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X-ray Visualization of Carbon-Particle Oxidation Process in Supercritical Water

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Abstract : A real-time X-ray visualization system for low X-ray absorption materials has been developed. The system is mainly composed of a multi-color scintillator based image intensifier and a real-time image-processing unit. The color image intensifier has such advantages as the high sensitivity, the wide dynamic range and the long lifetime over the conventional one. The dynamic imaging of low X-ray absorption materials was realized by the video-rate image subtraction function of the image processor. The system has been successfully applied for an observation of a carbon-particle oxidation process in supercritical water. The low X-ray absorption difference between carbon and supercritical water, surrounded by high X-ray absorption metal wall, is one of the most difficult objects to get good image. In our system, the carbon-particle image was taken at a 30 frame/sec video-rate by continuously subtracting the background image until at the instance of the carbon-particle disappearance by oxidation.

Keywords : X-ray imaging, Color image intensifier, Real-time image processing, Supercritical water, Oxidation process.

1. Introduction

An X-ray inspection is a traditional non-destructive inspection method both in medical and industrial fields. Among many objects, some are easily inspected and others not, depending on their X-ray transmission and absorption properties. Low X-ray transmission materials surrounded by high transmission materials, like the thighbone in human body or metal tips mixed in cheese, are easily visualized. On the other hand, low X-ray absorption materials in high absorption materials, such as the human lung or the heart surrounded by ribs and water or oil in a metal pipe, are difficult for visualization.

The color image intensifier (Color I.I.) developed by some of the present authors is distinguished in its higher sensitivity for RGB color picture signals, its wider dynamic range and the longer lifetime than the conventional one (Nittoh et al., 2004).

By combining this Color I.I. with a newly developed image processor, a novel real-time X-ray inspection system has been developed. By this system, the objects being difficult to be inspected by X-ray can be clearly imaged even in the video picture.

The system was applied for a visualization of the carbon-particle oxidation process in the supercritical water (Sugiyama et al., 2002). A carbon-particle was put inside a thick metal tube where the high temperature and high pressure supercritical water flows. The evolution of the carbon-particle shape was measured and analyzed along with its density change.

2. Method for Visualization

An image subtraction method is often used for visualizing low X-ray absorption materials hidden in backgrounds.

Let the intensity of the transmitted X-ray through the background material (density ρ_B, mass absorption coefficient μ_B, thickness D), and through the object (density ρ, mass absorption coefficient μ, thickness d) designate I_B and I_S respectively. Then I_B and I_S are expressed in the following equations,

$$I_B = I_0 \exp(-\mu_B \rho_B D) \tag{1}$$

$$I_S = I_0 \exp(-\mu_B \rho_B (D-d)) \exp(-\mu\rho d) \tag{2}$$

where I_0 is the incident X-ray intensity.

When the quantities μ and ρ of the object are very close to those of the background material, or the object thickness d is very small, then $I_B \approx I_S$. This means the transmission difference between the object and the background is quite small. Subtracted signal ΔI is expressed as follows.

$$\begin{aligned} \Delta I &= \left| I_S - I_B \right| \\ &= I_B - I_S \text{ (When } I_B > I_S\text{)} \\ &= I_0 \exp(-\mu_B \rho_B D)(1 - \exp(-\Delta\overline{\mu\rho} d)) \\ &= I_B (1 - \exp(-\Delta\overline{\mu\rho} d)) \end{aligned} \tag{3}$$

where

$$\Delta\overline{\mu\rho} = \mu\rho - \mu_B \rho_B \tag{4}$$

In the case of $\Delta\overline{\mu\rho} d << 1$, equation (3) is approximated in the first order expansion of the exponential term as

$$\Delta I = I_B \Delta\overline{\mu\rho} d \tag{5}$$

In the case of the carbon-particle measurement in supercritical water, the object corresponds to the carbon-particle and the background to the supercritical water on the above equations. X-ray average energy is 60 keV and mass absorption coefficient of carbon and water are $\mu = 0.175$ cm²/g, $\mu_B = 0.206$ cm²/g, respectively, based on the NIST Table (Hubbel and Seltzer, 2004). The density of carbon and supercritical water are $\rho = 1.73$ g/cm³, $\rho_B = 0.32$ g/cm³, respectively, based on the NBS/NRC Table (Haar et al., 1984). Taking into account that the diameter of the carbon-particle is 0.4 cm, $\Delta\overline{\mu\rho} d = 0.095 << 1$ in equation (3), so that the equation (5) is fulfilled.

