Reports

Anomalous Water: Characterization by Physical Methods

Abstract. *Anomalous water samples have been prepared in fused quartz capillaries from pure water in an unsaturated atmosphere. In agreement with observations of other investigators, water prepared in this manner, after concentration, exhibited an increased viscosity, a lowered vapor pressure, a phase separation at low temperatures, an index of refraction of 1.48 or greater, and a depression of the temperature of maximum density. However, electron microprobe examination indicated that a significant weight fraction of these concentrated anomalous water residues consists of sodium, boron, and oxygen. The presence of about 6 percent boron was also confirmed through neutron activation analyses and by mass spectrometric measurements. A parallel is drawn between the similar physical properties of anomalous water and highly concentrated sodium tetraborate solutions. The possibility of polymorphism in liquid water should be accepted only with serious reservations.*

Profound consequences for structural chemistry and for theories of the liquid state are implicit in the proposal of polymorphism in liquid water advanced by Deryagin and his co-workers (*1*). Although dimers, trimers, and higher polymers of water in a solid nitrogen matrix at 20°K have been reported from infrared studies (*2*), and liquid water has been likened to a hydrogen-bonded quasi-polymer whose structure is changing extremely rapidly, it was an entirely new and unexpected report that a relatively stable polymeric form of liquid water could be prepared under appropriate experimental conditions, even though on a very small scale. Lippincott and his co-workers (*3*) reinforced the Russian workers' conclusion that a modified form of water had been prepared, a form characterized by physical properties significantly different from those of ordinary water. Lippincott and his co-workers considered polywater, the concentrated form of anomalous water, to be a true water polymer with a symmetrical hydrogen bond that has an energy of 30 to 50 kcal/mole (*3*).

Proponents of the view that polywater is a new form of water have recognized that many of the physical properties ascribed to this material could be duplicated with salt solutions. For instance, a phase separation at low temperatures, an increased density, increased refractive index, increased viscosity, a shifted temperature of maximum density, and a lowered vapor pressure can all be produced in aqueous salt solutions. Many investigators have not analyzed their anomalous water samples because only submicrogram quantities have usually been available; others (*3*) have reported only small amounts of ionic materials in their products. From evidence gathered in this study, it is suggested that the container and capillary surfaces are sources of ions and that these materials contribute to a significant degree to properties assigned to polywater. A hydrated salt containing sodium, boron, and oxygen is strongly indicated as a principal constituent of polywater.

This report is chiefly concerned with anomalous water and the concentrated product, polywater, as prepared solely from water without the use of added salts for humidity control; however, we also present some experiments in which a saturated solution of potassium sulfate was used in the apparatus described by Anisimova *et al.* (*4*). These results are included not only because this procedure was originally suggested as an improved method for the preparation of anomalous water, but also because it is plausible that a related mechanism for filling the capillaries is operative when only pure water is used. These experiments are described first.

