DEVELOPMENT OF A PULSE RADIOLYSIS SYSTEM BY USING PICOSECOND ELECTRON PULSES AND FEMTOSECOND LASER PULSES

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Abstract

A new picosecond pulse radiolysis system has been developed at the Institute of Scientific and Industrial Research, Osaka University. The system is composed of a 38 MeV linear accelerator for a irradiation source and a Ti-sapphire laser for an analyzing light source. The electron pulses (FWHM = 20ps) and the laser pulses (FWHM = 60fs) are synchronized with a radio frequency of 27 MHz. The wavelength region of the analyzing light is from 300 nm to 1500 nm by using a fundamental light and a white light continuum by using a regenerative amplifier. The present time resolution of the system is about several ten picoseconds.

1. Introduction

The pulse radiolysis is a powerful method to investigate the time-dependent behavior of the short lived intermediates produced by a radiation. The first picosecond pulse radiolysis, so called the stroboscopic method, was planed [1] and done by J. W. Hunt at Toronto university in 1969 [2]. The irradiation source was an electron pulse and the analyzing light source was the Cherenkov light pulse produced by an electron pulse. The time profile was obtained by changing the time difference between an electron pulse and a Cherenkov light. The time resolution was decided by the pulse widths of the irradiation and analyzing light source. However, the observable time-scale was limited within 350 ps which corresponded to the interval of the electron pulse train of the S-band linac.

To solve such a problem, the two different picosecond pulse radiolysis system by using a single electron pulse was developed at Argonne national lab.[2] and Univ. of Tokyo[3]. In the Argonne, the analyzing light pulse could irradiate the sample before the electron pulse by using a 270^{o} magnet. On the other hand, the two linacs were used in the Univ. of Tokyo. One was used for a irradiation source and the another was used for a Cherenkov light source. However, in both methods the wavelength is limited in the visible region, because the intensity of the Cherenkov light in the infrared region is weak.

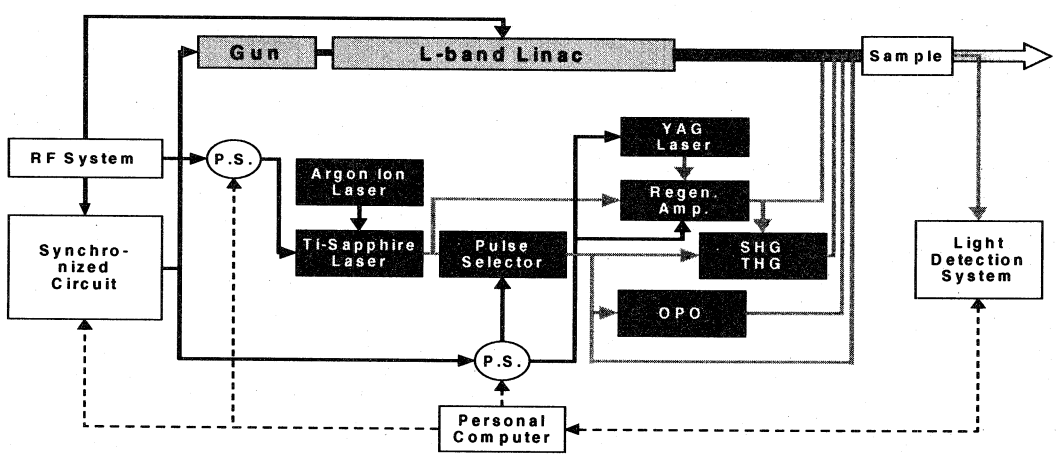

Fig.1 Block diagram of a new picosecond pulse radiolysis system

Later in order to measure in the infrared region, the LL-twin system[5] by using a diode laser was developed at Univ. of Tokyo. However, the wavelength was fixed.

To solve such two problems (the time-scale and the detectable wavelength), a new picosecond pulse radiolysis system by using an electron for an irradiation source and a Ti-sapphire laser for an analyzing light has been developed. Although the fundamental wavelength of the Ti-sapphire laser is from 750 nm to 850 nm, the wavelength is extended from 300 nm to 1500 nm by using the white light continuum.

