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FADING CHARACTERISTICS OF GAMMA INDUCED COLORATION IN HIGH DENSITY GLASS

ROBERT H. KERNOHAN AND GRACE M. McCAMMON

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FADING CHARACTERISTICS OF GAMMA INDUCED COLORATION IN HIGH DENSITY GLASS

Robert H. Kernohan, Fairchild Engine and Airplane Corporation - NEPA
Grace M. McCammon, Oak Ridge National Laboratory

SUMMARY

Specimens of a lead silicate glass manufactured by the Penberthy Instrument Company were measured for transmission characteristics. The specimens were given varying dosages from a 300 curie cobalt 60 gamma source. Transmission characteristics were measured as a function of time following irradiation. The gamma induced coloration faded and this fading could be accelerated by illuminating the specimens with an ordinary light bulb. All specimens nearly recovered their original transmission characteristics.

Introduction:

The investigation reported here originated in an auxiliary problem concerning the windows and other optical parts to be used in construction of hot cells for the testing of radioactive materials. The chief requirements for such an optical material would be optimum (1) gamma shielding ability, (2) transmission of thick sections in the visible wave length region, (3) resistance to coloration by gamma rays and (4) chemical and physical stability.

The glass for a good shielding window must contain a large weight percentage of an element of high atomic weight. The work reported in this paper is merely a preliminary investigation of an unusual characteristic found in one high density glass. Further tests and experiments have come immediately to mind on this glass as well as on others. However, this present report is of immediate practical rather than theoretical interest. Work on radiation resistant optical materials has been and is being ably carried on at Argonne National Laboratory (1) and at the University of Pittsburgh (2) in connection with the Corning Glass Company.

(1) Refer to Report ANL 4536, Geo. S. Monk, "Color of Optical Materials by High Energy Radiation."

(2) Rothermel, Sun, Silverman, "Phosphate Glass: $PbO-WO_3-P_2O_5$ System", Jour. Am. Ceramic Soc. Vol., 32, May 1949.

Glass Characteristics:

Five specimens of glass were made for use in this investigation by the Penberthy Instrument Company, 666 Adams Street, Seattle 8, Washington. The specimens were cylinders 7/8" in diameter and two inches long. The manufacturer furnished the following information:

Name: Hi-D glass.

Chemical composition by synthesis:

PbO	81.0%
SiO_2	17.3%
TiO_2	1.35%
As_2O_3	0.35%

Density: 6.4 g/cc 0.23 lb/cu. inch.

Color: Light yellow tint, colorless in thin sections.

Index of refraction: 1.96

Gamma Shielding characteristics:

Equivalent of steel and about 55% as effective as lead.

Uncoated surfaces reflect as much as 10 percent but this loss can be reduced to about $\frac{1}{2}$ percent by coating.

Experimental Details:

The original optical transmission was measured through the two inch thickness of the glass by means of a Beckman spectrophotometer, Model B. The uppermost curves labeled OT in Figures 1, 2, 3 and 4 are the original transmission curves. They agree fairly well with each other

and indicate excellent transmission for red (800 millimicrons) and yellow, fair for green, and practically no transmission around the violet end of the visible spectrum (400 millimicroms).

Specimen A was exposed in the center of the ORNL pile in a process tube for one week. Unfortunately the specimen container was dropped in transfer from the lead coffin to the hot lab and the glass cylinder fractured in two large pieces. The glass had been exposed to an integrated neutron flux of about 5×10^{17} neutrons/cm^2 and its induced activity two days after removal from the pile was about 5 R/hr. at about 6 inches. The color of the specimen was a dark reddish brown at this time. After three weeks the activity of the specimen decreased to nearly tolerance and its dark color had faded considerably. It was estimated to be about half as dark as immediately after removal from the reactor.

Specimens B, C, D and F were exposed to a 300 curie Co^{60} gamma source available in the Chemistry Division at ORNL. The center of this source yielded a dose rate of 4800 R/minute. Specimens B, C and D were irradiated in the center of the source for periods of 5, 31 and 90 minutes respectively, while F was exposed at a slight distance from the Co^{60} source in order that it might receive a total dose of 10^6 R overnight (approximately 17 hrs.). The Co^{60} source yields two monochromatic gammas of 1.17 and 1.33 Mev.

Specimens B, C, D and F were all a dull brownish black color when removed from the Co^{60} gamma source. D and F were noticeable darker than B and C, and B was lighter in color than C. It was apparent that Hi-D glass was not resistant to coloration by a gamma source. Color centers are usually readily induced in most crystals and optical materials

by ionizing radiations. Color centers can be removed quickly by the application of heat. On the other hand, it is well known that many ordinary colored glasses will fade particularly in strong sunlight. Monk at Argonne had noticed a slight fading in some glasses containing PbO.[1]

Coloration Fading:

The four specimens exposed to gamma irradiation were measured for transmission characteristics as a function of time after removal from the gamma source. Specimens B, C, and D were kept in a box in the dark after the gamma irradiation except for the brief period necessary to transfer them to the Beckman spectrophotometer. It soon appeared that these specimens no longer faded as time increased. They were then exposed to light. Specimen F was exposed to a source of light immediately after gamma irradiation and was not kept in the dark for any appreciable time.

TABLE I

SPECIMEN	GAMMA EXPOSURE	HISTORY AFTER GAMMA EXPOSURE
B	24,000 R	Kept in dark 31 days. Then exposed to light.
C	150,000 R	Kept in dark 19 days. Then exposed to light.
D	430,000 R	Kept in dark 13 days. Then exposed to light.
F	1,000,000 R	Exposed to light continuously.