Quartz tubing with an inside diameter of 0.5 mm was first cleaned with 5 percent hydrofluoric acid and then with sulfuric acid, and was finally rinsed with several liters of doubly distilled water. After the tubing had been air-dried, capillaries with diameters between 2 and 70 μm were drawn in an oxyacetylene flame. Approximately 25-cm lengths of drawn capillaries were sealed at each end and the outer surfaces were cleaned, first with acetone, then with alcohol, and then with ether. A degassed, saturated potassium sulfate solution was placed in the bottom of the tube with 6-cm lengths of the cleaned, open-ended capillaries placed in the ring-sealed upper section of the tube. Care was taken to prevent accidental contamination of the capillaries during the evacuation and seal-off operations. After a week at room temperature, the tube was opened. These procedures were repeated several times with the same results. Approximately 25 percent of the capillaries contained some liquid. For those columns at least 5 mm in length, the capillaries were sealed and individually examined in an apparatus designed for the determination of the temperature of maximum density (TMD). This apparatus consisted of a vacuum-jacketed cell which contained ethyl alcohol as the heat-transfer fluid and a traveling microscope which could be used to estimate column length to ± 2 μm. The temperature of the bath was read with a copper-constantan thermocouple and was controlled by a combination of gas flow and heat input regulation of a helium gas stream cooled to liquid-nitrogen temperatures which passed through a stainless steel U-tube placed in the cell. In accord with observations of earlier investigators, a TMD between 0° and −18°C was found for the liquid samples prepared in this manner. A sample of capillary liquid which exhibited a TMD of −17°C was forced with syringe pressure onto a cleaned and fired platinum disk 2.5 cm in diameter. The residue, visible upon microscopic examination, was analyzed with electron microprobe methods. The major constituents of the residue were potassium and sulfur (and possibly oxygen). Crystalline birefringent residues of capillary contents were also identified as potassium sulfate from measurements of the index of refraction. Fluorine, chlorine, sodium, and silicon were not present within the limits of detection, which are estimated to be between 10^{-6} and 10^{-8} μg for these elements. However, it is not possible to make use of this full sensitivity in the detection of these elements because of the presence in the substrate of trace impurities at this level or higher.

Because of the strong indication that the transport of the potassium sulfate solution to the capillaries occurred by way of the moisture film on the walls of the ring-sealed tube, attempts were made to minimize this possibility. One plastic and three Pyrex vacuum desiccators were used with saturated potassium sulfate solution to maintain the condition of 95 percent relative humidity. The saturated potassium sulfate solution was placed in petri dishes together with an inverted cover suspended by glass hooks. In each of the desiccators, the capillaries were placed on two parallel 6-mm glass rods about 40 mm apart. In addition, in the plastic desiccator the glass rods were supported on a polyethylene disk. In all, several hundred capillaries were placed in the desiccators. The air was removed from the desiccators at the beginning of the experiment, as usual. With this procedure, not a single capillary contained an observable column of liquid after a week. This result suggests that the earlier success in filling the capillaries was a consequence of contact with the vessel walls. The experiments of Rousseau and Porto (*5*) give support to this proposal.

For the remainder of this report, unless otherwise specified, references to anomalous water or polywater apply to a product prepared only with triply distilled, degassed water in an apparatus similar to that described by Karasev and Luzhnov (*6*). This Pyrex assembly consisted of a capillary chamber separated by a break seal from a water compartment. Provision was made for both the capillary and the water compartments to be connected separately to the vacuum system. Quartz capillaries prepared in the manner described above were placed on a stainless steel stand with the open ends several centimeters away from the points of support. Approximately 100 to 200 capillaries were used in each trial and these were baked at 375°C for 24 hours under a pressure of $\leqq 10^{-5}$ torr. A U-tube immersed in liquid nitrogen was placed between the capillary compartment and the stopcocks of the vacuum line during the bakeout period to minimize the possibility that vapor from stopcock grease would reach the capillary surfaces. The interior of this apparatus had also been cleaned in the same way as the capillaries were, and distilled water was boiled in the two chambers several times in an effort to remove water-soluble material. Distilled water which was redistilled from dilute alkaline permanganate solution was distilled a third time under vacuum into the water bulb after it was degassed by repeated freezing-thawing cycles. After the apparatus connection to the vacuum line was sealed and the break seal between the water and capillaries was broken with a glass-enclosed magnet, we maintained a condition of approximately 98 percent relative humidity around the capillaries by circulating water which was 0.3°C warmer than the bath water through a jacket which surrounded the capillary chamber. The entire assembly was immersed in a water bath held at 23.5°C and controlled to ± 0.01°C. After a brief period during which saturation conditions prevailed, the controlled temperature differential was maintained, usually for 1 to 2 weeks, before the capillaries were removed for examination.

