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DEVELOPMENT OF ANALYTICAL LIGHT SOURCE FOR CONSTRUCTION OF FEMTOSECOND PULSE RADIOLYSIS SYSTEM USING ER FIBER LASER

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Abstract

The initial processes in radiation chemistry occur from the ultrafast time scale (sub-femtosecond timescale) and are the dominant factor for the subsequent reaction. It is important to elucidate the initial process of the radiation chemical reaction. One of the methods to elucidate the initial process is pulse radiolysis. In pulse radiolysis, a substance is irradiated with ionizing radiation and at the same time irradiated with analytical light, and the absorption spectrum of the light is observed, which allows us to track the short-lived intermediate species. There are several requirements required for analytical light. We aimed to use the supercontinuum light generated by the second harmonic of the Er fiber laser as analytical light. In this conference, we report on the status of the development of this supercontinuum light and the prospects.

INTRODUCTION

Our research is aimed at elucidating the initial processes of radiation chemical reactions. Radiation chemical reactions are chemical reactions that occur due to the interaction between radiation and electrons in materials. To understand the whole picture of radiation chemical reactions, it is important to elucidate the interaction, i.e., the initial process. The interaction occurs in ultrafast time (sub-femtosecond timescale). Therefore, special techniques must be used to observe the interaction.

One of the methods for elucidating initial processes is pulse radiolysis. In pulse radiolysis, a material is irradiated with an electron beam and simultaneously allowed to pass through an analysing light to measure its absorbance, enabling temporal tracking of reaction intermediates that are formed and disappear in an ultrashort time range derived from bunch length of electron beam and pulse width of analyzing light. A conceptual diagram of pulse radiolysis is shown in Fig. 1.

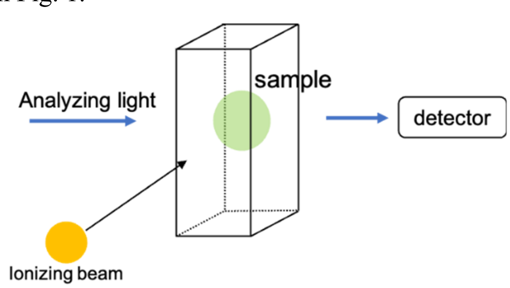

Figure 1: Diagram of pulse radiolysis.

There are three major requirements for analytical light. Firstly, it must have ultrashort pulse width. This is because the initial processes of radiation chemical reactions occur in the ultrafast time range, and pulse radiolysis systems need to have the same high temporal resolution. Secondly, wavelength is broadband. If the wavelength is broadband, a single pulse radiolysis system can observe many substances that absorb in that wavelength band. Thirdly, the intensity is high. A certain level of intensity is necessary so that the spectra of absorption wavelengths are not buried in noise, making it difficult to observe the spectra of absorption wavelengths. We aimed to use a supercontinuum (SC) light as the analysis light that satisfies the above three conditions. Supercontinuum light is broadband light generated by nonlinear optical effects in a nonlinear medium, which maintains the properties of the ultrashort laser pulse. We have constructed an optical laser system to generate this SC light.

In a previous study, a supercontinuum light with a spectral width of 750-1030 nm had been successfully generated by injecting a Yb laser (wavelength: 1030 nm) into a photonic crystal fiber (PCF) [1]. Figure 2 shows a spectral diagram of the generated supercontinuum light [1].

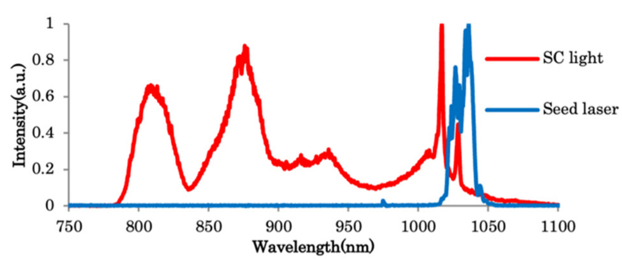

Figure 2: Spectral diagram of the generated supercontinuum light [1].

Figure 2 shows that the wavelength extends toward the short wavelength side with respect to the incident laser wavelength. On the other hand, active species observed in chemical radiation reactions seem to be absorbed in the visible wavelength region [2]. The SC light produced in previous studies does not cover this visible light region. Therefore, we have attempted to generate SC light that includes the visible light region by injecting the second harmonic (775 nm) of Erbium (Er) fiber laser (1550 nm) into a PCF, thinking that the wavelength of the laser injected into the PCF should be closer to the visible light region.

