DEVELOPMENT OF A FIBER LASER FOR IMPROVING THE PULSE RADIOLYSIS SYSTEM *

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Abstract

At Waseda University, we have been developing a pulse radiolysis system in order to clarify the early chemical reactions by ionizing radiation with S-band Cs-Te photo cathode RF-gun. According to our measurement, sample deterioration was observed even in the low absorption dose. Thus we need a high power and stable pulsed laser as a probe light. We constructed and tested Er fiber laser oscillator as a new probe laser. In this conference, we will introduce our Er fiber laser, experimental results of pulse radiolysis with Er fiber laser as a probe light and future prospective.

INTRODUCTION

In these days, the radiation is applied in various fields, for example semiconductor integrated circuit manufacturing. Semiconductor integrated circuit is conducted by transferring circuit pattern master called photomask on silicon wafer. To make this photomask, electron lithography is used. In this process, a resist material, which is high reactivity with the radiation, is used but most of resist's reaction mechanism was not proved yet.

When material is irradiated, a short-lived and highly reactive substance, radical, ion and exited states etc, called intermediate active species are made (fs~ps). Then the intermediate active species react with around substances (ps~μs). In this phase a principal chemical reaction is determined by intermediate active species in early process. So proving the behavior of intermediate active species is important for understanding and controlling radiation chemical reaction. A pulse radiolysis is one of the methods to measure the behavior of intermediate species by using an electron beam pulse. At Waseda University, we are using a Cs-Te RF electron gun for the radiation chemical analysis.

Now we introduce a purpose to investigate a Supercontinuum ray (SC ray) as a probe ray to improve pulse radiolysis system. In pulse radiolysis we measure a light absorption by intermediate active species. As a probe light, we have investigated a SC ray using Yb fiber laser and PCF (Photonic Crystal Fiber). But we haven't achieved a stable prove light in the visible region[1]. Therefore we built Er fiber laser oscillator as new prove ray source. Now we succeeded in generating a second harmonic of Er fiber laser and measuring of hydrated electron in ns time resolution. In this paper, we report current results about generation SC ray in the visible region, improvement of Er fiber for measurement of ps time resolution and analysis of dose rate effect against the hydrated electron.

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PULSE RADIOLYSIS

Pulse radiolysis experiment consists of sample, radiation (producing intermediate active species) and probe light. We use the Cs-Te cathode RF electron gun as a radiation source. An intermediate active species are produced by irradiating an electron beam on the sample. And the probe light, simultaneously incident to the sample, is absorbed depends on the intermediate active species concentration. Specifically, we measure the temporal change of O.D.(: Optical Density) :

$$\mathrm{O.D.} \equiv \log\frac{I_0}{I} = \varepsilon c l \quad (1)$$

where, I_0, I, ε, c, l are the incident light intensity, transmitted light intensity, the molar absorption coefficient, the active main concentration and the interaction length, respectively. From this O.D. we can obtain the amount of intermediate active species. We evaluate our pulse radiolysis system by measuring hydrated electrons. The hydrated electron is the free electrons that have been stabilized by coordination of water molecules. This has the broadband transient absorption around 720 nm and about 1 μs decay time. Relationship of the dose and decay time of hydrated electron is shown in Figure 1. We used Xe flash lamp as a probe light. As shown in Figure 1, the decay time of the hydrated electron changes immediately in the low absorbed dose. In particular, the decay time change of the high charge beam was significant in this pulse radiolysis system. It means that the sample deteriorates due to electron beam. When water is irradiated, many kinds of reactions occur. After a series of reaction final products (H_2O_2, H_2, etc) are generated and accumulated in water. Especially H_2O_2 reacts with hydrated electron well. So as more water is irradiated, the decay time of hydrated electron descends. On the other hands, the relationship of the dose and peak O.D. of hydrated electron is shown in Figure 2. Figure 2 shows that peak O.D. is constant against the dose. This also means that we can use the peak O.D. of hydrated electron to evaluate this system even from a deteriorated sample. However, it should be reduced the number of irradiations during the measurement (average number of times). To this purpose, the high intensity and stable probe light is required. Also, from the viewpoint of chemical reaction analysis, broadband and short pulse is required for the probe light. Thus, we have developed a SC light as the probe light. SC light is a broadband pulse laser. SC light is produced by ultrashort pulse IR laser from Yb fiber laser oscillator with a non-linear optical effect in PCF. The center oscillation

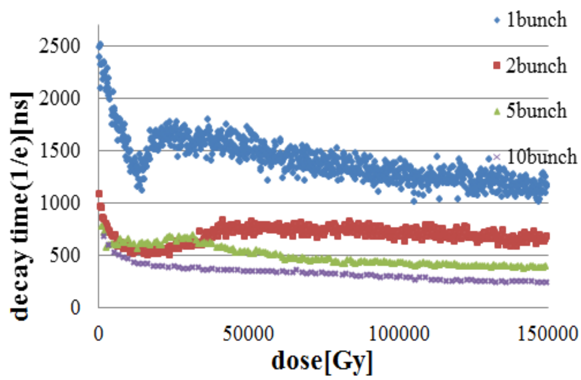

Figure 1: Relationship of the dose and decay time of hydrated electron.

