

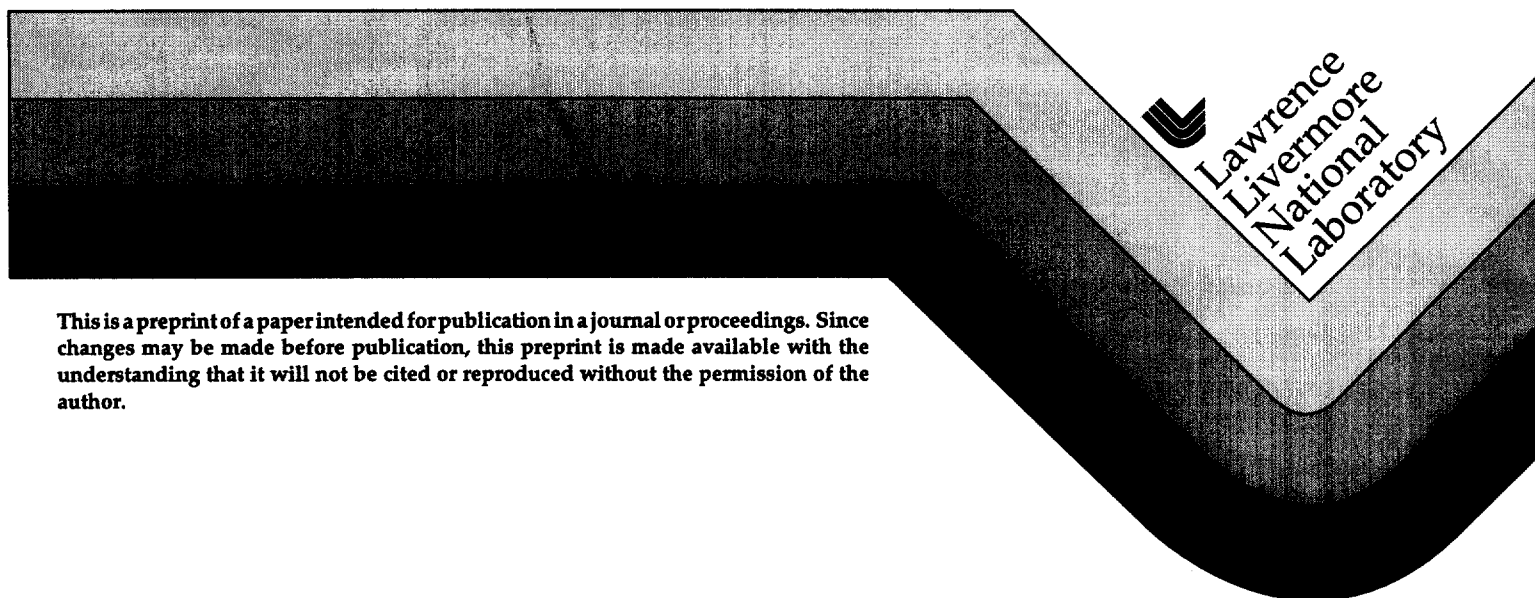
UCRL-JC-127588  
PREPRINT

# Long Term Bleaching of Optical Glasses Darkened by $\text{Co}^{60}$ Ionizing Radiation

G. Richard Wirtenson  
Richard H. White

This paper was prepared for submittal to the  
*SPIE's International Symposium on  
Optical Science, Engineering and Instrumentation*  
San Diego, CA  
July 27-August 1, 1997

May 1997



This is a preprint of a paper intended for publication in a journal or proceedings. Since changes may be made before publication, this preprint is made available with the understanding that it will not be cited or reproduced without the permission of the author.

#### DISCLAIMER

This document was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor the University of California nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or the University of California, and shall not be used for advertising or product endorsement purposes.