In order to get the above mentioned subtraction signal in two dimensional picture, an image subtraction is usually carried out by a personal-computer-based image processing software using the background and the object pictures which were taken separately.

In our newly developed image processor, the background image is stored in the frame memory and the object image can be continuously subtracted to produce the real-time subtraction video image. The system can visualize the low X-ray absorption materials in 30 frame/sec video-rates, which conventional systems can hardly visualize.

3. Construction of X-ray Imaging System

The block-diagram of the X-ray imaging system is shown in Fig. 1. The Color I.I. is approximately six times higher in its sensitivity and two orders wider in its dynamic range than the conventional image intensifiers (Nittoh et al., 2004). Low X-ray transmission objects through high transmission objects can be simultaneously taken in an image by using red, green and blue color component signals. The image processor has VGA sized (640×480) frame memory with 16 bit R,G,B color component and has various functions such as background subtraction, tone-curve adjustment, image inversion and pseudo-coloration. NTSC-based processed video signals can be recorded in a HDD video recorder or captured by a PC-based image capture system. One of the important functions of the image processor is an accumulation of signals into the frame memory. Twenty-four bit color (8 bit/color×3) VGA signals from Color I.I. are accumulated for 256 frames (corresponds to about 8.5 seconds) to produce the 48 bit signals (16 bit/color×3), which shows higher statistic accuracy especially in the case of the image subtraction.

