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TIME- AND SPACE-RESOLVED PHOTOCHROMISM OF HYDRATION ELECTRONS PRODUCED DURING NANOSECOND RADIATION PULSES

by

George J. Berzins, Ki S. Han, and Bruce W. Noel

ABSTRACT

Transient photochromism following the radiolysis of aqueous solutions is studied using existing instrumentation to provide a new technique for space- and/or time-resolved radiation dosimetry. The theory of photochromism due to hydration electrons is examined from microscopic and macroscopic viewpoints. Experimental data are presented. The results are discussed from the perspectives of applicability to radiation-pulse analysis and of correlation between theory and experiment.

1. INTRODUCTION

Past efforts by our group have been directed toward the development of techniques in space- and time-resolved radiation dosimetry. One result of this work is the gated intensifier-vidicon TV system, which has already been successfully demonstrated at rates of 2×10^6 frames/sec with image resolution exceeding 10,000 elements per frame.^{1,2} The utility of this camera for radiation-pulse analysis is currently limited by two factors: the gating electronics, and the response time of the scintillators that convert ionizing radiation to visible light.

Recent work in pulse radiolysis³⁻⁵ has prompted us to consider the applicability of some of those techniques to our camera. We are particularly interested in the concept of photochromism, which involves opacity changes in certain materials upon exposure to ionizing radiation. The possibility of merging a photochromic converter with the framing camera suggests a number of advantageous features that would add a new dimension to radiation-flux mapping technology.

Basic to a photochromic dosimetry system are: (1) a photochromic converter, (2) a light beam that interrogates the converter, and (3) an optical sensor that records the radiation-induced changes in light intensity. One obvious advantage of such a method is that a passive

radiation converter can be placed in a high radiation field, and a laser (or other collimated light) beam can transmit the dynamic information from the converter to a distant recording station. This permits locating radiation-sensitive electronic instrumentation away from the intense fields near radiation sources without using long signal cables. The latter is an important consideration when the radiation source, such as a flash x-ray machine, also generates electromagnetic noise.

The photochromic entity of dominant interest to us is the hydrated (or solvated) electron, e_{aq}^- . The time resolution of solutions containing e_{aq}^- can be decreased to less than a nanosecond (vs 2-3 nsec for currently used scintillators) and can be adjusted over a wide range. The sensitivity of such converters can also be adjusted over several orders of magnitude in dose rate. Another fundamental feature is that the signal is represented by the *absence* of light from a uniform field—a distinct advantage in pre-determining maximum illumination levels for a TV system.

During the past few months, we have investigated the feasibility of combining the concept of photochromism with the gated TV system, and have concluded that it is not only feasible but desirable. In the following sections we discuss the properties of e_{aq}^- from both theoretical and experimental viewpoints. We then present the results

of our laboratory work leading up to and including the initial fast-framing experiment that utilized photochromism of the hydration electron. Finally, we consider the implications of these results, the new problems encountered, and the points to be emphasized in future studies.

II. PHOTOCHROMISM OF THE HYDRATION ELECTRON—MICROSCOPIC MODELS

Experimental and theoretical studies of the physical and chemical characteristics of the solvated electron date back several years. Much of the experimental data has been acquired through pulse radiolysis and kinetic spectroscopy. In spite of the large volume of data, the nature of the binding of the electron still remains controversial.

One fundamental property, that of a unit negative charge for the major reducing species in irradiated aqueous solution, was obtained from measurements of rate constants over a range of ionic strength.⁶ The charge was then deduced from the Debye-Hückel theory⁷ and the Brönsted model⁸ of the ionic reaction. A negative charge is implicitly assumed in the macroscopic equations in Sec. III.

The approach to a model which should subsequently explain the spectral absorption and the lifetime of the absorbing species may be divided into two main sequences: the energy-transfer mechanism of incoming radiation to the solvent, and the rearrangement of transient species produced.

