IMPROVEMENT OF COMPACT PICO-SECOND AND NANO-SECOND PULSE RADIOLYSIS SYSTEMS AT WASEDA UNIVERSITY*

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Abstract

A pulse radiolysis method is very useful in studying primary processes of radiation chemistry. At Waseda University, the compact pico-second and nano-second pulse radiolysis systems have been developed with the photo-cathode RF gun. Recently, pico-second system has been improved due to the installation of a new RF gun cavity with Cs-Te cathode. A new nano-second pulse radiolysis system with Xe flush lamp has been developed. This system has successfully operated with enough S/N for the studying of primary processes on radiation chemistry from the system evaluation.

INTRODUCTION

A pulse radiolysis method is a powerful tool to study the primary processes in radiation chemistry. It has been contributed to both basic scientific and industrial researches, such as life science and lithography.

Many types of pulse radiolysis systems have been developed at many facilities, such as University of Tokyo [1], Osaka University [2], Argonne National Laboratory [3], Brookhaven National Laboratory [4], University of Paris-Sud[5] and so on. However, they have been developed using a large accelerator, so that the cost must be huge. Whereas, the compact pico-second and nano-second pulse radiolysis systems with photo-cathode RF gun only have been developing since in 2000 at Waseda University [6] [7].

A pico-second system is based on stroboscopic pump-probe method. Our system is based on a photo-cathode RF gun. Electron energy of RF gun is 4-5MeV, which is smaller than other facilities, but high enough to study radiation chemistry. We have been improved this system. To obtain higher time resolution, we have focused electron beam with quadrupole magnets before irradiating to sample, so we have succeeded in improving time resolution from 29ps to 18ps [6]. It has taken 14 hours to obtain a time profile, so damage of sample has been assumed. Consequently, we have raised measurement repetition rate from 5Hz to 10Hz, and minimize the average numbers, thus, we could obtain a time profile within 4 hours [7]. Recently, we have installed a new photo cathode RF gun cavity with Cs-Te cathode which has high quantum efficiency, and we have succeeded in improving optical density and S/N ratio of our pulse radiolysis system as the result.

A new nano-second system which can measure time profile with only one-shot and follow up wider time region than pico-second system has been developed. In the past, this system had been developed using He-Ne laser as proving light, however it can measure only 633nm. Instead of He-Ne laser, this system adopts Xe flush lamp which has broad spectrum. As system evaluation experiments, we tried to get time profile of some species.

In this paper, present status and future plans of our pulse radiolysis systems will be reported.

IMPROVEMENT OF PICO-SECOND PULSE RADIOLYSIS SYSTEM

Pulse Radiolysis System

A diagram of pico-second pulse radiolysis system at Waseda University is shown in Figure 1. This system is based on a stroboscopic pump-probe method. Electron beam ("pump") irradiates sample to generate transient species. Then, probe light ("probe") is irradiated to measure the absorption of transient species, so a transient phenomenon is measured. Optical delay changes irradiating timing of probe. Changing optical length enables us to get another timing data. Repeating this process, we can get a time profile of transient species. The time resolution of this system depends on not time resolutions of detectors but the longitudinal and transverse sizes of electron beam and probe light, thus we can achieve pico-second order time resolution for the measurement of transient species by the electron bunch and laser pulse with pico-second durations.

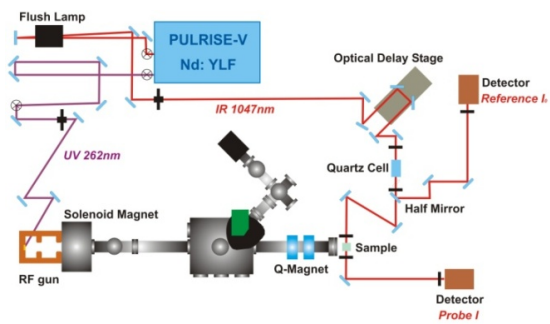

Figure 1: Pico-second pulse radiolysis system at Waseda University.

