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A 400 rad flash x-ray system for pulse radiolysis

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Design and working principle of a flash x-ray system is described. A two-stage Marx generator is used, the charging voltage is 150 kV, the stored energy 1125 J. Anode and cathode are arranged coaxially. The anode consists of a tungsten coated aluminium tube of 20 mm i.d. into which the substances to be irradiated are placed. Measurements of electrical and radiation parameters are described. The x-ray dose per pulse is 400 rad, the half-width of the pulse is 0.2 μsec and the mean quantum energy 44 keV. An example for the application of the system in microsecond pulse radiolysis is presented.

I. INTRODUCTION

For the pulse radiolytic investigation of reactions in biological and chemical systems, pulsed radiation sources are required having a pulse length less than a microsecond and a dose of some hundred rads per pulse. Such a system can be used for the study of primary processes and short lived intermediates in radiation chemical reactions, which play an important role in the early stages of the reaction chain of biological radiation effects.

For pulse radiolysis experiments, mainly electron accelerators are used. Owing to the small penetration depth of electrons in condensed matter, quantum energies of 1.5 MeV and more are necessary. With pulsed x-ray generators a similar penetration depth of the radiation is achieved at much lower accelerating voltage; thus the systems are simpler and less expensive.[1–3] Moreover the interfering effects of Čerenkov light and of excess charge introduced by the high energy electrons can be avoided. For these reasons the FXR-system described below was constructed. Electrode geometry and resulting spatial radiation distribution of the system were optimized for applications in pulse radiolysis.

II. APPARATUS

A. Electrical Part

The high voltage pulses are provided by a two-stage Marx generator. The circuit arrangement is shown in Fig. 1. With a charging voltage of 150 kV, two 0.05 μF capacitors[4] store an energy of 1125 J. 150 kV charging voltage was chosen with the consideration that, while both the efficiency of the x-ray production and the spectral distribution of the radiation become better with increasing voltage, technical difficulties on the other hand, especially with the electrical insulation, increase, which puts the simplicity of the basic concept in question.

The spark gaps consist of two aluminum spheres of 100 mm diam at a distance of 90 mm. The first spark gap is triggered by a trigger pin with a 200 kV pulse (50 nsec risetime) coming from the trigger unit.[4] In order to achieve a low inductance the components were arranged as compactly as permitted by the charging voltage. A liquid resistor was installed for the variation of the generator resistance. For the resistor an almost saturated NaCl solution is used.

B. X-ray Tube

The flash x-ray tube is made of steel and has a cubic geometry with 20 cm inner length (Fig. 2). There are openings at all sides covered by flanges for the different connections. The tube is continuously evacuated to 10^{-5} Torr by an oil diffusion pump. In order to avoid damage to the pumping system by the high voltage, the connection between pump and discharge vessel consists of two glass tubes, each 50 cm long. For the high voltage connection to the cathode, smaller dimensions than commercially available, but having sufficient insulation for pulses up to

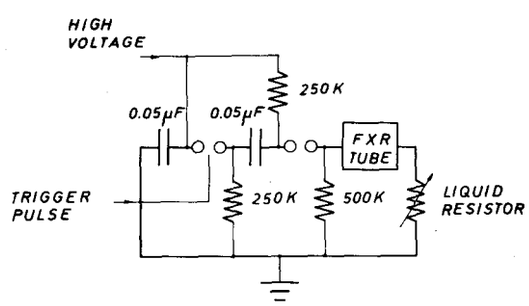

FIG. 1. Circuit arrangement.

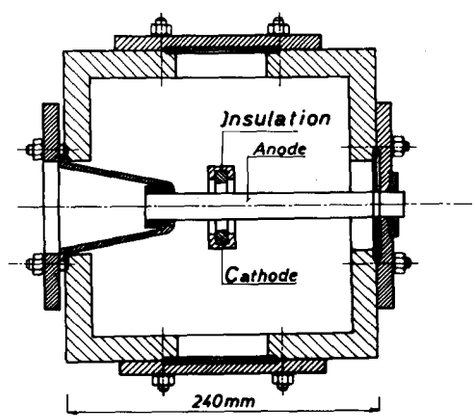

FIG. 2. Flash x-ray tube with coaxial electrode arrangement.