Mozumder and Magee⁹ proposed a model of ionizing-radiation tracks which may be envisioned as continuous "string beads." In their model, the "length of the string" is determined by the range-energy relationships and the different "sizes of beads" (called spurs, blobs, short tracks, and branch tracks) are calculated within a framework of diffusion kinetics. The "separation of beads" is made to agree with the linear-energy transfer. In the original work, a Monte Carlo technique was used to derive the distribution of the "beads." The results of the model showed, for example, a radius of approximately 17 Å for a spur of 100 eV of absorbed energy. The spur contained six pairs of radicals on the average. The "blobs" and "short tracks" are formed by absorbing energy in the ranges 100 to 500 eV and 500 to 5000 eV, respectively. Absorption of energy greater than 5000 eV causes formation of branch tracks.

By studying the reactions of the solvated electron, which are, by definition, electron-transfer processes, it has become possible to understand basic physical and chemical postulates associated with photochromic effects. The solvated electron undergoes reactions of electron-ion

combination, electron attachment, and dissociative electron attachment. In particular, the rate constants have been measured for these reactions¹⁰ and the optical-absorption spectra of the solvated electron have been reported for a number of different solvents, including pure water.

With regard to the rearrangement of the transient species, one of the successful models is based on simple molecular orbital theory using scaled H_2^+ wave functions. The model, proposed by Pohl and Raff¹¹ assumes that the energetic electron produced is preliminarily trapped or localized near the positive regions of the polar-solvent dipoles. Thus, a negative ion of solvent is formed whose lifetime is on the order of 10^{-11} sec. Following this quasi-localization of the electron, rapid reorientation of a local solvent molecule occurs to form a deeper trap whose estimated lifetime is 10^{-5} to 10^{-11} sec. These deeper traps can be viewed as analogs of the hydrogen-molecule ion. Further clustering of the solvent molecules near the deeper trap then occurs almost simultaneously, causing a modest further increase of trap depth and an increased lifetime.

The lifetimes quoted above are estimated on the basis of the measurement¹² of dielectric relaxation time of solvent dipoles such as H_2O . Essentially, the model depicts the solvated electron as being bound by two opposite solvent dipoles. A quantum-mechanical approximation treats the extra electron as localized, as in a scaled hydrogen molecule, in an orbital perturbed by the rest of the solvent. The model yields a theoretical expression for the optical-transition energies that is in reasonable agreement with experimental observations.

A correlation between the transition energy for the photoexcitation and the dielectric constant of the liquid was reported¹³ in a study of the solvated electron in organic liquids. The correlation arises naturally from the Clausius-Mossotti equation¹⁴ which relates the molecular polarizability and the dielectric constant.

Although a full quantum-mechanical treatment of the model is a formidable task, the physical picture of the solvated electron, with particular emphasis on its lifetime and optical transition, appears similar to that of the formation of mesic or kaonic atoms.

III. PHOTOCHROMISM OF THE HYDRATION ELECTRON—MACROSCOPIC MODEL

A. Experimental Considerations.

The hydration electron is hypothesized to be an electron that following radiolysis becomes trapped in a potential well formed by several adjacent molecules or ions. The electron is excited from this state through absorption

of visible or near-infrared light. The absorption spectrum has been found^{15,16} empirically to contain a peak near 7000 Å, with the half-maximum points near 5500 and 8500 Å. The spectrum has been shown to shift to lower energy (longer wavelength) as the sample temperature is increased.¹⁷

Transmission of light through a solution containing e_{aq}^- follows³ the Beer-Lambert law:

$$I = I_0 e^{-\epsilon \ell n} \quad (1)$$

where the absorption coefficient ϵ is a function of wavelength, and ℓ is the pathlength of the light through the solution containing e_{aq}^- in a density n .

The differential equation that governs³ the electron concentration contains a growth term that is proportional to the incident dose rate dR/dt and decay terms that are dependent upon the scattering of e_{aq}^- from its absorptive state by H^+ ions, by other electrons, and by other ion species (such as OH^-). Solution of the differential equation

$$\frac{dn}{dt} = k_1 \frac{dR}{dt} - k_2 [H^+]n - k_3 n^2 - \sum_i k_i [X_i]n \quad (2)$$

allows one to determine the instantaneous electron concentration and, in effect, the opacity of the photochromic sample. The constants k_1 , k_2 , and k_3 have the values 2.76×10^{-9} mol/liter·rad, 2.3×10^{10} liter/mol·sec, and 4.5×10^9 liter/mol·sec, respectively.³ The k_i are empirical constants that have been measured and tabulated by various experimenters. If an acidic solution such as dilute $HClO_4$ is used, all terms in the summation are small and can be neglected.

