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Method for In Situ Visualization of Tritium Diffused in Stainless Steel using a Digital Autoradiographic Imaging System

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Abstract

At the end of their service lives, various stainless steel components of nuclear weapons that have been exposed to tritium gas are evaluated to determine the extent of the tritium permeation. This information is then used to assess the decrement to performance caused by hydrogen (tritium) embrittlement. This evaluation is currently performed using a photo-emulsion based method and requires 24 hours or longer to complete. A system based on digital imaging technology has recently been designed and built at the Savannah River National Laboratory that performs this evaluation in 10 minutes or less on typical samples.

Introduction

Stainless steel tritium gas reservoirs and tubing, which are components of a nuclear weapon's gas delivery system, are exposed to tritium for a prolonged period of time. These components are destructively evaluated at the end of their service lives for a variety of reasons. One reason is to assess embrittlement due to tritium permeation into the material. The current method used for this assessment is autoradiography [1]. This technique requires the destructive removal of one or more samples from various components of the gas delivery system. These samples are prepared as metallographic mounts, which consists of mounting the sample in either bakelite or acrylic media. This mount is then ground, polished, and etched with an appropriate solution to reveal the microstructure of the sample. Next a liquid photographic emulsion (photo-emulsion) is applied to the surface of the metallographic mount. The liquid coating cures to form a transparent layer that is sensitive to the tritium beta (18 keV) and associated secondary low energy x-rays. Areas of the photographic emulsion that are exposed to the tritium radiation change from transparent to an opaque gray color. This change in the photo-emulsion then provides an indication of the location and depth of tritium

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permeation in the underlying base metal. Typical results are shown in Figure 1, where the dark gray color indicates the location of tritium. The developed emulsion on the sample is then imaged via an optical microscope and attached camera. The images are later used to measure the depth of tritium permeation from the inside surface of the reservoir component (i.e., where the tritium is located during service) to the edge of the exposed photo-emulsion. From the images in Figure 1, the concentration of tritium appears constant except at the very edges where there is an apparent grayscale density gradient. In fact, the concentration of tritium is not constant and the uniform appearance is due to saturation of the photographic emulsion, which results in the loss of the concentration gradient information.

<Location of Figure 1>

The extent of the tritium permeation is then correlated to the amount of embrittlement and the related decrement in material properties. The disadvantages of this technique include the long exposure times (24 hours or more), the generation of mixed waste (chemical and radiological), the cost and limited shelf life of the photo-emulsion, and the delay that results when rework is required. A digital imaging system, to replace the current photo-emulsion technique, has been designed, assembled and field tested at the Savannah River National Laboratory (SRNL). Images from this system are typically obtained in 10 minutes or less.

Experiment and Discussion

In this section, a description of the digital autoradiographic equipment and methodology and its associated capabilities is provided. The advantages of the digital method are the reduced time of exposure, increased sample throughput, reduced rework of samples, the elimination of chemical processing and the related mixed waste, and a notable cost savings.

Various commercial autoradiography systems are available; however, they are optimized for biological research and can not be readily adapted to the present application. Furthermore, the present application requires a synthesis of both visible and tritium images to replicate the result from the photo-emulsion technique. The goal in this project has been to provide an instrument optimized to the unique requirements of this application.

The proposed solution was an imaging system based on a lens-coupled CCD (charge coupled device) camera. Due to the contamination issues related to working with tritium, the feasibility of the proposed solution was demonstrated with an isotope in a sealed source configuration. Due to the low energy of the tritium beta particle another beta emitter was required due to attenuation in source encapsulation. It was calculated that a 0.1 μCi Strontium-90 (Sr-90) source would produce an equivalent amount of visible light in the scintillator as the expected average tritium concentration (on the order of 100 parts per million atomic, appm) in stainless steel. The initial feasibility tests indicated that integration times on the order of minutes would be needed. The relatively long integration time necessitated the use of a cooled CCD camera to minimize dark current and the associated dark current noise. Based on this information, the system described below was fabricated and assembled.

