DEVELOPMENT OF A NEW PICOSECOND PULSE RADIOLYSIS SYSTEM BY USING FEMTOSECOND LASER SYNCRHONIZED WITH PICOSECOND ELECTRON BEAMS

Y. Yoshida, Y. Mizutani, T. Kozawa, M. Miki, S. Seki, T. Yamamoto,
K. Hori, Y. Izumi*, K. Ushida**, and S. Tagawa
The Institute of Scientific and Industrial Research, Osaka University
Mihogaoka 8-1, Ibaraki, Osaka 567, Japan
*Department of Nuclear Engineering, Osaka University
Yamadaoka 2-1, Suita, Osaka, 565, Japan
**The Institute of Physical and Chemical Research (RIKEN)
Hirosawa 2-1, Wako-shi, Saitama, 351-01 Japan

Abstract

A new picosecond pulse radiolysis system for the absorption spectroscopy to observe ultra fast phenomena was developed. The system was composed of a 38 MeV picosecond electron L-band linac for the irradiation source and a femtosecond laser system for the analyzing light. The laser pulse was synchronized with the electron pulse by using the common radio frequency. The time-resolution of the system was several tens picoseconds. The studies on the primary processes of the radiation chemistry have been started by using the system.

1 INTRODUCTION

Primary processes of radiation chemistry is very important to know the whole reaction processes induced by high energy radiation. Pulse radiolysis technique is one of the most powerful method to research the primary processes, because very fast reaction can be detected directly. The studies on excited states and energy transfer were done by using picosecond pulse radiolysis for emission spectroscopy[1-5] with a very fast response light detector such as a streak camera. In absorption spectroscopy, the streak camera[6,7] and very fast response detector[8] also were used. However, the experiment had the difficulty to make very intense analyzing light.

The stroboscopic method does not require the very fast light detector. The time resolution is mainly decided by the pulse width of the radiation source and the analyzing light. The several picosecond pulse radiolysis systems were developed since the first picosecond pulse radiolysis by Hunt et al.[9] in 1970. Argonne group[10] developed the picosecond pulse radiolysis by using a single electron pulse from a L-band linac in 1980. Later so called twin-linac system[11-13] were developed at Tokyo Univ. in 1985. In these systems, the Cherenkov light pulse produced by the high energy electron was used for the analyzing light. Although the primary processes of radiation chemistry were studied by using these pulse radiolysis systems, they were not good at the infrared spectroscopy, because of the low intensity of the Cherenkov light in the region.

A picosecond pulse radiolysis system[14,15] by using the laser diode instead of the Cherenkov light was developed in 1991. The system enabled the absorption spectroscopy from visible to near infrared region. However, it is difficult to obtained complete spectra, because the laser diode was not tunable light source.

Recently, a new picosecond pulse radiolysis system has been developed in Osaka University. The femtosecond laser which is synchronized with the picosecond electron pulse is used for the analyzing light. The tunable laser pulse could cover the wide wavelength region from ultra violet to infrared by the non-linear optical effect. In this paper, the new system and its performance were described.

2. SYSTEM OF NEW PICOSECOND PULSE RADIOLYSIS

Figure 1 shows the block diagram of the new picosecond pulse radiolysis system. The system was mainly composed of an 38 MeV L-band electron linac[16], a femtosecond laser system, a control rf / trigger system, a light detection system and a personal computer.

The linac is composed of a thermal electron gun, two 1/12 (108MHz) sub harmonic pre-bunchers, a 1/6 (208 MHz) sub harmonic pre-buncher, a 1.3 GHz pre buncher, a 1.3 GHz buncher and a 1.3GHz acceleration tube. The pulse width and maximum pulse charge are 20 ps and 67 nC, respectively. The pulse beam is conducted to the irradiation room by a achromatic beam transport.

The sample contained in the quartz cell (the pass length is from 0.5 cm to 2 cm) was irradiated by the picosecond electron pulse from the L-band linac. The repetition of the electron pulse was below 60 Hz. The single femtosecond laser pulse as the analyzing light pulse also was passed through the sample. Both pulses were synchronized by using the radio frequency (rf). .