In an experiment, the measured quantity is the intensity of the light beam I , or, essentially, the fractional transmission I/I_0 . Denoting this ratio by S , one can obtain the instantaneous dose rate for a dilute acidic solution in terms of the known parameters and the measured signal as

$$\frac{dR}{dt} = -\frac{1}{\epsilon k_1 \ell} \left[\frac{1}{S} \frac{dS}{dt} + k_2 [H^+] \ell n S - \frac{k_3}{\epsilon \ell} (\ell n S)^2 \right] \quad (3)$$

in which $R = R(t)$ and $S = S(t)$. The total dose can be obtained by simply integrating over the pulse duration.

For low dose rates (less than 10^4 rad/nsec) and H^+ concentrations greater than 10^{-3} mol/liter, the quadratic term becomes negligible, simplifying the treatment. At high enough dose rates, however, the quadratic term may dominate the linear-decay term. Then, the reaction dynamics may be further complicated by depletion of the H^+ concentration through the reaction $e_{aq}^- + H^+ \rightarrow H$. Therefore, a differential equation describing the rate of

change of H^+ must be included as well. Although this does not affect the early part of the pulse shape (i.e., the quadratic decay term is dominant), it may add a tail to the signal.

B. Predicted Behavior of e_{aq}^- in a Photochromic Converter.

Quantitative predictions of signal amplitude and width resulting from a known radiation-dose profile can be made only from solutions of Eq. (2). The dose rate, on the other hand, can be obtained from Eq. (3), once the signal has been recorded. To investigate the theoretical limitations on experiments involving e_{aq}^- , we performed computer calculations using an artificial pulse formed by superimposing two Gaussian waveforms, and catalogued predicted signals for a wide range of conditions.

To simplify the analysis, we neglected the summation in Eq. (2), an assumption we believe to be consistent with our experimental conditions. Furthermore, for large enough $[H^+]$ and small n , the quadratic term may also be neglected, and the resulting linear differential equation can be solved using standard integrating factors.¹⁸ But, because the latter condition is unduly restrictive, we elected to solve the more general equation by using the Cauchy-Euler method of numerical integration.¹⁹ In Fig. 1 we compare the solutions to the linear and quadratic equations. For electron concentrations greater than 10^{-2} moles/liter, the two approaches predict radically different results, a fact of no small consequence for experiments in which repetitive radiation pulses are not available.

The time resolution of a converter is examined in Figs. 2 and 3. Figure 3 illustrates the nonlinear relationship between dose rate and photochromic response. We used the resulting peak 1-to-valley-to-peak 2 ratio as an index of the converter's resolution ability. It is evident that to preserve good time resolution, either a high H^+ concentration or a high dose rate is necessary. Note that the transmitted signal follows Eq. (1), and therefore is exponential, a disadvantage if linear recording systems are used. This is demonstrated in the lower part of Fig. 3. If a logarithmic recording system is used, the pulse shape is preserved over the entire range of the recording apparatus.

The predicted dynamic range of the converter is limited at the upper end (corresponding to high dose rates) by the saturation behavior of e_{aq}^- . Figure 4 shows that contrast and time resolution will become affected at $[e_{aq}^-] \gtrsim 10^{-1}$ mol/liter. For the artificial pulses used here, this corresponds to $\approx 10^{18}$ rads/sec, which is equivalent to an incident flux of 10^{27} neutron-MeV or gamma-MeV per $cm^2 \cdot sec$.

The low end of the dynamic range can be estimated by calculating the e_{aq}^- concentration needed to produce an optical-signal change just greater than the noise level.

Fig. 1.
Comparison of solutions to the differential equation (2) when the quadratic term is and is not assumed negligible.

Fig. 2.

Calculated response of a photochromic converter to an artificial radiation pulse (top). The fidelity of reproduction increases as $[H^+]$ increases.

Fig. 3.

