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STABILIZATION OF POLYMETHYL ALPHA-CHLOROACRYLATE PLASTIC SHEET

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WRIGHT AIR DEVELOPMENT CENTER
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Carpenter Litho & Prtg. Co., Springfield, O.
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FOREWORD

This report was prepared by the Central Research Laboratory of General Aniline and Film Corporation, Easton, Pa. under USAF Contract No. AF 33(600)-23883. The contract was initiated under Project No. 7340, "Rubber, Plastic and Composite Materials", Task No. 73403, "Transparent Materials", formerly RDO No. 616-12, "Transparent Materials", and was administered under the direction of the Materials Laboratory, Directorate of Research, Wright Air Development Center, with Lts. R. C. Smith and R. M. Mandel acting as project engineers.

This report covers work conducted from May 1953 to May 1954.

ABSTRACT

When this contract was initiated, it already had been shown that polymethyl α-chloroacrylate plastic sheet had outstanding physical properties - high heat distortion temperature, high tensile and flexural strengths, excellent craze resistance, low notch sensitivity, unique self-extinguishing property in burning tests, and complete formability - which made it particularly suited for use as an aircraft glazing material especially if its heat and light stability could be improved.

In order to obtain this plastic as a completely acceptable aircraft glazing material, Wright Air Development Center awarded this contract for the improvement of the heat and light stability of polymethyl α-chloroacrylate plastic sheets.

As a result of the work completed under this contract, polymethyl α-chloroacrylate plastic sheets possessing outstanding heat and light stability were prepared and were submitted to Wright Air Development Center.

This report summarizes the research and development work involved in obtaining these heat and light stabilized polymethyl α-chloroacrylate plastic sheets.

The detailed outstanding properties of polymethyl α-chloroacrylate (PMACA) established by testing programs on previous samples of polymethyl α-chloroacrylate plastic sheets are summarized below:

1. PMACA possesses a heat distortion temperature of 130-140°C (266-284°F).

2. PMACA possesses a tensile strength of 17,000 psi at room temperature and even at 110°C (230°F) it possesses a tensile strength of 8,000 psi.

3. PMACA possesses a flexural strength of 23,000 psi at room temperature.

4. Even though PMACA is quite hard (Barcol Hardness of 65), it still exhibits excellent strength properties at room temperature after notching; for its notched tensile strength is 12,000 psi and its notched flexural strength is 13,000 psi.

5. PMACA exhibits good resistance to solvent and stress crazing. PMACA samples withstood 11,000 psi flexural stress for 23 hours before crazing and 5,000 hours before failing in a long time flexural test. They also withstood 9,000 psi stress under toluene, 8,000 psi stress under isopropyl alcohol, and 4,500 psi stress under methyl ethyl ketone, for over 300 seconds without crazing.

6. PMACA is completely formable. At 177°C (350°F) a 6 in. diameter hemispherical dome was blown from 1/4 in. thick sheet with 18 psi air pressure.

7. PMACA is rapidly self-extinguishing. When tested for flammability by Method 2021 of Federal Specification L-P-406a, the flame extinguished immediately after removal of the external burner flame.

Stabilization of PMACA plastic sheets against heat and light is believed to overcome the principal deterrent to the use of PMACA, with its superior properties, as an aircraft glazing material.

PUBLICATION REVIEW

This report has been reviewed and is approved.

FOR THE COMMANDER:

M. R. Whitmore
Technical Director
Materials Laboratory
Directorate of Research

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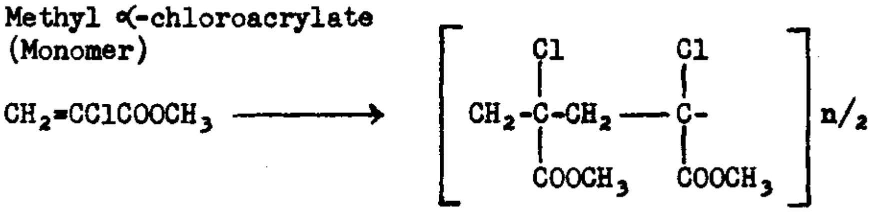
INTRODUCTION

Methyl α-chloroacrylate is a mobile, colorless, organic liquid which polymerizes under the influence of light, heat or catalysts to a transparent, hard, tough polymer (1, 2, 3, 4).

General Aniline and Film Corp. has adopted the trade-mark "Gafite" for polymers of methyl α-chloroacrylate produced by GAF.

Methyl α-chloroacrylate
(Monomer)



$$CH_2{=}CClCOOCH_3 \longrightarrow \left[CH_2-\overset{Cl}{\underset{COOCH_3}{C}}-CH_2-\overset{Cl}{\underset{COOCH_3}{C}}- \right]_{n/2}$$

Polymethyl
α-chloroacrylate
(Polymer)

(n) is an integer representing the number of monomer units in the polymer chain.

Methyl α-chloroacrylate is a powerful lachrymator and skin vesicant which makes its preparation and handling an operation requiring extreme care. It also reacts readily with oxygen. The reaction of the monomer with oxygen imparts a yellow color to polymer prepared from such monomer. In order to obtain colorless polymer, it is necessary to exclude air from contact with monomer. Despite the problems of handling the monomer, the Central Research Laboratory of GAF had investigated this polymer and learned much about its properties prior to April 1949 when the interest of the U. S. Air Force in the use of "Gafite" as a transparent material for aircraft enclosures resulted in the award to GAF of Air Force Contract AF 33(038)-2370 whose objective was the development of an aircraft glazing with a heat distortion temperature of at least 250°F (121°C).

The knowledge about the properties of polymethyl α-chloroacrylate made it a likely material to be investigated as a possible aircraft glazing material. After a year of research on improved methods of preparation of methyl α-chloroacrylate, and on the casting of this monomer, and its polymerization in glass casting cells, plastic sheet samples of the polymer meeting the objective of the contract were supplied to Wright Air Development Center (5).

After expiration of the first contract, GAF continued its own investigations on methyl α-chloroacrylate and made several improvements in the processes for production of methyl α-chloroacrylate polymer. The Materials Laboratory of Wright Air Development Center, after completing examination of the first contract samples of "Gafite", ordered an additional quantity for further testing under Contract AF 33(038)-15528. These samples were supplied by GAF using its improved processes. The National Bureau of Standards published the results of extensive tests on these samples, which tests were made with funds supplied by the Air Force (6).

At the third conference on transparent materials and aircraft enclosures, (1951) data on the physical properties of polymers of methyl α-chloroacrylate were presented by the Air Force and the National Bureau of Standards, by GAF and by Imperial Chemical Industries (7).

The data presented at that time established that the methyl α-chloroacrylate polymer possessed several properties favoring its use for military aircraft glazing. These properties included its high heat distortion temperature and its excellent resistance to crazing.

The polymer available at that time possessed a serious drawback in that surface distortion appeared on methyl α-chloroacrylate polymer sheets when they were heated at the forming temperature (320°F; 160°C) prior to shaping. GAF overcame this difficulty through its own research and for testing purposes supplied the Aircraft Industries Association with sheets of "Gafite" which did not develop surface distortion on heating (8).

Later, GAF produced sheets of the nondistorting "Gafite" 36 in. x 60 in. x 1/4 in. in size, cast to thickness tolerances, for the Air Force under Contract AF 33(038)-24249.

At the fourth meeting on transparent materials for aircraft enclosures (1952), GAF presented data obtained by the Aircraft Research and Testing Committee, W-38 subcommittee, of the Aircraft Industries Association on the nondistorting "Gafite", which showed that the nondistorting "Gafite" possessed physical properties equal to or slightly superior to the physical properties of earlier "Gafite" samples. Nondistorting "Gafite" had been obtained with no sacrifice in the physical properties of the methyl α-chloroacrylate polymer. Tests conducted by the Air Force on nondistorting "Gafite" indicated that it was possible to form half-scale canopies from "Gafite" (9).

However, studies on the thermal and ultraviolet degradation of polymethyl α-chloroacrylate made at Wright Air Development Center indicated that it would be desirable to improve the color stability of "Gafite" on weathering and to increase its heat stability (10, 11).

The absence of color is important for good visibility through the glazing, especially during the twilight hours. Increased heat stability is important in the forming of the glazing to prevent bubbles arising in the plastic by decomposition occurring during the heating required for forming. Air Force Contract AF 33(600)-23883 for research on improving the heat and light stability of methyl α-chloroacrylate polymer was awarded to GAF in May 1953. This report describes the work accomplished under this contract.

Objective

The objective of this project was the development of methyl α-chloroacrylate polymer of improved light and heat stability without sacrifice of the desirable physical properties obtained on earlier polymer samples. Specifically the polymer was to possess the following properties.

1. Specific gravity shall not exceed 1.50.
2. Rate of burning shall not exceed 0.75 in./min for a 1/2 in. width of 1/4 in. material.
3. Material must be formable into hemispheric shapes with an outside diameter of 10 in. and a draw of at least 4 1/2 in.

4. Flexural deformation temperature shall be not less than 130°C (266°F).
5. Index of refraction shall not exceed 1.52.
6. Requirements which vary with temperature:

Temperature	Tensile Strength	Elongation	Flexural Strength
23°C	16,000 psi	3%	22,000 psi
50°C	13,000 psi	4%	19,000 psi
70°C	11,000 psi	5%	17,000 psi

7. The material shall show no signs of thermal instability such as bubbling, pimpling, discolorations, etc., after exposure to 375° ± 5°F for 45 minutes.
8. Original haze, haze after accelerated weathering (L-P-406a, Method No. 6021), and haze after six months exposure in southern Florida shall not exceed 4.0%.
9. Original luminous transmittance determined by use of a pivotable sphere meter shall be not less than 91%, luminous transmittance after accelerated weathering (L-P-406a, Method No. 6021) shall be not less than 90%, and luminous transmittance after six months exposure in southern Florida shall be not less than 89%.
10. There shall be no appreciable development of yellow color in the material after six months outdoor exposure in southern Florida. This requirement is defined as follows:

Luminous transmittance determined by use of a pivotable sphere (Hunter hazemeter incorporating a No. 5543 Corning filter (thickness = 4.04 mm) shall be not less than 80% after six months exposure in southern Florida.

Research Program

Studies made at GAF prior to the receipt of this Air Force Contract led us to believe that there were definite possibilities for the achievement of the objectives of the contract. Early work done under the contract confirmed these beliefs and established that methyl α-chloroacrylate polymer itself was not unstable to light but rather that yellowing on light exposure was most likely due to small amounts of impurities present in the polymer. Because of this belief in the stability of pure methyl α-chloroacrylate polymer, research was directed toward obtaining as pure a polymer as possible. In addition, work was concentrated on adding stabilizers with the hope that they would overcome the light and heat instability due to impurities present in the polymer. In order to avoid undesirable effects on the physical properties of the polymer, the work on stabilizers was confined to the addition of those stabilizers which would be effective in very small concentrations (0.1%). For the improvement of light stability three lines of research were undertaken. These included elimination of residual monomer from the polymer, removal of impurities from the monomer, and addition of stabilizers to the polymer. For improvement of the heat stability, investigations centered on the following approaches: monomer of high purity was to be prepared and polymerized in such a manner that oxygen and other compounds known to have deleterious effects on the heat stability of the polymer were to be avoided. Heat stabilizers such as epoxy resins were to be incorporated in the polymer. The results obtained from this research program are discussed in the following report.

SECTION I. EXAMINATION OF LIGHT AND HEAT STABILITIES OF EARLY "GAFITE" SAMPLES

One and one-half years ago, around 400 samples of "Gafite" illustrating many different modifications of preparation and polymerization were placed on the roof of the Central Research Laboratory of General Aniline and Film Corp. at Easton, Pa. for exposure to the weather. A simplified optical measurement which served as an index of the depth of yellow color present in each sample was made on each sample prior to its being placed on the roof. When these samples were removed, they were measured again to determine the change in yellowing index on roof exposure. Another set of samples stored in the dark indoors in file drawers was measured similarly prior to and after 1.5 years of dark storage. The data obtained are presented in this section. The conditions of preparation of the most color-stable samples following light exposure were reviewed so that possible leads for research on light stabilization of "Gafite" could be uncovered.

In addition, the results of heat stability tests run in the past on a large number of "Gafite" samples are reported. These results indicated that the process of contacting methyl ∝-chloroacrylate monomer with phosphorus pentoxide (a process developed during noncontractual research by GAF on "Gafite") was a likely method to be investigated further in research and development work on the heat stabilization of "Gafite" during the period of this contract.

1.1 Light Stability of "Gafite" Samples

Since GAF had a large number of "Gafite" samples undergoing roof exposure, it was deemed advisable to assemble whatever information could be obtained from these samples and to examine these data for possible leads to investigate further during this contract. Several leads for research on the light stabilization of "Gafite" were already known, but it was felt that these samples should be reviewed at the start of this contract to make sure that no leads would be overlooked. The data obtained are presented in tables and charts in this section. The review of these data in relation to the method of preparation of each "Gafite" sample is presented in a later section.

1.2 Method of Determining Yellowing Index

An abridged colorimetric method for measuring degree of yellowness in polymethyl ∝-chloroacrylate was employed since work at GAF had indicated that data of sufficient accuracy and precision could be obtained more quickly and easily by the abridged method than by using the I.C.I. method (Internation Commission on Illumination--Standard Colorimetric Coordinate System).

This abridged method employed a light source followed by a lens, ground glass diffuser, a diaphragm, a Corning 5900 filter, the sample being measured, a Century No. 12 filter, and a blue sensitive phototube connected to a Photovolt Corp. Model No. 500 Meter as sketched in Figure 1.

With no sample, but only air space, the Photovolt was set at zero density. The sample was then placed in front of the Photovolt head and the optical density change was read. This density figure multiplied by 100 is referred to in this report as the yellowing index.

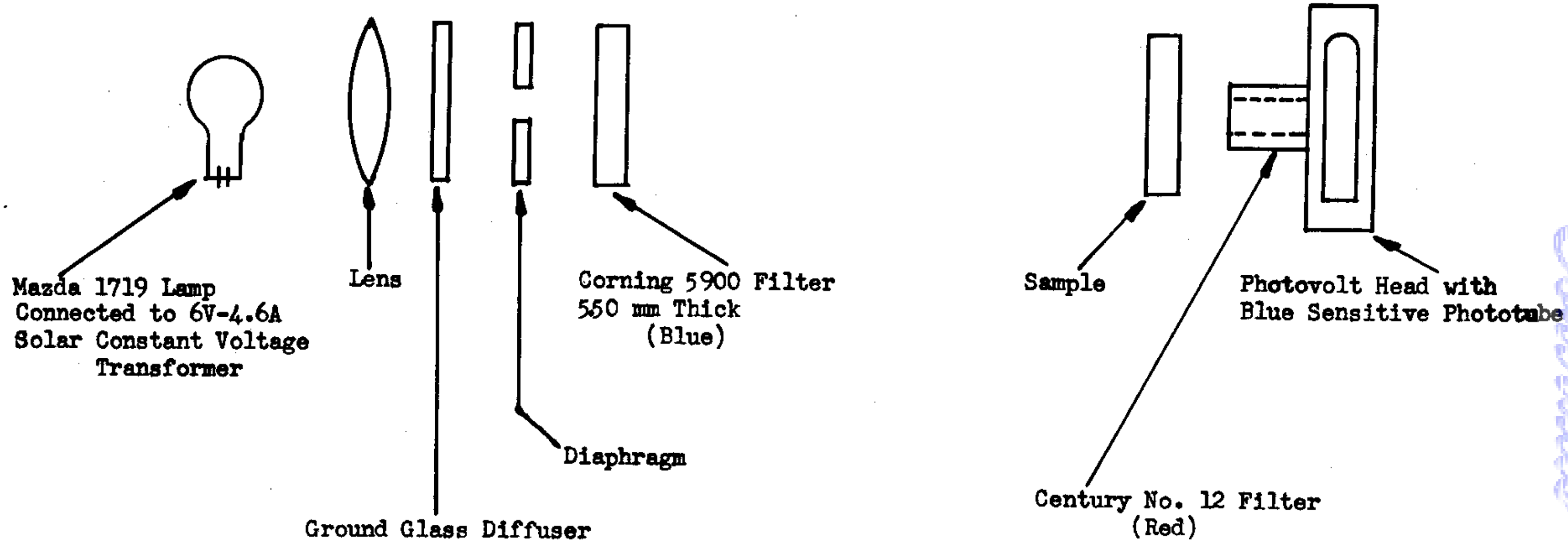


FIGURE 1. APPARATUS FOR MEASURING OPTICAL DENSITY AND YELLOWING INDEX

This method is an empirical one and the results depend on the exact manner in which the apparatus is constructed. The same apparatus was employed to measure the density and yellowing index of the samples before they were placed on the roof and after exposure for approximately 1.5 years on the roof.

A Century No. 53 filter which possessed an optical density of 0.36 measured on this apparatus was employed to check the instrument readings and good agreement was obtained between readings on this filter at the time the samples were placed on the roof and at the time the samples were removed.

The simple density value was used because it could be easily corrected for any variation in thickness of the sample since the density varies directly with thickness.

This method of measuring light transmission and optical density of the samples is similar to the method employed independently at Wright Air Development Center in Technical Note WCRTY 52-10 on "Thermal and Ultraviolet Degradation of Polymethyl Alpha-Chloroacrylate" using a blue filter to obtain "a more salient and pronounced measure of the degradation" caused by light (10).

1.2.1 Sample Size and Details of Exposure Test

The samples cut out for the exposure tests measured 1 1/2 in. x 3 in. x the thickness of the cast sheet (usually 1/8 in. or 1/4 in.). They were mounted on the roof by drilling a hole through the sample as sketched in Figure 2 and inserting Nichrome wire through the hole and allowing the sample to hang from the wire which was strung horizontally about 3 ft above the roof. Pyrex glass spacers of 1/4 in. OD Pyrex tubing in lengths of 1 in. to 1 1/4 in. were strung on the wire between the samples so that the sides of the samples were not protected by adjacent samples.

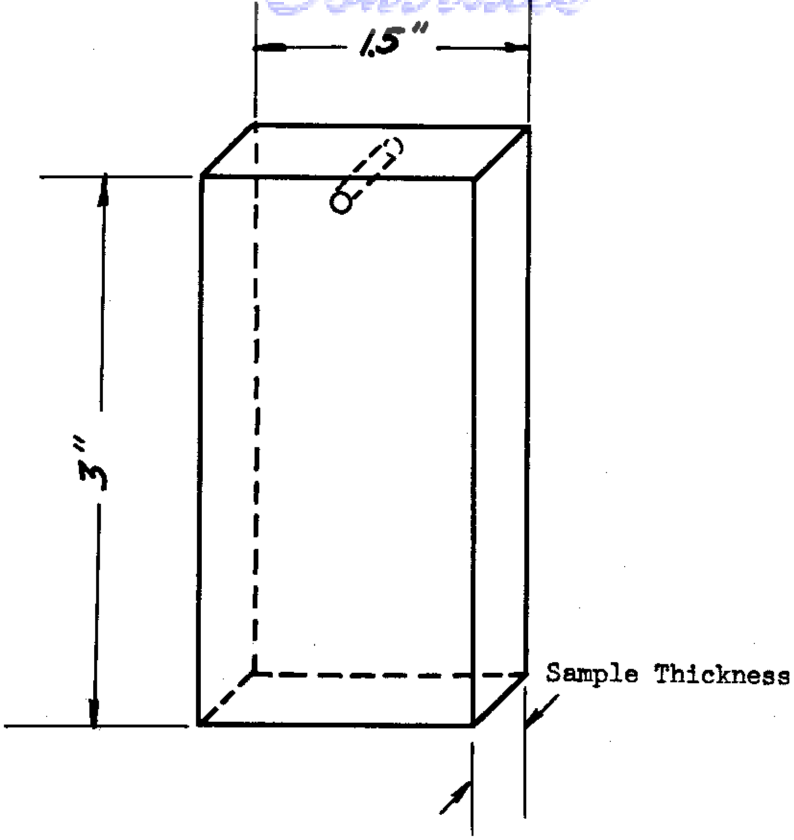
Each sheet casting tested was represented by two samples, one of which was mounted on the roof as described above and another of the same size which was stored in the dark indoors in file drawers over the same period of time as the roof exposure tests. Only samples possessing uniformly smooth surfaces are reported in the following tables so that surface conditions affecting optical density would not be an important factor.

The results of the tests are tabulated and shown graphically later in this section. A limited discussion of these results also follows, but the relating of these results to the methods of preparation and polymerization of the cast sheets is included in the next section.

1.3 Yellowing on Roof Exposure

If the samples yellowed uniformly, the changes in optical density or change in yellowing index corrected to a standard thickness would express the change in yellowing index caused by light exposure. If the samples yellowed only on the surfaces, no correction for thickness would be required since all the samples possessed two smooth exposed surfaces.

The samples actually appeared to undergo both types of yellowing, but the surface yellowing was usually more pronounced and the samples appeared to possess a



Edge View After Light Exposure

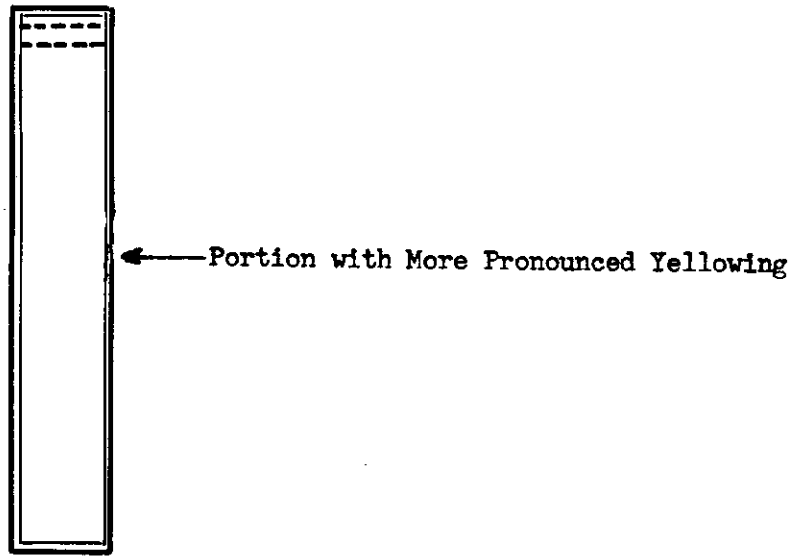


FIGURE 2. SAMPLE SIZE FOR COLOR STABILITY TESTS ON "GAFITE"

layer of darker yellow polymer next to the surfaces than in the center as shown in Figure 2. Accordingly, the sample results are not corrected for thickness although it probably is a factor but a minor one which appeared to be very difficult to assess. For reference purposes, the thickness measurements are included in Table 1 which lists the sample numbers and the observed optical densities. The letter F refers to the sample stored in the dark while the letter R refers to the roof exposure sample. The difference in original optical density is the difference between the original F and R samples and is expressed as a positive number when the original density of R was greater than that of F and as a negative number when the reverse was true. The density change on dark storage is the difference between the densities of the F sample before and after dark storage. It is expressed as a positive number when the F sample became more yellow on dark storage.

The density change on roof exposure is the difference between the densities of the R sample before and after roof exposure. It is expressed as a positive number when the sample became more yellow on roof exposure. The density difference between dark storage and roof exposure is the difference in densities between the F and R samples at the end of 1.5 years. It is expressed as a positive number when the roof exposure sample was more yellow than the dark storage sample.

The exact dates of exposure are recorded for each sample. The period of exposure varied from two weeks to seven weeks less than 1.5 years. This variation is very likely of little consequence since observations on yellowing of "Gafite" at GAF and at Wright Air Development Center have indicated that the rate of yellowing is greatest at the beginning of exposure and that this rate of yellowing slows down with increase in the time of exposure.

1.3.1 Probable Accuracy of Results

An indication of the accuracy and reproducibility of the readings of optical density or yellowing index (100 x optical density) is obtained from the differences in yellowing index values for the original "Gafite" samples (F and R). As indicated in Figure 3, over 90% of the yellowing index values agreed within ± 2.0 (± 0.02 in optical density).

Table 2 lists the samples and their differences in optical densities for original "Gafite" samples. This information was used in plotting Figure 3.

1.3.2 Changes During Dark Storage

Figure 4 indicates the changes in yellowing index of "Gafite" samples on dark storage. The data in Table 3 were employed to plot Figure 4. This figure indicates that the majority of the dark storage samples did not change more than the probable experimental error (± 2.0 in yellowing index values) but that quite a few samples did increase in yellowing index on dark storage whereas very few decreases in yellowing index beyond experimental error were noted.

1.3.3 Comparison of Yellowing in Dark Storage with Yellowing on Roof Exposure

In Figure 5 are plotted the data assembled in Table 4 on the differences in yellowing index between dark storage samples and roof exposure samples after 1.5 years. As expected, the majority of the roof exposure samples were more yellow than

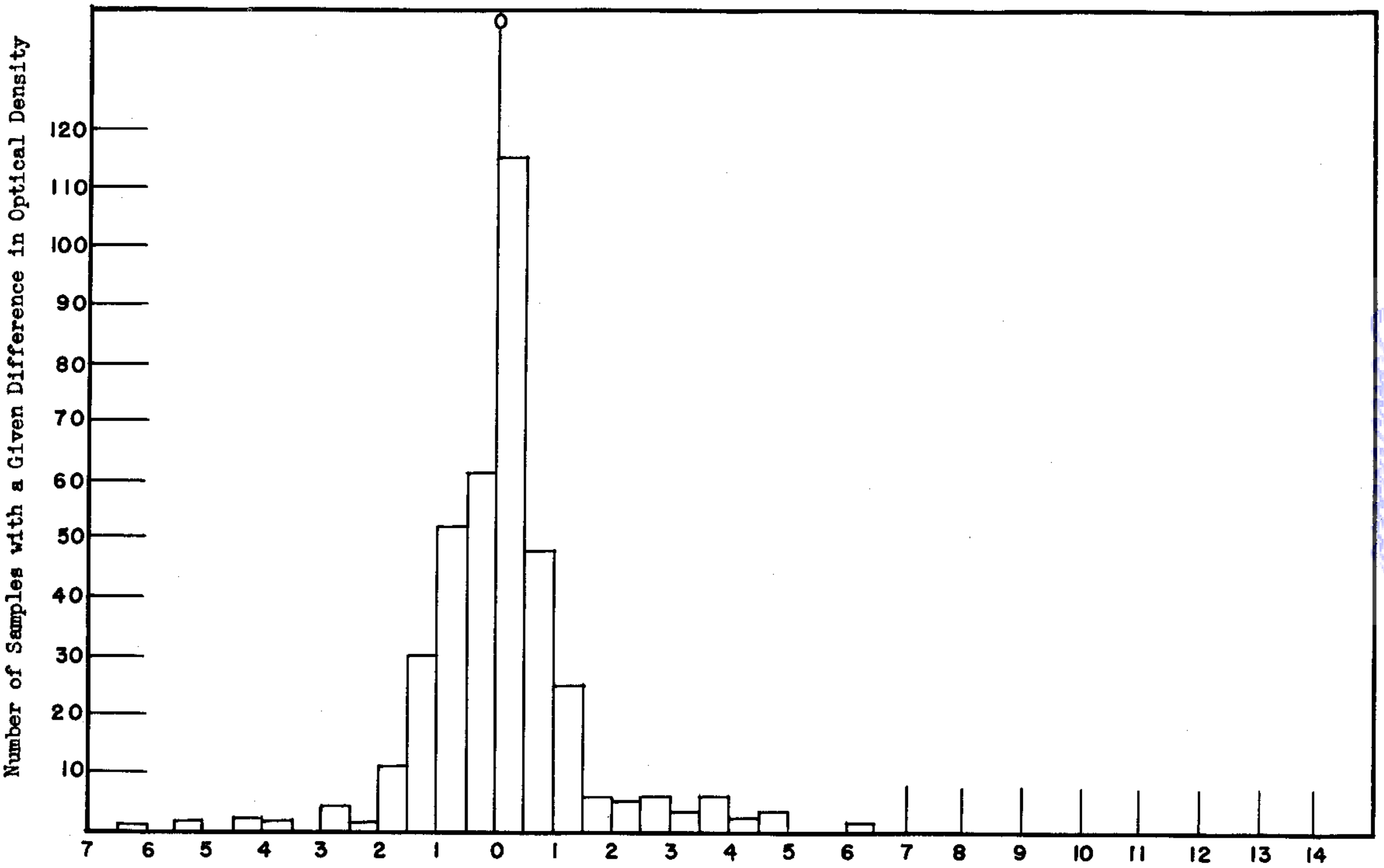


FIGURE 3. DIFFERENCES IN OPTICAL DENSITIES BETWEEN ORIGINAL "GAFITE" SAMPLES FROM 385 SHEETS

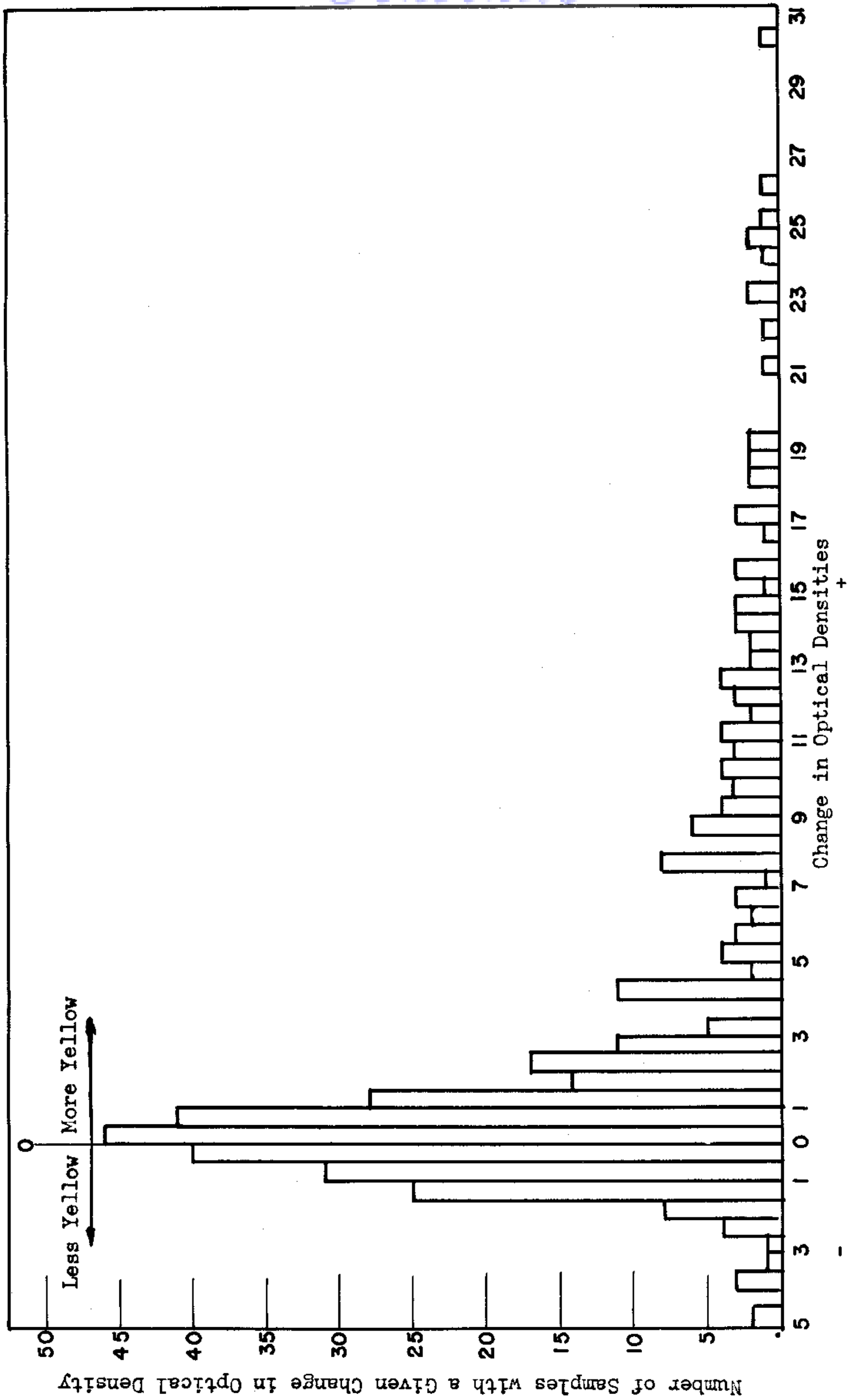


FIGURE 4. CHANGES IN OPTICAL DENSITIES OF 385 "GAFITE" SAMPLES ON DARK STORAGE FOR 1.5 YEARS

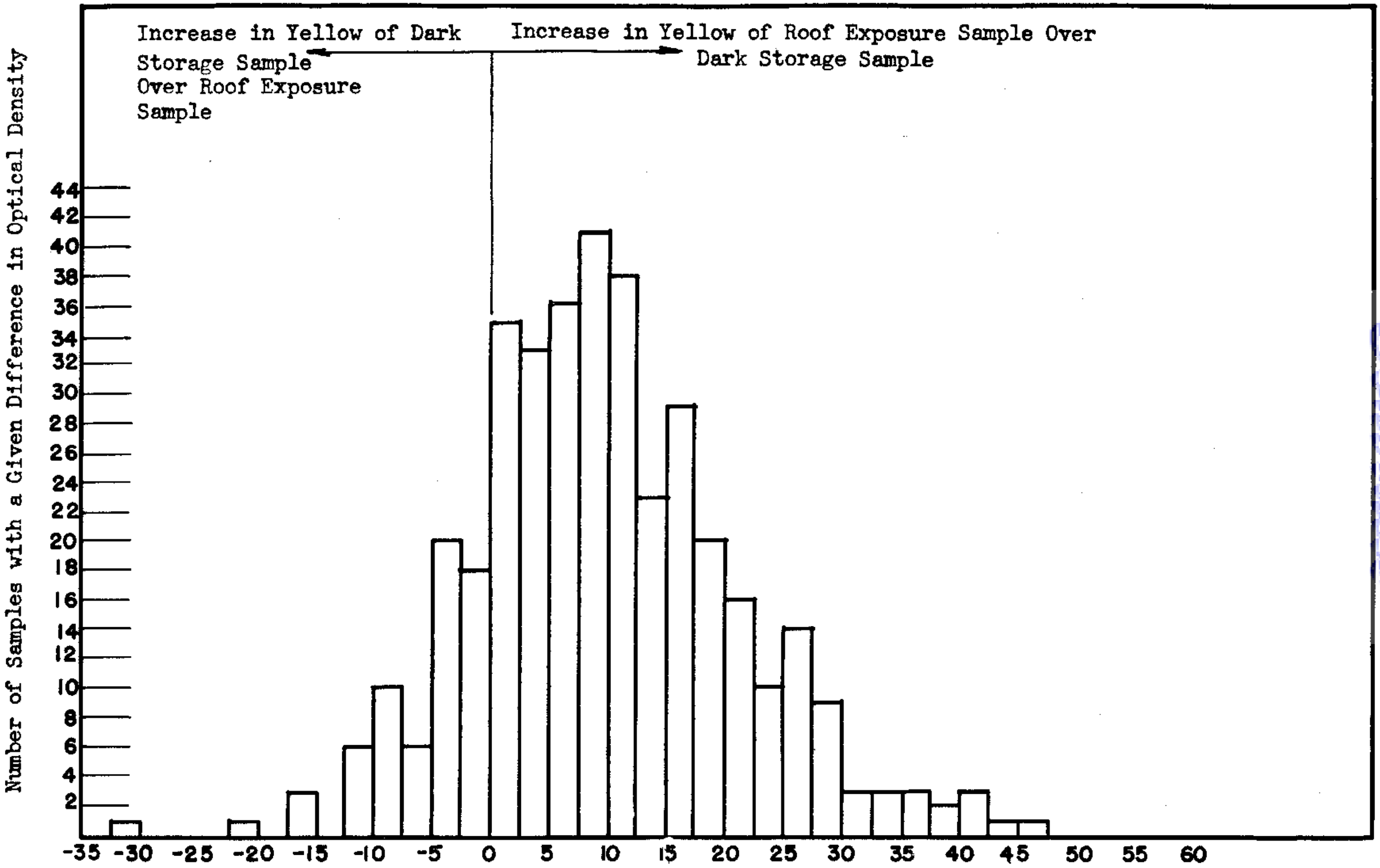


FIGURE 5. DIFFERENCES IN OPTICAL DENSITIES BETWEEN "GAFITE" DARK STORAGE AND ROOF EXPOSURE SAMPLES AFTER 1.5 YEARS AT EASTON, PA.

TABLE 1. LIGHT STABILITY TESTS ON "GAFITE"

Sample Number	Thickness in Inches	Original Densities	Densities After Dark Storage and Roof Exposure	Difference in Original Density Values	Density Change on Dark Storage	Density Change on Roof Exposure	Density Difference Between Dark Storage and Roof Exposure
January 21, 1952 to July 7, 1953							
1 F	0.106-0.105	0.065	0.065		+0.000		
1 R	0.105-0.107	0.065	0.330	+0.000		+0.265	+0.265
2 F	0.112-0.111	0.095	0.085		-0.010		
2 R	0.111-0.111	0.096	0.190	+0.001		+0.094	+0.105
3 F	0.132-0.128	0.065	0.055		-0.010		
3 R	0.126-0.130	0.065	0.231	+0.000		+0.166	+0.176
4 F	0.135-0.134	0.060	0.054		-0.006		
4 R	0.133-0.135	0.060	0.210	+0.000		+0.150	+0.156
5 F	0.114-0.118	0.058	0.050		-0.008		
5 R	0.121-0.118	0.050	0.400	-0.008		+0.350	+0.350
6 F	0.128-0.130	0.069	0.076		+0.007		
6 R	0.129-0.130	0.070	0.381	+0.001		+0.311	+0.305
7 F	0.103-0.101	0.063	0.060		-0.003		
7 R	0.101-0.099	0.064	0.298	+0.001		+0.234	+0.238
8 F	0.137-0.134	0.041	0.039		-0.002		
8 R	0.132-0.135	0.041	0.110	+0.000		+0.069	+0.071
9 F	0.126-0.132	0.069	0.058		-0.011		
9 R	0.131	0.025	0.240	-0.044		+0.215	+0.182
10 F	0.133-0.133	0.044	0.037		-0.007		
10 R	0.136-0.134	0.041	0.212	-0.003		+0.171	+0.175
11 F	0.124-0.122	0.056	0.039		-0.017		
11 R	0.122-0.124	0.050	0.245	-0.006		+0.195	+0.206
14 F	0.112-0.123	0.066	0.055		-0.011		
14 R	0.110-0.125	0.064	0.210	-0.002		+0.146	+0.155
15 F	0.124-0.126	0.045	0.040		-0.005		
15 R	0.125-0.127	0.046	0.250	+0.001		+0.204	+0.210
18 F	0.132-0.129	0.040	0.037		-0.003		
18 R	0.133-0.132	0.049	0.185	+0.009		+0.136	+0.148
19 F	0.125-0.121	0.044	0.030		-0.014		
19 R	0.121-0.124	0.044	0.187	+0.000		+0.143	+0.157
20 F	0.123-0.125	0.066	0.057		-0.009		
20 R	0.123-0.125	0.066	0.301	+0.000		+0.235	+0.244
21 F	0.123-0.123	0.060	0.065		+0.005		
21 R	0.123-0.123	0.049	0.300	-0.011		+0.251	+0.235

TABLE 1. (Continued) LIGHT STABILITY TESTS ON "GAFITE"

Sample Number	Thickness in Inches	Original Densities	Densities After Dark Storage and Roof Exposure	Difference in Original Density Values	Density Change on Dark Storage	Density Change on Roof Exposure	Density Difference Between Dark Storage and Roof Exposure
22 F	0.130-0.137	0.085	0.085		+0.000		
22 R	0.139-0.133	0.076	0.180	-0.009		+0.104	+0.095
23 F	0.123-0.127	0.090	0.105		+0.015		
23 R	0.130-0.123	0.085	0.180	-0.005		+0.095	+0.075
25 F	0.125-0.130	0.140	0.160		+0.020		
25 R	0.122-0.119	0.155	0.165	-0.015		+0.010	+0.005
36 F	0.117-0.125	0.046	0.055		+0.006		
36 R	0.128-0.130	0.045	0.300	-0.001		+0.255	+0.245
37 F	0.123-0.124	0.054	0.065		+0.011		
37 R	0.122-0.125	0.054	0.220	+0.000		+0.166	+0.155
38 F	0.123-0.126	0.057	0.056		-0.001		
38 R	0.121-0.125	0.058	0.190	+0.001		+0.132	+0.134
40 F	0.117-0.115	0.054	0.050		-0.004		
40 R	0.111-0.115	0.054	0.141	+0.000		+0.087	+0.091
41 F	0.118-0.120	0.054	0.058		+0.004		
41 R	0.117-0.118	0.054	0.210	+0.000		+0.156	+0.152
42 F	0.118-0.118	0.071	0.073		+0.002		
42 R	0.118-0.118	0.075	0.175	-0.004		+0.100	+0.102
43 F	0.102-0.108	0.085	0.075		-0.010		
43 R	0.101-0.108	0.087	0.155	+0.002		+0.068	+0.080
44 F	0.118-0.121	0.094	0.089		-0.005		
44 R	0.110-0.103	0.110	0.110	+0.016		+0.000	+0.021
48 F	0.115-0.114	0.051	0.048		-0.003		
48 R	0.115-0.115	0.054	0.130	+0.003		+0.076	+0.082
49 F	0.109-0.112	0.040	0.035		-0.005		
49 R	0.111-0.108	0.045	0.064	+0.005		+0.019	+0.029
51 F	0.127-0.134	0.075	0.063		-0.012		
51 R	0.127-0.118	0.064	0.144	-0.011		+0.080	+0.081
52 F	0.115-0.118	0.049	0.045		-0.004		
52 R	0.116-0.119	0.045	0.160	-0.004		+0.115	+0.115
54 F	0.112-0.112	0.040	0.040		+0.000		
54 R	0.113-0.110	0.038	0.160	-0.002		+0.122	+0.120
55 F	0.116-0.124	0.110	0.095		-0.015		
55 R	0.116-0.120	0.140	0.095	+0.030		-0.045	+0.000
57 F	0.120-0.112	0.045	0.039		-0.006		
57 R	0.119-0.111	0.046	0.200	+0.001		+0.154	+0.161
58 F	0.122-0.122	0.079	0.064		-0.015		
58 R	0.119-0.126	0.081	0.100	+0.002		+0.019	+0.036

TABLE 1. (Continued) LIGHT STABILITY TESTS ON "GAFITE"

Sample Number	Thickness in Inches	Original Densities	Densities After Dark Storage and Roof Exposure	Difference in Original Density Values	Density Change on Dark Storage	Density Change on Roof Exposure	Density Difference Between Dark Storage and Roof Exposure
63 F	0.106-0.106	0.042	0.043		+0.001	+0.103	+0.106
63 R	0.102-0.101	0.046	0.149	+0.004			
64 F	0.085-0.082	0.046	0.075		+0.029	+0.175	+0.135
64 R	0.095-0.087	0.035	0.210	-0.011			
65 F	0.110-0.108	0.031	0.030		-0.001	+0.082	+0.087
65 R	0.105-0.105	0.035	0.117	+0.004			
66 F	0.121-0.121	0.041	0.030		-0.011	+0.075	+0.090
66 R	0.123-0.122	0.045	0.120	+0.004			
67 F	0.102-0.103	0.050	0.055		+0.005	+0.069	+0.065
67 R	0.103-0.102	0.051	0.120	+0.001			
68 F	0.114-0.113	0.051	0.060		+0.009	+0.080	+0.070
68 R	0.119-0.117	0.050	0.130	-0.001			
69 F	0.090-0.092	0.045	0.045		+0.000	+0.185	+0.180
69 R	0.094-0.091	0.040	0.225	-0.005			
70 F	0.114-0.113	0.050	0.042		-0.008	+0.102	+0.113
70 R	0.113-0.107	0.053	0.155	+0.003			
71 F	0.091-0.092	0.066	0.060		-0.006	+0.080	+0.090
71 R	0.091-0.090	0.070	0.150	+0.004			
72 F	0.091-0.086	0.060	0.050		-0.010	+0.125	+0.135
72 R	0.092-0.087	0.060	0.185	+0.000			
73 F	0.078-0.076	0.055	0.085		+0.030	+0.072	+0.044
73 R	0.081-0.083	0.057	0.129	+0.002			
74 F	0.097-0.101	0.060	0.125		+0.065	+0.140	+0.070
74 R	0.107-0.108	0.055	0.195	-0.005			
75 F	0.115-0.113	0.065	0.055		-0.010	+0.079	+0.080
75 R	0.114-0.116	0.056	0.135	-0.009			
76 F	0.112-0.116	0.050	0.042		-0.008	+0.134	+0.132
76 R	0.117-0.114	0.040	0.174	-0.010			
77 F	0.113-0.108	0.055	0.055		+0.000	+0.072	+0.067
77 R	0.110-0.108	0.050	0.122	-0.005			
78 F	0.110-0.112	0.056	0.035		-0.021	+0.080	+0.105
78 R	0.114-0.108	0.060	0.140	+0.004			
79 F	0.112-0.107	0.060	0.085		+0.025	+0.041	+0.015
79 R	0.112-0.109	0.059	0.100	-0.001			
80 F	0.109-0.107	0.064	0.062		-0.002	+0.087	+0.097
80 R	0.106-0.114	0.072	0.159	+0.008			
81 F	0.098-0.099	0.059	0.055		-0.004	+0.053	+0.055
81 R	0.097-0.100	0.057	0.110	-0.002			

TABLE 1. (Continued) LIGHT STABILITY TESTS ON "GAFITE"

Sample Number	Thickness in Inches	Original Densities	Densities After Dark Storage and Roof Exposure	Difference in Original Density Values	Density Change on Dark Storage	Density Change on Roof Exposure	Density Difference Between Dark Storage and Roof Exposure
82 F	0.109-0.110	0.059	0.062		+0.003		
82 R	0.113-0.111	0.054	0.126	-0.005		+0.072	+0.064
83 F	0.111-0.114	0.050	0.055		+0.005		
83 R	0.112-0.110	0.054	0.165	+0.004		+0.111	+0.110
84 F	0.091-0.088	0.054	0.046		-0.008		
84 R	0.088-0.091	0.098	0.123	+0.044		+0.025	+0.077
January 23, 1952 to July 7, 1953							
86 F	0.118-0.112	0.059	0.071		+0.012		
86 R	0.108-0.103	0.085	0.134	+0.026		+0.049	+0.063
87 F	0.108-0.111	0.070	0.065		-0.005		
87 R	0.110-0.112	0.065	0.158	-0.005		+0.093	+0.093
88 F	0.102-0.103	0.089	0.070		-0.019		
88 R	0.106-0.106	0.085	0.100	-0.004		+0.015	+0.030
89 F	0.121-0.117	0.065	0.062		-0.003		
89 R	0.113-0.106	0.062	0.160	-0.003		+0.098	+0.098
90 F	0.103-0.104	0.056	0.051		-0.005		
90 R	0.104-0.108	0.065	0.151	+0.009		+0.086	+0.100
91 F	0.114-0.110	0.059	0.100		+0.042		
91 R	0.111-0.115	0.055	0.140	-0.004		+0.085	+0.040
93 F	0.118-0.121	0.075	0.067		-0.008		
93 R	0.120-0.118	0.064	0.140	-0.011		+0.076	+0.073
94 F	0.107-0.107	0.060	0.060		+0.000		
94 R	0.110-0.110	0.064	0.167	+0.004		+0.103	+0.107
95 F	0.113-0.110	0.040	0.040		+0.000		
95 R	0.113-0.115	0.048	0.185	+0.008		+0.137	+0.145
96 F	0.082-0.084	0.055	0.045		-0.010		
96 R	0.081-0.082	0.057	0.120	+0.002		+0.063	+0.075
98 F	0.110-0.106	0.058	0.054		-0.004		
98 R	0.105-0.099	0.064	0.150	+0.006		+0.086	+0.096
99 F	0.090-0.085	0.055	0.045		-0.010		
99 R	0.087-0.083	0.052	0.199	-0.003		+0.147	+0.154
102 F	0.109-0.112	0.053	0.045		-0.008		
102 R	0.110-0.111	0.052	0.180	-0.001		+0.128	+0.135
103 F	0.117-0.115	0.055	0.050		-0.005		
103 R	0.115-0.124	0.069	0.210	+0.014		+0.141	+0.160
104 F	0.083-0.084	0.114	0.101		-0.013		
104 R	0.089-0.086	0.114	0.160	+0.000		+0.046	+0.059

TABLE 1. (Continued) LIGHT STABILITY TESTS ON "GAFITE"

Sample Number	Thickness in Inches	Original Densities	Densities After Dark Storage and Roof Exposure	Difference in Original Density Values	Density Change on Dark Storage	Density Change on Roof Exposure	Density Difference Between Dark Storage and Roof Exposure
105 F	0.112-0.115	0.092	0.136		+0.044		
105 R	0.103-0.103	0.092	0.166	+0.000		+0.074	+0.030
106 F	0.098-0.100	0.106	0.095		-0.011		
106 R	0.097-0.099	0.094	0.165	-0.012		+0.071	+0.070
107 F	0.106-0.110	0.076	0.075		-0.001		
107 R	0.109-0.108	0.075	0.175	-0.001		+0.100	+0.100
109 F	0.103-0.103	0.067	0.065		-0.002		
109 R a/	0.099-0.100	0.068	0.200	+0.001		+0.132	+0.135
110 F	0.106-0.111	0.065	0.062		-0.003		
110 R a/	0.115-0.110	0.078	0.185	+0.013		+0.107	+0.123
112 F	0.124-0.119	0.090	0.080		-0.010		
112 R a/	0.119-0.124	0.080	0.170	-0.010		+0.090	+0.090
115 F	0.112-0.112	0.056	0.055		-0.001		
115 R	0.112-0.113	0.058	0.260	+0.002		+0.202	+0.205
116 F	0.115-0.120	0.078	0.074		-0.004		
116 R	0.114-0.116	0.069	0.229	-0.009		+0.160	+0.155
117 F	0.087-0.095	0.058	0.060		+0.002		
117 R	0.085-0.080	0.056	0.200	-0.002		+0.144	+0.140
118 F	0.115-0.114	0.035	0.045		+0.010		
118 R	0.116-0.114	0.049	0.202	+0.014		+0.153	+0.157
120 F	0.100-0.103	0.058	0.054		-0.004		
120 R	0.102-0.102	0.045	0.155	-0.013		+0.110	+0.101
121 F	0.113-0.114	0.062	0.059		-0.003		
121 R	0.120-0.117	0.064	0.188	+0.002		+0.124	+0.129
122 F	0.108-0.106	0.057	0.053		-0.004		
122 R	0.112-0.105	0.056	0.157	-0.001		+0.101	+0.104
125 F	0.130-0.137	0.051	0.045		-0.006		
125 R a/	0.139-0.134	0.046	0.340	-0.005		+0.294	+0.295
126 F	0.081-0.082	0.049	0.040		-0.009		
126 R	0.076-0.079	0.045	0.323	-0.004		+0.278	+0.283
127 F	0.116-0.114	0.039	0.045		+0.006		
127 R	0.114-0.116	0.049	0.229	+0.010		+0.180	+0.184
128 F	0.115-0.117	0.061	0.066		+0.005		
128 R	0.119-0.119	0.064	0.210	+0.003		+0.146	+0.154
129 F	0.113-0.115	0.063	0.065		+0.002		
129 R	0.114-0.115	0.065	0.202	+0.002		+0.137	+0.137
130 F	0.126-0.133	0.055	0.055		+0.000		
130 R	0.144-0.139	0.046	0.230	-0.009		+0.184	+0.175

TABLE 1. (Continued) LIGHT STABILITY TESTS ON "GAFITE"

Sample Number	Thickness in Inches	Original Densities	Densities After Dark Storage and Roof Exposure	Difference in Original Density Values	Density Change on Dark Storage	Density Change on Roof Exposure	Density Difference Between Dark Storage and Roof Exposure
131 F	0.115-0.109	0.052	0.059		+0.007		
131 R	0.109-0.111	0.053	0.219	+0.001		+0.166	+0.160
133 F	0.137-0.140	0.104	0.109		+0.005		
133 R	0.137-0.136	0.099	0.175	-0.005		+0.076	+0.066
139 F	0.138-0.135	0.170	0.157		-0.013		
139 R a/	0.134-0.133	0.159	0.195	-0.011		+0.036	+0.038
142 F	0.135-0.130	0.039	0.051		+0.012		
142 R	0.128-0.121	0.039	0.103	+0.000		+0.064	+0.052
143 F	0.131-0.124	0.045	0.040		-0.005		
143 R	0.122-0.128	0.045	0.065	+0.000		+0.020	+0.025
149 F	0.119-0.118	0.060	0.048		-0.012		
149 R b/	0.116-0.115	0.061	0.198	+0.001		+0.137	+0.150
150 F	0.120-0.119	0.045	0.047		+0.002		
150 R a/	0.119-0.122	0.050	0.114	+0.005		+0.064	+0.067
152 F	0.124-0.121	0.040	0.040		+0.000		
152 R b/	0.121-0.124	0.041	0.260	+0.001		+0.219	+0.220
155 F	0.122-0.121	0.045	0.051		+0.006		
155 R	0.123-0.133	0.040	0.090	-0.005		+0.050	+0.039
January 28, 1952 to July 28, 1953							
157 F	0.121-0.125	0.032	0.065		+0.033		
157 R b/	0.123-0.125	0.036	0.075	+0.004		+0.039	+0.010
161 F	0.122-0.119	0.084	0.085		+0.001		
161 R	0.123-0.122	0.085	0.145	+0.001		+0.060	+0.060
163 F	0.112-0.119	0.070	0.079		+0.009		
163 R	0.112-0.119	0.071	0.089	+0.001		+0.018	+0.010
167 F	0.118-0.114	0.050	0.059		+0.009		
167 R b/	0.121-0.116	0.043	0.094	-0.007		+0.051	+0.035
169 F	0.129-0.134	0.051	0.095		+0.044		
169 R b/	0.131-0.134	0.049	0.155	-0.002		+0.106	+0.060
174 F	0.117-0.120	0.055	0.057		+0.002		
174 R	0.117-0.120	0.065	0.230	+0.010		+0.165	+0.173
179 F	0.108-0.106	0.061	0.055		-0.006		
179 R b/	0.108-0.106	0.061	0.202	+0.000		+0.141	+0.147
182 F	0.117-0.122	0.062	0.050		-0.012		
182 R b/	0.114-0.117	0.060	0.172	-0.002		+0.112	+0.122
185 F	0.112-0.108	0.045	0.042		-0.003		
185 R b/	0.111-0.108	0.042	0.069	-0.003		+0.027	+0.027

TABLE 1. (Continued) LIGHT STABILITY TESTS ON "GAFITE"

Sample Number	Thickness in Inches	Original Densities	Densities After Dark Storage and Roof Exposure	Difference in Original Density Values	Density Change on Dark Storage	Density Change on Roof Exposure	Density Difference Between Dark Storage and Roof Exposure
188 F	0.112-0.109	0.050	0.050		+0.000		
188 R	0.109-0.108	0.054	0.083	+0.004		+0.029	+0.033
193 F	0.116-0.108	0.072	0.084		+0.012		
193 R	0.116-0.116	0.080	0.184	+0.008		+0.104	+0.100
February 1, 1952 to July 7, 1953							
201 F	0.125-0.132	0.066	0.050		-0.016		
201 R	0.131-0.130	0.055	0.152	-0.011		+0.097	+0.102
202 F	0.118-0.117	0.035	0.033		-0.002		
202 R	0.117-0.118	0.043	0.205	+0.008		+0.162	+0.172
203 F	0.136-0.133	0.035	0.044		+0.009		
203 R	0.127-0.130	0.034	0.197	-0.001		+0.153	+0.153
207 F	0.132-0.130	0.032	0.043		+0.011		
207 R	0.131-0.131	0.037	0.165	+0.005		+0.128	+0.122
208 F	0.269-0.268	0.054	0.055		+0.001		
208 R	0.269-0.272	0.050	0.320	-0.004		+0.270	+0.265
210 F	0.245-0.247	0.075	0.095		+0.020		
210 R	0.246-0.254	0.075	0.500	+0.000		+0.425	+0.405
211 F	0.249-0.258	0.164	0.157		-0.007		
211 R	0.269-0.259	0.155	0.412	-0.009		+0.257	+0.255
213 F	0.273-0.289	0.229	0.245		+0.016		
213 R	0.276-0.273	0.220	0.402	-0.009		+0.182	+0.157
215 F	0.278-0.279	0.090	0.110		+0.020		
215 R	0.253-0.261	0.084	0.360	-0.006		+0.276	+0.250
216 F	0.269-0.250	0.075	0.075		+0.000		
216 R	0.266-0.275	0.062	0.360	-0.013		+0.298	+0.285
219 F	0.267-0.276	0.085	0.114		+0.029		
219 R	0.260-0.267	0.085	0.320	+0.000		+0.235	+0.206
220 F	0.268-0.252	0.099	0.100		+0.001		
220 R	0.256-0.247	0.084	0.361	-0.015		+0.277	+0.261
221 F	0.259-0.261	0.075	0.082		+0.007		
221 R	0.255-0.260	0.083	0.292	+0.008		+0.209	+0.210
222 F	0.268-0.279	0.146	0.165		+0.019		
222 R	0.281-0.270	0.149	0.360	+0.003		+0.211	+0.195
224 F	0.270-0.272	0.160	0.155		-0.005		
224 R	0.271-0.275	0.148	0.380	-0.012		+0.232	+0.225
225 F	0.267-0.263	0.116	0.115		-0.001		
225 R	0.269-0.277	0.112	0.377	-0.004		+0.265	+0.262

