water. The use of gadolinium as a contrast-enhancement in neutron radiography is well established (1, 2); however, there are very few quantitative evaluations of gadolinium contrast agents in open literature.

The Neutron RADiography (NRAD) reactor at Idaho National Laboratory, a 250 KW TRIGA (Training, Research, Isotopes, General Atomics) reactor, provides neutrons for two beamlines. The North Radiography Station is used for digital imaging. The East Radiography Station at the NRAD allows for objects to be mounted to a cassette containing activation foils. Two activation foils, dysprosium for thermal neutrons and indium for epithermal neutrons, are typically used to capture the neutrons that pass through the sample. As the cassette is exposed to the beam of neutrons, the neutron attenuation by the materials in the object being imaged creates an activated map in the activation foils. After a sufficient irradiation time, the activation foils are placed in contact with photographic film. As the activated atoms in the foils decay, they release radiation that is captured in the film. The film is then developed to provide a radiographic image of the object (3).

METHODOLOGY

A set of 10 numbered aluminum test blocks made following ASTM standards provided documented test objects for investigating the ability of potential contrast agents to infiltrate cracks and appear in subsequent radiographs. Conventional dye penetrant testing provides a useful comparison standard for the imaging of cracks via neutron radiography. To produce a reference crack image using dye penetrant, the block surface is cleaned, the penetrant is applied, and the penetrant is allowed to dwell for 15 minutes. A developing agent sprayed on the surface of the block pulls the dye out of the cracks and reveals the crack patterns. Optical photographs of the cracks taken immediately after developer application, and after a two minute development period, provide the reference images for the neutron radiography experiments (see Fig. 1a). Blocks 4 and 8 indicated no significant cracking and were excluded from further testing.

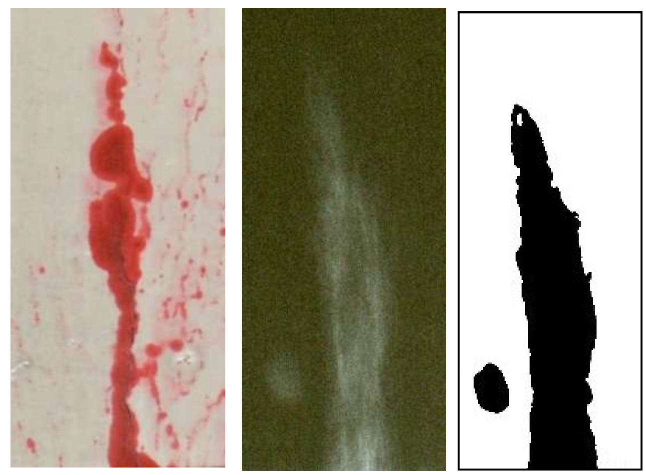
TABLE I. Infiltration and washing solutions.

	Contrast Agent	Solution Composition
Solution 1	0.3M $Gd(NO_3)_3$	water
Solution 2	0.3M $Gd(NO_3)_3$	50% water/50% methanol
Solution 3	0.3M $Gd(NO_3)_3$	50% water/50% methanol with 5% ammonium lauryl sulfate
Solution 4	none	water
Solution 5	none	50% water/50% methanol

Table I shows the different infiltrant and washing solutions used in this experiment. The different solutions test how lowering the viscosity and surface tension of the infiltrant solutions improves the ability to infiltrate gadolinium into cracks. Table II describes the infiltration and washing steps that each set of blocks went through. During the infiltration steps, the blocks were submerged in the solution, then allowed to sit or placed under a vacuum of approximately -0.03 MPa. Wash steps included soak steps at atmospheric pressure and under vacuum (~ -0.03 MPa), as well as submersion in an ultrasonic cleaning bath.

After preparing the blocks, they were attached to the foil cassette using aluminum tape, then exposed to the neutron beam in the East Radiography Station for 22 minutes. After exposure, the activation foils were removed from the cassette and allowed to decay overnight in contact with the radiographic film. The irradiated blocks were also held overnight to decay to background.

The next step of the process made digital scans of the radiographs, and performed image processing to create a metric to measure the amount of cracking measured in each image. To create the metric value, the image of each block was cropped and imported to ImageJ (4). Each image was converted to grayscale and intensity-shifted so the grayscale levels were consistent across all images. A rolling ball algorithm removed non-crack noise from the image. Finally, a threshold made the crack features in the image black, and the rest of the block image white, so the individual cracks could be isolated and cropped. The ratio of the black and white pixels in the cropped image provides a quantitative measure of the amount of gadolinium present in each crack. Fig. 1 shows Crack 6a as it appears in a dye penetrant test (Fig. 1a), a radiograph (Fig. 1b), and in a processed radiograph (Fig. 1c).



a. Dye penetrant. b. Radiograph. c. Processed image.