Calculated response of a photochromic converter to various radiation pulse amplitudes, $[H^+]$ fixed at 0.1 mol/liter (pH 1.0).

From Eq. (1), the measured signal S is given by

$$S = I/I_0 = e^{-\epsilon \ell n}; \quad (4)$$

then

$$\frac{dS}{S} = -\epsilon \ell \, dn. \quad (5)$$

In a typical experiment using He-Ne laser light ($\lambda = 6328 \text{ \AA}$, $\epsilon(\lambda) = 1.3 \times 10^4 \text{ liters/mol}\cdot\text{cm}$) and a 1-cm-thick photochromic cell, a 5% darkening is implied for an $\bar{c}_{sq}^-$ concentration of $4 \times 10^{-6} \text{ mol/liter}$. For the artificial pulse referred to above, this corresponds to $\approx 10^{12} \text{ rads/sec}$.

The other problem, that of determining dR/dt from the measured signal, is solved by Eq. (3). This problem also has been programmed. A least-squares fitting routine is used to smooth fluctuations in the digitized data, reducing the uncertainties in dR/dt that result from noise and signal-reading errors.

IV. EXPERIMENTAL APPARATUS AND PROCEDURE

We ran two types of experiments, spatially integrated measurements to demonstrate time resolution of $\bar{c}_{sq}^-$, and framing-camera experiments directed toward space- and time-resolved radiation dosimetry.

A. Spatially Integrated Measurements.

Our first experiments were simply to observe the presence of transient photochromism due to $\bar{c}_{sq}^-$. These experiments, their semiquantitative results, and the associated problems became the basis for subsequent work.

A typical experiment is shown schematically in Fig. 5. The narrow beam from a 15-mW He-Ne laser illuminated small areas of the photochromic cell. Under steady-state conditions, the two intensities I and I_0 in the optical beam were equal, and a dc current proportional to I_0 flowed in the detector. Changes in the detector current were recorded on an oscilloscope, single-sweep triggered

Fig. 4.

Electron concentration as a function of absorbed flux (initial wave shape and amplitude at bottom).

from the Febetron.* A radiation pulse from the Febetron created $n(t)$ hydration electrons, causing a transient decrease in I which was recorded as a pulse across the load resistor.

In the early experiments, a solid-state photodetector (rise time 5-10 nsec) was supplemented by a gain-100 amplifier (rise time 2 nsec). This system was adequate for observation of slowly decaying pulses, but its usefulness was limited when we used low-pH samples.

To improve the measuring system's response time, we replaced the detector-amplifier combination with a photomultiplier, and replaced the laser with a 3- μ sec (FWHM) flashlamp. Narrow-band optical filters were used to select spectral regions of interest. The tapered dynode resistor string of the photomultiplier permitted linear operation up to a few volts output. The trigger pulse to

*Manufactured by Field Emission Corporation, McMinnville, Oregon.

Fig. 5.

The experimental arrangement for spatially integrated but time-resolved photochromic dosimetry.

the Febetron was delayed 3.75 μ sec from the flashlamp trigger pulse, causing the radiation burst to occur during peak optical intensity. Some of the data recorded with this system are shown in Figs. 6 and 7. This system had rise and fall times of $\lesssim 1$ nsec, compared to 0.5 nsec for the oscilloscope and 3.0 nsec for the Febetron. Another advantage was that the oscilloscope trace a few nanoseconds before burst provided a direct measurement of I_0 . One disadvantage was that the divergence of the flashlamp light required the use of auxiliary optics, a problem not encountered with a laser beam. Several parts of the experimental apparatus are shown in Figs. 8 and 9.

Some of the photochromic cells used in our experiments are displayed in the inset in Fig. 8. The optical windows and most of the remaining structure are

Fig. 6.

Time response of a photochromic converter to electron pulses, pH 3.58-2.22. The pH values are shown below each photograph. The decay time of the photochromic response decreases with decreasing pH, in good agreement with the theory. (Oscilloscope traces have been artificially enhanced for clarity.)

made of high-purity quartz to minimize discoloration and luminescence from radiation. Even so, we observed luminescence in some of the experiments and had to include it in the data analysis. The entrance windows of cells used in electron beams were typically lapped to 0.25-mm thickness. Range-energy calculations²⁰ showed that 600-keV electrons are reduced to 467 keV in passing through an 0.25-mm-thick quartz window, and that they stop 1.3 mm into the photochromic liquid.