Results of the work are shown best by the curves of Figure 1, 2, 3 and 4. The time indicated on the curves indicates the elapsed time

(1) Op. cit.

from the end of the gamma irradiation to the time of reading in the spectrophotometer.

The source of light used to illuminate the specimen was merely a 150 watt tungsten light bulb placed one foot away from the optical face of the specimen. This sort of illumination would be very easy to duplicate in a hot cell. The experiment was performed in a rather warm room in addition to which there was some heating from the light bulb. A mercury thermometer placed next to the specimens usually registered about 90°F (32°) with a maximum of 100°F (38°C).

In Figure 1, 2 and 3 the curves labeled X include an illumination exposure of 21 hours, and the curves marked Y include an illumination exposure of 160 hours.

After exposure it was difficult to detect with the naked eye any difference between any of the four specimens and an original specimen.

Transmission curves were taken at intermediate times in order to have some indication of recovery rates. Graphs of the transmission at 500 mμ and 600 mμ versus time after gamma irradiation or recovery time are shown in Figures 5 and 6. These curves indicate a logarithmic sort of recovery during the initial stages. A plot of transmission at 500 mμ versus log of recovery time is shown in Figure 7.

It is apparent from a study of the curves that the illumination from an ordinary light bulb can greatly accelerate the fading of the gamma induced coloration in this particular glass.

In order to obtain some indication of the peak absorption of the induced color centers two graphs were made of the change in absorbance as a function of wave length. These are shown in Figure 8 and Figure 9.

Absorbance is calculated as follows:

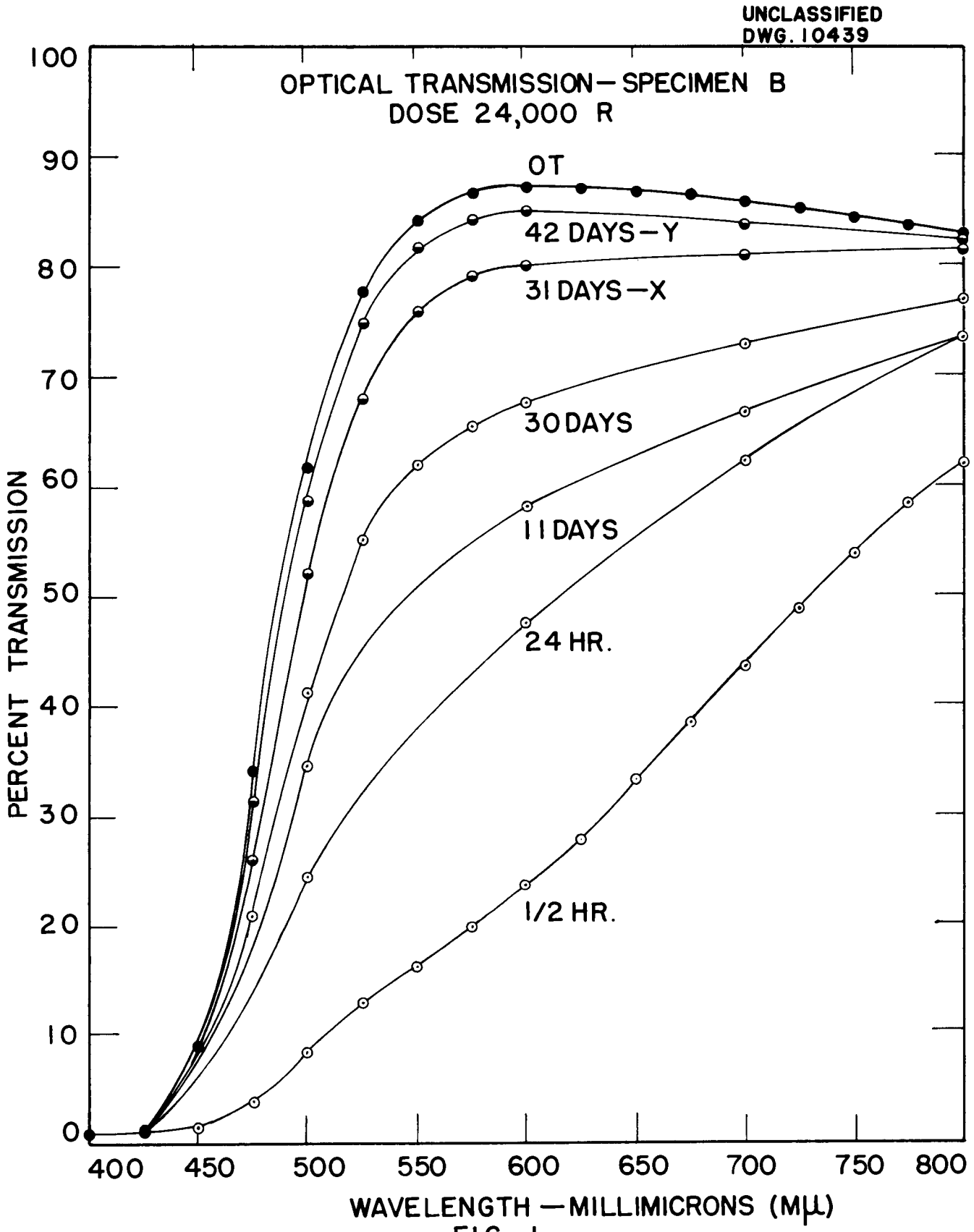
$$\text{Absorbance} = \log_{10} \frac{100}{\text{percent transmission}}$$

Points on the curves were found by substracting the calculated absorbance at any one time from the original absorbance. Unfortunately too few readings were taken in order to determine with any exactness the peaks of the change in absorbance curves. The curves show that the absorption is greatest in the range from 550 mμ to 425 mμ and that as the color centers fade the maximum absorbance gradually shifts from the region above 500 mμ to the region below.