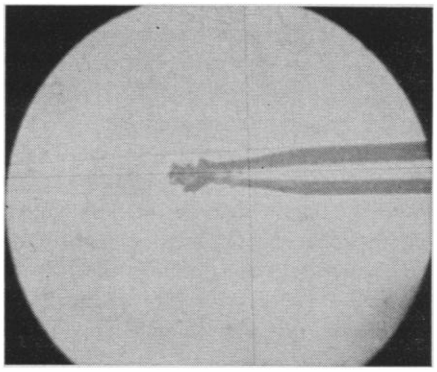

Fig. 1. Air-dried anomalous water residue prepared from pure water. Gel-like material forms long, tapered menisci and has a refractive index which closely approximates that of the quartz capillary.

Generally between 5 and 10 percent of the capillaries had liquid in them. Column lengths varied, but lengths equal to or less than 1 mm were much more common than those of a centimeter or more. In several trials, not a single capillary out of nearly 200 had a liquid column. It is believed that the variable success ratio was dependent upon whether the capillaries had come in contact with the capillary chamber wall, as has been suggested by other experiments discussed here. Although in this series of experiments the capillaries were suspended from a stainless steel stand, on occasion some of them were found to have been jarred by vibrations in the water bath so that some were in contact with the wall for at least part of the time. These capillaries usually contained some liquid. However, it is not possible to state that all capillaries which contained liquid had contacted the wall.

Upon exposure of the capillary liquid to room air, there was a gradual decrease in the length of the column and an increase in the index of refraction as measured with a Zeiss interference microscope. During the concentration process, it was noted that many droplets of anomalous water half filled the bore diameter and moved freely in a wave-like motion along the capillary. This creep characteristic has been cited as one of the features of anomalous water. If the material of high refractive index was left in contact with room air, the column would dry further and form long, tapered menisci (Fig. 1). A somewhat granular appearance was occasionally noted. Further drying resulted in a gel-like deposit.

A depression of the TMD was found for the anomalous water formed in the quartz capillaries under the conditions described above. Usually the TMD was between -5°C and -11°C, with the lower temperatures obtained with partially concentrated samples. In samples examined at low temperatures, we have also observed the phase separation ascribed to anomalous water. It was not possible to perform measurements of the thermal expansion with the desired precision on the highly concentrated samples because these columns were generally less than 3 mm in length, and, in addition, the definition of the meniscus was often poor.

It was considered to be of significance to determine whether a weighable residue could be obtained from anomalous water samples. If the product were principally water with only a trace of impurities, it would not be possible to obtain a weighable residue with the small volumes of material present in a single capillary. However, if the material were composed of a relatively nonvolatile polymer of water, or if there were appreciable quantities of inorganic salts present, a weighable residue would be obtained. Liquid columns were transferred to platinum tabs 0.5 mm in diameter under a protective plastic canopy to keep out dust particles. The tabs were weighed on an ultramicrochemical balance (Rodder series E) (sensitivity = 3.7×10^{-9} g per division). Residues which approximated 1 percent of the total weight of the anomalous water samples were obtained, and these weight determinations not only demonstrated that there was an appreciable quantity of nonvolatile matter in the anomalous water, but also made possible quantitative estimations of the sample compositions. An anomalous water sample, when dried in the capillary in air, would have a residual column which was approximately 1 percent of the original length.