We measured the dose by placing calibrated Avisco 195 cellophane (supplied by Field Emission Corporation) directly in front of the cell. The difference in I_0 immediately before and immediately after the radiation pulse gave the dose *in situ*. Because we used the optical beam to sample both the photochromic cell and the cellophane, the dose measurement was in exactly the

same geometry as the photochromic measurement. Typical doses used ranged from 0.5 to 2.0 Mrad. The dose rate was obtained from the measured dose and the known time profile of the radiation pulse.

We eliminated most of the ambient electromagnetic interference from the Febetron discharge by placing the photochromic cell in a metallic enclosure that was grounded to the Febetron. The detector, also in a grounded aluminum box, was located ~ 10 ft from the Febetron. Lead shielding reduced scattered x-ray background to levels that did not interfere with the measurements. Some of the shielding can be seen in Figs. 8 and 9.

Fig. 7.

Time response of a photochromic converter to electron pulses, pH 1.76-0.5. The decay times decrease with pH, in agreement with theory. As pH decreases, the decay time follows that of the radiation pulse more closely, and the response amplitude decreases.

B. Framing-Camera Experiments.

We ran our initial fast-framing experiment to demonstrate that photochromism due to e_{aq}^- can provide space- and time-resolved data simultaneously. Several series of 5-nsec-duration "snapshots" yielded data that agreed with that presented in the previous section.

The experiment is shown schematically in Fig. 10. The framing camera consisted of a gated generation-II image intensifier coupled to a vidicon, and was similar to that described previously.^{1,2} A monitor pulse from the laser was used to discharge the Febetron. A jitterless Febetron monitor pulse, derived from the electron discharge, was used to trigger the pulser that gated the generation-II image intensifier. Known delays were successively inserted between the Febetron and the pulser to provide a

sequence of 5-nsec pictures. The TV pictures were displayed on a monitor and recorded on Polaroid film. The monitor was unblanked two fields after the video coincidence trigger unit initiated the pulse sequence that fired the 0.5-MW dye laser. Optical and electronic delays were used for timing adjustments.

Some of the results of the first experiment are shown in Fig. 11. Light transmitted through the cross section of the 2.0-cm-diam cell (which appears elliptical in the picture) is strongly and weakly attenuated in frames 4 and 7, respectively. Unattenuated light (wedge-shaped bright areas at top center and bottom right center of each frame) bypassing the cell provided reference data, reducing the effects of laser-amplitude jitter. The figure includes a comparison of the time response deduced from the

Fig. 8.

Febetron-room part of the experimental apparatus. In foreground is the 600-keV electron machine. The photochromic converter is inside the hexagonal lead-lined box at the output-beam end of the machine. The light source is at the far end of the optical bench. The inset shows closeups of several of the spectroscopic cells used to contain the photochromic solution.

framing-camera data with the time response obtained from spatially integrated experiments.

V. DISCUSSION

During this study, we demonstrated that photochromism of ϵ_{aq}^- can be photographed with our fast-framing intensifier-TV camera, providing space- and time-resolved radiation dosimetry. The initial experiments were designed to be solely feasibility studies. Because of the relatively long camera-gating pulse (5-nsec FWHM) compared to the Febetron pulse (3-nsec FWHM), these experiments were necessarily only semiquantitative. Clearly, a faster intensifier-gating technique must be developed before the radiation pulse from our Febetron

can be meaningfully dissected with the camera. The photochromic-TV combination can, however, be usefully applied to examine radiation pulses of longer than 5-nsec duration.

The spatially integrated data confirmed that the decay time and sensitivity of an HClO_4 converter can be easily adjusted to meet the needs of a given experiment. The data also showed that the photochromic converter's decay time can be made significantly shorter than that of an NE-102 fluor, a scintillator commonly used in time-resolved (and space-resolved) radiation-intensity measurements. Pulse shapes produced by NE-102 and by a pH 0.96 HClO_4 cell are compared in Fig. 12. The two converters provide radiation-pulse response waveforms of very similar shape. However, the time scale for the

Fig. 9.