# Long term bleaching of optical glasses darkened by $\text{Co}^{60}$ ionizing radiation

G. Richard Wirtenson\* and Richard H. White

Participating Guests  
University of California  
Lawrence Livermore National Laboratory  
P.O. Box 808, L-17  
Livermore, CA 94551

## ABSTRACT

Typical camera designs include optical glass elements that may be affected by the ionizing radiation present in the natural space environment. Ordinary optical glasses darken at low ( $10^3$  rad) dose levels when exposed to ionizing radiation. This darkening decreases the sensitivity of optical sensors. Optical glass flats of FK 51, LaK 9, PK 51A, and ZK N7 were exposed to a 10.6 krad dose of ionizing radiation. Spectrophotometer traces determined the transmittance of the samples as a function of wavelength in the range 350 to 850 nm before and at various time intervals after the irradiation. These measured values were then used to evaluate the rate of recovery or "bleaching" of the exposed samples. To prevent accelerated bleaching, the samples were kept at room temperature and away from light, except during measurement. Tables of the measured data and plots of the transmissivity vs. wavelength at various times after irradiation are presented.

**Keywords:** radiation darkening, bleaching, optical glasses, space based optical systems.

## 1. INTRODUCTION

A typical camera design includes optical glass elements that may be affected by the ionizing radiation present in the natural space environment.<sup>1</sup> Ordinary optical glasses darken when exposed to ionizing radiation. This darkening decreases the sensitivity of optical sensors.

In most glasses, radiation-induced darkening fades with time. The rate of fading depends upon the material and wavelength. This effect is called bleaching if the color center is depopulated (a trapped electron or hole is thermally activated out of the color center); it is called annealing if the defect center is removed by rearrangement of the ions surrounding the defect.<sup>2</sup> Here we follow Treadaway<sup>3,4</sup> and ignore this distinction and refer to both processes as bleaching.

Exposure of the radiation darkened material to light or heat accelerates bleaching. Conversely, it is likely that the reduced temperatures and light intensities anticipated for a spacecraft optical system may slow bleaching. Data on bleaching acceleration is sparse; some is reported in references 3, 4, 5 and 6.

Studies reported here, measured the recovery of four optical glasses, three of which were designed into an optical system planned for a multi-year lifetime in orbit. The design anticipated the fact that darkening would occur at a slow rate and could be partially mitigated by the bleaching. A 10.6 krad dose was selected as representative of an average total dose over a 3 to 5 year lifetime of the optical system. Also, this dose produces a level of darkening sufficient to enable more accurate measurement of bleaching changes. These studies explored the bleaching of a variety of optical glasses over a period of one year, which is a much longer time than typically reported. This report does not include the effect of dose rates on bleaching rates which we have measured for FK 51 glass and will report elsewhere. Dose rate and total dose for these experiments and those reported in the companion paper were measured using ionization chamber dosimetry for pool dose rate and total dose in the high dose rate (10.6 krad/hr) experiments. Opti-chromic dosimetry was used for total dose measurements in the low dose rate (7 rad/hr) studies reported elsewhere.

---

\*Current address: Technical Consulting Services, Optical Thin Film Design and Consulting, P.O. Box 2347, Livermore, CA 94551, Email: wirt@compuserve.com

The absorption coefficients implied by the data collected for FK 51 do not agree with those reported by either Liepmann<sup>5</sup> or Scheller<sup>6</sup> (reporting data from Schott Glass Technologies). Our values fall between their values, being approximately half those reported by Liepman and double those given by Scheller.

## 2. THE BLEACHING STUDY GLASSES AND EXPERIMENT

Four glasses; FK 51, LaK 9, PK 51A, and ZK N7 supplied by Schott Glass Technologies, Duryea, Pennsylvania, were included in this study. These glasses are from the principal glass type designated "crown", which, generally, is harder, has a lower index of refraction, and lower dispersion than the other principal glass type: "flint." Type FK (fluoro-crown) is a phosphate or borosilicate glass with a fairly high fluorine content; its index of refraction ( $n$ ) and Abbe number ( $v$ ) are in the ranges: [ $n < 1.49$ ,  $v > 62$ ]. PK is a phosphate or borosilicate glass with a lower fluorine content [ $n \sim 1.49$  to  $1.54$ ,  $v > 62$ ]. ZK is a silicate or borosilicate glass with  $> 10\%$  by weight ZnO [ $n \sim 1.51$  to  $1.53$ ,  $v \sim 57$  to  $61$ ]. LaK is a silico-borate glass containing rare earth oxides [ $n = 1.64$ ,  $v > 50$ ].<sup>7</sup> Optical flats of each glass type,  $\sim 0.5$  cm in thickness were exposed to 10.6 krad of ionizing radiation in LLNL's Co<sup>60</sup> pool in the Standards and Calibration Laboratory.<sup>8</sup>