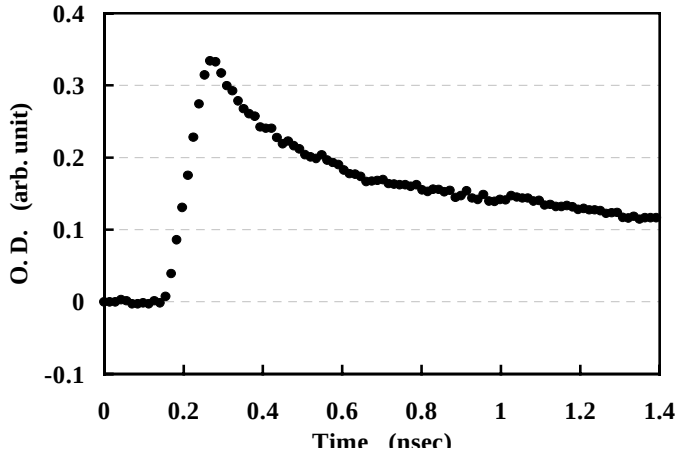

Fig. 1 Picosecond pulse radiolysis system synchronized wit femtosecond laser

3. SYNCHRONIZATION AND TRIGGERING SYSTEM

Figure 2 shows the rf / trigger system for the synchronization between the electron and the laser pulses. The basic clock was 27 MHz. The multiplied rf of 27 MHz were provided to the subharmonic pre bunchers (x4), the subharmonic pre bunchers (x8), the pre-buncher (x12), the buncher (x12) and acceleration tube (x12) of the linac. Also, the rf of 81 MHz which is three times of 27 MHz was provided to the laser system.

The trigger signals generated by the triple synchronized system were provided to both the electron gun of the linac and the laser system. At first, the trigger was synchronized between a start trigger from the computer and the 60 Hz. Then the triggers were synchronized with 27 MHz. Finally, the triggers were synchronized with 108 MHz. Therefore, the timing jitter depended on the timing jitter of the rf of 108 MHz.

In the measurement of the optical absorption, the four operation modes of the linac and the laser (mode A, B, C and D) were required for the reason described in the section 5. In each mode, both the linac and the laser, only the laser, only the linac and nothing were operated, respectively. The change of the modes was controlled by switching the trigger signal with the two enabled circuits which were controlled by the computer.

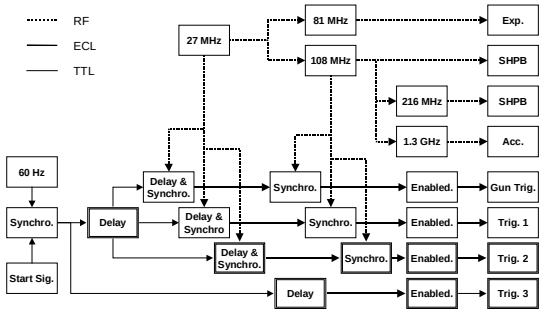

Fig. 2 Rf and Triggering system

4. SYSTEM OF FEMTOSECOND LASER

A femtosecond Ti-sapphire laser is excited by a 12 W Ar ion laser. The mode locker (A/O) of the Ti-sapphire laser was driven by 81 MHz. A compensation system of the time jitter (lock to clock system) was equipped to keep the stability of the laser. The system could keep the laser cavity to be constant length by the feed back system comparing the oscillation frequency of the cw laser with the 81 MHz. The pulse width of the laser observed by using an autocorrelator was about 60 fs under the operation of 81 MHz.

A single laser pulse synchronized with the electron pulse was extracted from the continuous laser pulse train of 81 MHz by the pulse selector (A/O) located after the Ti-Sapphire laser. The timing of the extraction was decided by the trigger signal from the rf / trigger system.

The tunable wavelength region of the Ti-Sapphire laser is from 720 nm to 900 nm. The second harmonic generator (SHG) and the third harmonic generator (THG) produced the analyzing pulse from 360 nm to 450 nm and from 240 nm to 300 nm , respectively. More wide wavelength region can be covered by using the optical parametric oscillation (from 1 μm to 2 μm) and its second harmonic generation (500nm to 1μm). The continuum analyzing light from 300 nm to 900 nm can be produced by using a regenerated Ti-sapphire amplifier.

In order to obtain time-dependent optical absorption, the time difference between the electron pulse and the laser pulse was changed by varying both the phase of 81 MHz provided to the laser system and the trigger timing provided to the pulse selector. The two phase shifters were inserted in the rf line and trigger signal line.

5. LIGHT DETECTION SYSTEM

The time resolution of transient absorption is decided by the width of the electron pulse, the width of the laser pulse, and the timing jitter between the electron pulse and the laser pulse. It is important for the light detection system to measure the correct amount of analyzing light. The detection system was composed of a Si-photodiode (Hamamatsu:S1722-02), a charge sensitive amplifier, a spectroscopy amplifier (EG&G Ortec:172) and a digital oscilloscope (SONY Tektronix:TDS684B).

The signal obtained in the mode A consists of the analyzing light containing the absorption signal, the Cherenkov radiation, the other radiation noise derived from the electron pulse and the rf noise from the linac. The signal in the mode B contains the analyzing light and the rf noise. The signal in the mode C contains the Cherenkov radiation, the other radiation noise and the rf noise. The rf noise is measured in the mode D. Therefor, real optical density were calculated by

optical density = log [(b-c)/(a-d)] (1)

where a, b, c and d is signal intensity obtained in the mode A, B, C and D, respectively.

