DEVELOPMENT OF THE PULSE RADIOLYSIS SYSTEM WITH A SUPERCONTINUUM RADIATION USING PHOTONIC CRYSTAL FIBER*

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Abstract

In usage of radiation, it is important to study the process of chemical effects of ionizing radiation in a material. Pulse radiolysis is a method to trace these rapid initial chemical reactions by ionizing radiation. As a pump beam, we are using about 4 MeV electron beam. In nanosecond timescale pulse radiolysis, it is required the stable probe light of a broad spectra. And especially in picosecond timescale pulse radiolysis, probe light should have short pulse width to use stroboscopic method. Therefore, in order to develop a unified timescale experimental system, we developed Supercontinuum (SC) light as a probe light, which is generated by nonlinear optical process of short pulse IR laser in photonic crystal fiber (PCF). To apply SC light as a probe light of pulse radiolysis, we measured its properties. We tried to measure the absorption spectra of hydrated electron by SC light. Then we successfully observed good signal-noise ratio data both in nanosecond and in picosecond experiment with the same system. In a further attempt, we succeeded to improve stability of SC light.

INTRODUCTION

Radiation Chemical Reaction

The radiation chemical reactions are used for radiation therapy, semiconductor integration, and nuclear energy development. In primary processes of the radiation chemical reaction, molecules of materials are ionized and/or excited, and many active species are generated. After these processes, intermediate active species are reacted each other as an ordinary chemical reaction. Thus, researching the behavior of the active species leads to forecast all over chemical reactions by ionizing radiation.

Pulse Radiolysis

Although the properties of irradiated materials can be researched by radioisotope, the intermediate active species have too short life to observe. Pulse radiolysis is a powerful method to observe these active species and trace the rapid chemical reactions using pulse radiation, which introduced by accelerator. We have been studying and developing pump-probe absorption spectroscopy [2], which is well- known method of pulse radiolysis. In this method, the pump beam and the probe light are necessary. As a pump beam, we are using 4 MeV electron beam introduced by photocathode RF-Gun [1]. When we observe the picosecond time scale behavior of active species, pump-probe pulse radiolysis with stroboscopic method is used. In stroboscopic method, the probe light should have short pulse width compared with the need time scale. We generate short pulse white light from short pulse IR laser by nonlinear optical effect. We used to use water cell to generate the white light, however, it was not stable enough and needed about 500 μJ/pulse or higher intensity laser to generate the white light.

Photonic Crystal Fiber

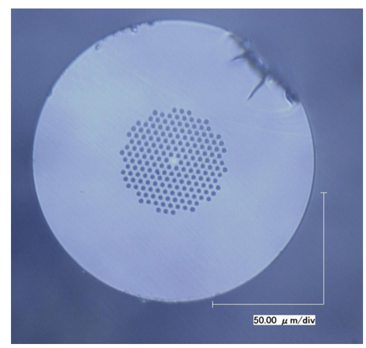

Figure 1: The cross sectional micrograph of PCF

By using photonic crystal fiber (PCF), we can introduce short pulse white light. PCF is optical fiber that is made of silica, and clad layer has micro air holes. The cross sectional micrograph of PCF obtained by microscope is shown in Fig.. 1. Because PCF has the several micrometer diameter core layer, IR laser concentrates very narrow space and gets high peak intensity, short pulse laser spreads its spectra by nonlinear optical effect through PCF [3]. Then we can obtain short pulse white light (Supercontinuum (SC) light). To generate SC light, required energy of laser is ~40 nJ/pulse. This energy is much lower than the energy that of water cell.

EXPERIMENTAL SETUP

Supercontinuum Generation

To apply SC light as a probe light of pulse radiolysis, we firstly tried to generate a SC radiation by passing IR laser through PCF, and measured its spectra and stability. Properties of IR laser are shown in Table 1. IR laser was amplified up to about 2000 times by Nd: YLF crystal pumped by flash lamp, and injected into PCF by achromatic objective lens. In front of the objective lens, laser average power was about 20 mW without amplification. We used 10 m-long F-NL-5/1040 (Newport) PCF. Generated SC was measured the spectrum by using monochromator and photodiodes.

