present. On the basis of work which has been reported thus far, it is suggested that strong reservations must be held on the interpretation of polywater as a true polymer of water.

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References and Notes

- 1. B. V. Deryagin and N. N. Fedyakin, Dokl. Akad. Nauk SSSR 147, 403 (1962); B. V. Deryagin, I. G. Ershova, B. V. Sheleznyi, Deryagin, Deryagin, I. G. Ershova, B. V. Sneleznyl, N. V. Churaev, *ibid.* 170, 876 (1966), and references cited in these papers.
 M. Van Thiel, E. D. Becker, G. C. Pimental, *J. Chem. Phys.* 27, 486 (1957).
- J. Chem. Phys. 21, 486 (1957).
 B. R. Lippincott, R. R. Stromberg, W. H. Grant, G. L. Cessac, Science 164, 1482 (1969);
 L. Bellamy, A. R. Osborn, E. R. Lippincott, A. R. Bandy, Chem. Ind. London 1969, 686 (1960)
- (1969).
 V. I. Anisimova, B. V. Deryagin, I. G. Ershova, D. S. Lychnikov, Ya. I. Rabino-

- vich, V. Kh. Simonova, N. V. Churaev, Zh. Fiz. Khim. 41, 2377 (1967).
 5. D. L. Rousseau and S. P. S. Porto, Science
- 167, 1715 (1970). 6. V.
- V. B. Karasev and Yu. M. Luzhnov, *Russ.* J. Phys. Chem. Engl. Transl. 42, 1255 (1968). For applications and descriptions of this technique, see L. S. Birks, *Electron Probe*
- Microanalysis (Interscience, New York, 1963).
 S. L. Kurtin, C. A. Mead, W. A. Mueller, B. C. Kurtin, E. D. Wolf, Science 167, 1720

- B. C. KUTHI, E. D. W.C., (1970).
 T. F. Page, Jr., R. J. Jakobsen, E. R. Lippincott, *ibid.*, p. 51.
 G. A. Petsko, *ibid.*, p. 171.
 R. M. Umarkhodzhaev, N. M. Ievskaya, L. V. Matveets, V. A. Gromov, Russ. J. Phys. Chem. Engl. Transl. 42, 1138 (1968); D. E. O'Reilly, H. P. Leftin, W. K. Hall, J. Chem. Phys. 29, 970 (1958).
- Phys. 29, 970 (1958).
 12. A. G. Leiga, D. W. Science 168, 114 (1970) Vance, A. T. Ward,
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X-ray Measurements near High-Power Klystrons

Abstract. The intensity and quality of the x-radiation outside the lead shielding around ultrahigh-frequency and S-band klystrons were measured as a function of high voltage, pulse frequency, and microwave power output by use of ionization chambers. Independent 20 percent increases in each variable gave, respectively, 8-, 1.2-, and 1.5-fold increases in the intensity, and 36, 1, and 4 percent increases in the mean energy of the x-rays.

Many thousands of high-power klystrons are presently in service in radar and communications transmitters and in linear high-energy particle accelerators. Incidental to their normal function as microwave power amplifiers, klystrons generate x-rays with internal intensities approaching 1000 times those produced by commercial x-ray equipment. The hazard is well recognized, and today manufacturers supply klystrons already fitted with carefully designed external lead shields around the tube body and collector; moreover, the collector is designed to absorb locally most of the x-radiation that is generated in it. However, since rectangular wave-guide structures, water lines,

and power cables penetrate into most high-power klystrons, usually there are minor breaks and leakage points in the external lead shielding that permit xrays to scatter out of the tube into working areas.

Very little has been published about the intensity and quality of stray x-rays around klystrons since the Lockport incident called attention to it almost 10 years ago (1). The present study was undertaken in order to obtain some useful information on the potential x-ray hazard associated with certain microwave power transmitters under test or in use at M.I.T. facilities.

X-ray intensities were measured in the ion chamber near a leakage point in

Table 1. Summary of x-ray measurements.