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EXPERIMENTAL SCHEMES

Figure 3 shows a schematic diagram of the laser system.

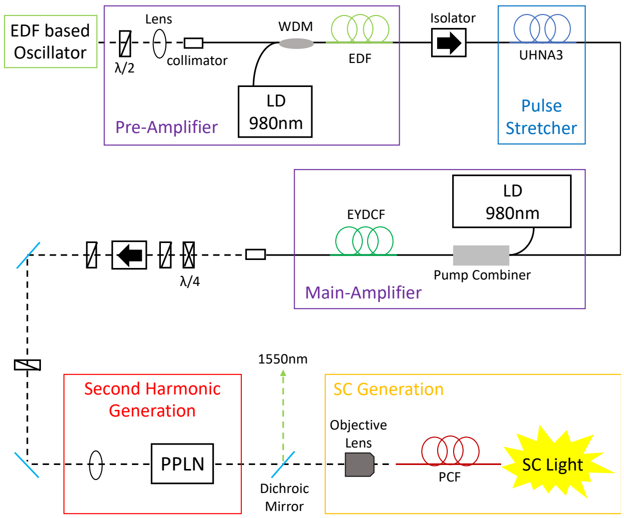

Figure 3: Schematic diagram of the laser system.

The laser system consisted of an oscillator, pulse stretcher, amplifier, second harmonic generator, and SC light generator. Each part is described below.

Oscillator

The oscillator outputted femtosecond laser pulse at a wavelength of 1550 nm with 40 mW using Er-doped fiber under 980 nm LD excitation. It oscillated in pulses due to mode-locking by nonlinear polarization rotation. Figure 4 shows a schematic diagram of the oscillator. Figure 5 shows a spectrum of the lase after the oscillator, and Table 1 shows the performance of the oscillator.

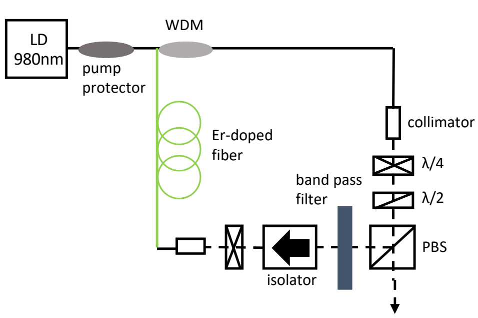

Figure 4: Schematic diagram of the oscillator.

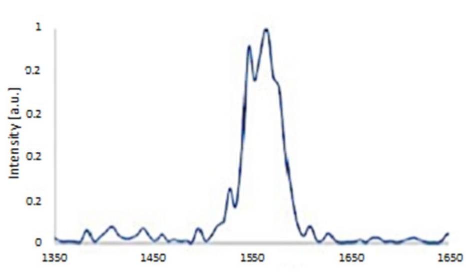

Figure 5: Spectrum of the laser after the oscillator.

Table 1: Performance of the Oscillator

Repetition frequency	70.4 MHz
Average power	43.0 mW
Pulse width (FWHM)	676 fs
Peak power	904 W
Center wavelength	1561 nm
Spectral width (FWHM)	41.6 nm

Pulse Stretcher

We used a fiber-based stretcher. The fiber used as a stretcher was UHNA3, which was an ultra-high NA fiber. The GVD of this fiber was about 0.10 ps^2/m around 1550 nm and can be used as a pulse stretcher to provide normal dispersion. The length of the fiber was 1.5 m.

Amplifier

The amplifiers consisted of a pre-amplifier and a main-amplifier for two-stage amplification. A double-clad fiber was used to withstand the intensity of the excitation light because the main amplifier uses powerful excitation light. Since the laser after the amplification mechanism passed through the isolator and was reflected by two mirrors to the crystal in Fig. 3, these optical elements caused a loss of output power. This amplifier succeeded in increasing the peak power by a factor of approximately 29. Figure 6 shows a spectrum of the laser before crystal injection, and Table 2 shows the laser parameters before crystal injection.

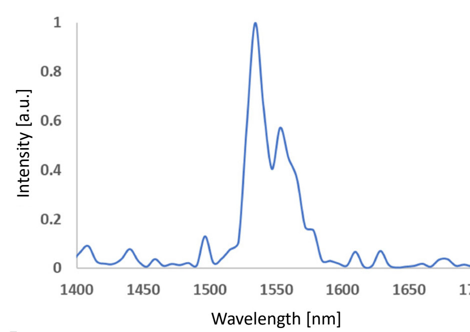

Figure 6: Spectrum of the laser before crystal injection.