TABLE 1. (Continued) LIGHT STABILITY TESTS ON "GAFITE"

Sample Number	Thickness in Inches	Original Densities	Densities After Dark Storage and Roof Exposure	Difference in Original Density Values	Density Change on Dark Storage	Density Change on Roof Exposure	Density Difference Between Dark Storage and Roof Exposure
226 F	0.271-0.281	0.141	0.149		+0.008		
226 R	0.266-0.275	0.143	0.322	+0.002		+0.179	+0.173
227 F	0.257-0.264	0.098	0.110		+0.012		
227 R	0.256-0.262	0.098	0.540	+0.000		+0.442	+0.430
228 F	0.261-0.264	0.210	0.195		-0.015		
228 R	0.272-0.267	0.219	0.610	+0.009		+0.391	+0.415
230 F	0.263-0.258	0.093	0.093		+0.000		
230 R	0.259-0.255	0.104	0.371	+0.011		+0.267	+0.278
232 F	0.257-0.264	0.072	0.075		+0.003		
232 R	0.258-0.265	0.078	0.390	+0.006		+0.312	+0.315
233 F	0.249-0.248	0.127	0.128		+0.001		
233 R	0.247-0.237	0.110	0.420	-0.017		+0.310	+0.292
234 F	0.263-0.253	0.097	0.085		-0.012		
234 R	0.260-0.251	0.089	0.380	-0.008		+0.291	+0.295
235 F	0.257-0.263	0.081	0.080		-0.001		
235 R	0.257-0.264	0.081	0.362	+0.000		+0.281	+0.282
236 F	0.272-0.277	0.089	0.090		+0.001		
236 R	0.278-0.282	0.079	0.349	-0.010		+0.270	+0.259
237 F	0.252-0.261	0.389	0.340		-0.049		
237 R	0.251-0.264	0.372	0.610	-0.017		+0.238	+0.270
239 F	0.250-0.260	0.095	0.072		-0.023		
239 R	0.252-0.242	0.103	0.410	+0.008		+0.307	+0.338
240 F	0.265-0.255	0.060	0.063		+0.003		
240 R	0.250-0.260	0.055	0.232	-0.005		+0.177	+0.169
242 F	0.130-0.123	0.050	0.060		+0.010		
242 R	0.121-0.124	0.054	0.156	+0.004		+0.102	+0.096
247 F	0.243-0.250	0.261	0.260		-0.001		
247 R	0.246-0.236	0.269	0.441	+0.008		+0.172	+0.181
248 F	0.256-0.263	0.310	0.300		-0.010		
248 R	0.257-0.251	0.298	0.528	-0.012		+0.230	+0.228
249 F	0.118-0.121	0.110	0.130		+0.020		
249 R	0.112-0.114	0.140	0.296	+0.030		+0.156	+0.166
250 F	0.120-0.123	0.172	0.197		+0.025		
250 R	0.126-0.120	0.174	0.400	+0.002		+0.226	+0.203
251 F	0.130-0.123	0.189	0.180		-0.009		
251 R	0.123-0.130	0.180	0.340	-0.009		+0.160	+0.160
253 F	0.116-0.114	0.351	0.330		-0.021		
253 R	0.114-0.116	0.389	0.550	+0.038		+0.191	+0.170
254 F	0.121-0.126	0.365	0.359		-0.006		
254 R	0.125-0.120	0.370	0.440	+0.005		+0.070	+0.081

TABLE 1. (Continued) LIGHT STABILITY TESTS ON "GAFITE"

Sample Number	Thickness in Inches	Original Densities	Densities After Dark Storage and Roof Exposure	Difference in Original Density Values	Density Change on Dark Storage	Density Change on Roof Exposure	Density Difference Between Dark Storage and Roof Exposure
256 F	0.246-0.249	0.315	0.320		+0.005		
256 R	0.245-0.247	0.320	0.420	+0.005		+0.100	+0.100
259 F	0.122-0.121	0.099	0.105		+0.006		
259 R	0.120-0.121	0.101	0.188	+0.002		+0.087	+0.083
260 F	0.117-0.119	0.104	0.114		+0.010		
260 R	0.120-0.121	0.108	0.180	+0.004		+0.072	+0.066
263 F	0.258-0.260	0.156	0.178		+0.022		
263 R	0.257-0.248	0.190	0.379	+0.034		+0.189	+0.201
264 F	0.244-0.247	0.101	0.111		+0.010		
264 R	0.241-0.248	0.091	0.425	-0.010		+0.334	+0.314
265 F	0.121-0.126	0.071	0.075		+0.004		
265 R	0.121-0.126	0.076	0.341	+0.005		+0.265	+0.266
266 F	0.112-0.113	0.051	0.058		+0.007		
266 R	0.111-0.110	0.055	0.175	+0.004		+0.120	+0.117
268 F	0.122-0.119	0.117	0.114		-0.003		
268 R	0.125-0.122	0.112	0.380	-0.005		+0.268	+0.266
269 F	0.233-0.223	0.143	0.150		+0.007		
269 R	0.224-0.235	0.162	0.349	+0.019		+0.187	+0.199
270 F	0.230-0.244	0.126	0.124		-0.002		
270 R	0.231-0.235	0.125	0.380	-0.001		+0.255	+0.256
271 F	0.222-0.223	0.115	0.124		+0.009		
271 R	0.229-0.223	0.132	0.220	+0.017		+0.088	+0.096
272 F	0.126-0.123	0.086	0.085		-0.001		
272 R	0.126-0.123	0.085	0.145	-0.001		+0.060	+0.060
274 F	0.237-0.239	0.130	0.128		-0.002		
274 R	0.236-0.245	0.130	0.290	+0.000		+0.160	+0.162
275 F	0.219-0.215	0.119	0.125		+0.006		
275 R	0.221-0.216	0.120	0.200	+0.001		+0.080	+0.075
276 F	0.234-0.229	0.099	0.100		+0.001		
276 R	0.235-0.224	0.089	0.329	-0.010		+0.240	+0.229
277 F	0.231-0.219	0.139	0.130		-0.009		
277 R	0.231-0.220	0.140	0.340	+0.001		+0.200	+0.210
278 F	0.106-0.103	0.141	0.140		-0.001		
278 R c/	0.110-0.102	0.148	0.139	+0.007		-0.009	-0.001
280 F	0.120-0.121	0.124	0.122		-0.002		
280 R b/	0.116-0.118	0.125	0.125	+0.001		+0.000	+0.003
284 F	0.240-0.231	0.220	0.196		-0.024		
284 R c/	0.232-0.240	0.228	0.230	+0.008		+0.002	+0.034

TABLE 1. (Continued) LIGHT STABILITY TESTS ON "GAFITE"

Sample Number	Thickness in Inches	Original Densities	Densities After Dark Storage and Roof Exposure	Difference in Original Density Values	Density Change on Dark Storage	Density Change on Roof Exposure	Density Difference Between Dark Storage and Roof Exposure
286 F	0.196-0.182	0.070	0.076		+0.006		
286 R	0.208-0.207	0.110	0.127	+0.040		+0.017	+0.051
291 F	0.185-0.183	0.072	0.067		-0.005		
291 R	0.180-0.180	0.071	0.109	-0.001		+0.038	+0.042
295 F	0.236-0.238	0.112	0.064		-0.048		
295 R	0.236-0.235	0.105	0.221	-0.007		+0.116	+0.157
298 F	0.252-0.248	0.112	0.123		+0.011		
298 R	0.249-0.252	0.109	0.150	-0.003		+0.041	+0.027
299 F	0.212-0.194	0.081	0.088		+0.007		
299 R	0.195-0.195	0.130	0.280	+0.049		+0.150	+0.192
February 7, 1952 to July 7, 1953							
302 F	0.242-0.238	0.062	0.063		+0.001		
302 R	0.240-0.233	0.057	0.184	-0.005		+0.127	+0.121
304 F	0.235-0.236	0.046	0.049		+0.003		
304 R	0.236-0.236	0.040	0.282	-0.006		+0.242	+0.233
305 F	0.223-0.225	0.072	0.070		-0.002		
305 R	0.225-0.222	0.068	0.218	-0.004		+0.150	+0.148
306 F	0.205-0.212	0.114	0.125		+0.011		
306 R	0.208-0.202	0.140	0.410	+0.026		+0.270	+0.285
307 F	0.236-0.234	0.059	0.055		-0.004		
307 R	0.229-0.228	0.059	0.114	+0.000		+0.055	+0.059
310 F	0.220-0.219	0.080	0.066		-0.014		
310 R	0.232-0.232	0.082	0.070	+0.002		-0.012	+0.004
312 F	0.257-0.253	0.070	0.060		-0.010		
312 R	0.252-0.253	0.065	0.275	-0.005		+0.210	+0.215
313 F	0.256-0.250	0.061	0.060		-0.001		
313 R	0.249-0.250	0.055	0.145	-0.006		+0.090	+0.085
314 F	0.237-0.238	0.050	0.052		+0.002		
314 R	0.239-0.245	0.056	0.257	+0.006		+0.201	+0.205
315 F	0.238-0.252	0.031	0.049		+0.018		
315 R	0.237-0.239	0.040	0.322	+0.009		+0.282	+0.273
317 F	0.228-0.230	0.074	0.076		+0.002		
317 R	0.229-0.228	0.080	0.105	+0.006		+0.025	+0.029
318 F	0.237-0.238	0.120	0.085		-0.035		
318 R	0.239-0.238	0.085	0.185	-0.035		+0.100	+0.100
319 F	0.238-0.238	0.092	0.090		-0.002		
319 R	0.239-0.238	0.089	0.287	-0.003		+0.198	+0.197

TABLE 1. (Continued) LIGHT STABILITY TESTS ON "GAFITE"

Sample Number	Thickness in Inches	Original Densities	Densities After Dark Storage and Roof Exposure	Difference in Original Density Values	Density Change on Dark Storage	Density Change on Roof Exposure	Density Difference Between Dark Storage and Roof Exposure
320 F	0.241-0.244	0.080	0.080	+0.006	+0.000	+0.074	+0.080
320 R	0.242-0.245	0.086	0.160				
321 F	0.231-0.231	0.105	0.089	-0.009	-0.016	+0.069	+0.076
321 R	0.234-0.233	0.096	0.165				
322 F	0.242-0.246	0.073	0.070	+0.007	-0.003	+0.165	+0.175
322 R	0.229-0.223	0.080	0.245				
323 F	0.297-0.300	0.155	0.161	+0.000	+0.006	+0.045	+0.039
323 R	0.298-0.296	0.155	0.200				
324 F	0.225-0.224	0.060	0.060	-0.010	+0.000	+0.139	+0.129
324 R	0.223-0.226	0.050	0.189				
325 F	0.237-0.238	0.066	0.070	+0.002	+0.004	+0.162	+0.160
325 R	0.238-0.237	0.068	0.230				
326 F	0.121-0.120	0.035	0.048	+0.005	+0.013	+0.170	+0.162
326 R	0.118-0.120	0.040	0.210				
327 F	0.110-0.108	0.040	0.055	+0.013	+0.015	+0.103	+0.101
327 R	0.109-0.108	0.053	0.156				
328 F	0.116-0.116	0.049	0.052	-0.005	+0.003	+0.131	+0.123
328 R	0.121-0.123	0.044	0.175				
329 F	0.131-0.133	0.089	0.075	-0.015	-0.014	+0.096	+0.095
329 R	0.132-0.133	0.074	0.170				
330 F	0.139-0.140	0.070	0.090	-0.005	+0.020	+0.145	+0.120
330 R	0.129-0.132	0.065	0.210				
331 F	0.132-0.128	0.068	0.070	-0.003	+0.002	+0.115	+0.110
331 R	0.128-0.129	0.065	0.180				
332 F	0.125-0.125	0.063	0.070	+0.007	+0.007	+0.089	+0.089
332 R	0.125-0.124	0.070	0.159				
333 F	0.140-0.140	0.082	0.090	-0.003	+0.008	+0.055	+0.044
333 R	0.134-0.133	0.079	0.134				
334 F	0.121-0.123	0.066	0.130	+0.013	+0.064	+0.011	+0.040
334 R	0.121-0.123	0.079	0.090				
335 F	0.092-0.095	0.060	0.061	+0.003	+0.001	+0.082	+0.084
335 R	0.094-0.089	0.063	0.145				
336 F	0.114-0.113	0.058	0.054	-0.004	-0.004	+0.102	+0.102
336 R	0.118-0.118	0.054	0.156				
337 F	0.131-0.132	0.370	0.387	-0.000	+0.017	+0.017	+0.000
337 R	0.132-0.133	0.370	0.387				
338 F	0.117-0.114	0.052	0.057	+0.005	+0.005	+0.023	+0.023
338 R	0.119-0.117	0.057	0.080				
341 F	0.449-0.420	0.210	0.225	-0.001	+0.015	+0.401	+0.385
341 R	0.441-0.418	0.209	0.610				

TABLE 1. (Continued) LIGHT STABILITY TESTS ON "GAFITE"

Sample Number	Thickness in Inches	Original Densities	Densities After Dark Storage and Roof Exposure	Difference in Original Density Values	Density Change on Dark Storage	Density Change on Roof Exposure	Density Difference Between Dark Storage and Roof Exposure
342 F	0.139-0.134	0.140	0.145		+0.005		
342 R	0.141-0.139	0.149	0.230	+0.009		+0.081	+0.085
344 F	0.238-0.234	0.250	0.260		+0.010		
344 R	0.235-0.235	0.249	0.280	-0.001		+0.031	+0.020
345 F	0.253-0.252	0.290	0.300		+0.010		
345 R	0.253-0.252	0.287	0.350	-0.003		+0.063	+0.050
348 F	0.246-0.239	0.440	0.435		-0.005		
348 R	0.245-0.245	0.428	0.425	-0.012		-0.003	-0.010
349 F	0.260-0.254	0.460	0.430		-0.030		
349 R	0.253-0.253	0.460	0.425	+0.000		-0.035	-0.005
350 F	0.268-0.259	0.425	0.417		-0.008		
350 R	0.258-0.248	0.408	0.414	-0.017		+0.006	-0.003
351 F	0.257-0.248	0.550	0.515		-0.035		
351 R	0.259-0.251	0.490	0.432	-0.060		-0.058	-0.083
355 F	0.250-0.250	0.502	0.519		+0.017		
355 R	0.238-0.235	0.502	0.449	+0.000		-0.053	-0.070
356 F	0.252-0.258	0.501	0.502		+0.004		
356 R	0.258-0.255	0.495	0.512	-0.006		+0.017	+0.007
357 F	0.256-0.257	0.490	0.500		+0.010		
357 R	0.245-0.245	0.480	0.410	-0.010		-0.070	-0.090
358 F	0.265-0.249	0.380	0.400		+0.020		
358 R	0.261-0.250	0.381	0.390	+0.001		+0.009	-0.010
359 F	0.242-0.239	0.339	0.380		+0.041		
359 R	0.245-0.250	0.338	0.357	-0.001		+0.019	-0.023
360 F	0.262-0.260	0.424	0.422		-0.002		
360 R	0.253-0.254	0.424	0.389	+0.000		-0.035	-0.033
361 F	0.235-0.237	0.320	0.329		+0.009		
361 R	0.229-0.233	0.310	0.330	-0.010		+0.020	+0.001
362 F	0.268-0.245	0.291	0.298		+0.007		
362 R	0.264-0.244	0.280	0.352	-0.011		+0.072	+0.054
363 F	0.255-0.256	0.379	0.387		+0.008		
363 R	0.250-0.250	0.392	0.350	+0.013		-0.042	-0.037
364 F	0.246-0.251	0.299	0.312		+0.013		
364 R	0.238-0.244	0.290	0.315	-0.009		+0.025	+0.003
365 F	0.244-0.255	0.260	0.389		+0.129		
365 R	0.242-0.235	0.390	0.370	+0.130		-0.020	-0.019
366 F	0.250-0.247	0.391	0.386		-0.005		
366 R	0.259-0.256	0.420	0.380	+0.029		-0.040	-0.006
368 F	0.247-0.243	0.257	0.370		+0.113		
368 R	0.247-0.244	0.253	0.279	-0.004		+0.026	-0.091

TABLE 1. (Continued) LIGHT STABILITY TESTS ON "GAFITE"

Sample Number	Thickness in Inches	Original Densities	Densities After Dark Storage and Roof Exposure	Difference in Original Density Values	Density Change on Dark Storage	Density Change on Roof Exposure	Density Difference Between Dark Storage and Roof Exposure
370 F	0.251-0.243	0.421	0.420	+0.007	-0.001	-0.013	-0.005
370 R	0.259-0.251	0.428	0.415				
371 F	0.256-0.255	0.305	0.330	-0.005	+0.030	+0.030	+0.000
371 R	0.262-0.259	0.300	0.330				
377 F	0.238-0.237	0.262	0.280	+0.008	+0.018	+0.010	+0.000
377 R	0.239-0.234	0.270	0.280				
378 F	0.124-0.133	0.129	0.149	-0.004	+0.020	+0.165	+0.141
378 R	0.129-0.120	0.125	0.290				
380 F	0.246-0.243	0.144	0.150	+0.004	+0.006	+0.162	+0.160
380 R	0.236-0.235	0.148	0.310				
381 F	0.237-0.240	0.115	0.126	-0.002	+0.011	+0.147	+0.134
381 R	0.239-0.241	0.113	0.260				
383 F	0.248-0.258	0.139	0.150	+0.002	+0.011	+0.279	+0.270
383 R	0.254-0.246	0.141	0.420				
384 F	0.261-0.260	0.108	0.126	+0.007	+0.018	+0.265	+0.254
384 R	0.272-0.256	0.115	0.380				
385 F	0.251-0.251	0.191	0.223	+0.004	+0.032	-0.011	-0.039
385 R	0.246-0.247	0.195	0.184				
386 F	0.259-0.252	0.310	0.330	+0.001	+0.020	+0.021	+0.002
386 R	0.254-0.252	0.311	0.332				
388 F	0.261-0.261	0.148	0.150	-0.006	+0.002	+0.100	+0.092
388 R	0.262-0.263	0.142	0.242				
389 F	0.253-0.252	0.126	0.180	-0.005	+0.054	+0.081	+0.022
389 R	0.250-0.250	0.121	0.202				
390 F	0.244-0.244	0.148	0.157	-0.003	+0.009	+0.050	+0.038
390 R	0.241-0.242	0.145	0.195				
391 F	0.258-0.255	0.150	0.175	-0.029	+0.025	+0.084	+0.030
391 R	0.259-0.259	0.121	0.205				
392 F	0.266-0.271	0.170	0.190	+0.002	+0.020	+0.028	+0.010
392 R	0.266-0.256	0.172	0.200				
393 F	0.278-0.278	0.159	0.210	-0.019	+0.051	+0.070	+0.000
393 R	0.258-0.259	0.140	0.210				
394 F	0.270-0.270	0.560	0.535	+0.000	-0.025	+0.440	+0.465
394 R	0.278-0.285	0.560	1.000				
396 F	0.253-0.254	0.166	0.175	-0.002	+0.009	+0.126	+0.115
396 R	0.250-0.259	0.164	0.290				
397 F	0.121-0.119	0.101	0.160	+0.004	+0.059	+0.085	+0.030
397 R	0.122-0.126	0.105	0.190				

TABLE 1. (Continued) LIGHT STABILITY TESTS ON "GAFITE"

Sample Number	Thickness in Inches	Original Densities	Densities After Dark Storage and Roof Exposure	Difference in Original Density Values	Density Change on Dark Storage	Density Change on Roof Exposure	Density Difference Between Dark Storage and Roof Exposure
398 F	0.119-0.118	0.095	0.120		+0.025		
398 R	0.117-0.118	0.105	0.170	+0.010		+0.065	+0.050
399 F	0.133-0.130	0.110	0.117		+0.007		
399 R	0.133-0.130	0.125	0.245	+0.015	•	+0.120	+0.128
February 18, 1952 to July 7, 1953							
400 F	0.275-0.290	0.252	0.275		+0.023		
400 R	0.250-0.259	0.230	0.240	-0.022		+0.010	-0.035
401 F	0.238-0.238	0.136	0.146		+0.010		
401 R	0.240-0.241	0.135	0.210	-0.001		+0.075	+0.064
404 F	0.243-0.244	0.130	0.142		+0.012		
404 R	0.238-0.237	0.130	0.289	+0.000		+0.159	+0.147
406 F	0.235-0.235	0.112	0.120		+0.008		
406 R	0.235-0.235	0.114	0.192	+0.002		+0.078	+0.072
407 F	0.252-0.258	0.150	0.155		+0.005		
407 R	0.247-0.235	0.145	0.278	-0.005		+0.133	+0.123
408 F	0.223-0.222	0.134	0.190		+0.056		
408 R	0.225-0.224	0.132	0.249	-0.002		+0.117	+0.059
409 F	0.243-0.238	0.115	0.149		+0.034		
409 R	0.242-0.237	0.117	0.247	+0.002		+0.130	+0.098
411 F	0.253-0.257	0.140	0.160		+0.020		
411 R	0.255-0.256	0.139	0.230	-0.001		+0.091	+0.070
412 F	0.246-0.265	0.168	0.182		+0.014		
412 R	0.243-0.261	0.155	0.192	-0.013		+0.037	+0.010
418 F	0.245-0.245	0.149	0.156		+0.007		
418 R	0.254-0.251	0.150	0.232	+0.001		+0.082	+0.076
419 F	0.246-0.251	0.149	0.157		+0.008		
419 R	0.251-0.250	0.145	0.270	-0.004		+0.125	+0.113
421 F	0.252-0.248	0.132	0.132		+0.000		
421 R	0.253-0.259	0.144	0.142	+0.012		-0.002	+0.010
422 F	0.233-0.238	0.176	0.190		+0.014		
422 R	0.232-0.231	0.148	0.259	-0.028		+0.111	+0.069
423 F	0.250-0.247	0.147	0.145		-0.002		
423 R	0.261-0.262	0.136	0.280	-0.011		+0.144	+0.135
425 F	0.239-0.239	0.138	0.157		+0.019		
425 R	0.240-0.240	0.133	0.280	-0.005		+0.147	+0.123
429 F	0.273-0.269	0.166	0.220		+0.054		
429 R	0.260-0.260	0.214	0.255	+0.048		+0.041	+0.035

TABLE 1. (Continued) LIGHT STABILITY TESTS ON "GAFITE"

Sample Number	Thickness in Inches	Original Densities	Densities After Dark Storage and Roof Exposure	Difference in Original Density Values	Density Change on Dark Storage	Density Change on Roof Exposure	Density Difference Between Dark Storage and Roof Exposure
431 F	0.251-0.249	0.208	0.230		+0.022		
431 R	0.248-0.249	0.207	0.260	-0.001		+0.053	+0.030
432 F	0.255-0.254	0.178	0.240		+0.062		
432 R	0.250-0.247	0.185	0.320	+0.007		+0.135	+0.080
434 F	0.246-0.247	0.320	0.360		+0.040		
434 R	0.256-0.250	0.330	0.241	+0.010		-0.089	-0.119
435 F	0.253-0.252	0.149	0.168		+0.019		
435 R	0.256-0.255	0.142	0.170	-0.007		+0.028	+0.002
437 F	0.260-0.255	0.262	0.358		+0.096		
437 R	0.255-0.249	0.237	0.258	-0.025		+0.021	-0.100
438 F	0.247-0.244	0.318	0.330		+0.012		
438 R	0.246-0.243	0.382	0.213	+0.064		-0.169	-0.117
439 F	0.249-0.246	0.290	0.300		+0.010		
439 R	0.246-0.246	0.272	0.279	-0.018		+0.007	-0.021
440 F	0.253-0.254	0.282	0.309		+0.027		
440 R	0.254-0.256	0.271	0.195	-0.011		-0.076	-0.114
442 F	0.247-0.264	0.212	0.240		+0.028		
442 R	0.257-0.264	0.216	0.246	+0.004		+0.030	+0.006
443 F	0.257-0.253	0.260	0.300		+0.040		
443 R	0.247-0.248	0.255	0.230	-0.005		-0.025	-0.070
444 F	0.237-0.238	0.170	0.240		+0.070		
444 R	0.236-0.238	0.166	0.225	-0.004		+0.059	-0.015
445 F	0.250-0.252	0.229	0.272		+0.043		
445 R	0.257-0.251	0.223	0.265	-0.006		+0.042	-0.007
446 F	0.247-0.248	0.225	0.249		+0.024		
446 R	0.248-0.249	0.229	0.262	+0.004		+0.033	+0.013
447 F	0.260-0.261	0.150	0.165		+0.015		
447 R	0.248-0.243	0.156	0.290	+0.006		+0.134	+0.125
449 F	0.240-0.246	0.520	0.510		-0.010		
449 R	0.240-0.239	0.520	0.410	+0.000		-0.110	-0.100
452 F	0.250-0.241	0.390	0.353		-0.037		
452 R	0.224-0.243	0.337	0.353	-0.053		+0.016	+0.000
455 F	0.231-0.233	0.257	0.301		+0.044		
455 R	0.234-0.232	0.260	0.270	+0.003		+0.010	-0.031
456 F	0.231-0.231	0.338	0.340		+0.002		
456 R	0.244-0.234	0.340	0.315	+0.002		-0.025	-0.025
457 F	0.233-0.231	0.427	0.428		+0.001		
457 R	0.232-0.235	0.422	0.540	-0.005		+0.118	+0.112

TABLE 1. (Continued) LIGHT STABILITY TESTS ON "GAFITE"

Sample Number	Thickness in Inches	Original Densities	Densities After Dark Storage and Roof Exposure	Difference in Original Density Values	Density Change on Dark Storage	Density Change on Roof Exposure	Density Difference Between Dark Storage and Roof Exposure
458 F	0.266-0.257	0.289	0.310		+0.021		
458 R	0.262-0.248	0.328	0.510	+0.039		+0.182	+0.200
460 F	0.241-0.244	0.265	0.290		+0.025		
460 R	0.258-0.249	0.310	0.399	+0.045		+0.089	+0.109
461 F	0.254-0.254	0.305	0.290		-0.015		
461 R	0.260-0.255	0.292	0.245	-0.013		-0.047	-0.045
462 F	0.240-0.243	0.221	0.230		+0.009		
462 R	0.247-0.240	0.237	0.301	+0.016		+0.064	+0.071
463 F	0.236-0.225	0.429	0.418		-0.011		
463 R	0.244-0.242	0.425	0.268	-0.004		-0.157	-0.150
467 F	0.270-0.257	0.340	0.340		+0.000		
467 R	0.250-0.254	0.299	0.277	-0.041		-0.022	-0.063
469 F	0.247-0.254	0.278	0.360		+0.082		
469 R	0.266-0.265	0.291	0.384	+0.013		+0.093	+0.024
472 F	0.246-0.255	0.210	0.262		+0.052		
472 R	0.254-0.263	0.219	0.228	+0.009		+0.009	-0.034
475 F	0.241-0.254	0.094	0.105		+0.011		
475 R	0.250-0.241	0.091	0.212	-0.003		+0.121	+0.107
478 F	0.230-0.231	0.155	0.196		+0.041		
478 R	0.230-0.230	0.154	0.340	-0.001		+0.186	+0.144
479 F	0.249-0.240	0.300	0.345		+0.045		
479 R	0.255-0.245	0.305	0.320	+0.005		+0.015	-0.025
480 F	0.235-0.240	0.259	0.260		+0.001		
480 R	0.243-0.248	0.261	0.439	+0.002		+0.178	+0.179
481 F	0.248-0.252	0.248	0.350		+0.102		
481 R	0.259-0.264	0.249	0.376	+0.001		+0.127	+0.026
484 F	0.230-0.235	0.189	0.203		+0.014		
484 R	0.227-0.220	0.190	0.390	+0.001		+0.200	+0.187
487 F	0.259-0.256	0.170	0.191		+0.021		
487 R	0.255-0.252	0.170	0.309	+0.000		+0.139	+0.118
488 F	0.259-0.254	0.213	0.231		+0.018		
488 R	0.254-0.257	0.226	0.442	+0.013		+0.216	+0.211
489 F	0.246-0.247	0.289	0.280		-0.009		
489 R	0.246-0.247	0.328	0.505	+0.039		+0.177	+0.225
490 F	0.273-0.267	0.312	0.420		+0.108		
490 R	0.278-0.268	0.306	0.405	-0.006		+0.099	-0.015
491 F	0.238-0.255	0.402	0.500		+0.098		
491 R	0.253-0.238	0.409	0.410	+0.007		+0.001	-0.090

TABLE 1. (Continued) LIGHT STABILITY TESTS ON "GAFITE"

Sample Number	Thickness in Inches	Original Densities	Densities After Dark Storage and Roof Exposure	Difference in Original Density Values	Density Change on Dark Storage	Density Change on Roof Exposure	Density Difference Between Dark Storage and Roof Exposure
492 F	0.281-0.275	0.391	0.520		+0.129		
492 R	0.269-0.276	0.401	0.490	+0.010		+0.089	-0.030
493 F	0.273-0.271	0.291	0.380		+0.089		
493 R	0.239-0.236	0.320	0.360	+0.029		+0.040	-0.020
494 F	0.251-0.252	0.249	0.380		+0.131		
494 R	0.257-0.255	0.240	0.359	-0.009		+0.119	-0.021
497 F	0.232-0.237	0.260	0.361		+0.101		
497 R	0.237-0.232	0.262	0.415	-0.002		+0.153	+0.054
498 F	0.235-0.243	0.258	0.359		+0.101		
498 R	0.237-0.231	0.259	0.360	+0.001		+0.101	+0.001
499 F	0.239-0.241	0.332	0.423		+0.111		
499 R	0.239-0.237	0.320	0.512	-0.012		+0.192	+0.089
February 28, 1952 to July 7, 1953							
500 F	0.265-0.262	0.276	0.440		+0.164		
500 R	0.268-0.261	0.281	0.629	+0.005		+0.348	+0.189
501 F	0.228-0.230	0.249	0.409		+0.160		
501 R	0.228-0.229	0.253	0.290	+0.004		+0.037	-0.119
502 F	0.234-0.236	0.245	0.362		+0.117		
502 R	0.233-0.233	0.255	0.343	+0.010		+0.088	-0.019
504 F	0.224-0.229	0.340	0.389		+0.049		
504 R	0.209-0.210	0.251	0.390	+0.011		+0.139	+0.001
505 F	0.255-0.254	0.620	0.700		+0.080		
505 R	0.254-0.252	0.625	0.710	+0.005		+0.085	+0.010
506 F	0.250-0.248	0.400	0.432		+0.032		
506 R	0.248-0.247	0.420	0.386	+0.020		-0.034	-0.046
507 F	0.275-0.269	0.425	0.500		+0.075		
507 R	0.263-0.280	0.422	0.540	-0.003		+0.118	+0.040
508 F	0.257-0.259	0.420	0.509		+0.089		
508 R	0.261-0.263	0.405	0.700	-0.015		+0.295	+0.191
509 F	0.245-0.245	0.270	0.412		+0.142		
509 R	0.245-0.245	0.280	0.448	+0.010		+0.168	+0.036
510 F	0.243-0.241	0.291	0.435		+0.144		
510 R	0.243-0.242	0.292	0.621	+0.001		+0.329	+0.186
511 F	0.244-0.243	0.258	0.430		+0.172		
511 R	0.244-0.244	0.268	0.639	+0.010		+0.371	+0.209
512 F	0.238-0.235	0.320	0.502		+0.182		
512 R	0.238-0.235	0.349	0.730	+0.029		+0.381	+0.228
513 F	0.241-0.237	0.320	0.422		+0.102		
513 R	0.241-0.237	0.301	0.630	-0.019		+0.329	+0.208

TABLE 1. (Continued) LIGHT STABILITY TESTS ON "GAFITE"

Sample Number	Thickness in Inches	Original Densities	Densities After Dark Storage and Roof Exposure	Difference in Original Density Values	Density Change on Dark Storage	Density Change on Roof Exposure	Density Difference Between Dark Storage and Roof Exposure
514 F	0.238-0.233	0.235	0.310		+0.075		
514 R	0.239-0.232	0.238	0.370	+0.003		+0.132	+0.060
515 F	0.248-0.247	0.219	0.318		+0.099		
515 R	0.247-0.250	0.220	0.380	+0.001		+0.160	+0.062
516 F	0.239-0.238	0.202	0.289		+0.087		
516 R	0.239-0.240	0.212	0.248	+0.010		+0.036	-0.041
518 F	0.246-0.243	0.319	0.319		+0.067		
518 R	0.243-0.246	0.349	0.349	-0.013		+0.110	+0.030
519 F	0.249-0.250	0.240	0.325		+0.085		
519 R	0.250-0.253	0.230	0.350	-0.010		+0.120	+0.025
521 F	0.261-0.269	0.301	0.425		+0.124		
521 R	0.250-0.249	0.305	0.380	+0.004		+0.075	-0.045
522 F	0.267-0.263	0.126	0.184		+0.058		
522 R	0.261-0.260	0.126	0.540	+0.000		+0.414	+0.356
523 F	0.253-0.251	0.265	0.400		+0.135		
523 R	0.250-0.251	0.279	0.380	+0.014		+0.101	-0.020
525 F	0.246-0.243	0.312	0.438		+0.126		
525 R	0.252-0.253	0.340	0.403	+0.028		+0.063	-0.035
526 F	0.241-0.242	0.195	0.235		+0.040		
526 R	0.247-0.242	0.200	0.532	+0.005		+0.332	+0.297
527 F	0.276-0.281	0.250	0.409		+0.159		
527 R	0.273-0.277	0.251	0.320	+0.001		+0.069	-0.089
528 F	0.261-0.258	0.224	0.301		+0.077		
528 R	0.255-0.258	0.229	0.521	+0.005		+0.292	+0.220
529 F	0.231-0.225	0.100	0.125		+0.025		
529 R	0.237-0.232	0.101	0.516	+0.001		+0.415	+0.391
530 F	0.240-0.242	0.306	0.350		+0.044		
530 R	0.237-0.240	0.310	0.350	+0.004		+0.040	+0.000
531 F	0.249-0.245	0.154	0.243		+0.089		
531 R	0.245-0.242	0.190	0.270	+0.036		+0.080	+0.027
532 F	0.236-0.232	0.114	0.180		+0.066		
532 R	0.237-0.234	0.114	0.548	+0.000		+0.434	+0.368
533 F	0.238-0.234	0.269	0.347		+0.078		
533 R	0.235-0.231	0.281	0.313	+0.012		+0.032	-0.034
535 F	0.250-0.250	0.105	0.187		+0.082		
535 R	0.246-0.248	0.080	0.600	-0.025		+0.520	+0.413
537 F	0.249-0.250	0.090	0.170		+0.080		
537 R	0.247-0.248	0.095	0.515	+0.005		+0.420	+0.345

TABLE 1. (Continued) LIGHT STABILITY TESTS ON "GAFITE"

Sample Number	Thickness in Inches	Original Densities	Densities After Dark Storage and Roof Exposure	Difference in Original Density Values	Density Change on Dark Storage	Density Change on Roof Exposure	Density Difference Between Dark Storage and Roof Exposure
538 F	0.245-0.245	0.390	0.410		+0.020		
538 R	0.255-0.246	0.426	0.412	+0.036		-0.014	+0.002
542 F	0.241-0.237	0.189	0.376		+0.187		
542 R	0.237-0.239	0.191	0.430	+0.002		+0.239	+0.054
543 F	0.244-0.245	0.075	0.195		+0.120		
543 R	0.250-0.248	0.072	0.525	-0.003		+0.453	+0.330
544 F	0.266-0.273	0.137	0.286		+0.149		
544 R	0.271-0.264	0.139	0.442	+0.002		+0.303	+0.156
545 F	0.245-0.242	0.225	0.380		+0.155		
545 R	0.248-0.251	0.231	0.349	+0.006		+0.118	-0.031
546 F	0.235-0.234	0.272	0.451		+0.079		
546 R	0.238-0.237	0.283	0.410	+0.011		+0.127	+0.059
547 F	0.248-0.246	0.228	0.320		+0.092		
547 R	0.253-0.250	0.251	0.510	+0.023		+0.259	+0.190
548 F	0.257-0.260	0.248	0.429		+0.181		
548 R	0.258-0.260	0.250	0.513	+0.002		+0.263	+0.084
549 F	0.259-0.249	0.134	0.221		+0.087		
549 R	0.254-0.252	0.120	0.420	-0.014		+0.300	+0.199
550 F	0.265-0.262	0.288	0.435		+0.147		
550 R	0.264-0.259	0.310	0.525	+0.022		+0.215	+0.090
551 F	0.248-0.252	0.168	0.273		+0.105		
551 R	0.246-0.248	0.174	0.432	+0.006		+0.258	+0.159
552 F	0.247-0.248	0.217	0.329		+0.112		
552 R	0.245-0.245	0.230	0.432	+0.013		+0.202	+0.103
553 F	0.241-0.241	0.337	0.430		+0.093		
553 R	0.241-0.243	0.321	0.380	-0.016		+0.059	-0.050
554 F	0.243-0.244	0.289	0.442		+0.153		
554 R	0.245-0.247	0.290	0.350	+0.001		+0.060	-0.092
556 F	0.241-0.242	0.120	0.360		+0.240		
556 R	0.236-0.236	0.115	0.376	-0.005		+0.261	+0.016
557 F	0.245-0.243	0.189	0.380		+0.191		
557 R	0.243-0.242	0.180	0.401	-0.009		+0.221	+0.021
558 F	0.248-0.244	0.221	0.409		+0.188		
558 R	0.245-0.250	0.205	0.339	-0.016		+0.134	-0.070
559 F	0.245-0.244	0.172	0.290		+0.118		
559 R	0.241-0.244	0.165	0.405	-0.007		+0.240	+0.115
561 F	0.264-0.266	0.228	0.320		+0.092		
561 R	0.270-0.267	0.231	0.407	+0.003		+0.176	+0.087
562 F	0.258-0.256	0.221	0.330		+0.109		
562 R	0.252-0.255	0.227	0.420	+0.006		+0.193	+0.090

TABLE 1. (Continued) LIGHT STABILITY TESTS ON "GAFITE"

Sample Number	Thickness in Inches	Original Densities	Densities After Dark Storage and Roof Exposure	Difference in Original Density Values	Density Change on Dark Storage	Density Change on Roof Exposure	Density Difference Between Dark Storage and Roof Exposure
563 F	0.251-0.252	0.241	0.412	-0.010	+0.171	+0.270	+0.089
563 R	0.249-0.248	0.231	0.501				
564 F	0.301-0.315	0.200	0.369	+0.009	+0.169	+0.223	+0.063
564 R	0.302-0.271	0.209	0.432				
565 F	0.257-0.261	0.145	0.221	+0.020	+0.076	+0.245	+0.189
565 R	0.261-0.256	0.165	0.410				
566 F	0.262-0.255	0.145	0.277	+0.035	+0.132	+0.245	+0.148
566 R	0.257-0.252	0.180	0.425				
567 F	0.259-0.267	0.199	0.292	+0.001	+0.093	+0.119	+0.027
567 R	0.271-0.264	0.200	0.319				
568 F	0.225-0.225	0.137	0.226	+0.002	+0.089	+0.181	+0.094
568 R	0.224-0.225	0.139	0.320				
569 F	0.236-0.236	0.155	0.230	-0.005	+0.075	+0.200	+0.120
569 R	0.238-0.238	0.150	0.350				
571 F	0.242-0.243	0.204	0.330	+0.011	+0.126	+0.075	-0.040
571 R	0.242-0.241	0.215	0.290				
572 F	0.254-0.256	0.189	0.330	-0.009	+0.141	+0.175	+0.025
572 R	0.248-0.247	0.180	0.355				
573 F	0.240-0.240	0.180	0.302	-0.003	+0.122	+0.183	+0.058
573 R	0.241-0.241	0.177	0.360				
574 F	0.243-0.244	0.157	0.336	-0.009	+0.079	+0.182	+0.094
574 R	0.241-0.239	0.148	0.330				
576 F	0.136-0.131	0.083	0.196	-0.003	+0.113	+0.250	+0.134
576 R	0.133-0.138	0.080	0.330				
577 F	0.246-0.242	0.225	0.362	-0.002	+0.137	+0.096	-0.043
577 R	0.242-0.247	0.223	0.319				
578 F	0.267-0.266	0.220	0.410	-0.004	+0.190	+0.104	-0.090
578 R	0.264-0.265	0.216	0.320				
579 F	0.254-0.252	0.254	0.409	+0.006	+0.155	+0.121	-0.028
579 R	0.265-0.262	0.260	0.381				
580 F	0.159-0.160	0.190	0.200	+0.001	+0.010	+0.048	+0.039
580 R	0.157-0.157	0.191	0.239				
581 F	0.271-0.270	0.260	0.430	-0.001	+0.170	+0.092	-0.079
581 R	0.270-0.272	0.259	0.351				
582 F	0.237-0.237	0.229	0.375	+0.002	+0.146	+0.149	+0.005
582 R	0.237-0.237	0.231	0.380				
584 F	0.234-0.235	0.249	0.510	+0.003	+0.261	+0.054	-0.204
584 R	0.234-0.234	0.252	0.306				
585 F	0.238-0.239	0.291	0.540	-0.002	+0.249	+0.091	-0.160
585 R	0.236-0.235	0.289	0.380				

TABLE 1. (Continued) LIGHT STABILITY TESTS ON "GAFITE"

Sample Number	Thickness in Inches	Original Densities	Densities After Dark Storage and Roof Exposure	Difference in Original Density Values	Density Change on Dark Storage	Density Change on Roof Exposure	Density Difference Between Dark Storage and Roof Exposure
586 F	0.243-0.242	0.265	0.512		+0.247		
586 R	0.245-0.244	0.262	0.352	-0.003		+0.090	-0.160
587 F	0.262-0.261	0.129	0.380		+0.251		
587 R	0.260-0.262	0.150	0.349	+0.021		+0.199	-0.031
588 F	0.262-0.263	0.198	0.410		+0.212		
588 R	0.263-0.266	0.190	0.322	-0.008		+0.132	-0.088
589 F	0.259-0.261	0.182	0.405		+0.223		
589 R	0.261-0.260	0.185	0.315	+0.003		+0.130	-0.090
590 F	0.266-0.267	0.169	0.400		+0.231		
590 R	0.264-0.262	0.165	0.385	-0.004		+0.220	-0.015
591 F	0.261-0.260	0.175	0.407		+0.232		
591 R	0.260-0.260	0.170	0.348	+0.005		+0.178	-0.059
592 F	0.188-0.187	0.320	0.620		+0.300		
592 R	0.194-0.194	0.325	0.300	-0.005		-0.025	-0.320

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a/ Indicates samples were put on roof Jan. 28, 1952 to July 7, 1953.
b/ Indicates samples were put on roof from Feb. 29, 1952 to July 7, 1953.
c/ Indicates samples were put on roof Feb. 7, 1952 to July 7, 1953.

TABLE 2. DIFFERENCES IN OPTICAL DENSITIES FOR ORIGINAL "GAFITE" SAMPLES

Sheet Numbers with Differences in Optical Densities Falling in the Following Ranges

Density Range of Differences	No. of Sheets	Percent of Total	Sheet Numbers
-0.060 to -0.064	1	0.3	351
-0.050 to -0.054	1	0.3	452
-0.040 to -0.044	2	0.5	9 467
-0.035 to -0.039	1	0.3	318
-0.025 to -0.029	4	1.0	391 422 437 535
-0.020 to -0.024	1	0.3	400
-0.015 to -0.019	11	2.8	220 233 237 329 350 393 439 508 513 553 558
-0.010 to -0.014	30	7.8	21 51 64 76 93 106 112 120 139 201 216 224 236 248 264 276 324 348 357 361 362 412 423 440 461 499 518 519 549 563
-0.005 to -0.009	52	13.5	5 11 22 23 69 74 75 77 82 87 116 125 130 133 155 167 211 213 215 234 240 251 268 295 302 304 312 313 321 328 330 356 364 371 388 389 407 425 435 443 445 457 490 494 556 557 559 569 572 574 588 592
-0.000 to -0.004	61	15.8	10 14 36 42 52 54 68 79 81 88 89 91 99 102 107 117 122 126 169 182 185 203 208 225 270 272 291 298 305 319 331 333 336 341 344 345 359 368 378 381 390 396 401 408 411 419 431 444 463 475 478 507 543 573 576 577 578 581 585 586 590
+0.000 to +0.004	115	29.9	1 2 3 4 6 7 8 15 19 20 37 38 40 41 43 48 57 58 63 65 66 67 70 71 72 73 78 83 94 96 104 105 109 115 121 128 129 131 142 143 149 152 157 161 163 179 188 210 219 222 226 227 235 242 250 259 260 266 274 275 277 280 307 310 323 325 335 337 349 355 358 360 380 383 385 386 392 394 397 404 406 409 418 442 446 449 455 456 480 481 484 487 497 498 501 510 514 515 521 522 527 529 530 532 542 544 548 554 561 567 568 580 582 584 589
+0.005 to +0.009	48	12.5	18 49 80 90 95 98 150 193 202 207 221 228 232 239 247 254 256 265 278 284 314 315 317 320 322 326 333 338 342 370 377 384 432 447 472 479 491 500 505 526 528 537 545 551 562 564 579 591
+0.010 to +0.014	25	6.5	103 110 118 127 174 230 327 334 363 398 421 434 469 488 492 502 504 509 511 516 523 533 546 552 571

TABLE 2. (Continued) DIFFERENCES IN OPTICAL DENSITIES FOR ORIGINAL "GAFITE" SAMPLES

Sheet Numbers with Differences in Optical Densities Falling in the Following Ranges

Density Range of Differences	No. of Sheets	Percent of Total	Sheet Numbers
+0.015 to +0.019	6	1.5	25 44 269 271 399 462
+0.020 to +0.024	5	1.3	506 547 550 565 587
+0.025 to +0.029	6	1.5	86 306 366 493 512 525
+0.030 to +0.034	3	0.8	55 249 263
+0.035 to +0.039	6	1.5	253 458 489 531 538 566
+0.040 to +0.044	2	0.5	84 286
+0.045 to +0.049	3	0.8	299 429 460
+0.060 to +0.064	1	0.3	438
+0.130 to +0.134	1	0.3	365

TABLE 3. CHANGES IN OPTICAL DENSITIES OF DARK STORAGE SAMPLES AFTER 1.5 YEARS

Sheet Numbers with Differences in Optical Densities Falling in the Following Ranges

Density Range of Differences	No. of Sheets	Percent of Total	Sheet Numbers
-0.045 to -0.049	2	0.5	237 295
-0.035 to -0.039	3	0.8	318 351 452
-0.030 to -0.034	1	0.3	349
-0.025 to -0.029	1	0.3	394
-0.020 to -0.024	4	1.0	78 239 253 284
-0.015 to -0.019	8	2.1	11 55 58 88 201 228 321 461
-0.010 to -0.014	25	6.5	2 3 9 14 19 43 51 66 72 75 96 99 104 106 112 139 149 182 234 248 310 312 329 449 463
-0.005 to -0.009	31	8.0	4 5 10 15 20 44 49 57 70 71 76 84 87 90 93 102 103 125 126 143 179 211 224 251 254 277 291 348 350 366 489
-0.000 to -0.004	40	10.4	7 8 18 38 40 48 52 65 80 81 89 98 107 109 110 115 116 120 121 122 185 202 225 235 247 268 270 272 274 278 280 305 307 313 319 322 336 360 370 423
+0.000 to +0.004	46	11.9	1 22 41 42 54 63 69 77 82 94 95 117 129 130 150 152 161 174 188 208 216 220 230 232 233 236 240 265 276 302 304 314 317 320 324 325 328 331 335 356 388 421 456 457 467 480
+0.005 to +0.009	41	10.6	6 21 36 67 68 83 127 128 131 133 155 163 167 203 221 226 256 259 266 269 271 275 286 299 323 332 333 338 342 361 362 363 380 390 396 399 406 407 418 419 462
+0.010 to +0.014	28	7.3	37 86 118 142 193 207 227 242 260 264 298 306 326 344 345 357 364 381 383 401 404 412 422 438 439 475 484 580
+0.015 to +0.019	14	3.6	23 212 222 315 327 337 341 355 377 384 425 435 447 488
+0.020 to +0.024	17	4.4	25 210 215 249 263 330 358 378 386 392 400 411 431 446 458 487 538
+0.025 to +0.029	11	2.8	64 79 219 250 371 391 398 440 442 460 529
+0.030 to +0.034	5	1.3	73 157 385 409 506
+0.040 to +0.044	11	2.8	91 105 169 359 434 443 445 478 526 530
+0.045 to +0.049	2	0.5	479 504
+0.050 to +0.054	4	1.0	389 393 429 472
+0.055 to +0.059	3	0.8	397 408 522
+0.060 to +0.064	2	0.5	334 432
+0.065 to +0.069	3	0.8	74 518 532
+0.070 to +0.074	1	0.3	444
+0.075 to +0.079	8	2.1	507 514 528 533 546 565 569 574
+0.080 to +0.084	4	1.0	469 505 535 537
+0.085 to +0.089	7	1.9	493 508 516 519 531 549 568

TABLE 3. (Continued) CHANGES IN OPTICAL DENSITIES OF DARK STORAGE SAMPLES AFTER 1.5 YEARS

Sheet Numbers with Differences in Optical Densities Falling in the Following Ranges

Density Range of Differences	No. of Sheets	Percent of Total	Sheet Numbers
+0.090 to +0.094	4	1.0	547 553 561 567
+0.095 to +0.099	3	0.8	437 491 515
+0.100 to +0.104	4	1.0	481 497 498 513
+0.105 to +0.109	3	0.8	490 551 562
+0.110 to +0.114	4	1.0	368 499 552 576
+0.115 to +0.119	2	0.5	502 559
+0.120 to +0.124	3	0.8	521 543 573
+0.125 to +0.129	4	1.0	365 492 525 571
+0.130 to +0.134	2	0.5	494 566
+0.135 to +0.139	2	0.5	523 577
+0.140 to +0.144	3	0.8	509 510 572
+0.145 to +0.149	3	0.8	544 550 582
+0.150 to +0.154	1	0.3	554
+0.155 to +0.159	3	0.8	527 545 579
+0.160 to +0.164	2	0.5	500 501
+0.165 to +0.169	1	0.3	564
+0.170 to +0.174	3	0.8	511 563 581
+0.180 to +0.184	2	0.5	512 548
+0.185 to +0.189	2	0.5	542 558
+0.190 to +0.194	2	0.5	557 578
+0.210 to +0.214	1	0.3	588
+0.220 to +0.224	1	0.3	589
+0.230 to +0.234	2	0.5	590 591
+0.240 to +0.244	1	0.3	556
+0.245 to +0.249	2	0.5	585 586
+0.250 to +0.254	1	0.3	587
+0.260 to +0.264	1	0.3	584
+0.300 to +0.304	1	0.3	592

TABLE 4. DIFFERENCES IN OPTICAL DENSITIES BETWEEN DARK STORAGE SAMPLE AND ROOF EXPOSURE SAMPLE AFTER 1.5 YEARS

Sheet Numbers with Changes in Optical Densities Falling in the Following Ranges

Density Range of Differences	No. of Sheets	Percent of Total	Sheet Numbers
-0.300 to -0.324	1	0.2	592
-0.200 to -0.224	1	0.2	584
-0.150 to -0.174	3	0.8	463 585 586
-0.100 to -0.124	6	1.6	434 437 438 440 449 501
-0.075 to -0.099	10	2.7	351 357 368 491 527 554 578 581 588 589
-0.050 to -0.074	6	1.6	355 443 467 553 558 591
-0.025 to -0.049	20	5.2	360 363 385 400 455 456 461 472 479 492 506 516 521 525 533 545 571 577 579 587
-0.000 to -0.024	18	4.7	278 348 349 350 358 359 365 366 370 439 444 445 490 493 494 502 523 590
+0.000 to +0.024	35	9.1	25 44 55 79 157 163 280 310 337 338 345 356 361 364 371 377 386 389 392 393 412 421 435 442 446 452 469 498 504 505 530 538 556 557 582
+0.025 to +0.049	33	8.5	49 58 73 88 91 105 139 143 155 167 185 188 284 291 298 317 323 333 334 390 391 397 429 431 481 507 509 518 519 531 567 572 580
+0.050 to +0.074	36	9.3	8 67 68 74 77 81 82 86 93 104 106 133 142 150 161 169 260 272 286 307 345 362 398 401 406 408 411 422 462 497 514 515 542 546 564 573
+0.075 to +0.099	41	10.6	22 23 40 43 48 51 65 66 71 75 80 84 87 89 96 98 112 242 254 259 271 275 313 320 321 329 332 335 342 388 409 418 432 499 548 550 561 562 563 568 574
+0.100 to +0.124	38	9.9	2 42 52 54 63 70 78 83 90 94 107 110 120 122 182 193 201 207 256 266 302 318 327 328 330 331 336 396 407 419 425 457 460 475 487 552 559 569
+0.125 to +0.149	23	6.0	18 38 64 72 76 95 102 109 117 121 129 179 305 324 379 381 399 404 423 447 478 566 576
+0.150 to +0.174	29	7.5	4 14 19 37 41 57 99 103 116 118 128 131 149 174 202 203 213 226 240 249 251 253 274 295 325 326 380 544 551
+0.175 to +0.199	20	5.2	3 9 10 69 127 130 222 247 269 299 319 322 480 500 508 510 547 549 565
+0.200 to +0.224	16	4.2	11 15 115 152 219 221 250 263 277 312 314 358 488 511 513 528
+0.225 to +0.049	10	2.7	7 20 21 36 224 248 276 304 489 512
+0.250 to +0.274	14	3.6	1 208 211 215 220 225 236 237 265 268 270 315 383 384

TABLE 4. (Continued) DIFFERENCES IN OPTICAL DENSITIES BETWEEN DARK STORAGE SAMPLE AND ROOF EXPOSURE SAMPLE AFTER 1.5 YEARS

Sheet Numbers with Changes in Optical Densities Falling in the Following Ranges

Density Range of Differences	No. of Sheets	Percent of Total	Sheet Numbers
+0.275 to +0.299	9	2.3	125 126 216 230 233 234 235 306 526
+0.300 to +0.324	3	0.8	6 232 264
+0.325 to +0.349	3	0.8	239 537 543
+0.350 to +0.374	3	0.8	5 522 532
+0.375 to +0.399	2	0.5	341 529
+0.400 to +0.424	3	0.8	210 228 535
+0.425 to +0.449	1	0.2	227
+0.450 to +0.474	1	0.2	394

the dark storage samples but a few samples were actually more yellow on dark storage than on roof exposure.

1.3.4 Final Yellowing Index of Samples

While the previous figures dealt with changes in optical density or yellowing index, Figure 6 plotted from Table 5, lists the final yellowing index values for all the samples after 1.5 years of roof exposure irrespective of the amount of yellow in the sample before roof exposure. This figure indicates that although the majority of the samples possessed quite a bit of yellow color after roof exposure, there were a few samples which possessed only trace amounts of yellow color after 1.5 years of roof exposure at Easton, Pa. The history of these more light stable samples was investigated, and is reported in the next section.

1.3.5 Changes in Yellowing Index of "Gafite" Samples on Roof Exposure

Figure 7 presents the data assembled in Table 6 on the changes in the yellowing index of "Gafite" samples on roof exposure for 1.5 years at Easton, Pa. This figure illustrates that there were a few "Gafite" samples which actually became less yellow on roof exposure and that there were quite a few samples which changed very slightly.

The results presented in this figure and the preceding Figures 4, 5, and 6 hardly could be accounted for if methyl α-chloroacrylate polymer itself was unstable to light, but they could be explained if the cause of the yellowing on light exposure were small amounts of impurities present in the polymer. Since most of the monomer employed for casting these sheets possessed a purity of 99.0 mole percent or better, all traces of impurities probably have to be eliminated from the monomer and polymer to achieve maximum light stability. This approach toward increasing the light stability of "Gafite" sheets was investigated extensively.

