PULSE RADIOLYSIS WITH SUPERCONTINUUM PROBE GENERATED BY PCF*

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Abstract

We have been studying a pump-probe pulse radiolysis as an application of the S-band photo cathode RF-Gun. Pump-probe spectroscopy is well-known method of pulse radiolysis measurement. We had used 5MeV electron beam obtained from the photo cathode RF-Gun as a pump beam, and used the white light emitted from Xe flash lamp or generated by self-phase modulation in the water cell as a probe light. However, the white probe light with high intensity, good stability and broad spectrum is a key issue for pump-probe pulse radiolysis.

Supercontinuum light with photonic crystal fiber (PCF) is a new technique of white light generation. Short pulse laser through PCF spreads its spectrum by nonlinear optical effect. Supercontinuum light has very continuous spectrum, and it is studied for various applications recently.

For applying supercontinuum light as a probe of pulse radiolysis experiment, we have generated a supercontinuum radiation with 7 picoseconds pulse width IR (1064nm) laser and PCF, and measured its properties. The experimental results of supercontinuum generation and design of a supercontinuum based pulse radiolysis system will be presented.

INTRODUCTION

Radiation Chemistry

Radiation chemistry is the study of the chemical effects of ionizing radiation in the material. The chemical effects of ionizing radiation are unique effects in comparison to other chemical effects and applied to various fields. For example, increasing rubber tire strength with cross-linking reaction and preparation of electrolyte film for button battery are well known.

Primary processes of chemical reactions by ionizing radiation consist mainly of ionization and excitation, and these effects make intermediate active species. After these reactions, intermediate active species react according to their density and diffusion constant as an ordinary chemical reaction. Therefore the figures of initial state of intermediate active species inform of all over chemical reactions by ionizing radiation. Because many intermediate active species decay in a very short time within ns or μs, particular measuring method is necessary to trace these reactions.

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Pulse Radiolysis

We have been developing photocathode RF-Gun and studying various applications of electron beam. [1]

Pulse radiolysis is a method to trace these rapid initial chemical reactions by ionizing radiation. Pump-probe absorption spectroscopy is well-known method of pulse radiolysis, and we have been studying and developing this method. [2, 3, 4] The pump beam and the probe light are necessary for pump-probe pulse radiolysis. When the lifetime of intermediate active species is shorter than ns, pump-probe pulse radiolysis is measured by stroboscopic method. In stroboscopic method, the probe light should have short pulse width compared to the lifetime of intermediate active species. The short pulse white light used to be generated from IR laser by nonlinear optical effect in the water cell. The white probe light with high intensity, good stability and broad spectrum is a key issue for high quality pump-probe pulse radiolysis. However, the light generated from pulse laser in the water cell is not stable enough and its instability complicates pulse radiolysis measurement.

Supercontinuum

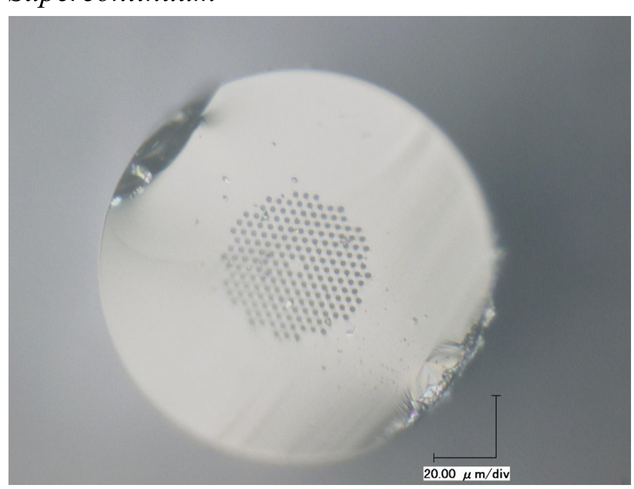

Figure 1: The cross-sectional micrograph of PCF obtained by optical microscope. One division is 20 μm.

Supercontinuum (SC) with photonic crystal fiber (PCF) is a new technique of white light generation. PCF is optical fiber which is made of silica, and clad layer of which has many micro air holes. The cross-sectional micrograph of PCF obtained by optical microscope is shown in Figure 1. Because the core layer of PCF is several micrometers diameter, photon density is constantly high while passing through PCF. Thus short pulse laser through PCF spreads its spectrum by nonlinear optical effect. [5] SC light has a very continuous spectrum, and it is studied for various applications recently. The required energy of laser for SC generation is lower than other methods of white light generation, and the improvement of pulse radiolysis system can be expected by using SC generation.