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Pico-second UV (262nm) and IR (1047nm) laser pulses, which are generated from PULRISE-V, Nd:YLF mode-locked laser, are used for "pump" and "probe", respectively. "Pump" is pico-second electron pulse which generates transient species in the sample. The electron pulse is generated with photo-electric effect by irradiating UV laser pulse to the photo-cathode and accelerated by RF electric field in the RF gun cavity. "Probe" is white light continuum made by focusing IR laser pulse into Milli-Q in the quartz cell. Timing of probe light pulse irradiating into sample cell is controlled by optical delay stage. Probe light is separated into two as probe and reference by a half mirror. Probe light passes sample cell, because transient species in the sample cell absorb light of their own characteristic wavelengths. After passing through the sample cell, the white light is monochromated with interference filter and guided to a detector (PIN photo diode). Reference light is directly monochromated and guided to a detector. We can calculate optical density as follows:

$$\mathrm{O.D.} = \log_{10} \frac{I_0}{I} \qquad (1)$$

where I_0 is the intensity of reference light, I is the intensity of probe light, respectively.

New Photo-cathode RF Gun with Cs-Te

Our photo-cathode RF gun is BNL type 1.6 cells of S-band RF cavity. Recently, we have installed a new cavity with Cs-Te cathode which has higher quantum efficiency and the performance has been more improved than copper cathode we had used before [8]. Optical density, which is the signal of pulse radiolysis system, depends on beam charge (and partly energy), so optical density and S/N ratio would be improved. Comparison of performances of Cu and Cs-Te cathode is shown in table 1.

Table 1: Comparison of Performances of Copper Cathode and Cs-Te

	Cu	Cs-Te
Q.E.	10^{-4}	10^{-2}
Energy	4.6MeV	5.5MeV
Charge	-1nC	-7nC

Evaluation Experiment of Pico-second System

After installing new cavity, we have tried to obtain a time profile of hydrated electron as an evaluation of pico-second system. Hydrated electron has been researched since Hart and Boag, university of Toronto, had observed it in 1960s [9] [10]. Hydrated electron is formed in sub pico-second after irradiating electron beam pulse, and decays in micro-second order time scale. Experimental conditions are shown in Table 2.

Results and Discussions

The evaluation results of pico-second pulse radiolysis system in 2006 and 2007 are shown in Table 3 and Figure 2. We evaluated the optical density (O.D.), the S/N ratio and the time resolution. Signal is obtained as the optical density (O.D). Noise is determined by standard deviation at base part. Time resolution is evaluated be the rise time of this profile. Theoretical prediction of time resolution can be given by following equation:

Table 2: Typical Conditions of Evaluation of Pico-second System (left:2006, right:2007, respectively.)

	2006	2007
Repetition rate	10Hz	10Hz
Cathode	Cu	Cs-Te
Charge	0.67nC	2.1nC
Energy	4.6MeV	5.3MeV

$$\sigma = \sqrt{{\sigma_{Bd}}^2 + {\sigma_{Ls}}^2 + n^2({\sigma_{Bs}}^2 + {\sigma_{Ld}}^2)} \qquad (2)$$

where σ_{Bd} is bunch length of electron beam, σ_{Bs} is beam size of electron beam, σ_{Ls} is the spot size of proving light, σ_{Ld} is pulse length of proving light, n is refractive index of sample, respectively. The O.D. and the S/N ratio results in 2007, have been improved in comparison to 2006. Nevertheless, time resolution in 2007 has been worse. It is considered that bunch length should not be controlled enough way under the experiments.

Table 3: Results in 2006 and 2007

	2006	2007
O.D.	0.024	0.101
S/N ratio	6.1	19.9
Time resolution	18ps	28ps

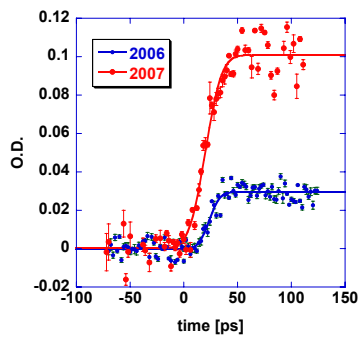

Figure 2: Time profiles of hydrated electron measured at 720 nm in 2006 and 2007.