Conclusions:

The Hi-D glass manufactured by the Penberthy Instrument Company would make a suitable material for windows exposed to gamma irradiation, not so much because of its resistance to coloration, but because of the ease of recovery from, or fading of the induced coloration.

Other questions immediately pose themselves and would be suitable for further investigation. Among them are: (1) Would successive cycling of gamma irradiation and illumination cause any deterioration in transmission properties? (2) Could the fading effect be accelerated by varying the intensity and/or the spectrum distribution of the source of illumination? (3) Could a balance of gamma irradiation and illumination be arranged so that no coloration would occur? (4) Would an increase in temperature of the specimen enhance fading?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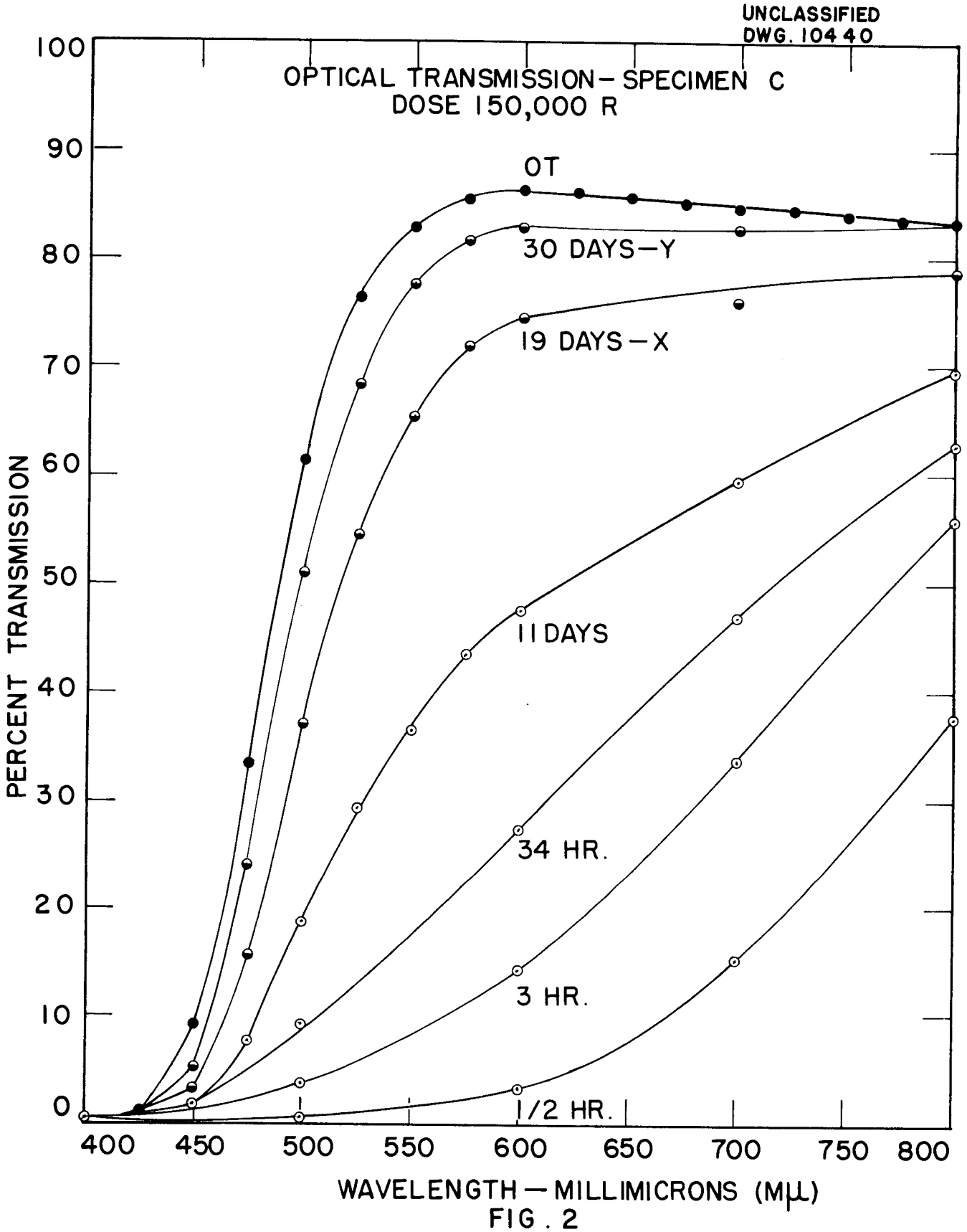
DWG. 10439
OPTICAL TRANSMISSION—SPECIMEN B
DOSE 24,000 R
OT
42 DAYS—Y
31 DAYS—X
30 DAYS
11 DAYS
24 HR.
1/2 HR.
PERCENT TRANSMISSION
0
10
20
30
40
50
60
70
80
90
100
400
450
500
550
600
650
700
750
800
WAVELENGTH —MILLIMICRONS (Mμ)

