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CHARACTERIZATION OF A PULSED X-RAY SOURCE FOR FLUORESCENT LIFETIME MEASUREMENTS*

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ABSTRACT

To search for new, fast, inorganic scintillators, we have developed a bench-top pulsed x-ray source for determining fluorescent lifetimes and wavelengths of compounds in crystal or powdered form. This source uses a light-excited x-ray tube which produces x-rays when light from a laser diode strikes its photocathode. The x-ray tube has a tungsten anode, a beryllium exit window, a 30 kV maximum tube bias, and a 50 μA maximum average cathode current. The laser produces 3 x 10^7 photons at 650 nm per ~100 ps pulse, with up to 10^7 pulses/sec. The time spread for the laser diode, x-ray tube, and a microchannel plate photomultiplier tube is less than 120 ps fwhm. The mean x-ray photon energy, at tube biases of 20, 25, and 30 kV, is 9.4, 10.3, and 11.1 keV, respectively. We measured 140, 230, and 330 x-ray photons per laser diode pulse per steradian, at tube biases of 20, 25, and 30 kV, respectively. Background x-rays due to dark current occur at a rate of 1 x 10^6 and 3 x 10^6 photons/sec/steradian at tube biases of 25 and 30 kV, respectively. Data characterizing the x-ray output with an aluminum filter in the x-ray beam are also presented.

I. INTRODUCTION AND MOTIVATION

This pulsed x-ray source was developed as a new tool in an ongoing search for scintillators that could improve the performance of PET (positron emission tomography) detectors. Faster and brighter scintillators may improve both the detector recovery time, the ability to reject events that Compton scatter in the patient., and could also reduce the cost. Previous efforts to find new scintillators used an electron synchrotron in single-bunch mode to measure the x-ray excited fluorescence of over 400 compounds [1, 2], but use of a synchrotron is costly and time-consuming. We have developed this bench-top pulsed x-ray source to reduce the cost and increase the ease of making these measurements.

The design priorities for the pulsed x-ray source included compact size, low cost, the capability to observe fluorescence from powders as well as crystals, and the capability to determine scintillation time structure on the order of tens of picoseconds. The incorporation of a light-excited x-ray tube manufactured to produce very brief (≈ 100 ps) pulses of x-rays [3] made these priorities realistic. The design of the pulsed x-ray source is described in [4]; the primary purpose of this paper is to characterize its performance.

II. SYSTEM DESIGN

A. General Design

The two primary components of the pulsed x-ray source are the light-excited x-ray tube and the diode laser, as shown diagramatically in Figure 1. The x-ray tube is essentially a single-stage photomultiplier tube, with a photocathode which releases electrons when light is absorbed. The electrons are accelerated across 30 kV (typically) into a tungsten anode, and x-rays are produced when the electrons impact the anode.

The light-excited design of the x-ray tube makes it possible to generate short pulses of x-rays simply by directing short pulses of light onto the photocathode of the tube. The repetition rate of the x-ray pulses can be varied by changing the repetition rate of the light pulses. For the pulsed x-ray source presented here, a diode laser was used as the light source because of its short laser pulse duration, easily varied repetition rate, and relatively low cost.

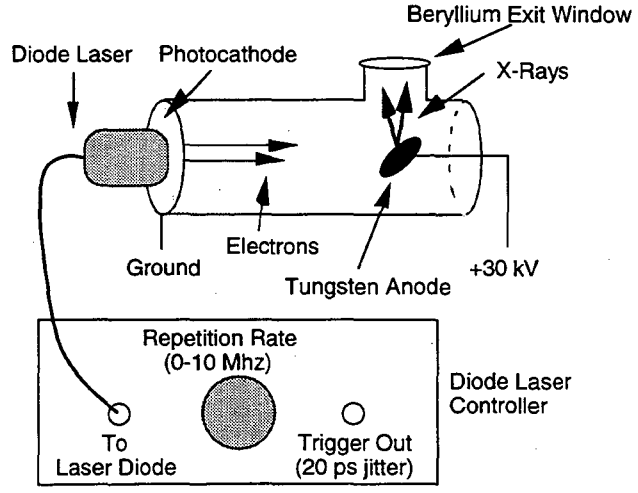

Figure 1. Schematic of the pulsed x-ray source. A laser diode excites the photocathode with <97 ps pulses at ≤10 MHz.