A schematic representation of the system design is shown in Figure 2. The primary components of the system are a scientific grade cooled CCD camera (Photometrics Versarray, 1024 x 1024 Array), lens (Nikon 60 mm f/2.8 Micro), an expandable light-tight bellows, and sample holder and manipulator. This camera is maintained at a temperature of -70 °C by a thermoelectric cooler. At this temperature the dark current and associated dark current noise is essentially negligible and the system noise is dominated by the readout noise. The readout noise level is minimized by using the slowest readout speed (50 kHz) and lowest gain settings on the camera. The system provides the ability to change the magnification or field-of-view (FOV) by changing the working distance using the vertically oriented linear translation stage.

<Location of Figure 2>

The digital autoradiography method can be described as follows. A metallurgical mount is loaded into the sample holder, shown in Figure 3. The sample is placed on a self-leveling vise to ensure that the top surface of the sample is parallel to the image plane of the camera. The vise is tightened using a thumb wheel so that the sample is pressed into a recess on the bottom of the top plate, thus preventing sample movement during the inspection. The centering cup, attached to the self-leveling vise, aligns the sample with a recess in the top plate. The sample table allows for variation in sample height beyond the 0.5-inch travel range of the self-leveling vise. The 2-axis manipulator is used to move the sample within the system field-of-view (FOV).

<Location of Figure 3>

After the sample is loaded, a visible image is obtained with the bellows and scintillator removed from the system. Next, the scintillator sheet is placed in direct contact with the sample surface through the opening in the holder top plate. A glass cover plate is placed on top of the scintillator to hold the scintillator in intimate contact with the sample. The scintillator converts the energy deposited by the beta and associated x-rays into visible light which is collected by the optical system. The selected scintillator (Kodak BioMax™ Transcreen LE) is optimized for low energy beta emitters such as tritium. It is typically used as an intensifying screen in film-based autoradiography of biological specimens. The bellows assembly is installed to provide a light-tight environment. The tritium image is then acquired, with typical integration times ranging from 5 to 10 minutes. The tritium image is automatically processed by the system software to remove unwanted contributions, such as the CCD bias level and readout noise, so that a useful composite image can be created. The resulting tritium image is merged with the visible image to form a composite image. The composite image displays the visible data in gray-scale and the tritium data in shades of green. This presentation clearly differentiates the visible and tritium information. Representative images are displayed in Figure 4 with the visible image in 4a, the processed tritium image in 4b, and the color composite in 4c.

<Location of Figure 4>

The system was calibrated for dimensional measurements and for tritium concentration. Dimensional calibration of the digital autoradiography system was needed to provide accurate quantitative measurement of tritium permeation depth. The calibration involved correlating the vertical stage position, which is related to the camera-to-sample distance, to the CCD image pixel size. This calibration was performed using a set of calibrated gage blocks.

The calibration of the CCD intensity to tritium concentration (C_T) was performed as follows. The permeation depth to the 1 appm tritium concentration was calculated using an approved diffusion code for a number of the samples in the study population. Samples for which the digital, photo-emulsion, and calculated permeation values were in agreement were used to “bench mark” the CCD intensity for tritium concentration. The 1 appm level is near the sensitivity limit of both methods and thus corresponds roughly to the concentration value where permeation depth measurements are made in practice. The empirical relationship of CCD intensity to the system FOV under constant lighting conditions and the intrinsic linearity of the intensity with exposure time (T_{exp}) were also used to formulate a relationship between CCD intensity and tritium concentration. Lastly, the CCD camera analog to digital converter has a gain factor (G_F) setting that allows signal multiplication by a factor of 1, 2, or 4. Using the information above, the working model (Eq. 1) for the CCD intensity (I), as a function of the FOV, C_T, T_{exp} and $G_{F,}$ is obtained.

