Development and Application of Pulse Radiolysis Systems in Time Region From Femtosecond To Picosecond

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Abstract

Several types of pulse radiolysis systems in the time region from femtosecond to picosecond have been developed. New techniques, such as generation of ultra fast electron pulse, synchronization of laser with electron linac, and timing detection, are adopted. The performance of the system and its application are reported.

1. Introduction

Radiation application is spreading to wide files, such as device fabrication, new material, and medicine. It is important for the high technical application to investigate the primary processes in radiation chemistry [1,2], because the characteristic of the radiation-induced reaction appears in short time region within picosecond order.

Picosecond pulse radiolysis by using electron linear accelerator has been developed since the first stroboscopic method by Hunt et al. in 1979 [3]. However, the time-resolution was kept about several tens picoseconds for the 30 years. Recently, the time-resolution is being broken into subpicosecond time-scale by using several new techniques. The new techniques are as follows.

1) Generation of subpicosecond and femtosecond electron pulse from linac [4,5]
2) Synchronization of femtosecond laser with electron linac [6-9]
3) Timing detection between electron pulse and laser pulse [5]

2. Development of pulse radiolysis system

Figure 1 shows the development history of pulse radiolysis at ISIR, Osaka University. Several types of pulse radiolysis systems for absorption spectroscopes have been developed since 1992. The important factors are time resolution and wavelength region of analyzing light.

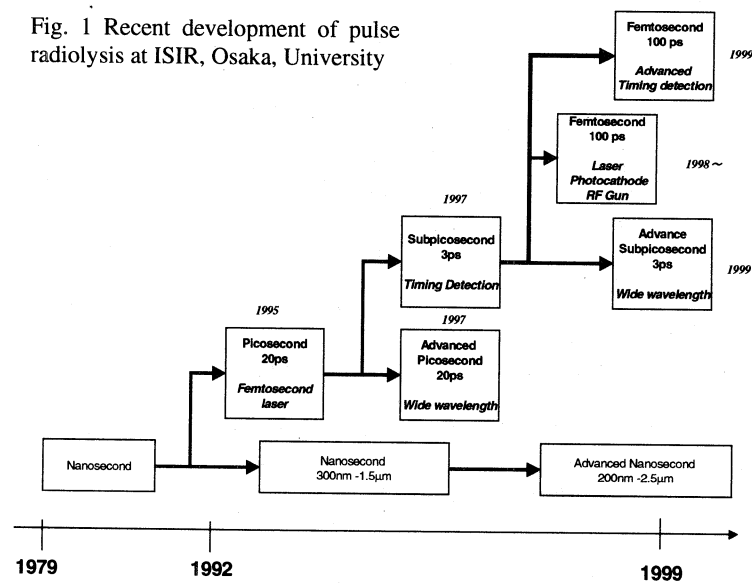
In the stroboscopic method, a sample is irradiated by a picosecond electron pulse to occur a radiation–induced reaction. At the same time, a picosecond analyzing light is injected into the sample to detect the absorption of very short-lived intermediates. Cherenkov radiation was used as the analyzing light in many picosecond pulse radiolysis system [3,10 11]. We used femtosecond laser as the analyzing light. The merits of the laser pulse are as follows.

1) S/N of data is improved due to high intensity of laser pulse.
2) Wide wavelength is available by using the non-liner effect of laser.
3) Whole system is simple.

Figure 2 shows concepts of the stroboscopic pulse radiolysis system in the time region from femtosecond to picosecond.

In picosecond pulse radiolysis as shown fig.2a, electron beams from L-band linac [12] and analyzing light pulse from femtosecond laser are used. The synchronization technique was used between the linac and laser. The available wavelength was extended from 300 nm to 1000 nm by generation of conium light. The

Fig. 1 Recent development of pulse radiolysis at ISIR, Osaka, University



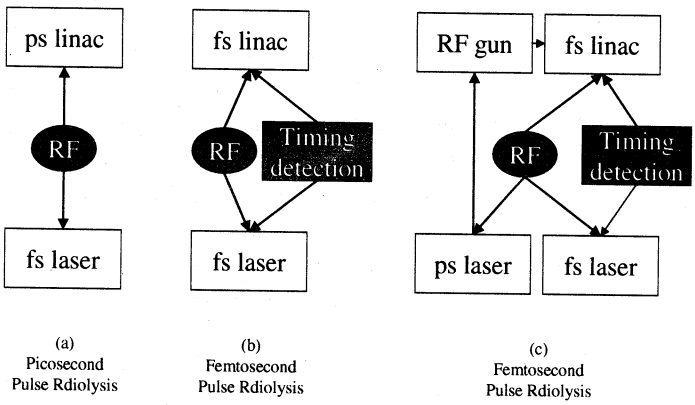


Fig. 2 Cross section of laser photocathod RF electron gun.

primary process of radiation chemistry has been studied by this system.

By generating subpicosecond or femtosecond electron pulse with the pulse compression technique, the time resolution can be improved, as shown fig. 2b. To obtained high time resolution, the timing detection technique was used. The first subpicosecond data were obtained in 1998.

Another method for femtosecond pulse radiolysis is shown fig.2c. The linac is equipped with a laser photocathod RF electron gun [13] to produce high quality electron beam from which ultra short pulse can be generated easily. Two type lasers are used for the analyzing light and for the driver of the gun. The synchronization and timing detection techniques are also used. Preliminary experiment by using the s-band linac has been started since 1998.

The details of the techniques for pulse radiolysis will be mentioned below.