A pre-exposure transmittance was measured for each glass. After exposure the transmittance was measured at 8 times from 1 hour to more than one year after exposure. (Transmittance is defined as the ratio of incident intensity to transmitted intensity. It differs from unity primarily because of reflection at the air/glass interfaces, but it may include some absorption. Here we assume the absorption in the un-irradiated samples to be zero and define the *transmissivity* for the irradiated samples to be the ratio of the transmittance of the irradiated sample to the transmittance before irradiation.) A system baseline was run before and after each measurement set, to allow correction for systematic variations.

Transmittances were measured in the range 350 to 850 nm using the Hitachi U-2000 Spectrophotometer at JLE Associates, Pleasanton, CA. The data are presented in Tables I-V and Figures 1-5.

## 3. RESULTS

We report the results of these experiments in both tabular and graphical form at the end of this paper. Table I gives the transmittances as a function of wavelength for un-irradiated samples of the four glasses studied. Tables II through V give the internal transmissivities for various wavelengths ( $\lambda$ ) and for various times after irradiation. Figures 1 through 5 are plots of the data from Tables I through V.

## 4. SUMMARY

While the analysis is not presented in this paper, we determined that use of these glasses in the optical system, as originally designed, would introduce an unacceptable level of darkening even though the anticipated fluence was only of the order of 1 krad/yr during a period of low solar activity. More stable cerium dioxide, CeO<sub>2</sub>, doped glasses were substituted for the original glass types in the following way: BK 7 G18 for ZK N7 and LaK 9 G15 for LaK 9. The designation G15 for instance, is used by Schott to identify a glass with a doping level of 1.5% CeO<sub>2</sub>. FK 51 cannot be effectively stabilized by the addition of CeO<sub>2</sub>.<sup>9</sup> Therefore, if it is used in space applications, thicknesses must be minimized and the designer must be willing to accept the increasing loss in transmission attributable to radiation induced darkening.

## ACKNOWLEDGMENTS

We are indebted to Dr. John L. Emmett and JLE Associates 22721 NE 123 Circle, Brush Prairie, WA, 98606, for the use of their spectrophotometer and Dr. Norbert Wild for reviewing this paper.

## REFERENCES

1. R.H. White and G.R. Wirtenson, "Radiation Induced Darkening of the Optical Elements in the Startracker Camera", UCRL-ID-113713, Lawrence Livermore National Laboratory, March 1993.
2. Wirtenson and White, "Effects of Ionizing Radiation on Selected Optical Materials: An Overview", UCRL-ID-111453, Lawrence Livermore National Laboratory, July 30, 1992.
3. M.J. Treadaway, B.C. Passenheim and B.D. Kitterer "Transient Radiation Effects in Optical Materials",

Intelcom Rad Tech Inc. Report No. SAMSO-TR-75-174, April 29, 1975

4. M.J. Treadaway, B.C. Passenheim, and B.D. Kitterer "Luminescence and Absorption of Electron-Irradiated Common Optical Glasses, Sapphire, and Quartz", IEEE Transaction on Nuclear Science, Vol. NS-22, December 20, 1976
5. M. Liepmann, "Gamma Radiation Effects on some Optical glasses" SPIE Vol. 1761, 1992.
6. R. Scheller (Schott Glass Technologies Inc.) "Radiation Effects on Glasses" private communication to T. Simpson (JAYCOR), July 17, 1992
7. R. Scheller (Schott Glass Technologies) "Schott Glass Designations" private communication to G.R. Wirtenson, May 22, 1996
8. G.R. Wirtenson and R.H. White, "Calibration of the Standards and Calibration Laboratory's Co<sup>60</sup> Radiation Pool", UCRL-ID-112675, Lawrence Livermore National Laboratory, January, 1993.
9. W. Beier and B. Speit, Schott Glaswerke, Mainz, Germany, private communication to G.R. Wirtenson, September 12, 1994.