Table 2: Laser Parameters Before Crystal Injection

Average power	239 mW
Pulse width (FWHM)	129 fs
Peak power	26.3 kW
Center wavelength	1.54 μm
Spectral width (FWHM)	36.9 nm

Second Harmonic Generation

The second harmonic (775 nm) was generated by injecting a short-pulse laser pulse, whose intensity was increased by the amplifiers, into a nonlinear optical crystal called a PPLN (periodically poled lithium niobate) crystal. Figure 7 shows a spectrum of the second harmonic, and Table 3 shows the parameters of the second harmonics.

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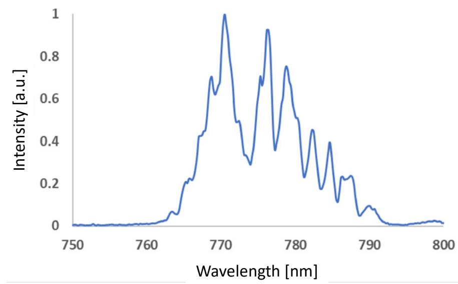

Figure 7: Spectrum of the second harmonic.

Table 3: Parameters of the Second Harmonic

Average power	480 µW
Center wavelength	774 nm
Spectral width (FWHM)	16.1 nm
Conversion efficiency	0.20%

DISCUSSION

In this study, the generation of the second harmonic was successful, however, the conversion efficiency from the fundamental to the second harmonic in the PPLN crystal was low (0.20%), and the average output of the second harmonic was not reached for the analytical light source in femtosecond pulse radiolysis. With the current output, wavelength broadening cannot be expected even when injected into a PCF. We believe there are two reasons for the low conversion efficiency. First, the peak power was low. Nonlinear optical effects are larger the higher the peak power. Therefore, further increasing the current peak power may improve the conversion efficiency. Second, the spectrum is not symmetrical with respect to the center wavelength. The process of second harmonic generation in PPLN crystals exists in two ways, second harmonic generation (SHG) and sum frequency generation (SFG) [3]. Eq. (1) shows the transformation of frequencies in SHG and Eq. (2) shows the transformation of frequencies in SFG.

$$\text{SHG: } \omega \rightarrow 2\omega \tag{1}$$
$$\text{SFG: } (\omega + \omega_0) + (\omega - \omega_0) \rightarrow 2\omega \tag{2}$$

ω is the center frequency of the fundamental wave, ω_0 is the difference from the center frequency. Frequency components that satisfy the quasi-phase matching condition of the PPLN crystal cause second harmonic generation. Conversely, the frequency components that do not satisfy the quasi-phase matching condition cause sum frequency generation. The pseudo-phase matching condition is shown in Eq. (3).

$$d = \frac{\lambda_\mathrm{F}}{4\{n_\mathrm{SH}(T) - n_\mathrm{F}(T)\}} \tag{3}$$

λ_F is wavelength of fundamental wave, $n_\mathrm{SH}(T)$ is refractive index of second harmonic in PPLN depending to temperature T, and $n_\mathrm{F}(T)$ refractive index of fundamental wave. As can be seen from Eq. (2), sum frequency generation is the combination of two frequencies with equal difference with respect to the center frequency to generate a frequency that is twice the center frequency. Therefore, in order to convert all frequency components in the fundamental to second harmonics, it is necessary to generate a fundamental such that the quasi-phase matching condition is satisfied at the center frequency of the fundamental and the spectrum is symmetrical with respect to the center frequency. However, as Fig. 6 shows, the spectrum of the laser before crystal injection was not symmetrical. This results in a low conversion efficiency because only some components of the fundamental wave were converted to second harmonic waves.

CONCLUSION

We aimed to generate SC light by injecting the second harmonic (775 nm) of an Er fiber laser (1550 nm) into the PCF. We have succeeded in generating the second harmonic, however, the lack of output power of the second harmonic was still an issue. The cause of low conversion efficiency seemed to be low peak power and asymmetric spectrum. The entire system needs to be reviewed to increase the peak power and to obtain a spectrum symmetrical to the center. After improving the conversion efficiency to the second harmonic, the SHG light will be injected into the PCF to generate SC light, and its performance as analyzing light for the pulse radiolysis system will be evaluated.

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