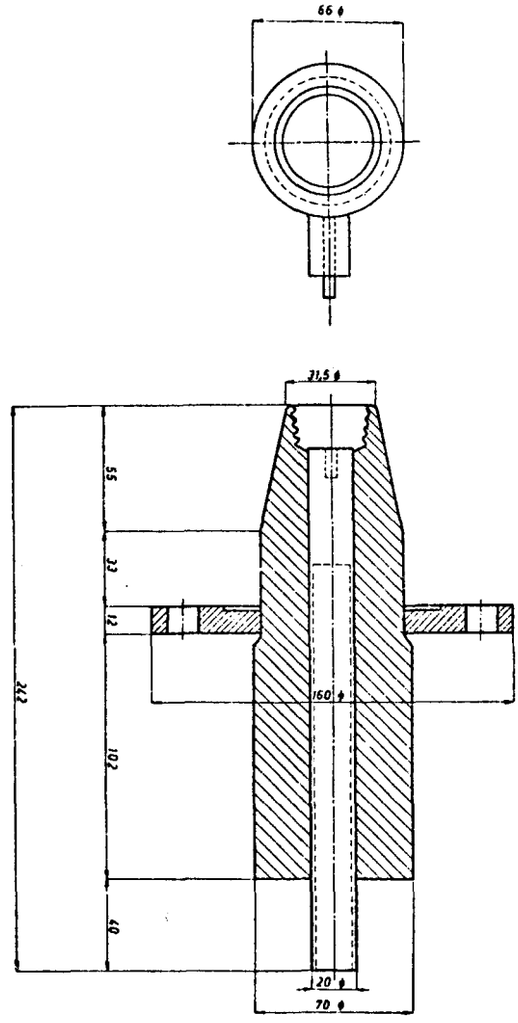

FIG. 3. High voltage connection to the tube and ring cathode.

300 kV, were required. After tests with various materials, insulations using cast Araldit[5] turned out to be the best. A copper rod is surrounded by 25 mm of Araldit; this assembly is sealed to a flange with which it is fixed to the tube (Fig. 3). The cathode consists of a ring of copper or brass, surrounded by Araldit, and is screwed to the copper rod. The anode, in form of a tube, is placed coaxial to the cathode and is sealed vacuum tight to the discharge vessel at both ends. Inside the anode, outside the vacuum, the materials to be irradiated are placed.

Experiments with different cathode geometries (sharp edges or needles) showed no significant dependency of the x-ray dose on cathode geometry. This corresponds to a theory by Fünfer[6] on the first stages of the discharge: only a few electrons leave the cathode by field emission. At the anode they set free positive ions which in turn generate further electrons at the cathode. As field emission plays a minor role, the cathode geometry is not decisive for the current rise.

For the material of the transmission type anode there are contradictory requirements: it should have a high efficiency for x-ray production, but small absorption for the x-radiation, a high melting point to withstand the thermal stress, and sufficient stability against atmospheric pressure. This can only be reached with a two-layer structure. Here aluminum tubes are used with a tungsten layer put on by means of a plasma jet technique.[7] The tubes have an i.d. of 20 mm, the Al wall is 0.8 mm thick. 200 μ tungsten gave the best results if both dose per pulse and lifetime of the anode are considered. An anode of this type lasts for 200–300 discharges; owing to the evaporation of anode material during each discharge the tube walls are not vacuum tight any longer. With increasing damping of the discharge circuit the lifetimes of the anodes are increased, accompanied however by a decrease in x-ray output, down to 50% of the maximum value in the case of nearly critical damping.

III. EXPERIMENTAL RESULTS

The discharge current was measured by means of a magnetic probe placed at the tube near the high voltage connection. Both the signal from the probe, proportional to dI/dt, and the integrated signal were recorded for different damping of the circuit. From the oscillating part of the current curve, that is for a period when the interelectrode space has a small resistance, the inductance L and the resistance R were calculated, which in this case can be put equal to the values L_G and R_G of the generator. The inductance is 1.25 μH, the Ohmic resistance lies between 0.6 Ω without liquid resistor and 3.5 Ω for the case of nearly critical damping. The peak current was calculated to be 41 kA.

The time dependent x-ray output was recorded with a plastic scintillator and a photomultiplier in time correlation to the dI/dt signal. The scintillator was placed inside the anode tube and was shielded with 3 mm of lead to avoid saturation. Via a light guide the signal was transmitted to the multiplier. Near the anode the light guide had to be surrounded by lead in order to avoid fluorescence in the fiber glass. A typical pulse form is shown in Fig. 4. The mean length of the radiation pulse is 0.20 μsec (FWHM) without and 0.14 μsec with an additional resistor. The figure shows that at the beginning of the x-ray emission there is a fast oscillation of the dI/dt signal. The maximum of the radiation pulse corresponds to a dip in the dI/dt curve. This is in agreement with the interpretations of the FXR-discharge mechanism developed by Händel,[8] which say that the x-radiation is emitted at a time when the formation of a gas discharge is going on in the interelectrode space. The discharge finally becomes a pure gas discharge without x-ray emission. Owing to the migration time of the ions, the durations of the different discharge phases, and thus the x-ray output, are strongly dependent on the distance