Analyses of the anomalous water residues were made with an electron microprobe (Applied Research Laboratories model EMX), an instrument of extreme sensitivity which is particularly suited to the determination of all but the lightest elements in particles of a few micrometers in diameter (*7*). The principal components by weight were sodium, boron, and oxygen. Chlorine and silicon were found, but these were small weight fractions of the residue. Nitrogen, sulfur, phosphorus, potassium, and titanium were looked for but were not detected; however, the sensitivity of detection of nitrogen suffers somewhat from high background levels. In Fig. 2, a–c, are shown the x-ray intensity distributions from Na, Si, and Cl obtained by scanning for each element independently over the same sample area. In order to make estimates of weight percentages of the elements, suitable standard solutions of sodium chloride, sodium tetraborate, and sodium silicate were prepared. A measured column length of the standard solution in a capillary of known diameter was transferred to a platinum tab in the same manner as that used for the anomalous water transfer. Expected residue weights were confirmed by direct weighing on the ultramicrochemical balance; these weights approximated a microgram. Residue weights from the standard solutions were chosen to provide x-ray intensities which bracketed those given by the anomalous water residues under the same instrumental conditions. Although only estimates of the weights of the elements present in the residues are possible because of the uncertainty of the sample areas, the variable sample thickness, and possible segregations and self-absorption of the x-rays, the estimates are rendered somewhat more reliable by the use of standards measured under like conditions. With these experimental limitations in mind, we found that the results from the electron microprobe analysis of an anomalous water sample indicated the presence of at least 4 percent boron and 6 percent sodium. (Although oxygen was found in the anomalous water residues at intensities comparable to those observed in the sodium tetraborate–sodium silicate standards, the lack of knowledge of the state of hydration of these materials in the vacuum of the electron microprobe instrument precluded a direct quantitative estimation of this element.) These weight percentages coupled with the knowledge of the presence of oxygen (inferred from chemical considerations to be roughly three times the amount of sodium) account for at least a third of the residue weight. Another anomalous water residue, about eight times the original weight of the previous sample, contained 3 percent boron and 5 percent sodium. The ratio of boron to sodium was the same in the two samples.

The determination of boron in the anomalous water residues was also made in an isotopic dilution experiment. A known amount of enriched (96 percent) $^{10}\mathrm{B}$ as sodium borate was added to a weighed residue. From the mass spectrometric measurement of the $^{10}\mathrm{B}/^{11}\mathrm{B}$ ratios as the $\mathrm{Na_2BO_2^+}$ ion, it was found that the anomalous water contained 6 percent boron (by weight). Neutron activation analyses of boron in anomalous water gave confirmatory results. The gamma radiation emitted from the excited lithium atom $({}^{10}_{5}\mathrm{B} + {}^{1}_{0}\mathrm{n} \rightarrow {}^{7}_{3}\mathrm{Li} + {}^{4}_{2}\mathrm{He})$ was detected by a gamma-ray spectrometer with a germanium crystal detector and compared with the intensities obtained from known standards.

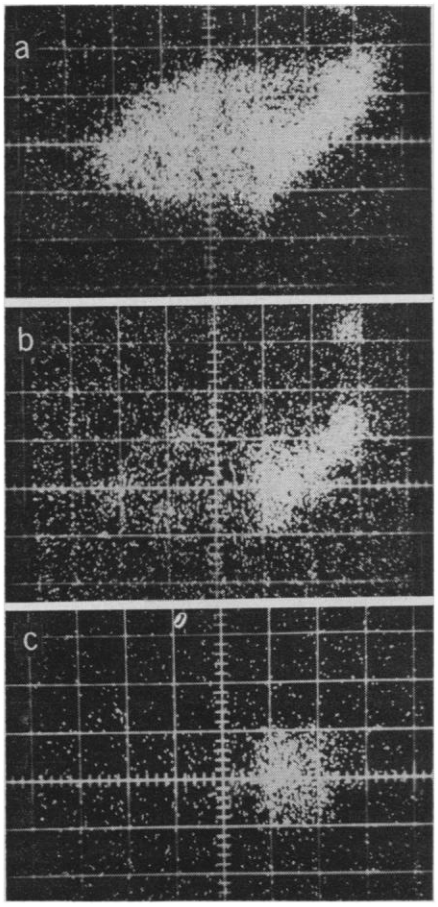

Fig. 2. X-ray intensities of different elements obtained from the same sample area of an anomalous water residue with the electron microprobe (× 2000). (a) Sodium; (b) chlorine; (c) silicon. Boron and oxygen were also present but scanning patterns are not shown. Trace impurities in platinum tab are shown outside the sample area.