FIG. 1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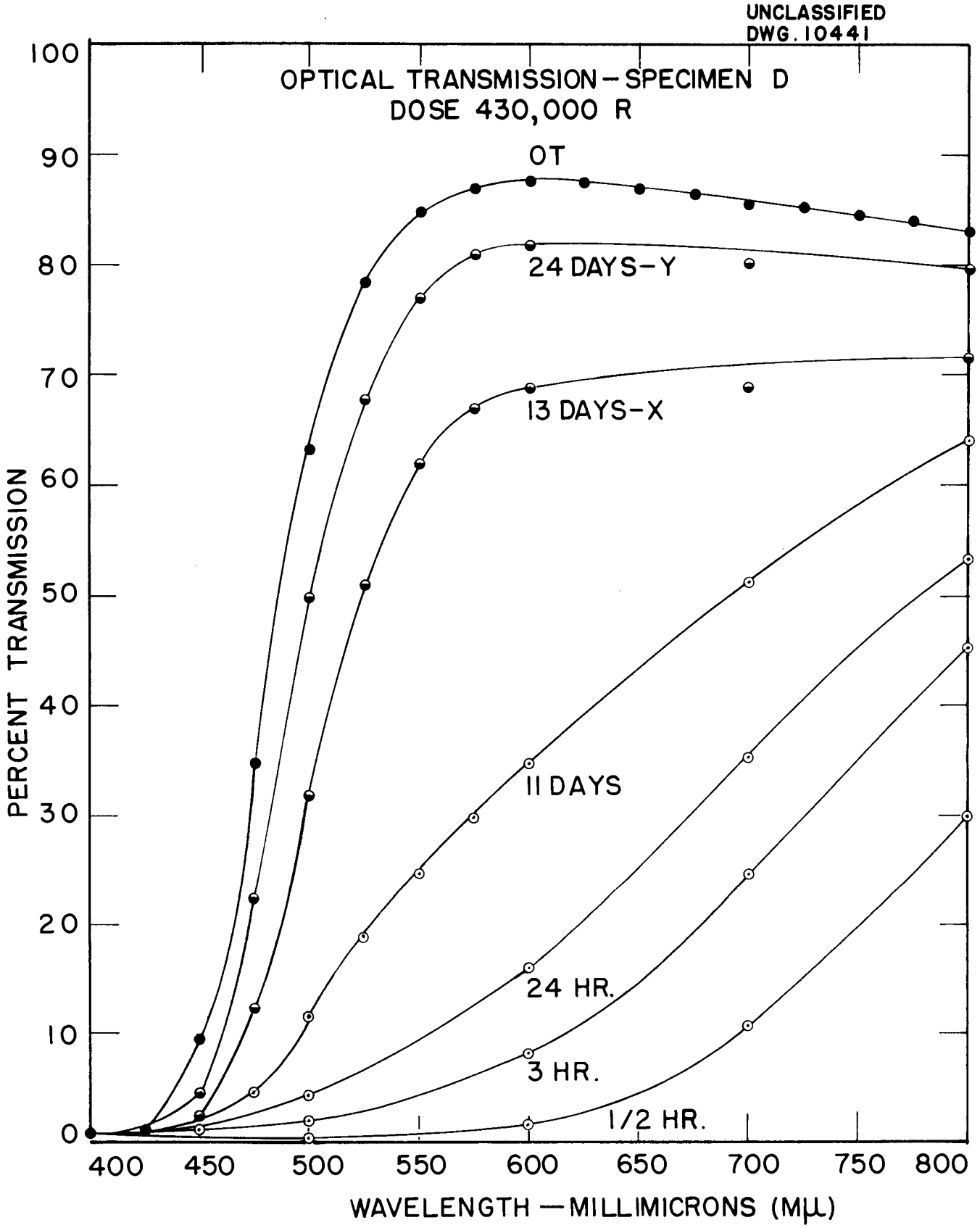
DWG. 10440
OPTICAL TRANSMISSION – SPECIMEN C
DOSE 150,000 R
OT
30 DAYS–Y
19 DAYS – X
11 DAYS
34 HR.
3 HR.
1/2 HR.
PERCENT TRANSMISSION
100
90
80
70
60
50
40
30
20
10
0
400
450
500
550
600
650
700
750
800
WAVELENGTH – MILLIMICRONS (Mμ)