**Table I: Transmittances (%)  
of Un-Irradiated Glass Samples**

$\lambda$ (nm)	FK 51	LaK 9	PK 51A	ZK N7
	sample thickness			
	.494 cm	.494 cm	.506 cm	.496 cm
850	93.5	89.0	93.5	93.5
800	93.5	88.9	93.6	93.5
750	93.0	88.8	93.5	93.5
700	93.0	88.9	93.6	93.5
650	93.0	88.8	93.5	93.5
600	92.5	88.8	93.5	93.4
550	92.5	88.8	93.6	93.4
500	92.0	88.8	93.5	93.3
450	91.0	88.5	93.5	93.0
400	90.0	88.1	93.6	92.6
350	89.0	81.4	88.1	86.2

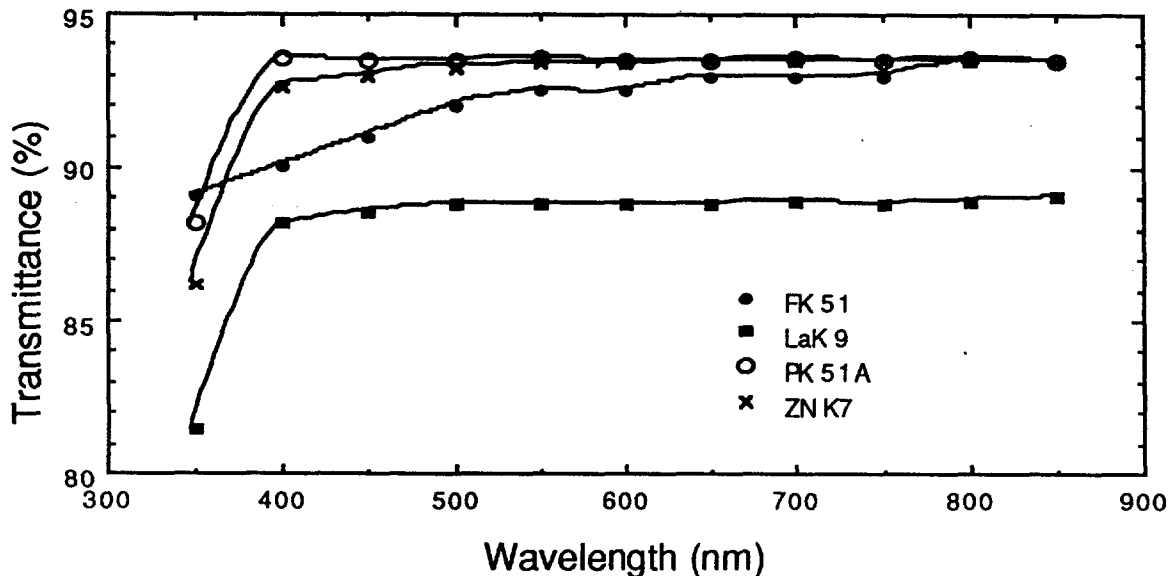


Fig 1: Transmittances Vs wavelength for the four un-irradiated glass samples.

Table II: Transmissivities (%) for FK 51 (0.494 cm)

$\lambda$ (nm)	time following irradiation (s)							
	$2.40 \times 10^3$	$6.06 \times 10^3$	$2.35 \times 10^4$	$8.75 \times 10^4$	$1.09 \times 10^5$	$1.91 \times 10^6$	$5.37 \times 10^6$	$3.04 \times 10^7$
850	99.2	99.2	99.4	99.2	99.7	100.	100.	100.
800	98.4	98.5	98.8	98.8	99.3	99.9	100.	100.
750	97.6	97.8	98.5	98.6	99.2	99.9	100.	100.
700	95.1	95.8	96.9	97.2	97.8	99.0	99.4	99.6
650	90.2	91.4	93.2	94.1	94.8	96.6	97.2	97.8
600	81.3	83.1	86.0	87.4	88.4	91.3	92.5	93.8
550	69.8	72.2	75.8	78.0	79.1	83.1	84.8	87.0
500	63.8	66.4	70.4	72.8	73.9	78.6	80.4	83.1
450	63.8	66.5	70.7	73.3	74.5	79.2	81.2	83.8
400	67.9	70.5	74.6	77.2	78.3	82.9	84.8	87.2
350	72.9	74.9	78.3	80.9	81.9	86.2	87.9	90.0