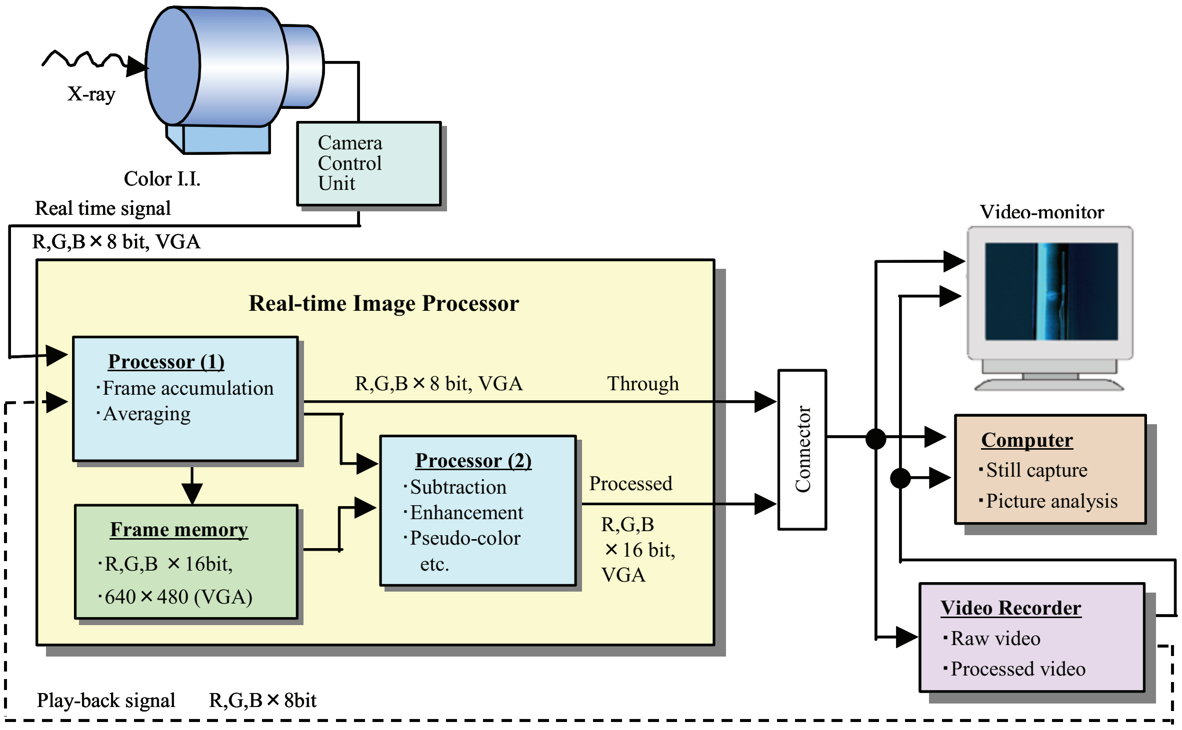


Fig.1. Block-diagram of X-ray image processing system.

4. Carbon-Particle Measurement in Supercritical Water

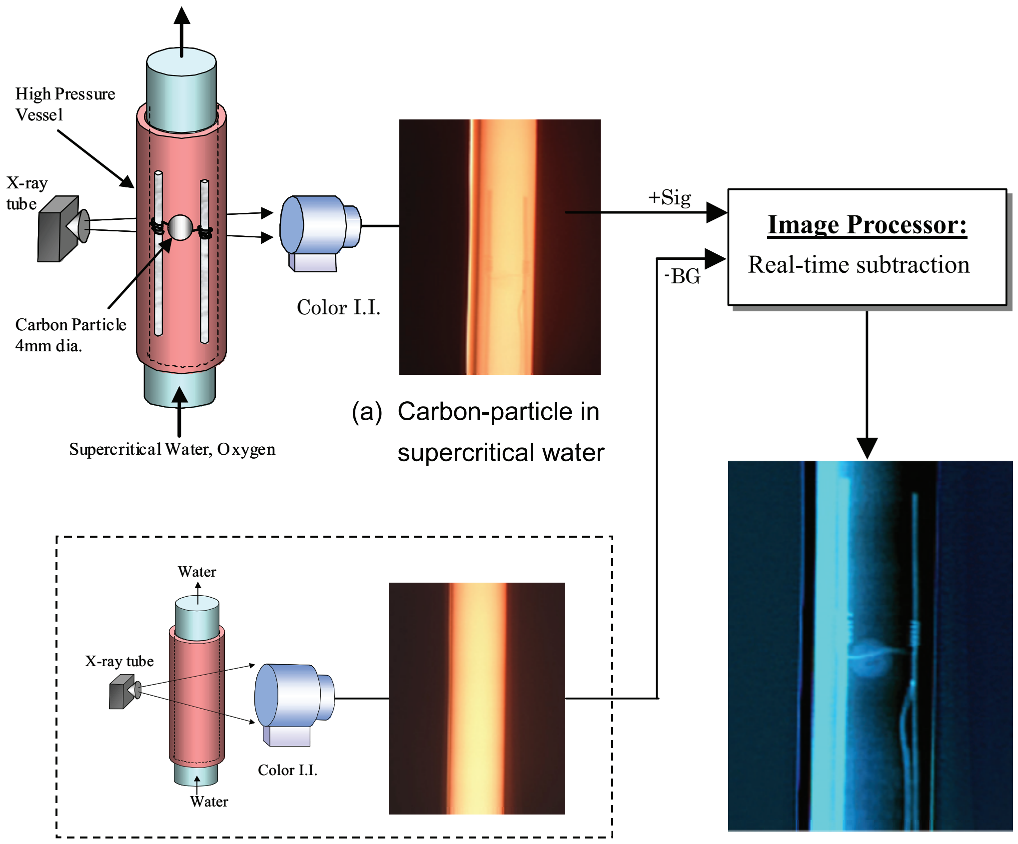
The X-ray imaging system was applied to direct observation of the carbon-particle oxidation process in supercritical water along with injection of H_2O_2.