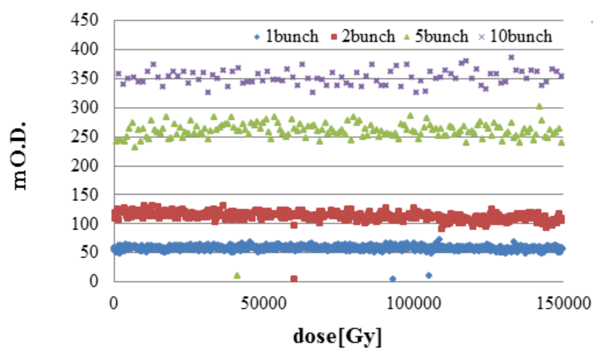

Figure 2: Relationship of the dose and peak O.D..

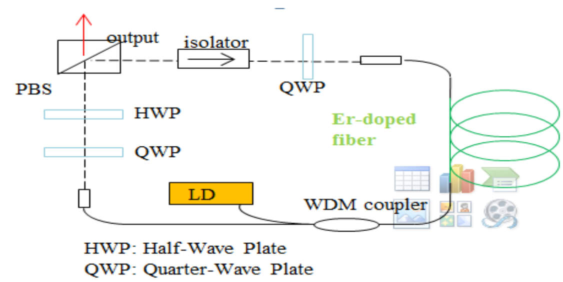

Figure 3: Schematic of Er fiber laser.

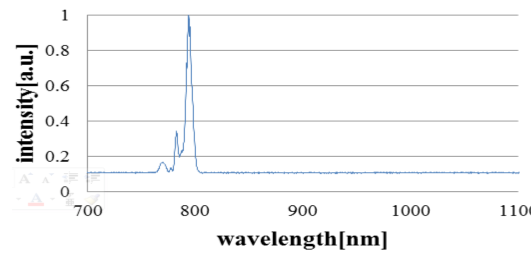

Figure 4: Spectrum of second harmonic.

Table 1: Performance of Er Fiber Laser

Center wavelength	1550 nm
Spectrum width (FWHM)	About 20 nm
Pulse width (FWHM, calculated)	About 50 fs
Repetition frequency	43.4 MHz
Output	26.8 mW
LD-Pulse Conversion efficiency	8.8 %

wavelength of Yb fiber laser is 1030 nm and we have been successful to broadband IR laser from 800 nm to 1030 nm [1]. However, it is difficult to broad to visible light region because of laser power shortage. Moreover intensity of the short wavelength part was unstable. Therefore we started to build Er fiber laser oscillator for the purpose of a further strengthening of the probe light and stabilization. The center oscillation wavelength of Er fiber laser is 1550 nm. It is expected that generating highly stable and high power at visible light region by broading a second harmonic(775 nm) of Er fiber laser.

Er FIBER LASER

Er fiber laser oscillator has a 1550 nm center wavelength. It is possible to oscillate a fs pulsed laser by a mode-locking by NLPR (Non-Linear Polarization Rotation) [2]. Figure 3 shows a schematic of Er fiber laser oscillator. Broadening of the pulse width due to the dispersion in the fiber is suppressed by the negative dispersion of the Er-doped fiber. We adopted a backward pumping to the Er fiber in order to increase the power. The performance of Er oscillator is shown in Table 1. Here, the pulse width is calculated value from the Fourier limited pulse width determined from the spectral width. We expect that the actual pulse width would be longer than Tab. 1. The resulting IR pulse laser was converted by the nonlinear optical crystal to the second harmonic. Spectrum of the second harmonic is shown in figure 4.