1.3.6 Visual Color Observations on "Gafite" Samples

In Table 7 are listed the less yellow sheets after roof exposure as ranked by eye with the sheets increasing in yellow color proceeding down each column and with the yellow color of the sheets increasing proceeding across the columns. The sheet whose number was at the bottom of each column appeared less yellow than the sheet whose number appeared at the top of the next column to the right. For example, Sheet 349 in Column A appeared less yellow to the eye than Sheet 385 in Column B.

1.3.7 Visual Observation of Color Changes in "Gafite" Samples

In Table 8 are listed the sheet numbers of those sheets whose dark storage sample and roof exposure sample appeared very little different to the eye. The samples are ranked in order according to the difference in color between the two types of samples from each sheet after 1.5 years of being on test. The difference in color increases as one goes down each column and as one moves to the right in columns. Samples of sheets with number at bottom in each column exhibit less color difference between samples than do samples of sheet with number at top in next column to the right. For example, samples of Sheet 385 in Column A show less color difference to the eye than do samples of Sheet 348 in Column B.

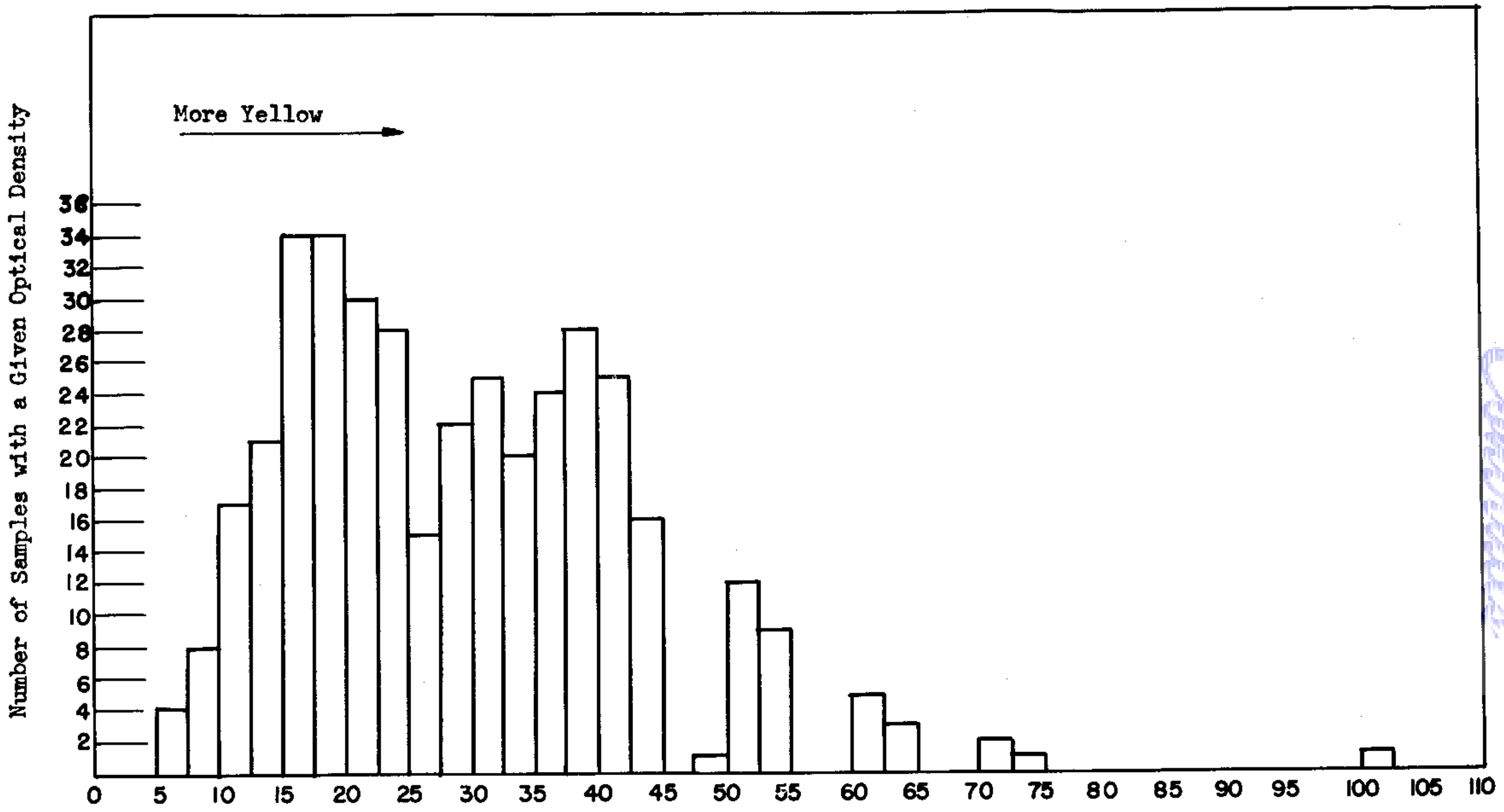

FIGURE 6. FINAL OPTICAL DENSITIES OF 385 "GAFITE" SAMPLES AFTER 1.5 YEARS ROOF EXPOSURE AT EASTON, PA.

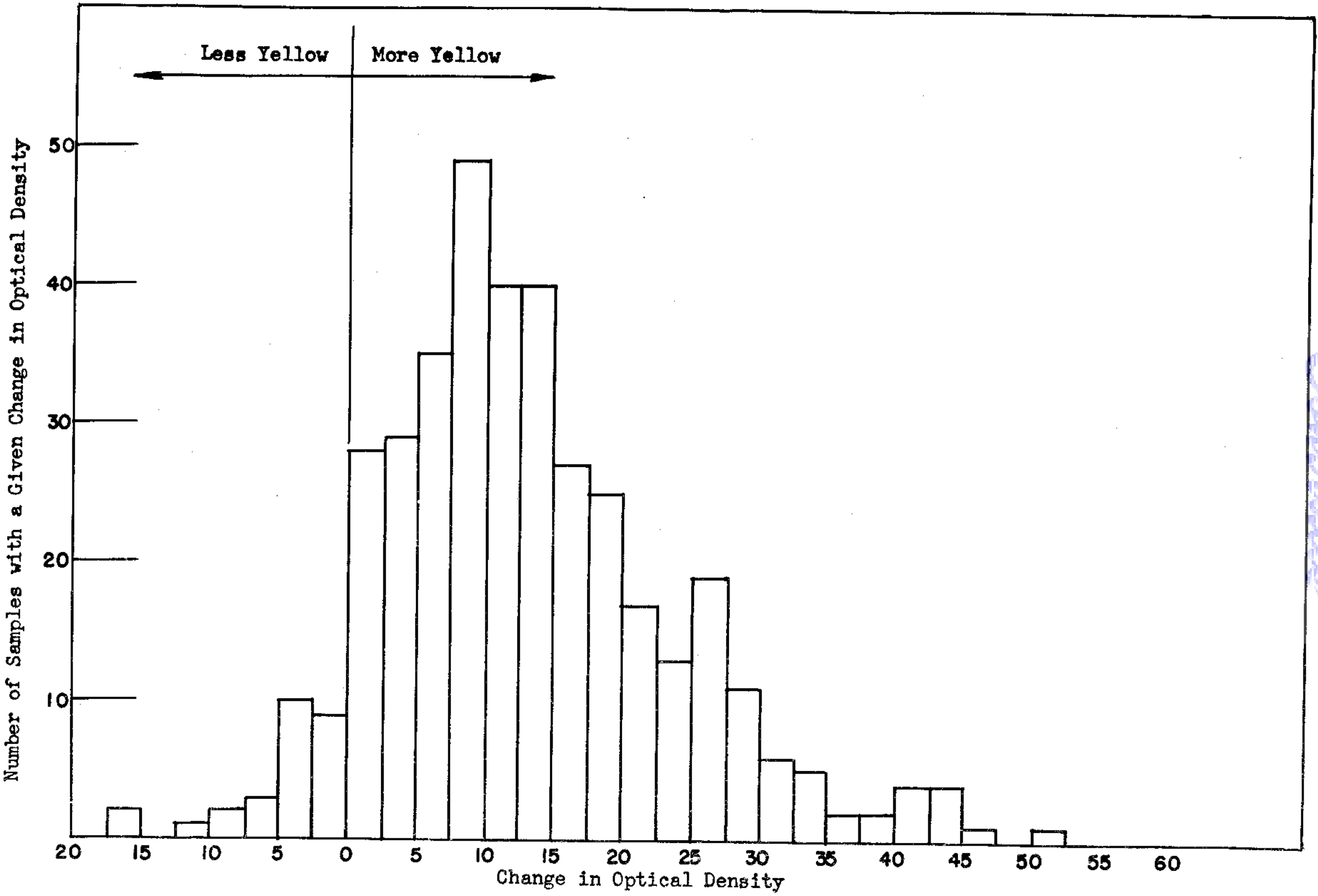

FIGURE 7. CHANGES IN OPTICAL DENSITIES OF 385 "GAFITE" SAMPLES ON 1.5 YEARS' ROOF EXPOSURE AT EASTON, PA.

TABLE 5. FINAL OPTICAL DENSITIES OF "GAFITE" SAMPLES AFTER 1.5 YEARS' ROOF EXPOSURE AT EASTON, PA.

Sheet Numbers with Changes in Optical Densities Falling in the Following Ranges

Density Range of Differences	No. of Sheets	Percent of Total	Sheet Numbers
0.050 to 0.074	4	1.0	49 143 185 310
0.075 to 0.099	8	2.1	55 155 157 163 167 188 334 338
0.100 to 0.124	17	4.4	8 44 58 65 66 67 77 79 81 84 88 96 142 150 291 307 317
0.125 to 0.149	21	5.4	40 48 51 63 68 73 75 78 82 86 91 93 161 272 278 280 286 313 333 335 421
0.150 to 0.174	34	8.8	25 43 52 54 70 71 76 80 83 87 89 90 94 98 104 105 106 112 120 122 169 182 201 207 242 298 320 321 327 329 332 336 398 435
0.175 to 0.199	34	8.8	2 18 19 22 23 38 42 72 74 95 99 102 107 110 121 133 139 149 193 203 259 260 266 302 318 324 328 331 385 390 397 406 412 440
0.200 to 0.224	30	7.8	4 10 14 37 41 57 64 103 109 117 118 128 129 131 179 202 271 275 295 305 323 326 330 389 391 392 393 401 438 475
0.225 to 0.249	28	7.3	3 9 11 69 116 127 130 174 240 284 322 325 342 388 399 400 408 409 411 418 434 442 443 444 461 472 516 580
0.250 to 0.274	15	3.9	15 115 152 314 381 419 422 429 431 437 445 446 455 463 531
0.275 to 0.299	22	5.7	7 221 249 274 299 304 312 319 344 368 377 378 396 404 407 423 425 439 447 467 501 571
0.300 to 0.324	25	6.5	20 21 36 126 208 219 226 315 364 380 432 456 462 479 487 527 533 567 568 577 578 584 588 589 592
0.325 to 0.349	20	5.2	1 125 236 251 265 269 276 277 361 371 386 478 502 518 545 558 574 576 587 591
0.350 to 0.374	24	6.2	215 216 220 222 230 235 345 359 362 363 365 452 493 494 498 514 519 530 554 569 572 573 581 586
0.375 to 0.399	28	7.3	6 224 225 232 234 263 268 270 337 358 360 366 384 460 469 481 484 504 506 515 521 523 553 556 579 582 585 590
0.400 to 0.424	25	6.5	5 211 213 233 239 250 256 306 350 357 370 383 449 490 491 497 525 538 546 549 557 559 561 562 565

TABLE 5. (Continued) FINAL OPTICAL DENSITIES OF "GAFITE" SAMPLES AFTER 1.5 YEARS' ROOF EXPOSURE AT EASTON, PA.

Sheet Numbers with Changes in Optical Densities Falling in the Following Ranges

Density Range of Differences	No. of Sheets	Percent of Total	Sheet Numbers
0.425 to 0.449	16	4.2	247 254 264 348 349 351 355 480 488 509 542 544 551 552 564 566
0.475 to 0.499	1	0.3	492
0.500 to 0.524	12	3.1	210 253 356 458 489 499 528 529 537 547 548 563
0.525 to 0.549	9	2.3	227 248 457 507 522 526 532 543 550
0.600 to 0.624	5	1.3	228 237 341 510 535
0.625 to 0.649	3	0.8	500 511 513
0.700 to 0.724	2	0.5	505 508
0.725 to 0.749	1	0.3	512
1.000 to 1.024	1	0.3	394

TABLE 6. CHANGES IN OPTICAL DENSITIES OF "GAFITE" SAMPLES ON 1.5 YEARS' ROOF EXPOSURE AT EASTON, PA.

Sheet Numbers with Changes in Optical Densities Falling in the Following Ranges

Density Range of Differences	No. of Sheets	Percent of Total	Sheet Numbers
-0.150 to -0.174	2	0.5	438 463
-0.100 to -0.124	1	0.3	449
-0.075 to -0.099	2	0.5	434 440
-0.050 to -0.074	3	0.8	351 355 357
-0.025 to -0.049	10	2.6	55 349 360 363 366 443 456 461 506 592
-0.000 to -0.024	9	2.3	278 310 348 365 370 385 421 467 538
+0.000 to +0.024	28	7.3	25 44 49 58 88 143 163 280 284 286 334 337 338 350 356 358 359 361 377 386 400 437 439 452 455 472 479 491
+0.025 to +0.049	29	7.5	79 84 86 104 139 157 185 188 291 298 317 327 344 364 368 371 392 412 429 435 442 445 446 493 501 516 530 533 538
+0.050 to +0.074	35	9.1	8 43 67 73 77 81 82 96 105 106 142 150 155 161 167 254 260 272 307 320 321 333 345 362 390 393 398 431 444 462 525 527 553 554 584
+0.075 to +0.099	49	12.7	2 23 40 48 51 65 66 68 71 75 78 80 87 89 90 91 93 98 112 133 201 259 271 275 313 329 332 335 342 389 391 397 401 406 411 418 460 461 490 492 502 505 521 531 571 577 581 585 586
+0.100 to +0.124	40	10.4	22 42 52 54 63 70 83 94 107 110 120 121 122 169 182 193 242 256 266 295 318 327 331 336 388 399 408 422 457 475 494 498 507 518 519 523 545 567 578 579
+0.125 to +0.149	40	10.4	14 18 19 38 72 74 76 95 99 102 103 109 117 128 129 149 179 207 302 324 328 330 381 396 407 409 419 423 425 432 447 481 487 504 514 546 558 582 588 589
+0.150 to +0.174	27	7.0	3 4 10 37 41 57 116 118 131 174 202 203 247 249 251 274 299 305 322 325 326 378 380 404 497 509 515
+0.175 to +0.199	25	6.5	11 64 69 127 130 213 226 240 253 263 269 319 458 478 480 489 499 561 562 568 572 573 574 587 591
+0.200 to +0.224	17	4.4	9 15 115 152 221 222 277 312 314 484 488 550 552 557 564 569 590
+0.225 to +0.249	13	3.4	7 20 219 224 237 248 250 276 304 542 559 565 566
+0.250 to +0.274	19	4.9	1 21 36 208 211 225 230 236 265 268 270 306 384 547 548 551 556 563 576
+0.275 to +0.299	11	2.9	125 126 215 216 220 234 235 315 383 508 528
+0.300 to +0.324	6	1.6	6 232 233 239 544 549

TABLE 6. (Continued) CHANGES IN OPTICAL DENSITIES OF "GAFITE" SAMPLES ON 1.5 YEARS' ROOF EXPOSURE AT EASTON PA.

Sheet Numbers with Changes in Optical Densities Falling in the Following Ranges

Density Range of Differences	No. of Sheets	Percent of Total	Sheet Numbers
+0.325 to +0.349	5	1.3	264 500 510 513 526
+0.350 to +0.374	2	0.5	5 511
+0.375 to +0.399	2	0.5	228 512
+0.400 to +0.424	4	1.0	341 522 529 537
+0.425 to +0.449	4	1.0	210 227 394 532
+0.450 to +0.474	1	0.3	543
+0.500 to +0.524	1	0.3	535

TABLE 7. VISUAL COLOR OBSERVATIONS ON "GAFITE" SAMPLES AFTER ROOF EXPOSURE FOR 1.5 YEARS AT EASTON, PA.

Colorless to Very Light Yellow A	Very Light Yellow B	Very Light Yellow to Light Yellow C	Light Yellow D	Light Yellow E	Light Yellow F	Yellow G	Yellow H	Yellow I	Yellow J	Yellow K	Yellow L	Yellow M	Yellow N
143	385	77	370	71	68	10	9	94	392	418	115	268	249
142	88	201	8	72	242	19	323	182	391	411	127	527	381
49	317	344	364	43	438	112	302	179	362	463	174	571	380
155	56	260	363	327	435	106	305	193	393	422	116	442	399
167	55	259	358	70	336	90	271	328	421	419	295	446	404
163	291	280	366	335	121	152	388	64	398	434	299	487	423
185	75	351	368	342	120	149	401	51	397	467	117	478	431
338	74	357	371	96		390	63	42	429	407	130	274	425
161	73	356	377	207		284	109	44	406	409	131	265	378
337	86	361	202	169		208	107	41	405	408	118	234	580
188	84	355	67	321		133	105	38	444	445	76	304	
58	82	359	66	98		275	104	37	475	443	128	396	
91	81	386	65	25		266	103	22	472	455	57	319	
310	79	298	333	23		389	110	48	18	479	69	315	
157	93	278	365	331		318	102	87	437	469	129	314	
89	286	313	332	330		122	99	52	440	460	126	312	
334	78	345	272	324		40	95	54	439	456	325	322	
350	150	348	360	329			83	80	240	36	400	125	
349	307		203	320					412	139	326		

TABLE 8. VISUAL COLOR OBSERVATIONS ON "GAFITE" SAMPLES WITH LEAST COLOR CHANGE BETWEEN ROOF EXPOSURE SAMPLE AND DARK STORAGE SAMPLE AFTER 1.5 YEARS

No Change or Almost None A	Very Slight Change B	Slight Change C
143	348 [a]	358
142	359	323
49	351	444
155	357 [a]	437 [a]
167	356 [a]	439
163	361	440 [a]
185	355 [a]	438 [a]
338	386	435
161	280	389
337	365	284
188	370 [a]	412
58	364	390
91	377	401
310	278	400
157	360 [a]	445
89	344	443
334	363 [a]	434
350	368	429
349	371	472
385	366 [a]	580
		592 [a]

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[a] Possibly less yellow after exposure.

1.4 Heat Stability of "Gafite" Samples

In Table 9 which lists graphically the optical density values recorded numerically in Table 1, there is also listed the color appearance of the samples to the eye and whether the samples showed any crazing. Most of the samples showed no crazing. Along with the above information, there is listed the heat stability values which were obtained on the sheets shortly after they were cast. (Recent noncontractual work at GAF indicates that these heat stability values decrease for some sheets on storage.) The heat stability values were obtained by immersing a (1 1/2 in. x 3 in. x sample thickness) specimen in a Dow-Corning Type 550 Silicone Oil Bath maintained at 160°C. The specimen was examined with the naked eye at the end of 1/2 an hour in this bath for the presence of any bubbles in the plastic. The sample was immersed and heated for consecutive 1/2 hour periods until bubbles were present in the plastic. The heat stability value is that maximum number of 1/2 hour periods of heating at 160°C at the end of which there were no bubbles in the test specimen. The specimen contained bubbles visible to the naked eye at the end of the next 1/2 hour period of heating following the number of 1/2 hour heat stability periods.

There probably are several variables in the preparation of cast "Gafite" sheet which are responsible for changes in the heat stability of "Gafite" sheet. Because it frequently was difficult to control all of the variables in sheet casting, the value of a specific treatment toward improving the heat stability of "Gafite" sometimes was established only after comparison of a number of samples prepared using the process alteration with a number of samples prepared in other ways. A considerable bit of noncontractual research has established the value of treating methyl α-chloroacrylate monomer employed for sheet casting with phosphorus pentoxide (P_2O_5) prior to casting. It was believed that this process merited study under this contract as a means of obtaining "Gafite" possessing improved heat stability. The evidence supporting this conclusion is presented in Figures 8, 9, and 10. Figure 8 summarizes the range of heat stabilities obtained on all the "Gafite" sheets tested to date. This figure illustrates the great variance in results of heat stability tests. In Figure 9 are presented the heat stability results on all "Gafite" sheets, the monomer for which had not been treated with phosphorus pentoxide (P_2O_5). In Figure 10 are presented the heat stability results on all "Gafite" sheets, the monomer for which had been treated with phosphorus pentoxide (P_2O_5). The marked shift toward increased heat stability when the process of treatment with phosphorus pentoxide was employed is evident. It was thought that control of other variables causing lower heat stability coupled with further development of the process of treatment with phosphorus pentoxide might offer a way of obtaining "Gafite" sheets possessing consistently high heat stabilities.

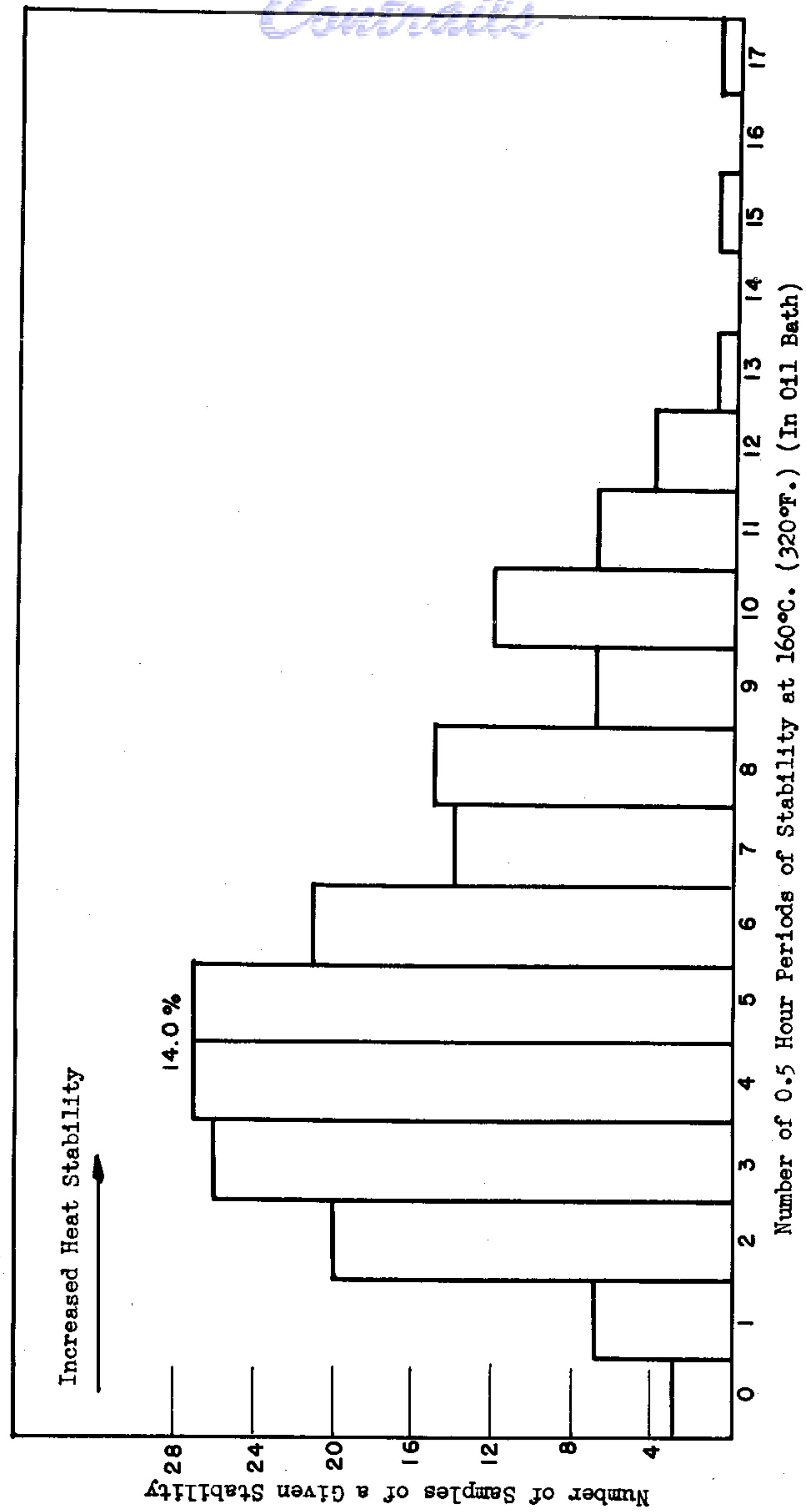


FIGURE 8. HEAT STABILITY OF "GAFITE" AT 160°C. (193 SAMPLES)

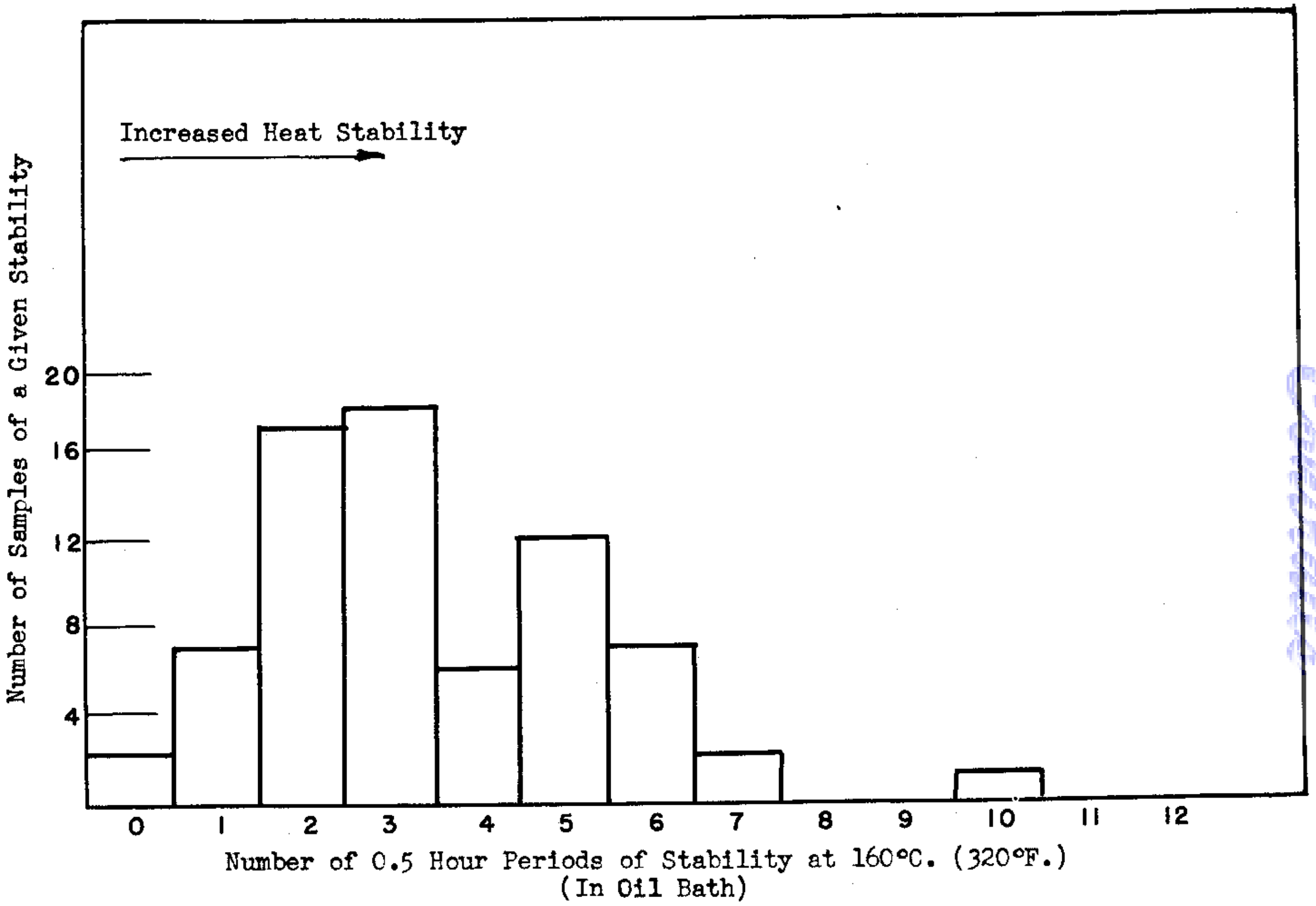


FIGURE 9. HEAT STABILITY OF "GAFITE" at 160°C.

72 SAMPLES--MONOMER NOT TREATED WITH P_2O_5

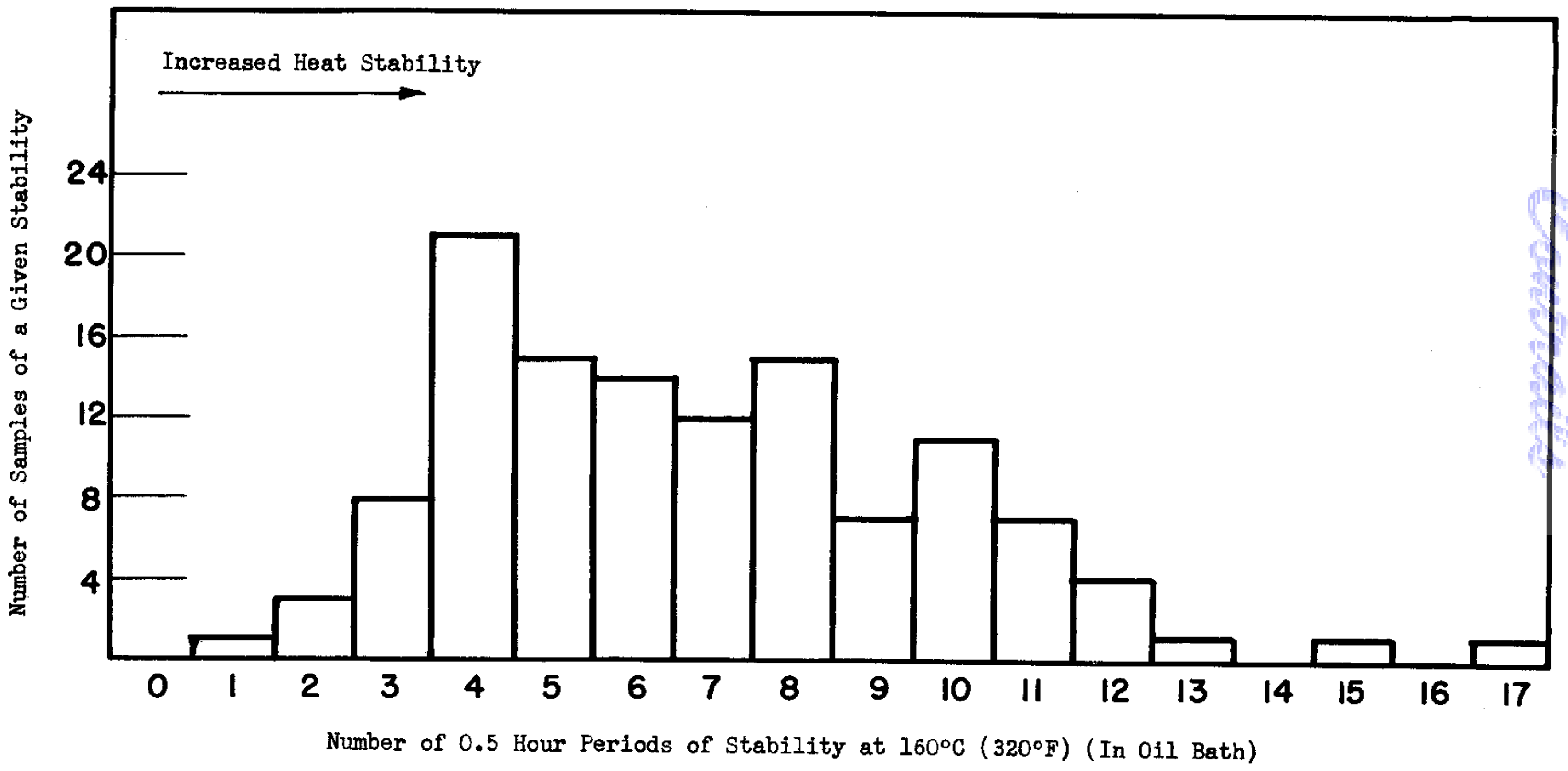


FIGURE 10. HEAT STABILITY OF "GAFITE" AT 160°C

121 SAMPLES-MONOMER TREATED WITH P_2O_5

CODE TO TABLE 9

Light and Heat Stability Tests on "Gafite"

Density Readings for Each Sample:

First (Top) Line is density value for original dark storage sample.
Second Line is density value for sample after dark storage.
Third Line is density value for original roof exposure sample.
Fourth (Bottom) Line is density value for sample after roof exposure.

Color Designations:

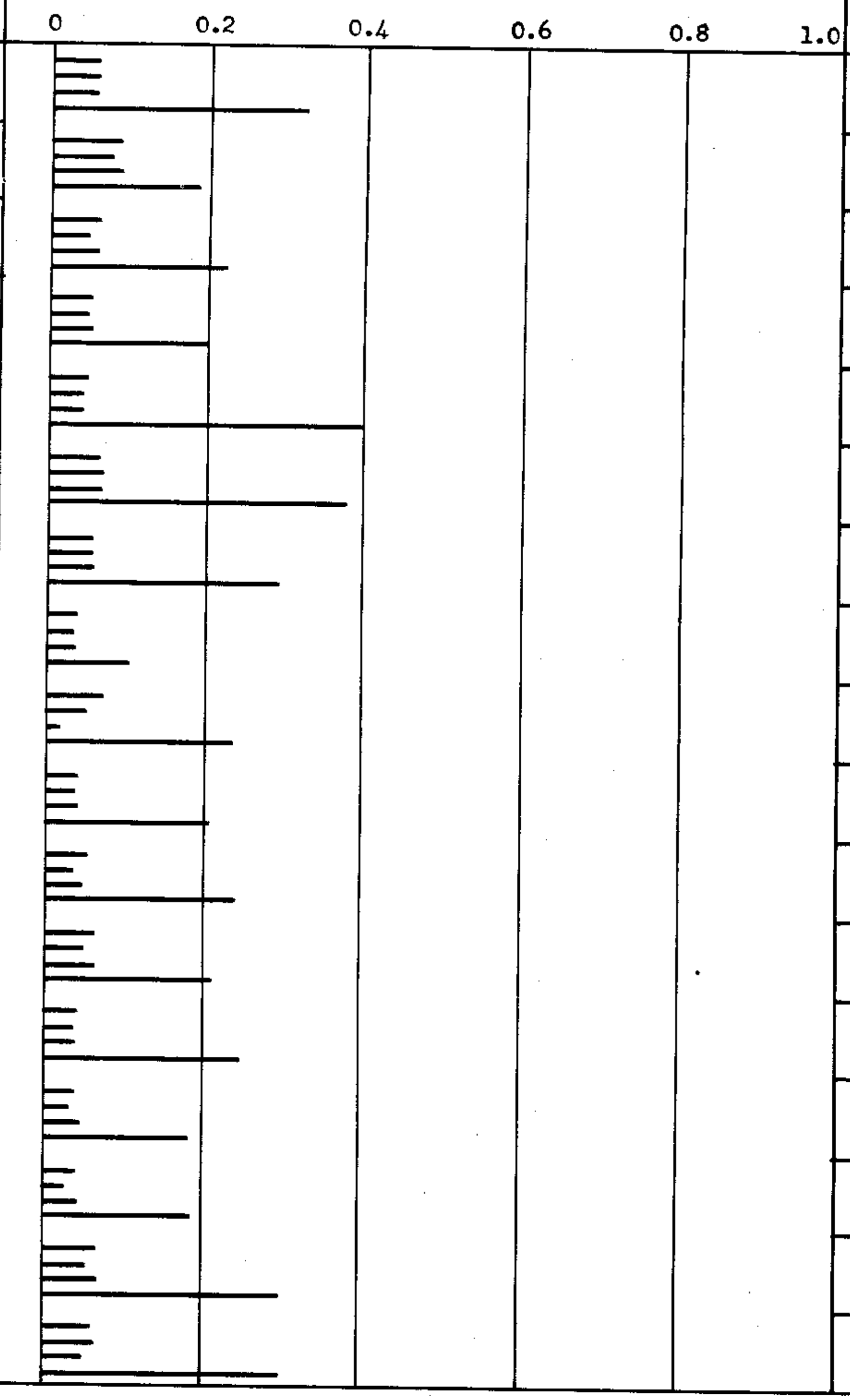
C - Colorless
VLY - Very light yellow
LY - Light yellow
Y - Yellow
VY - Very yellow
DY - Dark yellow

Crazing Designations (for Roof Exposure Samples):

0 - No crazing
1 - Light crazing
2 - Severe crazing

TABLE 9. LIGHT AND HEAT STABILITY TESTS ON "GAFITE"

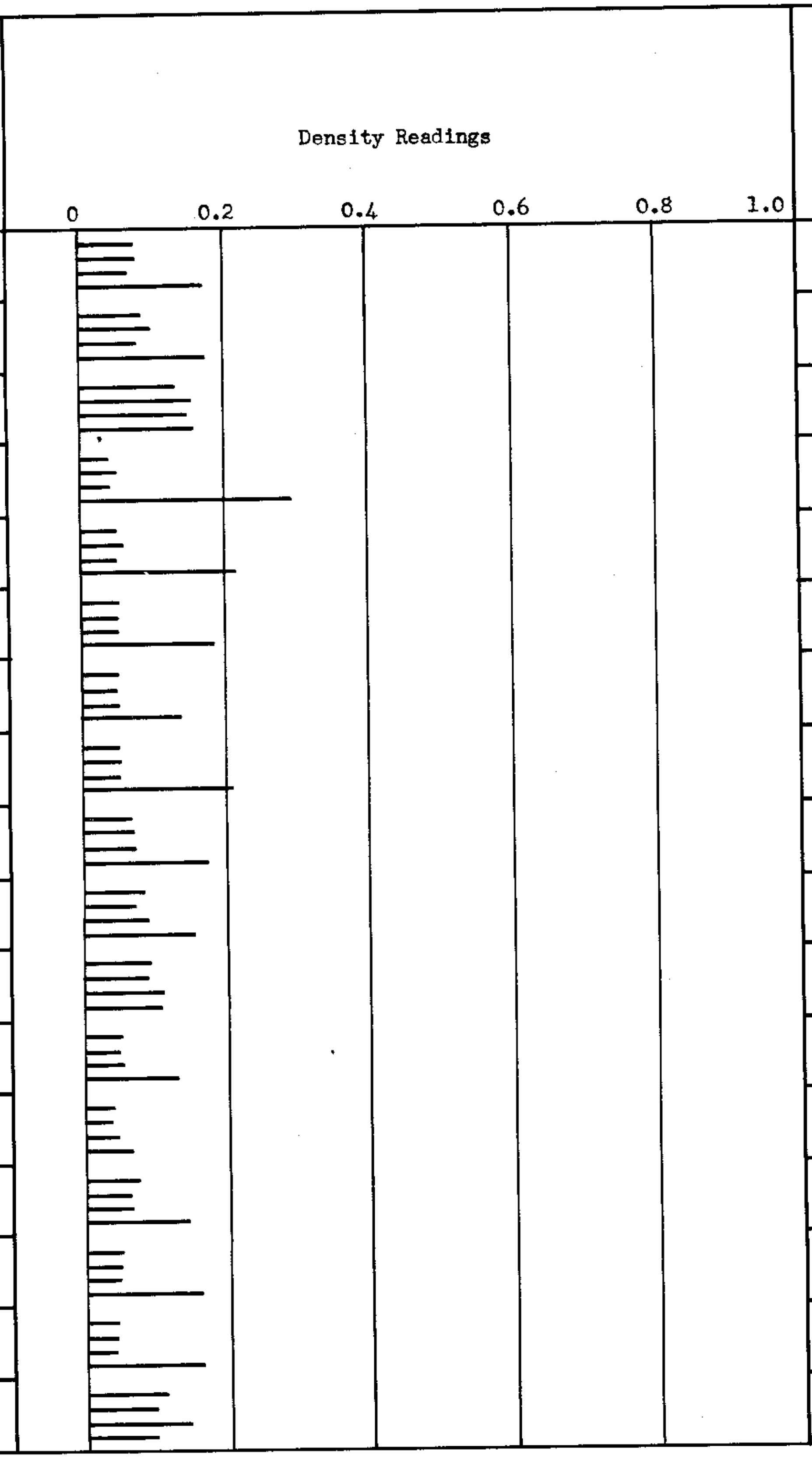
Exposed January 21, 1952 to July 7, 1953



Sample Number	Density Readings	Crazing	Color After		Heat Stability
			Dark Storage	Roof Exposure	
1		0	VLY	Y	
2		0	VLY	Y	
3		1	C	Y	
4		1	C	Y	
5		1	C	DY	
6		2	VLY	Y	
7		0	C	Y	
8		2	C	LY	
9		0	C	LY	
10		0	C	LY	
11		0	C	LY	
14		0	C	LY	
15		0	C	LY	
18		0	C	LY	
19		1	C	LY	
20		1	C	Y	
21		1	C	Y	

TABLE 9. (Continued) LIGHT AND HEAT STABILITY TESTS ON "GAFITE"

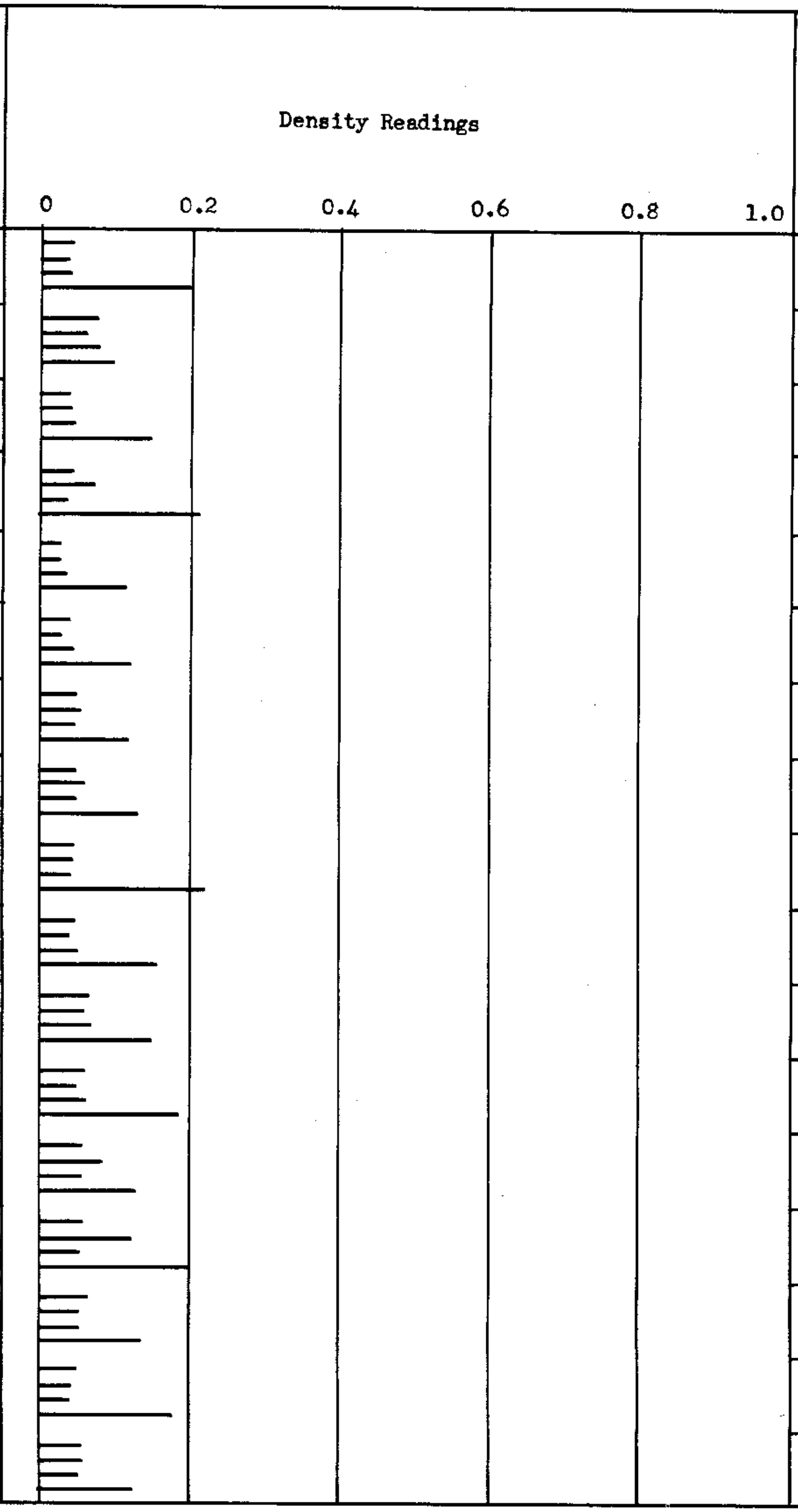
Exposed January 21, 1952 to July 7, 1953



Sample Number	Crazing	Color After: Dark Storage	Color After: Roof Exposure	Heat Stability
22	1	C	LY	
23	1	VLY	LY	
25	2	LY	LY	
36	2	C	LY	
37	1	C	LY	
38	1	C	LY	
40	1	C	LY	
41	0	C	LY	
42	2	C	LY	
43	1	C	LY	
44	2	VLY	LY	
48	1	C	LY	
49	1	C	VLY	
51	1	VLY	LY	
52	1	C	LY	
54	0	C	Y	
55	2	C	VLY	

TABLE 9. (Continued) LIGHT AND HEAT STABILITY TESTS ON "GAFITE"

Exposed January 21, 1952 to July 7, 1953



Sample Number	Crazing	Color After Dark Storage	Color After Roof Exposure	Heat Stability
57	1	C	LY	
58	2	C	VLY	
63	1	C	LY	
64	1	C	LY	
65	2	C	VLY	
66	2	C	LY	
67	2	C	LY	
68	2	C	LY	
69	1	C	Y	
70	1	C	LY	
71	1	C	LY	
72	2	C	VLY	
73	2	C	VLY	
74	1	C	VLY	
75	1	C	VLY	
76	2	C	LY	
77	0	C	LY	

TABLE 9. (Continued) LIGHT AND HEAT STABILITY TESTS ON "GAFITE"

Exposed January 21, 1952 to July 7, 1953

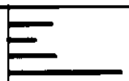
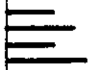
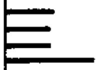
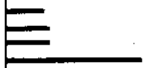
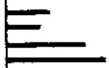
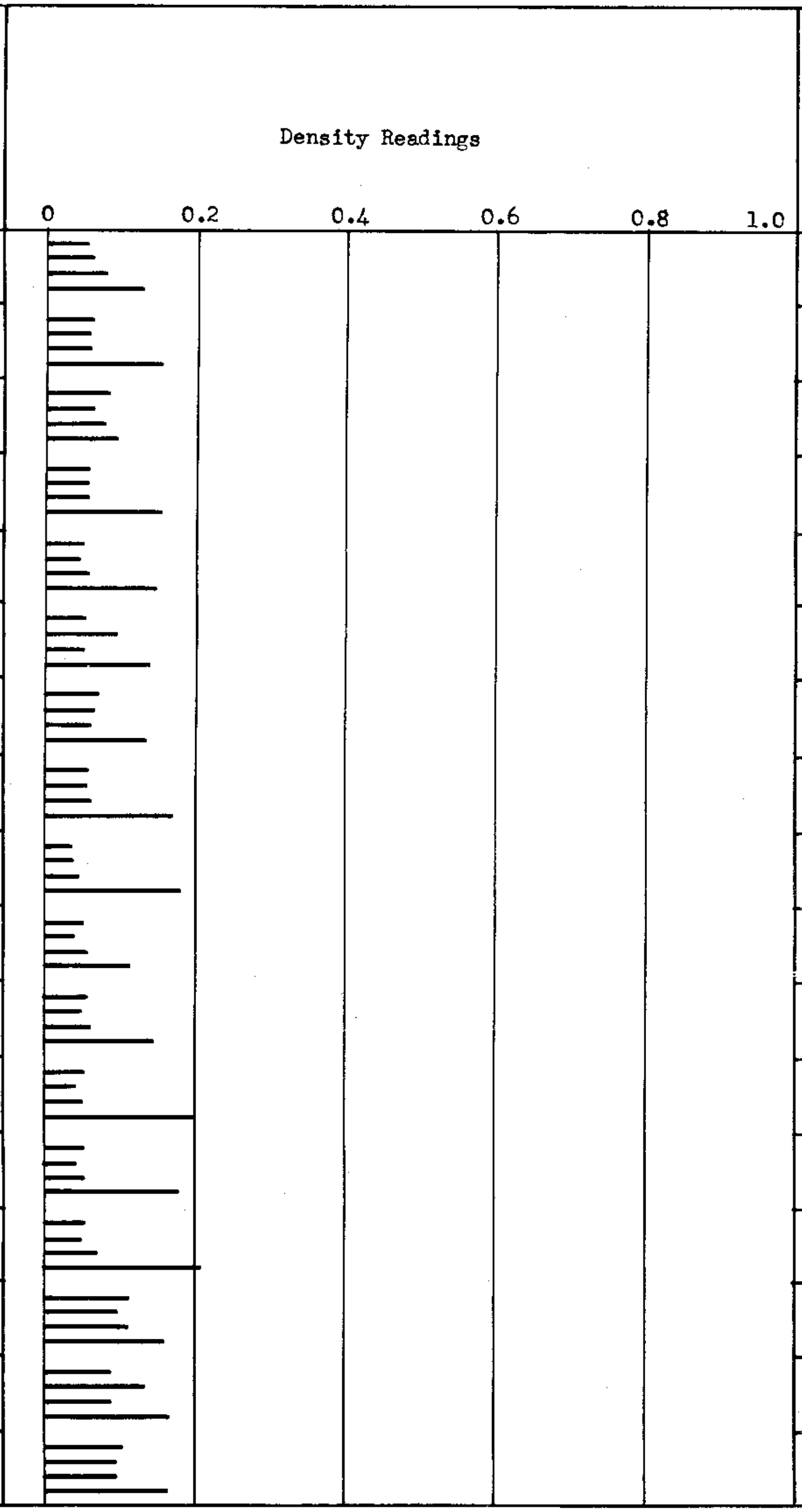
Sample Number	Density Readings 0.0 0.2 0.4 0.6 0.8 1.0	Crazing	Color After Dark Storage	Color After Roof Exposure	Heat Stability
78		1	C	LY	
79		1	C	VLY	
80		0	C	Y	
81		2	C	LY	
82		2	C	VLY	
83		1	C	Y	
84		2	C	LY	

TABLE 9. (Continued) LIGHT AND HEAT STABILITY TESTS ON "GAFITE"

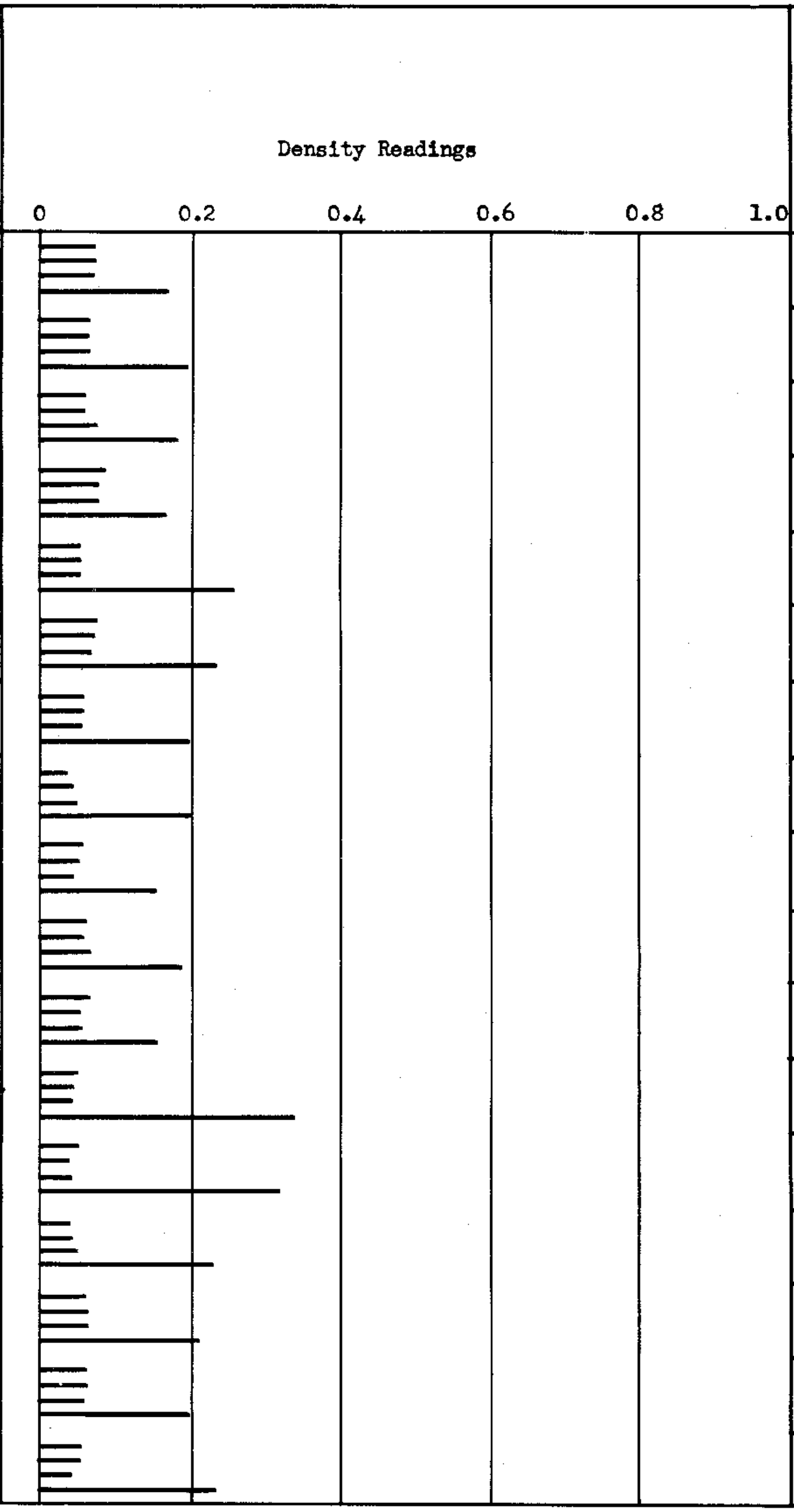
Exposed January 23, 1952 to July 7, 1953



Sample Number	Crazing	Color After Dark Storage	Color After Roof Exposure	Heat Stability
86	1	C	VLY	
87	0	C	Y	
88	1	C	VLY	
89	1	C	VLY	
90	0	C	Y	
91	2	C	VLY	
93	1	C	LY	
94	0	C	LY	
95	1	C	LY	
96	1	C	LY	
98	1	C	LY	
99	1	C	LY	
102	1	C	LY	
103	1	C	LY	
104	2	LY	Y	
105	0	C	Y	
106	2	VLY	Y	

TABLE 9. (Continued) LIGHT AND HEAT STABILITY TESTS ON "GAFITE"