FIG. 2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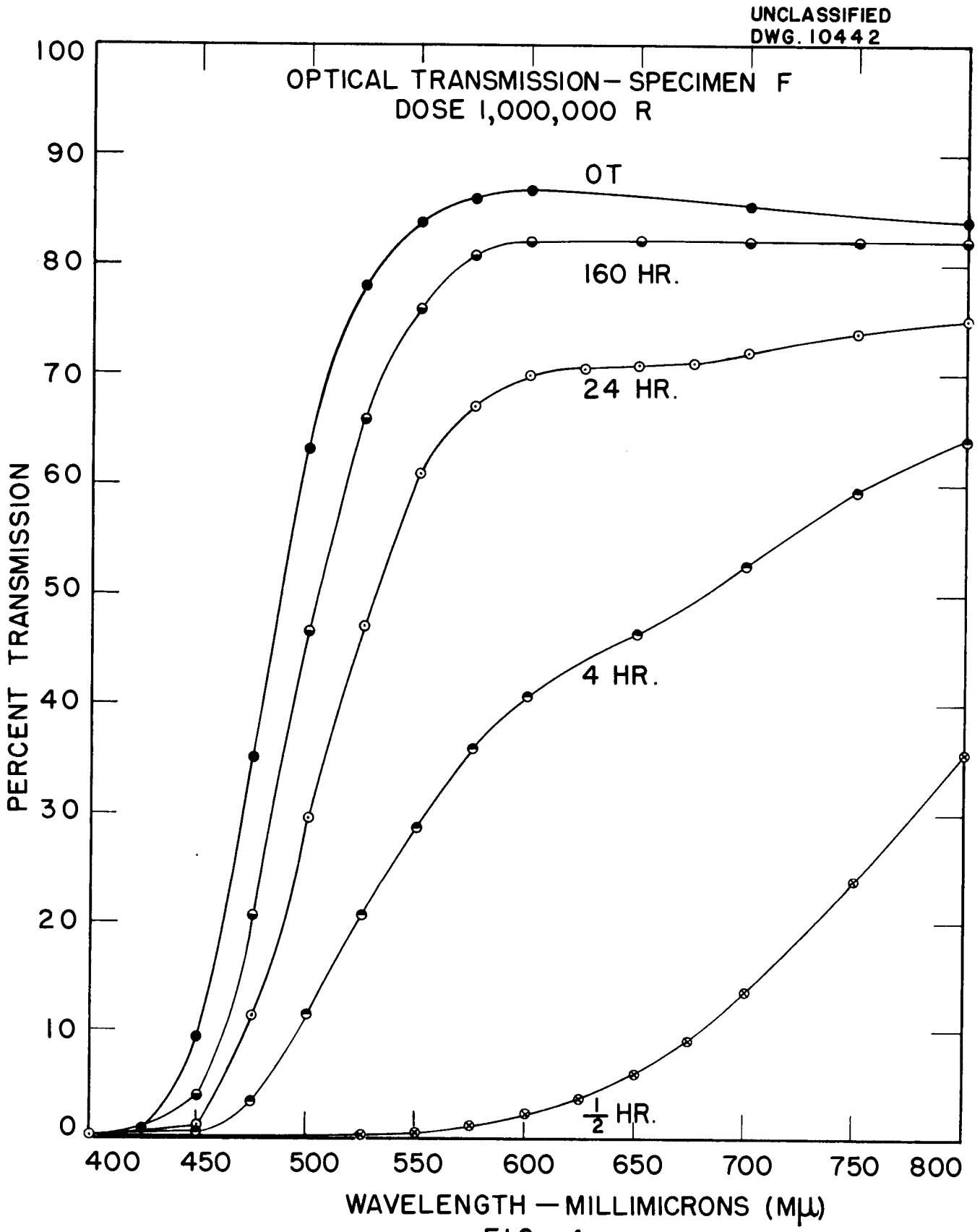
DWG. 10441
OPTICAL TRANSMISSION – SPECIMEN D
DOSE 430,000 R
OT
24 DAYS–Y
13 DAYS–X
11 DAYS
24 HR.
3 HR.
1/2 HR.
PERCENT TRANSMISSION
0
10
20
30
40
50
60
70
80
90
100
400
450
500
550
600
650
700
750
800
WAVELENGTH — MILLIMICRONS (Mμ)

FIG. 3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DWG. 10442
OPTICAL TRANSMISSION – SPECIMEN F
DOSE 1,000,000 R
OT
160 HR.
24 HR.
4 HR.
½ HR.
PERCENT TRANSMISSION
100
90
80
70
60
50
40
30
20
10
0
400
450
500
550
600
650
700
750
800
WAVELENGTH — MILLIMICRONS (Mμ)