EXPERIMENT

We operated nanosecond pulse radiolysis to observe hydrated electron using a second harmonic of Er fiber laser at 775 nm. Figure 5 shows the setup of pulse radiolysis system and Table 2 shows the parameters of the RF electron gun. In figure 5, DET and SHG means detector and nonlinear optical crystal generating the second harmonic. Second harmonic from Er fiber laser was split into two by the half mirror. One passed through the sample (:I) and the other was used for the reference (:I_0). Beam and probe light were injected at same axis to the sample. FCT (Fast Current Transformer) and dipole magnet are used for measuring the charge and the energy. We finely tuned the beam with steering magnet and Q-magnet. The sample of water was bubbled for 10 minutes by Ar. Results of 1 bunch and 10 bunches are shown in Figure 6. Peak O.D., noise and decay time of 1 bunch were 60.5 mO.D., 4.84 mO.D.(rms) and 1132.9 ns respectively. The decay time of the signal is defined as the time until 1 / e of the peak O.D. and calculated by fitting exponential function. Decay time was about 1 μs and it means that hydrated electrons were correctly observed. On the other hand, results of 10 bunches were 427.5 mO.D., 4.96mO.D. (rms) and 306.4 ns respectively. This decay time is shorter than that

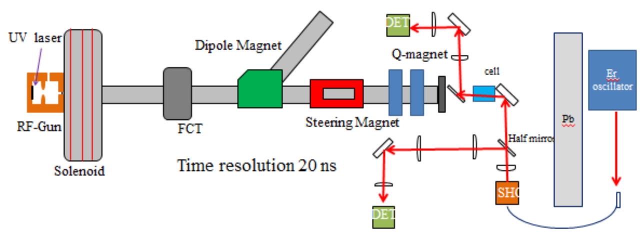

Figure 5: Schematic of our pulse radiolysis system.

Table 2: The Parameter of Electron Beam

Charge	2 – 3 nc/pulse
Energy	4 – 4.5 MeV
ビームサイズ(rms)	X – 1.37 mm
	Y – 1.22 mm
バンチ長(rms)	4 ps

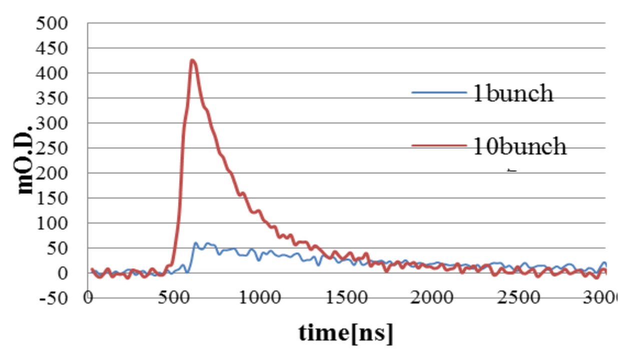

Figure 6: Optical density of hydrated electron at 775nm.

of 1 bunch. It is considered that the sample was deteriorated by high charged beam. Even the same average number of times in 1 bunch and 10 bunches, dose of 10 bunches is 10 times larger than 1 bunch. Therefore, deterioration of the sample is prominent towards the 10 bunch in comparison with the 1 bunch. Concerning to the decay time difference between 1 bunch and 10 bunches, the dose rate effect would enhance the deterioration of the sample. The dose rate effect is a phenomenon that radiation chemical reaction changes in different absorbed dose. The main decay reactions of hydrated electrons are secondary reactions. Thus the decay time strongly depends on the dose. Figure 6 shows that we obtain sufficient peak against noise with Er fiber laser as a probe light. It also means that we could measure hydrated electron much less number of average with Er fiver laser than Yb fiber laser. And we succeeded in observing the signal in the 1 bunch, 16 averages. However this was insufficient in the view of the radiation chemistry analysis yet. In the future, further improvement the strength of the probe light is required in order to reduce the number measurement times to avoid the sample deterioration.

CONCLUSION

In this paper, it was confirmed that second harmonic from Er fiber laser is useful as probe light in pulse radiolysis. Using Er fiber laser reduces the number of times of irradiation as compared with Yb fiber laser. Also, it is expected to realize picosecond pulse radiolysis system by observing the signal at 1 bunch. In the future, we plan to construct a picosecond pulse radiolysis system in the visible light region. SC light is generated by broadening the second harmonic of Er fiber laser in the PCF. It is expected that the strength of probe light descends significantly than that of the current situation. Now we are working to increase the power of the second harmonic for stabilization SC light generation in the visible light region. After improvement, picosecond pulse radiolysis, which is based on the stroboscopic method, would be realized.

REFERENCE

[1] Y. Soeta *et al.*,“Improvement pulseradyolysis system by introducing femtosecond pulsed laser”, in *Proc. of the 12th Annual meeting of Particle Accelerator Society of Japan*, August 2015, Tsuruga, Japan.

[2] R. Suzuki *et al.,* “Development of a mode-locked Yb fiber laser based on nlpr”, in *Proc. 11th Annual meeting of Particle Accelerator Society of Japan,* August 911, 2014, Aomori, Japan.