IMPROVEMENT OF NANO-SECOND PULSE RADIOLYSIS SYSTEM

Measurement System

A nano-second pulse radiolysis system which can obtain a time profile only one shot in shorter time, and follow up wider region than pico-second system has been developed. Probe light in nano-second system was relatively long pulse white light. In order to measure broad spectrum, we adopted Xe flush lamp. Light-emitting time of Xe flush lamp is several hundreds of micro second, so we can use Xe flush lamp as CW light in the nano-second time range.

A diagram of new nano-second pulse radiolysis system at Waseda University is shown in Figure 3. Probe light passes sample cell and passed light is monochromated with a monochromator and guided into the detector.

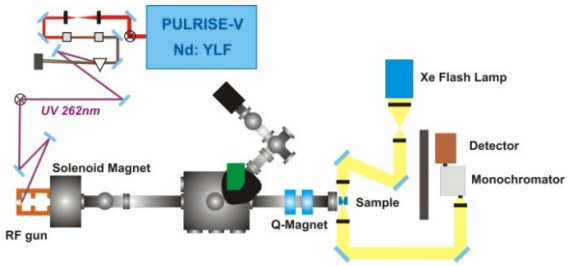

Figure 3: Nano-second pulse radiolysis system

Evaluation Experiment of New Nano-second System

We have tried to obtain a time profile and absorption spectrum of hydrated electron for the evaluation of nano-second system. The experimental conditions are shown in Table 4.

Table 4: Experimental Conditions for Nano-second System

Charge	4.0nC
Energy	4.4MeV
Sampling rate	1GHz
Average	16Ave.
Measurement wavelength	520-920nm

Results and Discussions

Figure 4 shows a time profile of hydrated electron. It decays in micro-second order. The O.D. and the S/N ratio are 0.0853, and 37.1, respectively. Figure 5 shows absorbance spectrum of hydrated electron. It is known that hydrated electron has broad spectrum, and peak wavelength is at 720nm [9] [10]. We have obtained the similar result as in the past reports with only 1 hour. It is obvious that nano-second system has been successfully operated and can be concluded that this system is used for studying on early events in radiation chemistry.

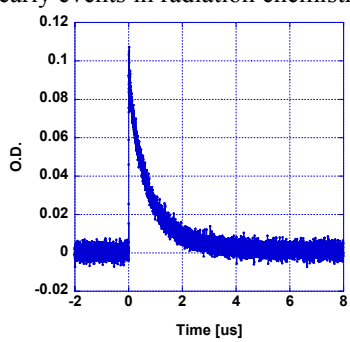

Figure 4: Time profile of hydrated electron at 720nm.

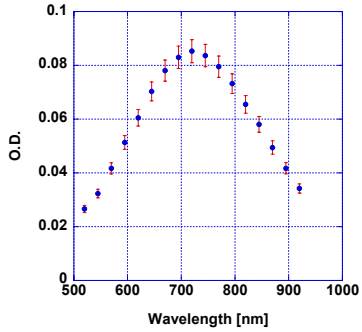

Figure 5: Spectrum of hydrated electron.

SUMMARY

Pico-second and nano-second pulse radiolysis systems at Waseda University have been improved. In pico-second system, O.D. and S/N ratio have been improved by installing a Cs-Te photo cathode RF gun which has high quantum efficiency. In nano-second system, a new system with Xe flush lamp has been developed. Then, we have tried to obtain time profile and spectrum of hydrated electron as evaluation experiment. From these data, nano-second system has successfully operated.

In near future, we will perform the primary process in radiation chemistry, such as geminate ion recombination process in n-dodecane etc.

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