FIG. 4

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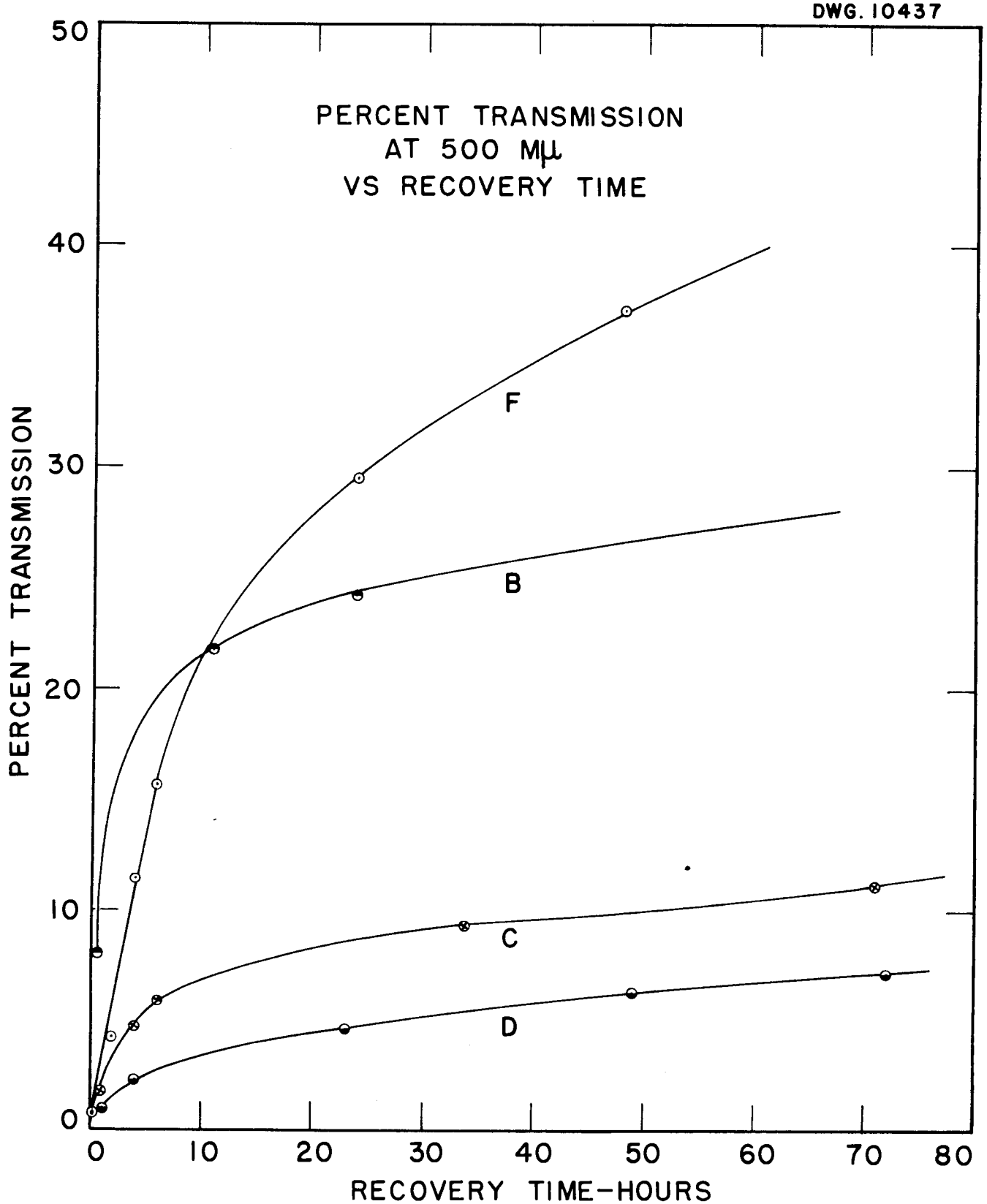
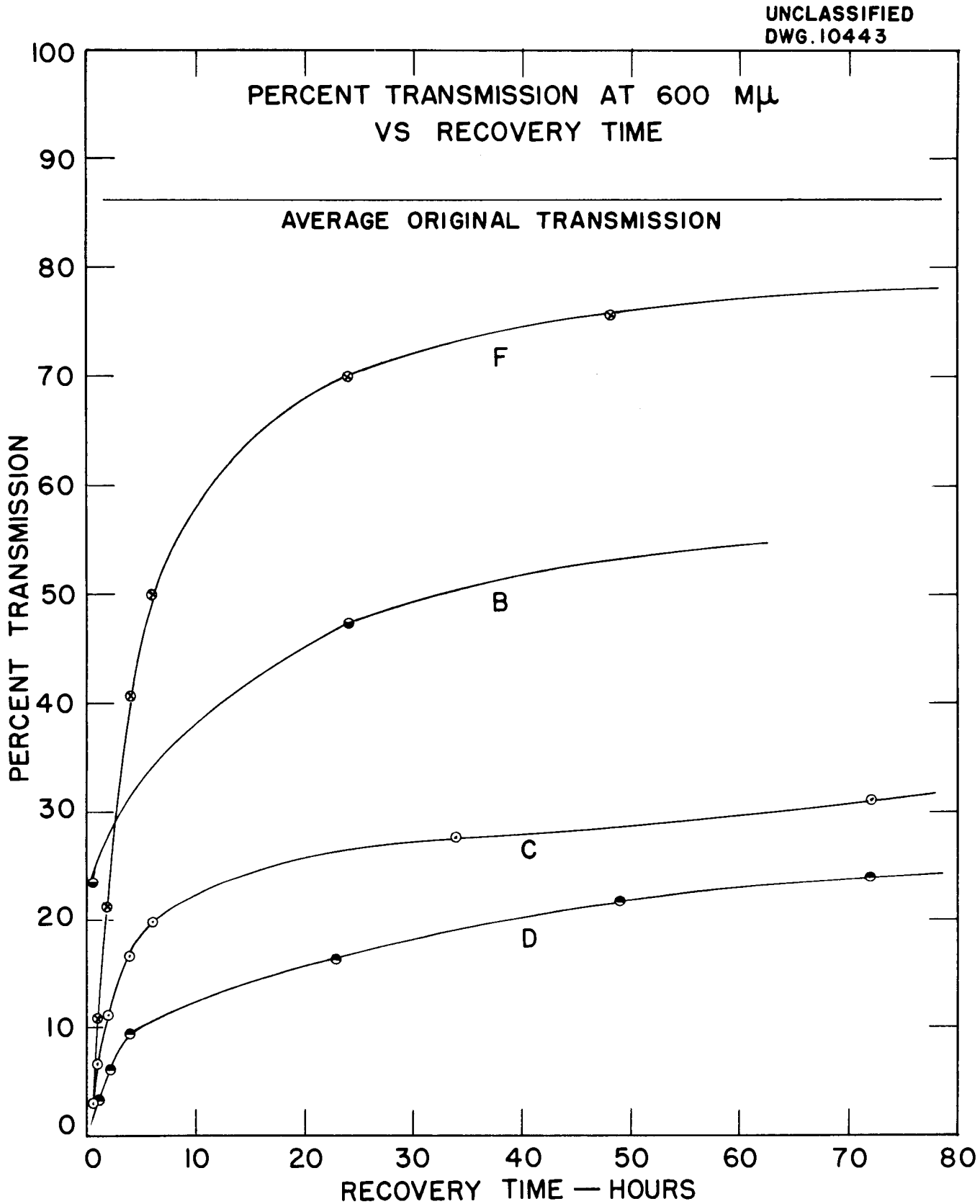


FIG. 5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PERCENT TRANSMISSION AT 600 Mμ
VS RECOVERY TIME
AVERAGE ORIGINAL TRANSMISSION
F
B
C
D
PERCENT TRANSMISSION
RECOVERY TIME — HOURS
0
10
20
30
40
50
60
70
80
90
100