(1) $$I = [6.537 \times \ln(FOV) - 11.91] \times \left[\frac{T_{\exp}}{1000}\right] \times [C_T] \times \left[\frac{G_F}{4}\right]$$

The FOV is in units of mm, T_{exp} is in units of seconds, C_T is in units of appm, and G_F is unitless. The relationship in Eq. 1 is used to calculate the appropriate threshold intensity at which to measure the permeation depth in a consistent and unbiased fashion. The resulting nonlinear relationship is due to the fact that the CCD intensity is directly proportional to the sample area per pixel, which increases with working distance, and that the solid angle subtended by the lens aperture decreases, with increasing working distance, at a different rate.

The system software consists of two major modules, the data acquisition module and the image measurement and analysis module. These modules are custom designed software created using the LabVIEW ™ programming language. LabVIEW ™ is a graphical programming language where each program has a "front panel" that serves as the user interface and an underlying "wiring diagram" where data flow and processing are mapped. The data acquisition module controls basic system functions, such as camera and stage control, the overall inspection flow control, image processing functions, and data archiving.

The measurement and analysis module assists the user in obtaining unbiased and consistent measurements of tritium permeation at a user selected location or locations. The user selects a subregion of the image for analysis as defined by the red box in Figure 5a. After the subregion is specified, the software automatically rotates the image subregion data such that the selected profile line (yellow line in Figure 5a) is oriented in the vertical direction as shown in Figures 5b and 5c. The user then adjusts the two red horizontal cursors in the visible image (Figure 5b) window to set the limits for the tritium permeation measurement. These cursors are usually set at the edges of the sample, since there should be no tritium outside the sample. The user then adjusts the vertical cursor in the visible image to select the desired column for the tritium profile. The cursors in the tritium image (Figure 5c) automatically follow the movement of the cursors in the visible image. When the cursor adjustment is complete, the user presses a control button, which causes the software to automatically extract the profile data from the tritium image data as shown in Figure 5d. The profile data points are fit automatically using a polynomial fitting subroutine. The user now adjusts the threshold (horizontal cursor) in Figure 5d. As the user adjusts the threshold, the permeation measurement is updated in real-time. The threshold is determined using the CCD intensity calculated at the 1 appm tritium concentration using Eq. 1. The permeation measurement is made between the intersection points of the threshold level and the polynomial fit, which reduces the impact of noise on the measurement. For this sample, the permeation measurement obtained form the digital system was 0.102 inches, which compared well to a measured value of 0.101 inches from the photo-emulsion method.

<Location of Figure 5>

Figure 6 shows a comparison between the extracted tritium profile of Figure 5d and an ideal "Gaussian" function. The Gaussian function of the form in (Eq. 2) is the theoretical solution of the diffusion equation for a point source of tritium. This figure shows excellent agreement between the expected theoretical distribution and the experimentally measured result. Lastly, Figure 7 shows a comparison between the photo-emulsion method and the digital system results for several representative samples that were evaluated during the course of this study.

(2) $$I = I_0 \exp\left[-p \left(\frac{r - r_0}{s} \right)^2 \right]$$

<Location of Figure 6>

<Location of Figure 7>

Conclusion and Future Work

The general conclusion from the comparison study is that the digital autoradiography technique produces similar results as the photo-emulsion autoradiography technique both qualitatively and quantitatively. However, the digital method has the advantage of producing these images in 10 minutes or less, whereas the photo-emulsion method requires 24 hours or more. Future work in this area could include the replacement of the off-the-shelf f/2.8 lens with a custom designed lens with greater light collection efficiency. Other possibilities for improvement would be to replace the lens altogether in favor of a direct coupling between the sample and the CCD chip with a fiber optic coupler, or to employ an intensified CCD camera. All these methods could be used to reduce noise and or exposure time beyond what has already been achieved.

Acknowledgements

Metallography and photo-emulsion autoradiography of the samples in this report were performed in the metallurgical laboratory of the Material Test Facility at the Savannah River Site by Brenda Bordon, Josie Fraley, Pam Morgan, Sandra Stallings, Chiquita Johnson, and Lin Thacker.