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Figure 2 is a photograph of the pulsed x-ray source. The steel box on the table top is the sample chamber, and directly behind it is the x-ray tube. The laser diode is mounted above the x-ray tube, and the laser diode controller is on the stand above the table top. The sample chamber is evacuated by the pump under the table top. As the system is configured in this photograph, scintillation light passes through the quartz telescope on the right side of the sample chamber and into a microchannel plate photomultiplier tube. The high-voltage power supply, which powers the x-ray tube, and the x-ray control panel, which includes extensive safety interlocks and a current meter for monitoring the x-ray tube cathode current, are both mounted in the lower right portion of the table.

B. Component Characteristics

The characteristics of the diode laser and light excited x-ray tube are summarized in Table I and Table II.

III. System Characterization

A. System Impulse Response

We measured a 97 ps fwhm time spread for the laser diode coupled to a microchannel photomultiplier tube. A 41 ps fwhm (full-width at half-maximum) time spread for the photomultiplier tube was derived from a factory-measured combined time spread of 50.8 ps for the photomultiplier tube and a diode laser by assuming addition in quadrature of the pulse widths and using the known 30 ps laser pulse width for that laser. Deconvolving this 41 ps contribution from the 97 ps measurement yields a value of 88 ps fwhm for the pulse duration of our diode laser. The theoretical estimate for the time spread of the x-ray tube is 31 ps, based on the design of the tube. By combining in quadrature the 88 ps fwhm laser diode pulse width with the 31 ps estimated time spread of the x-ray tube, we obtain a 93 ps fwhm predicted x-ray pulse width. (See [5] for further discussion of the predicted impulse response.)

We are not equipped to directly measure the x-ray pulse width. As an approximation to this measurement, we added an ultra-fast scintillator and a microchannel plate photomultiplier tube to the system in order to obtain fluorescence data as an upper limit on the impulse response of the laser diode, the x-ray tube, and the photomultiplier tube. The scintillator used was malachite green oxalate in crystal form, which likely has a fluorescence decay time of 10-30 ps (based on fluorescence decay times of malachite green solutions [6]). The data were collected over 235,000 seconds.

The observed fluorescence time structure of the malachite green is shown in Figure 3. As noted in the figure, the full-width at half-maximum for the peak is 120

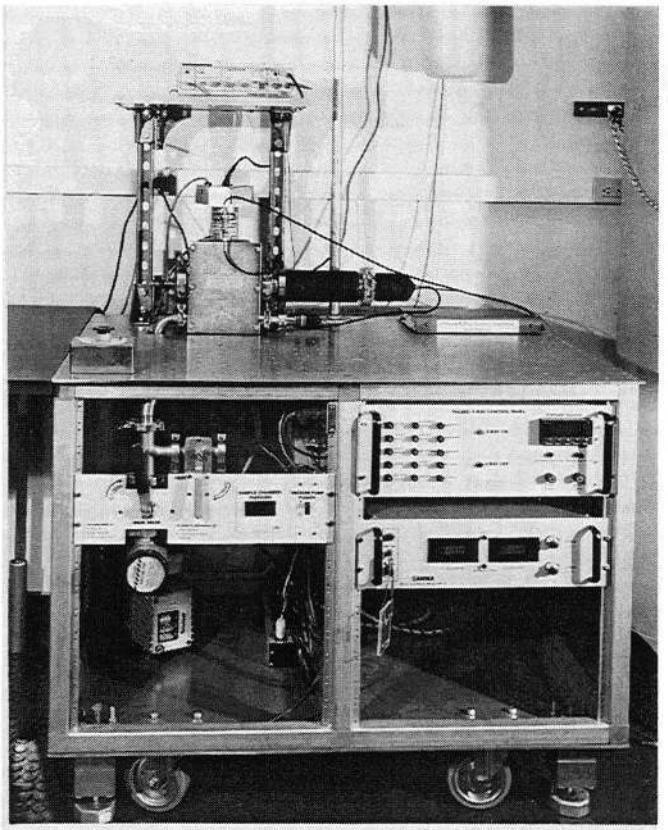

Figure 2. Photograph of the pulsed x-ray source.