If, in addition to boron, sodium, and oxygen, consideration is given to water of hydration, an even larger fraction of the original sample weight can be accounted for. In any case, it has been demonstrated from the quantities of inorganic salts found in the anomalous water preparations that there is reason for concern that these substances may contribute in a significant degree to the properties of polywater. Recent electron microprobe examinations of anomalous water (*5, 8*) have also provided evidence for the presence of significant quantities of inorganic salts.

In order to determine whether sodium, boron, and oxygen could have been transferred from the Pyrex container wall to the capillaries during the high-temperature bakeout procedure referred to above, a cleaned platinum disk was subjected to 30 hours at 375°C under vacuum in the Pyrex apparatus. Electron microprobe examinations showed no traces of these elements above the background of a comparable platinum blank. If this heat treatment directly contributes to the presence of sodium, boron, and oxygen in the contents of the capillaries, the transfer to the capillary interior would have to be remarkably efficient.

Proton nuclear magnetic resonance measurements of anomalous water were made at 60 Mhz with a nuclear magnetic resonance spectrometer (Varian DA-60). Increased signal-to-noise ratios were obtained with the C-1024 (Varian Associates) time-averaging computer. Precision-made, coaxially spaced capillary sample tubes (Wilmad Glass Company) were used. Although the proton resonance of ordinary water was observed in quartz capillaries 10 to 20 μm in diameter after 10 to 100 scans, the report of a broad resonance attributed to polywater protons 300 hz downfield from that of bulk water was not confirmed (*9, 10*). Although an apparent broad absorption signal was observed approximately 300 hz downfield from the ordinary proton resonance, a corresponding number of scans at the same radiofrequency levels with an empty capillary gave a hump in this same region. Background spectral traces were not shown in previous reports. The absence of a downfield proton resonance in the work reported here, in spite of over 10,000 scans, may be the result of a smaller sample or an unfavorable spin-lattice relaxation time. A broadened proton resonance shifted 300 hz downfield from that of bulk water has been observed for protons adsorbed on silica gel (*11*). In the application of time-averaging techniques to extremely small samples of water, the possibility of a contribution of surface-adsorbed water to the reported shifted resonance should be recognized.

Because electron microprobe examinations indicated the presence of impurities in the anomalous water, it was of interest to try to reproduce some of the properties ascribed to polywater with concentrated salt solutions. A 1 percent aqueous solution of reagent grade $Na_2B_4O_7 \cdot 10\ H_2O$ was used to fill quartz capillaries, the contents of which were then allowed to dry in room air. These samples showed an increased refractive index upon evaporation until a match with that of the quartz was noted. Just as shown in Fig. 1, the gel-like residue from the dried borax solution developed long, tapered menisci.

Capillaries filled with a room-temperature saturated solution of borax were dried overnight under vacuum and displayed a lowered vapor pressure as shown by slow evaporation rates paralleling our anomalous water samples. Although some of the vacuum-dried capillaries had liquid columns which had decreased to about one-half the original lengths, others appeared to be empty. However, a thin film of residue was noted upon examination with a Zeiss interference microscope. The very deliquescent nature of the dried residue was noted upon exposure to an atmosphere saturated with water vapor. The wave-like nature of the liquid adhering to the wall and of the pendant drops which slowly moved in the capillary were identical to the behavior observed for partially concentrated anomalous water samples. No crystals were visible upon microscopic examination with polarized light. Stable supersaturated borax solutions can be readily prepared in micrometer-sized capillaries. Although capillaries filled with the saturated borax solution had a TMD of − 5°C, the columns partially dried in a vacuum showed TMD's of − 21°C. The thermal expansion curve was almost flat between − 10°C and − 25°C. A sodium metaborate solution was prepared from equimolar quantities of sodium hydroxide and boric acid. This solution in a quartz capillary showed a gradual decrease of column length as the liquid was cooled to − 50°C in the manner of polywater. No sudden expansion such as that which accompanies the formation of ice was seen in this gel-like material.