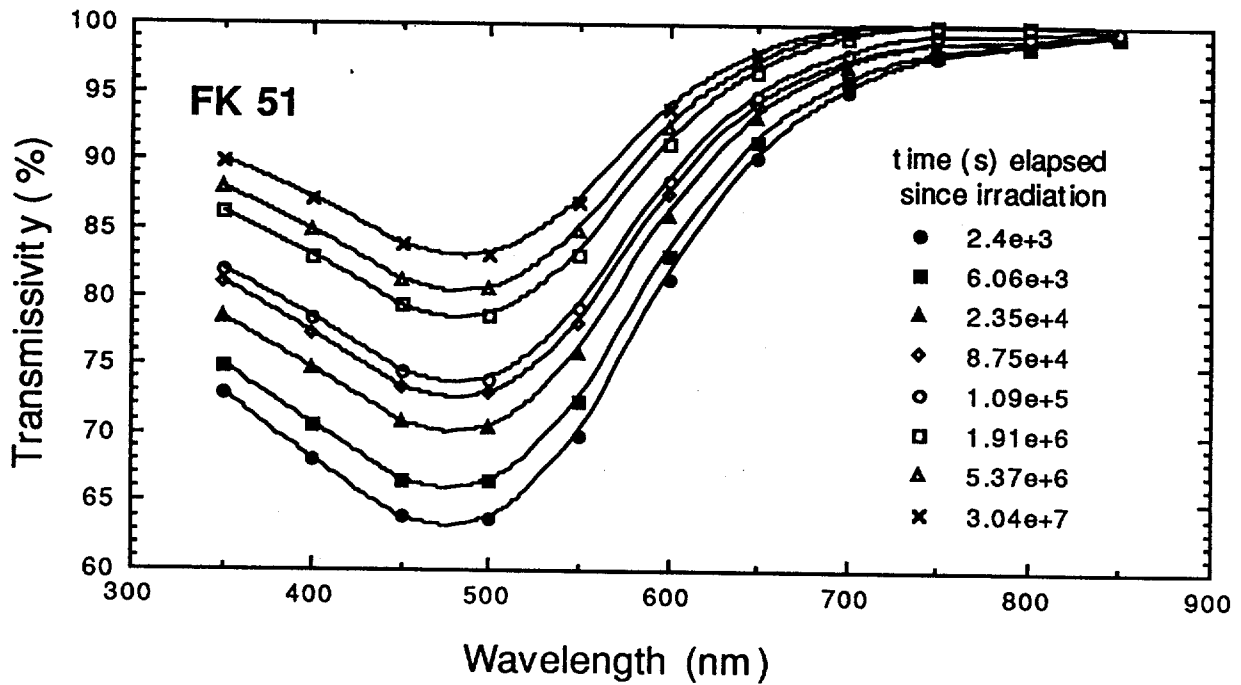
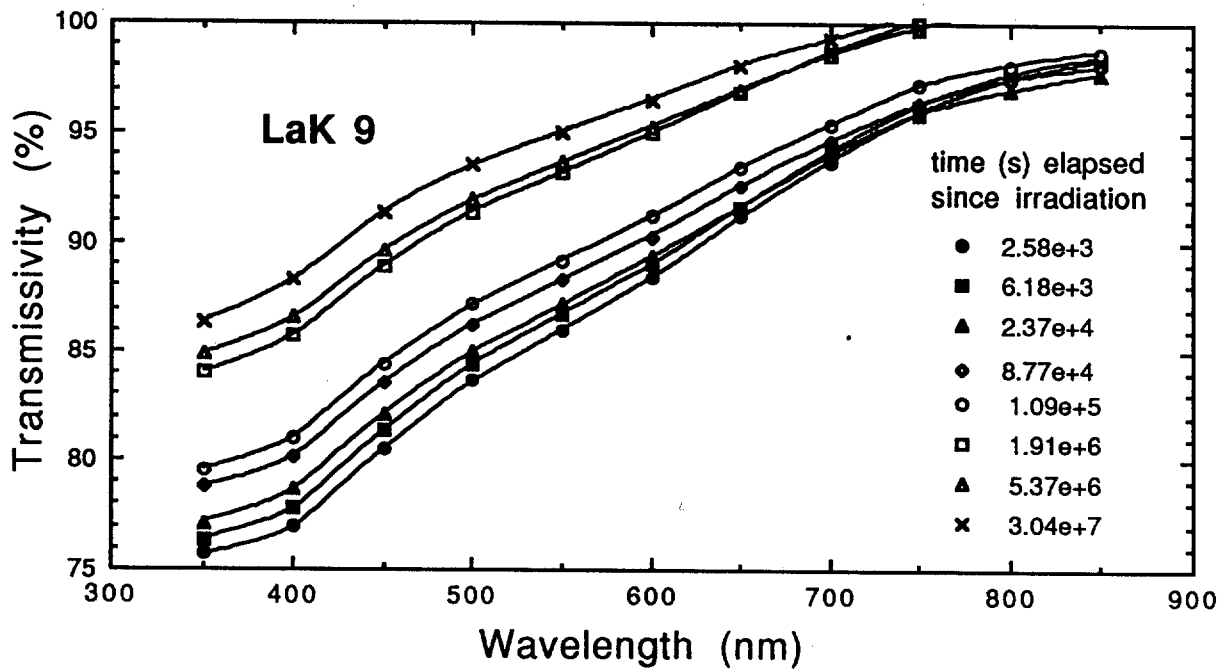