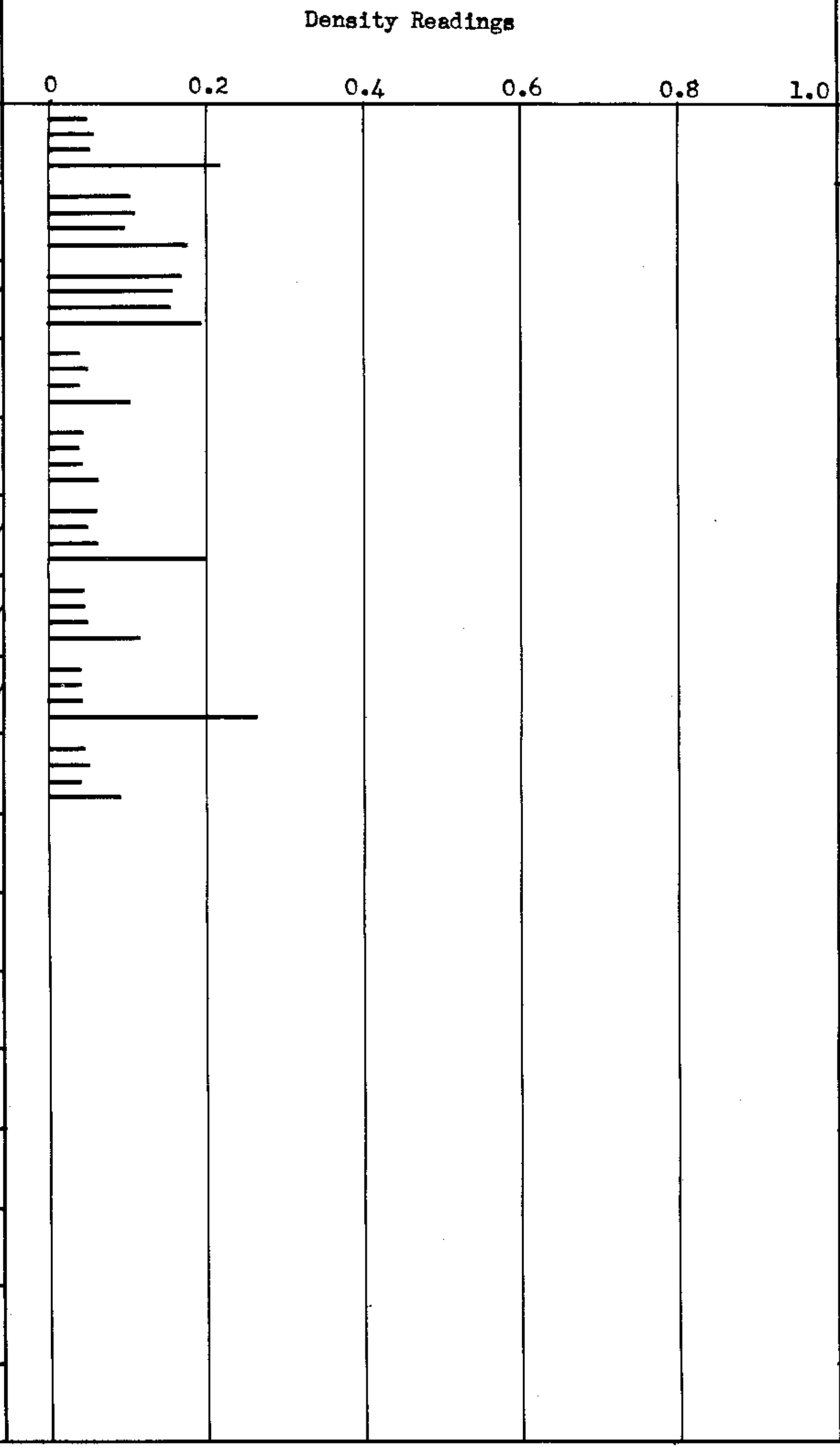
Exposed January 23, 1952 to July 7, 1953



Sample Number	Crazing	Color After Dark Storage	Color After Roof Exposure	Heat Stability
107	0	C	Y	
109 [a]	1	C	Y	
110 [a]	1	C	Y	
112 [a]	0	C	Y	
115	0	C	DY	
116	0	VLY	DY	
117	0	C	DY	
118	0	C	Y	
120	0	C	Y	
121	0	C	Y	
122	0	C	Y	
125 [a]	0	C	Y	
126	2	VLY	Y	
127	1	C	Y	
128	1	C	Y	
129	1	C	Y	
130	1	VLY	Y	

TABLE 9. (Continued) LIGHT AND HEAT STABILITY TESTS ON "GAFITE"

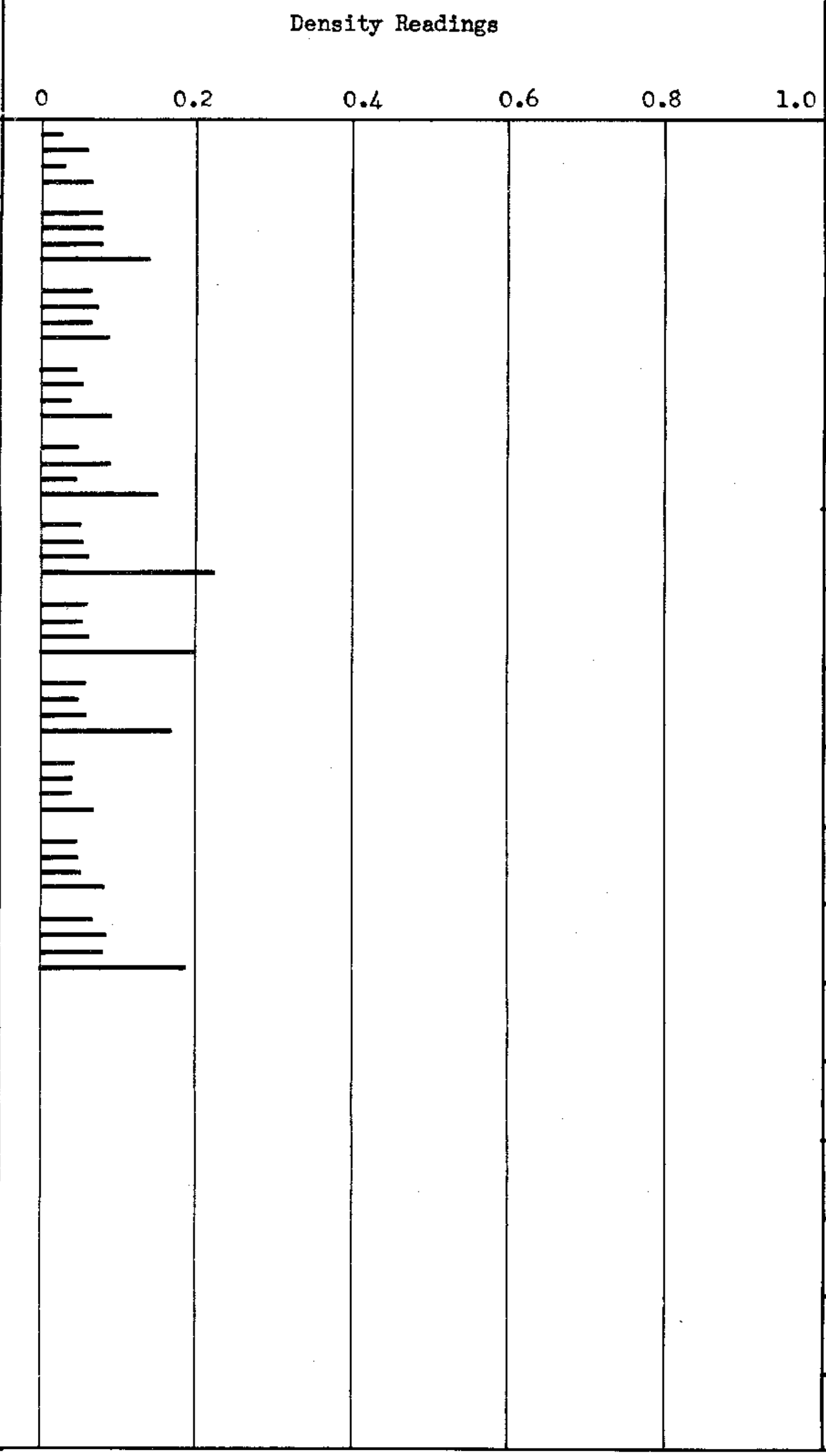
Exposed January 23, 1952 to July 7, 1953



Sample Number	Density Readings	Crazing	Color After Dark Storage	Color After Roof Exposure	Heat Stability
131		1	VLY	Y	
133		1	LY	DY	
139 a/		0	Y	Y	
142		2	C	C	
143		2	C	C	
149 b/		0	C	Y	
150 a/		0	VLY	LY	
152 b/		0	C	LY	
155		1	C	C	

TABLE 9. (Continued) LIGHT AND HEAT STABILITY TESTS ON "GAFITE"

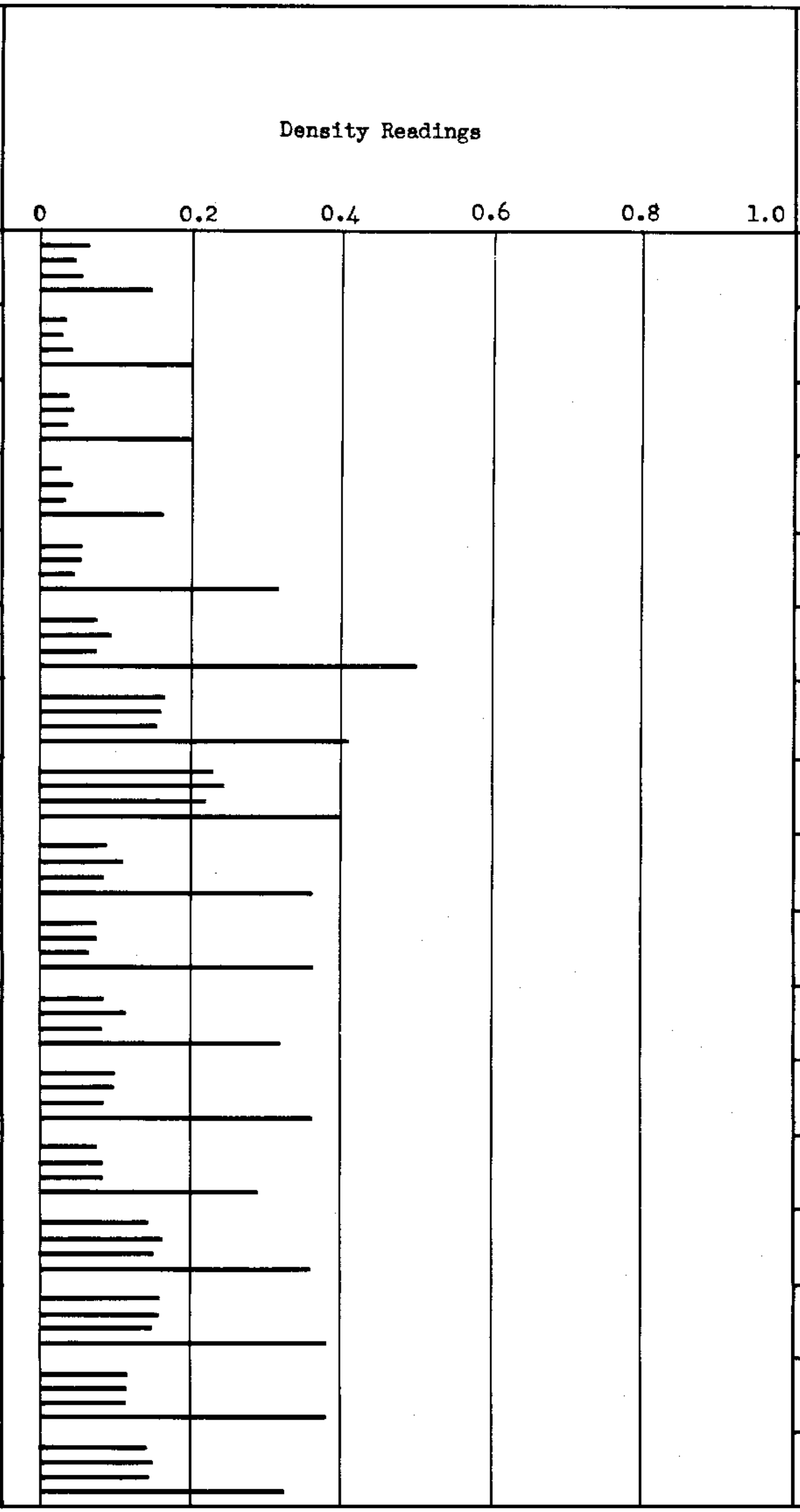
Exposed January 28, 1952 to July 7, 1953



Sample Number	Crazing	Color After Dark Storage	Color After Roof Exposure	Heat Stability
157 b/	1	C	VLY	
161	2	C	C	
163	2	VLY	VLY	
167 b/	2	C	VLY	
169 b/	2	LY	LY	
174	0	C	VY	
179 b/	0	VLY	Y	
182 b/	1	VLY	LY	
185 b/	0	C	C	
188	0	C	VLY	
193	0	C	Y	

TABLE 9. (Continued) LIGHT AND HEAT STABILITY TESTS ON "GAFITE"

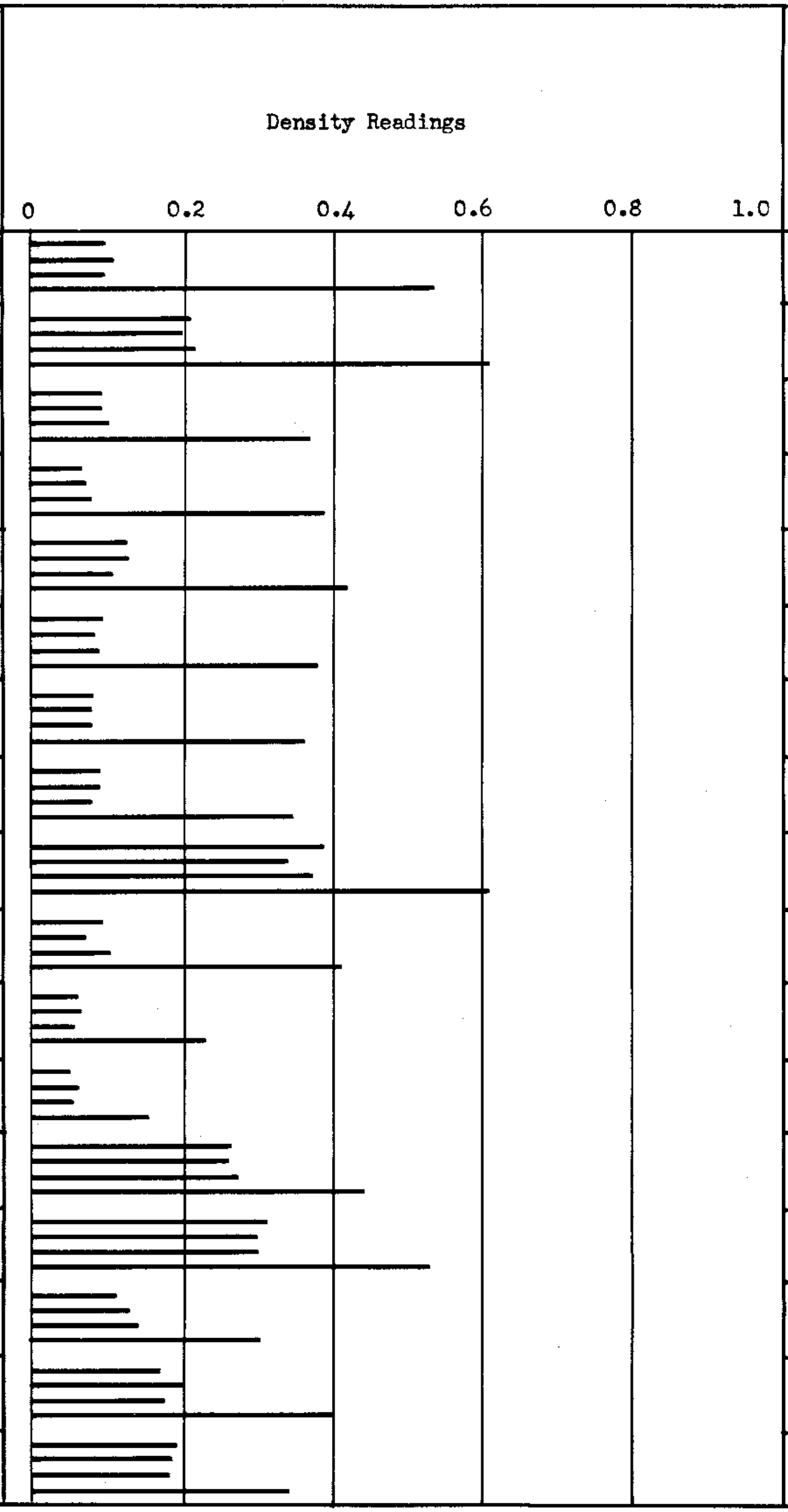
Exposed February 1, 1952 to July 7, 1953



Sample Number	Crazing	Color After Dark Storage	Color After Roof Exposure	Heat Stability
201	1	LY	VLY	
202	1	C	LY	
203	0	C	LY	
207	2	C	Y	
208	0	C	Y	
210	0	VLY	VY	
211	0	LY	VY	
213	0	Y	VY	
215	0	VLY	DY	
216	1	C	VY	
219	0	VLY	DY	
220	0	VLY	DY	
221	0	VLY	DY	
222	1	LY	DY	
224	0	LY	DY	
225	0	VLY	DY	
226	1	LY	DY	

TABLE 9. (Continued) LIGHT AND HEAT STABILITY TESTS ON "GAFITE"

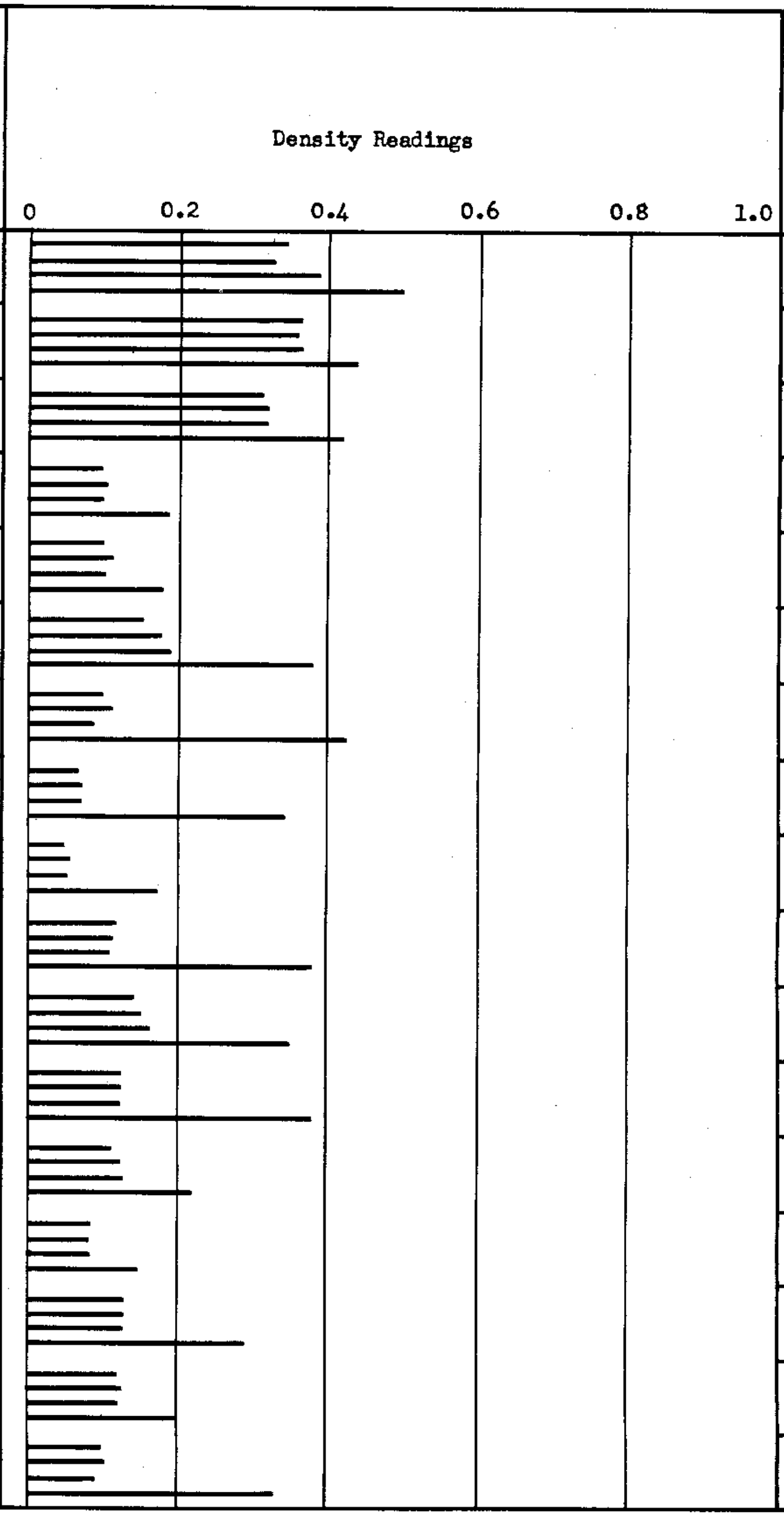
Exposed February 1, 1952 to July 7, 1953



Sample Number	Crazing	Color After Dark Storage	Color After Roof Exposure	Heat Stability
227	1	VLY	DY	
228	0	Y	DY	
230	0	VLY	DY	
232	0	C	Y	
233	0	LY	DY	
234	0	VLY	Y	
235	0	C	Y	
236	0	C	Y	
237	0	C	Y	
239	1	VLY	DY	
240	0	C	Y	
242	0	C	Y	
247	0	Y	DY	
248	0	Y	DY	
249	0	Y	DY	
250	1	Y	DY	
251	1	Y	DY	

TABLE 9. (Continued) LIGHT AND HEAT STABILITY TESTS ON "GAFITE"

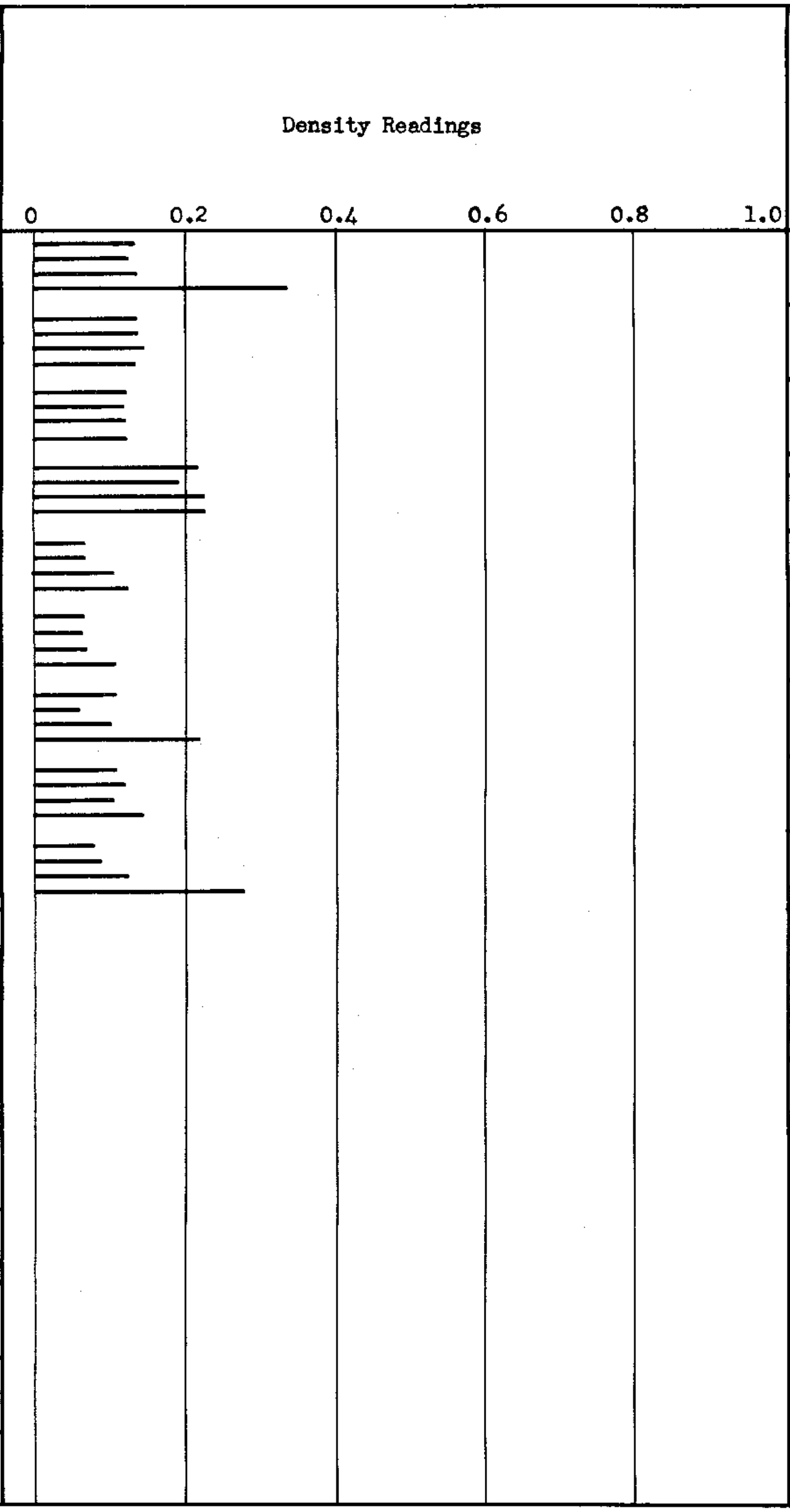
Exposed February 1, 1952 to July 7, 1953



Sample Number	Crazing	Color After		Heat Stability
		Dark Storage	Roof Exposure	
253	0	Y	DY	
254	0	VY	DY	
256	0	Y	DY	
259	2	VLY	VLY	
260	2	VLY	LY	
263	1	VLY	DY	
264	0	C	DY	
265	0	C	DY	
266	0	VLY	Y	
268	0	LY	Y	
269	0	LY	DY	
270	0	VLY	DY	
271	0	VLY	Y	1
272	0	LY	Y	
274	0	VLY	Y	2
275	0	LY	Y	2
276	0	C	Y	2

TABLE 9. (Continued) LIGHT AND HEAT STABILITY TESTS ON "GAFITE"

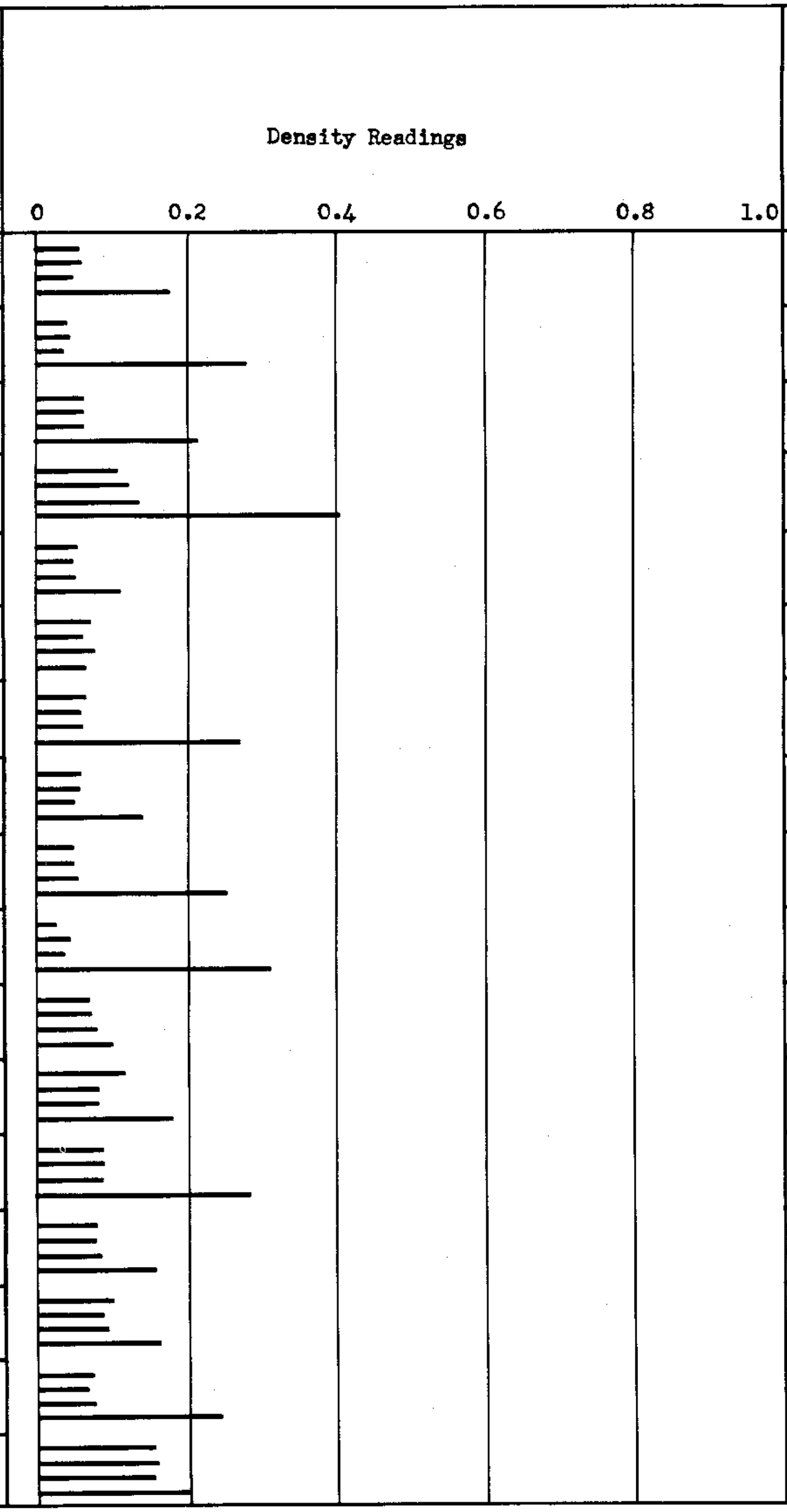
Exposed February 1, 1952 to July 7, 1953



Sample Number	Density Readings	Crazing	Color After Dark Storage	Color After Roof Exposure	Heat Stability
277		0	VLY	Y	1
278 c/		0	LY	LY	10
280 b/		0	LY	LY	
284 c/		0	VLY	Y	
286		0	C	LY	1
291		1	C	LY	3
295		0	C	DY	
298		0	VLY	Y	
299		0	C	DY	3

TABLE 9. (Continued) LIGHT AND HEAT STABILITY TESTS ON "GAFITE"

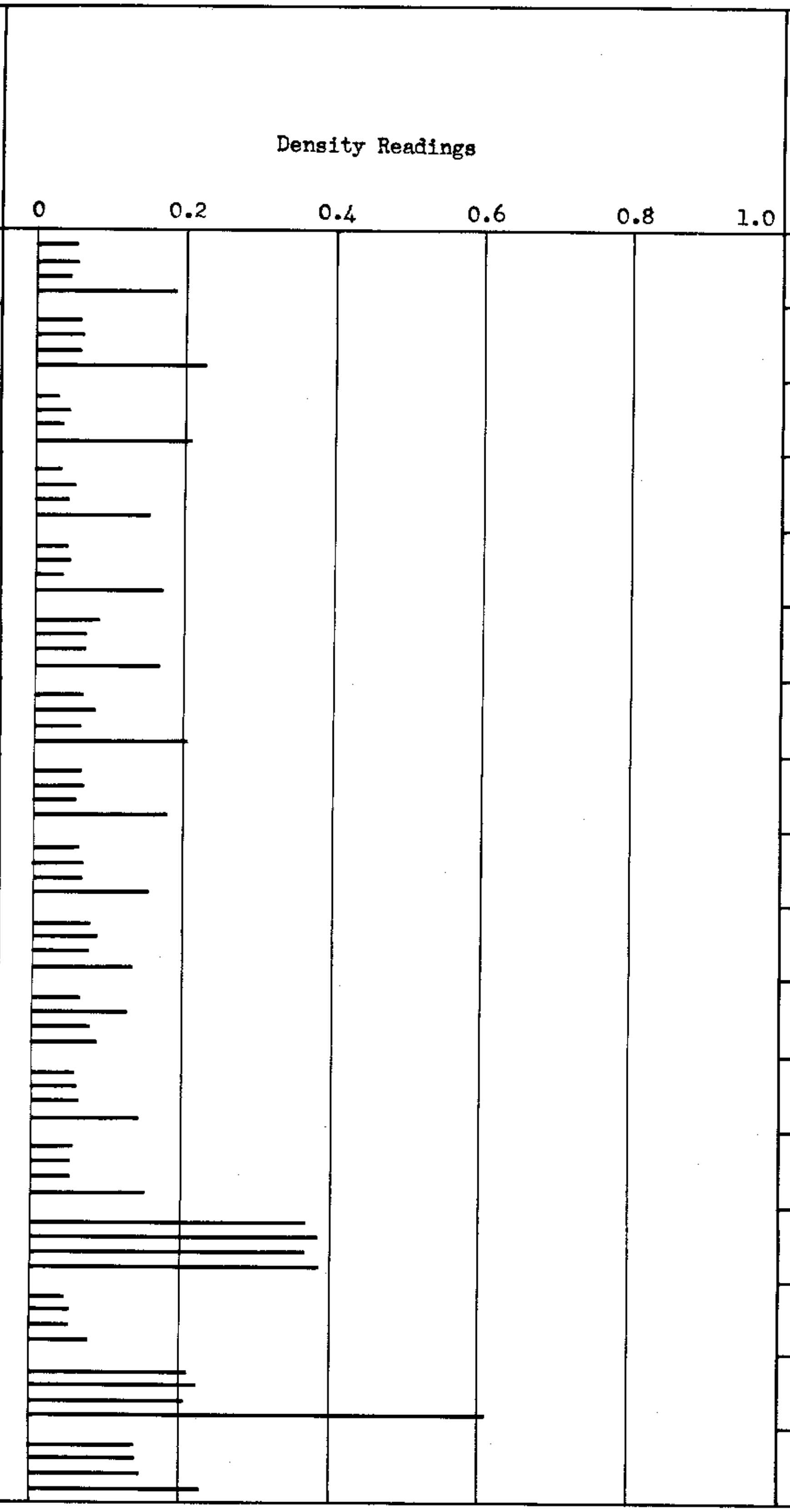
Exposed February 7, 1952 to July 7, 1953



Sample Number	Crazing	Color After Dark Storage	Color After Roof Exposure	Heat Stability
302	0	C	Y	
304	0	C	Y	3
305	0	C	Y	4
306	0	VLY	DY	
307	0	C	LY	
310	0	C	VLY	
312	0	C	Y	4
313	0	C	LY	5
314	0	C	Y	
315	0	C	Y	5
317	0	C	LY	2
318	0	C	Y	3
319	0	C	Y	
320	0	VLY	LY	
321	0	VLY	Y	5
322	0	C	Y	6
323	0	LY	LY	

TABLE 9. (Continued) LIGHT AND HEAT STABILITY TESTS ON "GAFITE"

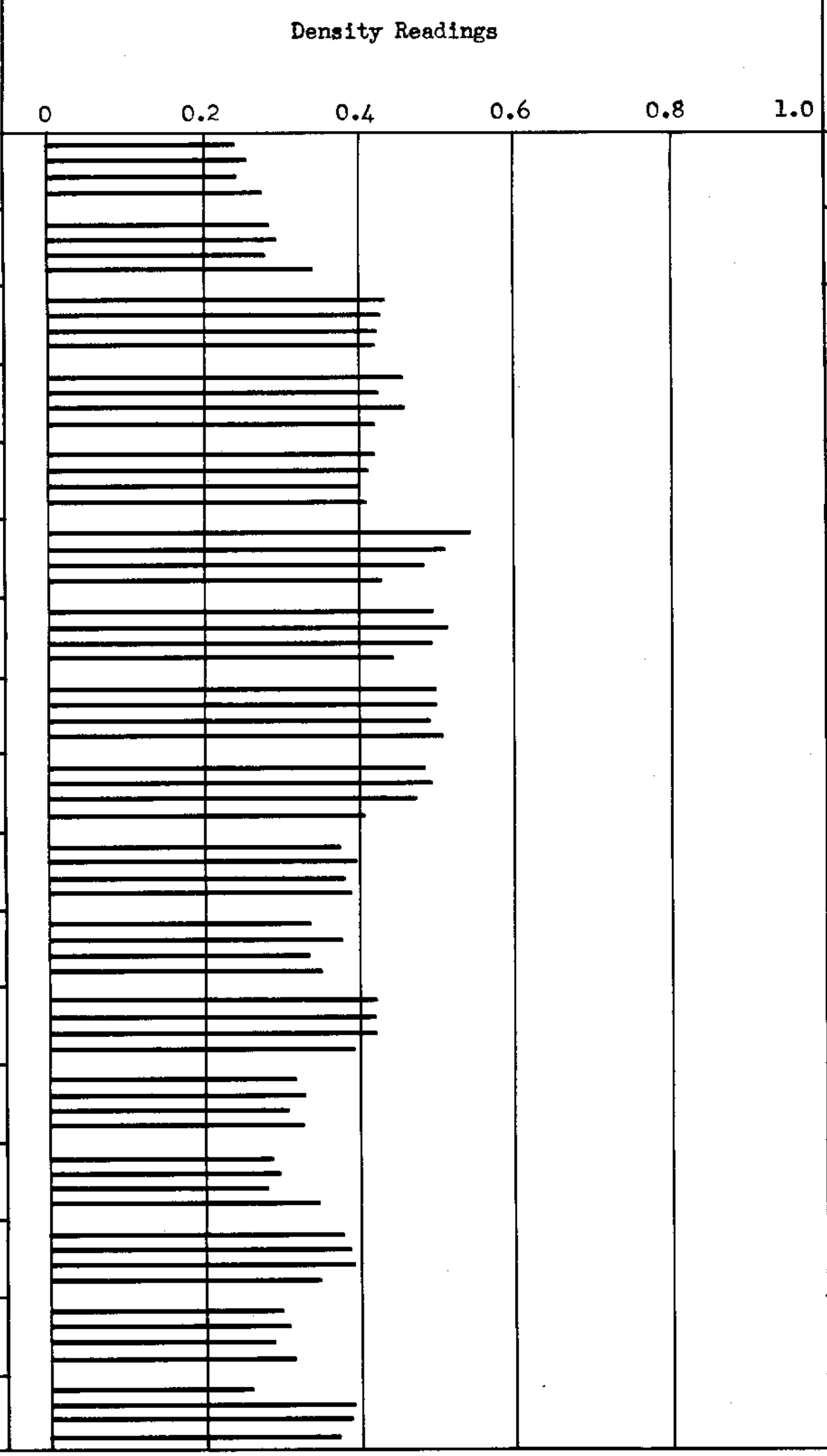
Exposed February 7, 1952 to July 7, 1953



Sample Number	Crazing	Color After Dark Storage	Color After Roof Exposure	Heat Stability
324	0	C	Y	6
325	0	C	Y	6
326	0	C	DY	6
327	0	C	Y	6
328	0	C	DY	3
329	0	VLY	Y	3
330	0	C	DY	5
331	0	VLY	Y	5
332	0	C	Y	5
333	0	VLY	LY	3
334	0	C	VLY	5
335	1	C	Y	5
336	0	C	Y	5
337	0	VLY	VLY	5
338	1	C	VLY	6
341	0	VLY	DY	
342	1	C	LY	

TABLE 9. (Continued) LIGHT AND HEAT STABILITY TESTS ON "GAFITE"

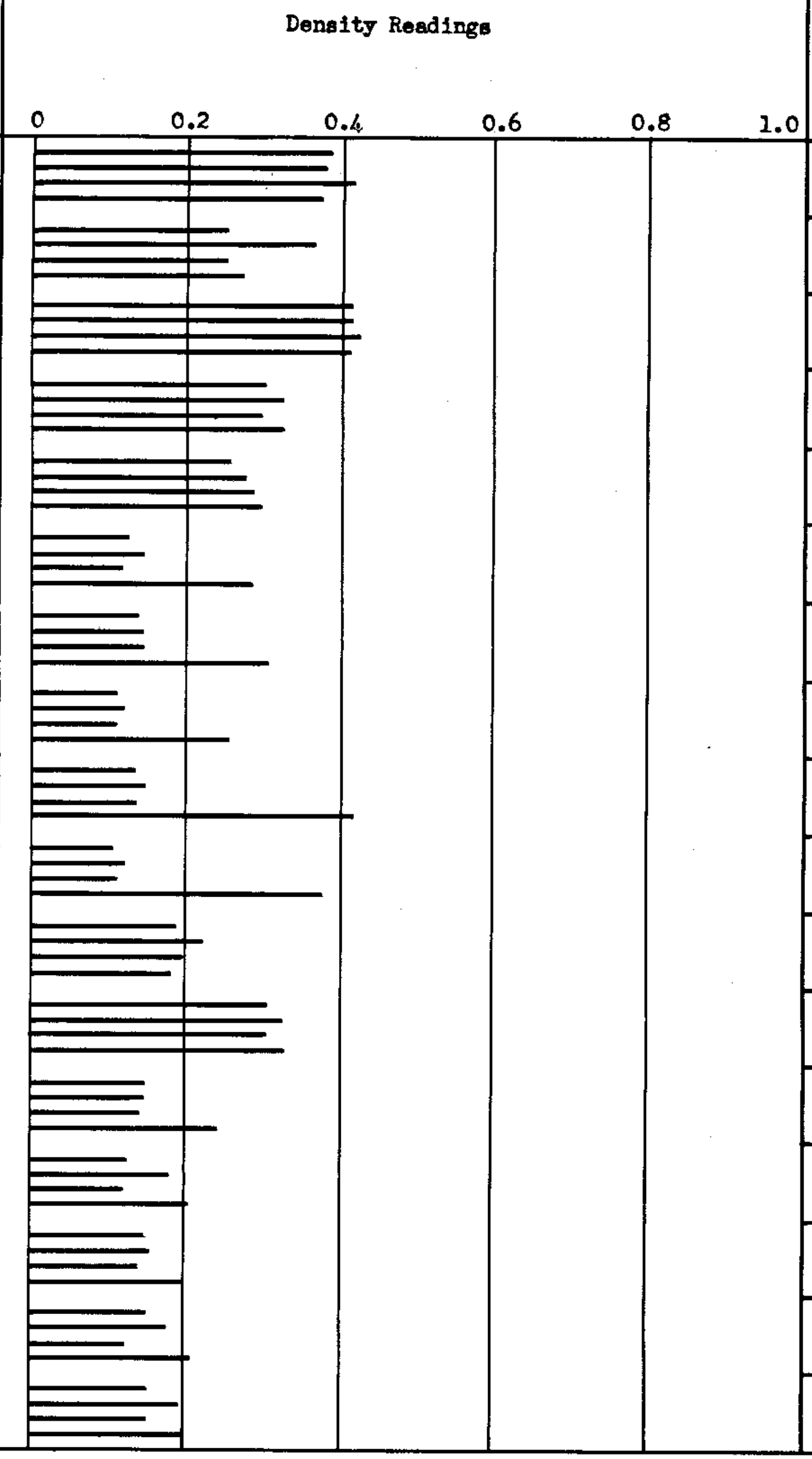
Exposed February 7, 1952 to July 7, 1953



Sample Number	Crazing	Color After Dark Storage	Color After Roof Exposure	Heat Stability
344	0	C	VLY	6
345	0	C	LY	7
348	0	LY	LY	7
349	0	LY	VLY	
350	0	VLY	VLY	
351	0	VLY	VLY	
355	0	LY	LY	
356	0	LY	VLY	3
357	0	LY	VLY	3
358	0	LY	LY	2
359	0	LY	VLY	3
360	0	Y	LY	0
361	0	VLY	VLY	3
362	0	VLY	LY	2
363	1	LY	LY	3
364	0	VLY	LY	2
365	0	LY	LY	1

TABLE 9. (Continued) LIGHT AND HEAT STABILITY TESTS ON "GAFITE"

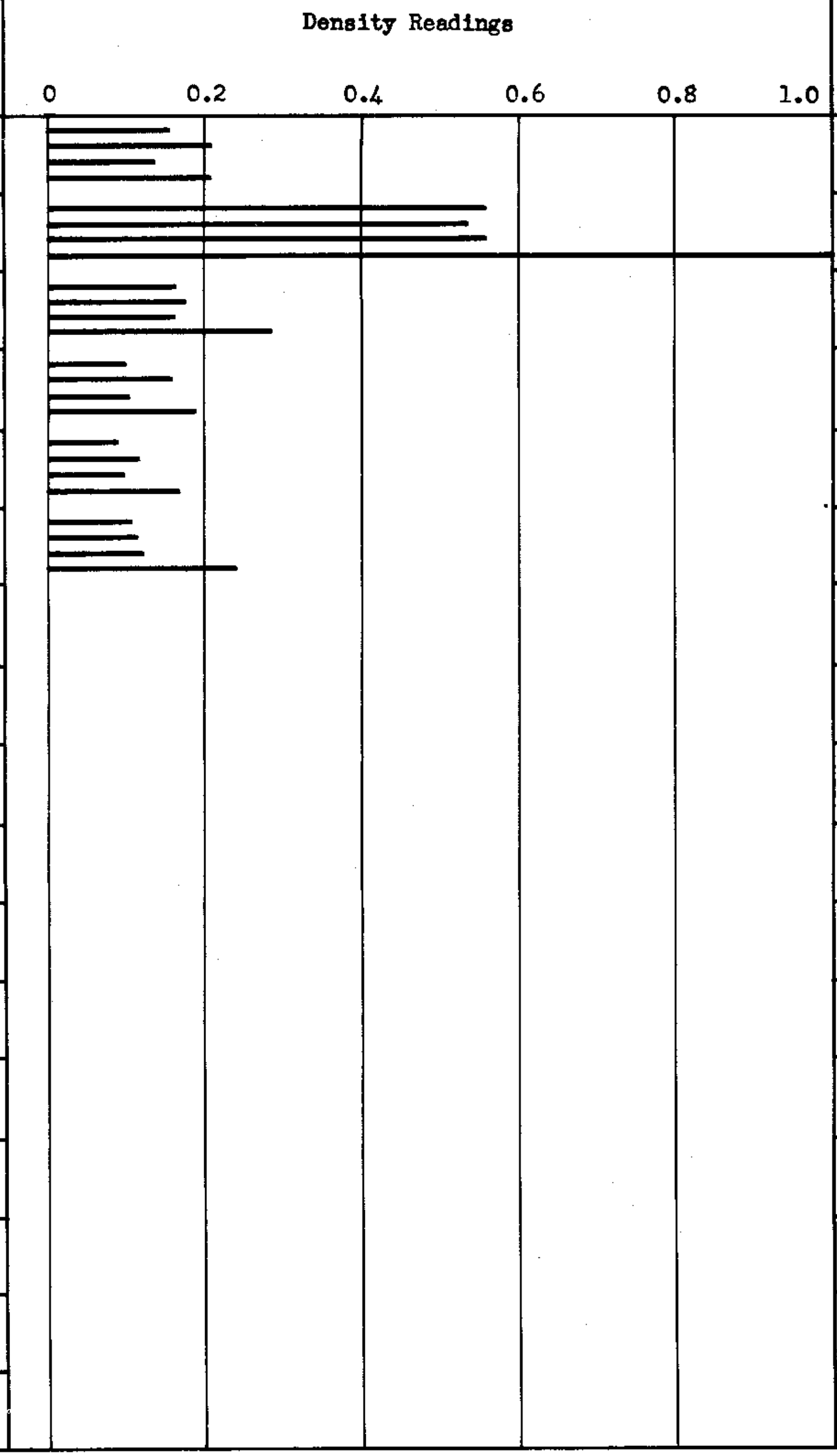
Exposed February 7, 1952 to July 7, 1953



Sample Number	Crazing	Color After		Heat Stability
		Dark Storage	Roof Exposure	
366	0	LY	VLY	2
368	0	VLY	VLY	3
370	0	LY	LY	
371	0	VLY	LY	2
377	0	LY	LY	2
378	2	LY	Y	
380	0	VLY	Y	2
381	0	VLY	Y	3
383	0	VLY	DY	2
384	0	VLY	DY	5
385	0	LY	VLY	3
386	0	LY	VLY	2
388	0	LY	LY	3
389	0	VLY	LY	3
390	0	LY	Y	7
391	0	LY	Y	3
392	0	LY	Y	3

TABLE 9. (Continued) LIGHT AND HEAT STABILITY TESTS ON "GAFITE"

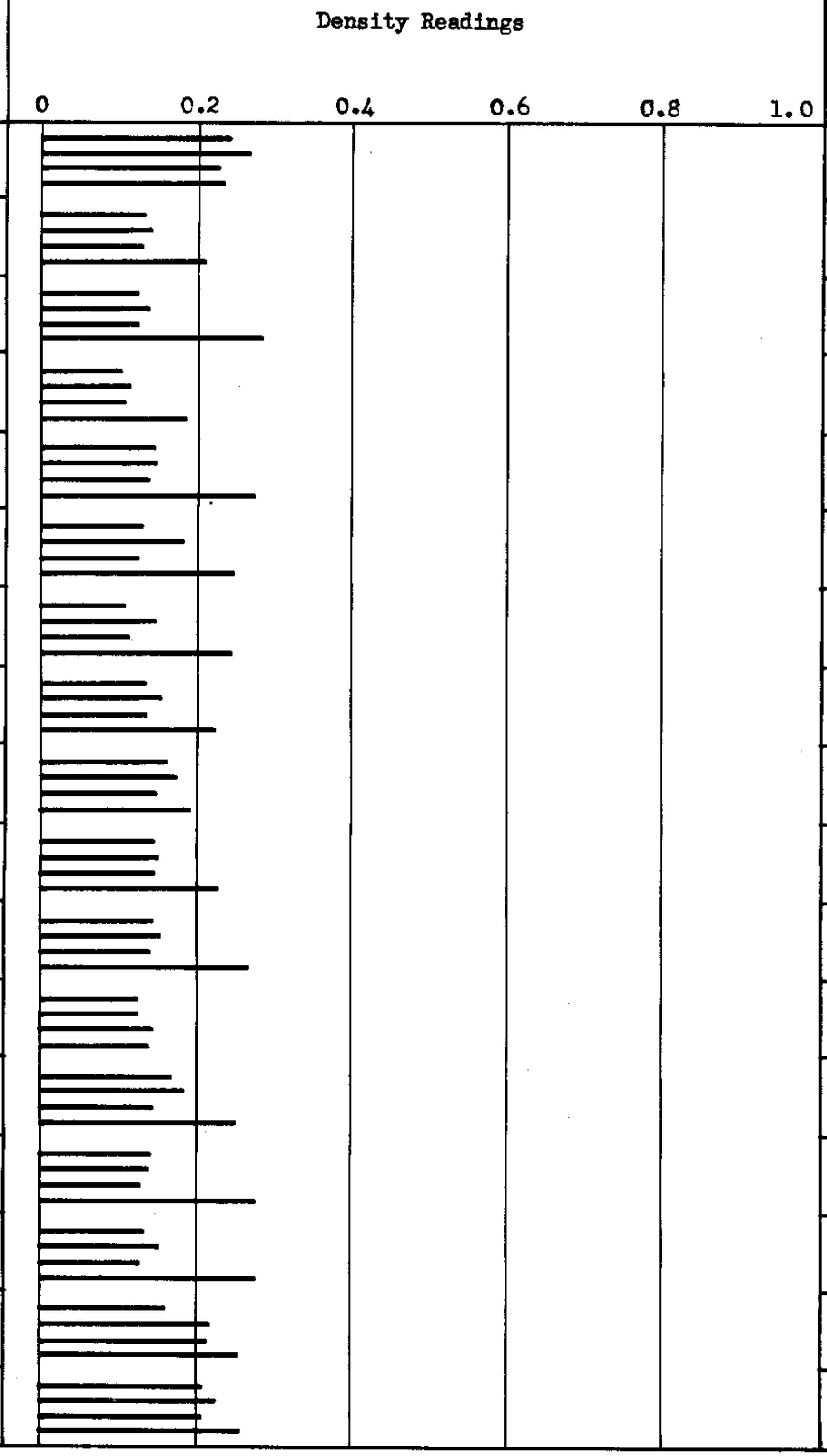
Exposed February 7, 1952 to July 7, 1953



Sample Number	Density Readings	Crazing	Color After Dark Storage	Color After Roof Exposure	Heat Stability
393		0	LY	Y	4
394		0	DY	DY	
396		0	LY	Y	4
397		2	LY	DY	
398		2	LY	Y	
399		1	LY	Y	

TABLE 9. (Continued) LIGHT AND HEAT STABILITY TESTS ON "GAFITE"

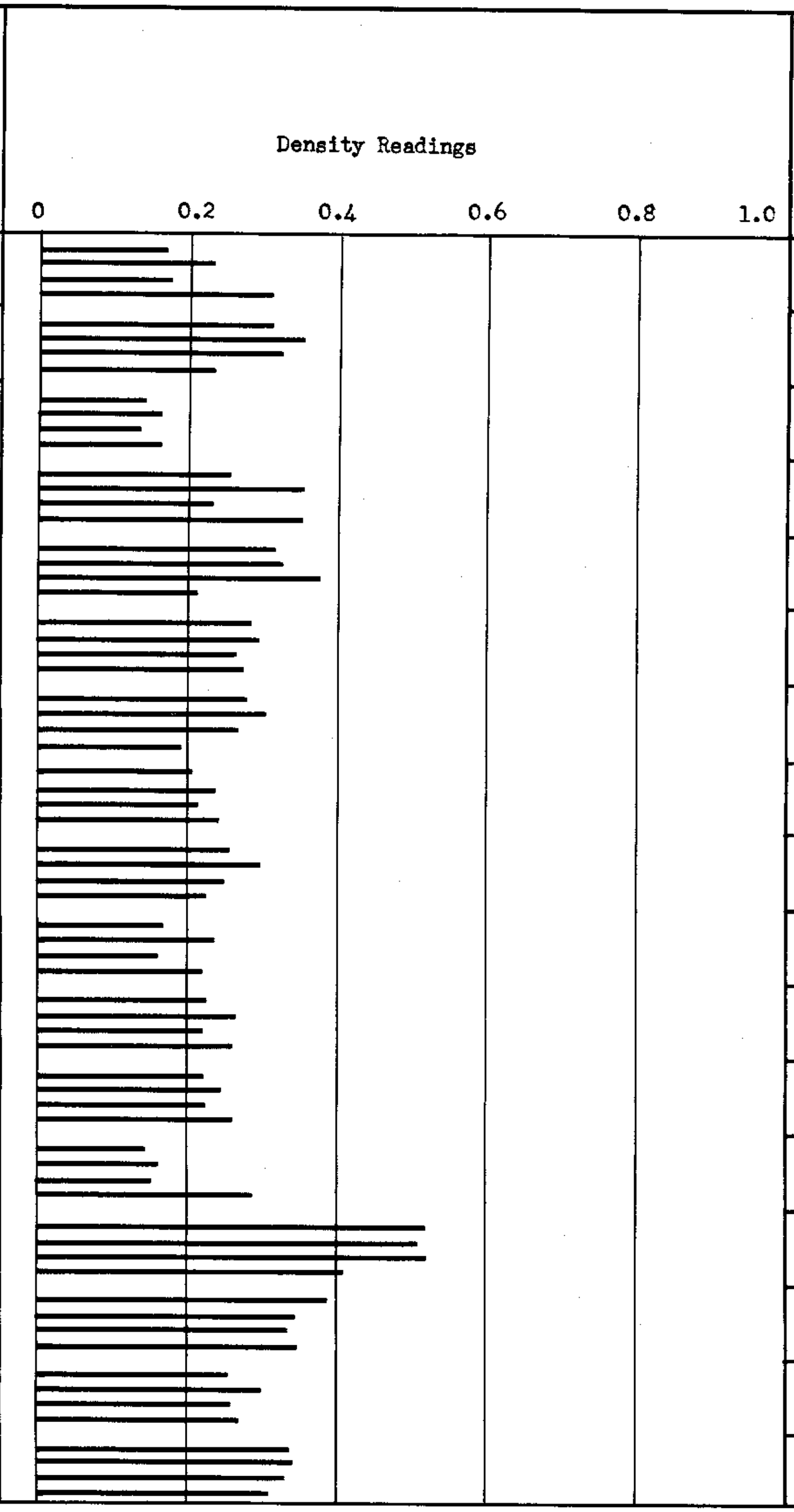
Exposed February 18, 1952 to July 7, 1953



Sample Number	Crazing	Color After		Heat Stability
		Dark Storage	Roof Exposure	
400	0	LY	Y	5
401	0	LY	Y	5
404	0	LY	Y	5
406	0	LY	Y	4
407	0	LY	Y	1
408	0	LY	Y	6
409	0	LY	Y	4
411	0	LY	Y	6
412	0	LY	Y	8
418	0	LY	Y	4
419	0	LY	Y	4
421	0	LY	Y	6
422	0	VLY	Y	12
423	0	VLY	Y	10
425	0	LY	DY	15
429	0	LY	Y	8
431	0	LY	Y	10

TABLE 9. (Continued) LIGHT AND HEAT STABILITY TESTS ON "GAFITE"