Variable	Volues	X-radiation		Body
	varues	Intensity	Mean energy	(μa)
	S-band	klystron		s
RF output/diode power	0 to 0.30	1 to $30 \times$	1 to $1.5 \times$	8 to 13
PRF	400 to 3000/sec	1 to $8\times$	1 to $1.1 \times$	8
	UHF	klystron		
RF output/diode power	0 to 0.12	1 to $5\times$	1 to $1.2 \times$	200 to 4100
High voltage	139 to 176 kv	1 to $13 \times$	1 to $1.5 \times$	200 to 400

the lead shielding around a 10-Mw Sband klystron operated at constant 120ky high voltage. When the tube was diode pulsing with no radio-frequency (RF) drive, the x-ray intensity rose directly with the pulse rate frequency (PRF) in the range 400 to 3000 per second; the mean energy increased from 90 to 99 kev. When the tube was activated by RF drive to produce the maximum microwave power output at a given PRF, the x-ray intensity increased to about 30 times the undriven values, and the mean x-ray energy rose by about 50 percent to values well in excess of the peak high voltage of the tube. The curves (Fig. 1) resemble the saturation curves that are obtained when RF output is plotted against RF drive.

Similar measurements were taken near a 40-Mw ultrahigh-frequency (UHF) klystron tube operated with fixed diode pulsing conditions: the xray intensity rose 13-fold when the high voltage was increased from 139 to 176 key, and the mean energy increased from 78 to 108 kev. When the tube was RF-driven to the level of 12 percent microwave power output efficiency at fixed voltage points, the x-ray intensity increased about fivefold over the undriven values, and the mean xray energy rose by about 20 percent. The data for the UHF tube are given in Fig. 2. The lower points demonstrate an extraordinary dependence of the xray intensity on the high voltage of the tube; the three upper points show how strongly the x-radiation rises when the tube body is modestly activated by RF drive. The x-ray field condition for the latter points was found to be quite unstable during the fine tuning done to optimize the RF output: the x-ray intensity fluctuated by as much as 50 percent and the mean energy by as much as 18 kev.

The measured x-ray intensities are given on a relative scale for the following reasons: (i) the radiation intensity levels changed greatly with the position of the ion chambers in the neighborhood of the breaks in the lead shielding, (ii) the field-measurement points were arbitrarily chosen, and (iii) the amount and the effectiveness of the lead shielding around different types of high-power klystrons varies. Therefore only relative changes in x-ray intensity and quality with changes in the operating conditions of the klystrons have general significance. The actual x-ray

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Fig. 1. Radio-frequency output dependence of the stray x-ray intensity near a wellshielded klystron. The curves represent eight values of the average diode power, as determined by the PRF. Numbers near points represent mean energy in kiloelectron volts. Tube: VA938c (S-band); 120 kv; 10-Mw peak; $10-\mu$ sec video pulse.

intensities recorded in this study were between 2 and 260 mr/hr (11 r/hr was the peak pulse level) for the modified PIC-6 (Eberline Instrument Company), between 0.6 and 70 mr/hr (peak of 4 r/hr) for the 8004 (EG&G), and between 5 and 25 mr/hr (peak of 25 r/hr) for the 440RF instrument (Victoreen Instrument Company).

Table 1 contains a general summary of the results. The body current of the tube reflects the number of defocused



Fig. 2. High-voltage dependence of the stray x-ray intensity near a well-shielded klystron. Numbers below the lower points refer to the average diode power and the mean x-ray energy; those below the upper points refer to the average radiated RF power when the tube was RF-driven at the given diode power. Tube: L3775 (UHF); 100 20- μ sec pulses per second.

beam electrons striking the klystron body or microwave resonance structure. While one variable was changed over the given range of values to produce the changes in x-radiation, the other variables were held constant. The pulse duration for each klystron was fixed; therefore the PRF was directly proportional to the duty cycle that was 0.002 for the UHF klystron and between 0.004 and 0.03 for the S-band klystron.

Ion recombination and space charge effects are known to distort the responses of ionization chambers operating in pulsed x-ray fields. This problem becomes increasingly important as the chamber volume increases, the collection voltage falls, the duty cycle of the x-ray source decreases, and the source strength increases (2). High-power klystrons are excellent examples of intense, low-duty-cycle sources of pulsed x-radiation; moreover, when RF-driven they are also sources of pulsed microwave radiation that can induce disabling voltages in the electric circuits of x-ray detection instruments. One of the detectors (the 8004) used in this work was in fact inoperable for this reason when the UHF tube was RF pulsing. Curiously, its operation was not disturbed by the RF fields around the S-band klystron. For these reasons, RF-insensitive ion chambers having high collector voltage (the 8004), small volume (the 440RF), or both (the modified PIC-6), were used to make the measurements reported here.