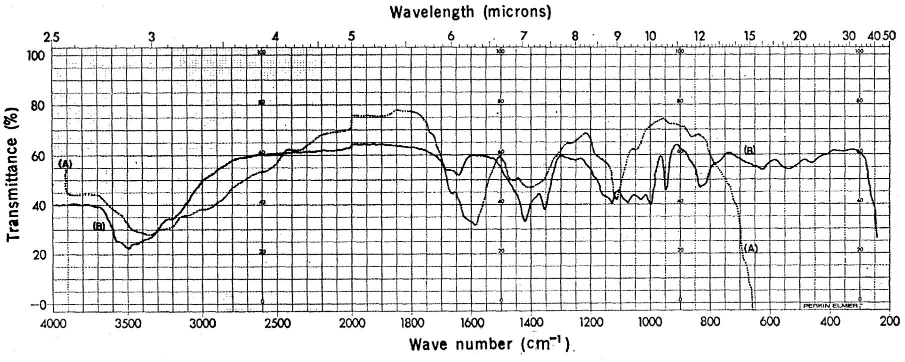

Fig. 3. Infrared spectrum of polywater obtained by Page *et al.* (*9*) (curve *A*) compared with that of a pressed disk formed from 0.5 mg of $Na_2B_4O_7 \cdot 10H_2O$ ground with 200 mg of KBr (curve *B*).

Evidence to support the existence of polywater as well as the postulated symmetrical hydrogen bond has been based in large measure on the interpretation of the infrared spectrum. Lippincott and his co-workers (*3*) have pointed out notable features which include intense bands at 1400 and 1600 cm^{-1} and the lack of an O–H absorption band in the 3000- to 4000-cm^{-1} region. Because of the presence of "residual water" and a significant absorption by the Irtran plate, the complete absence of an absorption by polywater in this 3450-cm^{-1} region was not demonstrated in the more recently published polywater spectrum (*9*).

Since sodium, boron, and oxygen were found in our preparations of anomalous water, we examined for comparative purposes the infrared spectrum of a compound of these elements. The infrared spectrum of a pressed disk of a ground sample of 0.5 mg of sodium tetraborate decahydrate combined with 200 mg of spectroscopic quality potassium bromide was recorded with a spectrometer (Perkin-Elmer model 621) (Fig. 3). The background of pure KBr used is essentially flat with only about a 5 percent absorption in the O–H stretching region. There is a relatively larger absorption in the 3450-cm^{-1} region for the borax than that reported for either of the polywater infrared spectra of Lippincott and his co-workers (*3, 9*). A relatively strong doublet-like absorption band near 1400 cm^{-1} and a band at approximately 1650 cm^{-1}, which are not unlike those of polywater, are observed for borax, although the reasons for the absorptions at these wave numbers may not be the same. Rousseau and Porto (*5*), however, have obtained infrared spectra of polywater samples which not only contained significant quantities of impurities but also showed absorptions at 1400 and 1600 cm^{-1} and a relatively weak absorption at 3450 cm^{-1} as well.

Although there are similarities in the spectra shown in Fig. 3, we do not propose that polywater is necessarily a single inorganic compound dissolved in water nor that it is even chiefly sodium tetraborate. This spectrum does point up the fact that care must be exercised in drawing conclusions about the character of a polywater preparation from observations on a single band in the infrared spectrum since a number of substances, including nitrate (*12*), appear to give spectra which show resemblances to the polywater spectrum.

This study of the character of anomalous water prepared by published techniques has indicated that a significant weight fraction of inorganic salts are present in material prepared in quartz capillaries from water alone. Although a number of similarities are noted between concentrated borax solutions and polywater, other compounds in the sodium-boron-oxygen system may be 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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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 x-rays 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 the lead shielding around a 10-Mw S-band klystron operated at constant 120-kv 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.

Table 1. Summary of x-ray measurements.

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

Similar measurements were taken near a 40-Mw ultrahigh-frequency (UHF) klystron tube operated with fixed diode pulsing conditions: the x-ray intensity rose 13-fold when the high voltage was increased from 139 to 176 kev, 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 x-ray 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 x-ray 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