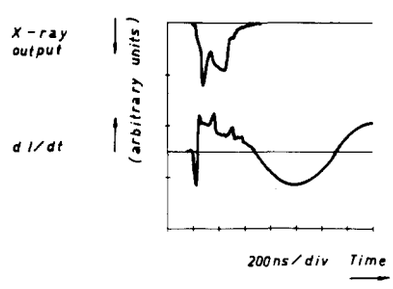

FIG. 4. X-ray pulse and current time derivative in time correlation.

between anode and cathode. A pronounced dose maximum was found for an anode–cathode spacing of 8 mm.

The dose inside the anode was usually measured with LiF–Teflon thermoluminescence dosimeters, sometimes Fricke dosimetry was also employed. The maximum output is about 400 R per pulse, the dose rate 2×10^9 R/sec. The dose is reproducible within an rms deviation of 10%. By using iron filters the mean quantum energy of the radiation was evaluated to 44 keV.

For the application of the system in pulse radiolysis not only the maximum value of the dose is of interest, but also the distribution along the axis of the anode. For, if absorption spectrometry is used as detection method,[9] the light absorption caused by primary products of radiolysis depends on the line integral of the dose along the analyzing light beam. To measure the dose distribution, nine TLD disks were arranged in the anode, their planes being parallel to the x-radiation. The disks were separated by pieces of plastic material, so that the dose could be measured over a length of 5 cm. Figure 5 shows the distribution curve; the points were taken from three different pulses, each with a different maximum dose and were normalized to 100 R. The line integral of the dose was calculated to be 454 R·cm under these conditions.

IV. APPLICATIONS

The x-ray output of the system is high enough to enable investigations of dose rate effects on biological systems, using one or several pulses. The main application, however, is intended to be in the field of pulse radiolysis of dissolved matter, whereby the method of spectrophotoelectric recording is employed for studying the formation and decay of transient species.

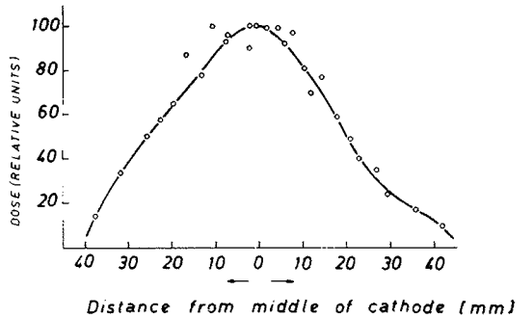

FIG. 5. Dose distribution along the axis of the anode tube.

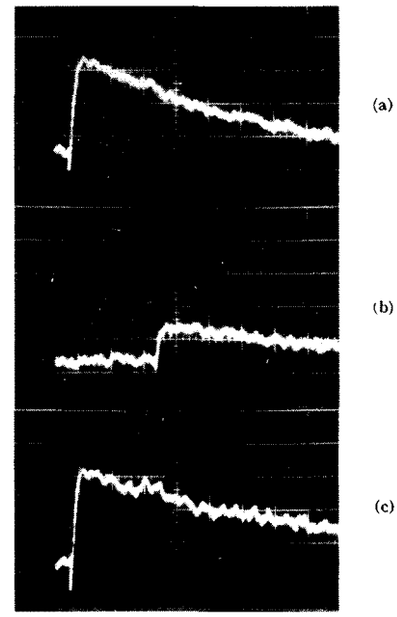

FIG. 6. Pulse radiolysis of eosin. Transient absorption signal at 405 nm; dose 110 rad in (a) and (c), 50 rad in (b). Horizontal scale: 500 μsec/div.

As an example of such a measurement the formation and decomposition of half-reduced eosin was studied (Fig. 6). The oscilloscope was triggered before the x-ray system so that the zero-line is visible. The negative value of the absorption at the beginning is caused by an increase in intensity of the light source due to magnetic interference. During the irradiation hydrated electrons are formed, one of the products of the radiolysis of water. They react with the dissolved eosin, forming a half-reduced substance R, which has an absorption maximum at 405 nm. By another reaction R is transferred back to the primary form. The figures show the transient absorption signal at 405 nm for a 5×10^{-5} molar eosin solution. For the first and third picture the dose was 110 rad, for the second 50 rad.

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