Table I. Characteristics of Hamamatsu PLP-01 Light Pulser with C3551-01 Controller and LDH065 Laser Diode Head

Emission wavelength	650 nm
Peak power	>100 mW
Average power (max)	0.1 mW
Pulse width	<97 ps fwhm
Pulse repetition rate	dc to 10 MHz
Photons per pulse	$> 10^7$
Timing pulse jitter	±10 ps

Table II. Characteristics of Hamamatsu N5084 Light-Excited X-Ray Tube

Overall tube length	152 mm
Tube Diameter	52 mm
Photocathode	S-20
Quantum efficiency @ 650 nm	> 10%
Photocathode diameter	12 mm
Target material (anode)	Tungsten (45°)
Output window material	Beryllium
Output window diameter	20 mm
Output window thickness	0.5 mm
Cooling	Natural air
Tube voltage (max)	30 kV
Average tube current (max)	50 μA
Peak tube current	2 A (2 μs duration)
Photocathode photon rate (max)	3×10^{15}/s @ 10% QE

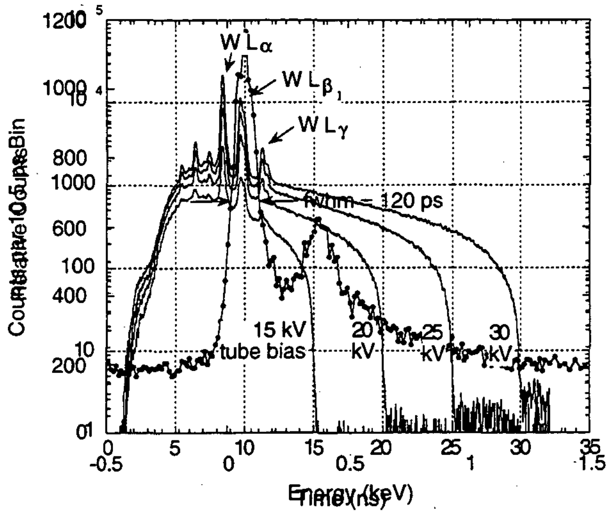

Figure 3. Energy spectra of the x-ray source at various tube biases. oxalate crystals, which provides an upper limit of 120 ps for the system impulse response.

ps. We can conclude, then, that the system impulse response for the laser diode, x-ray tube, and microchannel plate photomultiplier tube has a width less than 120 ps, which is consistent with the above estimate of 93 ps for the x-ray pulse width. The secondary peak in Figure 3 has not yet been explained and further measurements are required to determine its origin, but it is a stable artifact.

B. Energy Spectra of X-Rays

The measured spectral output of the x-ray source was obtained using a lithium-drifted silicon detector. The detector was cooled to liquid nitrogen temperatures and had an energy resolution of 180 eV fwhm at 2 µs peaking time. The detector was positioned 14 cm from the x-ray tube anode, with a lead pinhole collimator placed over the detector to limit the count rate. Each spectrum was collected over 1800 seconds using a 200 kHz laser diode pulse repetition rate. Corrections were made for limited absorption of high-energy x-rays in the 6 mm-thick detector (a 21% adjustment at 30 keV). The data in these energy spectra were used for the remaining figures. (The background flux of the x-ray source in the absence of laser light has been subtracted from the following figures, with an extrapolated value used for the case with a 32 kV tube bias and no filtration. This background flux will be discussed in a later section.)

Figure 4 shows the spectral output of the pulsed x-ray source at various tube biases. These spectra are typical for x-rays generated by an x-ray tube with a tungsten anode, with bremsstrahlung radiation at high energies and the characteristic tungsten peaks around 10 keV. The additional peaks between 5 keV and 8 keV correspond to iron, chromium, manganese, and copper characteristic peaks from fluorescence of various system components.

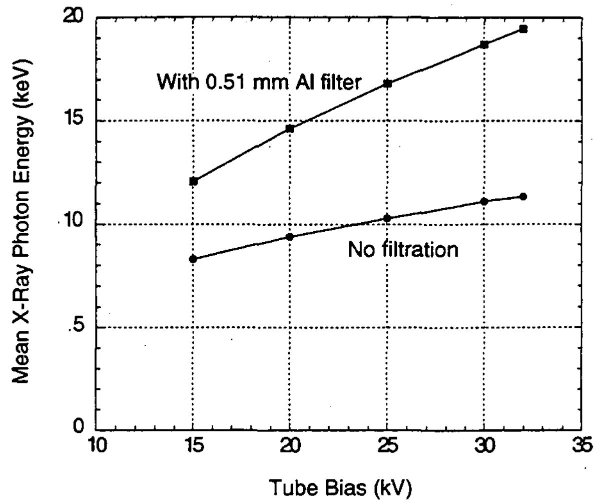

Figure 6. Mean energy of x-ray photons at various tube biases, with and without the aluminum filter.