Fig. 2 Transmissivity vs. Wavelength for 0.494 cm of FK 51 at Various Times after Exposure to Radiation.

**Table III: Transmissivities (%) for LaK 9 (0.494 cm)**

$\lambda$ (nm)	time following irradiation (s)							
	$2.58 \times 10^3$	$6.18 \times 10^3$	$2.37 \times 10^4$	$8.77 \times 10^4$	$1.09 \times 10^5$	$1.91 \times 10^6$	$5.37 \times 10^6$	$3.04 \times 10^7$
850	98.3	98.4	97.6	98.0	98.7	100.	100.	100.
800	97.4	97.6	96.9	97.4	98.1	100.	100.	100.
750	95.9	96.2	95.9	96.3	97.2	100.	100.	100.
700	93.7	94.2	94.0	94.6	95.4	98.6	98.6	99.2
650	91.2	91.6	91.6	92.6	93.4	96.8	97.0	98.1
600	88.4	89.0	89.4	90.3	91.2	95.0	95.3	96.5
550	86.0	86.7	87.1	88.3	89.2	93.2	93.6	95.0
500	83.6	84.4	85.0	86.3	87.2	91.4	91.9	93.5
450	80.4	81.3	82.1	83.5	84.4	88.9	89.6	91.4
400	76.9	77.8	78.6	80.2	81.0	85.8	86.6	88.3
350	75.7	76.4	77.1	78.7	79.5	84.0	84.9	86.4



**Fig. 3 Transmissivity vs. Wavelength for 0.494 cm of LaK 9 at Various Times after Exposure to Radiation.**

Table IV: Transmissivities (%) for PK 51A (0.506 cm)