Fig. 1. Crack 6a shown by different techniques

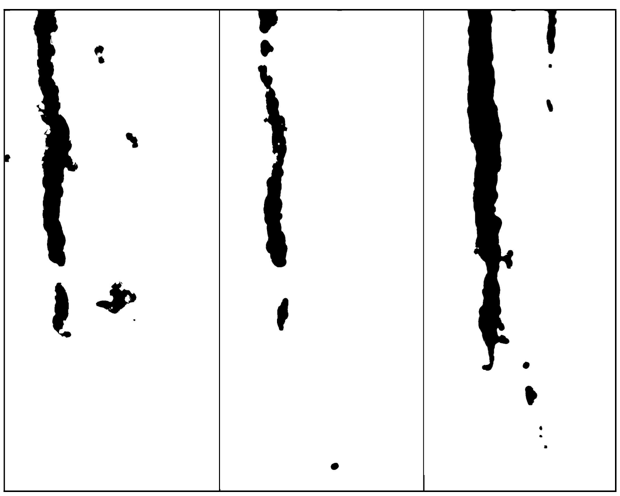
TABLE II. Infiltration and washing steps.

		Blocks 1&2	Blocks 5&7	Blocks 6&9	Blocks 3&10
Infiltration 1 (I1)	Solution	2	1	1	2
	Process	5 min. soak*	5 min. vacuum†	5 min. soak	5 min. vacuum
Wash 1 (W1)	Solution	4	4	4	4
	Process	5 min. ultrasonic + 5 min. vacuum	5 min. ultrasonic + 5 min. vacuum	5 min. ultrasonic	5 min. ultrasonic
Infiltration 2 (I2)	Solution	3	2	2	3
	Process	15 min. soak	15 min. vacuum	15 min. soak	15 min. vacuum
Wash 2 (W2)	Solution	5	4	4	4
	Process	10 min. ultrasonic	10 min. ultrasonic	10 min. ultrasonic	10 min. ultrasonic
Wash 3 (W3)	Solution	5	4	4	4
	Process	15 min. soak + 15 min. vacuum	15 min. soak + 15 min. vacuum	15 min. soak + 15 min. vacuum	15 min. soak + 15 min. vacuum
Infiltration 3 (I3)	Solution	3	2	2	3
	Process	7.5 min vacuum x4	7.5 min vacuum x4	7.5 min vacuum x4	7.5 min vacuum x4
Wash 4 (W4)	Solution	5	5	5	4
	Process	(22.5 m vacuum + 5 min. ultrasonic) x3	(22.5 m vacuum + 5 min. ultrasonic) x3	(22.5 m vacuum + 5 min. ultrasonic) x3	(22.5 m vacuum + 5 min. ultrasonic) x3
Wash 5 (W5)	Solution	4	4	4	4
	Process	25 min. vacuum + (5 min. vacuum + 5 min. ultrasonic) x3	25 min. vacuum + (5 min. vacuum + 5 min. ultrasonic) x3	25 min. vacuum + (5 min. vacuum + 5 min. ultrasonic) x3	25 min. vacuum + (5 min. vacuum + 5 min. ultrasonic) x3

*atmospheric pressure †approximately -0.03 MPa vacuum

RESULTS

Because each block featured multiple cracks, the eight blocks yielded 21 distinct cracks. Each crack was isolated as shown in Fig. 1. Fig. 2 shows a series of processed images for Crack 6b, Fig. 2a is the processed crack image after Infiltration 1; Fig. 2b shows the processed crack image after Wash 1 in Table II; and Fig. 2c displays the processed image after Infiltration 2 in Table II. The percentages of black pixels in the processed image area determined the measured area of the cracks. In Fig. 2a, the percent area cracked is 8.5%. Since the crack had no infiltrant prior to Infiltration 1, the change in crack area for Infiltration 1 step for Crack 6b is 8.5%. The wash step shown in Fig. 2b resulted in a measured crack area of 4.9%, resulting in a -3.6% change from Fig. 2a. For Infiltration 2 (Fig. 2c), the change was +8.5% for a total area of 13.4%.



a. Infiltration1 b. Wash 1 c. Infiltration 2

Fig. 2. Processed images of Crack 6b through three treatment steps

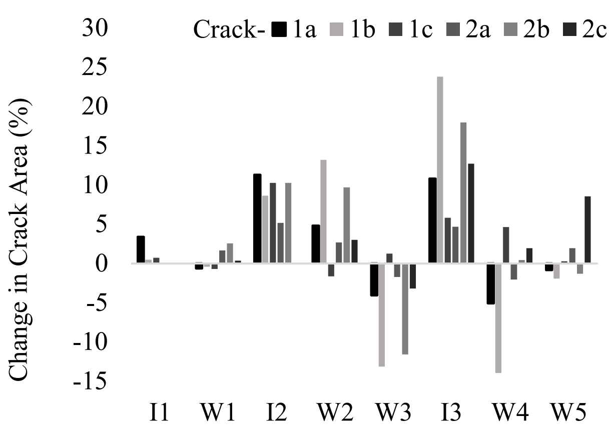


Fig. 3. Change in crack area for each step in Table II for Blocks 1 and 2.