In practice, we place a 0.51 mm aluminum filter in the x-ray beam to eliminate most of the low-energy x-ray photons. These low energy photons would be absorbed in the quartz cuvette holding the sample; they would not contribute to the fluorescence signal from the sample and in fact would generate a low level of fluorescence from the quartz. The spectra which result after aluminum filtering are shown in Figure 5.

Figure 6 shows the mean x-ray photon energies as a function of tube bias for the spectra shown in figures 4 and 5 and for spectra obtained at a tube bias of 32 kV. As expected, the aluminum filter significantly increases the mean photon energy.

C. X-Ray Flux

The relative flux of x-ray photons was determined by integrating the counts in the spectra and making minor corrections (<2%) for system dead time. This relative

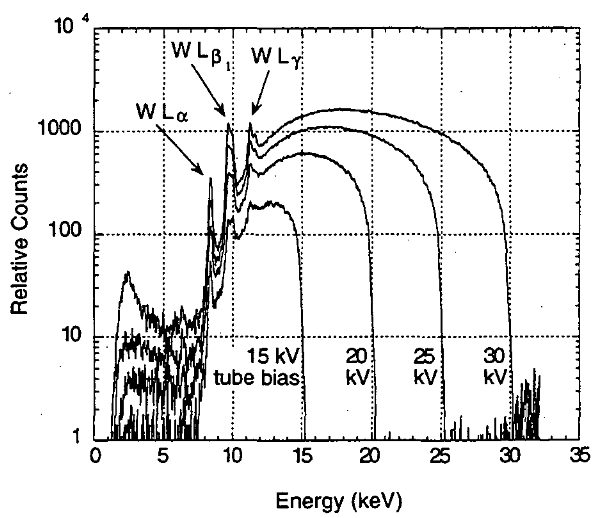

Figure 5. Energy spectra of the x-ray source with a 0.51 mm aluminum filter.

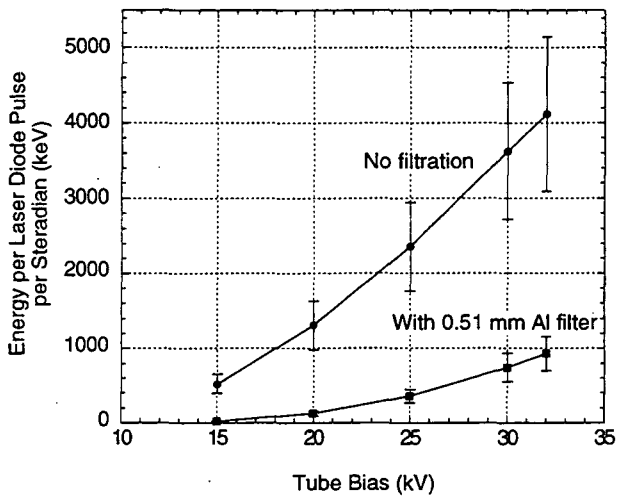

Figure 8. Mean energy deposited per laser diode pulse.

flux was converted to the absolute flux in number of x-ray photons per laser diode pulse per steradian by using the distance from the x-ray tube anode to the detector, the size of the lead collimator pinhole, and the laser pulse repetition rate. The result is shown in Figure 7. The flux increases nearly linearly with tube bias.

Multiplying the flux of Figure 7 by the mean photon energy of Figure 6 yields the energy deposited per laser diode pulse per steradian, which is shown in Figure 8. As expected, the higher mean photon energy for the filtered case compensates somewhat for the lower flux, so that the filtration loss is not as dramatic for energy deposited as for photon flux. Knowledge of the energy deposited per laser diode pulse is important for determining fluorescence efficiencies.

The pulsed x-ray source generates a background x-ray flux in the absence of laser diode light. In the model of the light-excited x-ray tube as a single stage photomultiplier tube, this would be the x-ray flux resulting from dark current. Characterizing this background flux is necessary to determine the extent to which it contributes noise to fluorescence time spectra.