FIG. 6

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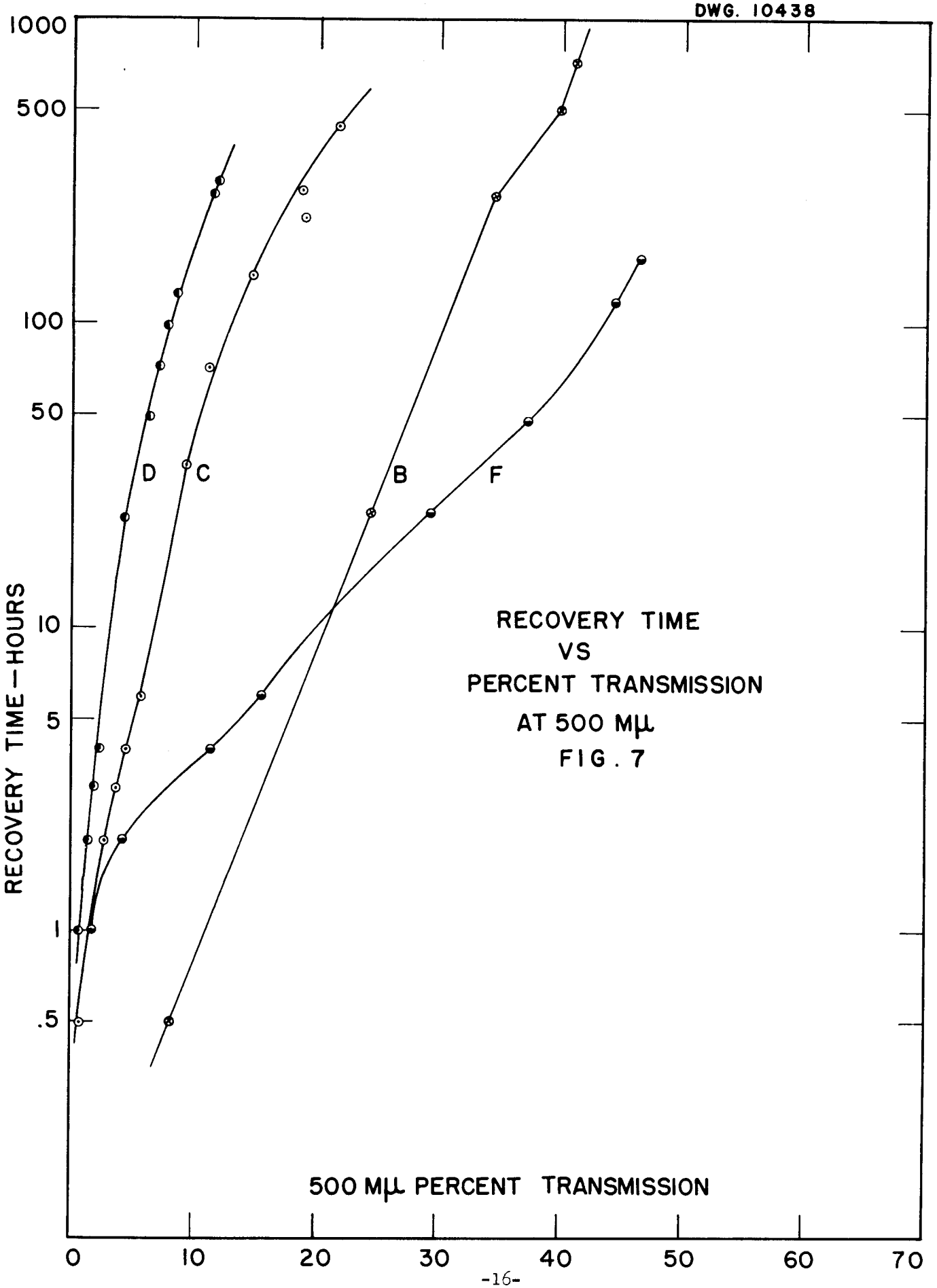