EXPERIMENTAL SETUP

Supercontinuum Generation

To apply SC light as a probe light of pulse radiolysis, we generated a SC radiation with PCF and laser, and measured its properties. Laser whose wavelength is 1064 nm, laser medium crystal is Nd: VAN, pulse width is 7 ps, repetition frequency is 357 MHz, and average power is 500 mW is used for SC generation. IR laser is amplified up to 120 times by Nd: YAG crystal pumped by flash lamp, and entered into the fiber by the achromatic objective lens. Laser average power in front of objective lens is about 170 mW without amplification. Used PCF is F-NL-5/1040 (Newport) and it is 10 m long. After passing through PCF, SC is monochromated by grating monochromator and measured by photo diode detector.

ns Pulse Radiolysis with Supercontinuum

Experimental setup of ns pulse radiolysis with SC is shown in Figure 2.The pump beam and the probe light are necessary for pump-probe pulse radiolysis. About 4 MeV electron beam obtained by photocathode RF-Gun, RF frequency is 2.8 GHz and cathode material is Cs-Te, is used as pump beam.

As the charge per pulse of the electron beam is too low to experiment [1], 10 electron pulses are assumed as one bunch train. Thus, the bunch length is 84 ns and the charge per bunch is about 1.0 nC. SC generated by IR laser and PCF is used as the probe light.

The sample of this experiment is pure water that is sealed after being freeze-thawed and distilled. Because hydrated electron generated by irradiating pure water marks a high molar absorbance coefficient and a long lifetime, pure water is an often-used sample for the evaluation experiment of pulse radiolysis system.

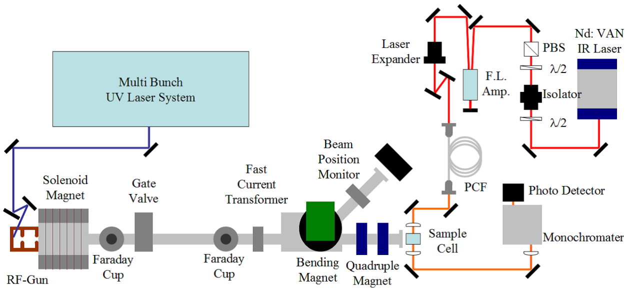

Figure 2: Experimental setup of ns pulse radiolysis with SC.

RESULTS AND DISCUSSION

Supercontinuum Generation

The intensity spectra of generated SC at each amplification ratio are shown in Figure 3. When IR laser amplified to 120 times by flash lamp amplifier is entered into the fiber, a signal is detected over about 600 nm. The probe light in ultraviolet range is necessary for complete pulse radiolysis system. But, when IR laser is amplified over 120 times, the edge face of PCF is broken by laser power. The stability of intensity is shown in Figure 4. The vertical axis presents standard deviation divided by average of measured intensity. The stability of intensity was relatively fine and approximately constant around near IR range, but it got worse in other range as the measured wavelength is far from 1064 nm.

Though it is not strong and stable enough to apply as prove light of pulse radiolysis, nanosecond pulse radiolysis with SC is conducted because we resulted it difficult to improve SC parameter with the current system.

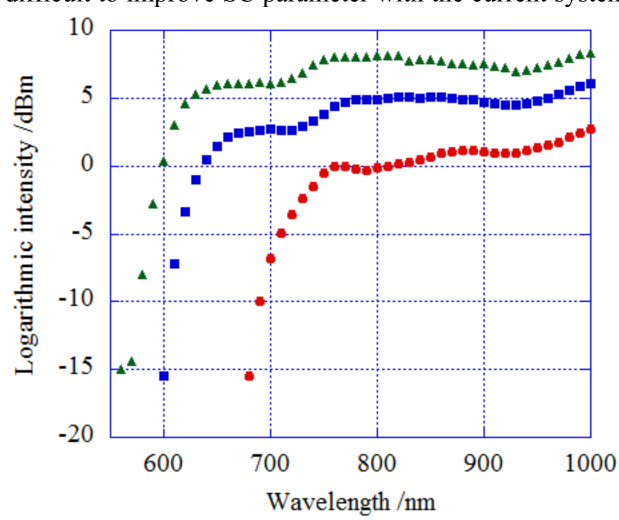

Figure 3: The intensity spectra of generated SC at each amplification ratio. 40 times (●), 80 times (■), and 120 times (▲).

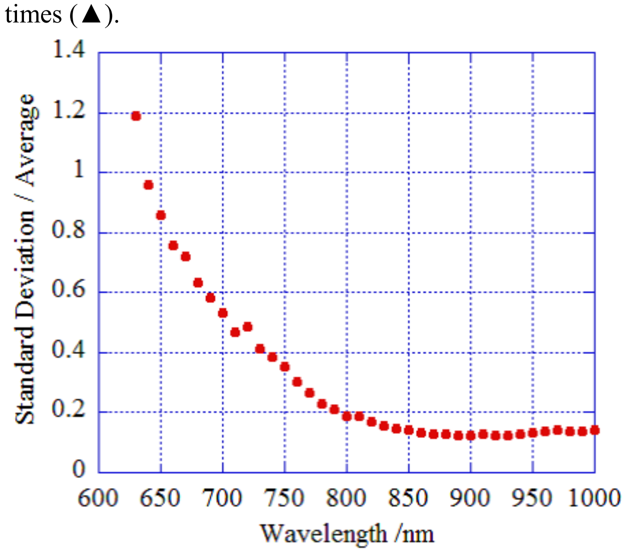

Figure 4: The intensity stability of generated SC. The amplification ratio is 80 times.

ns Pulse Radiolysis with Supercontinuum

The transient absorption decay of water sample at 820nm is shown in Figure 5. The vertical axis indicates optical density (O.D.). O.D. is common logarithmic ratio of reference intensity to measured intensity, and it is proportional to the concentration of intermediate active species according to Lambert-Beer law. This data indicates the absorption of hydrated electron. As a result, we firstly succeeded in nanosecond pulse radiolysis with SC probe.