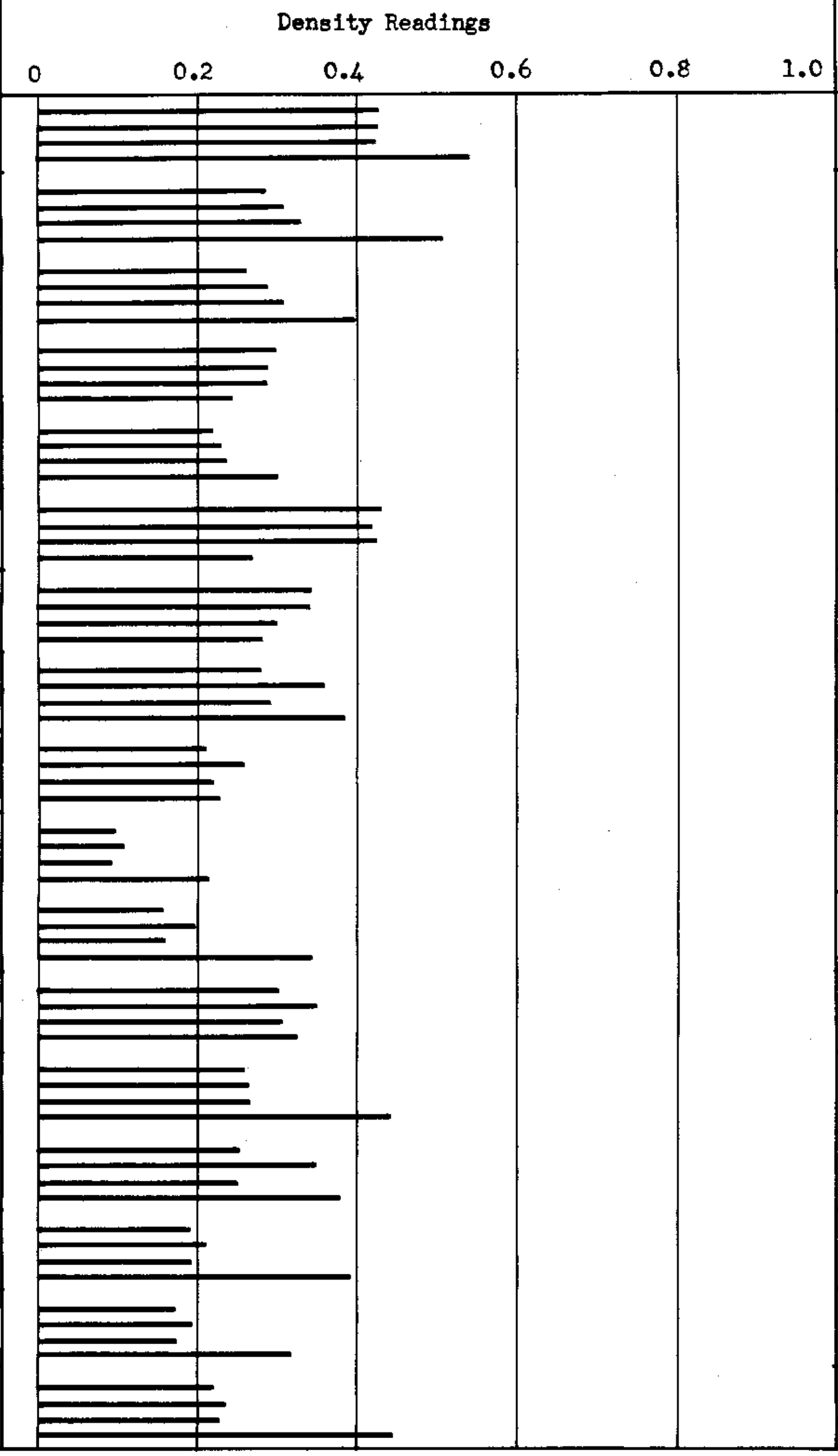
Exposed February 18, 1952 to July 7, 1953



Sample Number	Crazing	Color After		Heat Stability
		Dark Storage	Roof Exposure	
432	0	Y	DY	17
434	0	Y	Y	7
435	0	Y	Y	8
437	0	DY	Y	7
438	0	Y	Y	8
439	0	Y	Y	8
440	0	Y	Y	6
442	0	LY	DY	6
443	0	LY	DY	6
444	0	LY	Y	
445	0	Y	Y	5
446	0	LY	Y	6
447	0	VLY	DY	5
449	0	Y	DY	4
452	0	Y	DY	4
455	0	Y	Y	5
456	0	Y	Y	7

TABLE 9. (Continued) LIGHT AND HEAT STABILITY TESTS ON "GAFITE"

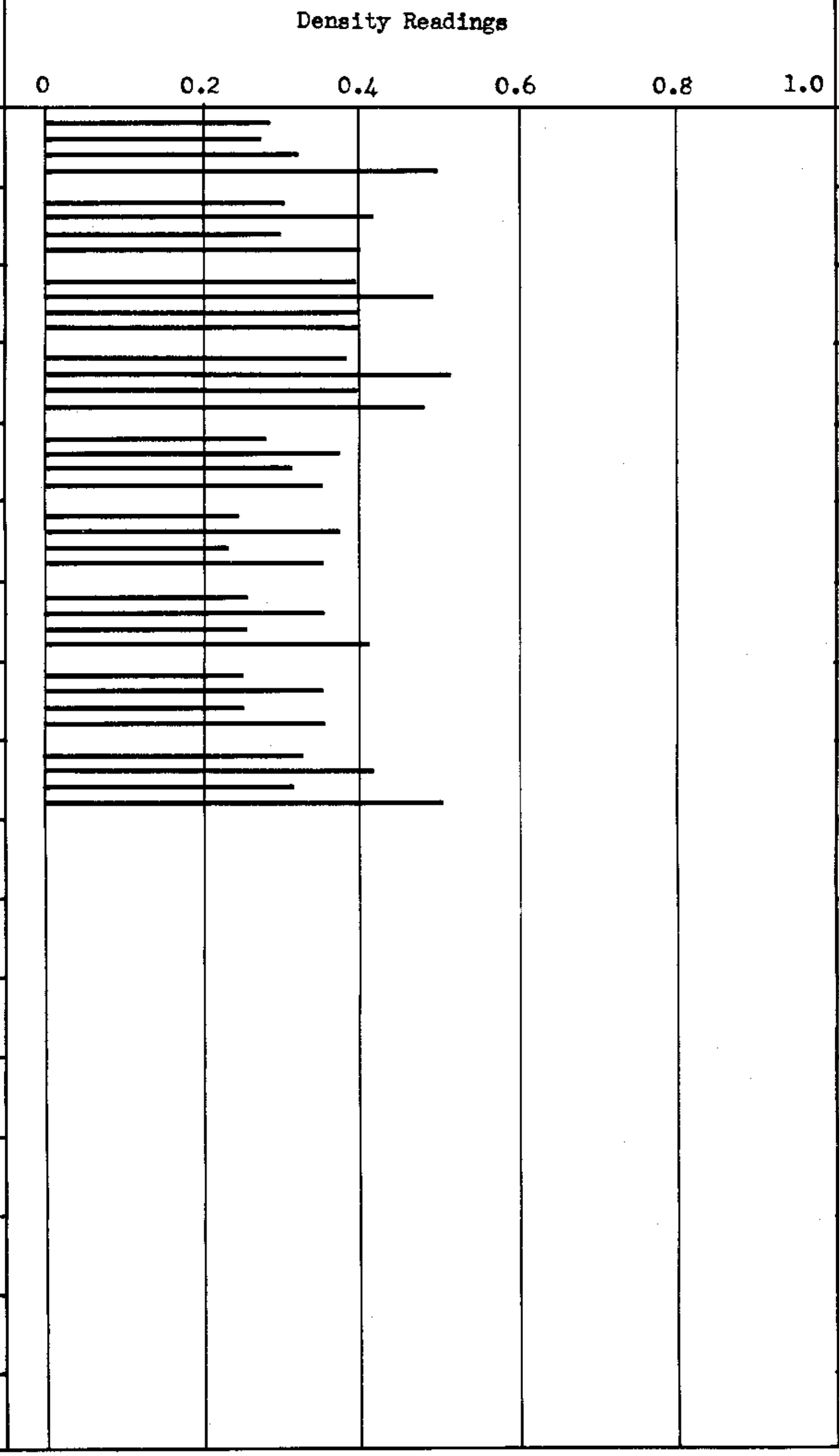
Exposed February 18, 1952 to July 7, 1953



Sample Number	Crazing	Color After Dark Storage	Color After Roof Exposure	Heat Stability
457	0	LY	DY	7
458	0	LY	DY	8
460	0	LY	Y	
461	0	LY	Y	6
462	0	LY	Y	5
463	0	Y	Y	3
467	0	LY	Y	
469	0	Y	Y	2
472	0	Y	Y	
475	0	VLY	LY	
478	0	LY	Y	
479	0	Y	Y	
480	2	Y	DY	
481	0	Y	DY	
484	2	LY	DY	
487	0	LY	Y	
488	0	LY	Y	

TABLE 9. (Continued) LIGHT AND HEAT STABILITY TESTS ON "GAFITE"

Exposed February 18, 1952 to July 7, 1953



Sample Number	Crazing	Color After		Heat Stability
		Dark Storage	Roof Exposure	
489	0	LY	DY	8
490	0	Y	DY	5
491	0	Y	DY	
492	0	DY	DY	
493	0	Y	DY	11
494	0	Y	Y	11
497	0	Y	DY	8
498	0	Y	DY	11
499	0	Y	DY	

TABLE 9. (Continued) LIGHT AND HEAT STABILITY TESTS ON "GAFITE"

Exposed February 28, 1952 to July 7, 1953

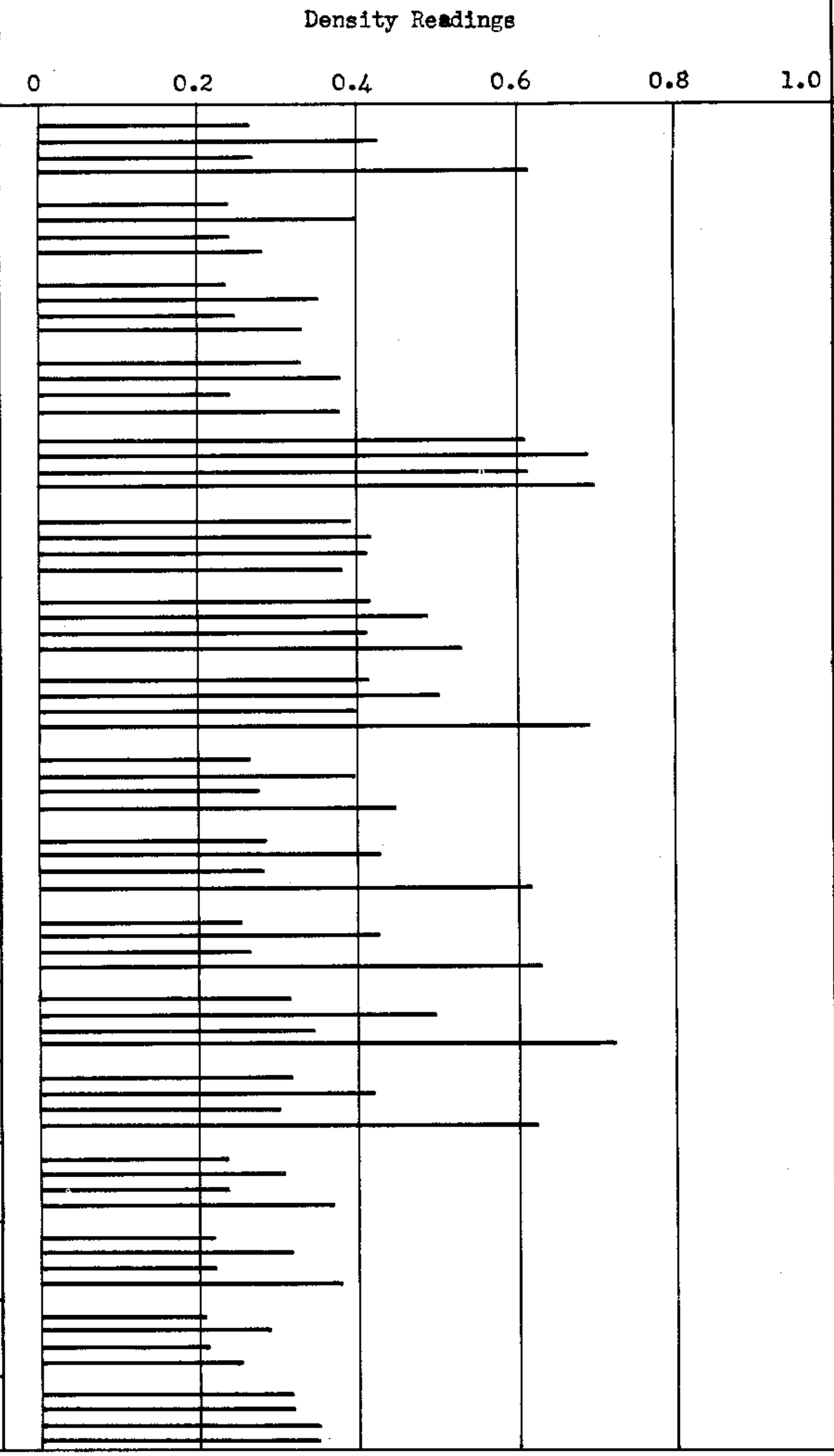
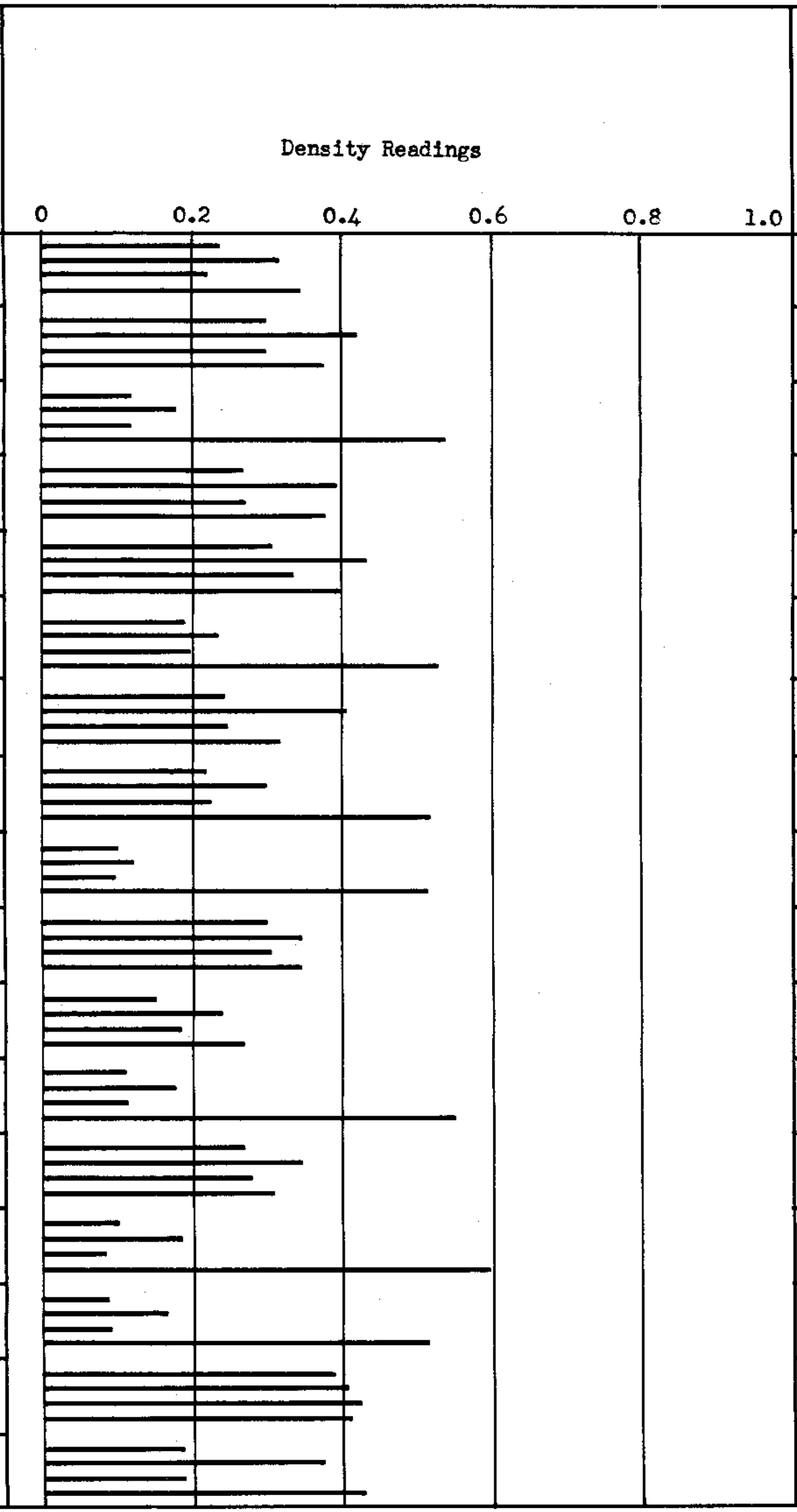
Sample Number	Density Readings 	Crazing	Color After		Heat Stability
			Dark Storage	Roof Exposure	
500		0	DY	DY	
501		0	DY	Y	11
502		0	LY	Y	9
504		0	Y	Y	11
505		0	DY	DY	12
506		0	Y	Y	10
507		0	Y	DY	9
508		0	Y	DY	9
509		0	Y	DY	
510		0	Y	DY	
511		0	Y	DY	
512		0	Y	DY	
513		0	Y	DY	
514		0	LY	Y	
515		0	LY	Y	
516		0	LY	Y	
518		0	LY	Y	

TABLE 9. (Continued) LIGHT AND HEAT STABILITY TESTS ON "GAFITE"

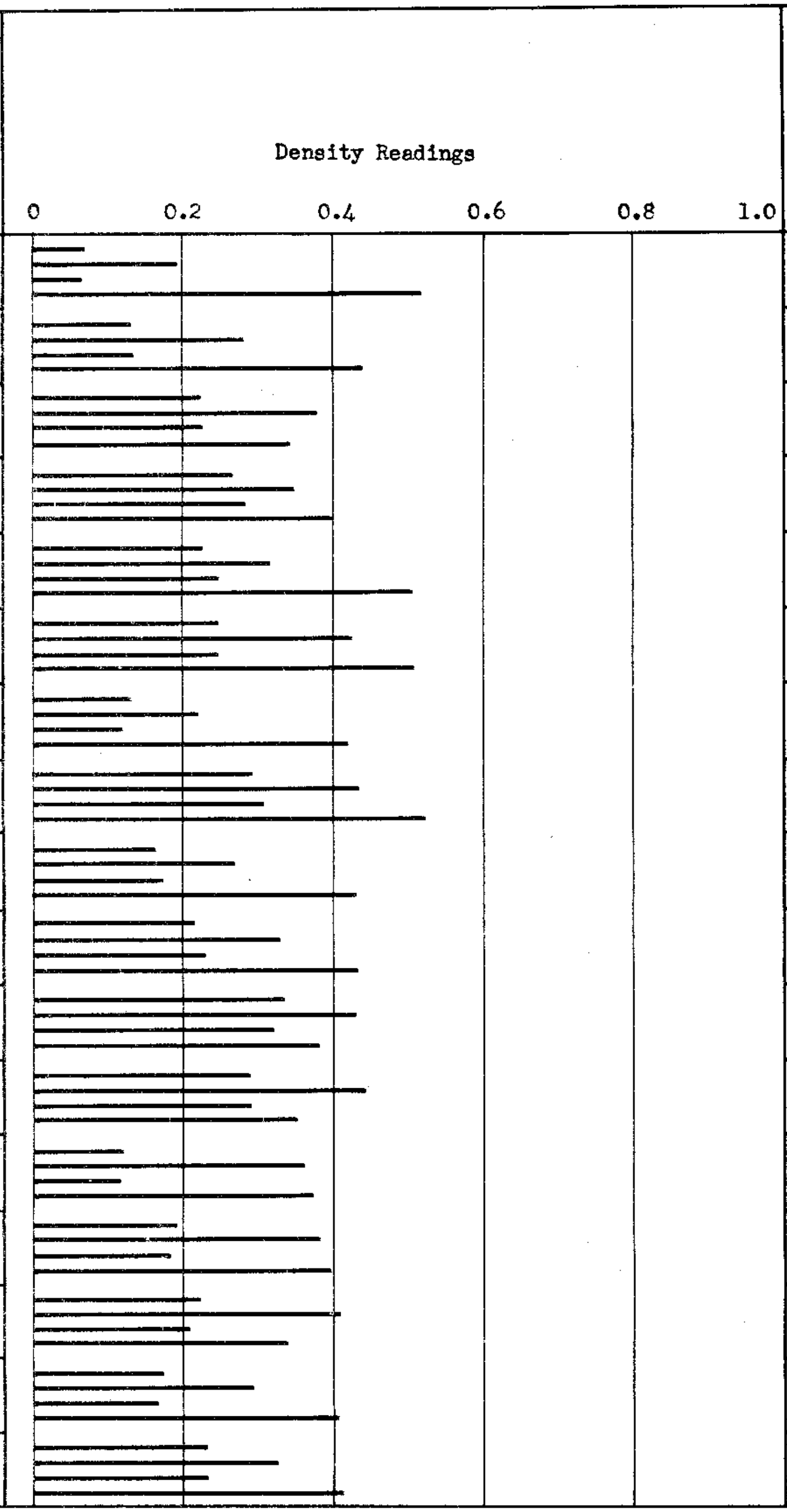
Exposed February 28, 1952 to July 7, 1953



Sample Number	Crazing	Color After Dark Storage	Color After Roof Exposure	Heat Stability
519	0	Y	Y	
521	0	Y	Y	5
522	0	LY	DY	
523	0	Y	Y	
525	0	Y	Y	5
526	0	LY	DY	9
527	0	Y	Y	
528	0	LY	DY	
529	0	LY	DY	6
530	0	Y	Y	7
531	0	LY	Y	8
532	0	LY	DY	5
533	0	Y	Y	9
535	0	LY	DY	4
537	0	LY	DY	
538	0	Y	Y	
542	0	Y	DY	7

TABLE 9. (Continued) LIGHT AND HEAT STABILITY TESTS ON "GAFITE"

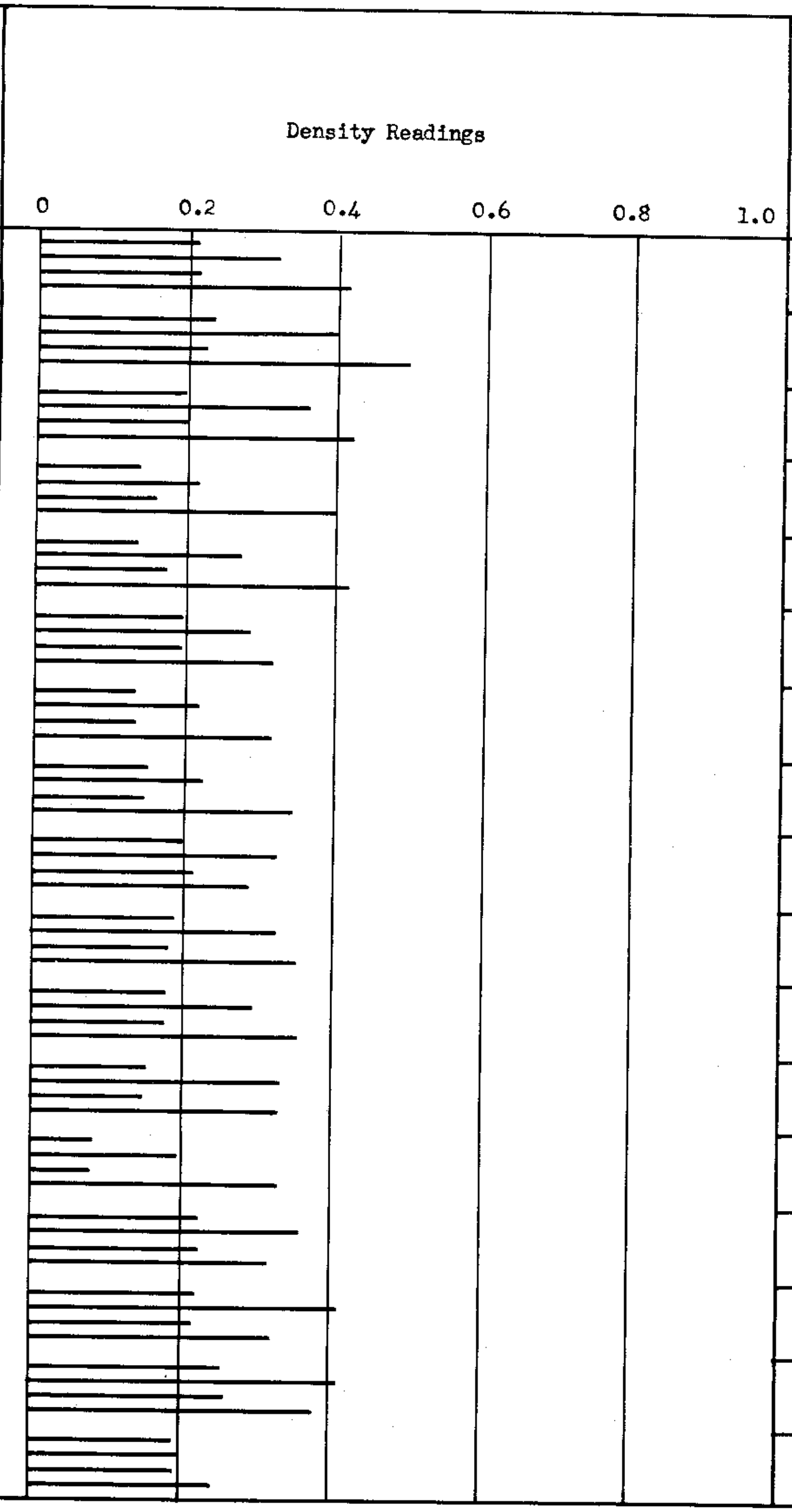
Exposed February 28, 1952 to July 7, 1953



Sample Number	Crazing	Color After Dark Storage	Color After Roof Exposure	Heat Stability
543	0	LY	DY	6
544	0	Y	DY	
545	0	Y	Y	
546	0	Y	DY	12
547	0	Y	DY	
548	0	Y	DY	
549	0	LY	DY	10
550	0	DY	DY	
551	0	LY	DY	
552	0	Y	DY	
553	0	Y	DY	
554	0	Y	DY	
556	0	LY	DY	7
557	0	Y	DY	
558	0	Y	DY	7
559	0	LY	DY	
561	0	Y	DY	8

TABLE 9. (Continued) LIGHT AND HEAT STABILITY TESTS ON "GAFITE"

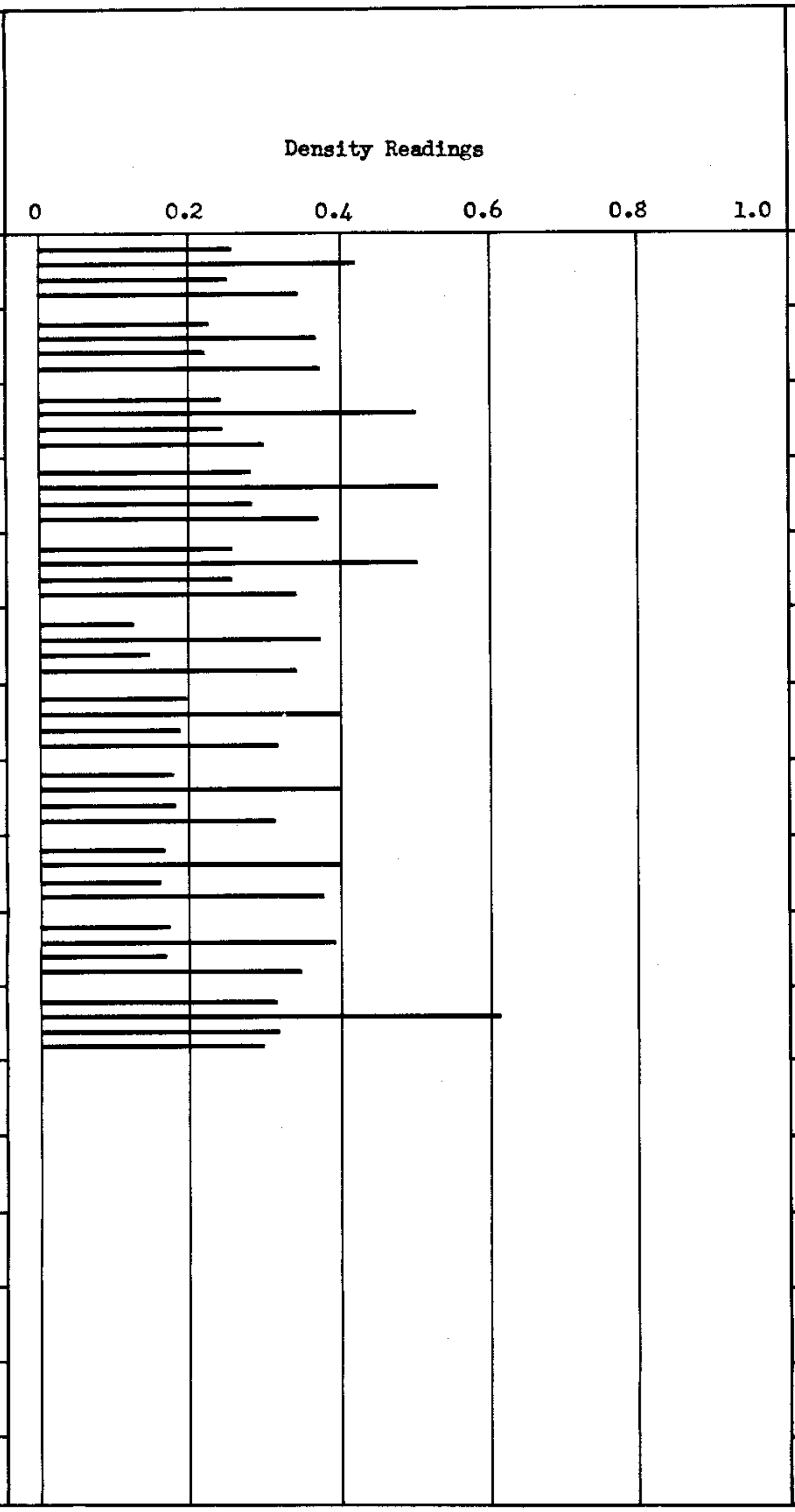
Exposed February 28, 1952 to July 7, 1953



Sample Number	Crazing	Color After Dark Storage	Color After Roof Exposure	Heat Stability
562	0	LY	DY	8
563	0	Y	DY	8
564	0	Y	DY	10
565	0	LY	DY	11
566	0	LY	DY	5
567	0	Y	Y	4
568	0	LY	Y	4
569	0	Y	Y	4
571	0	Y	Y	4
572	0	Y	Y	3
573	0	LY	Y	8
574	0	Y	Y	6
576	0	Y	DY	
577	0	Y	Y	4
578	0	Y	Y	8
579	0	Y	Y	2
580	0	Y	DY	

TABLE 9. (Continued) LIGHT AND HEAT STABILITY TESTS ON "GAFITE"

Exposed February 28, 1952 to July 7, 1953



Sample Number	Crazing	Color After Dark Storage	Color After Roof Exposure	Heat Stability
581	0	Y	Y	4
582	0	Y	Y	4
584	0	Y	Y	4
585	0	Y	Y	3
586	0	Y	Y	5
587	0	LY	Y	3
588	0	LY	Y	5
589	0	Y	Y	4
590	0	Y	Y	5
591	0	Y	Y	6
592	0	DY	DY	

a/ Indicates samples were put on roof January 28, 1952 to July 7, 1953.
b/ Indicates samples were put on roof February 29, 1952 to July 7, 1953.
c/ Indicates samples were put on roof February 7, 1952 to July 7, 1953.

SECTION II. REVIEW OF COMPOSITION OF EARLY "GAFITE" SHEETS POSSESSING GOOD LIGHT STABILITY

The results of light stability tests on about 400 samples of "Gafite" exposed for 1.5 years on the roof of the Central Research Laboratory of GAF at Easton, Pa., were reported in Section I. These 400 samples were ranked by their final visual color, by their final yellowness index, and by their changes in yellowness index on light exposure. The best 187 samples of these 400 samples were reviewed for their conditions of preparation, which are recorded in this section. These results are surveyed and the most likely approaches to solution of the light stability problem are outlined in this section.

2.1 Selection of Samples

Table 7 of Section I lists "Gafite" sheet samples ranked by visual color observations after 1.5 years of roof exposure. Each column of this table was numbered consecutively from 1 (colorless to light yellow) to 14 (yellow) with the numbers increasing with increasing visual yellowness in the samples. All the sheets in columns to and including Column 8 (labeled H in Section I) were selected for review.

Table 5 of Section I lists "Gafite" sheets according to their final yellowness index (100 x optical density values listed). The columns in this table were numbered consecutively with the numbers increasing with increasing yellowness index. It was found that most of the sheets selected from Table 7 (up to and including Column 8) were listed in Table 5 up to and including Column 6. All sheets listed in Table 5 up to and including Column 6 were selected. This list includes a few sheets not selected originally from Table 7. The correlation between the tables is an indication that the visual observation of yellowness agreed fairly well with the measured yellowness index.

Table 6 of Section I lists "Gafite" samples ranked according to their changes in yellowness index on 1.5 years roof exposure. The columns in this table were numbered with all columns possessing an increase of optical density of 0.024 or less being numbered 1. By this method those samples becoming less yellow on roof exposure were included in column 1. The other columns were numbered consecutively with increasing numbers denoting greater increases in yellowness index on roof exposure. All sheets listed in columns up to and including column 2 were selected for study. This selection contains sheets not previously chosen from Tables 5 and 7. However, many of the selections from Tables 5 and 7 were present in this list. By this method the "Gafite" sheets possessing the lowest final yellowness indices and showing the least increase in yellowness indices were chosen from Tables 5, 6, and 7. All of these 187 sheets were reviewed for their methods of preparation and the data on these sheets together with their ranking in Tables 5, 6, and 7 were assembled in Table 10 of this section. The "Gafite" sheets with the lowest final yellowness indices and the highest stabilities toward light possess lower numbers across this table. For comparison purposes the heat stabilities are included where available. With the heat stability results, the higher numbers denote the more stable sheets toward heat.

In Table 10, the samples derived from the same batch of monomer are designated by use of identical letters following the sheet number. For example Sheets 8A and 9A were cast from the same batch of monomer just as Sheets 461DD and 467DD were both cast from another batch of monomer. In Table 10, the mole percent purity is calculated from the freezing point of the methyl α-chloroacrylate monomer (5).

TABLE 10. SELECTED LIST OF SHEETS POSSESSING LEAST YELLOWNESS AFTER LIGHT EXPOSURE AND SHOWING LEAST CHANGE IN YELLOWNESS ON LIGHT EXPOSURE

Sample Number	Methyl ∝-Chloroacrylate Purity (Mole %)	Inhibitors Added	Purification Steps	Catalysts and Other Added Agents	Polymerization Conditions		Final Visual Color Rating	Final Yellowness Index Rating	Yellowing Color Change Rating	Heat Stability
					Initial Polymerization Steps	Final Polymerization Step				
2			Vacuum distilled Dried over P_2O_5 Vacuum distilled		Light polymerized	16 hr at 118°C		6	4	
8A			Vacuum distilled Dried over P_2O_5 Vacuum distilled		Light polymerized	24 hr at 121°C	4	3	3	
9A			Vacuum distilled Dried over P_2O_5 Vacuum distilled		Light polymerized	24 hr at 121°C	8	8	9	
10			Vacuum distilled twice		Light polymerized	34 hr at 121°C	7	7	7	
18B		p-tert-Butylcatechol	Vacuum distilled twice		Light polymerized	24 hr at 121°C	10	6	6	
19B		p-tert-Butylcatechol	Vacuum distilled twice		Light polymerized	23 hr at 121°C	7	6	6	
22C		p-tert-Butylcatechol	Steam distilled Dried over P_2O_5 Vacuum distilled		Light polymerized	24 hr at 119°C	9	6	5	
23C		p-tert-Butylcatechol	Steam distilled Dried over P_2O_5 Vacuum distilled	0.09% Benzoyl peroxide	Light polymerized	24 hr at 121°C	5	6	4	
25		p-tert-Butylcatechol	Steam distilled Dried over P_2O_5 Vacuum distilled		Light polymerized	23 hr at 121°C	5	5	1	
38		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.02% Benzoyl peroxide	144 hr at 25°C	24 hr at 121°C	9	6	6	
40		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.02% Porofor N	Light polymerized	24 hr at 121°C	7	4	4	

TABLE 10. (Continued) SELECTED LIST OF SHEETS POSSESSING LEAST YELLOWNESS AFTER LIGHT EXPOSURE AND SHOWING LEAST CHANGE IN YELLOWNESS ON LIGHT EXPOSURE

Sample Number	Methyl α-Chloroacrylate Purity (Mole %)	Inhibitors Added	Purification Steps	Catalysts and Other Added Agents	Polymerization Conditions: Initial Polymerization Steps	Polymerization Conditions: Final Polymerization Step	Final Visual Color Rating	Final Yellowness Index Rating	Yellowing Color Change Rating	Heat Stability
42D		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled		Light polymerized	24 hr at 121°C	9	6	5	
43D		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled		Light polymerized	98 hr at 122°C	5	5	3	
44D		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled		Light polymerized	72 hr at 122°C	9	3	1	
48E		p-tert-Butylcatechol	Steam distillation Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled Dried over P_2O_5 Vacuum distilled		Light polymerized	24 hr at 120°C	9	4	4	
49E		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled twice	3% CH_3OH	Light polymerized	24 hr at 121°C	1	1	1	
51F		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled		Light polymerized	24 hr at 122°C 24 hr at 127°C	9	4	4	
52F		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled		Light polymerized	24 hr at 121°C	9	5	5	
54G		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled		Light polymerized	24 hr at 121°C	9	5	5	

TABLE 10. (Continued) SELECTED LIST OF SHEETS POSSESSING LEAST YELLOWNESS AFTER LIGHT EXPOSURE AND SHOWING LEAST CHANGE IN YELLOWNESS ON LIGHT EXPOSURE

Sample Number	Methyl α-Chloroacrylate Purity (Mole %)	Inhibitors Added	Purification Steps	Catalysts and Other Added Agents	Polymerization Conditions		Final Visual Color Rating	Final Yellowness Index Rating	Yellowing Color Change Rating	Heat Stability
					Initial Polymerization Steps	Final Polymerization Step				
55G		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.02% Benzoyl peroxide 3% Methanol	Light polymerized	24 hr at 121°C	2	2	1	
56G		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.02% Benzoyl peroxide 3% Methanol	Light polymerized	24 hr at 121°C	2			
58		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.02% Porofor N 3% Methanol	Light polymerized	21 hr at 121°C	1	3	1	
63		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.002% Benzoyl peroxide	17 hr at 47°C 8 hr at 62°C	24 hr at 121°C	8	4	5	
65H		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.002% Benzoyl peroxide 1% Methanol	87 hr at 45°C 7 hr at 60°C	24 hr at 120°C	4	3	4	
66H		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.002% Benzoyl peroxide 1% Methanol	87 hr at 45°C 7 hr at 60°C	24 hr at 120°C	4	3	4	
67H		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.002% Benzoyl peroxide 1% Methanol	87 hr at 45°C 7 hr at 60°C	24 hr at 120°C	4	3	3	
68H		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.002% Benzoyl peroxide 1% Methanol	87 hr at 45°C 7 hr at 60°C	24 hr at 120°C	6	4	4	

TABLE 10. (Continued) SELECTED LIST OF SHEETS POSSESSING LEAST YELLOWNESS AFTER LIGHT EXPOSURE AND SHOWING LEAST CHANGE IN YELLOWNESS ON LIGHT EXPOSURE

Sample Number	Methyl α-Chloroacrylate Purity (Mole %)	Inhibitors Added	Purification Steps	Catalysts and Other Added Agents	Polymerization Conditions		Final Visual Color Rating	Final Yellowness Index Rating	Yellowing Color Change Rating	Heat Stability
					Initial Polymerization Steps	Final Polymerization Step				
70I		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.02% Benzoyl peroxide 1% Methanol	72 hr at 25°C 17 hr at 48°C 11 hr at 61°C 13 hr at 46°C	Heated at 120°C	5	5	5	
71I		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distillation	0.02% Benzoyl peroxide 1% Methanol	72 hr at 25°C 17 hr at 48°C 11 hr at 61°C 13 hr at 46°C	Heated at 120°C	5	5	4	
72I		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.02% Benzoyl peroxide 1% Methanol	72 hr at 25°C 17 hr at 48°C 11 hr at 61°C 13 hr at 46°C	Heated at 120°C	5	6	6	
73J		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.02% Benzoyl peroxide 2% Methanol	29 hr at 25°C 40 hr at 46°C	Heated at 120°C	2	4	3	
74J		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.02% Benzoyl peroxide 2% Methanol	29 hr at 25°C 40 hr at 46°C	Heated at 120°C	2	6	6	
75J		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.02% Benzoyl peroxide 2% Methanol	29 hr at 25°C 40 hr at 46°C	Heated at 120°C	2	4	4	
76K		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled		Light polymerized	20 hr at 121°C	12	5	6	
77K		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.02% Benzoyl peroxide 3% Methanol	17 hr at 25°C 26 hr at 46°C	Heated at 120°C	3	3	3	

TABLE 10. (Continued) SELECTED LIST OF SHEETS POSSESSING LEAST YELLOWNESS AFTER LIGHT EXPOSURE AND SHOWING LEAST CHANGE IN YELLOWNESS ON LIGHT EXPOSURE

Sample Number	Methyl ∝-Chloroacrylate Purity (Mole %)	Inhibitors Added	Purification Steps	Catalysts and Other Added Agents	Polymerization Conditions		Final Visual Color Rating	Final Yellowness Index Rating	Yellowing Color Change Rating	Heat Stability
					Initial Polymerization Steps	Final Polymerization Step				
78K		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.02% Benzoyl peroxide 3% Methanol	17 hr at 25°C 26 hr at 46°C	Heated at 120°C	2	4	4	
79K		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.02% Benzoyl peroxide 3% Methanol	17 hr at 25°C 26 hr at 46°C	Heated at 120°C	2	3	2	
80L		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled		Light polymerized	24 hr at 120°C	9	5	4	
81L		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.05% Benzoyl peroxide 3% Methanol	136 hr at 25°C	8 hr at 45°C 16 hr at 60°C 24 hr at 120°C	2	3	3	
82L		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.05% Benzoyl peroxide 3% Methanol	136 hr at 25°C	8 hr at 45°C 16 hr at 60°C 24 hr at 120°C	2	4	3	
83M		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled		Light polymerized	24 hr at 120°C	8	5	5	
84M		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.02% Benzoyl peroxide 3% Methanol	Light polymerized	8 hr at 45°C 16 hr at 60°C 24 hr at 120°C	2	3	2	
86M		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.02% Benzoyl peroxide 3% Methanol	112 hr at 25°C	8 hr at 45°C 16 hr at 60°C 24 hr at 120°C	2	4	2	

TABLE 10. (Continued) SELECTED LIST OF SHEETS POSSESSING LEAST YELLOWNESS AFTER LIGHT EXPOSURE AND SHOWING LEAST CHANGE IN YELLOWNESS ON LIGHT EXPOSURE

Sample Number	Methyl α-Chloroacrylate Purity (Mole %)	Inhibitors Added	Purification Steps	Catalysts and Other Added Agents	Polymerization Conditions		Final Visual Color Rating	Final Yellowness Index Rating	Yellowing Color Change Rating	Heat Stability
					Initial Polymerization Steps	Final Polymerization Step				
87N		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled		Light polymerized	24 hr at 120°C	9	5	4	
88N		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.02% Benzoyl peroxide 3% Methanol	Light polymerized	24 hr at 120°C	2	3	1	
89N		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.02% Benzoyl peroxide 3% Methanol	87 hr at 25°C 16 hr at 60°C	24 hr at 120°C	1	5	4	
90-0		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled		Light polymerized	24 hr at 120°C	7	5	4	
91-0		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.02% Benzoyl peroxide 3% Methanol	8 hr at 48°C 21 hr at 59°C	20 hr at 121°C	1	4	4	
93-0		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.02% Benzoyl peroxide 3% Methanol	16 hr at 25°C 8 hr at 48°C 22 hr at 59°C	24 hr at 121°C	2	4	4	
94		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled		Light polymerized	21 hr at 120°C	9	5	5	
95P		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled		Light polymerized	24 hr at 120°C	8	6	6	

TABLE 10. (Continued) SELECTED LIST OF SHEETS POSSESSING LEAST YELLOWNESS AFTER LIGHT EXPOSURE AND SHOWING LEAST CHANGE IN YELLOWNESS ON LIGHT EXPOSURE

Sample Number	Methyl α-Chloroacrylate Purity (Mole %)	Inhibitors Added	Purification Steps	Catalysts and Other Added Agents	Polymerization Conditions		Final Visual Color Rating	Final yellowness Index Rating	Yellowing Color Change Rating	Heat Stability
					Initial Polymerization Steps	Final Polymerization Step				
96P		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled		Light polymerized	24 hr at 120°C	5	3	3	
98P		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled		Light polymerized	20 hr at 120°C	5	5	4	
99P		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled		Light polymerized	24 hr at 120°C	8	6	6	
102Q		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled		Light polymerized	24 hr at 121°C	8	6	6	
103Q		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled		Light polymerized	24 hr at 121°C	8	7	6	
104R		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled		Light polymerized	24 hr at 121°C	8	5	2	
105R		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled		Light polymerized	24 hr at 121°C	8	5	3	
106R		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled		Light polymerized	24 hr at 121°C	7	5	3	

TABLE 10. (Continued) SELECTED LIST OF SHEETS POSSESSING LEAST YELLOWNESS AFTER LIGHT EXPOSURE AND SHOWING LEAST CHANGE IN YELLOWNESS ON LIGHT EXPOSURE

Sample Number	Methyl α-Chloroacrylate Purity (Mole %)	Inhibitors Added	Purification Steps	Catalysts and Other Added Agents	Polymerization Conditions: Initial Polymerization Steps	Polymerization Conditions: Final Polymerization Step	Final Visual Color Rating	Final Yellowness Index Rating	Yellowing Color Change Rating	Heat Stability
107R		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled		Light polymerized	24 hr at 121°C	8	6	5	
109S		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled		Light polymerized	24 hr at 121°C	8	7	6	
110S		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	5 ml. of air passed through 1000 gm of monomer at (-35°C)	Light polymerized	24 hr at 121°C	8	6	5	
112S		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.02% Benzoyl peroxide	17 hr at 25°C 8 hr at 49°C 88 hr at 60°C	24 hr at 121°C	7	5	4	
120T		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.013% Benzoyl peroxide	16 hr at 25°C 9 hr at 48°C 17 hr at 60°C	24 hr at 120°C	6	5	5	
121T		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.013% Benzoyl peroxide	Light polymerized	24 hr at 120°C	6	6	5	
122		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled		Light polymerized	24 hr at 120°C	7	5	5	
133	99.9%	p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled		Light polymerized	24 hr at 120°C	7	6	4	

TABLE 10. (Continued) SELECTED LIST OF SHEETS POSSESSING LEAST YELLOWNESS AFTER LIGHT EXPOSURE AND SHOWING LEAST CHANGE IN YELLOWNESS ON LIGHT EXPOSURE

Sample Number	Methyl α-Chloroacrylate Purity (Mole %)	Inhibitors Added	Purification Steps	Catalysts and Other Added Agents	Polymerization Conditions		Final Visual Color Rating	Final Yellowness Index Rating	Yellowing Color Change Rating	Heat Stability
					Initial Polymerization Steps	Final Polymerization Step				
139	99.1%	p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.01% Aerosol OT 0.1% di-tert-Butyl peroxide	Light polymerized	24 hr at 118°C	11	6	2	
142U	98.2%	p-tert-Butylcatechol	Steam distillation Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distillation	0.01% Aerosol OT	Light polymerized	Heated at 120°C	1	3	3	
143U	98.2%	p-tert-Butylcatechol	Steam distillation Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distillation	0.01% Aerosol OT	Light polymerized	Heated at 120°C	1	1	1	
149	99.9%	p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.12% di-tert-Butyl peroxide 0.01% Aerosol OT	64 hr at 45°C	24 hr at 120°C	7	6	6	
150	99.6%	p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.014% Aerosol OT 0.28% di-tert-Butyl peroxide	Light polymerized	24 hr at 120°C	2	3	3	
152	99.9%	p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.0067% Aerosol OT 0.067% di-tert-Butyl peroxide	89 hr at 57°C	27 hr at 118°C	7	9	9	
155	99.0%	p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.024% Aerosol OT 0.062% di-tert-Butyl peroxide	Light polymerized	22 hr at 119°C 22 hr at 88°C	1	2	3	

TABLE 10. (Continued) SELECTED LIST OF SHEETS POSSESSING LEAST YELLOWNESS AFTER LIGHT EXPOSURE AND SHOWING LEAST CHANGE IN YELLOWNESS ON LIGHT EXPOSURE

Sample Number	Methyl ∝-Chloroacetylate Purity (Mole %)	Inhibitors Added	Purification Steps	Catalysts and Other Added Agents	Polymerization Conditions		Final Visual Color Rating	Final Yellowness Index Rating	Yellowing Color Change Rating	Heat Stability
					Initial Polymerization Steps	Final Polymerization Step				
157		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.04% di-tert-Butyl peroxide 0.016% DC-550 Silicone oil	40 hr at 60°C	22 hr at 119°C	1	2	2	
161		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.03% DC-550 Silicone oil 0.04% di-tert-Butyl peroxide	Heated at 60°C	Heated at 120°C	1	4	3	
163		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.025% DC-550 Silicone oil 0.031% di-tert-Butyl peroxide	Heated at 59°C	Heated 24 hr at 120°C	1	2	1	
167		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.07% di-tert-Butyl peroxide 0.05% DC-550 Silicone oil	Heated at 60°C	24 hr at 120°C	1	2	3	
169		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.06% di-tert-Butyl peroxide 0.05% DC-550 Silicone oil 0.24% Dibutyl tin diacetate	Light polymerized	24 hr at 119°C	5	5	5	
182		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.046% DC-550 Silicone oil 0.46% Dibutyl tin diacetate	16 hr at 60°C	24 hr at 118°C	9	5	5	
185		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.08% DC-550 Silicone oil 0.05% di-tert-Butyl peroxide	40 hr at 60°C	24 hr at 120°C	1	1	2	

TABLE 10. (Continued) SELECTED LIST OF SHEETS POSSESSING LEAST YELLOWNESS AFTER LIGHT EXPOSURE AND SHOWING LEAST CHANGE IN YELLOWNESS ON LIGHT EXPOSURE

Sample Number	Methyl α-Chloroacrylate Purity (Mole %)	Inhibitors Added	Purification Steps	Catalysts and Other Added Agents	Polymerization Conditions		Final Visual Color Rating	Final Yellowness Index Rating	Yellowing Color Change Rating	Heat Stability
					Initial Polymerization Steps	Final Polymerization Step				
188		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.08% di-tert-Butyl peroxide 0.05% DC-550 Silicone oil	63 hr at 60°C	44 hr at 120°C	1	2	2	
193		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled		Light polymerized	78 hr at 118°C	9	6	5	
201		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.04% DC-550 Silicone oil 0.05% di-tert-Butyl peroxide	Light polymerized	Heated at 120°C	3	5	4	
202V		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.04% DC-550 Silicone oil 0.05% di-tert-Butyl peroxide	3 hr at 25°C 240 hr at 60°C	31 hr at 116°C	4	7	7	
203V		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.04% DC-550 Silicone oil 0.05% di-tert-Butyl peroxide	3 hr at 25°C 240 hr at 60°C	31 hr at 116°C	4	6	7	
207		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.04% DC-550 Silicone oil 0.05% di-tert-Butyl peroxide	Light polymerized	24 hr at 116°C	5	5	6	
208		p-tert-Butylcatechol	Steam distilled Dried over $MgSO_4$ Dried over P_2O_5 Vacuum distilled	0.04% DC-550 Silicone oil 0.05% di-tert-Butyl peroxide	Light polymerized	24 hr at 116°C	7	11	11	

TABLE 10. (Continued) SELECTED LIST OF SHEETS POSSESSING LEAST YELLOWNESS AFTER LIGHT EXPOSURE AND SHOWING LEAST CHANGE IN YELLOWNESS ON LIGHT EXPOSURE

Sample Number	Methyl α-Chloroacrylate Purity (Mole %)	Inhibitors Added	Purification Steps	Catalysts and Other Added Agents	Polymerization Conditions		Final Visual Color Rating	Final Yellowness Index Rating	Yellowing Color Change Rating	Heat Stability
					Initial Polymerization Steps	Final Polymerization Step				
242	99.6%	p-tert-Butylcatechol	Steam distilled twice Freeze dried	0.05% DC-550 Silicone oil	Light polymerized	45 hr at 119°C	6	5	5	
259W	93.5%	p-tert-Butylcatechol	Steam distilled twice Freeze dried	0.05% DC-550 Silicone oil 0.05% di-tert-Butyl peroxide	Light polymerized	140 hr at 120°C	3	6	4	
260W	93.5%	p-tert-Butylcatechol	Steam distilled twice Freeze dried	0.05% DC-550 Silicone oil 0.05% di-tert-Butyl peroxide	Light polymerized	140 hr at 120°C	3	6	3	
266	98.9%	p-tert-Butylcatechol	Steam distilled twice Freeze dried Chromatographed through alumina	0.05% DC-550 Silicone oil 0.05% di-tert-Butyl peroxide	Light polymerized	24 hr at 120°C	7	6	5	
271X	98.7%	p-tert-Butylcatechol	Steam distilled twice Freeze dried Chromatographed through alumina	0.05% DC-550 Silicone oil 0.05% di-tert-Butyl peroxide	Light polymerized	Heated at 120°C	8	7	4	1
272X		p-tert-Butylcatechol	Steam distilled twice Freeze dried Chromatographed through alumina	0.05% DC-550 Silicone oil 0.05% di-tert-Butyl peroxide	Light polymerized	Heated at 120°C	4	4	3	
275		p-tert-Butylcatechol	Steam distilled twice Freeze dried Chromatographed through alumina	0.05% DC-550 Silicone oil 0.05% di-tert-Butyl peroxide	Light polymerized	24 hr at 121°C	7	7	4	2

TABLE 10. (Continued) SELECTED LIST OF SHEETS POSSESSING LEAST YELLOWNESS AFTER LIGHT EXPOSURE AND SHOWING LEAST CHANGE IN YELLOWNESS ON LIGHT EXPOSURE

Sample Number	Methyl α-Chloroacrylate Purity (Mole %)	Inhibitors Added	Purification Steps	Catalysts and Other Added Agents	Polymerization Conditions: Initial Polymerization Steps	Polymerization Conditions: Final Polymerization Step	Final Visual Color Rating	Final Yellowness Index Rating	Yellowing Color Change Rating	Heat Stability
278Y	99.7%	Picric Acid	Steam distilled Freeze dried	0.05% DC-550 Silicone oil 0.05% di-tert-Butyl peroxide	Light polymerized	48 hr at 121°C	3	4	1	10
280Y	99.7%	Picric Acid	Steam distilled Freeze dried	0.05% DC-550 Silicone oil 0.05% di-tert-Butyl peroxide	Light polymerized	48 hr at 121°C	3	4	1	
284	99.3%	Picric Acid	Steam distilled Freeze dried	0.05% DC-550 Silicone oil 0.05% di-tert-Butyl peroxide	Light polymerized	29 hr at 120°C	7	8	1	
286	98.8	Picric Acid	Steam distilled Freeze dried	0.05% DC-550 Silicone oil 0.05% di-tert-Butyl peroxide	Light polymerized	Heated at 120°C	2	4	1	1
291	98.6	Picric Acid	Steam distilled Freeze dried	0.05% DC-550 Silicone oil	Light polymerized	25 hr at 121°C	2	3	2	3
298	98.7	Picric Acid	Steam distilled Freeze dried	0.05% DC-550 Silicone oil	Light polymerized	24 hr at 120°C	3	5	2	
302	98.3	Picric Acid	Steam distilled Freeze dried	0.05% DC-550 Silicone oil	Light polymerized	24 hr at 120°C	8	6	6	
305	98.5	Picric Acid	Steam distilled Freeze dried	0.05% DC-550 Silicone oil 0.05% di-Butyl tin diacetate	Light polymerized	20 hr at 120°C	8	7	7	4
307	99.1	Picric Acid	Steam distilled Freeze dried	0.05% DC-550 Silicone oil	Light polymerized	20 hr at 124°C	2	3	3	

TABLE 10. (Continued) SELECTED LIST OF SHEETS POSSESSING LEAST YELLOWNESS AFTER LIGHT EXPOSURE AND SHOWING LEAST CHANGE IN YELLOWNESS ON LIGHT EXPOSURE