Here, the carbon-particle imaging method and time dependent carbon-particle size and density change measurement are presented. Precise oxidation process analysis will be presented elsewhere (Fujie et al., 2005).

4.1 Visualization of Carbon-Particle by Subtraction Method

A carbon-particle of 4mm diameter with the density of 1.73 g/cm^3 (graphite) was hung by metal rods and a wire inside the Inconel tubular vessel of 11mm inner diameter and 4mm thickness as schematically shown in Fig. 2(a). A thermal insulator covers the vessel, although not shown in the figure, and supercritical water typically with 500°C temperature and 30 MPa pressure flows upward. The X-ray tube and 4 inch Color I.I. is set perpendicular to the flow with 70 cm distance.

The X-ray image of the carbon-particle under 500 °C, 30 MPa condition is shown in Fig. 2(a), where the X-ray condition is 140 kVp and 3 mA with a 1.5 mm Cu filter. The carbon-particle could hardly be imaged due to its low X-ray absorption difference between the carbon and the supercritical water. The background image without the carbon-particle in 500 °C, 30 MPa supercritical water had been stored in the frame memory of the image processor, as shown in Fig. 2(b), prior to the carbon-particle oxidation experiment. By subtracting the background image, the carbon-particle could be imaged as in Fig. 2(c). The image processor works at 30 frame/sec video-rates from NTSC input signal to processed output signal in real-time, which can be monitored on TV. Figure 2 shows the 256 frame accumulated 48 bit (16 bit for each color) image for better statistic accuracy. The subtracted image in Fig. 2(c) is not spatially accurate for pixel-to-pixel basis, because the experimental arrangement moved slightly between object and background measurement, due to the carbon-particle installation inside and repeated high temperature cycles. Despite the subtraction inaccuracy, the real-time visualization is important for the process control, otherwise we can't continue the experiment.



(a) Carbon-particle in supercritical water

(b) Background without carbon-particle

(c) Background subtracted image

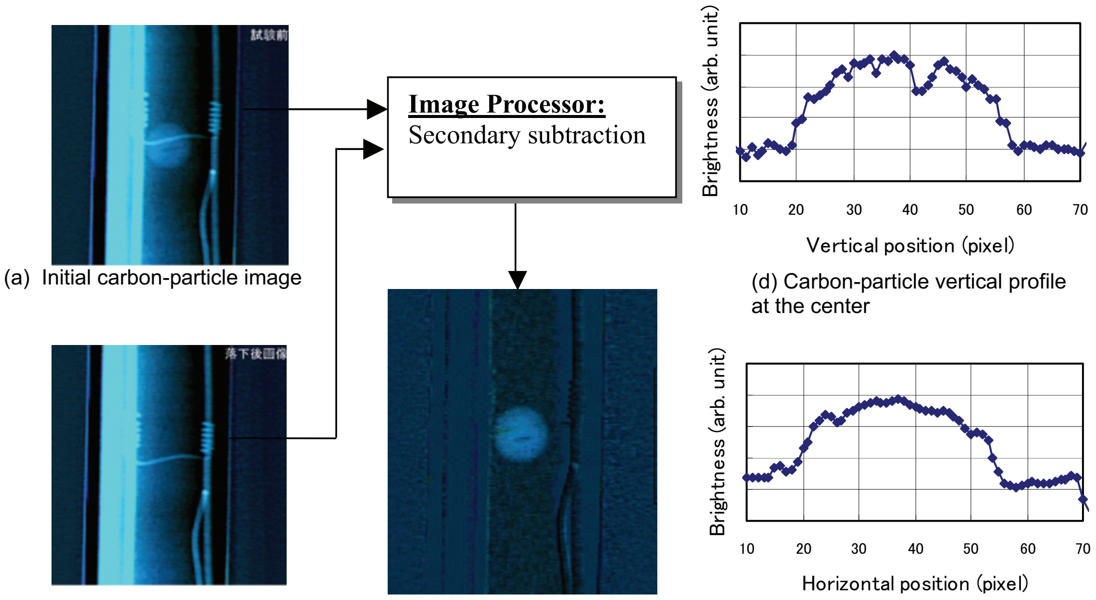
Fig. 2. Dynamic carbon-particle image extraction by subtraction of background image.