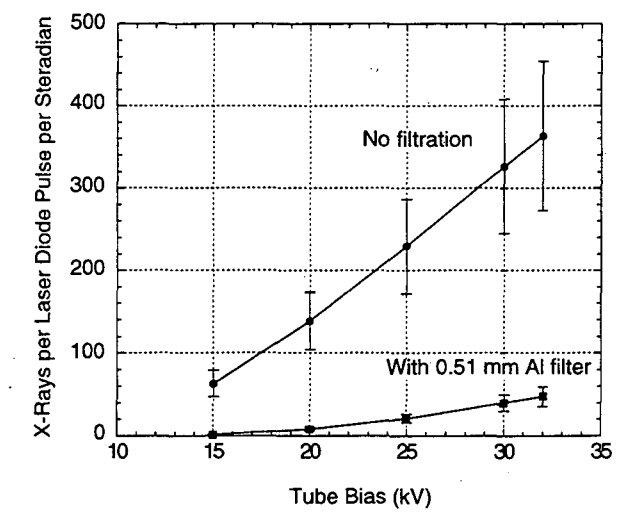

Figure 7. X-ray flux resulting from laser pulses. The 25% error bars (in this figure and figures 8 and 9) are based on the estimated precision of the lead collimator pinhole diameters.

We have observed a significant variation in the background flux over the life of the tube thus far. Two months after we began using the tube, the dark current at 30 kV tube bias decreased by a factor of twenty, apparently as a result of a spontaneous discharge within the x-ray tube. Since that time, the dark current at 30 kV tube bias has increased slightly but remains a factor of ten below the original level.

Figure 9 presents both pre-discharge and post-discharge data for the background flux. The pre-discharge flux data were calculated from the current generated in a photodiode placed in the x-ray beam, with scaling applied (based on laser-induced flux measurements) to compensate for limitations of this method. The post-discharge flux data were determined from spectra of the background flux by the same technique as used for the laser-induced flux above. The background flux tends to increase exponentially with tube bias.

Observations suggest that the background flux rate is independent of laser diode repetition rate. This has not been directly verified, but the tube cathode current at a given tube bias with the laser on appears to be the sum of the dark current at that bias (as measured with the laser off) and a value which is essentially independent of tube bias. The dark current, then, appears to be independent of laser operation, which suggests that the background flux is independent of laser operation as well.

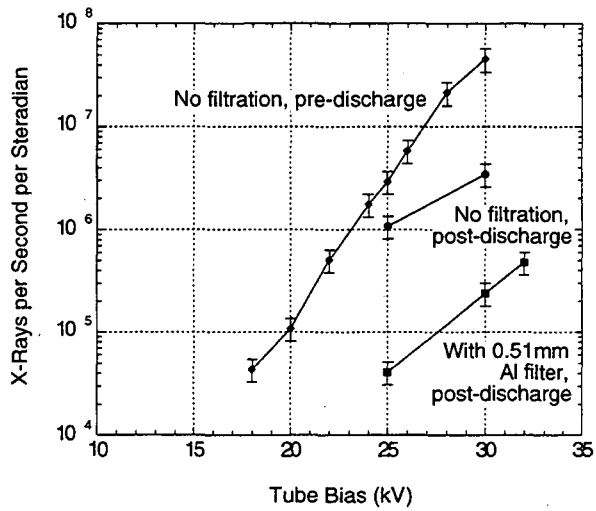

Figure 9. Background x-ray flux in the absence of laser light.

IV. CONCLUSIONS

The pulsed x-ray source has several appealing features for the characterization of fast scintillators. It is a compact, table-top device, and is relatively inexpensive with a parts cost of about $50,000 (U.S.). It has a pulse width of less than 120 ps fwhm and with a well character-

ized impulse response it can be used to determine fluorescence decay times to within 50 ps. The repetition rate of the x-ray pulses can easily be varied by adjusting the laser diode pulse repetition rate. Single fluorescence photons can be detected and spectra can be averaged over time, so that accurate measurements of weak fluorescence can be made. Finally, a monochromator can readily be incorporated to select fluorescence wavelengths or to obtain fluorescence spectra.

The pulsed x-ray source also has a few limitations, which include the existence of a background flux, a low total flux, and a 30 keV maximum x-ray photon energy. The background flux is a limitation for applications in which timing information is required, but with the use of a high laser repetition rate (or windowing of data acquisition around the laser pulse, or use of a brighter light source) a high ratio of desired x-rays to background x-rays can be achieved. Also, the background flux simply generates a low uniform background in time-averaged fluorescence time spectra. The low total flux is only a limitation for applications requiring a high flux (such as acquisition of fluorescence wavelength spectra) and could be overcome by using a more intense light source. (The laser diode used here generates about 1/500 of the rated x-ray tube cathode current of 50 μA.) The 30 keV maximum photon energy may not be high enough for some applications, but it is sufficient for the intended purpose of scintillator characterization.

V. ACKNOWLEDGMENTS

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