OBSERVATION OF OZONE EXPLOSION OF LIQUID NITROGEN INDUCED BY IRRADIATION WITH ELECTRON LINEAR ACCELERATOR*

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Abstract

A pulsed electron radiography system has been developed, which consisted of an electron linear accelerator, a scintillation screen and a high sensitivity image sensor. High contrast penetrating images were obtained by modulating of energy of electron beam. In addition, the system was capable for high speed strobo-imaging with the time resolution of about a few micro-second. By the use of this system, we observed successfully an ozone explosion phenomenon of liquid nitrogen induced by electron irradiation, which has been considered to be a serious problem for low temperature irradiation experiments.

INTRODUCTION

Some reports pointed out that liquid nitrogen becomes to be dangerous to explode by radiation irradiation[1)]. The phenomenon is called as the ozone explosion, for the reason that the ozone was observed to be accumulated in the liquid nitrogen by irradiation. Many considered to be induced the explosion by radically decomposition of the ozone. However, the detail of this phenomenon is not clear by the reason that the reproducibility of the phenomenon was insufficient and it occurred only in the severe condition such as intense radiation field.

In order to investigate the phenomenon, we have been developing a new type of radiography system by the use of an electron linear accelerator. The electron radiography system is considered to have two advantages. One is the time resolution. The beam from an electron linear accelerator is pulsed basically. The penetrating image obtained by such beams is strobo-images with the time resolution corresponds to the pulse width of electron beam. The system is considered to be suitable to observe a moving object such as the explosion phenomenon. The other point is that the electron beam can be use for double purpose, the probing beam and the irradiation beam.

Additionally, the attenuation behaviour of high energy electrons in materials is quite different from that of X rays, γ rays and neutrons which are used in conventional radiography. The attenuation curve of electron is not similar to exponential, rather to rectangular. Near the region of the electron range, the electron beam becomes to decrease suddenly. Therefore, a high contrast penetrating image would be obtained by modulating of energy of electron beam.

STROBO-IMAGING

An example of the pulse shape of electron linear accelerator used is shown in Fig. 1. The pulse width was about 2 μ sec. A high speed strobo-radiograph is expected to be realized by the use of the electron beam. The outline of the electron radiography system developed is shown in Fig. 2. Electrons pass through a sample and are converted to photons with a ZnS(Ag) scintillation screen. The electron penetrating image projected on the scintillation screen are measured by a SIT-TV camera and sent to a computer.

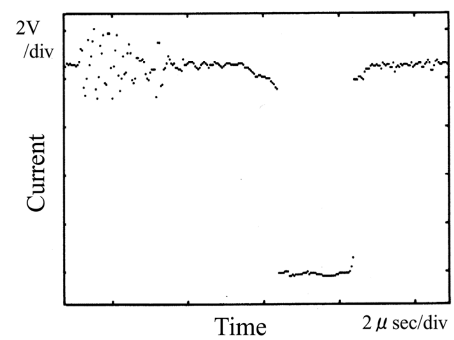

Figure 1: Pulse shape of the electron linear accelerator.

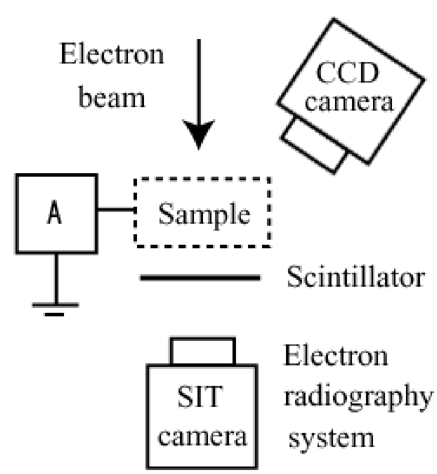

Figure 2: Electron radiography system.

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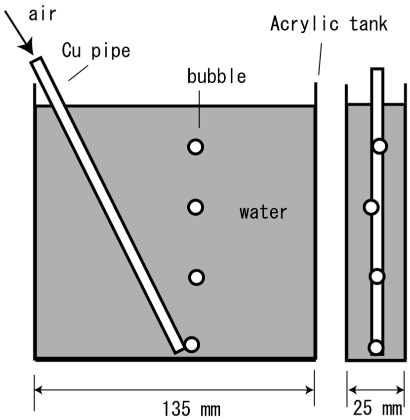
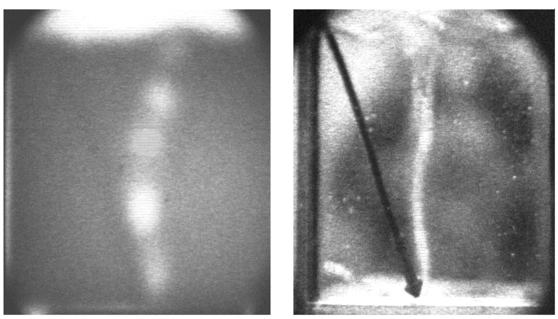


Figure 3: Moving object for testing the electron strobo-radiography system.



