

MEASUREMENTS OF THE YIELD OF HYDRATED ELECTRONS BY MEANS OF PICOSECOND PULSE RADIOLYSIS

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Abstract

Direct measurements of the initial yield of hydrated electrons in aqueous solutions were carried out using a stroboscopic picosecond pulse radiolysis system. The G value 30 ps after irradiation was determined to be 4.8 ± 0.3 products per 100 eV compared to that of 2.7 after 100 ns.

1. Introduction

The initial yield of the hydrated electron before it escapes the spur is an extremely important parameter for spur diffusion model. Calculations by Schwarz¹⁾ predict it is 4.78. However, there is a substantial difference between the experimental G values obtained in the time region 30–100 ps. Wolff et al.²⁾ reported $G = 4.0 \pm 0.2$ at 30 ps by measurements using a stroboscopic picosecond pulse radiolysis (SPR) system with a train of fine structure electron pulse. Recently Jonah et al.³⁾ reported $G = 4.6 \pm 0.2$ at 100 ps using a single picosecond electron pulse. This significant disagreement prompted us to measure the G value at 30 ps.

2. Experimental

The experiments were carried out using an SPR system⁴⁾ with a time resolution of about 30 ps and the radiation source of a 45 MeV electron pulse from the S-band LINAC at Hokkaido University. The cell used had a 2 cm optical path length and two circular apertures of 3 mm at its front and rear. The pure water in 1 dm³ reservoir was bubbled with pure argon before and during irradiation and then circulated through the cell by a pump.

3. Results and discussion

Figure 1 shows the SPR signals observed at 633 nm in pure water and 0.5 M HClO₄. The yield of hydrated electrons at 30 ps was estimated by simulations considering the pulse shape and radiation doses. The shape of the macro pulse was measured by monitoring the Cerenkov light with a photomultiplier and it could be approximated to a Gaussian with a half width of 11 ns. The dose measurement was carried out by a conventional nanosecond pulse radiolysis technique using the SPR system, which allows a He-Ne laser (633 nm) to pass through the irradiation volume of the sample. Since all measurements were carried out at 633 nm, the yield of hydrated electrons at 30 ps was determined relative to the definite G value of 2.7 at 100 ns.⁵⁾ The dashed lines in Fig. 1 are computer simulated kinetic traces of hydrated electrons. The decay of hydrated electrons during the pulse, which brings about the decrease in the initial step height by a factor of 0.88, was taken into account, and the correction for the density (1.03) was made in the case of 0.5 M HClO₄. The experimental kinetic traces were best reproduced by adapting $G = 4.8$ at 30 ps for both samples. Simulated traces with $G = 4.0$ are also shown in Fig. 1 for comparison. For several experimental results, we concluded that $G = 4.8 \pm 0.3$ at 30 ps. It should be noted that the present experiments give a higher G value than that reported by Wolff et al.²⁾ irrespective of the similar experimental conditions. The G value close to those of the present study has been suggested in certain steady state studies.^{6,7)}

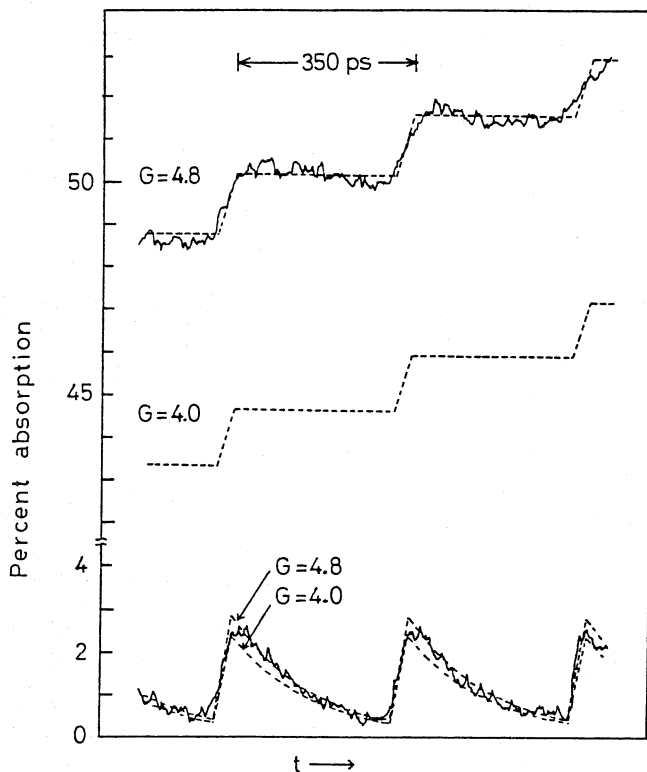


Figure 1. The kinetic traces of hydrated electrons observed in pure water (upper trace) and 0.5 M HClO_4 (lower trace) at 633 nm. The dashed lines are simulated traces with indicated G values.

4. References

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