References

[1] J. D. Braun, G. L. Downs, and G. W. Powell, “An Autoradiographic Investigation of Tritium in 304L Stainless Steel,” Metallography (1971), pp. 231-242.

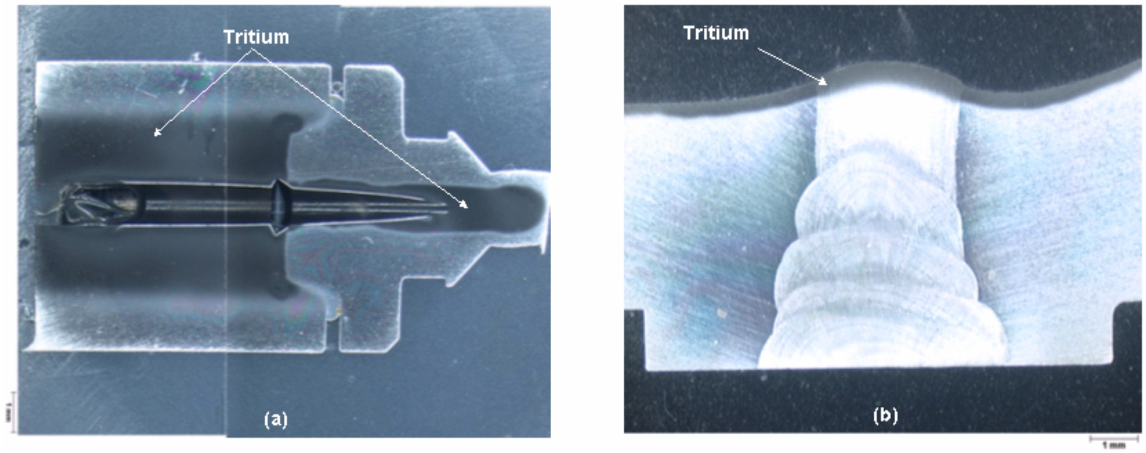

Figure 1: Typical photo-emulsion autoradiography results. Reclamation weld (a) and girth weld (b). The presence of tritium is indicated by the dark gray bands.

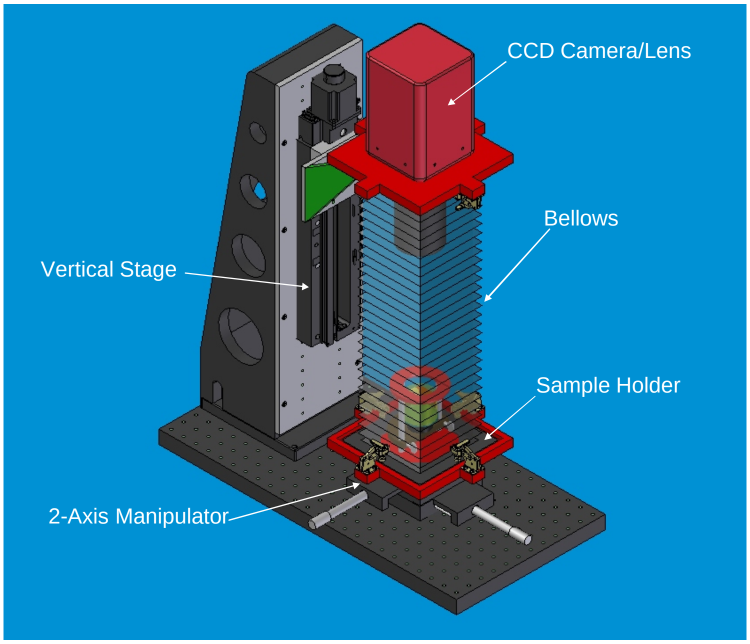

Figure 2: Schematic representation of the digital autoradiography system.