2. Outline of new picosecond pulse radiolysis system

An schematic of a new pulse radiolysis system is shown in Fig.1. The system is mainly composed of the ISIR 38MeV L-band linac[6], a femtosecond Ti-sapphire laser system, a rf system, a synchronized circuit, a light detection system and a personal computer.

A sample was irradiated by a 20 ps electron pulse from the linac. At the same time, the femtosecond analyzing light (FWHM=60fs) from the laser system passed through the sample cell. The analyzing light was detected by the light detection system and the time-dependent absorption was calculated by the computer. Also, all equipment was controlled by the computer.

Usually, it is difficult to synchronize a femtosecond laser pulse with a picosecond electron pulse within the timing jitter of several picoseconds. In the new system, the timing jitter was controlled by the RF synchronization method. The frequency of the master oscillator of the linac was 108 MHz which was multiplied by 12 to drive the klystrons at 1296 MHz. On the other hand, the Ti-sapphire laser is operated at 81 MHz. Therefore, the rf of 27 MHz which was the greatest common divisor was used as the standard radio frequency. The rfs of 81 MHz (27 MHz multiplied by 3) and 108 MHz (27 MHz multiplied by 4) were provided to the laser system and the linac, respectively.

Although the laser timing was controlled by the rf of 81 MHz, there still remains the timing jitter due to the change of the laser cavity length. To solve the problem, the compensation system of the cavity length (so called the lock-to-clock-system) worked on very well. The system keeps the cavity length to be constant by the PZT. The laser pulse could be synchronized with the electron pulse completely.

Output laser from the Ti-sapphire laser is a pulse train (81 MHz). On the other hand, the maximum

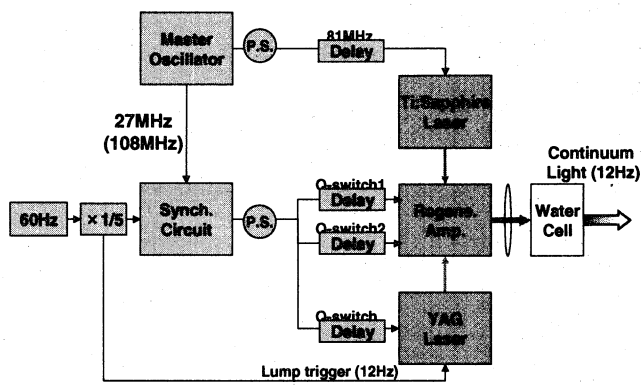

Fig.2 Trigger system for the generation of the white light continuum.

repetition of the single electron pulse was 60 Hz. It is necessary to pick up a single laser pulse which is synchronized with a single electron pulse. The pulse selector (the acoustic-optic modulator) was inserted after the Ti-sapphire laser. The timing trigger to the pulse selector was provided from the synchronized circuit.

The time difference between an electron pulse and a laser pulse should be changed continuously to obtain a time profile of the absorption. In Toronto and Argonne, the time difference was changed by a optical delay line system. In this system, two electric delay lines (phase shifters) were used in stead of the optical delay line to make an alignment easily in the radiation field. The phase shifters were installed into the 81 MHz line for the Ti-sapphire laser and the trigger line for the pulse selector.

The fundamental wavelength region was from 750 to 850 nm. The wavelength region was extended by using of the nonlinear optics such as SHG(second harmonics generation), THG(third harmonics generation), and OPO(optical parametric oscillation) as shown in Fig. 1. However, these techniques can not cover the all wavelength region from 300 to 1500 nm. The system of the white light continuum will mention in the next section.

3. Generation of the white light continuum

The system for the generation of the white light continuum is shown in Fig.2. The white light continuum was generated by focusing a single high intensity laser into an appropriate material, such as H_2O, D_2O and quartz block. The high intensity laser was produced by the Ti-sapphire regenerative amplifier excited by the YAG laser.