The variable quality of the x-ray spectra that the klystrons generate under different operating conditions (see below) also presents difficulties in the interpretation of ion chamber readings unless the x-ray energy response of the chamber is either flat in the region of interest or known and corrected. The energy response of the 8004 and 440RF instruments is flat, but that of the modified PIC-6 ion chamber varies strongly in the 70- to 150-kev region. Prior to the present study, the absolute x-ray energy response of this instrument was determined with and without the use of plates of copper 0.16 and 0.32 cm thick placed against the base of the instrument between the standard x-ray sources and the detector. The x-ray field measurements reported here were interpreted by use of such calibrations; that is, the ratio of the instrument readings with and without the copper plates defined to ± 1 percent

the mean x-ray energy, and hence gave the correction for energy response.

Two klystron tubes were used in the present study. One was a Varian Associates VA938c five-cavity S-band (2.8 kilomegacycles) tube with 10-Mw diode and 4-Mw RF peak power and microperveance 2.0. It was operated at 120 kv and variable PRF with 10-µsec diode and 9- μ sec RF pulsing. The other was a Litton Industries L3775 fourcavity UHF (0.40 kilomegacycle) tube with 40-Mw diode and 15-Mw RF peak power and microperveance 3.1. The tube was operated between 139 and 176 kv and at fixed PRF (100 per second) with 20- μ sec diode pulsing and 15- μ sec RF pulsing. Both tubes were diode-pulsed by use of a d-c high voltage supply, a line type modulator with a pulse-forming network, and a high-voltage pulse transformer.

Klystrons are active elements in the electrical circuit; the peak tube current I is related to the pulsed high voltage E by the relation

$I \equiv k E^{3/2}$

where k, the perveance, is fixed when the tube is manufactured. The peak diode (or video) power is therefore





proportional to the 5/2 power of E. The product of the peak tube power and the duty cycle gives the average tube power, a quantity to which the x-ray yield is related (see below).

The procedure for the measurement of x-ray intensity was as follows: The ion chambers were mounted in arbitrary positions near the break in the lead shielding around a wave-guide penetration; the chambers were not disturbed during a given set of measurements. Changes in the x-ray fields as registered by at least two different types of ion chambers were recorded for each step change in the PRF, the high voltage, or the RF output.

Although the linear rise in x-ray intensity with PRF was expected, the concomitant gradual rise in mean energy was not. This effect may have been caused by a gradual loading of the beam magnetic focusing fields that cause small changes in the internal x-ray scattering geometry.

The x-ray intensity rose with the 11.5th power of the high voltage E, a dependency that remains extraordinary (E^9) even when the intensity values are normalized to constant diode (video) power. There are two general high-voltage-dependent factors that could contribute to this effect: the collector (copper target) yield of x-rays, and the photoelectric effect (PE) in the copper structures and in the lead shielding. The yield factor is difficult to estimate because it depends on the collector beam-target geometry which cannot readily be determined inside a klystron; moreover, this geometry will vary with beam voltage in a fixedfocusing magnetic field. Finally, the relevant yield is only that yield above a certain photon energy, somewhere between 35 and 50 kev, that has a significant probability of penetrating the copper body or wave guide. By graphically integrating published spectral yield curves above an arbitrary cutoff value, one can obtain 3rd to 5th power dependencies on E(3, 4).

In order for the primary x-ray beam to reach the external ion chambers, it must channel along the copper drift tube and output cavity, through the wall of the output wave guide, and through a crack in the external lead shield (Fig. 3). The energy-dependent PE resonance absorption is the dominant photon interaction in these structures (4). Over a broad range of energy, the x-ray intensity transmitted through typical wall thicknesses of 0.2

to 0.4 cm can exhibit an E^5 dependence. Therefore, the observed high-voltage dependency may be explained in terms of the PE and the source yield factors.