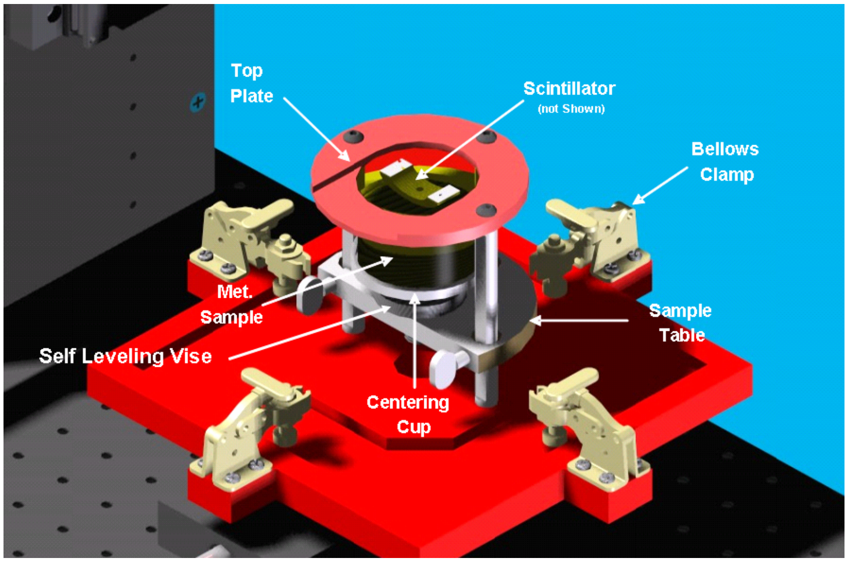

Figure 3: Details of the digital autoradiography system sample holder.

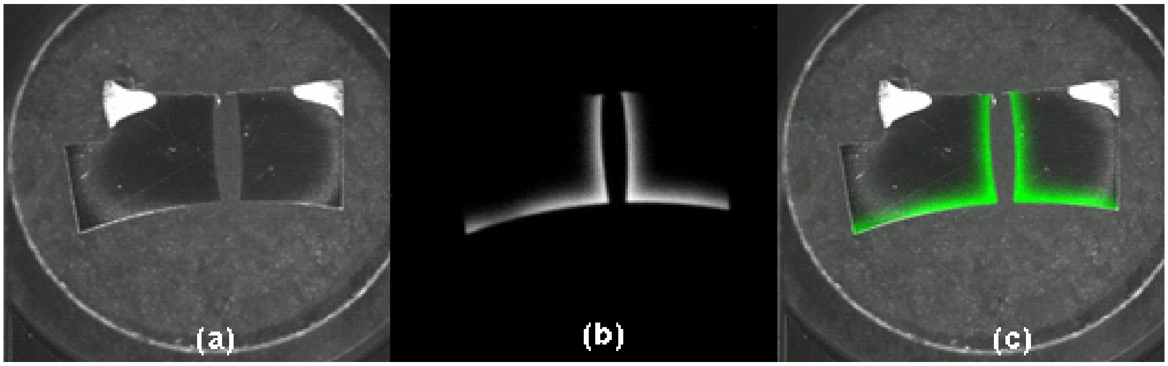

Figure 4: Digital autoradiography images of a gas delivery component: (a) visible image; (b) tritium image; (c) color composite image.