6. PRIMARY PROCESSES OF RADIATION CHEMISTRY OF N-DODECANE

The geminate ion recombination[17,18] in n-dodecane was studied by using the new system. The most geminate pairs of an electron and a n-dodecane cation radical produced by high energy electron recombined rapidly due to the long range of the Coulomb potential. Figure 3 shows the typical time-dependent behavior of transient absorption of n-dodecane cation radicals obtained in the picosecond pulse radiolysis of liquid n-dodecane monitored at 820 nm. The cation radicals formed within the time resolution of the system. The decay showed a typical geminate decay. The theoretical decay line was obtained by the Smoluchowski equation which describe the kinetics of the geminate ion recombination based on the diffusion theory. In the analysis, the sum of the diffusion coefficients of 6 x 10^{-4} cm^2/s for the electron and the radical cations and the exponential initial distribution[i,ii] with the initial separation of 6.6 nm were used. These parameters agreed with reported values[20,21].

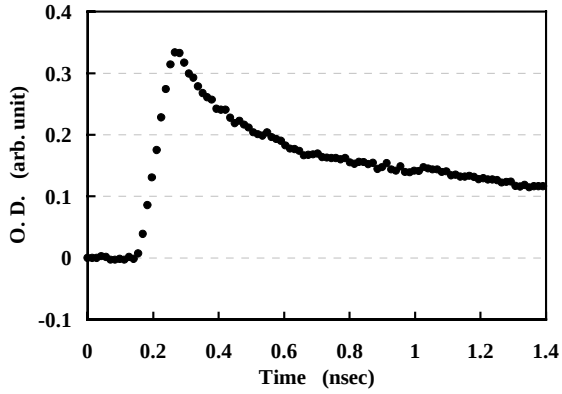

Fig. 3 Time-dependent behavior of the n-dodecane cation radical obtained in the picosecond pulse radiolysis of n-dodecane monitored at 820 nm.

7.REFERENCES

[1] S. Tagawa, Y. Katsumura, Y. Tabata, Chem. Phys. Lett., 64, 258 (1979).
[2] Y. Katsumura, S. Tagawa, Y. Tabata, J. Chem. Phys., 84, 833 (1980).
[3] S. Tagawa, Y. Katsumura, Y. Tabata, Radiat. Phys. Chem., 15, 287 (1980).
[4] S. Tagawa, Y. Katsumura, Y. Tabata, Radiat. Phys. Chem., 19, 125 (1982).
[5] Y. Katsumura, S. Tagawa, Y. Tabata, Radiat. Phys. Chem., 19, 243 (1982).
[6] H. Kobayashi, T. Ueda, T. Kobayashi, S. Tagawa, Y. Yoshida, Y. Tabata, Radiat. Phys. Chem., 23, 4 (1984).
[7] Y. Yoshida, S. Tagawa, H. Kobayashi, Y. Tabata, Radiat. Phys. Chem., 30, 83 (1987).
[8] S. Tagawa, M. Washio, H. Kobayashi, Y. Katsumura, Y. Tabata, Radiat. Phys. Chem., 21, 45 (1983).
[9] M. J. Bronskill, W. B. Taylor, R. K. Wolff, J. W. Hunt, Rev. Sci. Instr., 41, 333 (1970).
[10] C. D. Jonah, Rev. Sci. Instr., 46, 62 (1975).
[11] Y. Tabata, H. Kobayashi, M. Washio, S. Tagawa and Y. Yoshida, Radiat. Phys. Chem. 26, 473 (1985).
[12] H. Kobayashi, Y. Tabata, T. Ueda, T. Kobayashi, Nucl. Instr. Meth. B24/25, 1073 (1987).
[13] Y. Yoshida, S. Tagawa, M. Washio, H. Kobayashi Y. Tabata, Radiat. Phys. Chem., 34, 493 (1989).
[14] Y. Yoshida, T. Ueda, T. Kobayashi, H. Shibata and S. Tagawa, Nucl. Instr. Meth. A327, 41(1993).
[15] Y. Yoshida and S. Tagawa, Dynamics and Mechanisms of Photoinduced Electron Transfer and Related Phenomena, (Edited by N. Mataga, T. Okada and H. Masuhara, Elsevier Sci. Pub., 435 (1992).
[16] S. Takeda, K. Tsumori, N. Kimura, T. Yamamoto, T. Hori, T. Sawai, J. Ohkuma, S. Takamuku, T. Okada, K. Hayashi, M. Kawanishi, IEEE Trans. Nucl. Sci., NS-32, 3219 (1985).
[17] J. M. Warman, The Study of Fast Processes and Transient Species by Electron Pulse Radiolysis (Edited by J. H. Baxendale and F. Busi) Reidel, Dordrecht, (1982).
[18] Y. Yoshida, S. Tagawa and Y. Tabata, Pulse Radiolysis, (Edited by Y. Tabata), CRC Press, 343 (1991).
[19] S. Tagawa, N. Hayashi, Y. Yoshida, M. Washo, Y. Tabata, Radiat. Phys. Chem., 34, 503 (1989).
[20] Y. Yoshida, S. Tagawa, Y. Tabata, Radiat. Phys. Chem., 23, 279 (1984).
[21] Y. Yoshida, S. Tagawa, Y. Tabata, Radiat. Phys. Chem., 28, 201 (1986).