In order to discuss the changes in x-radiation with relative RF output, it is necessary to list the effects of an impressed RF field on the electron beam in the body of a klystron (5). (i) The electron beam is velocity-modulated so that intense bunches of electrons arrive simultaneously at the microwave output cavity. If the electrons are initially at 120 kev, the bunch at the output cavity can contain electrons with energies between 90 and 150 kev. (ii) The electron beam is defocused, an effect that changes the x-ray source distribution within the klystron tube and the internal scattering geometry. (iii) The bunched electrons lose energy to the output cavity in crossing the cavity gap and arrive partially spent at the collector. (iv) Outof-phase (unbunched) electrons are strongly accelerated in crossing the output cavity gap. This acceleration depends in a complicated fashion on the tube voltage, the gap transit time, tuning factors, and RF power-coupling factors. However, a typical S-band klystron output cavity gap of 2 cm could be charged with accelerating electric fields of the order of 100 kev/cm. The large increase in x-ray intensity and in mean energy with increasing RF output is evidence that effects (i), (ii), and especially (iv) more than compensate for effect (iii) on the x-ray yield.

I conclude that local x-ray shielding that is adequate for safe klystron operation at a given high voltage, PRF, and microwave power output may be entirely inadequate if there are significant increases in any of these operating parameters; and that x-ray intensity and mean energy measurements should prove to be a simple useful means of analyzing klystron function since the external x-radiation level is a sensitive indicator of output cavity tuning and power coupling factors.

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References and Notes

- 1. Nine workers at the Lockport, New York, Air Force Station were severely overexposed to x-Force Station were severely overexposed to x-radiation while tuning a high-power klystron [J. W. Howland, M. Ingram, H. Mermagen, C. L. Hansen, Jr., in *Diagnosis and Treatment* of Acute Radiation Injury (World Health Organization, Geneva, 1961), pp. 11–26]. See also M. Ingram, J. W. Howland, C. L. Hansen, Jr., H. Mermagen, C. R. Angel, Health Phys. 519 (1962).
- 2. J. W. Boag, in Radiation Dosimetry (Aca-
- J. W. Boag, in Radiation Dosimetry (Academic Press, New York, 1966), vol. 2, pp. 17-27; R. Zendle and E. Goodale, Health Phys. 2, 78 (1959); ibid., p. 316.
 H. W. Koch and J. W. Motz, Rev. Mod. Phys. 31, 950 (1959); A. H. Comptom and S. K. Allison, X-Rays in Theory and Experiment (Van Nostrand, New York, 1935), pp. 89-90.
 R. D. Evans, The Atomic Nucleus (McGraw-Hill, New York, 1955), pp. 614-617, 698-699.
 Bruce E. Nelson, "Introduction to Klystron Amplifiers" (Publication AEB No. 19, Varian Associates, Palo Alto. California, April 1963).
- Amplifiers" (Publication AEB No. 19, Varian Associates, Palo Alto, California, April 1963). I thank E. Storm of the Los Alamos Scien-6. tific Laboratory, Los Alamos, New Mexico, for carrying out the energy-response calibrations of the PIC-6 ion chamber, C. Heon and N. Efremos of the M.I.T. Lincoln Laboratory, Etremos of the M.I.T. Lincoln Laboratory, Bedford, Massachusetts, for operating the klys-trons, D. Searl for assisting in the x-ray measurements, and A. Browne of the Lincoln Laboratory and R. V. Keating of the M.I.T. Laboratory for Nuclear Science, Cambridge, Massachusetts, for helpful discussions. Sup-ported in part under AEC contract AT(30-1)-2098.

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Droplet Countercurrent Chromatography

Abstract. A new form of countercurrent chromatography, named droplet countercurrent chromatography, has been developed. This all-liquid separation technique is based on the partitioning of solute between a steady stream of droplets of moving phase and a column of surrounding stationary liquid phase. Miliigram quantities of dinitrophenyl (DNP) amino acids were separated with an efficiency comparable to that of gas chromatography.

Liquid-liquid chromatography and countercurrent distribution (1) are powerful analytical procedures for the purification and identification of a wide variety of compounds. These techniques have certain limitations, however. Adsorption of solute to the support used in liquid-liquid partition chromatography often causes peak tailing, and the capacity of this technique is low. Although solid supports are not used in countercurrent distribution, this technique is more cumbersome and generally has less resolving power but higher capacity. Recently, Ito and Bowman reported on a simple all-liquid microtechnique called countercurrent chromatography, which involves partitioning in a long helical tube in a centrifugal field and which has an efficiency comparable to that of