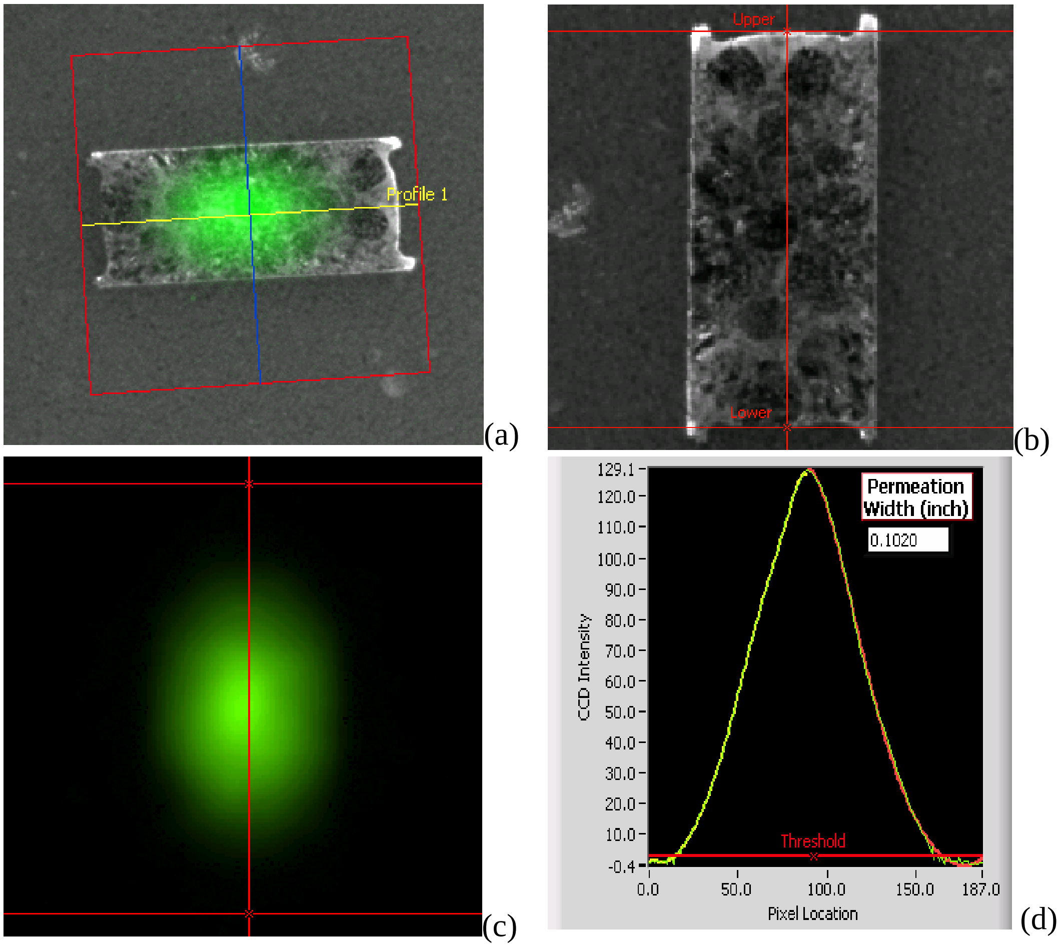

Figure 5: Images taken from the measurement and analysis software. (a) user defined analysis subregion; (b) visible image showing extracted data profile line and user imposed limits; (c) tritium image with profile line and limits shown; (d) extracted tritium profile showing threshold for permeation width measurement.

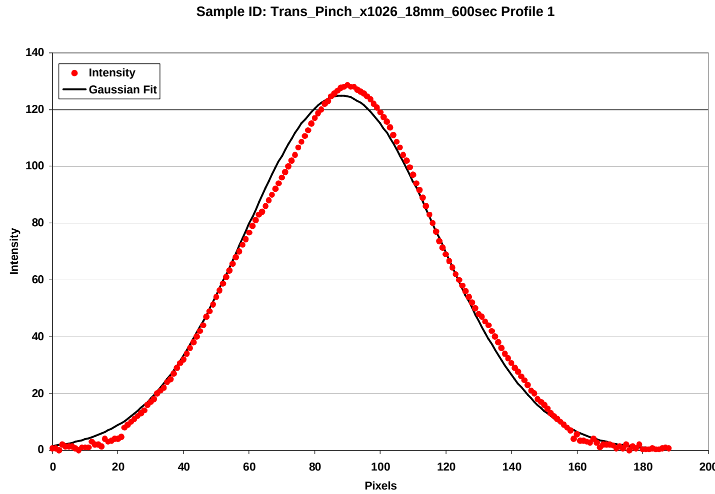

Figure 6: Comparison between the experimentally measured tritium concentration profile and an ideal Gaussian distribution.