$\lambda$ (nm)	time following irradiation (s)							
	$2.94 \times 10^3$	$6.48 \times 10^3$	$2.41 \times 10^4$	$8.80 \times 10^4$	$1.09 \times 10^5$	$1.91 \times 10^6$	$5.37 \times 10^6$	$3.04 \times 10^7$
850	98.4	98.8	98.4	99.0	99.1	99.3	98.8	98.8
800	97.8	98.2	97.8	98.4	98.6	98.8	98.4	98.6
750	97.0	97.4	97.2	97.9	98.1	98.3	98.1	98.3
700	95.1	95.6	95.5	96.4	96.6	97.2	97.0	97.4
650	90.3	91.0	91.4	92.6	92.8	94.2	94.3	95.3
600	80.3	81.5	82.6	84.4	84.7	87.4	88.2	89.9
550	68.8	70.3	72.0	74.2	74.6	78.5	79.9	82.7
500	62.8	64.4	66.2	68.5	68.9	73.3	75.0	78.1
450	63.3	64.8	66.5	68.7	69.1	73.3	74.9	78.0
400	67.9	69.2	70.7	72.7	73.00	76.6	77.9	80.5
350	71.0	72.1	73.4	75.2	75.5	78.2	79.1	81.2

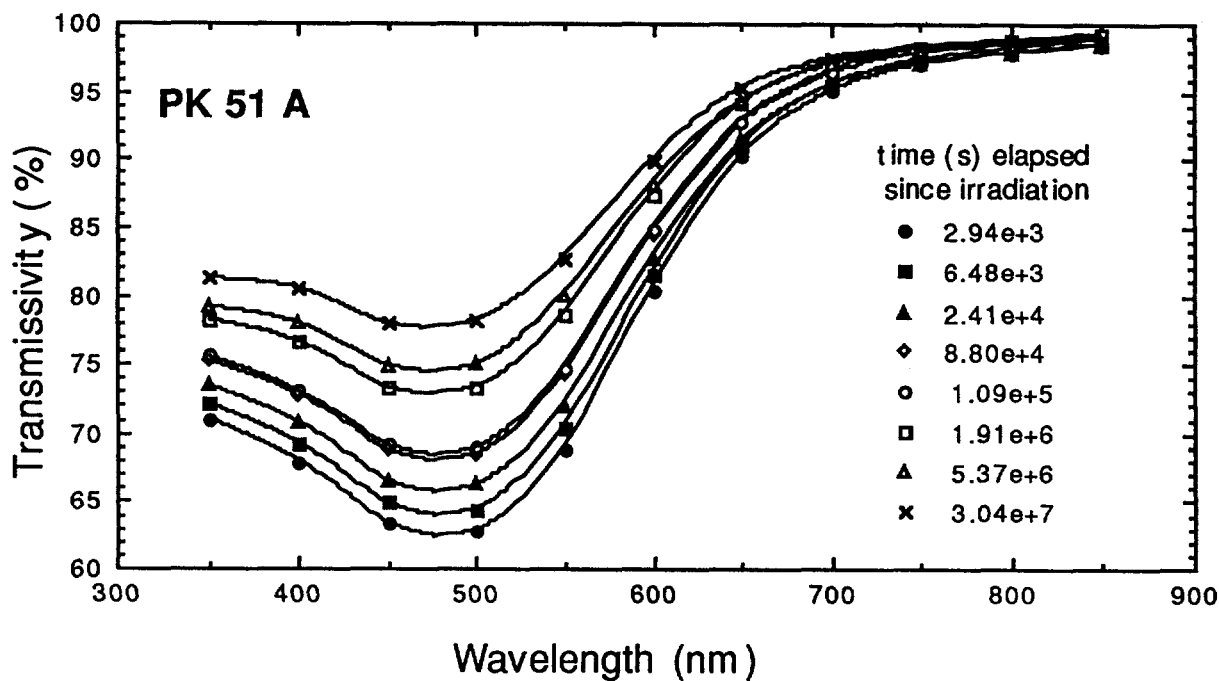


Fig. 4 Transmissivity vs. Wavelength for 0.506 cm of PK 51A at Various Times after Exposure to Radiation.