Table1: Main properties of IR laser

Wavelength	1047 nm
Laser Medium	Nd: YLF
Pulse Width (FWHM)	10 ps
Average Power	~200 mW
Repetition Rate	119 MHz

Picosecond Pulse Radiolysis with SC Light

Experiamental setup of pump-probe pulse radiolysis with SC is shown in Fig. 2. The pump beam was about 4 MeV electron beam obtained by photocathode RF-Gun, RF frequency is 2.8 GHz and cathode material is Cs-Te. The repetition frequency of laser pulse is synchronized with RF. IR laser is converted into UV laser by using the two nonlinear optical crystals, and the UV pulse is irradiated to cathode of RF-Gun. UV pulse number is able to change by using pulse picker. Number of the bunch and bunch length depend on UV pulse number and pulse width, so it is also changeable [1]. To obtain the beam which has ~10 ps bunch length for picosecond pulse radiolysis, only 1 pulse laser was taken out of 119MHz pulse laser, and ~ 5 nC electron beam was obtained. As a probe light, SC light generated from IR laser by PCF was used. As a sample of this experiment, pure water that was sealed after Ar bubbling was used. This is because hydrated electron has a high absorptivity rapid production time and a long lifetime. The probe light was divided into two with the half mirror. The reference intensity and the absorbed intensity are measured with photo detector put in two places respectively, and optical density (O.D.) was obtained. O.D. is common logarithmic ratio of reference intensity to measured intensity. According to Lambert-Beer law, O.D. is proportional to the density of the intermediate active species.

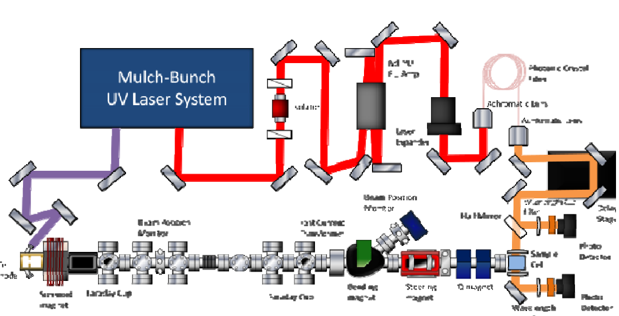

Figure 2: Experiamental setup of pump-probe pulse radiolysis with SC

Nanosecond Pulse Radiolysis with SC Light

We also performed nanosecond time scale pulse radiolysis by the same setup. The change from picosecond system was only the half mirror remove. To increase the beam charge and the signal, number of the beam bunch was increased 1 to 10. Accordingly, the bunch length changed ~10 ps to ~84 ns.

RESULTS AND DISCUSSION

Supercontinuum Generation

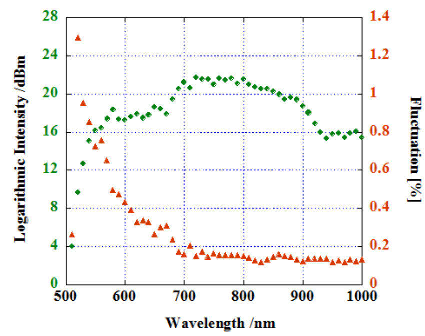

Figure 3: The intensity spectra (●) and stability (▲) of SC generated from the 119 MHz laser

The intensity spectra and stability of generated SC are shown in Fig. 3. The spectrum was broadened as far as 500 nm. The "fluctuation" means standard deviation divided by average of measured intensity. As a result, the intensity was stable enough above 700 nm, but it becomes unstable in shorter wavelength. When incident laser intensity gets higher, stability of SC light becomes better. However, when incident strength exceeds about 30W, the edge face of PCF is destroyed by laser power. Then, as a further attempt, we generated SC light from single pulse IR laser. By using single pulse laser, pulse energy of breakdown threshold has increased about 4 times compared with before. As a result, the SC stability was improved according to Fig. 4.

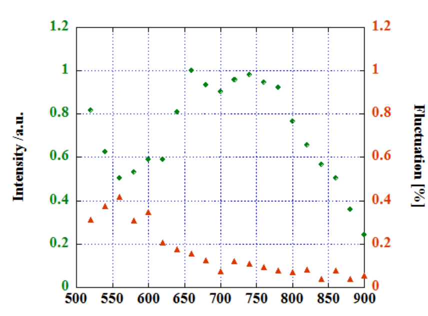

Figure 4: The intensity spectra (●) and stability (▲) of SC generated from the single laser