4.2 Time-Dependent Carbon-Particle Size and Density Measurement

After H_2O_2 was injected into the supercritical water, the carbon-particle began oxidation. Real-time subtracted images corresponding to Fig. 2(c) were continuously recorded on HDD video recorder and the still picture was also captured in about every 30 minutes until the instance of its disappearance.

The carbon-particle disappeared about 5 hour 33 minutes after the injection of H_2O_2, and then the experiment finished. The picture after the carbon-particle has disappeared is shown in Fig. 3(b) together with its initial picture in Fig. 3(a). These pictures contain unnecessary rods and wire, and they also have brightness gradient due to the subtraction misalignment.

Secondary image processing was carried out for the purpose of the carbon-particle oxidation analysis. It was done by putting the initially subtracted HDD video signal to the image processor as "play-back" mode shown in dashed line of Fig. 1. The image of Fig. 3(b), after the carbon-particle had disappeared, was stored in the frame memory as the background image. Figure 3(c) is a typical image of the secondary subtraction. Background gradient, the wire or the rods are canceled and the clear graphite image can be taken by this method. Spatial intensity profiles of the carbon-particle for vertical and horizontal axes are exhibited in Fig. 3(d) and Fig. 3(e), respectively.



(a) Initial carbon-particle image

(d) Carbon-particle vertical profile at the center

(b) Image after carbon-particle disappeared

(c) Secondary subtracted carbon-particle image

(e) Carbon-particle horizontal profile at the center

Fig. 3. Carbon-particle image after secondary subtraction and its intensity profile.

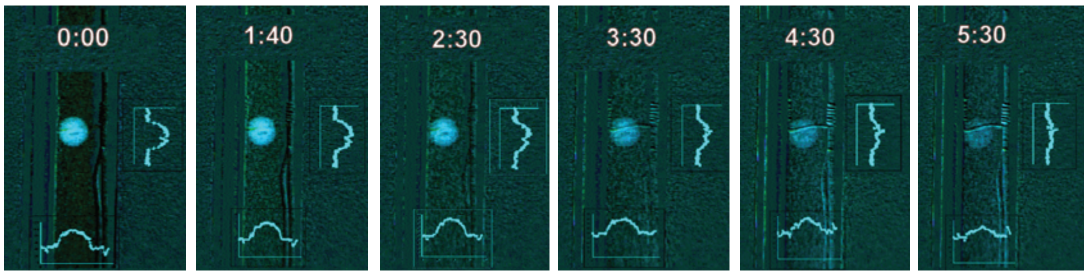
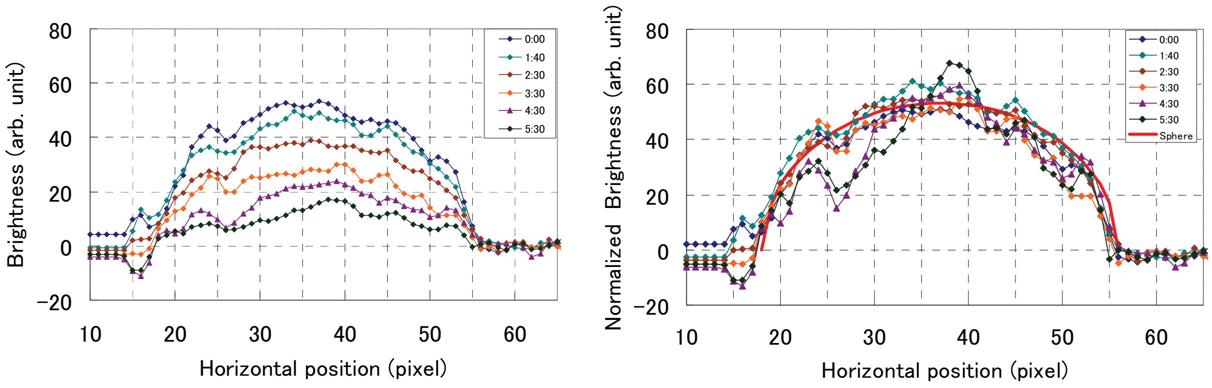


Fig. 4. Time-dependent carbon-particle image change until just before its disappearance.