Closeup of lead-lined box, shown in Fig. 8, with cover removed. The photochromic cell is held in place in the electron-beam path. The light enters from the right-hand side of the box and passes through a small aperture immediately before the cell. An exit aperture is shown at the left-hand side of the box. Light from this aperture passes through the lens at left and into the monitor room.

photochromic converter is 2.5 times as fast, indicating the time-integrating effect of the NE-102.

Where valid comparisons could be made, our spatially integrated measurement data partially agreed with the calculated values. For example, as seen in Fig. 13, the decay of $\bar{e}_{aq}^-$ agreed with that predicted by Eq. (2) for low pH values only, and was noticeably faster than predicted as the concentration was decreased. We believe that the slight deviations for pH 1.27 and 1.76 in Fig. 13 are due to the photomultiplier fall time, ≈ 1 nsec for 90% to 10%. Possible explanations for the larger deviations at lower HClO_4 concentrations are that unknown impurities could provide species other than H^+ which remove $\bar{e}_{aq}^-$ from solution and/or that recombination between $\bar{e}_{aq}^-$ and H_2O^+ could occur at a greater rate than expected, possibly due to our high dose rate (see below).

The absolute I/I_0 ratios, on the other hand, are difficult to compare with theoretical values, for several reasons. First, because we do not know the detailed energy spectrum of the Febetron, we cannot estimate the electron range, the value of ℓ in Eq. (1), accurately. Direct measurements of the dose within the cell are impossible, and precise determinations using substitute materials are difficult. Second, although we used narrow-band optical filtration, there were finite contributions to the signal

Fig. 10.

The experimental configuration using the fast-framing camera for space- and time-resolved dosimetry.

from other wavelengths. This problem is accentuated by the predominantly blue response of the S-11 photocathode. Third, and perhaps most significant, at dose rates of several megarad per nanosecond, the ionized electron concentration is somewhat less than 1% of the concentration of H_2O molecules in the volume of interest. The measured I/I_0 values in this dose-rate region depart significantly from those predicted by theory. The theory predicts saturation, as seen in Fig. 4, but it appears at much higher dose rates. Possibly, this concentration level saturates the number of available potential wells, a possibility not predicted by existing theory.

The saturation hypothesis may be important also from an academic standpoint. Recent literature²¹⁻²³ has suggested that a precursor to $\bar{e}_{aq}^-$, or a "dry electron," is present in highly concentrated HClO_4 (and other) solutions. We believe that there may be a relationship between the "dry electron" and the saturation effects observed, and plan to investigate this problem in more detail.

Frame 1

Frame 4

Frame 7

Framing Expt.

Photo Diode Expt.

Fig. 11.

Some fast-framing results. Densitometer comparison of the cell transmission vs the reference (wedge-shaped areas) is used to determine I/I_0 . This ratio is plotted for eight frames in the "framing experiment" curve (bottom left). This curve compares favorably with one obtained from a spatially integrated experiment (bottom right).

pH 0.96

NE102 Scintillator

Fig. 12.

The response of an NE-102 scintillator and a photochromic converter (pH 0.96) compared to Febetron radiation pulses (note different time scales).

In other future experiments, we will attempt a comprehensive study of the spatial-resolution capabilities of HClO_4 converters. We will have to emphasize selecting light sources that are suitable from the standpoints of intensity, jitter, and spatial uniformity. We also plan to explore the above saturation hypothesis further in the hope of clarifying our understanding of anomalies observed in our work or reported elsewhere. Another topic of interest is the behavior of HClO_4 in neutron, as opposed to x-ray or electron, fields.

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Fig. 13.

Relative abundance of hydration electrons in four concentrations of HClO_4 following the Febetron pulse. The "observed" values are proportional to the measured $\ln(I/I_0)$. The calculated values were obtained from solutions to Eq. (2). The ordinate values have been arbitrarily shifted to facilitate comparison. The slope (the only meaningful quantity in this case) of the calculated and observed densities agrees well for the lower pH values.

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