Sample Number	Methyl α-Chloroacrylate Purity (Mole %)	Inhibitors Added	Purification Steps	Catalysts and Other Added Agents	Polymerization Conditions		Final Visual Color Rating	Final Yellowness Index Rating	Yellowing Color Change Rating	Heat Stability
					Initial Polymerization Steps	Final Polymerization Step				
310	98.6%	Picric Acid	Steam distillation Freeze dried	0.05% DC-550 Silicone oil	Light polymerized	Heated at 120°C	1	1	1	
313	97.4%	Picric Acid	Steam distilled Freeze dried	0.05% DC-550 Silicone oil	Light polymerized	18 hr at 125°C	3	4	4	5
317	96.7%	Picric Acid	Steam distilled Freeze dried	0.01% DC-550 Silicone oil	Light polymerized	20 hr at 120°C	2	3	2	
318	97.3%	Picric Acid	Steam distilled Freeze dried	0.05% DC-550 Silicone oil	Light polymerized	23 hr at 126°C	7	6	5	
320	96.7%	Picric Acid	Steam distilled Freeze dried	0.05% DC-550 Silicone oil	Light polymerized	Heated at 120°C	5	5	3	
321	98.6%	Picric Acid	Steam distilled Freeze dried Chromatographed through alumina	0.05% DC-550 Silicone oil	Light polymerized	19 hr at 125°C	5	5	3	5
323		Picric Acid	Steam distilled Freeze dried		Light polymerized	16 hr at 126°C	8	7		
324	96.5%	Picric Acid	Steam distilled Freeze dried Chromatographed through alumina	0.05% DC-550 Silicone oil	Light polymerized	20 hr at 126°C	5	6	6	6
327	96.8%	Picric Acid	Steam distilled Freeze dried Chromatographed through alumina	0.05% DC-550 Silicone oil	Light polymerized	19 hr at 125°C	5	5	2	6
328	97.6%	Picric Acid	Steam distilled Freeze dried Chromatographed through alumina	0.05% DC-550 Silicone oil	Light polymerized	31 hr at 122°C	9	6	6	3

TABLE 10. (Continued) SELECTED LIST OF SHEETS POSSESSING LEAST YELLOWNESS AFTER LIGHT EXPOSURE AND SHOWING LEAST CHANGE IN YELLOWNESS ON LIGHT EXPOSURE

Sample Number	Methyl ∝-Chloroacrylate Purity (Mole %)	Inhibitors Added	Purification Steps	Catalysts and Other Added Agents	Polymerization Conditions: Initial Polymerization Steps	Polymerization Conditions: Final Polymerization Step	Final Visual Color Rating	Final Yellowness Index Rating	Yellowing Color Change Rating	Heat Stability
329	97.0%	Picric Acid	Steam distilled Freeze dried	0.05% DC-550 Silicone oil	Light polymerized	19 hr at 124°C	5	5	4	3
330	97.5%	Picric Acid	Steam distilled Freeze dried	0.05% DC-550 Silicone oil	Light polymerized	19 hr at 126°C	5	7	6	5
331	97.5%	Picric Acid	Steam distilled Freeze dried	0.05% DC-550 Silicone oil	Light polymerized	19 hr at 126°C	5	6	5	5
332	96.7%	Picric Acid	Steam distilled Freeze dried	0.05% DC-550 Silicone oil	Light polymerized	21 hr at 125°C	4	5	4	5
333	97.2%	Picric Acid	Steam distilled Freeze dried	0.05% DC-550 Silicone oil	Light polymerized	18 hr at 125°C	4	4	3	3
334	96.6%	Picric Acid	Steam distilled Freeze dried	0.05% DC-550 Silicone oil	Light polymerized	20 hr at 126°C	1	2	1	5
335	96.7%	Picric Acid	Steam distilled Freeze dried	0.05% DC-550 Silicone oil	Light polymerized	26 hr at 124°C	5	4	4	5
336	97.7%	Picric Acid	Steam distilled Freeze dried	0.05% DC-550 Silicone oil	Light polymerized	21 hr at 122°C	6	5	5	5
337	97.1%	Picric Acid	Steam distilled Freeze dried	0.05% DC-550 Silicone oil 0.0047% "Uvinul 490"	Light polymerized	Heated 22 hr at 124°C	1	14	1	5
338	96.9%	Picric Acid	Steam distilled Freeze dried	0.05% DC-550 Silicone oil	Light polymerized	Heated at 120°C	1	2	1	6
342	97.8%	Picric Acid	Steam distilled Freeze dried	0.05% DC-550 Silicone oil 0.005% "Uvinul 490"	Light polymerized	22 hr at 121°C	5	8	4	
344		Picric Acid	Steam distilled Freeze dried	0.05% DC-550 Silicone oil 0.01% "Uvinul 490"	Light polymerized	23 hr at 121°C	3	10	2	6
345		Picric Acid	Steam distilled Freeze dried	0.05% DC-550 Silicone oil 0.01% "Uvinul 490"	Light polymerized	20 hr at 121°C	3	13	3	7

TABLE 10. (Continued) SELECTED LIST OF SHEETS POSSESSING LEAST YELLOWNESS AFTER LIGHT EXPOSURE AND SHOWING LEAST CHANGE IN YELLOWNESS ON LIGHT EXPOSURE

Sample Number	Methyl α-Chloroacrylate Purity (Mole %)	Inhibitors Added	Purification Steps	Catalysts and Other Added Agents	Polymerization Conditions		Final Visual Color Rating	Final Yellowness Index Rating	Yellowing Color Change Rating	Heat Stability
					Initial Polymerization Steps	Final Polymerization Step				
348		Picric Acid	Steam distilled Freeze dried	0.05% DC-550 Silicone oil 0.05% "Uvinul 490"	Light polymerized	Heated at 120°C	3	16	1	7
349Z	99.8%	Picric Acid	Steam distilled Freeze dried	0.05% DC-550 Silicone oil 0.02% "Uvinul 490"	Light polymerized	24 hr at 120°C	1	16	1	
350Z	99.8%	Picric Acid	Steam distilled Freeze dried	0.05% DC-550 Silicone oil 0.03% "Uvinul 490"	Light polymerized	24 hr at 120°C	1	15	1	
351Z	99.8%	Picric Acid	Steam distilled Freeze dried	0.05% DC-550 Silicone oil 0.04% "Uvinul 490"	Light polymerized	24 hr at 120°C	3	16	1	
355		Picric Acid	Steam distilled Freeze dried	0.05% "Uvinul 490"	Light polymerized	24 hr at 120°C	3	16	1	
356		Picric Acid	Steam distilled Freeze dried	0.05% "Uvinul 490"	Light polymerized	22 hr at 120°C	3	18	1	3
357		Picric Acid	Steam distilled Freeze dried	0.05% "Uvinul 490"	Light polymerized	24 hr at 121°C	3	15	1	3
358	98.3%	Picric Acid	Steam distilled Freeze dried	0.01% "Uvinul 490"	Light polymerized	23 hr at 121°C	4	14	1	2
359	97.3%	Picric Acid	Steam distilled Freeze dried	0.01% "Uvinul 490"	Light polymerized	14 hr at 120°C	3	13	1	3
360	97.3%	Picric Acid	Steam distilled Freeze dried	0.01% "Uvinul 490"	Light polymerized	21 hr at 120°C	4	14	1	0
361	98.2%	Picric Acid	Steam distilled Freeze dried	0.01% "Uvinul 490"	Light polymerized	21 hr at 120°C	3	12	1	3

TABLE 10. (Continued) SELECTED LIST OF SHEETS POSSESSING LEAST YELLOWNESS AFTER LIGHT EXPOSURE AND SHOWING LEAST CHANGE IN YELLOWNESS ON LIGHT EXPOSURE

Sample Number	Methyl α-Chloroacrylate Purity (Mole %)	Inhibitors Added	Purification Steps	Catalysts and Other Added Agents	Polymerization Conditions		Final Visual Color Rating	Final Yellowness Index Rating	Yellowing Color Change Rating	Heat Stability
					Initial Polymerization Steps	Final Polymerization Step				
363	97.1%	Picric Acid	Steam distilled Freeze dried	0.01% "Uvinul 490"	Light polymerized	24 hr at 121°C	4	13	1	3
364	97.1%	Picric Acid	Steam distilled Freeze dried	0.01% "Uvinul 490"	Light polymerized	19 hr at 119°C	4	11	2	2
365	97.3%	Picric Acid	Steam distilled Freeze dried	0.01% "Uvinul 490"	Light polymerized	Heated at 120°C	4	13	1	1
366	97.0%	Picric Acid	Steam distilled Freeze dried	0.01% "Uvinul 490"	Light polymerized	Heated at 120°C	4	14	1	2
368	96.7%	Picric Acid	Steam distilled Freeze dried	0.005% "Uvinul 490"	Light polymerized	28 hr at 120°C	4	10	2	3
370		Picric Acid	Steam distilled Freeze dried		Light polymerized	26 hr at 121°C	4	15	4	
371	96.7%	Picric Acid	Steam distilled Freeze dried	0.01% "Uvinul 490"	Light polymerized	26 hr at 121°C	4	12	2	2
377	97.7%	Picric Acid	Steam distilled Freeze dried	0.005% "Uvinul 490"	Light polymerized	17 hr at 120°C	4	10	1	2
385AA	98.1%	Picric Acid	Steam distilled Freeze dried	0.05% "Uvinul 400"	Light polymerized	24 hr at 117°C	2	6	1	3
386AA	98.1%	Picric Acid	Steam distilled Freeze dried	0.01% "Uvinul 490"	Light polymerized	18 hr at 120°C	3	12	1	2
388		Picric Acid	Steam distilled Freeze dried	0.05% Benzoyl peroxide	24 hr at 35°C 16 hr at 45°C	18 hr at 120°C	8	8	5	3
389		Picric Acid	Steam distilled Freeze dried	0.05% Benzoyl peroxide	18 hr at 45°C	20 hr at 120°C	7	7	4	3
390	98.5%	Picric Acid	Steam distilled Freeze dried Dried over P_2O_5	0.05% Benzoyl peroxide	62 hr at 45°C	24 hr at 120°C	7	6	3	7

TABLE 10. (Continued) SELECTED LIST OF SHEETS POSSESSING LEAST YELLOWNESS AFTER LIGHT EXPOSURE AND SHOWING LEAST CHANGE IN YELLOWNESS ON LIGHT EXPOSURE

Sample Number	Methyl α-Chloroacrylate Purity (Mole %)	Inhibitors Added	Purification Steps	Catalysts and Other Added Agents	Polymerization Conditions		Final Visual Color Rating	Final Yellowness Index Rating	Yellowing Color Change Rating	Heat Stability
					Initial Polymerization Steps	Final Polymerization Step				
392	95.9%	Picric Acid	Steam distilled Freeze dried Dried over P_2O_5	0.05% Benzoyl peroxide	24 hr at 45°C	21 hr at 120°C	10	7	2	3
397BB		Picric Acid	Steam distilled Freeze dried		Light polymerized	24 hr at 119°C	10	6	4	
398BB		Picric Acid	Steam distilled Freeze dried		Light polymerized	24 hr at 119°C	10	5	3	
400CC		Picric Acid	Steam distilled Freeze dried Dried over P_2O_5	0.025% Benzoyl peroxide	23 hr at 44°C	25 hr at 117°C	12	8	1	5
401CC	97.5%	Picric Acid	Steam distilled Freeze dried	0.025% Benzoyl peroxide	25 hr at 44°C	20 hr at 120°C	8	7	4	5
406	98.3%	Picric Acid	Steam distilled Freeze dried	0.025% Benzoyl peroxide	21 hr at 45°C	18 hr at 121°C	10	6	4	4
412	98.8%	p-tert-Butylcatechol	Steam distilled Freeze dried Dried over P_2O_5	0.025% Benzoyl peroxide 0.05% DC-550 Silicone oil	23 hr at 43°C	23 hr at 120°C	10	6	2	8
421	98.9%	p-tert-Butylcatechol	Steam distilled Freeze dried Dried over P_2O_5	0.05% DC-550 Silicone oil 0.025% Benzoyl peroxide	33 hr at 49°C	27 hr at 118°C	10	4	1	6
429	98.2%	p-tert-Butylcatechol	Steam distilled Freeze dried Dried over P_2O_5	0.025% Benzoyl peroxide 0.05% DC-550 Silicone oil	Light polymerized	24 hr at 120°C	10	9	2	8
434		p-tert-Butylcatechol	Steam distilled Freeze dried Dried over P_2O_5	0.05% DC-550 Silicone oil	Light polymerized	19 hr at 120°C	11	8	1	7
435	98.7%	Picric Acid	Steam distilled Freeze dried Dried over P_2O_5	0.05% DC-550 Silicone oil	Light polymerized	2 hr at 125°C 2 hr at 140°C	6	5	2	8

TABLE 10. (Continued) SELECTED LIST OF SHEETS POSSESSING LEAST YELLOWNESS AFTER LIGHT EXPOSURE AND SHOWING LEAST CHANGE IN YELLOWNESS ON LIGHT EXPOSURE

Sample Number	Methyl α-Chloroacrylate Purity (Mole %)	Inhibitors Added	Purification Steps	Catalysts and Other Added Agents	Polymerization Conditions		Final Visual Color Rating	Final Yellowness Index Rating	Yellowing Color Change Rating	Heat Stability
					Initial Polymerization Steps	Final Polymerization Step				
437		p-tert-Butylcatechol	Steam distilled Freeze dried Dried over P_2O_5	0.05% DC-550 Silicone oil 0.05% Benzoyl peroxide	Light polymerized	24 hr at 120°C	10	9	1	7
438		Picric Acid p-tert-Butylcatechol (5% of weight of picric acid)	Steam distilled Freeze dried Dried over P_2O_5	0.025% Benzoyl peroxide 0.05% DC-550 Silicone oil	Light polymerized	23 hr at 121°C	6	7	1	8
439	98.8%	Picric Acid p-tert-Butylcatechol (5% of weight of picric acid)	Steam distilled Freeze dried Dried over P_2O_5	0.025% Benzoyl peroxide 0.05% DC-550 Silicone oil	Light polymerized	21 hr at 120°C	10	10	1	8
440		Picric Acid p-tert-Butylcatechol (2 1/2% of weight of picric acid)	Steam distilled Freeze dried Dried over P_2O_5	0.025% Benzoyl peroxide 0.05% DC-550 Silicone oil	Light polymerized	22 hr at 120°C	10	6	1	6
442		Picric Acid p-tert-Butylcatechol (2.5% of weight of picric acid)	Steam distilled Freeze dried Dried over P_2O_5	0.025% Benzoyl peroxide 0.05% DC-550 Silicone oil	58 hr at 25°C 9 hr at 50°C	33 hr at 127°C	11	8	1	6
443		Picric Acid p-tert-Butylcatechol (2.5% of weight of picric acid)	Steam distilled Freeze dried Dried over P_2O_5	0.025% Benzoyl peroxide	12 hr at 49°C	17 hr at 121°C	11	8	1	6
445		Picric Acid p-tert-Butylcatechol (2 1/2% of weight of picric acid)	Steam distilled Freeze dried Dried over P_2O_5	0.025% Benzoyl peroxide	20 hr at 50°C	19 hr at 122°C	11	9	2	5
446		Picric Acid p-tert-Butylcatechol (2.5% of weight of picric acid)	Steam distilled Freeze dried Dried over P_2O_5	0.025% Benzoyl peroxide	20 hr at 49°C	19 hr at 120°C	13	9	2	6

TABLE 10. (Continued) SELECTED LIST OF SHEETS POSSESSING LEAST YELLOWNESS AFTER LIGHT EXPOSURE AND SHOWING LEAST CHANGE IN YELLOWNESS ON LIGHT EXPOSURE

Sample Number	Methyl α-Chloroacrylate Purity (Mole %)	Inhibitors Added	Purification Steps	Catalysts and Other Added Agents	Polymerization Conditions		Final Visual Color Rating	Final Yellowness Index Rating	Yellowing Color Change Rating	Heat Stability
					Initial Polymerization Steps	Final Polymerization Step				
449		Picric Acid p-tert-Butylcatechol (2.5% of weight of picric acid)	Steam distilled Freeze dried	0.021% Porofor N	18 hr at 49°C 14 hr at 60°C	23 hr at 122°C		15	1	4
452		Picric Acid p-tert-Butylcatechol (2.5% of weight of picric acid)	Steam distilled Freeze dried Dried over P_2O_5 Flash vacuum distilled	0.025% Benzoyl peroxide 1 mole % of acetic acid	12 hr at 60°C	24 hr at 121°C		13	1	4
455		Picric Acid p-tert-Butylcatechol (2.5% of weight of picric acid)	Steam distilled Freeze dried Dried over P_2O_5 Flash vacuum distilled	0.025% Benzoyl peroxide 0.05% DC-550 Silicone oil 0.5% mole of acetic acid	12 hr at 60°C	29 hr at 121°C	11	9	1	5
456	98.1%	p-tert-Butylcatechol	Steam distilled Freeze dried Dried with silica gel	0.025% Benzoyl peroxide	37 hr at 51°C 9 hr at 60°C	Heated at 120°C	11	11	1	7
461DD		p-tert-Butylcatechol	Steam distilled Freeze dried Dried over P_2O_5 Flash vacuum distilled	0.025% Benzoyl peroxide Cast in glass treated with sodium stearate	37 hr at 51°C 9 hr at 60°C	Heated at 120°C		8	1	6
463		Picric Acid p-tert-Butylcatechol (2 1/2% of weight of picric acid)	Steam distilled Freeze dried	0.44% Acetic anhydride 0.025% Benzoyl peroxide	19 hr at 60°C	24 hr at 120°C	11	9	1	3
467DD		p-tert-Butylcatechol	Steam distilled Freeze dried Dried over P_2O_5 Flash vacuum distilled	0.025% Benzoyl peroxide 0.0075% HCl	24 hr at 25°C 14 hr at 60°C	24 hr at 120°C	11	10	1	

TABLE 10. (Continued) SELECTED LIST OF SHEETS POSSESSING LEAST YELLOWNESS AFTER LIGHT EXPOSURE AND SHOWING LEAST CHANGE IN YELLOWNESS ON LIGHT EXPOSURE

Sample Number	Methyl α-Chloroacrylate Purity (Mole %)	Inhibitors Added	Purification Steps	Catalysts and Other Added Agents	Polymerization Conditions		Final Visual Color Rating	Final Yellowness Index Rating	Yellowing Color Change Rating	Heat Stability
					Initial Polymerization Steps	Final Polymerization Step				
472		Picric Acid p-tert-Butylcatechol (2.5% of weight of picric acid)	Steam distilled Freeze dried Dried over P_2O_5	0.025% Benzoyl peroxide 0.05% DC-550 Silicone oil	66 hr at 30°C 7 hr at 50°C	24 hr at 121°C	10	8	1	
479		Picric Acid p-tert-Butylcatechol (2.5% of weight of picric acid)	Steam distilled Freeze dried Dried over P_2O_5	0.5 mole % of chloroacrylic acid 0.025% Benzoyl peroxide	14 hr at 60°C	14 hr at 118°C	11	11	1	
491		p-tert-Butylcatechol	Steam distilled Freeze dried Dried over P_2O_5 Flash vacuum distilled	0.032% di-tert-Butyl peroxide 0.024% DC-550 Silicone oil Glass casting plates washed with a 1% solution of diethyl aniline in methanol	Light polymerized	24 hr at 120°C		15	1	
493		p-tert-Butylcatechol	Steam distilled Freeze dried Dried over P_2O_5 Flash vacuum distilled	0.032% di-tert-Butyl peroxide 0.024% DC-550 Silicone oil 0.12% Dibutyl tin diacetate	24 hr at 57°C	19 hr at 121°C		13	2	11
501		p-tert-Butylcatechol	Steam distilled Freeze dried Dried over P_2O_5 Flash vacuum distilled	0.032% di-tert-Butyl proxide 0.024% DC-550 Silicone oil 0.12% Dibutyl tin diacetate	24 hr at 25°C 19 hr at 58°C	25 hr at 120°C		10	2	11
506		p-tert-Butylcatechol	Steam distilled Freeze dried Dried over P_2O_5 Flash vacuum distilled twice	0.024% DC-550 Silicone oil 0.032% di-tert-Butyl peroxide 0.12% Dibutyl tin diacetate	24 hr at 25°C 14 hr at 58°C	24 hr at 120°C		14	1	10

TABLE 10. (Continued) SELECTED LIST OF SHEETS POSSESSING LEAST YELLOWNESS AFTER LIGHT EXPOSURE AND SHOWING LEAST CHANGE IN YELLOWNESS ON LIGHT EXPOSURE

Sample Number	Methyl α-Chloroacrylate Purity (Mole %)	Inhibitors Added	Purification Steps	Catalysts and Other Added Agents	Polymerization Conditions		Final Visual Color Rating	Final Yellowness Index Rating	Yellowing Color Change Rating	Heat Stability
					Initial Polymerization Steps	Final Polymerization Step				
516		p-tert-Butylcatechol	Steam distilled Freeze dried Dried over P_2O_5 Flash vacuum distilled	0.024% DC-550 Silicone oil 0.032% di-tert-Butyl peroxide 0.12% Dibutyl tin diacetate	30 hr at 25°C 14 hr at 58°C	24 hr at 120°C		8	2	
530		p-tert-Butylcatechol	Steam distilled Freeze dried Dried over P_2O_5 Flash vacuum distilled	0.06% Dibutyl tin diacetate 0.032% di-tert-Butyl peroxide	16 hr at 25°C 13 hr at 59°C	Heated at 120°C		13	2	7
533		p-tert-Butylcatechol	Steam distilled Freeze dried Dried over P_2O_5 Flash vacuum distilled	0.06% Dibutyl tin diacetate 0.032% di-tert-Butyl peroxide	16 hr at 25°C 20 hr at 58°C	Heated at 120°C		11	2	9
538		p-tert-Butylcatechol	Steam distilled Freeze dried Dried over P_2O_5 Flash vacuum distilled	0.06% Dibutyl tin diacetate 0.032% di-tert-Butyl peroxide	194 hr at 25°C 28 hr at 60°C	24 hr at 120°C		7	2	

2.2 Analysis of Results

For comparisons among these selected sheets, the final yellowness index rating obtained from Table 5 and the yellowness color change rating obtained from Table 6 are the most significant values. The final visual color rating obtained from Table 7 was included to show that for most of these sheets, the final visual color rating was generally the same order of magnitude as the final yellowness index rating. Notable exceptions occurred among the sheets from 337 to 386 which included many sheets containing ultraviolet absorbers. The ultraviolet absorbers introduced a yellow color to the sheets which did not appear very yellow to the eye, but whose yellowness index as measured on the photovolt meter was high.

In comparing the final yellowness index rating with the yellowness color change rating, two trends were noted. The first trend to be noted was that for a large number of these samples, the numbers for the two ratings were often the same. This observation means that for most of the samples possessing little yellowness after 1.5 years roof exposure, the change in yellowness is small if the final yellowness is slight. Thus, most of these samples were nearly colorless originally and their final yellowness was due only to a change in yellowness during light exposure. This behavior occurred in samples inhibited with picric acid and also in those inhibited with p-tert-butylcatechol; in samples light polymerized in the initial steps and in samples polymerized in the dark using heat and catalysts; and in samples purified by a variety of methods.

The yellowing of these samples is believed to be due to an impurity common to all these samples. The most likely impurity common to all these samples and present in varying amounts in the samples is thought to be residual (unpolymerized) monomer remaining in the polymer, which residual monomer reacts with oxygen of the air in the presence of light to cause yellowing in the polymer sheet. As part of our research program we therefore investigated conditions of polymerization, particularly the final polymerization step, to determine those conditions required to reduce or remove the residual monomer from the polymer.

It has been noted earlier that by incorporation of various hydroxyl containing compounds in methyl α-chloroacrylate a colorless polymer results. Examination of sheets containing methanol which were exposed to light indicated that the methanol also acted, to a certain extent, as a light stabilizer. In particular, it was noted in several sheets cast from the same monomer (notably sheets 48 and 49; 54 and 55, and 56; 76, 77, 78, and 79; 80, 81, and 82; 83, 84, and 86; 87, 88, and 89; 90, 91, and 93) that the sheets containing methanol possessed less final yellow color than the sheets cast from the same monomer to which no methanol was added. Unfortunately methanol even in low concentration lowers the heat stability of the polymer sheet as well as its heat distortion temperature so that it cannot be incorporated in "Gafite" sheets where these properties are important.

The second trend noted in these selected sheets was evidenced by a high number representing considerable yellowness for the final yellowness index accompanied by a very low number representing the change in yellowness on light exposure. These sheets were yellow after light exposure, but virtually all of the yellowness was present in the sheets as cast. This effect is first seen in the series of sheets starting with 337 and continuing to 386. With a few exceptions, this series of sheets was prepared with one of the ultraviolet absorbers, "Uvinul 400" or "Uvinul 490" present in the sheets. This series of sheets demonstrated that the presence of these ultraviolet absorbers in "Gafite" was effective in markedly reducing the increase of yellowness on light exposure. These absorbers did introduce some yellowness in the sheets originally, but as mentioned previously the eye does not see them to be as yellow as might be inferred from their final yellowness index rating. Noncontractual research previously had indicated that these ultraviolet absorbers manufactured by GAF stabilized "Gafite" sheets on exposure in a Fadeometer. These results are definite confirmations of the earlier experiments as extended to outdoor exposure tests. We believed that the incorporation of ultraviolet absorbers in "Gafite" merited considerable study under this contract as a means of increasing the light stability of "Gafite". Much of the experimental work was devoted to such a study.

From sheet 386 on, many of the sheets were prepared from monomer which had been treated with phosphorus pentoxide as the final step or next to last step prior to flash vacuum distillation. Of these selected sheets in the range from 386 to 592 several exhibit the same type of behavior as noted in the sheets containing ultraviolet absorbers. The sheets possess an initial yellow color which changes very little on light exposure. This behavior might be due to incorporation of the reaction product of the inhibitor, p-tert-butylcatechol, and phosphorus pentoxide in the sheet causing the initial yellowness coupled with low residual monomer content which would tend to prevent much color change on light exposure. Of the large number of sheets cast from 386 to 592 only a few of these sheets, those listed in Table 1 of this report, show a small color change on light exposure. This small percentage of sheets, possibly with low residual monomer content, roughly corresponds to the small percentage of the earlier sheets showing only a little color change on light exposure.

The summary analysis of the results of light stability tests on "Gafite" samples was extended by comparison of methods of preparation of the least yellow samples and the samples exhibiting least change in yellowness on light exposure. These comparisons and previous research on "Gafite" indicated that the following avenues of research were of merit in the development of light stable "Gafite" sheets:

1. Reduction of residual monomer content in "Gafite" sheets.
2. Incorporation of ultraviolet absorbers in "Gafite".
3. Incorporation of aliphatic hydroxyl containing compounds in "Gafite".

SECTION III. EXPERIMENTAL WORK ON IMPROVEMENT OF LIGHT AND HEAT STABILITY OF "GAFITE"

Investigations described in Sections I and II established the value of certain avenues of approach in obtaining "Gafite" of improved light and heat stability. Experimental work directed toward these approaches is described in this section together with some observations on heat and light stability tests.

3.1 Light Stability Tests on "Gafite"

In considering light stability tests on "Gafite" sheets we wished to choose or devise a method which would provide a rapid sensitive test for changes in "Gafite" sheet produced by light exposure. A simplified method had been employed as described in Section I. We believed that the test for light stability might be made more sensitive if instead of using sheets with polished surfaces, we used sheets with ground surfaces. It was thought that the grooves ground in the sheet would trap more light by reflection and scatter the light more than polished sheets. In addition such a method would be readily adaptable to use on small scale experiments in which the casting was made in glass tubes or vessels and not in casting cells yielding polished surfaces. A sample could be machined from such a casting and rough ground for use in a light exposure test.

To test the possibility and effects of such grinding with light exposure tests, the following experiments were performed. Four 1 1/2 in. x 3 in. samples were cut from "Gafite" Sheet 313 and finished as described in Table 11.

The opacity of these samples increased in order from 313I which was transparent to 313F to 313E to 313D which was translucent white from grinding.

The optical densities of these samples were measured using the simplified method for measuring yellowness index described in Section I.

TABLE 11. FINISHES ON SAMPLES FROM "GAFITE" SHEET 313

Sample No.	Finish	Thickness
313I	Polished surface as cast	0.249-0.251 in.
313F	Polished surfaces hand ground with 600A Emery Paper by wet polishing	0.247-0.253 in.
313E	Polished surfaces hand ground with No. 1 Dry Emery Paper	0.253-0.255 in.
313D	Polished surfaces wet ground with 220 Grit Emery Powder on rotating grinding wheel	0.253-0.255 in.

TABLE 12. OPTICAL DENSITIES OF SAMPLES OF "GAFITE" SHEET 313 USING SIMPLIFIED METHOD

Sample No.	Optical Density Simplified Method	Average
313I	0.059 0.050	0.055
313F	0.380 0.380	0.380
313E	0.900 0.890	0.895
313D	1.100 1.130	1.115

In Section I the previous samples of "Gafite" Sheet 313 possessed the following values (samples with polished surfaces).

TABLE 13. PREVIOUS OPTICAL DENSITIES OBTAINED ON "GAFITE" SHEET 313 USING SIMPLIFIED METHOD

Sample No.	Thickness	Optical Density Simplified Method
313F	0.250-0.256 in.	0.061
313R	0.249-0.250 in.	0.055

These results check those of the polished Sample 313I.

Next, the transmission curves were run on these same samples in a General Electric Recording Spectrophotometer. These curves are reproduced in Figures 11, 13, 15 and 17 inclusive. The curve (N) in these figures is the normal transmission curve with the sample in the normal position close to the measuring photocell. The curve (F) is the result obtained with the sample close to the entering light beam. The differences between the positions of curves (N) and (F) are representative of the haze or light scattering at the surface (and internally) of the samples. The greater the difference between curves (N) and (F) the greater the scattering or haze on and in the sheet samples.

After obtaining the above measurements on the samples, the samples were exposed for 500 hours in an Atlas Fadeometer and the above measurements were repeated.

The results obtained by the simplified method for measuring yellowness index after the 500 hours in a Fadeometer are recorded in Table 14.

TABLE 14. OPTICAL DENSITIES AFTER EXPOSURE IN FADEOMETER ON SAMPLES FROM "GAFITE" SHEET 313 USING SIMPLIFIED METHOD

Sample No.	Optical Density Simplified Method
313I	0.185
313F	0.540
313E	1.050
313D	1.500

The increase in optical density, as measured by the simplified method, due to exposure in the Fadeometer is tabulated in Table 15.

TABLE 15. INCREASE IN OPTICAL DENSITIES ON EXPOSURE IN FADEOMETER FOR SAMPLES FROM "GAFITE" SHEET 313 USING SIMPLIFIED METHOD

Sample No.	Increase in Optical Density Simplified Method
313I	0.130
313F	0.160
313E	0.155
313D	0.385

The sample 313D with the greatest grinding gives a density change about three times as great as the transparent sample 313I with the polished surfaces. The progression from transparent sample 313I through the other samples to sample 313D is not uniform, however. This may be due partly to the fact that the photovolt instrument employed is more responsive to small changes in optical density in samples with low optical density than in samples with high optical density.

We turn now to the results obtained with the more precise General Electric Recording Spectrophotometer equipped with integrator which was used in obtaining the transmission curves. The transmission curves on these samples after 500 hours in a Fadeometer are reproduced in Figures 12, 14, 16, and 18. The (N) and (F) designations have the meaning previously explained. Casual inspection of the curves shows that for all samples transmission in the wavelength region about 420 millimicrons is decreased by exposure in the Fadeometer. However, in the wavelength region about 700 millimicrons the transmission drop seems to progressively increase from samples 313I to 313F to 313E to 313D with increased opacity of the sample. To determine the exact changes, the transmission values were converted to optical densities, which are listed in Table 16.

TABLE 16. OPTICAL DENSITY VALUES FOR SAMPLES OF "GAFITE" SHEET 313 BEFORE AND AFTER FADEOMETER EXPOSURE AS DETERMINED BY GE RECORDING SPECTROPHOTOMETER

Sample No.	Type Curve		Wavelength	Percent Transmission	Optical Density	Increase in Optical Density on Fadeometer Exposure
313I	Before Fadeometer Exposure	(N)	420	89.3	0.049	---
		(F)	420	88.0	0.056	---
	After Fadeometer Exposure	(N)	420	69.0	0.161	0.112
		(F)	420	65.4	0.184	0.128
	Before Fadeometer Exposure	(N)	700	92.5	0.034	---
		(F)	700	91.6	0.038	---
	After Fadeometer Exposure	(N)	700	92.0	0.036	0.002
		(F)	700	89.0	0.051	0.013
313F	Before Fadeometer Exposure	(N)	420	81.3	0.090	---
		(F)	420	39.0	0.409	---
	After Fadeometer Exposure	(N)	420	56.4	0.249	0.159
		(F)	420	28.2	0.550	0.141
	Before Fadeometer Exposure	(N)	700	89.5	0.048	---
		(F)	700	52.2	0.282	---
	After Fadeometer Exposure	(N)	700	83.9	0.076	0.028
		(F)	700	53.0	0.276	0.006 (Decrease)
313E	Before Fadeometer Exposure	(N)	420	53.2	0.274	---
		(F)	420	9.2	1.036	---
	After Fadeometer Exposure	(N)	420	29.0	0.538	0.264
		(F)	420	5.7	1.244	0.208
	Before Fadeometer Exposure	(N)	700	65.0	0.187	---
		(F)	700	14.4	0.842	---

TABLE 16. (Continued) OPTICAL DENSITY VALUES FOR SAMPLES OF "GAFITE" SHEET 313
BEFORE AND AFTER FADEOMETER EXPOSURE AS DETERMINED
BY GE RECORDING SPECTROPHOTOMETER

Sample No.	Type Curve		Wavelength	Percent Transmission	Optical Density	Increase in Optical Density on Fadeometer Exposure
	After Fadeometer Exposure	(N)	700	54.1	0.267	0.080
		(F)	700	14.1	0.851	0.009
313D	Before Fadeometer Exposure	(N)	420	41.1	0.386	---
		(F)	420	5.60	1.252	---
	After Fadeometer Exposure	(N)	420	18.8	0.726	0.340
		(F)	420	2.50	1.602	0.350
	Before Fadeometer Exposure	(N)	700	51.5	0.288	---
		(F)	700	7.6	1.119	---
	After Fadeometer Exposure	(N)	700	34.3	0.465	0.177
		(F)	700	4.9	1.310	0.191

A survey of these results shows:

1. At 420 millimicrons wavelength in the (N) normal position there is a progressive increase in the differences in optical densities of these samples before and after Fadeometer exposure which increase corresponds to the increase in haze or grinding of the sample. The heavily ground sample 313D increases in optical density almost three times as much as does the polished sample 313I on Fadeometer exposure measured under these conditions.

2. At 420 millimicrons wavelength in the (F) far position there is a progressive increase in the difference in optical densities of these samples before and after Fadeometer exposure which increase corresponds to the increase in haze or grinding of the sample. The heavily ground sample 313D increases in optical density almost three times as much as does the polished sample 313I on Fadeometer exposure measured under these conditions.

3. Neither at 420 millimicrons nor at 700 millimicrons is there any regular difference within samples between the optical density increase on Fadeometer exposure in the (N) position as compared to the (F) position at the same wavelength. Whether the (N) or (F) value is larger varies with the samples in no discernible regular way.

4. At 700 millimicrons wavelength in the (N) normal position there is a progressive increase in the differences in optical densities of these samples before and after Fadeometer exposure. The increase in optical density corresponds to the increase in haze or grinding of the sample. The heavily ground sample 313D increases in optical density about 88 times as much as does the polished sample 313I on Fadeometer exposure measured under these conditions. With increase in grinding the changes in optical density on Fadeometer exposure also become apparent in wavelengths (around 700 millimicrons) far removed from the wavelengths (around 420) associated with yellowing in polished samples.

5. At 700 millimicrons wavelength in the (F) far position there is no regular increase in the differences in optical densities of these samples before and after Fadeometer exposure. The increase in optical density does not correspond in a regular way with increase of haze or grinding of the sample. However, the heavily ground sample 313D increases in optical density about 15 times as much as does the polished sample 313I on Fadeometer exposure measured under these conditions.

To the eye the difference between exposed and unexposed areas of the same sheet appears greater with the polished sheet than with the ground sheet, however. This behavior probably can be explained by the fact that it is easier to see a small change in a clear sheet than a large change in a more optically dense sheet. Also the fact that the change in a transparent sheet is concentrated more in the wavelength region associated with yellowing may make the color change more pronounced to the eye than with yellowing accompanied with decreased optical density at other wavelengths and colors besides the yellow region.

In summary, Fadeometer tests on ground samples produce a greater measurable optical density change than do tests on polished specimens and the change is noted over the whole spectra from 420 to 700 millimicrons rather than concentrated in the 420 millimicron wavelength region as noted with polished samples. When using visual observations to interpret Fadeometer results, the differences between exposed and unexposed areas of the same samples are more pronounced with polished samples. This ease of seeing color changes in polished sheets probably also means that visual comparisons of yellowing between different samples is easier with polished samples than with ground samples.

The simplified method for measuring yellowness index is very useful for polished samples, but with use of ground samples the density change occurs in a region which is difficult to read as precisely as desired on the photovoltmeter employed.

Depending on sample size available and whether the Fadeometer test is a screening test or is one whose optical density change is to be measured quantitatively, one may prefer polished surfaces or ground surfaces on the sample exposed in the Fadeometer. This information was useful in designing and interpreting experiments on light stability of "Gafite" sheets. For screening tests, the use of polished samples is probably preferable, whereas for measurement of optical changes occurring during Fadeometer tests the use of samples possessing ground surfaces will yield greater measurable changes in optical density.

Since most of the tests on effect of composition changes of "Gafite" on its light stability were screening tests, these light stability tests were carried out on samples with polished surfaces with visual evaluation of the results.

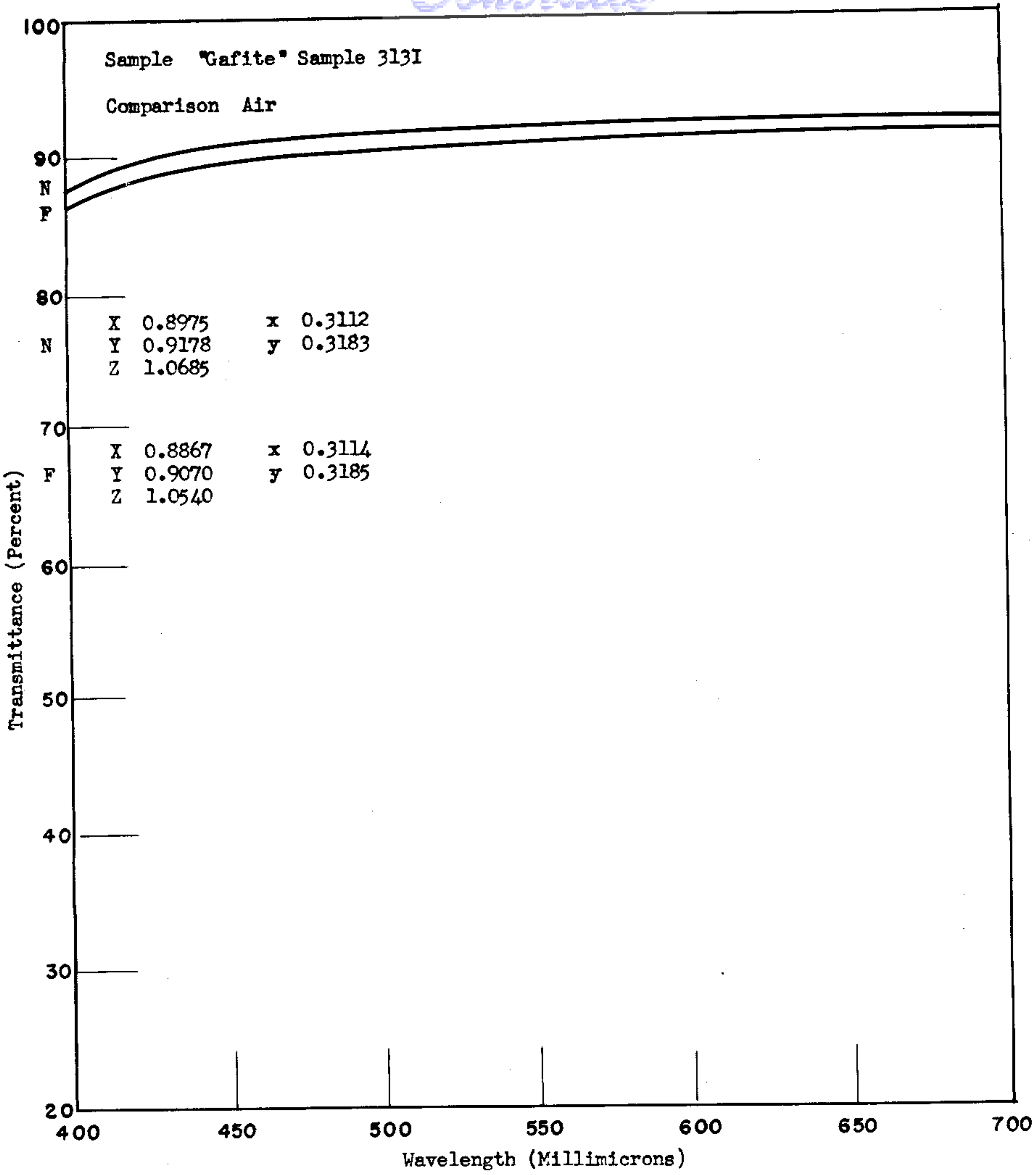


FIGURE 11. "GAFITE" SAMPLE 313I (WITH POLISHED SURFACES) PRIOR TO FADEOMETER TEST

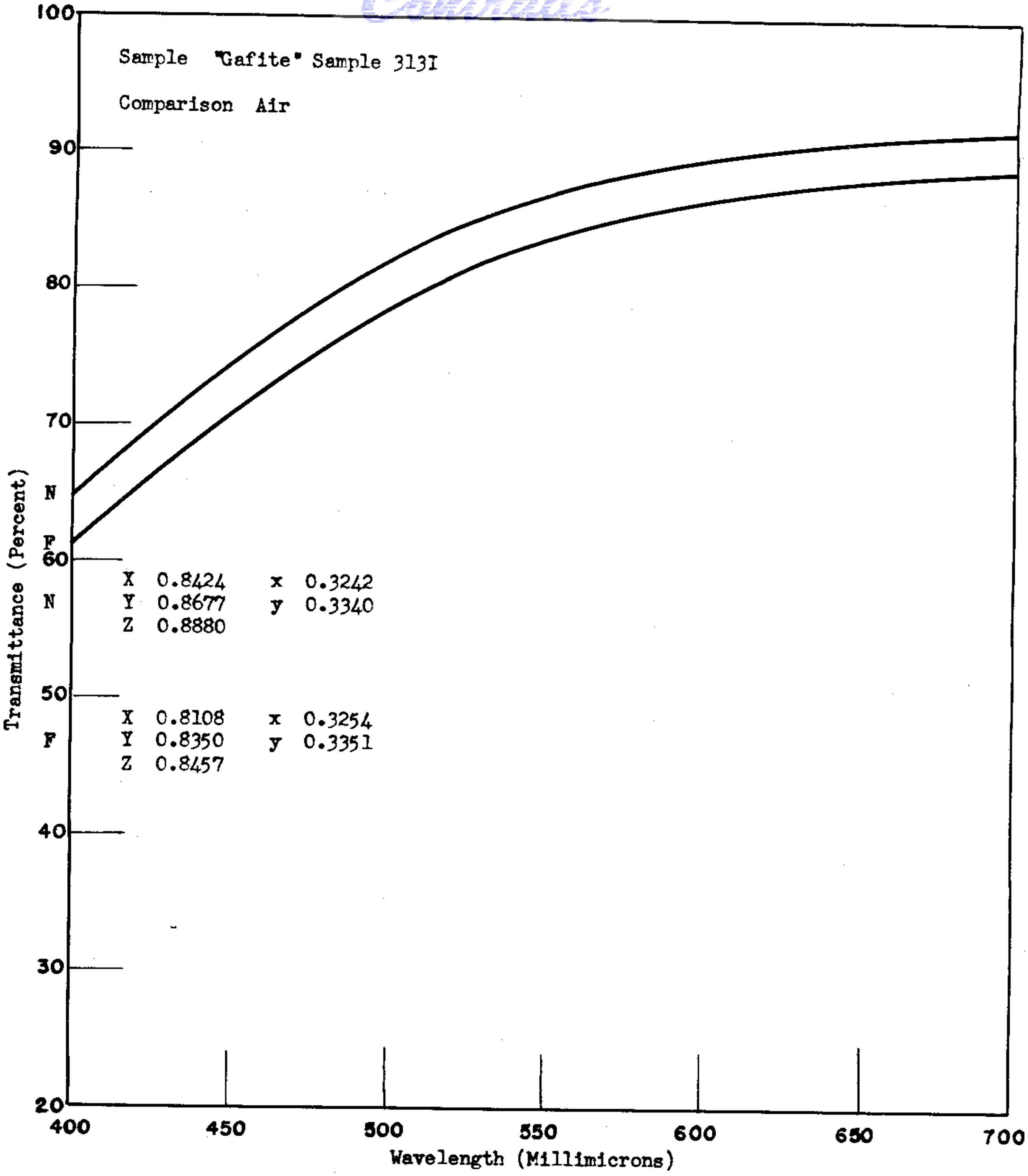


FIGURE 12. "GAFITE" SAMPLE 313I (WITH POLISHED SURFACES) AFTER 500 HOURS IN FADEOMETER

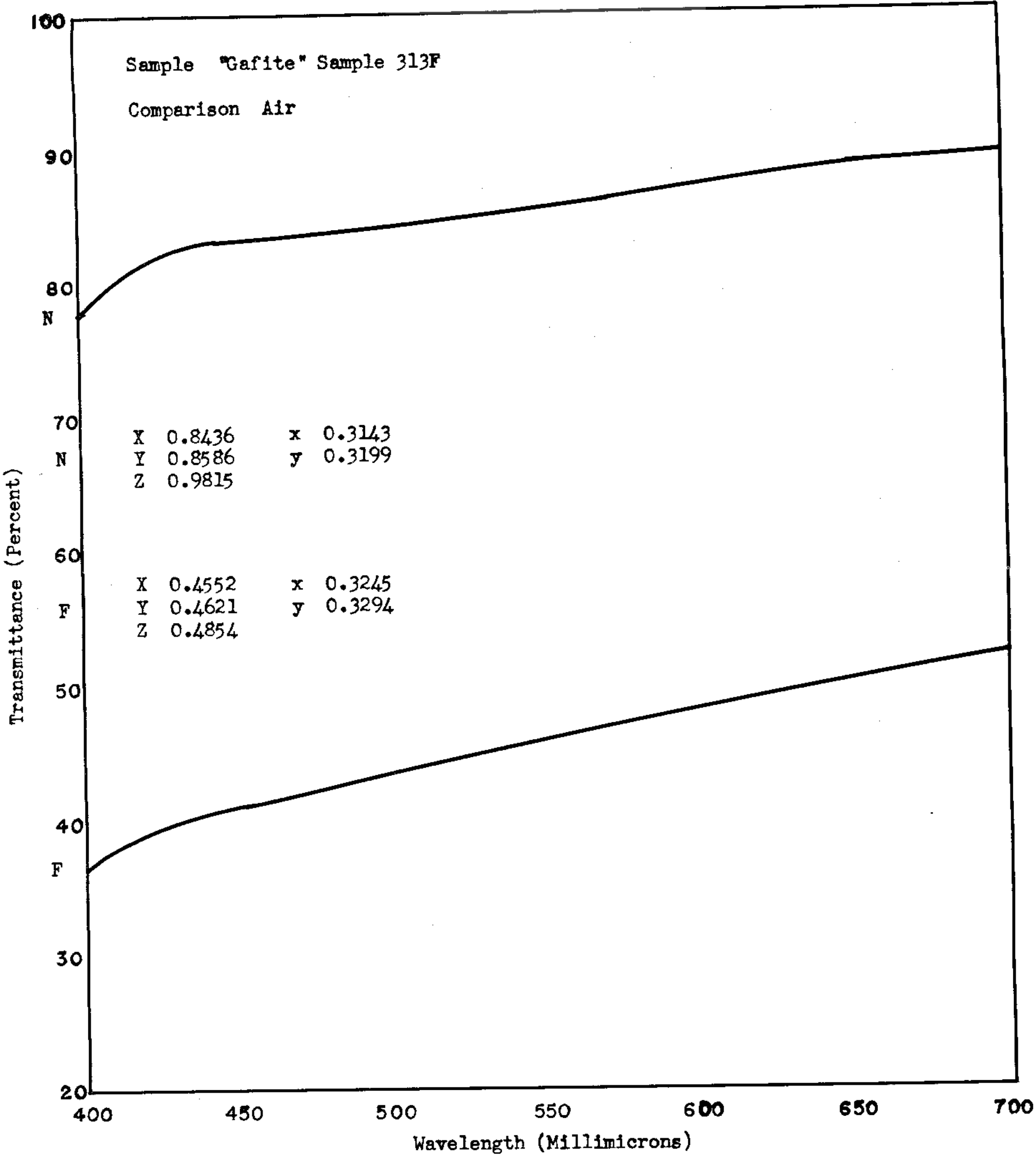


FIGURE 13. "GAFITE" SAMPLE 313F (GROUND BY HAND WITH 600A EMERY PAPER BY WET POLISHING) PRIOR TO FADEOMETER TEST

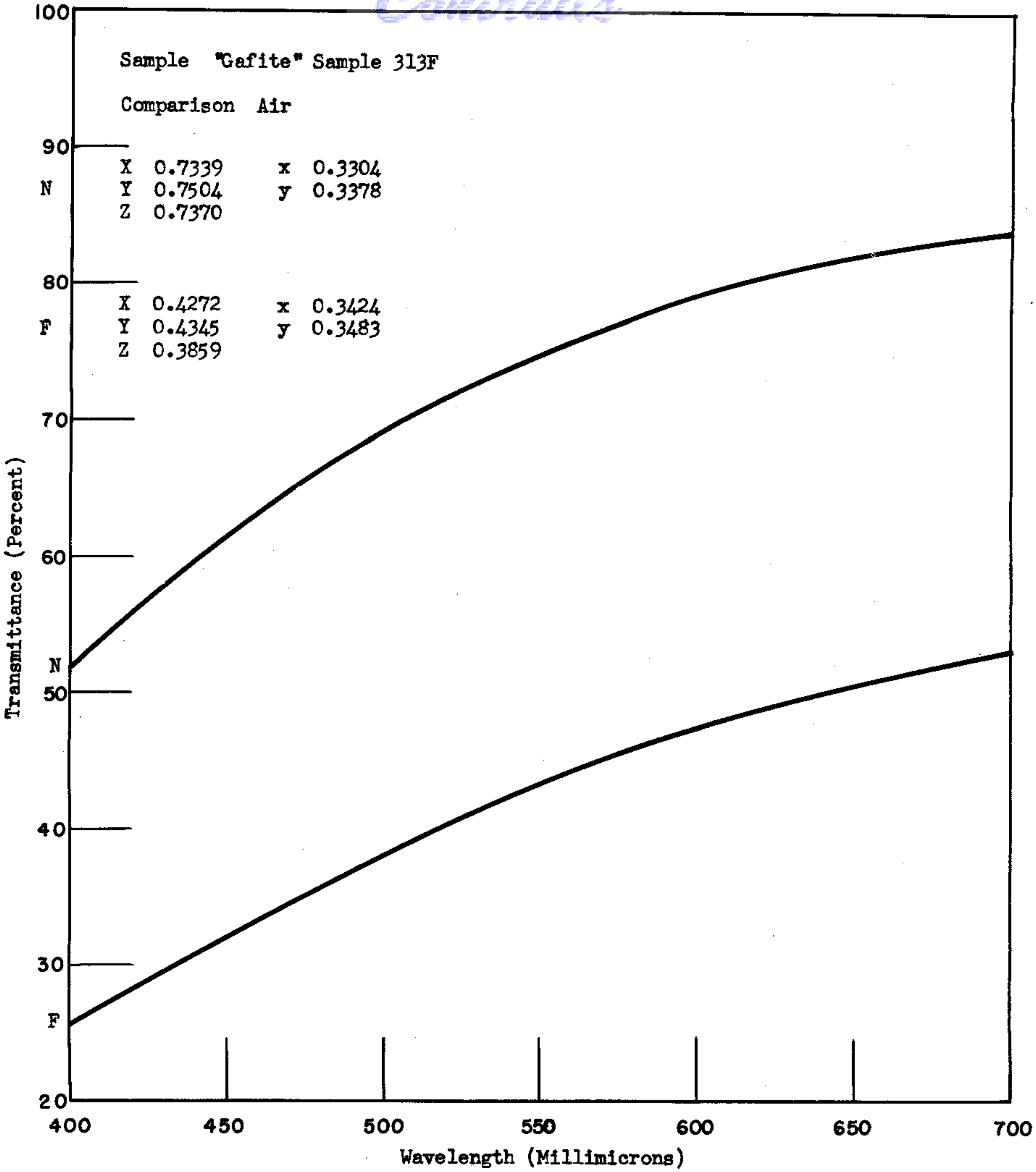


FIGURE 14. "GAFITE" SAMPLE 313F (GROUND BY HAND WITH 600A EMERY PAPER BY WET POLISHING) AFTER 500 HOURS IN FADEOMETER

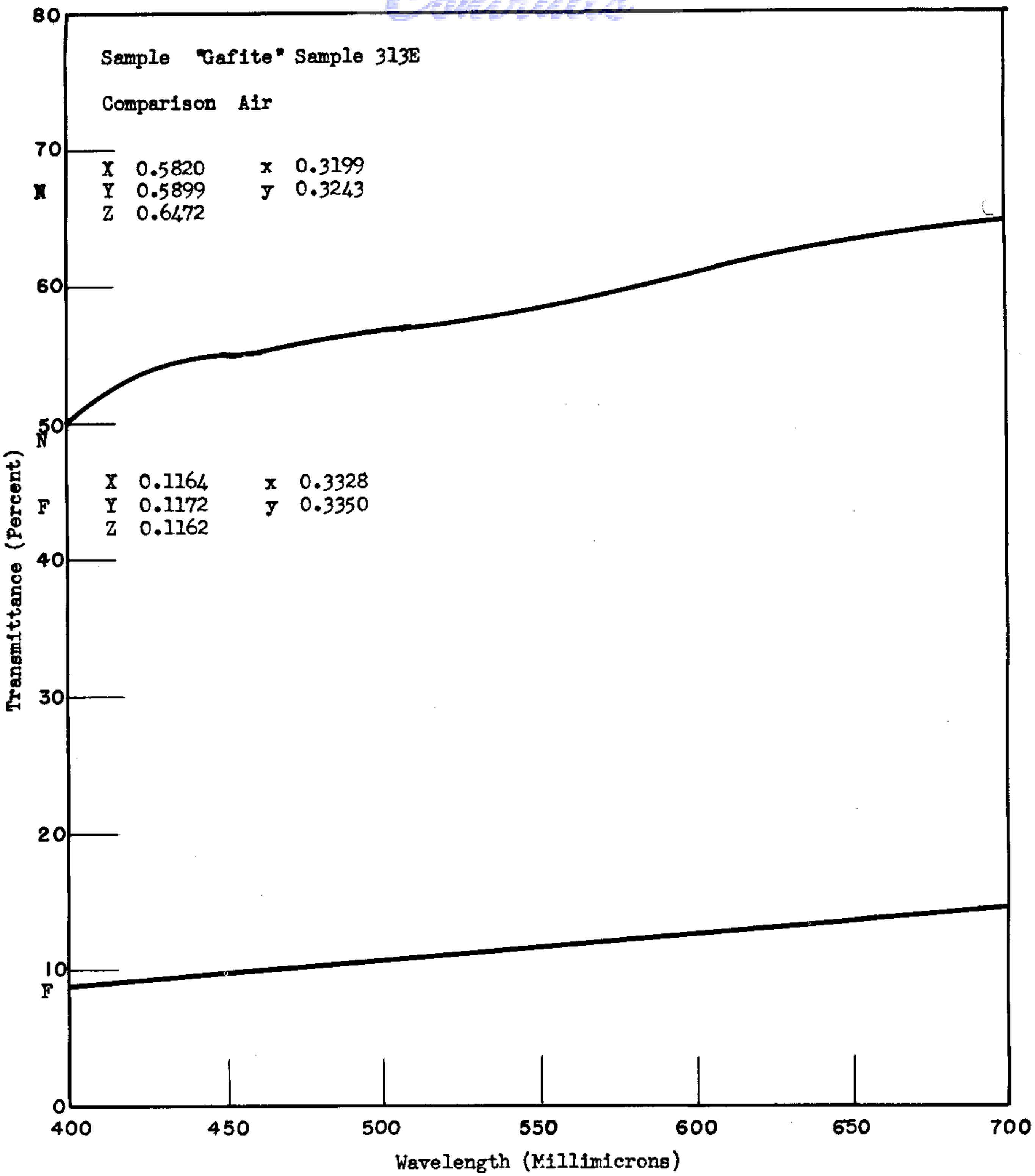


FIGURE 15. "GAFITE" SAMPLE 313E (GROUND BY HAND WITH DRY NO. 1 EMERY PAPER) PRIOR TO FADEOMETER TEST

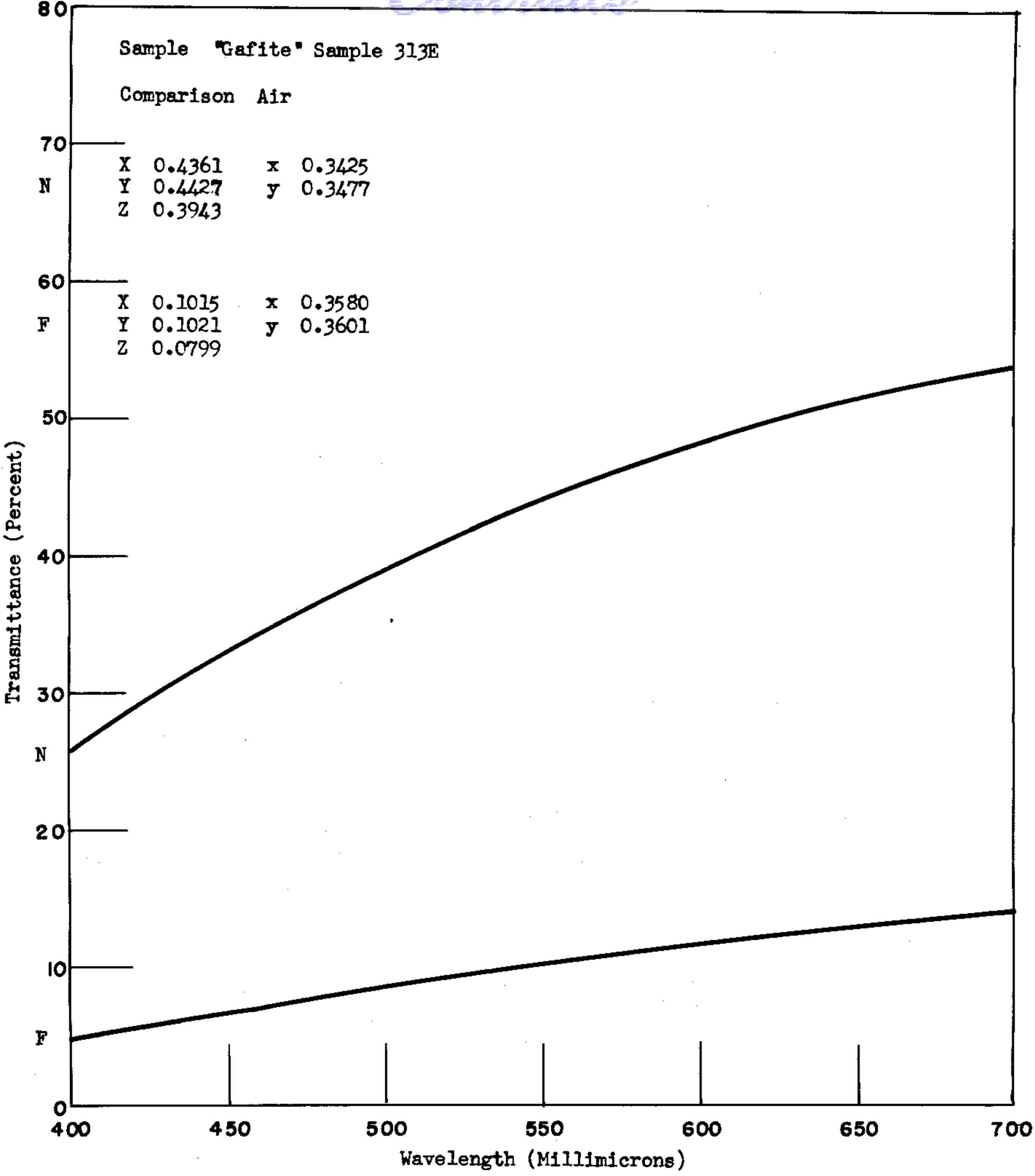


FIGURE 16. "GAFITE" SAMPLE 313E (GROUND BY HAND WITH DRY NO. 1 EMERY PAPER) AFTER 500 HOURS IN FADEOMETER

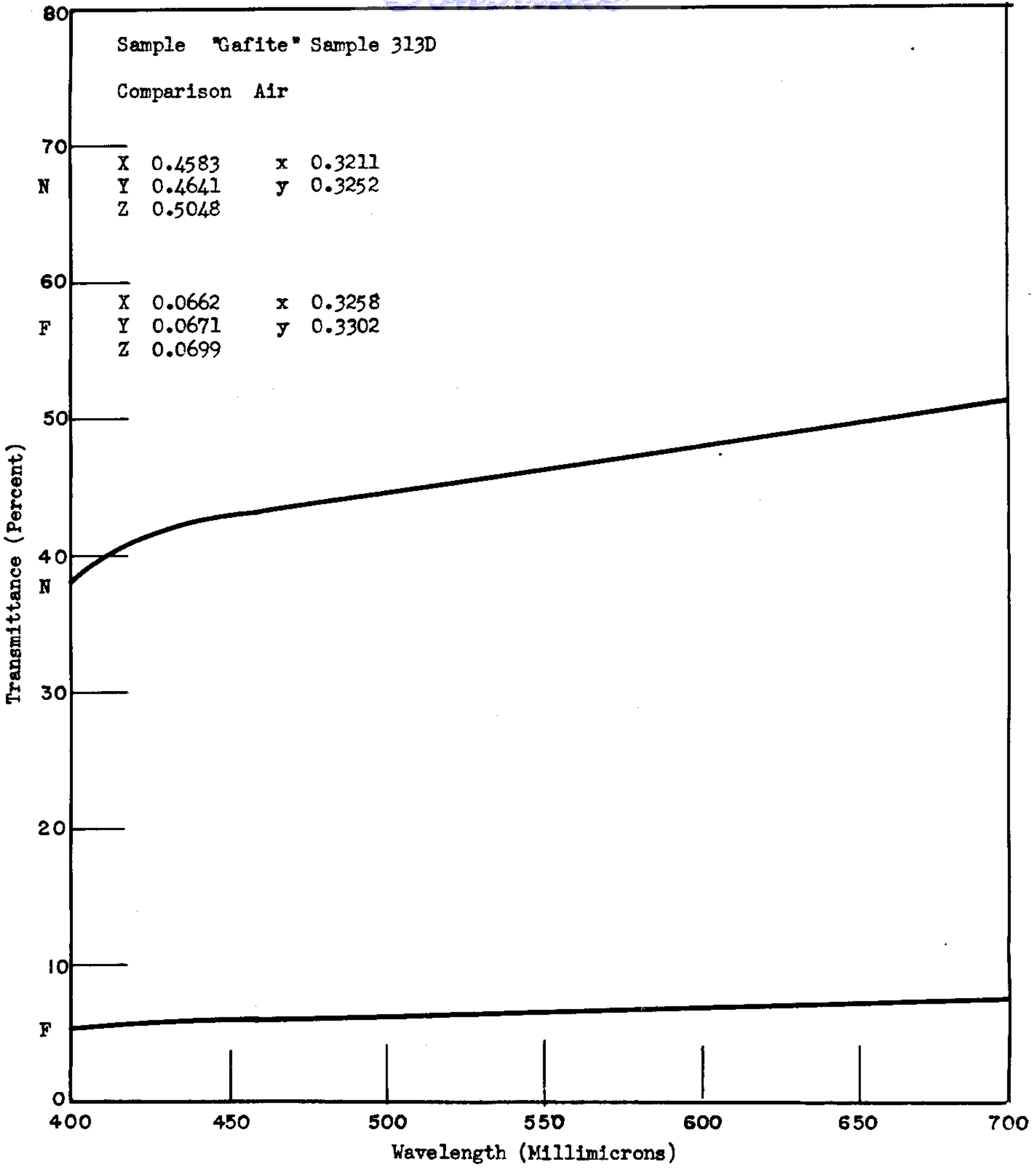


FIGURE 17. "GAFITE" SAMPLE 313D (GROUND WITH 220 GRIT EMERY POWDER ON ROTATING WET GRINDING WHEEL) PRIOR TO FADEOMETER TEST