Photo-emulsion Technique Results | Digital Technique Results

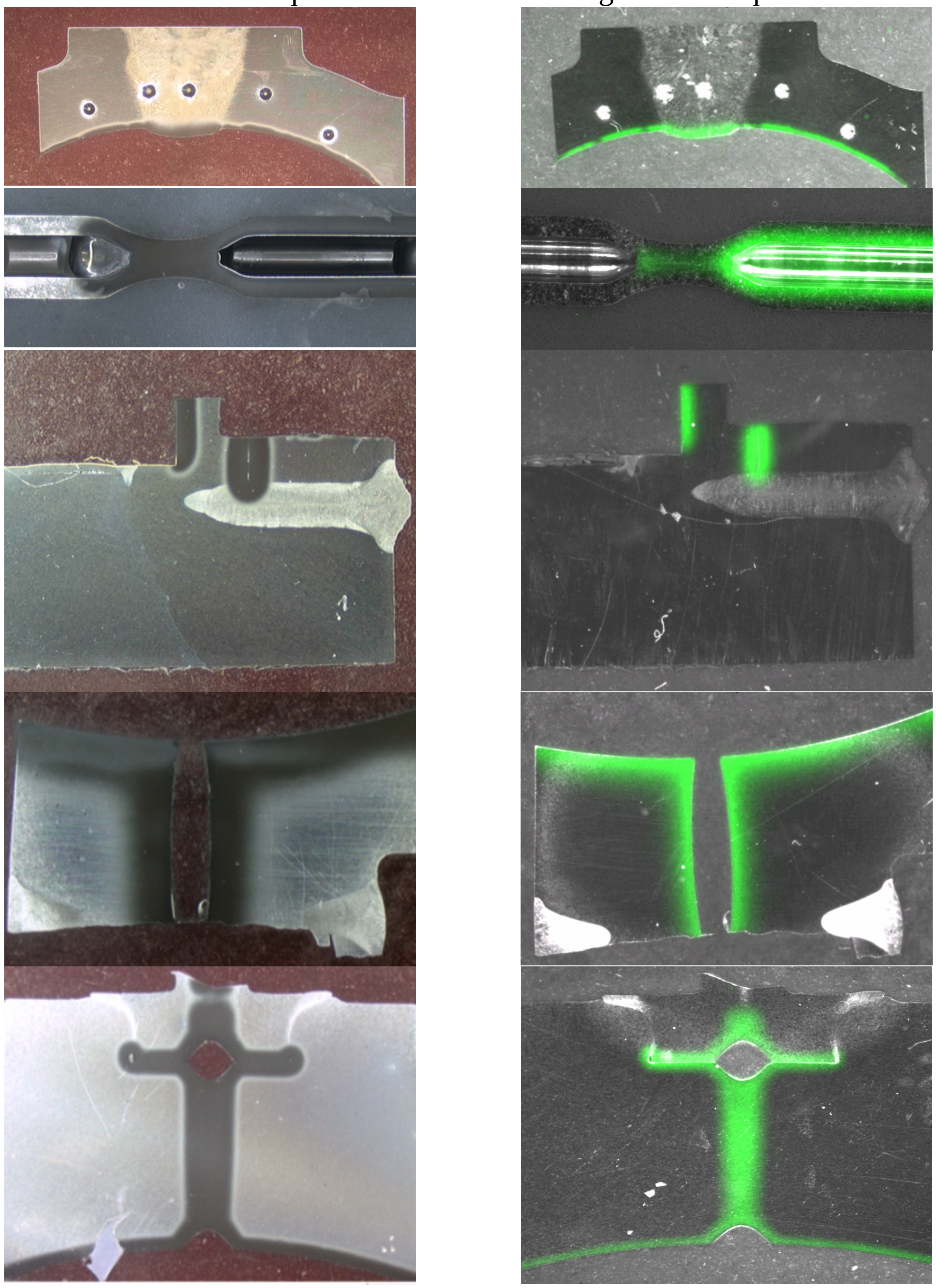

Figure 7: Comparison between the photo-emulsion method (left column) and the digital autoradiography method (right column) results for typical samples.