Table V: Transmissivities (%) for ZK N7 (0.496 cm)

$\lambda$ (nm)	time following irradiation (s)							
	$2.76 \times 10^3$	$6.36 \times 10^3$	$2.39 \times 10^4$	$8.78 \times 10^4$	$1.09 \times 10^5$	$1.91 \times 10^6$	$5.37 \times 10^6$	$4.13 \times 10^7$
850	99.6	98.8	99.2	99.3	99.5	99.8	100.	99.9
800	99.3	98.4	99.0	99.1	99.3	99.6	100.	99.8
750	98.8	97.9	98.6	98.7	98.9	99.3	99.8	99.5
700	97.8	97.1	97.8	98.0	98.2	98.7	99.3	99.1
650	96.4	95.7	96.5	96.8	97.1	97.7	98.3	98.4
600	94.6	94.0	95.0	95.3	95.6	96.5	97.3	97.4
550	92.4	91.9	92.99	94.2	93.7	94.8	95.7	96.1
500	89.7	89.3	90.5	91.1	91.4	92.8	93.8	94.3
450	85.0	84.8	86.3	87.2	87.6	89.3	90.4	91.3
400	74.8	75.1	77.1	78.5	79.0	81.5	83.0	84.5
350	59.8	60.5	62.9	65.2	65.6	69.1	71.2	73.7

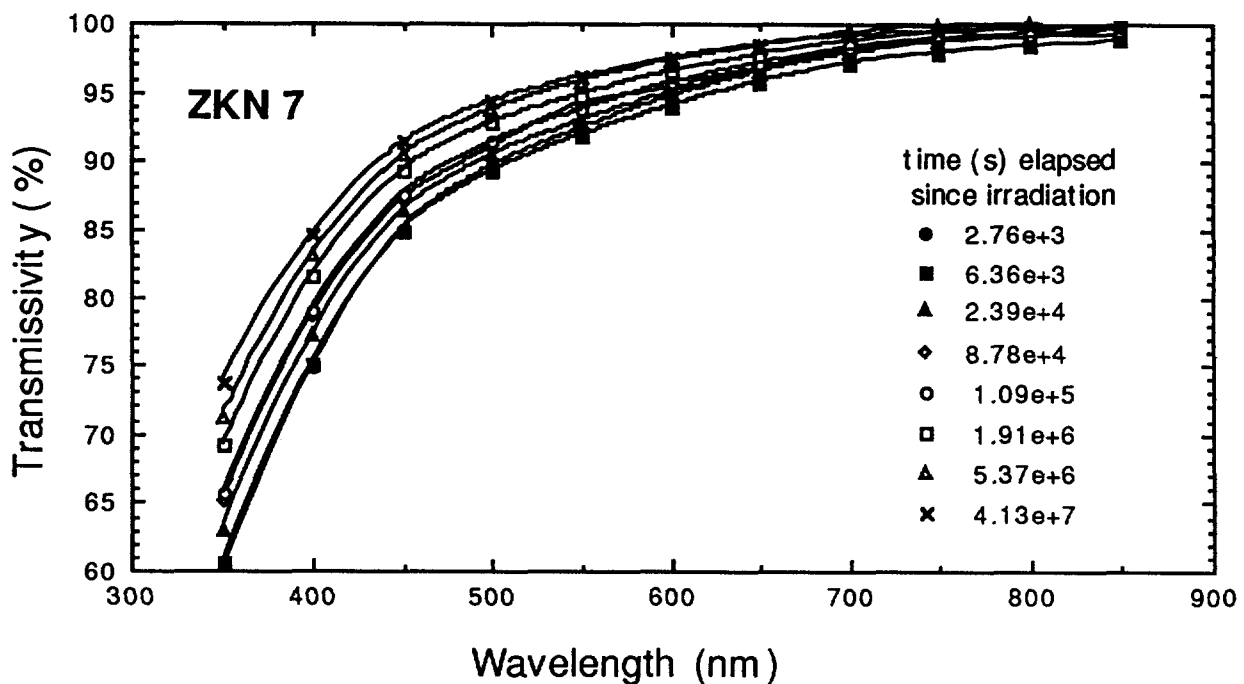


Fig. 5 Transmissivity vs. Wavelength for 0.496 cm of ZK N7 at Various Times after Exposure to Radiation.

This work was performed under the auspices of the U.S. Dept. of Energy at LLNL under contract # W-7405-Eng-48.

*Technical Information Department • Lawrence Livermore National Laboratory*  
*University of California • Livermore, California 94551*