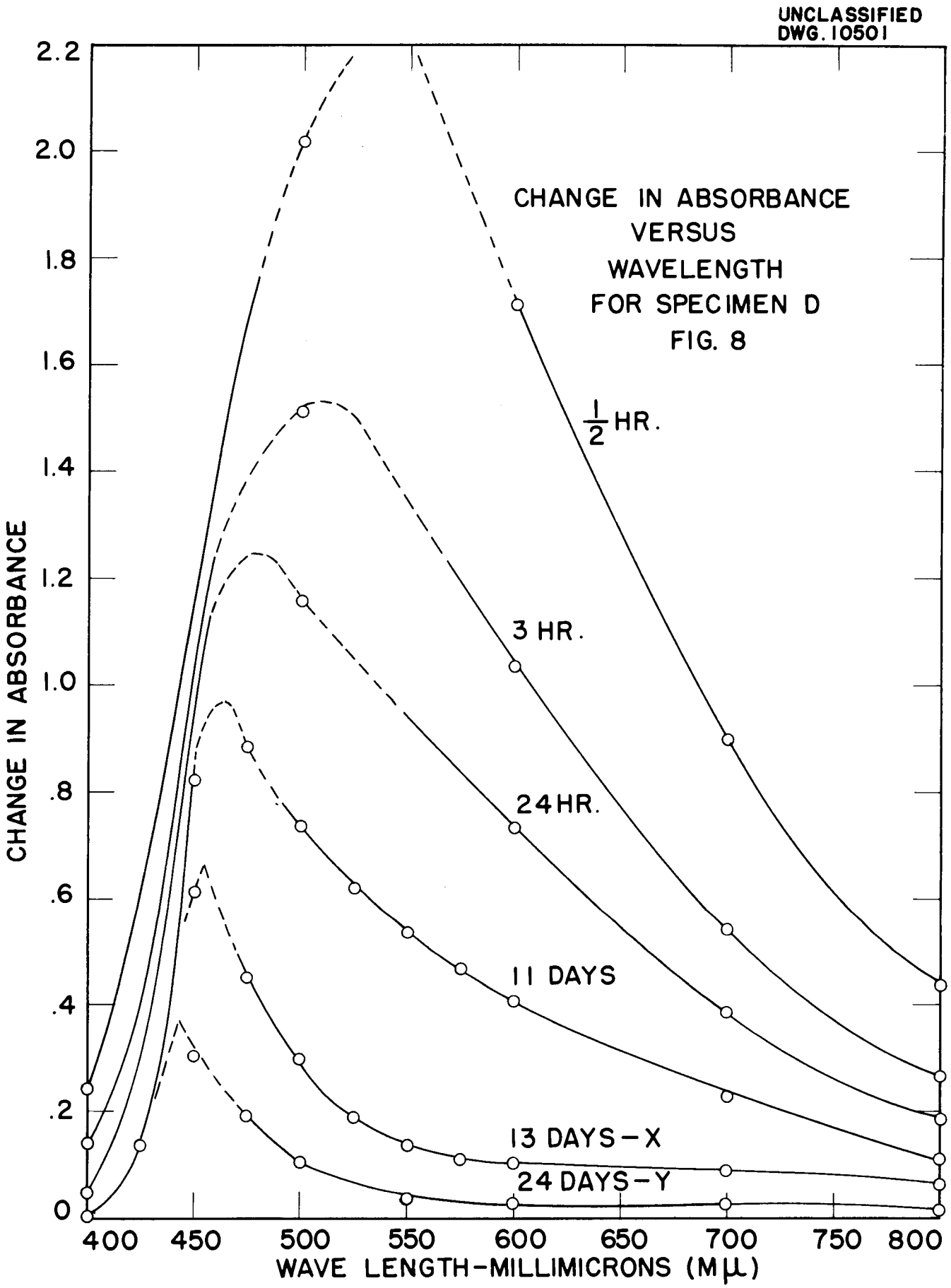
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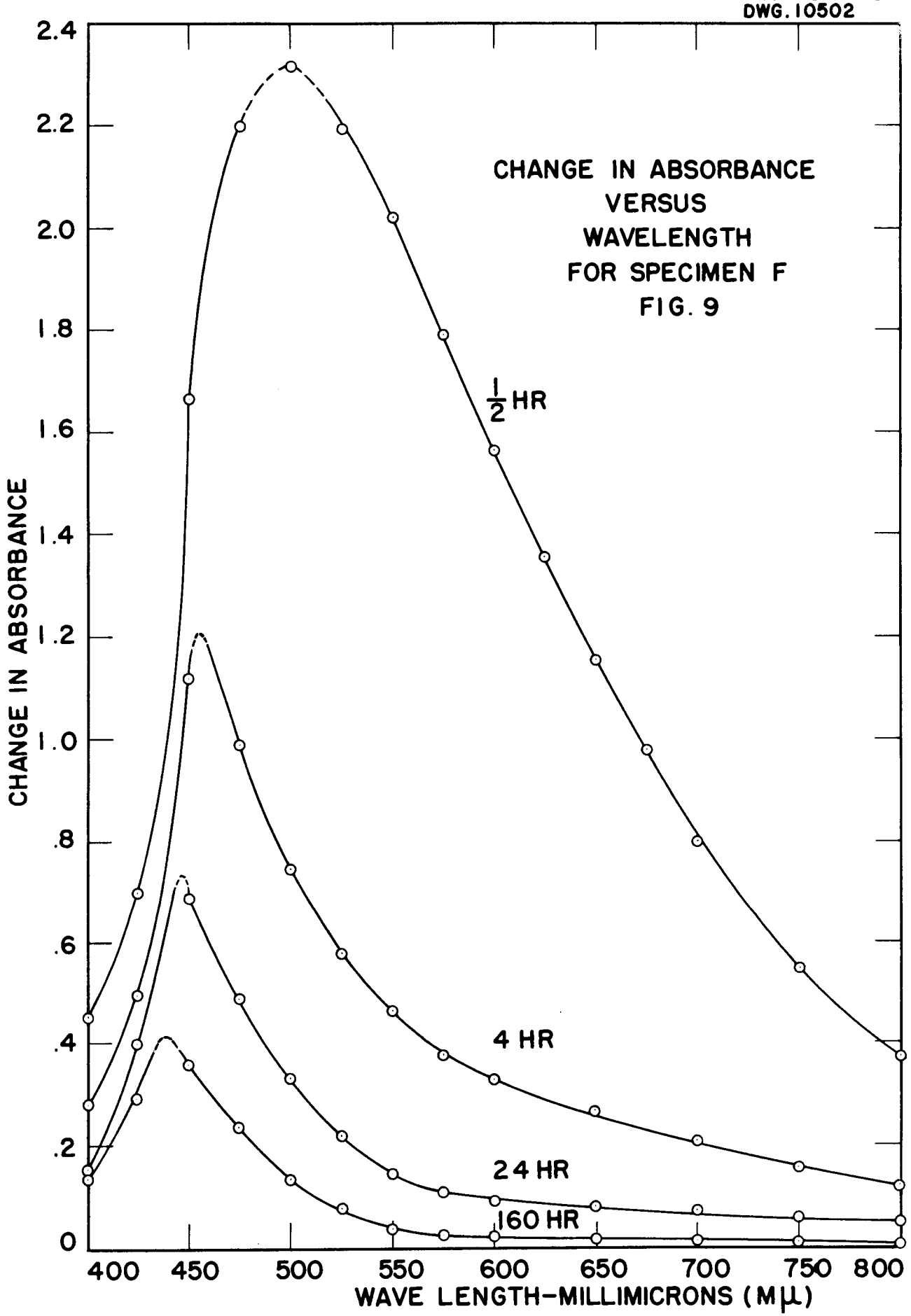
CHANGE IN ABSORBANCE
VERSUS
WAVELENGTH
FOR SPECIMEN D
FIG. 8
1/2 HR.
3 HR.
24 HR.
11 DAYS
13 DAYS-X
24 DAYS-Y
CHANGE IN ABSORBANCE
2.2
2.0
1.8
1.6
1.4
1.2
1.0
.8
.6
.4
.2
0
400
450
500
550
600
650
700
750
800
WAVE LENGTH-MILLIMICRONS (Mμ)

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