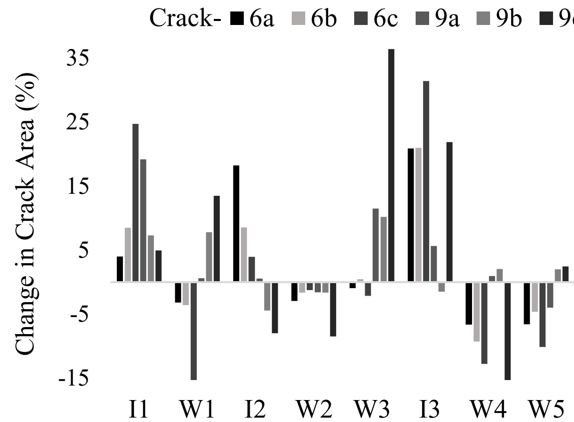


Fig. 4. Change in crack area for each step in Table II for Blocks 6 and 9.

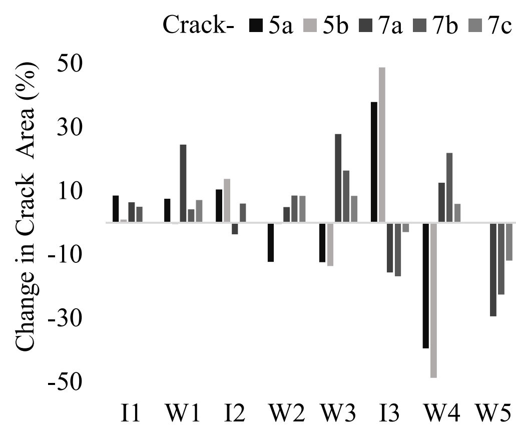


Fig. 5. Change in crack area for each step in Table II for Blocks 5 and 7.

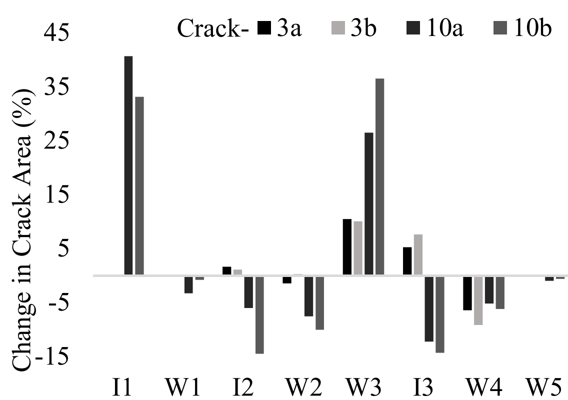


Fig. 6. Change in crack area for each step in Table II for Blocks 3 and 10.

Figs. 3-6 show these responses graphically for all of the cracks detected in this experiment. For Figs. 3-6 the four groups of two blocks undergoing the same infiltration and wash steps from Table II were kept together. The horizontal axis of the graph displays each infiltration or wash step from Table II for that set of blocks, with each crack displayed individually, and the vertical axis is the response of the measured crack area to that infiltration or wash.

From Figs. 3-6, Infiltration 3 increased the cracked area an average of 10.1%, higher than Infiltration 1 (8%) or Infiltration 2 (3.5%). This indicates that infiltration steps utilizing multiple vacuum cycles with solutions containing methanol have the greatest impact on the amount of gadolinium in the cracks. For Infiltration 3, the cracks in blocks 5, 6, 7, and 9 showed slightly better infiltration than in blocks 1, 2, 3, and 10. The infiltration solution used for blocks 1, 2, 3, and 10 contained ammonium lauryl sulfate, indicating that using the ammonium lauryl sulfate to lower the surface tension of the infiltration solution did not improve ability to penetrate the cracks. Wash 4 and 5 have the highest average removal rates of gadolinium, again indicating that multiple vacuum and ultrasonic cleaning steps have the most effect on the amount of contrast agent in the cracks. The results in Figs. 3-6 show that Wash 4 and 5 had the most success removing gadolinium from the cracks, indicating that shorter wash steps like Wash 1, 2, and 3 do not allow enough time for the wash solution to remove the gadolinium contrast agent.

SUMMARY AND CONCLUSIONS

This experiment demonstrated the ability to infiltrate a series of cracks with a contrast agent to enhance the visibility of the cracks in a neutron radiograph. A quantitative method was developed to determine the crack area by performing image processing on the radiographs. Testing different contrast agent solutions and varying the methods of infiltration allowed for documented development of the preferred infiltration and washing methods. Based on the results of this experiment, infiltration solutions containing a 50/50 volume ratio of water and methanol were the most successful in infiltrating the cracks. Adding ammonium lauryl sulfate to lower the surface tension of the solution did not increase the amount of contrast agent remaining in the cracks. For washing the contrast agent out of the cracks, it was determined that longer processes involving multiple vacuum and ultrasonic cycles were more effective in removing the infiltrant from cracks.

To verify the results of the image analysis process, neutron activation analysis based on an activated dysprosium infiltrant will directly measure the amount of infiltrant in the cracks. Combining the pixel counting from gadolinium-infiltrated radiographs with dysprosium activation analysis will allow for a correlation between processed radiographs and count data to provide a validation of the crack quantification metric, determine from the image processing of neutron radiographs.

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