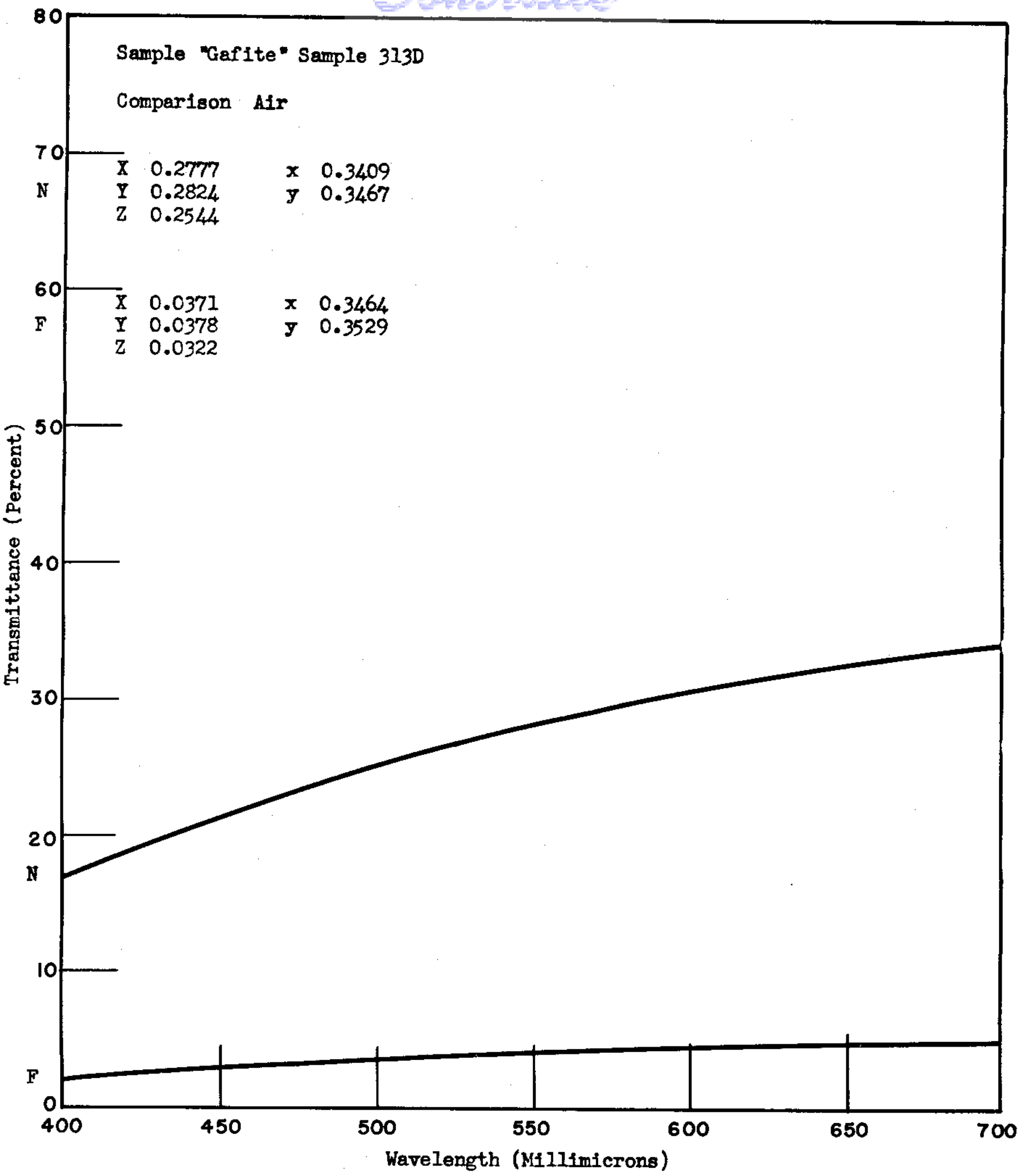


FIGURE 18. "GAFITE" SAMPLE 313D (GROUND WITH 220 GRIT EMERY POWDER ON ROTATING WET GRINDING WHEEL) AFTER 500 HOURS IN FADEOMETER

3.2 Effect of Heating Medium on Heat Stability

To determine the relation of heating medium on heat stability, several heat stability tests were run in different media on samples from the same sheet. The results shown in Table 17 indicate that the heat stabilities (time required to form bubbles in the sample visible to the naked eye) run about three times as great in an air oven with rapid circulation (250 ft/min) as they do in a liquid salt bath at the same temperature. Note that duplicate runs checked well except for one sample run at 160°C in the liquid salt bath.

TABLE 17. HEAT STABILITY OF SAMPLES FROM "GAFITE" SHEET 578

(Time in Minutes for Bubble Formation)

Temperature	Liquid Salt Bath [1]	Air Oven
160°C	79, 43, 74	230
177°C	17	75
191°C	7, 7	26, 24

The salt bath composition used for heat stability tests is described below; it is stable at 191°C and does not attack "Gafite". It does possess a serious drawback in that the gases released from "Gafite" on heating are insoluble in the salt bath and accumulate at the interface between the plastic and the liquid salt bath. These bubbles obscure the viewing of the formation of internal bubbles in "Gafite" and must be continually scraped from the surface in order to watch for internal bubbling of the plastic.

The salt bath was composed of equimolar amounts of potassium nitrate and sodium nitrite. For this bath, 10 parts by weight of potassium nitrate and seven parts be weight of sodium nitrite were mixed and melted. This bath possesses a melting point of 145°C. Although it possesses good stability at elevated temperatures, care must be taken to avoid the spattering of the hot nitrate onto organic material as this may result in an ignition. Nevertheless, a sample of "Gafite" sheet was heated as high as 250°C in this bath without ignition. The bath must be maintained in the molten state in glass vessels because the fused salt expands sufficiently on solidification to crack a glass vessel or a glass thermometer. When stored at room temperature, the nitrite and nitrate bath should be kept covered for it will absorb moisture which, however, can be readily driven out by stirring and heating the bath.

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[1] This liquid bath was made by melting a mixture of 10 parts by weight of potassium nitrate and seven parts by weight of sodium nitrite.

For screening tests, it is believed that heat stabilities run in a liquid bath are preferable for they will require considerably less time to run.

Table 18 lists the heat stabilities of "Gafite" sheets as determined by several methods. This table enables one to make a rough comparison of the difference between heating methods. It should be noted that correlation between methods is very rough and actually the particular section of a sheet sampled in some cases may make a difference in the value obtained for the stability. The sheets from 632 on were cast under this contract. The sheets 639 and 642 were cast from monomer which had had little air contact as evidenced by their very light yellow color. It should be noted that these sheets meet the heat stability requirements of this contract.

The heat stability by one-half hour periods was run by immersing a sample in a bath for this period and withdrawing it and examining it for bubbles. If no bubbles were present, the sheet was heated for additional periods until bubbles were noted in the sheet during the examination interval. In this table the stabilities are listed for the number of minutes of heating which had occurred when bubbles were noted. Expressed as one-half hour periods, this would be one 1/2 hour period more than our usual method of expressing heat stabilities which counts only the periods in which it has not bubbled. In determining the heat stabilities by the continuous method the sample is watched until bubbles are noted in the sample and this time is recorded.

TABLE 18. "GAFITE" HEAT STABILITY TESTS
(Results Expressed in Minutes)

Sheet No.	Bubbles Present After__Minutes in Salt Bath (30 Minute Periods) at 160°C	Bubbles Present After__Minutes in Salt Bath Continuous at 160°C	Bubbles Present After__Minutes in Salt Bath Continuous at 191°C	Bubbles Present After__Minutes in Air Oven (250 ft/ min Air Flow) Continuous at 191°C
493	90	105	10	50
497	90	105	8.5	37
508	90	113	7.5	39
529	60	76	8.5	50
533	120	190	8	45
535	90	141	11.5	55
542	30	67	6.5	33
546	90	127	7.5	32
549	120	84	8.5	42
601	90	74	2.5	30
604	90	114	4.0	42
632	60	70	5	30
633	120	80	5	23
634	90	80	4.5	23
635	120	102	5	36
636	120	118	6.5	36
637	30	60	4	24
638	90	52	4	26
639	120	149	10	52
640	60	112	6.4	34
641	60	92	6.4	23
642	60	102	9.4	45
643	30	66	4.5	23

The best correlation seems to be between the continuous method run at the same temperature (191°C) which gave bubbles as observed in a liquid salt bath in roughly one-fifth the time as observed in an air oven.

It was noted that the running of tests by one-half hour heating periods was dependent on the manner of cooling of the samples after heating. Rapid cooling induced bubble formation earlier than did slow cooling. For this reason a test method based on continuous heating in a liquid bath would be a preferred one. Heating in a liquid also would eliminate inconsistencies due to different heat transfer rates obtained in different air ovens.

3.3 Screening Tests on the Effect of Added Agents on the Light and Heat Stability of "Gafite"

In order to determine the effect of added agents on the light and heat stability of "Gafite" small scale castings were made of methyl α-chloroacrylate (about 120 milliliter) in Petroff flasks of pyrex glass to which had been attached a neck of glass tubing for sealing off purposes. The methyl α-chloroacrylate was added to the Petroff flask to which had been previously added the agent (in a small glass boat) under test. The flask containing the monomer and the added agent was cooled with dry ice and the gas space above the monomer was filled with prepurified nitrogen and closed with a cork. The glass tube neck was sealed off with the cork in place up to the point when the tube neck was placed in a burner to be sealed. Despite these precautions it was evident from some of the flask castings made in this way that air had entered the gas space above the monomer for duplicate samples cast varied in color after curing. This variation could only have been due to different amounts of air entering the neck of the flask during the sealing operation.

One possible solution to this problem would have been sealing off the necks of the flask under vacuum with the monomer cooled to prevent its evaporation. It was found that the walls of the Petroff flasks would not withstand evacuation. It was also found that the Petroff flasks frequently cracked during polymerization if they were filled up into the neck of the seal off tube, despite previous coating of the flask walls with a mold release agent to minimize adherence of the "Gafite" to glass. The difficulty appeared to lie in the geometry of the Petroff flask which would not allow for polymerization shrinkages.

Casting of the screening samples was changed to 25 mm OD test tubes, approximately 10 in. long, to which had been sealed a neck of 10 mm OD tubing about 3 in. long. These tubes were washed with Igepal and tap water followed by distilled water. They were then dried and a solution of a mold release agent was rinsed over the inside walls of the tubes. The tubes were then dried, washed with distilled water, and dried again.

To avoid vacuum sealing and to insure absence of air from the tube casting a method of closure using polyvinyl alcohol tubing was employed. Previous noncontractual research had established that polyvinyl alcohol was not attacked by methyl α-chloroacrylate as a solvent. The impermeability of polyvinyl alcohol to oxygen transmission is an established and valuable property of this polymer.

In the utilization of polyvinyl alcohol, a short length (about 3 in.) of polyvinyl alcohol tubing was attached to the glass tubing neck on the casting tube by slipping the polyvinyl alcohol tubing over the glass tubing for a distance of one-half inch. The added agent was weighed into a glass boat and the glass boat was slipped into the casting tube through the polyvinyl alcohol tubing and the glass tubing neck of the casting tube. The casting tube was filled with cold monomer up into the polyvinyl alcohol tubing at which point the polyvinyl alcohol tubing was pinched almost shut so that the monomer was present above the pinch-off point. After expansion of the monomer on warming to room temperature had ceased, the polyvinyl alcohol tubing was pinched completely shut. In this way the tube casting was filled with monomer which had contacted air only briefly at low temperature during the transfer of the monomer to the casting tube. After closing the polyvinyl alcohol tubing off the tubing was left in place until the polymerization and curing of the methyl α-chloroacrylate was complete. The glass boats in which the added agents had been introduced served as mixing bars when the casting tubes were inverted repeatedly to insure adequate and uniform mixing of the ingredients. By operating in the above manner, duplicate castings could be made which were consistently alike in appearance, particularly their color. With the ability to reproduce conditions almost entirely free of detrimental air contact, one could be sure that changes in color and stability of the cast samples were due to the effect of the added agents and not to accidental air contact.

Previous noncontractual research had established that contact of the monomer with air caused polymer cast from this monomer to yellow during its heat curing and the air contact caused the polymer to possess low heat stability. Although no definite comparisons were made, it appeared as if more pure monomer was more susceptible to discoloration by air contact than monomer of lower purity.

Although other contaminants besides air may cause cast polymer to be yellow and to possess low heat stability, we believe that we now are preparing monomer of such purity that air is the only contaminant causing yellow color and low heat stability in polymer cast from this monomer.

To determine whether air contact during pouring of a number of cast tubes had occurred, two tubes, one with and one without dibutyl tin diacetate catalyst, were poured at the start and at the end of any series of tubes cast. The appearances of these tubes serve as an indication of the amount of air contact of the monomer before and during pouring of the tubes of any series of experiments.

Some 62 screening castings were made in three series. The first series of 23 castings was made before use of the polyvinyl alcohol tubing closure method and the results are obscured by undefinable amounts of air contact. The second series of 25 castings utilized the polyvinyl alcohol tubing as did the third series of 14 castings. The results obtained supported the following general conclusions:

1. Dibutyl tin diacetate in "Gafite" sheet castings functions only as a polymerization catalyst. It does not improve the original as cast color of methyl α-chloroacrylate. It does not improve the light stability or heat stability of methyl α-chloroacrylate polymer.

2. "Gafite" which is yellow from contact of monomer with air appears to darken more on light exposure than "Gafite" which is colorless and has not contacted air.

The second series of castings employed monomer which from the light yellow appearance of the blank tubes had contacted air. It was noted that the following agents, ranked in order of decreasing effectiveness, caused the polymer castings in which they were incorporated to be markedly less yellow than the blanks:

Triethyl phosphite
"Thermolite 31" (Metal and Thermit Corp. Stabilizer)
Diethylene glycol
Diethyl hydrogen phosphite (formed white precipitate with dibutyl tin diacetate)
Benzyl alcohol
Tetrabutyl tin

The following compounds listed had little or no effect on decreasing the as cast color of the methyl α-chloroacrylate polymer in which they had been incorporated:

Diethyl ethyl phosphonate
"Saran" Light Stabilizer (Dow Chemical Co.)
Allyl chloride
Allyl glycidyl ether

On the other hand, di-tert-butyl peroxide increased the yellow as cast color of the methyl α-chloroacrylate polymer in which it had been incorporated.

Heat stabilities varied within this second series of castings, but oddly enough appeared to improve toward the end of the series. This result may have been due to some carbon dioxide dissolved in the monomer which may have separated from the monomer solution as the monomer warmed up closer to room temperature at the end of the series of castings and obscured the normal slight decrease in heat stability due to increased air exposure of the monomer at the end of a series of castings. However, the heat stabilities from first to last were poorer than those of the third series which was cast from another monomer batch. The monomer purity including absence of air contact appeared to determine the heat stabilities of these casting more than these particular added agents.

It was observed though that diethylene glycol, Thermolite 31, triethyl phosphite, and diethyl ethyl phosphonate did not possess lower heat stability than the blanks.

The third series of castings had blanks which indicated that the monomer employed had had very little contact with air. All of these castings possessed similar heat stabilities measured in a salt bath at 191°C (10 minutes) which indicated that they should have possessed heat stabilities of about 45 minutes at 191°C in an air oven. There was very little difference in color or heat stability among these samples. Samples containing "Uvinul 400" and "Uvinul 490" were very light yellow in color while the other samples were nearly colorless. These tubes contained the added agents:

Ethylene sulfite
Diallyl phosphite (forms white precipitate with dibutyl tin diacetate)
Diethyl ethyl phosphonate
"Uvinul 400"
"Uvinul 490"

The most interesting results obtained from these castings were on that one containing the "Uvinul 400". This casting of methyl α-chloroacrylate containing 0.05% "Uvinul 400" and 0.05% dibutyl tin diacetate showed no visual color changes after 240 hours in a modified weatherometer. (Estimated to be equivalent to six months exposure in south Florida.) Another interesting result was the very slight yellowing exhibited by the samples containing 0.2% ethylene sulfite, and 0.2% ethylene sulfite and 0.05% dibutyl tin diacetate. This agent was the only one aside from the ultraviolet light absorbers which has shown better light stability than the blank samples.

Although triethyl phosphite was outstanding in its ability to prevent the formation of yellow color in polymer from monomer whose blank samples indicated it had been exposed to air, this agent was disappointing in its resistance to yellowing on light exposure. However, the polymer containing triethyl phosphite was outstanding in its resistance to yellowing on extended heating. A combination of triethyl phosphite and "Uvinul 400" in methyl α-chloroacrylate provided light stability together with the other desirable properties conveyed by triethyl phosphite.

3.4 "Gafite" Sheet Containing Ultraviolet Absorbers

Among the more light stable "Gafite" sheets prepared in the past were sheets containing the ultraviolet absorbers, "Uvinul 490" and "Uvinul 400". These sheets which had contained ultraviolet absorbers and which had been prepared in the past were polymerized by light in the initial step. The dibutyl tin diacetate catalyst which was employed to effect uniform room temperature polymerization and to reduce strain pattern formation in "Gafite" sheets had not been employed together with "Uvinul 400" or "Uvinul 490". In order to reduce strain pattern development and to accomplish initial polymerization at room temperature, this catalyst is preferred for "Gafite" casting.

To determine the effect of the combination of "Uvinul 490" and dibutyl tin diacetate a casting was made of "Gafite" containing 0.1% dibutyl tin diacetate and 0.05% "Uvinul 490". It was hoped that the tin catalyst would still function as a catalyst in the presence of "Uvinul 490" causing polymerization at room temperature without strain pattern development and that the "Uvinul 490" would still serve to stabilize the "Gafite" casting against changes on light exposure. It was found that the tin catalyst apparently functioned in its proper role so far as the polymerization was concerned, but this combination of agents introduced a deep green-yellow color in "Gafite" which was not introduced by either agent alone. Variations in the concentrations of these agents were employed to determine whether a concentration could be found which would achieve the necessary strain free polymerization with a minimum of yellow color development together with satisfactory light stabilization.

It was found that 0.01% or 0.05% dibutyl tin diacetate in combination with 0.005% "Uvinul 490" functioned to catalyze the polymerization while only a slight yellow color was noted in the sample.

Saran Light Stabilizer incorporated in "Gafite" was found to give more color in the presence of dibutyl tin diacetate than in its absence. The tin catalyst's function as a catalyst did not appear to be impaired by the presence of the Saran Light Stabilizer. These effects were the same as those noted with "Uvinul 400" and "Uvinul 490".

In "Gafite" compositions containing dibutyl tin diacetate and one of the ultraviolet absorbers studied, it was found that compositions containing "Uvinul 400" were the least yellow as cast and possessed the greatest light stability.

A casting prepared containing 0.05% "Uvinul 400" and 0.05% dibutyl tin diacetate in "Gafite" was exposed for 240 hours in a modified weatherometer (estimated as the equivalent of six months in south Florida) and showed no visible change. This casting was the only casting prepared to date showing no visible change on light exposure. The original color of this casting was very nearly colorless and since it underwent no visible change on light exposure, the casting after light exposure was still very nearly colorless.

3.5 Chemical Drying Agents for Methyl α-Chloroacrylate

To obtain "Gafite" sheets possessing good heat stability it is desirable to remove all traces of moisture as well as low molecular weight alcohols such as methanol from methyl α-chloroacrylate. Phosphorus pentoxide has been used to accomplish this drying in the past. It was thought that barium oxide or calcium hydride might be equally as good or better drying agents because of their known efficiency in removal of water. It remained to be determined whether they could be employed with methyl α-chloroacrylate. Accordingly, the purity of vacuum distilled methyl α-chloroacrylate and portions of this same monomer dried separately by these three chemical drying agents for one hour at room temperature was determined by freezing point determinations. (The freezing points were determined on these monomers after passing them through a fairly coarse filter and traces of the drying agents may have been present during the course of the freezing point determination.) These data are summarized in Table 19.

TABLE 19. FREEZING POINTS OF METHYL α-CHLOROACRYLATE MONOMER AFTER DRYING

	Freezing Point
Vacuum distilled methyl α-chloroacrylate	-37.20°C
Vacuum distilled methyl α-chloroacrylate dried over P_2O_5	-36.78°C
Vacuum distilled methyl α-chloroacrylate dried over BaO	-36.78°C
Vacuum distilled methyl α-chloroacrylate dried over CaH_2	-36.58°C

The higher freezing point obtained with the monomer dried over calcium hydride indicates that it is the more pure monomer. The calcium hydride method of drying methyl α-chloroacrylate in addition to being more efficient is mechanically a more desirable method than the phosphorus pentoxide treatment process because the monomer can be treated with calcium hydride at room temperature with little or no increase in the viscosity of the monomer and the excess calcium hydride as well as its reaction products are readily filtered. With the process of treatment with phosphorus pentoxide, the monomer frequently increases in viscosity and the sticky particles of phosphorus pentoxide and its reaction products are difficult to filter.

The purification of methyl α-chloroacrylate by vacuum distillation followed by calcium hydride treatment has been run several additional times, and although the calcium hydride treatment has always raised the freezing point of the monomer so far it has not yielded a monomer of as high a freezing point as obtained in the initial experiment. Results on two different batches of monomer are reported in Table 20.

TABLE 20. FREEZING POINTS OF METHYL α-CHLOROACRYLATE DRIED WITH CALCIUM HYDRIDE

	Vacuum Distilled Monomer		Vacuum Distilled Monomer Dried with Calcium Hydride and Filtered
Freezing Points, Batch 3103-77	-37.10°C	⟶	-36.97
Freezing Points, Batch 3103-80	-36.97°C	⟶	-36.88

A check freezing point showed -38.94°C for the freezing point of mercury as compared to the literature values of -38.87 to -38.91°C (average -38.89°C) for pure mercury.

The monomer from Batch 3103-80 after drying with calcium hydride was stored at -20 to -37°C in the dark in a stoppered bottle for seven days and the freezing point was rerun. The sample apparently had picked up some moisture for its freezing point was -36.97°C. To a 50 milliliter sample of this monomer whose freezing point had just been determined was added one gram of calcium hydride and the freezing point was redetermined in the presence of calcium hydride and found to be -36.89°C. (The calcium hydride is insoluble in methyl α-chloroacrylate and does not affect the freezing point.) Apparently the original value of -36.88°C for this monomer dried with calcium hydride is quite reproducible. The freezing point differences reported are believed to be significant and beyond experimental error. The failure to obtain the high purity monomer (freezing point -36.58°C) in the second experiment cannot be attributed to moisture pickup during determination of the freezing point or the freezing point run in the presence of calcium hydride would not have duplicated the freezing point of the calcium hydride treated and filtered monomer from Batch 3103-80.

3.6 Drying by Distillation

Following investigation of chemical drying agents it was found that by efficient operation of fractional vacuum distillation one could prepare dry monomer of high purity (see Table 26 on composition of sheets supplied to the Air Force for representative freezing points). This monomer could be polymerized to yield polymer possessing the heat stability required for this contract. All sheets supplied to the Air Force under this contract were dried by fractional vacuum distillation without resort to chemical drying agents.

3.7 Study of Inhibitors for Methyl α-Chloroacrylate

In order to prepare methyl α-chloroacrylate polymer of high purity it was desirable to incorporate a fractional vacuum distillation step in its preparation. With a monomer which is as easily polymerized as methyl α-chloroacrylate, it was essential for the success of any vacuum distillation to have an effective inhibitor present during the distillation. It would be desirable if this inhibitor were nonvolatile so that it could be easily removed by the distillation. It also would be desirable if an inhibitor could be found to incorporate in the distillation packing in order to prevent polymerization in the upper part of the distillation column. To compare the relative value of several possible inhibitors, a test program on catalysts and inhibitors for polymerization of methyl α-chloroacrylate was undertaken. For this work methyl α-chloroacrylate (3099-138C) which had been vacuum distilled under prepurified nitrogen and which possessed a freezing point of -36.26°C was employed. This monomer was poured into 20 mm OD test tubes on which had been sealed 10 milliliter OD necks. These necks were attached to a 2 in. length of polyvinyl alcohol tubing which could be sealed off with a pinch clamp. The test tube had previously been coated with a solution of polyvinyl alcohol and Congo Red. This solution on drying deposited a red film over the glass through which observation of the contents of the tube could be made, but which would filter out light of the wave lengths responsible for polymerization. Each tube held approximately 33 milliliter of monomer when filled up to the neck at the point where the polyvinyl alcohol tubing was attached. The inhibitor or catalyst under test was weighed into each tube prior to the addition of the monomer and each tube was swept with prepurified nitrogen. After adding the monomer to the tube the polyvinyl alcohol tubing was pinched shut so that no air could contact the liquid monomer present below the pinch clamp. These tubes were sealed off and placed on a large mixing wheel which rotated at three revolutions per minute. The heating cycles employed for testing of catalysts and inhibitors for methyl α-chloroacrylate in chronological order of their use are listed below:

4 days at 20-25°C
5 hr at 50°C
10 days at 20-25°C
19 hr at 35-40°C
3 days at 45-50°C
5 days at 55-60°C
Remainder of time at 65-70°C

Each of the tubes on the mixing wheel was observed to determine the time required to gel or to form a nonflowing polymer in a tube rotating at three revolutions per minute. The results of this investigation are listed in Table 21.

TABLE 21. CATALYST AND INHIBITOR TESTING

Notebook No.	Catalyst or Inhibitor	Time Required to Gel or to Form Nonflowing Polymer in Tube Rotating at Three Revolutions Per Minute
3103-212	0.02 gm Dibutyl tin diacetate	5 hr
3103-205	0.04 gm Sodium hydroxide (pellets)	6 hr
3103-155	0.02 gm Dibutyl tin diacetate	7 hr
3103-206	0.04 gm Sodium methoxide	21 hr
3103-211	0.02 gm "Thermolite 99" (Metal and Thermit)	21 hr
3103-185	0.04 gm Sodium hydride	22 hr
3103-181	0.04 gm Sodium sulfide (fused flakes)	23 hr
3103-215	Diffuse light (tube not coated)	24 hr
3103-191	0.04 gm Antioxidant 2246 (American Cyanamid)	25 hr
3103-210	0.02 gm "Thermolite 31" (Metal and Thermit)	2 days
3103-213	11 milliliter Air in tube with 22 milliliter of monomer	2 days
3103-171	0.04 gm 9,10-Diformylaminoanthracene	3 days
3103-202	0.04 gm p-Nitroaniline	3 days and 4 hr
3103-203	0.04 gm Copper powder	3 days and 4 hr
3103-195	0.04 gm 2,4-Dinitrophenylhydrazine	3 days and 4 hr
3103-183	0.04 gm Calcium hydride	3 days and 6 hr
3103-201	0.04 gm Silver shavings	9 days
3103-162	0.04 gm 2-Methylanthraquinone	10 days
3103-163	0.04 gm 1,5-Diaminoanthraquinone	10 days
3103-164	0.04 gm 2,3-Dimethylanthraquinone	14 days
3103-193	0.04 gm Concentrated (36%) hydrochloric acid	14 days
3103-167	0.04 gm 1-Amino-4-hydroxyanthraquinone	18 days
3103-158	0.04 gm 1-Amino-2,3-dimethylanthraquinone	19 days
3103-196	0.04 gm 1,3-Dibutylthiourea	20 days
3103-197	0.04 gm Thiourea	20 days

TABLE 21. (Continued) CATALYST AND INHIBITOR TESTING

Notebook No.	Catalyst or Inhibitor	Time Required to Gel or to Form Nonflowing Polymer in Tube Rotating at Three Revolutions Per Minute
3103-200	0.04 gm p-tert-Butyl-m-cresol	23 days
3103-173	0.04 gm Methylene Blue	27 days
3103-186	0.04 gm N-Methylaniline	27 days
3103-187	0.04 gm Phenol	27 days
3103-198	0.04 gm 2-Aminoanthraquinone	27 days
3103-199	0.04 gm Universal Oil Products Inhibitor No. 1	27 days
3103-154	Pure monomer (no added agent)	28 days
3103-192	0.04 gm Sulfur	28 days
3103-172	0.04 gm Dihydro-1,4-dihydroxyanthraquinone	30 days
3103-194	0.04 gm 1,5-Dinitronaphthalene	30 days
3103-176	0.04 gm 2,3-Dichloronaphthoquinone	32 days
3103-188	0.04 gm Picryl chloride	32 days
3103-204	0.04 gm p-tert-Butylcatechol	32 days
3103-174	0.04 gm 2,5-di-tert-Butylquinone	33 days
3103-175	0.04 gm 2,5-di-tert-Butylhydroquinone	34 days
3103-182	0.04 gm 2,4-Dinitrochlorobenzene	34 days
3103-161	0.04 gm "Celliton" Fast Blue G Extra	36 days
3103-169	0.04 gm "Celliton" Fast Pink FF 3B Powder Conc.	43 days
3103-184	0.04 gm Universal Oil Products Inhibitor No. 3	43 days
3103-190	0.04 gm Picric acid	43 days
3103-159	0.04 gm 1,4-Diaminoanthraquinone	44 days
3103-178	0.04 gm Hydroquinone monomethyl ether	49 days
3103-180	0.04 gm Pyrogallic acid	49 days
3103-170	0.04 gm Violet BA	51 days
3103-156	0.04 gm "Celliton" Fast Blue B	56 days
3103-179	0.04 gm p-Phenylenediamine	56 days
3103-177	0.04 gm di-β-Naphthyl-p-phenylenediamine	56 days
3103-189	0.04 gm Hydroquinone	72 days
3103-160	0.04 gm Celliton Fast Violet 6B Pure	112 days
3103-208	0.04 gm Benzoquinone	112 days
3103-166	0.04 gm Dihydro-1,4,5,8-tetrahydroxyanthraquinone	147 + days
3103-207	0.04 gm Chloranil	147 + days
3103-209	0.04 gm Copper oxide	147 + days

All of these catalysts and inhibitors were tested in the absence of air and light, and this ranking for catalyst and inhibitors is only valid under this stipulation. Air is an effective catalyst as noted by Tube (3103-213) which contained only air and monomer and which polymerized in two days at 20-25°C in the absence of light. Light is an effective catalyst too as noted by Tube (3103-215) from which the polyvinyl alcohol and Congo Red coating had been omitted and which polymerized in 24 hours in diffuse light. Compounds requiring under 28 days to polymerize the monomer should be rated as catalysts for polymerization under these conditions, whereas those requiring over 28 days to polymerize the monomer should be rated as inhibitors for polymerization. Pure monomer polymerized in 28 days under these conditions. From the compounds studied, three outstanding inhibitors were found. These are actually still on test. Of these good inhibitors chloranil is too volatile to be as useful in distillation as dihydro-1,4,5,8-tetrahydroxyanthraquinone "Leukotetra". Both chloranil and "Leukotetra" are soluble in the monomer and could be used in the distillation pot as a soluble inhibitor. Copper oxide is both nonvolatile and insoluble in the monomer. It is ideally suited for inhibition of the monomer in packed distillation columns. Use of "Leukotetra" in distillation pots and of copper oxide distributed in the packing of distillation columns has made it possible to fractionally vacuum distill methyl α-chloroacrylate over periods of time as long as 10 hours with very little polymerization in the still pot or distillation column when operating at a pressure of 30 mm.

3.8 Purification of Methyl α-Chloroacrylate by Fractional Distillation

Distillation equipment permitting the cutting of several fractions and their maintenance under prepurified nitrogen have made possible detailed study of the changes occurring in purity during distillation. This work shows that there are present both low and high boiling impurities in methyl α-chloroacrylate as prepared by the dehydrohalogenation of methyl α,β-dichloropropionate with sodium adipate solution. Even after treatment of the monomer with calcium hydride, these impurities remain in the monomer and contaminate the distillate. The evidence for these conclusions is based upon the freezing points obtained for methyl α-chloroacrylate fractions after distillation through a six feet long 40 mm OD column packed with 0.24 x 0.24 in. protruded metal packing of 316 Stainless Steel (obtained from Scientific Development Co.). These distillations were performed at 30 mm pressure with no reflux, but only straight through distillation.

Freezing point determinations of methyl α-chloroacrylate samples taken during the course of vacuum distillations of crude methyl α-chloroacrylate indicate that the purity of the distilled monomer increases at first as distillation proceeds and then declines near the end of the distillation. Another distillation run with calcium hydride present in the still pot during distillation showed the same progression of freezing point changes, but with slightly higher purities throughout. These data are summarized in Table 22.

From these results it is evident that reflux and fractional distillation are required if even higher purity monomer is to be obtained. It would have been advantageous if this step could have been avoided for it increases the length of the distillation period and imposes severe requirements on the inhibitor to prevent polymerization of methyl α-chloroacrylate in the still pot during distillation.

With the discovery and use of copper oxide and "Leukotetra" as inhibitors for the polymerization of methyl α-chloroacrylate, it became possible to operate fractional vacuum distillation columns under more reflux with increased efficiency. In this way monomer was dried and purified for use in casting.

TABLE 22. FREEZING POINT CHANGES IN METHYL α-CHLOROACRYLATE DISTILLATE FRACTIONS

Distillation of Crude Methyl α-Chloroacrylate (5 Liters Approximately) Freezing Point of Original Monomer (3100-83) = -37.34°C

Fraction	Notebook No.	Volume (Milliliter)	Freezing Point, °C
Forerun	3099-100A	900	--
↓	3099-100B	50	-37.13
	3099-100C	1000	-37.00
	3099-100D	50	-37.07
	3099-100E	1000	-37.14
	3099-100F	1000	-37.38
	3099-100G	400	-37.65
Final Fraction	3099-100H	50	-37.90

Distillation of Crude Methyl α-Chloroacrylate (5 Liters Approximately) from Calcium Hydride, Freezing Point of Original Monomer (3100-82) = -37.83°C

Fraction	Notebook No.	Volume (Milliliter)	Freezing Point, °C
Forerun	3099-101B	250	--
↓	3099-101C	50	-37.44
	3099-101D	1000	-36.92
	3099-101E	50	-36.79
	3099-101F	1000	-36.99
	3099-101G	50	-36.95
	3099-101H	1000	-37.07
Final Fraction	3099-101I	50	-37.29

3.9 Purification of Methyl α-Chloroacrylate by Recrystallization

The freezing point of methyl α-chloroacrylate serves as an excellent criterion of overall purity of this monomer. In the early work on freezing points of methyl α-chloroacrylate a value of -36.63°C had been calculated to be the value for 100% pure methyl α-chloroacrylate as determined from freezing point depression of a solution of ethyl acetate in methyl α-chloroacrylate. During the period of this contract it became apparent that this calculated value for 100% pure methyl α-chloroacrylate did not represent the true value of the freezing point of 100% methyl α-chloroacrylate for freezing points higher than the calculated value were obtained on some samples of fractionally vacuum distilled monomer.

To determine more closely what the probable value of the freezing point of 100% methyl α-chloroacrylate was, a purification of methyl α-chloroacrylate by repeated crystallization of this monomer was undertaken to obtain as pure a sample as possible through repeated crystallizations. The purity changes during recrystallization were followed by freezing point determinations run on the fractions obtained.

These crystallizations were carried out by partially freezing vacuum distilled methyl α-chloroacrylate with stirring and sucking off the liquid layer. The solid layer was melted and after removing a sample for freezing point determination, another crystallization and separation of phases was performed. The monomer was stored in the dark at dry ice temperature under prepurified nitrogen between crystallizations.

Table 23 contains the results of the recrystallizations of methyl α-chloroacrylate.

It will be noted that the purity of the monomer increased (the freezing point was raised) on recrystallization in regular sequence through six recrystallizations. The seventh, eighth, and ninth recrystallizations failed to produce any significant changes in the freezing point on recrystallization. This behavior would be expected if the monomer had reached 100% purity. However, there were disturbing points in these results that do not fit in with such a conclusion. The monomer after the seventh, eighth, and ninth recrystallizations possessed a lower (poorer) freezing point than the monomer after six recrystallizations. In addition the check determination 3103-146C on 3103-146B in the eighth crystallization was poor and because the freezing point had raised indicated that the monomer had become more pure on storage.

To check the instrument, the freezing point of C.P. Mercury was run on 10 March 1954 and values for the freezing point of -38.82 and -38.80 were obtained. These values are acceptable checks on the averaged literature value of -38.89°C.

On 11 March 1954 two samples were withdrawn from the nine times crystallized monomer. The first freezing point of the Sample A (3103-146F) was determined and the sample was allowed to melt and the freezing point was redetermined. This process was repeated for one more time. The values obtained are listed in Table 24.

On Sample A, the freezing point was raised by repeated crystallization and thawing to a constant value of -35.93 which was raised further to -35.83 by the process of bubbling a rapid stream of oxygen through the monomer at 0°C for five minutes. The Sample A from the oxygen treatment after weekend storage was found to possess a lower freezing point which was not changed by the introduction of nitrogen over a period of five minutes into the monomer at 0°C.

On Sample B, the freezing point remained constant after overnight storage, but was raised when hydrogen was bubbled into the monomer at 0°C for five minutes. Weekend storage of the sample from the hydrogen treatment caused the freezing point to drop and bubbling nitrogen for five minutes through this monomer at 0°C did not change the freezing point materially.

Check determinations on mercury indicated that the freezing point results obtained were probably reliable. In addition this determination is subject to very little human error, for the freezing point is read from curves drawn by an electronic recorder which plots the cooling curve while the sample is being mechanically stirred in a cooling bath.

We believe that the above results indicated that the methyl α-chloroacrylate is readily contaminated with a gas, which is most likely carbon dioxide. The changes in freezing point occurring on storage and on recrystallization might be explained by a gas which is readily soluble in methyl α-chloroacrylate. This gas could dissolve in the monomer and lower its freezing point. The amount dissolved could be affected by recrystallization and the temperature which the monomer attained on

TABLE 23. FREEZING POINTS OF METHYL α-CHLOROACRYLATE ON RECRYSTALLIZATION

Date	Number of Re-crystallizations	Monomer Description	Freezing Point, °C	Reference
3/4/54	0	Original vacuum distilled monomer 3099-130C, G and H	-36.51	3103-143A
	1	Liquid Phase	-36.81	3103-143B
	1	Solid Phase	-36.43	3103-143C
3/5/54	2	Liquid Phase	-36.67	3103-144A
	2	Solid Phase	-36.36	3103-144B
	3	Liquid Phase	-36.93	3103-144C
	3	Solid Phase	-36.25	3103-144D
	4	Liquid Phase	-36.46	3103-144E
	4	Solid Phase	-36.28	3103-144F
3/8/54	4	Solid Phase - Repeat sample after storage	-36.23	3103-145A
	5	Liquid Phase	-36.31	3103-145B
	5	Solid Phase	-36.15	3103-145C
	6	Liquid Phase	-36.38	3103-145D
	6	Solid Phase	-36.09	3103-145E
3/9/54	6	Solid Phase - Repeat sample after storage	-36.10	3103-145F
	7	Liquid Phase	-36.38	3103-145G
	7	Solid Phase	-36.33	3103-145H
	8	Liquid Phase	-36.31	3103-146A
	8	Solid Phase	-36.35	3103-146B
3/10/54	8	Solid Phase - Repeat sample after storage	-36.19	3103-146C
	9	Liquid Phase	-36.38	3103-146D
	9	Solid Phase	-36.29	3103-146E

TABLE 24. FREEZING POINT OF NINE TIMES CRYSTALLIZED METHYL α-CHLOROACRYLATE

Date	Monomer Description	Freezing Point, °C	Reference
3/11/54	Nine times recrystallized - 1st freezing point - Sample A	-36.08	3103-146F
	- 2nd freezing point - Sample A	-35.92	3103-146F
	- 3rd freezing point - Sample A	-35.93	3103-146F
	Nine times recrystallized - 1st freezing point - Sample B	-36.27	3103-146G
3/12/54	Nine times recrystallized - 4th freezing point - Sample A	-35.93	3103-146F
	- 5th freezing point - Sample A + Oxygen	-35.83	3103-146F
	Nine times recrystallized - 2nd freezing point - Sample B	-36.26	3103-146G
	- 3rd freezing point - Sample B + Hydrogen	-36.16	3103-146G
3/15/54	Nine times recrystallized - 6th freezing point - Sample A + Oxygen	-36.28	3103-146F
	- 7th freezing point - Sample A + Oxygen and Nitrogen	-36.26	3103-146F
	- 4th freezing point - Sample B + Hydrogen	-36.38	3103-146G
	- 5th freezing point - Sample B + Hydrogen and Nitrogen	-36.42	3103-146G
3/17/54	It was noted that the thermopile had ridden up slightly in the tube surrounding it. To make sure that the freezing points were not affected by this occurrence, the freezing point for mercury was checked with the thermopile up in the tube and down in its normal position. The differences were less than 0.1°C.		
	Thermopile up in tube - C. P. Mercury	-38.74	
	Thermopile down in tube - C. P. Mercury	-38.79	
	Thermopile down in tube - C. P. Mercury (redistilled)	-38.84	

standing between recrystallizations. In addition introduction of an insoluble gas might be expected to sweep out some of the dissolved gas and raise the freezing point. The solubility of carbon dioxide in methyl α-chloroacrylate has been established from past experiments. Since it was used to attain the low temperatures involved in recrystallization and storage, it is possible that some of it contacted the monomer despite the precautions taken to keep it out. These precautions included keeping the monomer under an atmosphere of prepurified nitrogen.

The positive results from the above experiment are summarized below:

1) Pure methyl α-chloroacrylate possesses a freezing point at least as high as -35.83°C.

2) Oxygen, hydrogen and nitrogen are only very slightly, if at all soluble, in methyl α-chloroacrylate at 0°C.

3) It appears that carbon dioxide is very readily dissolved by methyl α-chloroacrylate.

3.10 Exclusion of Air from Methyl α-Chloroacrylate

Previous noncontractual research has shown the desirability and necessity of keeping air contact with methyl α-chloroacrylate to a minimum. Observations on castings made for screening tests have confirmed these earlier results. Indeed, the monomer of high purity now being produced seems even more sensitive to air exposure than monomer of lower purity made in the past.

To avoid air contact, which has such a serious effect on the color and heat stability of methyl α-chloroacrylate, this monomer is handled and stored under an atmosphere of prepurified nitrogen. Alterations on distillation equipment were made which permitted the cutting of several fractions without their contacting any gas except prepurified nitrogen.

Castings made from such air excluded monomer were markedly better in color than those obtained from monomer which had contacted air after their distillation.

3.11 Selection of Curing Conditions

Experimental work on leads uncovered in the study of the stability behavior of past "Gafite" sheets included study of the effect of incorporation of aliphatic hydroxyl containing compounds and ultraviolet absorbers in "Gafite" sheets. A number of experiments performed incorporating these agents in "Gafite" had to be repeated due to the discovery that curing at 160°C following initial polymerization of these sheets was not feasible because the remaining monomer content, estimated at 10-15%, polymerized so rapidly as to have caused heat evolution and boiling of the residual monomer in the sheet introducing bubbles in the sheets. This effect had not been noted in sheets cured at 120°C after initial polymerization. Stepwise heating must be employed to slowly polymerize the residual monomer until its content is reduced sufficiently to permit heating at 160°C without bubbling. The curing temperature of 160°C was employed in an effort to obtain as low a residual monomer content as possible.

Several sheets were cast and cured in this manner without premature bubbling. A typical heating schedule which was employed successfully after initial polymerization at room temperature was four hours at 60°C, followed by 16 hours at 100°C, and followed by three hours at 160°C. The most desirable curing temperature was worked out with infrared studies. The temperature of 160°C was chosen initially as a standard curing temperature because indirect evidence obtained in the past indicated it was far more effective in completing polymerization than a temperature of 120°C. The infrared studies confirmed the selection of this temperature for curing.

The curing conditions selected are those employed in an air oven with 250 ft/min air circulation. The curing conditions are dependent on the rate of heat transfer as well as the temperature of the oven employed for heating.

3.12 Residual Monomer in Methyl α-Chloroacrylate Polymer

It is our belief that the poor light stability of methyl α-chloroacrylate is due to the monomer remaining in the polymer sheet following polymerization and not to any instability of the polymer. For this reason we were desirous of selecting polymerization conditions such that the residual monomer content was at a minimum.

The most likely method of obtaining values for residual monomer content appeared to be through infrared spectrography. Past work at these laboratories indicated the feasibility of such analyses for residual monomer content. This work has been facilitated by our receipt and installation of the latest model Perkin-Elmer infrared spectrometer. With the aid of this new instrument we sought to determine the exact effect of curing conditions on monomer content of "Gafite".

The writer wishes to acknowledge his indebtedness to Dr. Hans J. Stolten of our Analytical Research Section for the following results on infrared determination of monomer content of "Gafite".

The following methods of sample preparation were examined in order to devise a useful method for the determination of the monomer content of "Gafite" by infrared absorption.

1) "Gafite" chips of various thicknesses.
2) "Gafite" chips of various thicknesses wet with Nujol.
3) Nujol mulls of "Gafite" filings of various thicknesses.
4) Nujol mulls of "Gafite" emulsion polymer.
5) Ethylene dichloride solutions of "Gafite".
6) Approximately one mil thick films of "Gafite" evaporated from ethylene dichloride solutions.
7) Films approximately three mils thick cast by the usual polymerization technique.

The first five methods tried did not yield infrared spectra suitable for quantitative determinations. The sixth method yielded excellent spectra, but the method of sample preparations is not suitable for quantitative measurement due to possible loss of monomer by evaporation. However, since these films yield suitable spectra, the seventh method is currently used for quantitative estimations of monomer content. The films are used as received after their removal from the miniature casting cells.

Since the "Gafite" films vary in thickness, and micrometer measurements are not sufficiently accurate, the intensity of the characteristic infrared absorption due to the C-H stretching vibration is employed as a measure of the film thickness. The absorption selected occurs at 3.33μ and is independent of the monomer content of the "Gafite". The intensity of the 6.23μ absorption, due to the "Gafite" monomer, is used as a measure of the monomer content. The intensity of this absorption is corrected for film thickness variations by dividing the absorbance at 6.23μ by the absorbance at 3.33μ. For different thicknesses of any film this ratio remains constant.

In the résumé of the experimental work that follows in Table 25, the numbers that appear in the monomer content column are the ratios of the absorbances described above. For a sample of pure monomer (MACA 3099-95C), this ratio has a value of approximately 3.0. An estimation of the monomer content of the films can be made by the proportion 100%/3.0 = x %/ratio value in the monomer column. Since the value 3.0 was obtained on liquid monomer in a sealed liquid cell, and the ratios listed were obtained from films between two NaCl plates, monomer values calculated in this manner are only approximate. Nevertheless, in examining the following data the changes in the value of this ratio are real and reflect real changes in the monomer content of the "Gafite" due to the varying polymerization and curing procedures employed.

The curing conditions empirically selected prior to this study were

4 hours at 60°C
16 hours at 100°C
3 hours at 160°C

From the infrared results, the most advantageous curing treatment for low residual monomer appears to be 2-8 hours at 160°C or more likely 2-4 hours at 160°C. These results show that slight decomposition definitely starts somewhere between four and eight hours at 160°C. Since the optimum could lie somewhere between two and eight hours at 160°C, the initial curing treatment of three hours at 160°C was continued.

These results indicate the desirability for further detailed study of the 140°C to 160°C curing temperature range. The temperature of curing has a profound effect on residual monomer content. For example, one hour of curing at 140°C is more efficient than 23.5 hours of curing at 120°C. These results would be of value in aiding in determining an economical curing cycle in point of time.

3.13 Composition of Cast Sheets Supplied to the Air Force

From the research results obtained in this report, three types of "Gafite" sheets were selected as being most promising for further study. "Gafite" sheets of these three types were cast under the contract. The first type contained only dibutyl tin diacetate catalyst and "Uvinul 400", ultraviolet absorber. The second type contained triethyl phosphite in addition to the tin catalyst and the ultraviolet absorber. The third type contained ethylene sulfite in addition to the tin catalyst and ultraviolet absorber. All three types of "Gafite" sheet appeared to meet the contract specifications. In addition to the sheets required under the contract there were additional sheets of the three types supplied gratis to the

TABLE 25. MONOMER CONTENT OF "GAFITE" FILMS CURED AT DIFFERENT TEMPERATURES

Sample No.	Time	Monomer Content		
	Ultraviolet Exposed (Room Temperature)	A6.23μ/A3.33μ	Percent Monomer	
3100-105	2 hr	1.17	39	a/
-106	4 hr	1.08	36	
-107	8 hr	1.13	38	
-108	16 hr	1.00	33	
-109	32 hr	1.02	34	
	Catalyst Polymerized (Room Temperature)			
3100-110	1 day	0.94	31	
-111	4 days	1.24	41	
-112	5 days	1.11	37	
-113	6 days	1.34	45	
-114	7 days	0.96	32	
	60°C			
3100-115	1 hr	1.13	38	
-116	2 hr	1.16	39	
-117	4 hr	1.14	38	
-118	8 hr	1.07	36	
-119	14 hr	0.93	31	
-120	23.5 hr	0.98	33	
	100°C			
3100-121	1 hr	0.82	27	
-122	2 hr	0.56	19	
-123	4 hr	0.50	17	
-124	8 hr	0.49	16	
-125	14 hr	0.37	12	
-126	23.5 hr	0.27	9.0	
	120°C			
3100-127	1 hr	0.26	8.7	
-128	2 hr	0.21	7.0	
-129	4 hr	0.18	6.0	
-131	8 hr	0.13	4.3	
-132	14 hr	0.17	5.7	
-133	23.5 hr	0.13	4.3	

TABLE 25. (Continued) MONOMER CONTENT OF "GAFITE" FILMS CURED AT DIFFERENT TEMPERATURES

Sample No.	Time	Monomer Content	
		A6.23μ/A3.33μ	Percent Monomer
	140°C		
3100-134	1 hr	0.10	3.3
-135	2 hr	0.056	1.9
-136	4 hr	0.056	1.9
-137	8 hr	0.035	1.2
-138	14 hr	0.032	1.1
-139	23.5 hr	0.10	3.3 b/
	160°C		
3100-140	1 hr	0.034	1.1
-141	2 hr	0.029	0.97
-142	4 hr	0.016	0.53
-143	8 hr	0.049 c/	---
-144	14 hr	0.10	---
-145	23.75 hr	0.076	---
	180°C		
3100-146	1 hr	0.067	---
-147	2 hr	0.11	---
-148	4 hr	0.076	---
-149	8 hr	0.12	---
-150	14 hr	0.08	---
-151	23.5 hr	0.19	---

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a/ These values may vary due to evaporation losses of monomer in the time required to transfer the film from the casting cell to the instrument. This time was not constant throughout these samples.

b/ This film yielded a poor curve for reading monomer content due to its thickness being too great.

c/ Beginning with this sample, the monomer absorption at 6.23μ, which is now very small, becomes somewhat obscured. This is probably due to the appearance of decomposition products of the "Gafite". Consequently the increase in value of the absorbency ratio is not indicative of an increase in monomer absorption but rather of the appearance of a new absorption close to 6.23μ which may be due to the above mentioned decomposition.