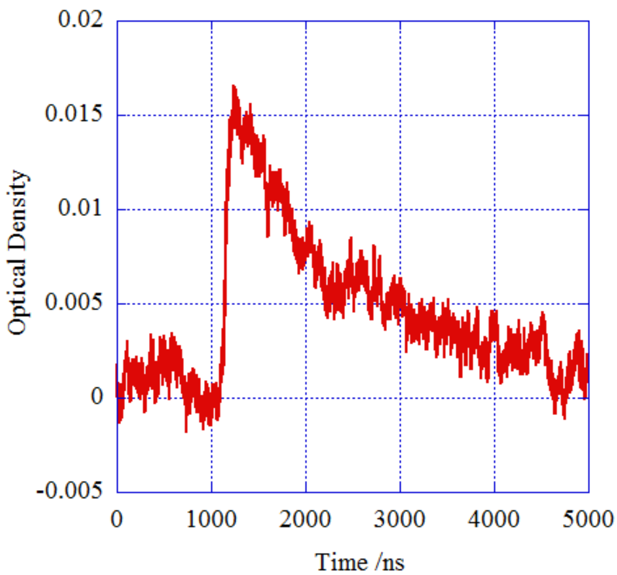

Figure 5: The transient absorption decay of pure water sample at 820nm probe light.

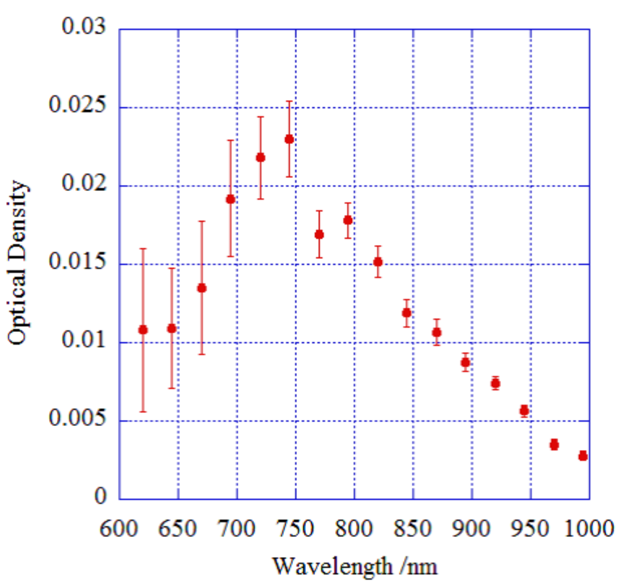

Figure 6: The absorption spectrum of hydrated electron.

The absorption spectrum of hydrated electron is shown in Figure 6. O.D. in Figure 6 means the maximum value at each transient absorption data and the error bar means the standard deviation of O.D. before electron beam interaction. The maximum absorption is around 720 nm, so it corresponds to the absorption of hydrated electron. As SC probe was not strong and not stable below 750 nm, the error bar became very large and the reliability of O.D. was low in this range.

CONCLUSIONS AND PROSPECTIVES

To apply SC light as a probe light of pulse radiolysis, we generated a SC radiation with PCF and laser, and measured its properties. 1064 nm, 7 ps pulse width laser was used for SC generation. IR laser is amplified up to 120 times by Nd: YAG crystal pumped by flash lamp, and entered into the fiber. When IR laser is amplified over 120 times, the edge face of PCF is broken by laser power. Laser average power in front of objective lens is about 170 mW without amplification. A signal is detected over about 600 nm, and the stability of intensity was relatively fine and approximately constant over 850 nm. In this range, we succeeded in pulse white light generation with better stability and broader spectra than the traditional method.

Nanosecond pulse radiolysis experiment was performed using 4MeV electron beam (pump) and SC light (prove). As a result, we succeeded in tentative nanosecond pulse radiolysis with SC probe. However SC probe is not strong and not stable enough below 750 nm, we are planning to improve the SC light generation to optimize the pulse duration, pulse repetition rate, and fiber length. In addition, it is planned to use different PCF appropriate to visible laser and generate SC from second harmonic of IR laser.

On the other hand, it is able to use Xe flash lamp and other white light source as probe light of nanosecond pulse radiolysis, and picosecond pulse radiolysis needs short pulse white light source. Picosecond pulse radiolysis with SC will be conducted when the necessary devices are prepared.

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