The trigger system was changed to control the

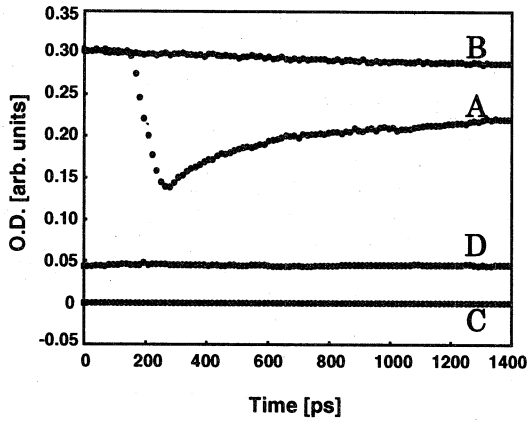

Fig.3 Typical signals obtained in the new picosecond pulse radiolysis. Signal A, B and C are detected by a light detection system. D is detected by a beam current monitor. (see in the content)

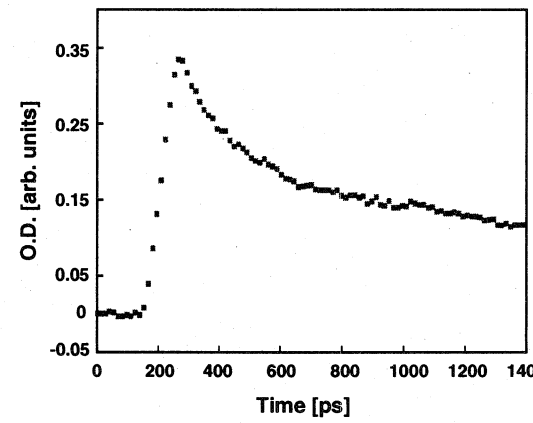

Fig.4 The time-dependent behavior of n-dodecane cation radical obtained in the new pulse radiolysis of liquid n-dodecane, monitored at 820 nm.

regenerative amplifier. A single amplified laser pulse (12Hz) was picked up by two Q-switches (the pockels cells) installed in the regenerative amplifier in stead of the pulse selector. One controlled the injection timing of the seed pulse (81 MHz) from the Ti-sapphire laser and the other controlled the output timing of the amplified laser pulse. To control the timing of the two Q-switches, the triggers from the synchronized circuit are supplied to the two Q-switches.

To change the time difference between a single electron pulse and a single amplified laser pulse, three phase sifters were installed into the trigger lines of the two Q-switches and the YAG laser. The detectable wavelength region was from 300 nm to 1500 nm.

4. Light detection system

The light detection system was consists of a monochromator, a photodiode (Si or InGaAs), and an oscilloscope. Fig 3 shows an example of the detected signals. The four types of the signals were measured. Signal A is the intensity of the analyzing light containing the absorption and the background (Cherenkov light and the emission from samples, if there is the emission from the samples). Signal B is the intensity of the analyzing light. Signal C is the intensity of the background. Signal D is the beam intensity which is detected by the beam current monitor. The optical density (O.D.) was calculated as follows.

$$\text{O.D.} = \log \{(B-C) / (A-C)\} \cdot D_0 / D$$

where D_0 is a initial beam intensity.

5. Example of the measurement

Fig. 4 shows the typical time-dependent absorbance of the n-dodecane cation radicals obtained in the new picosecond pulse radiolysis of liquid n-dodecane monitored at 820 nm. After the ionization, an electron and a radical cation of n-dodecane, so called the geminate ion pair, are produced. The cation radicals formed within the time resolution of the system. The decay part shows the geminate ion recombination[7] of the cation radical and the electron which is important reaction in primary processes.

6. Development in the future

The present time-resolution is about several ten picoseconds which depends mainly on the pulse width of the electron pulse (~20 ps). Recently, a plane for the generation of the femtosecond electron pulse has started at Osaka university. At present, a few picosecond electron pulse is obtained by using the magnetic pulse compression. In the future, the system will be developed into a femtosecond pulse radiolysis system.

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