Time dependent still pictures from the start to just before the disappearance are shown in Fig. 4 together with their spatial profiles. In order to analyze the density and the shape, horizontal profiles at each time are plotted in Fig. 5(a). The carbon-particle brightness is decreasing monotonically along with the lapse of time. Figure 5(b) is peak-area and the baseline normalized brightness profiles of Fig. 5(a) together with theoretical sphere transmitted intensity. As shown in the figure, the shape and the size of the carbon-particle were kept almost constant until it disappeared.



(a) Carbon-particle profile during the oxidation

(b) Brightness- normalized profile and theoretical sphere transmission profile

Fig. 5. Brightness and profile change of the carbon-particle during the oxidation.

4.3 Disappearance of Carbon-Particle

The moment of the carbon-particle disappearance was confirmed by secondary subtraction measurement of HDD recorded video signal that the particle disappeared in a very short time. Figure 6 shows the secondary subtracted video signals at the very instance of the carbon-particle collapse. Images were captured at each successive GOP (Group Of Pictures) of MPEG2 video signals in HDD recorder without any accumulation or averaging, so that they contain higher statistic noise. Despite the noise in the image, it could be clearly identified that the carbon-particle disappeared within two seconds. Of course it can be seen in dynamic images with 30 frame/sec video, where statistic noise seems slightly lower than the still image due to the compensation function of the human eyes.

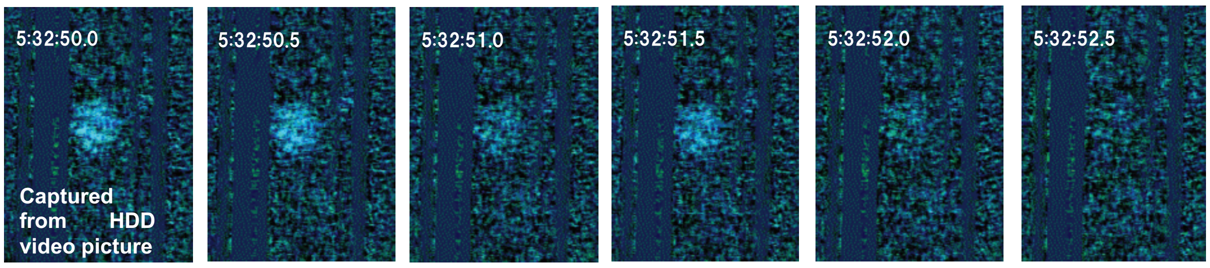


Fig. 6. Instantaneous disappearance of the carbon-particle
(Captured and processed from MPEG2 video in HDD recorder).

Based on the analysis of the chemists working together with the authors, the carbon-particle had gradually oxidized from the inside without any visible outer-shape change. Then, finally, the ash left behind suddenly collapsed (Fujie et al., 2005). They evaluate that this is the first observation of the latent reaction progress in supercritical water from outside.

5. Conclusion

A real-time, video-rate X-ray visualization system has been developed by combining a newly developed color image intensifier with also newly developed image processor. Objects of low X-ray absorption difference could be clearly imaged using its powerful video-rate subtraction and image manipulation function.

The system was successfully applied for visualization of a carbon-particle oxidation process in a thick metal vessel where high temperature and high pressure supercritical water flows. It was confirmed that the system is effective not only for the shape measurement but for the density measurement. Furthermore, by simply recording X-ray transmitted video images, just like recording TV program in VTR, desired image processing can be carried out using play-back mode of HDD recorded signals.

The authors hope the system will contribute to the visualization of latent objects or phenomena in wide area of X-ray inspections.

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