(a) Electron Strobo-image (b) Optical video image

Figure 4: Comparison of the electron strobo-image and the normal optical video image by the use of the image of bubbles rising in the water.

Next, the time resolution of the electron radiography for moving object is estimated. Figure 3 shows an outside view of an acrylic tank used in the evaluation experiment. The tank was filled with water. Air was blown into the water with a copper pipe and the bubbles were generated at the end of the pipe continuously. In the experiment, the bubbles played the moving objects. The electron radiography system and a normal CCD camera were observed the bubbles rising in water. Figure 4 shows examples of the results. The electron energy was 5MeV, the beam current was 1 μ A and the beam repetition was 15Hz. Both of the optical image and the electron radiograph was one frame image (1/30 sec exposures). In the optical image (Fig. 4(b)), the bubbles were seen as a continuous tube-like image and the individual bubble could not be discriminated. In the electron radiograph (Fig. 4(a)), the individual images of bubbles were seen. The rising speed of the bubble was measured about 0.33 m/sec. A bubble image moves about 1cm by one frame interval (1/30 sec). Normal video rate is considered to be insufficient for obtained a frozen image of the rising bubbles. In the case of the electron radiograph, the time resolution was determined by the electron pulse width (2 μ sec). The high seed strobo-imaging was considered to be realized by the electron radiography, which exposure time corresponded to the shutter speed of 1/500000 sec.

OZONE EXPLOSION

Next, we tried to explode liquid nitrogen artificially by electron irradiation and to observe the process by the use of the electron radiography system. Figure 5 shows a Pyrex dewar used for the liquid nitrogen target. During the electron irradiation, liquid nitrogen was supplied continuously. The outside view of the dewar was observed by a CCD camera and the inside condition was measured by the electron radiography system. The explosion occurred after 6 minutes of electron irradiation in the condition that the electron energy was 6 MeV and beam current was 2 μ A. The sequential frames of electron penetrating images at the explosion were shown in Fig. 6. At the moment of the explosion, a void appeared in the upper-left corner of the dewar.

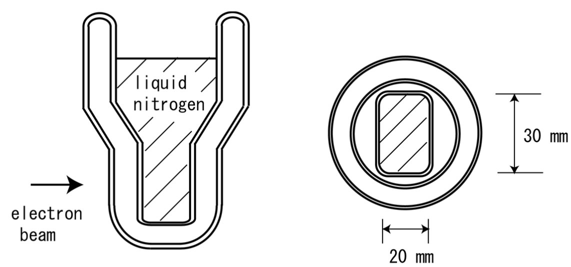
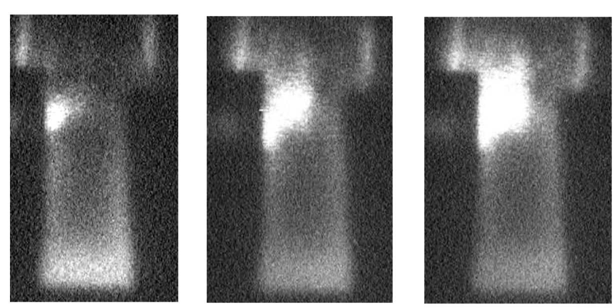


Figure 5: Dewar flask for liquid nitrogen using in the explosion experiment.



(a) 0/30sec (b) 1/30sec (c) 2/30sec

Figure 6: Sequential video frame of electron penetration image at the explosion.

DISCUSSION

Many researchers believe that the ozone is the major cause of the explosion. The process has been considered as following.

(1) Ozone is generated by irradiation from the solved oxygen in the liquid nitrogen.

(2) According to increase of ozone concentration, solid ozone starts to educe in the liquid nitrogen. Liquid nitrogen is colored gradually to deep blue.
(3) Finally, the ozone decomposes with heat generation. The heat enhances the decomposition of ozone and the reaction proceeds explosively.