Picosecond Pulse Radiolysis with SC Light

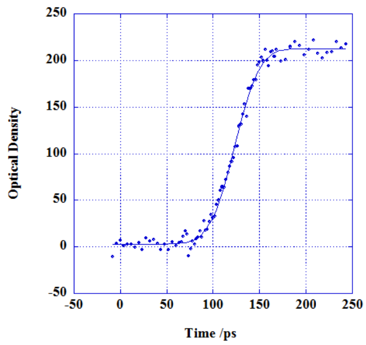

Figure 5: The transient absorption generation of water sample at 800 nm

The transient absorption of water sample at 800nm is shown in Fig. 5, which is obtained by picosecond pulse radiolysis. This data indicates the absorption of hydrated electron. The vertical axis indicates O.D. As a result, we firstly succeeded in performing picosecond pulse radiolysis with SC probe. Signal-to-noise ratio data of this experiment was 37.1. Time resolution σ can estimate theoretically by;

$$\sigma = \sqrt{{\sigma_{Bd}}^2 + {\sigma_{Ss}}^2 + n^2({\sigma_{Bs}}^2 + {\sigma_{Sd}}^2)} \qquad (1)$$

where n, σ_{Bd}, σ_{Ss}, σ_{Bs}, and σ_{Sd} is the refractive index, the bunch length, the SC size, the beam size, and the SC pulse width, respectively. According to Fig. 5, the time resolution of the picosecond pulse radiolysis was 45.5 ps in FWHM. It was worse than we expected. Therefore, it should be necessary to evaluate the beam parameters and SC properties to define the cause of the time resolution degradation.

Nanosecond Pulse Radiolysis with SC Light

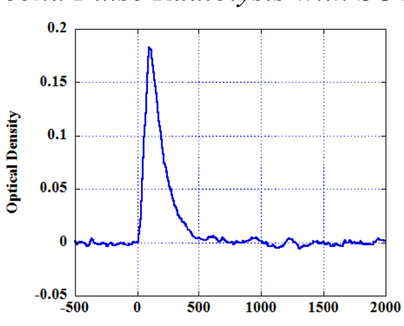

Figure 6: The transient absorption decay of water sample at 800 nm

The transient absorption decay of water sample at 800 nm is shown in shown in Fig. 6,

*In Fig. 6, lifetime of hydrated electron is strangely short compared with previous work [2]. It considered that air had include the sample during the experiment according to the looseness of the seal.

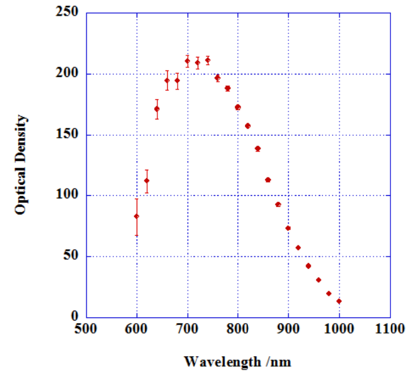

Figure 7: The absorption spectra of hydrated electron

which is obtained by nanosecond pulse radiolysis. Signal-to-noise ratio data of this experiment was 112.8. The absorption spectra of hydrated electron are Fig. 7. O.D. in Fig. 7 means the maximum value at each transient absorption data and the error bar means the standard deviation of O.D. The maximum absorption is around 720 nm, so it corresponds to the absorption of hydrated electron. As SC probe was not stable below 700 nm, the error bar became large and the reliability of O.D. was relatively low in this range.

CONCLUSIONS AND PROSPECTS

To apply SC light as a probe light of pulse radiolysis, we generated a SC radiation with PCF and IR laser, and measured its spectra and stability. We succeeded in generating pulse white light with broader spectra than the previous work. As a further attempt, we tried to generate SC light from single pulse IR laser. By using single pulse laser, pulse energy of breakdown threshold has increased about 4 times compared with before. Then we also succeeded in increasing SC stability than the traditional method. Pulse radiolysis experiment was performed using ~4 MeV electron beam as pump beam and SC light as probe light. As a result, we succeed to observe different time scale pulse radiolysis without making large change by using SC light. Both in picosecond and nanosecond experiment, fine signal-noise ratio data were obtained. We are planning to improve this unified system further more. The longer PCF and/or different specification PCF is the candidates for broadening the SC spectrum. Aiming to improve time resolution in picosecond experiment, we effort to evaluate the beam parameters and SC properties.

REFERENCES

[1] Y. Yokoyama et al., Proc. of IPAC'11, TUPC059 (2011).
[2] Y. Hosaka et al., Proc. of IPAC'10, MOPEA035 (2010).
[3] T. Schreiber, J. Limpert, H. Zellmer, A. Tunnermann, K.P. Hansen, Optics Communications 228 (2003)