3. Generation of ultra short electron pulse

The ultra short pulse was generated by magnetic pulse compression. The magnetic pulse compressor which consists of two 45° sector magnets, four quadrupole magnets and a vertical beam slit was installed in the L-band linac [12], as shown in Fig. 3.

The longitudinal energy distribution of the electron pulse was modulated so that the energy of electrons in the early phase of the pulse was higher than that in the later phase of the pulse. The phase of accelerating electric field was 70? . The peak energy of accelerated pulse was 26.5 MeV. The energy spread after the modulation was 9.4 %. The pulse length was approximately 30 ps at the end of the accelerating tube. In the magnetic pulse compressor, high energy electrons in the early phase take a long path and low energy electrons in the later phase take a short path. By translating the energy dispersion into the difference of the trajectory length, the electron pulse is compressed at the end of the magnetic pulse compressor. This system can compress the 30 ps electron single pulse to subpicosecond [4]. The electron pulse was used in subpicosecond pulse radiolysis (fig.2 b)

4. Synchronization of femtosecond laser

In the picosecond and subpicosecond pulse radiolysis (fig. 2a, b), mode locked Ti:Sapphire laser was synchronized to the L-band linac. The frequency of the laser was 81 MHz. On the other hand, the basic oscillator of the L-band linac was 108 MHz. The frequency of 27 MHz, which is the greatest common divisor, was used as a common master oscillator.

In the laser photocathod RF electron gun, (fig. 2c), the frequency of the picosecond YAG laser for the drive of the gun was 119MHz. To synchronize with the s-band linac, the basic frequency of 119 MHz was used.

The timing jitter between the electron pulse and the laser pulse was kept in the picosecond order by a timing stabilizer system equipped in the laser system.

5. Timing detection system

The jitter between the laser pulse and the electron pulse was several picoseconds. Therefore, it is difficult to obtain high time resolution in subpicosecond and femtosecond pulse radiolysis only by using the

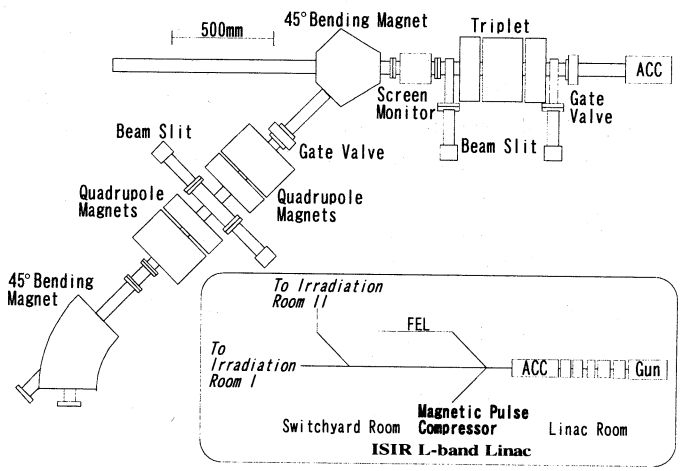


Fig.3 Magnetic pulse compression system

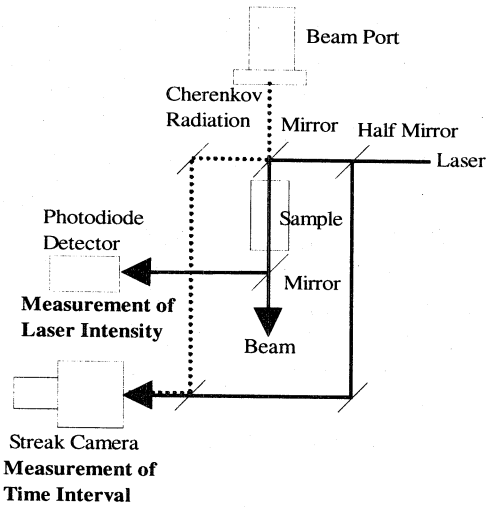


Fig. 4 Timing detection system

synchronizing technique. To break the time resolution below picosecond, the timing detection system was developed.

Figure 4 shows the timing detection system. The time interval between the electron pulse (Cherenkov light) and the laser pulse was measured by the streak camera. The Cherenkov radiation was emitted by the electron pulse in air at the end of the beam line. The laser pulse was separated from the analyzing light by a half mirror. The precise time interval could be obtained by the analysis of the streak image.

6. Time-resolution

The time-resolution are decided mainly by the pulse width both electron and laser in picosecond pulse radiolysis. However, in subpicosecond and femtosecond pulse radiolysis, the time resolution is limited by the difference between the velocity of the light and that of the electron pulse in a sample. The passing time of the electron pulse through a sample is given by $l/(\beta c)$, where l is an optical length of a sample, β the ratio of the velocity of electron to that of light in the vacuum and c the velocity of light in the vacuum. On the other hand, in the case of light, the passing time is given by ln/c, where n is a refractive index of a sample. Therefore, the time resolution is limited by the thickness of a sample. For example, the time resolution is limited to >1.8 ps (10-90% rise time) at n = 1.33 and l = 2 mm.

7. Pulse radiolysis application

The primary process of radiation chemistry has been studied by using the picosecond and subpicosecond pulse radiolysis.

The geminate ion recombination [9,14,15] is the one of the most important reaction in the primary process. A geminate pair of cation radical and electron produced by high energy electron recombines quickly in non-polar liquid. The time-dependent behavior of geminate recombination process was observed directly. The data was analyzed based on the diffusion theory [16].

The formation process of solvated electron [17-19] in polar liquid was also studied. The formation time of solvated electron is picoseconds order. Especially, that of hydrated electron in water is several hundred femtoseconds.

Other studies on primary process in many fields, such as organic chemistry, polymer chemistry and biology, will be started.

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