Accordingly, the phenomenon is called as ozone explosion. However, the detail of the process is not clear. For example, the liquid nitrogen did not explode only by injecting of ozone artificially. The explosion occurs only under the radiation condition. There is a probability that some radiation effects take part in the explosion.

Figure 7 shows the one flame image at the moment of the explosion. At the time, liquid nitrogen flashed luminescence. Some spark-like-patterns are seen on the surface of the dewar. Next moment (after 1/30 sec), the dewar exploded and the fragments flew off (Fig. 8). The figure shows that the explosion is similar to spark discharge rather than an ozone explosion. The liquid nitrogen is insulator and it can be charged up by electron irradiation. The discharge is considered to be one of the probable reasons of the explosion.

Figure 9 shows the images jointed of fragments of dewar flask exploded. The inner vessel is damaged radically. It suggests that the explosion is occurred in the inner vessel.

Some lighting patterns were seen in Fig. 7. However, the pattern did not correspond to the rupture line of outer vessel. This fact indicates that a large-scale discharge occurred on the surface of dewar independently of rupture of the vessel.

On the other side, the shape of fright fragment shown in Fig. 8 seems to be similar to the rupture pattern shown in Fig. 9. This fact indicates that the upper region of the outer vessel blew off in the first moment of the explosion.

On the whole, the following facts were confirmed by the experiment.

(1) The explosion started at the liquid surface of the nitrogen.
(2) The discharge occurred on the surface of the dewar.

The part of the discharge phenomenon played in the explosion process is not clear. The seemingly most reasonable explanation would be that the discharge is the ignition of the explosion.

CONCLUDING REMARKS

A pulsed electron radiography system by using an electron linear accelerator has been developed. By the use of this system, ozone explosion phenomenon of liquid nitrogen induced by electron irradiation was observed successfully. In this experiment, the explosion and the discharge are observed simultaneously. The phenomenon seems to be complex process combined of ozone and discharge.

REFERENCES

[1] M.Takehisa, K.Yoshida, "JAERI-M6363",1976.
[2] Strobo-Radiography System by using Electron Linear Accelerator, R. Taniguchi, T. Araki, T. Kojima, S. Okuda, Proc. 3rd US-Japan Symposium on Advancing Applications and Capabilities in NDE, pp401-406(Hawaii, US, June 20-24,2005).

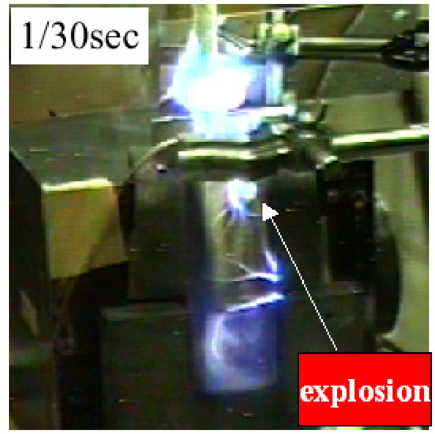


Figure 7: One flame image at the moment of explosion.

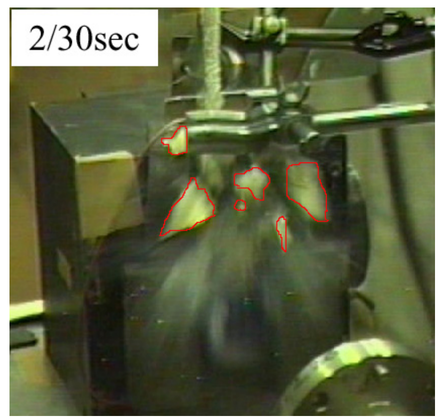


Figure 8: One flame image after the explosion. Fragments of broken dewar are enclosed by red line.

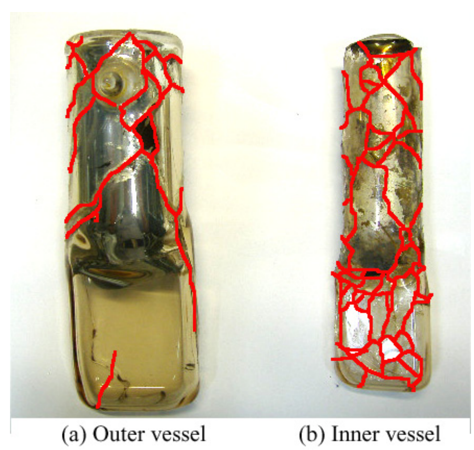


Figure 9: Restoration photographs by jointing of fragments of dewar flask exploded.