Photochemistry and Photobiology. 1968. Vol. 7, pp. 583–585. Pergamon Press. Printed in Great Britain

AN ULTRAVIOLET PULSE GENERATOR IN THE SUBMICROSECOND RANGE FOR FLASH PHOTOLYSIS STUDIES

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THE EXPERIMENTAL methods now in use for identification of short-lived species cannot be adapted for use in the shorter time range (10^{-7}–10^{-8} sec) necessary for observation of short-lived excited molecules. In order to achieve a spectroscopic analysis in this time range, the light pulses must be shortened by several orders of magnitude.

Modern equipment can produce electron and X-ray pulses of a few nsec, but the production of u.v. and visible-light pulses shorter than a microsecond, with sufficient intensity, presents far greater difficulties, and the shortcomings of these techniques are due to the lack of sources of this type.

The authors have much experience in short-range u.v. generation since they have been studying the measurement of the decay time of very fast scintillators (decay time less than a nsec) for several years[1–3]. After experimenting with about 100 different types of generator[4], they have finally developed a u.v. spark generator with fixed wet mercury electrodes and a hydrogen pressure of 30 atm, capable of producing high-intensity (40–50kW) pulses of a few tenths of a nsec. The pulse is very constant, and the lamps have a long life. The high pressure in the lamp has several functions: to quench the life of excited states (especially metastable states), to remove the corona on the gap previous to the spark, and to increase the spark tension and consequently the flash intensity.

In order to generate high-powered pulses, the spark mechanism for a very short time scale must be understood. It has been found that the liquid electrode (metal wetted by mercury) performs a very important function. The final shape attained by the electrode before the spark is a very fine point (radius 20–30 Å) and is produced by the very strong electric field. This strong electric field (10^{10} V/cm) gives rise to field emission; 10^4 simultaneous avalanches are produced, and with the electrodes 0·5 mm apart, the time needed for full ionization of the gas is less than 0·2 nsec. Solid electrodes require an interactive process of spark formation, which results in a spark formation time longer than 1–2 nsec.

The real time characteristics, of this pulse generator depend on the associated electrical circuit. The spark time characteristics and the power depend on (a) the electrode gap, (b) the gas pressure, and (c) the associated electrical circuit.

We designed some generators with mercury electrodes (in capillary) and variable electrode gap; these generators produced good pulses with the followng characteristics.

Power: 160 kW
Rise time: 0·3 nsec
Width (half intensity): 0·35 nsec
Decay time: 0·4 nsec
Spark tension: 15 kV.

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This is not the upper power limit for these pulses with this type of generator.

The electrical circuit capacity was increased by the use of coaxial cable, which preserved the pulse quality, and a square pulse of 10^6 W was obtained with the following characteristics.

Rise time: less than 1 nsec
Width: 40 nsec
Decay time: less than 1 nsec
Repetititon: greater than 10,000 pulses/sec.

This u.v. and visible-light generator can be used for spectroscopic analysis in flash photolysis or pulse radiolysis studies; and with modern photomultipliers, some improvement in the sensitivity can be obtained.

The main improvements in this generator as compared with conventional ones are the short rise time, the square shape, and the short decay time without tail, of which the short decay time is the most important.*

For production of high-energy u.v. pulses for photolytic action it is necessary to increase the electrode gap to 10–15 mm and the spark tension to 80–100 kV. Many practical problems must be solved before mercury-wet electrodes can be used for this generator. Some prototypes are already being tested.

Meanwhile, a generator has been developed with solid electrodes but incorporating some of the improvements developed after the early experiments. It has the following electrical pulse characteristics.

Rise time: 20 nsec
Width (half intensity): 40–60 nsec
Decay time: 60 nsec
Spark tension: 100 kV (or higher)
Electrical energy: 1500 J
Power: 30,000 mw.
Repetition: 8 sec/pulse (with 150 kV applied tension).

The hydrogen pressure in this generator is 15 atm and it has stainless steel electrodes and good u.v. transmitting glass. This generator has produced more than 1000 pulses without appreciable deterioration of the electrodes. The metallic surface of the electrodes appears to melt and solidify alternately during emission.

Some problems, such as insulation and construction difficulties, have still to be overcome. If the glass is not annealed very carefully, it is broken by the great violence of the shock wave. After much experimentation, glass tubes have been obtained that can withstand more than 1000 pulses without rupture, but after a large number of pulses, fine cracks appear in the zone in front of the electrodes. Tubes of quartz are now being tested.

The dependence of the spark tension on the gap distance, and the dependence of spark tension on the pressure show that it should be possible to increase the power by a factor of two by sparking the flash at over 150 kV. However, this increases also all the difficulties already discussed.

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*A detailed description of this pulse generator will be published shortly.

The condenser used in these experiments was specially designed for currents greater than 100,000 A and has very low inductance (resonance peak about 40 MHz ≃ 13 nsec 0·3 μF, 100 kV insulation).

In order to obtain large square pulses, coaxial cable can be used as a capacitor of very high insulation.

The light output power and spectral characteristics of the sparks thus produced are being studied.

RESUMEN

Los métodos experimentales que se emplean en la actualidad par identificar el electrón hidratado y otras especies transientes no pueden extenderse a escala de tiempos más cortos (10^{-7}–10^{-8} seg), que se requiere para la observación de moléculas excitadas de corta vida, que son precursoras de estas especies transientes, sin sustancial desarrollo. Para alcanzar estos tiempos, los pulsos de excitación sean de electrones, rayos X o luz, así como la luz pulsada para el análisis espectro-fotométrico debe acortarse en varios órdenes de magnitud. Hemos diseñado un generador de luz ultravioleta y visible que emite pulsos menores que 10^{-7} seg y potencia de miles de megavatios. El principio básico consiste en aumentar la tensión de chispa hasta próximo a 100 kV y decrecer la capacidad del circuito de descarga como $\sqrt{V}$, para mantener la energía de la descarga. Suponemos que estos no son los límites finales de intensidad y tiempo. Para obtener pulsos de luz netos, con tiempo de decrecimiento corto y sin cola se han mejorado considerablemente el circuito elétrico y el montaje del generador. Los primeros resultados en el estudio de transientes de corta vida muestran que esta fuente de luz mejorada permite extender el estudio de transientes en la región de 10^{-7} seg.

REFERENCES

1. J. T. D'Alessio and P. K. Ludwig, *Nucl. Sci.* **12**, 351 (1965).
2. J. T. D'Alessio, P. K. Ludwig and M. Burton, *Rev. Sci. Instr.* **35**, 1015 (1964).
3. P. Ludwig, J. T. D'Alessio and M. Burton, *Proc. Indiana Acad. Sci.* **74**, 181 (1965).
4. Patent: J. T. D'Alessio and P. Ludwig, U.S. 3.290.615 AEC Case No S-31.706- *Mercury Wetted Fixed Electrode arc Generator.* June 23, 1965.