TABLE 26. "GAFITE" SHEETS SUPPLIED UNDER USAF CONTRACT NO. 33(600)-23883

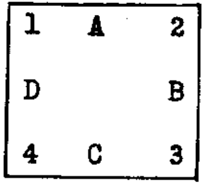
Sheet Number	Methyl α-Chloroacrylate Freezing Point (°C.)	Nominal Size (Inches)	Concentration of (Wt. %) Dibutyl Tin Diacetate Catalyst	Concentration of (Wt. %) Uvinul 400 U.V. Absorber	Concentration of (Wt. %) Added Agents	Thickness (in Inches) at  (Filling Corner at 1)
662	-36.23	24 x 24 x 1/4	0.05	0.05	None	1- 0.244 2- 0.249 3- 0.249 4- 0.241 A- 0.250 B- 0.252 C- 0.254 D- 0.248
663	-36.72	24 x 24 x 1/4	0.05	0.05	None	1- 0.242 2- 0.243 3- 0.249 4- 0.248 A- 0.247 B- 0.250 C- 0.249 D- 0.248
682	-36.35	24 x 24 x 1/4	0.05	0.05	0.05 Triethyl Phosphite	1- 0.251 2- 0.258 3- 0.251 4- 0.256 A- 0.249 B- 0.258 C- 0.256 D- 0.253
692	-36.43	24 x 24 x 1/4	0.05	0.05	0.05 Ethylene Sulfite	1- 0.240 2- 0.240 3- 0.238 4- 0.243 A- 0.230 B- 0.228 C- 0.232 D- 0.228
656	-36.36	12 x 12 x 1/4	0.05	0.05	None	1- 2- 3- 4- A- B- C- D-
659	-36.39	24 x 24 x 1/4	0.054	0.054	None	1- 2- 3- 4- A- B- C- D-
660	-36.39	24 x 24 x 1/4	0.05	0.05	None	1- 2- 3- 4- A- B- C- D-

TABLE 26. (Continued) "GAFITE" SHEETS SUPPLIED UNDER USAF CONTRACT NO. 33(600)-23883

Sheet Number	Methyl α-Chloroacrylate Freezing Point (°C.)	Nominal Size (Inches)	Concentration of (Wt. %)			Thickness (in Inches) at 1 A 2 D B 4 C 3 (Filling Corner at 1)	
			Dibutyl Tin Diacetate Catalyst	Uvinul 400 U.V. Absorber	Added Agents		
661 6R6D	-36.54	12 x 12 x 1/4	0.062	0.062	None	1- 3- A- C-	2- 4- B- D-
668	-36.45	24 x 24 x 1/4	0.05	0.05	None	1- 3- A- C-	2- 4- B- D-
669	-36.39	24 x 24 x 1/4	0.05	0.05	None	1- 3- A- C-	2- 4- B- D-
670	-36.39	24 x 24 x 1/4	0.05	0.05	None	1- 3- A- C-	2- 4- B- D-
671 3R4D	-36.36	12 x 12 x 1/4	0.05	0.05	None	1- 3- A- C-	2- 4- B- D-
671 3R16D	-36.36	12 x 24 x 1/4	0.05	0.05	None	1- 3- A- C-	2- 4- B- D-
671 15R4D	-36.36	12 x 12 x 1/4	0.05	0.05	None	1- 3- A- C-	2- 4- B- D-

TABLE 26. (Continued) "GAFITE" SHEETS SUPPLIED UNDER USAF CONTRACT NO. 33(600)-23883

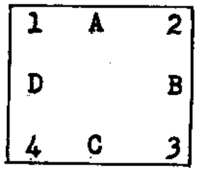
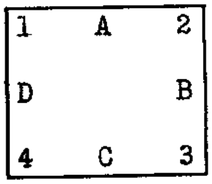
Sheet Number	Methyl α-Chloroacrylate Freezing Point (°C)	Nominal Size (Inches)	Concentration of (Wt. %) Dibutyl Tin Diacetate Catalyst	Concentration of (Wt. %) Uvinul 400 U.V. Absorber	Concentration of (Wt. %) Added Agents	Thickness (in Inches) at  (Filling Corner at 1)	
673	-36.24	24 x 24 x 1/4	0.05	0.05	None	1- 3- A- C-	2- 4- B- D-
674	-36.32	24 x 24 x 1/4	0.05	0.05	None	1- 3- A- C-	2- 4- B- D-
675	-37.70	24 x 24 x 1/4	0.05	0.05	None	1- 3- A- C-	2- 4- B- D-
677 3R2D	-36.35	12 x 24 x 1/4	0.05	0.05	0.05 Triethyl Phosphite	1- 3- A- C-	2- 4- B- D-
677 14.5R 14.5D	-36.35	12 x 12 x 1/4	0.05	0.05	0.05 Triethyl Phosphite	1- 3- A- C-	2- 4- B- D-
680 1R 12.5D	-36.28	12 x 12 x 1/4	0.05	0.05	0.05 Triethyl Phosphite	1- 3- A- C-	2- 4- B- D-
680 13.5R 2D	-36.28	12 x 12 x 1/4	0.05	0.05	0.05 Triethyl Phosphite	1- 3- A- C-	2- 4- B- D-

TABLE 26. (Continued) "GAFITE" SHEETS SUPPLIED UNDER USAF CONTRACT NO. 33(600)-23883

Sheet Number	Methyl α-Chloroacrylate Freezing Point (°C.)	Nominal Size (Inches)	Concentration of (Wt. %)			Thickness (in Inches) at  (Filling Corner at 1)	
			Dibutyl Tin Diacetate Catalyst	Uvinul 400 U.V. Absorber	Added Agents		
684 10R 3D	-36.45	12 x 12 x 1/4	0.05	0.05	0.05 Triethyl Phosphite	1- 3- A- C-	2- 4- B- D-
686	-36.88	24 x 24 x 1/4	0.05	0.05	0.05 Ethylene Sulfite	1- 3- A- C-	2- 4- B- D-
690	-36.94	24 x 24 x 1/4	0.05	0.05	0.05 Ethylene Sulfite	1- 3- A- C-	2- 4- B- D-
694	-36.43	24 x 24 x 1/4	0.048	0.048	0.048 Ethylene Sulfite	1- 3- A- C-	2- 4- B- D-
695	-36.36	12 x 12 x 1/4	0.05	0.05	0.05 Triethyl Phosphite	1- 3- A- C-	2- 4- B- D-
						1- 3- A- C-	2- 4- B- D-
						1- 3- A- C-	2- 4- B- D-

Air Force for additional tests. The exact compositions and the freezing points for the monomer employed to cast all these sheets are listed in Table 26 on "Gafite" sheets supplied to the Air Force.

3.14 Heat Stability of "Gafite" Sheets Supplied to the Air Force

All of the "Gafite" sheets listed in Table 26 were tested for heat stability and found to form no bubbles on heating in an air oven at 375°F (191°C) for 45 min with an air circulation of 250 ft/minute.

3.15 Light Stability and Color of "Gafite" Sheets for the Air Force

The specified tests on luminous transmittance and haze call for testing plastic samples on a pivotal sphere (Hunter) Hazemeter. Since this instrument was not available at GAF the transmittance and haze were determined on a General Electric recording spectrophotometer with a tristimulus integrator. In Table 27 are listed the values for luminous transmittance and haze as determined on a General Electric recording spectrophotometer. These same samples were run on the Hunter Hazemeter at Wright Field and the results on haze and luminous transmittance are recorded in Table 28. From comparison of the two tables it will be seen that the methods are in substantial agreement. It was known from qualitative examination of GAF casting tube samples containing "Uvinul 400" that their light stability after 240 hours exposure in a modified weatherometer estimated to be equivalent to six months in south Florida was sufficient so that it was difficult to visually detect any change in the sample. To confirm these results on cast sheet several of the cast sheets prepared under this contract were checked for luminous transmittance and haze after 240 hours exposure in a modified weatherometer. These results are listed in Table 29.

It will be noted from this table that all samples possessed a transmittance of 89% or greater and that all samples possessed less than 4% haze except for one sample (667) in which the haze was 6.4%.

The original luminous transmittance and haze values for all the sheets submitted to the Air Force are summarized in Table 30. The transmittance curves (Figures 19-40) obtained on a General Electric recording spectrophotometer for these samples follow Table 30.

The transmittance and haze were determined on the General Electric recording spectrophotometer as follows:

$$\%\ \text{Transmission} = Y_{(N)} \times 100$$

$$\%\ \text{Haze} = \frac{Y_{(N)} - Y_{(F)}}{Y_{(N)}} \times 100$$

$Y_{(N)}$ = Y value with sample in normal position (close) to photocell.

$Y_{(F)}$ = Y value with sample in far position (away) from photocell.

TABLE 27. LUMINOUS TRANSMITTANCE AND HAZE FROM TRANSMITTANCE CURVES OBTAINED ON GENERAL ELECTRIC RECORDING SPECTROPHOTOMETER FOR "GAFITE" SHEETS 1/4 INCH THICK

Sheet No.			Haze (%)	Luminous Transmittance (%)
655	1R	15D	0.8	91.1
656			0.5	91.2
657	1R	4.5D	1.5	90.7
658	3R	3D	1.6	91.1
659	26R	3D	1.3	91.2
660	20R	3D	1.1	91.3
661	7R	22D	1.8	91.1
662	17R	1D	0.8	91.6
663	6.5R	2.5D	0.03	91.4
664	2R	15D	1.3	91.0
666	4R	22D	1.0	92.2
668	5R	27D	0.3	91.7
669	17.5R	2.5D	0.8	92.0
670	5.5R	2.5D	0.4	94.0
671	14R	1.5D	0.8	91.6
673	2R	13D	0.9	92.0
674	24R	26.5D	0.9	92.0
675	26.5R	14.5D	0.6	91.6
676	2.5R	17D	0.3	93.7
677	17.5R	1D	0.5	93.4

TABLE 28. LUMINOUS TRANSMITTANCE AND HAZE BY PIVOTABLE SPHERE (HUNTER) HAZEMETER OF "GAFITE" SHEETS 1/4 INCH THICK

Sheet No.			Haze (%)	Luminous Transmittance (%)
655	1R	15D	1.7	91.3
656			0.6	91.1
657	1R	4.5D	1.4	91.0
658	3R	3D	2.0	91.4
659	26R	3D	1.3	91.4
660	20R	3D	0.9	91.6
661	7R	22D	2.0	91.4
662	17R	1D	0.5	91.7
663	6.5R	2.5D	0.6	91.7
664	2R	15D	1.0	91.6
666	4R	22D	0.7	91.9
668	5R	27D	0.6	91.5
669	17.5R	2.5D	0.6	91.9
670	5.5R	2.5D	0.9	92.0
671	14R	1.5D	0.5	91.5
673	2R	13D	0.7	91.7
674	24R	26.5D	0.8	91.7
675	26.5R	14.5D	0.5	91.8
676	2.5R	17D	1.0	91.8
677	17.5R	1D	0.9	92.0

TABLE 29. LUMINOUS TRANSMITTANCE AND HAZE FROM TRANSMITTANCE CURVES OBTAINED ON GENERAL ELECTRIC RECORDING SPECTROPHOTOMETER FOR "GAFITE" SHEETS 1/4 INCH THICK AFTER 240 HOURS EXPOSURE IN A MODIFIED WEATHEROMETER

Sheet No.			Haze (%)	Luminous Transmittance (%)
656	1R	23D	2.8	90.0
658	1R	5D	2.2	90.6
659	26R	15D	1.5	88.8
662	5R	1D	3.3	91.1
663	27R	8D	2.9	90.7
666	14R	4D	2.9	90.2
667	21R	5D	6.4	90.2
668	2.5R	21.5D	2.7	89.6
669	5.5R	2.5D	3.8	90.7
670	26R	5.5D	3.0	91.1
671	22R	1.5D	1.9	90.5
672	27.5R	22D	2.0	90.5
673	12R	27D	1.7	90.6
674	12R	26.5D	0.8	90.8
675	26.5R	24.5D	0.8	90.4

TABLE 30. LUMINOUS TRANSMITTANCE AND HAZE FROM TRANSMITTANCE CURVES OBTAINED ON GENERAL ELECTRIC RECORDING SPECTROPHOTOMETER FOR 1/4 INCH THICK "GAFITE" SHEETS SUBMITTED TO THE AIR FORCE

Sheet No.	Haze (%)	Luminous Transmittance (%)
662	0.6	91.7
663	0.7	91.6
682	0.7	92.8
692	0.9	91.7
656	0.5	91.2
659	0.4	91.6
660	1.4	91.8
661	0.3	90.5
668	0.6	91.6
669	0.9	91.8
670	0.5	91.7
671	1.1	91.8
673	0.9	91.8
674	0.9	91.9
675	0.4	91.9
677	0.6	93.1
680	0.9	92.0
684	0.6	93.6
686	0.2	91.8
690	0.7	91.8
694	0.6	91.8
695	0.7	92.5

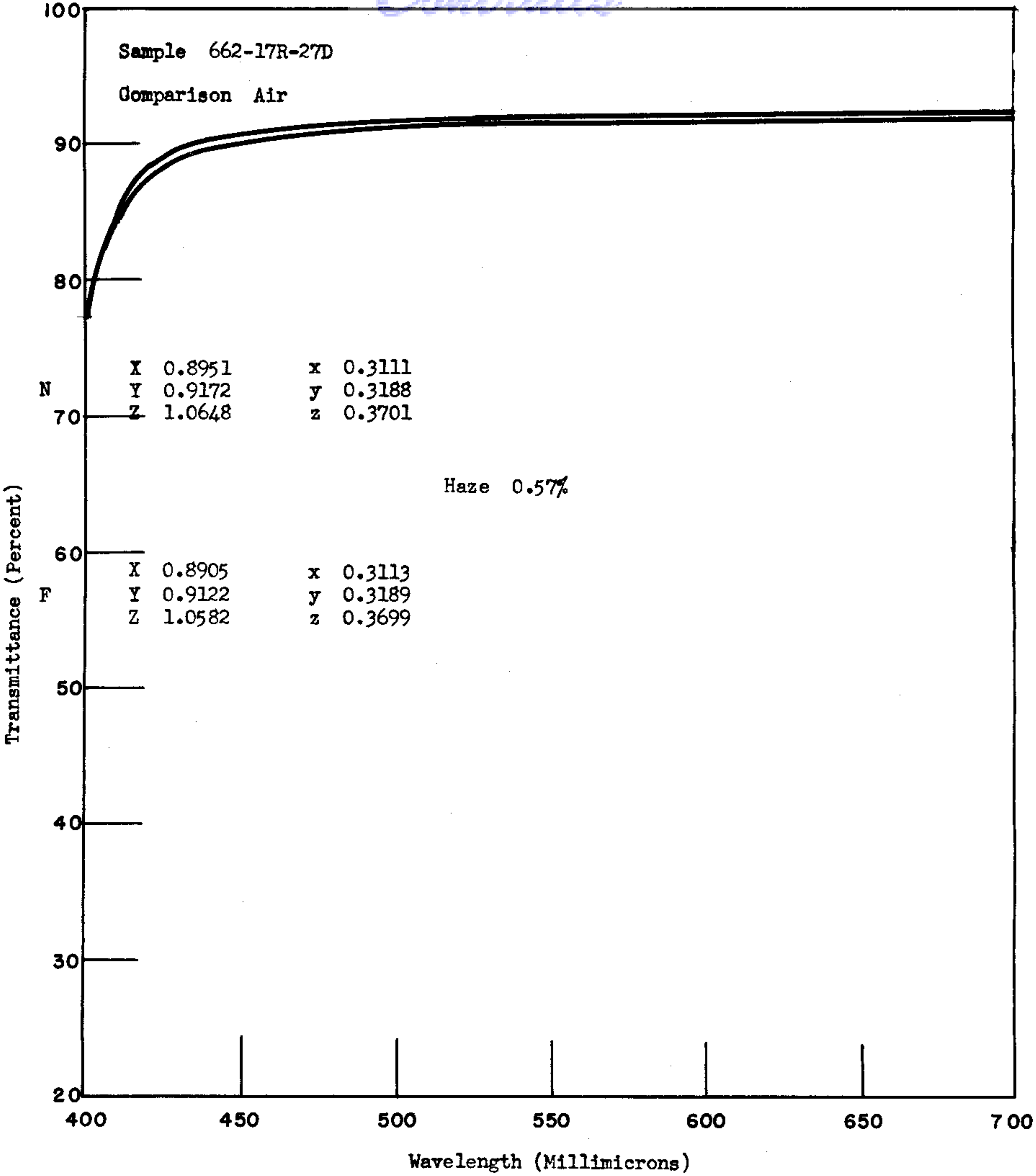


FIGURE 19. TRANSMISSION CURVES FOR "GAFITE" SAMPLE 662-17R-27D

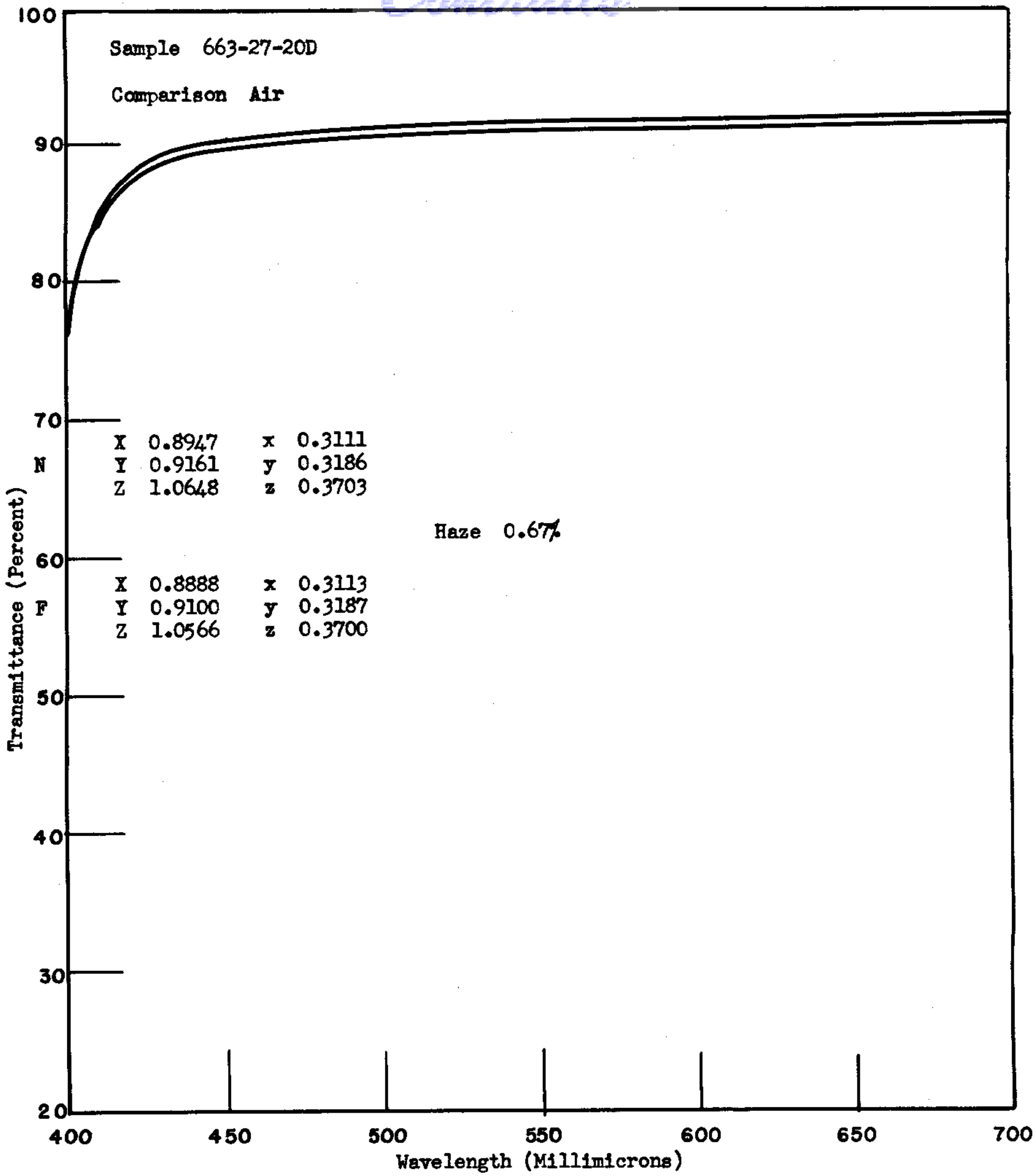


FIGURE 20. TRANSMISSION CURVES FOR "GAFITE" SAMPLE 663-27-20D

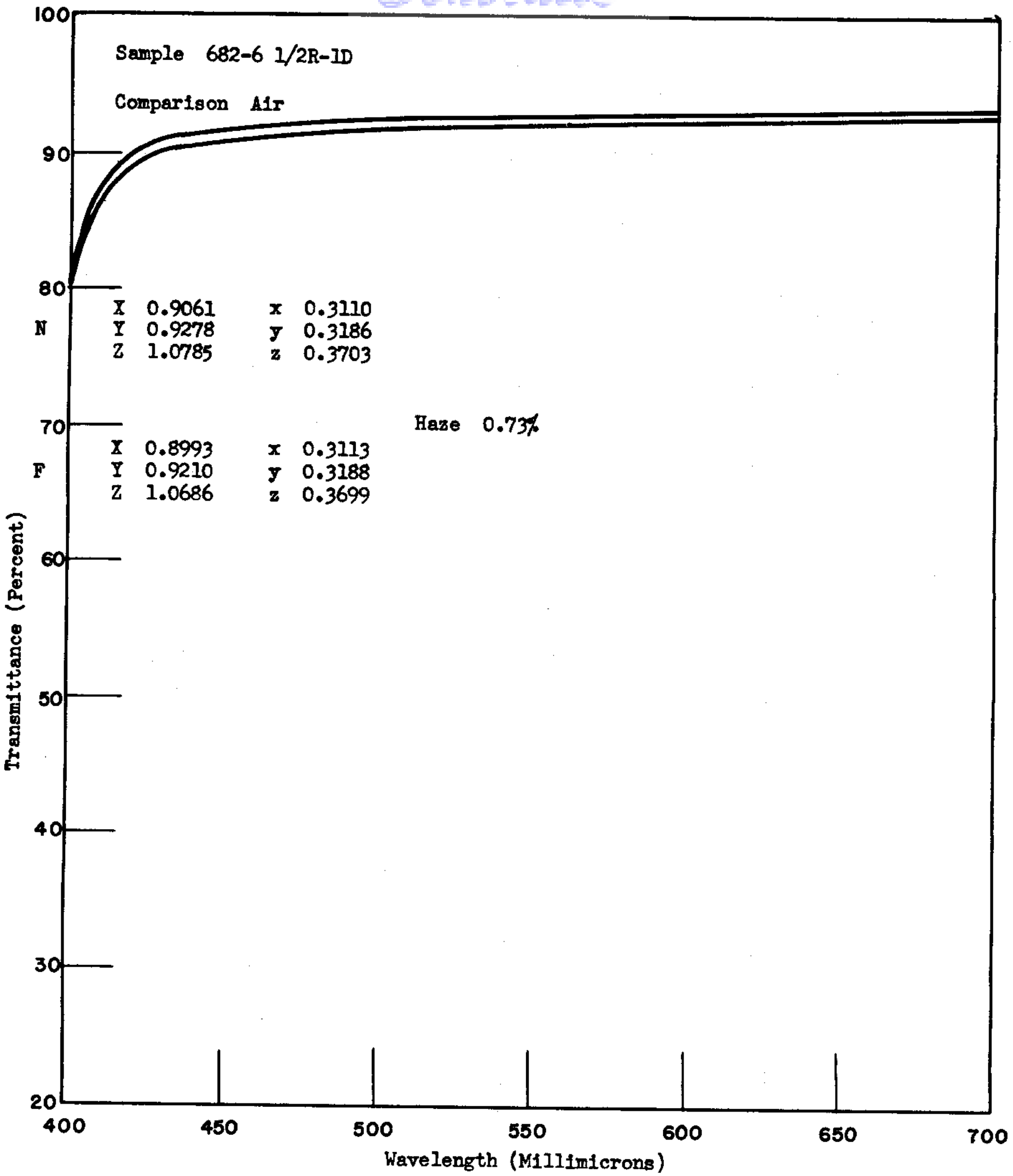


FIGURE 21. TRANSMISSION CURVES FOR "GAFITE" SAMPLE 682-6 1/2R-1D

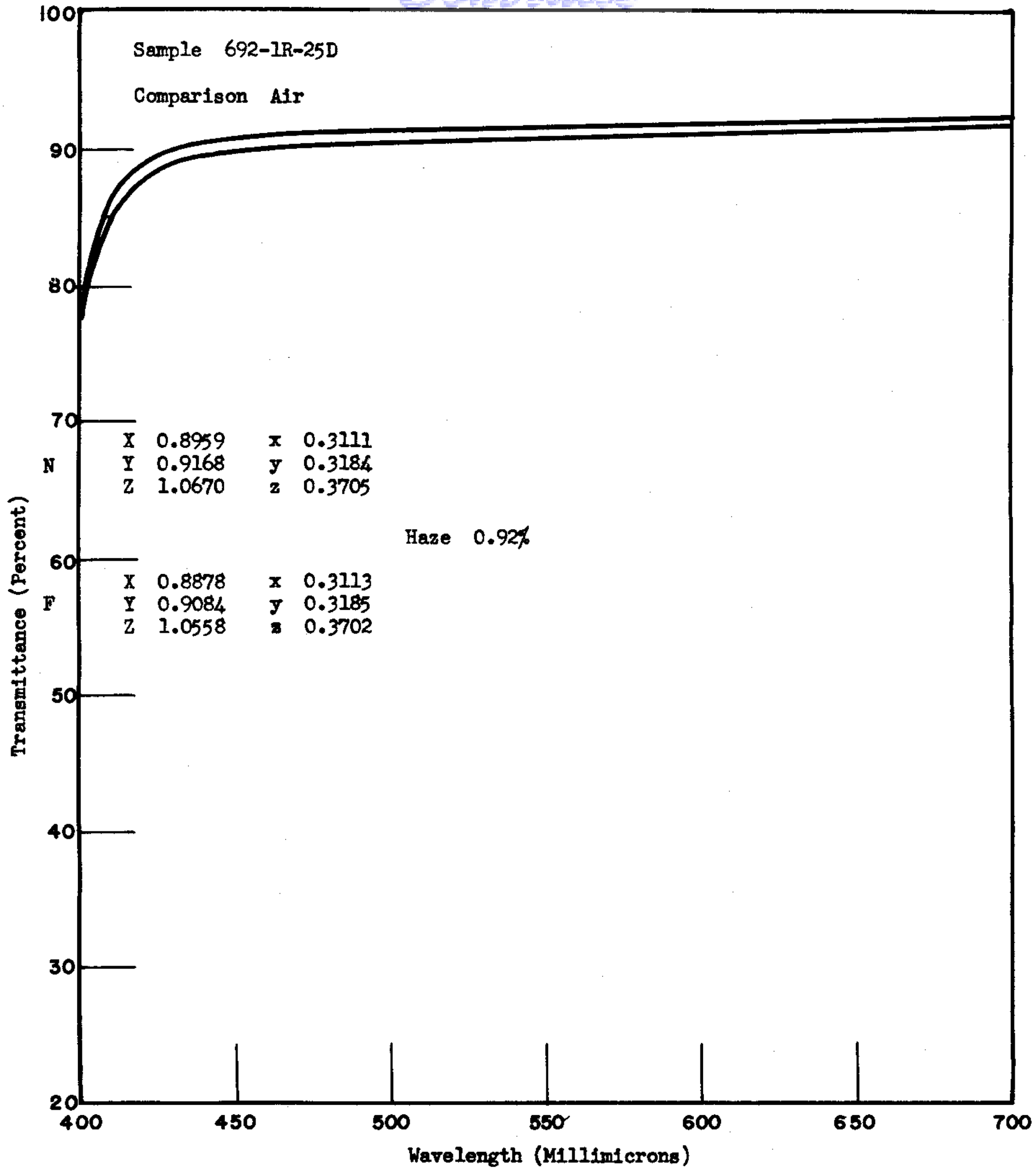


FIGURE 22. TRANSMISSION CURVES FOR "GAFITE" SAMPLE 692-1R-25D

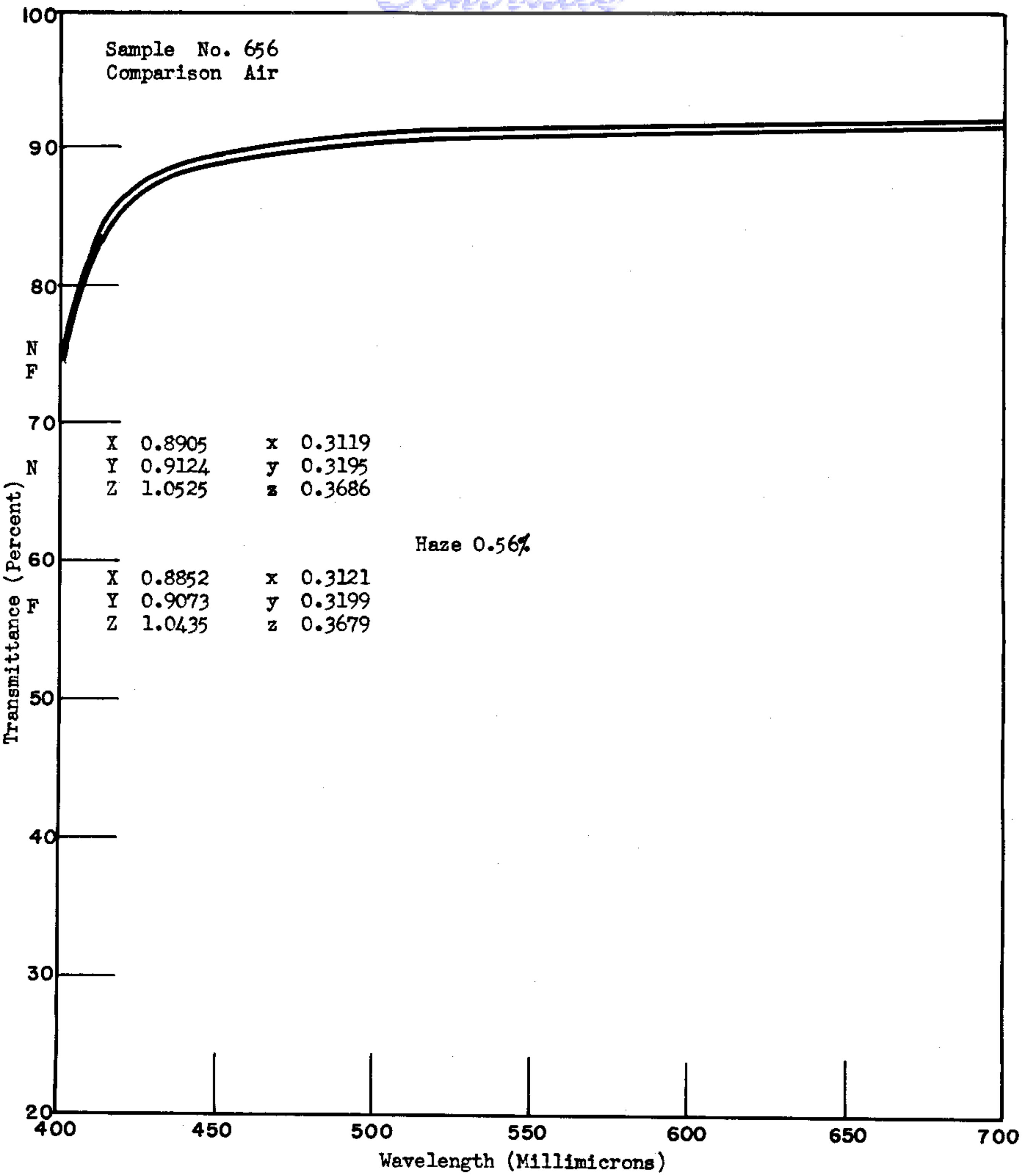


FIGURE 23. TRANSMISSION CURVES FOR "GAFITE" SAMPLE 656

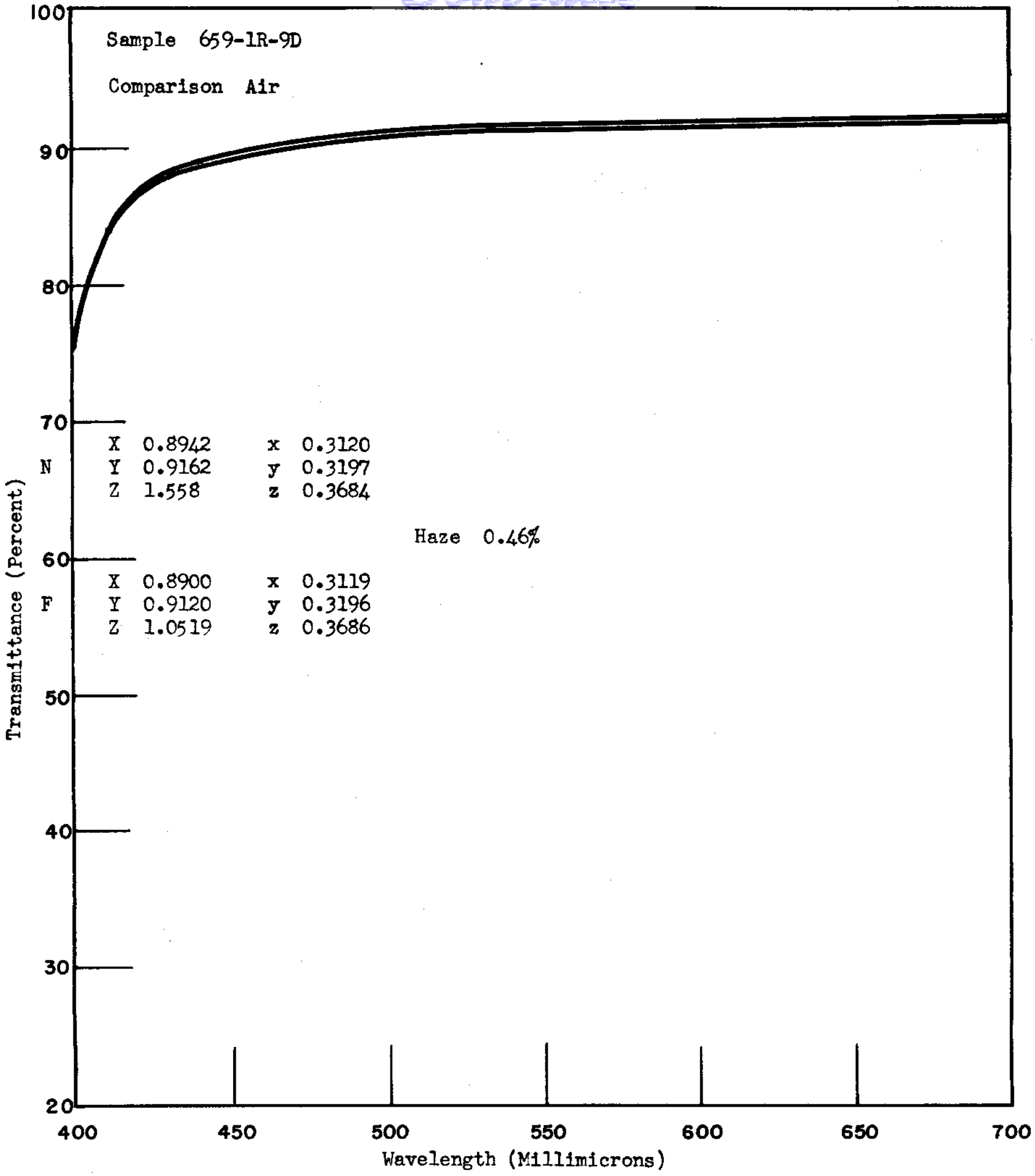


FIGURE 24. TRANSMISSION CURVES FOR "GAFITE" SAMPLE 659-1R-9D

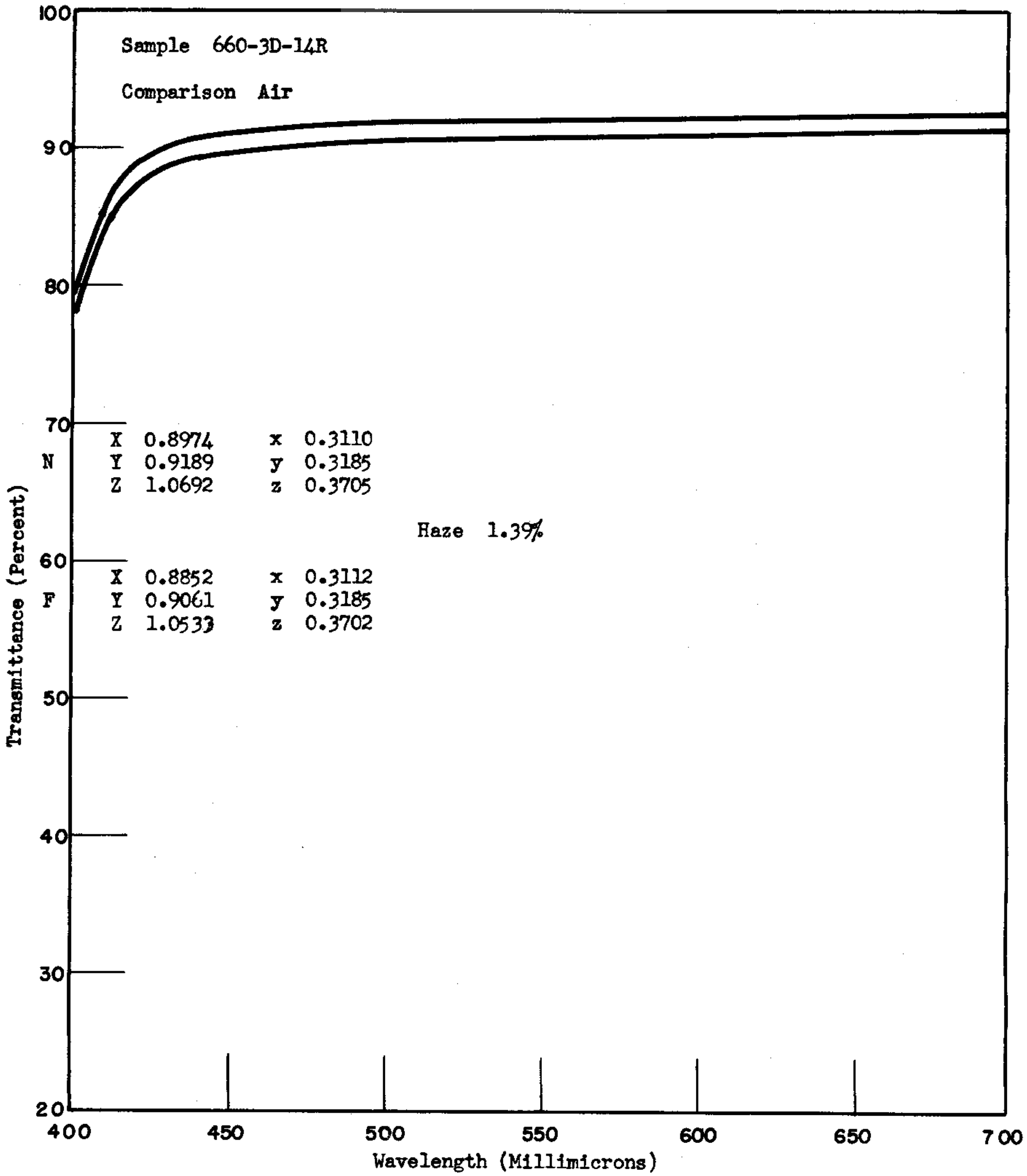


FIGURE 25. TRANSMISSION CURVES FOR "GAFITE" SAMPLE 660-3D-14R

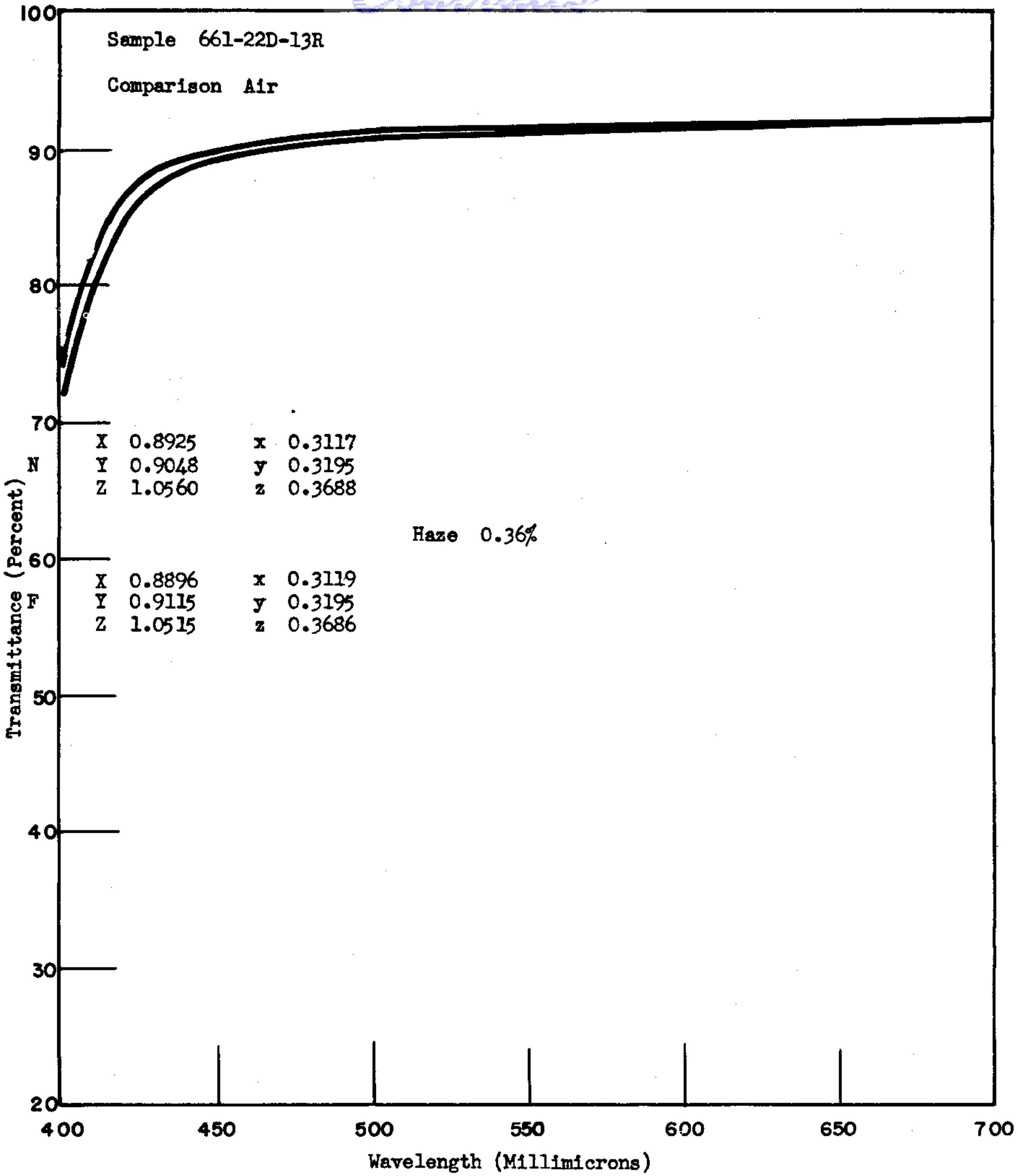


FIGURE 26. TRANSMISSION CURVES FOR "GAFITE" SAMPLE 661-22D-13R

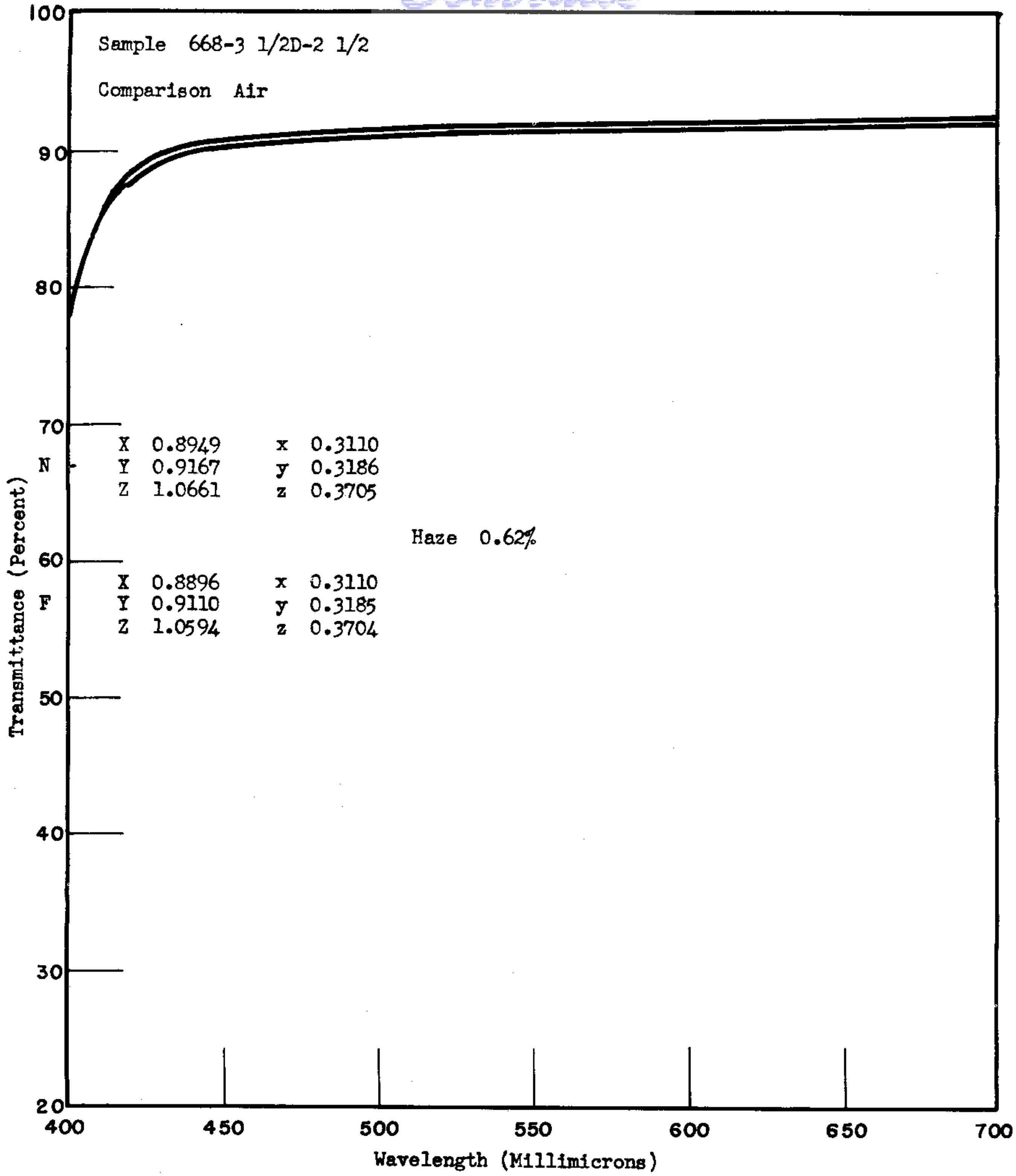


FIGURE 27. TRANSMISSION CURVES FOR "GAFITE" SAMPLE 668-3 1/2D-2 1/2

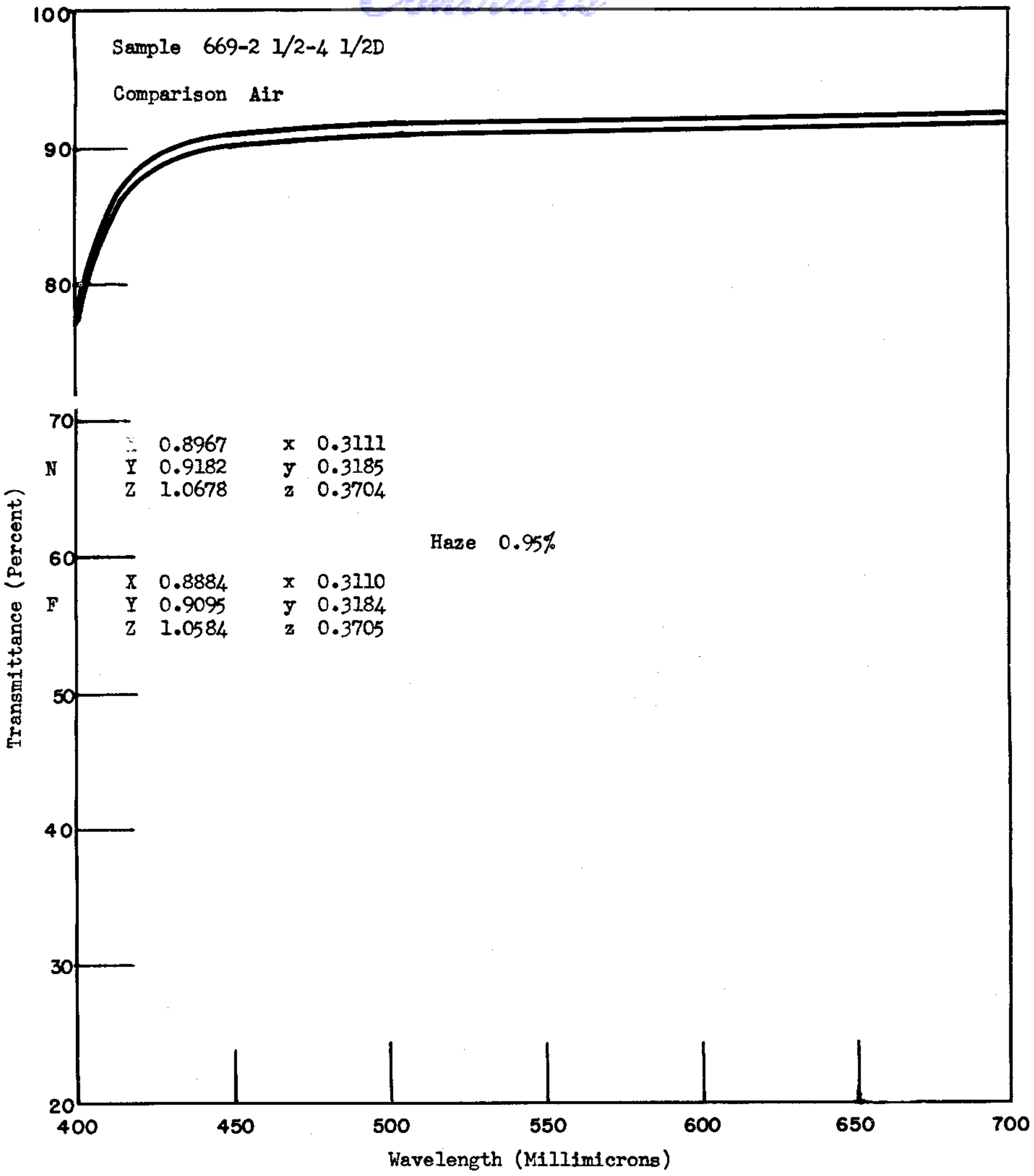


FIGURE 28. TRANSMISSION CURVES FOR "GAFITE" SAMPLE 669-2 1/2-4 1/2D

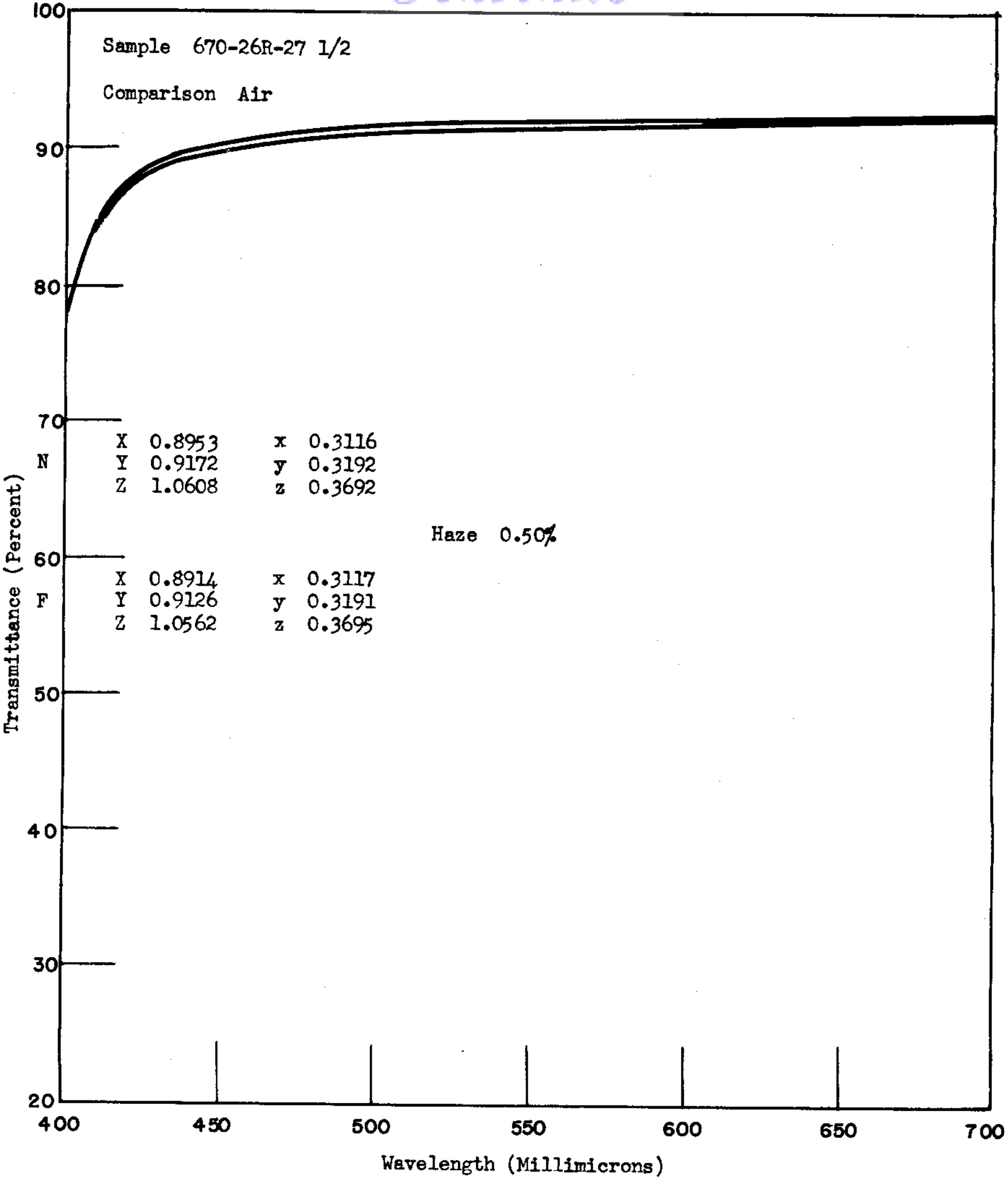


FIGURE 29. TRANSMISSION CURVES FOR "GAFITE" SAMPLE 670-26R-27 1/2

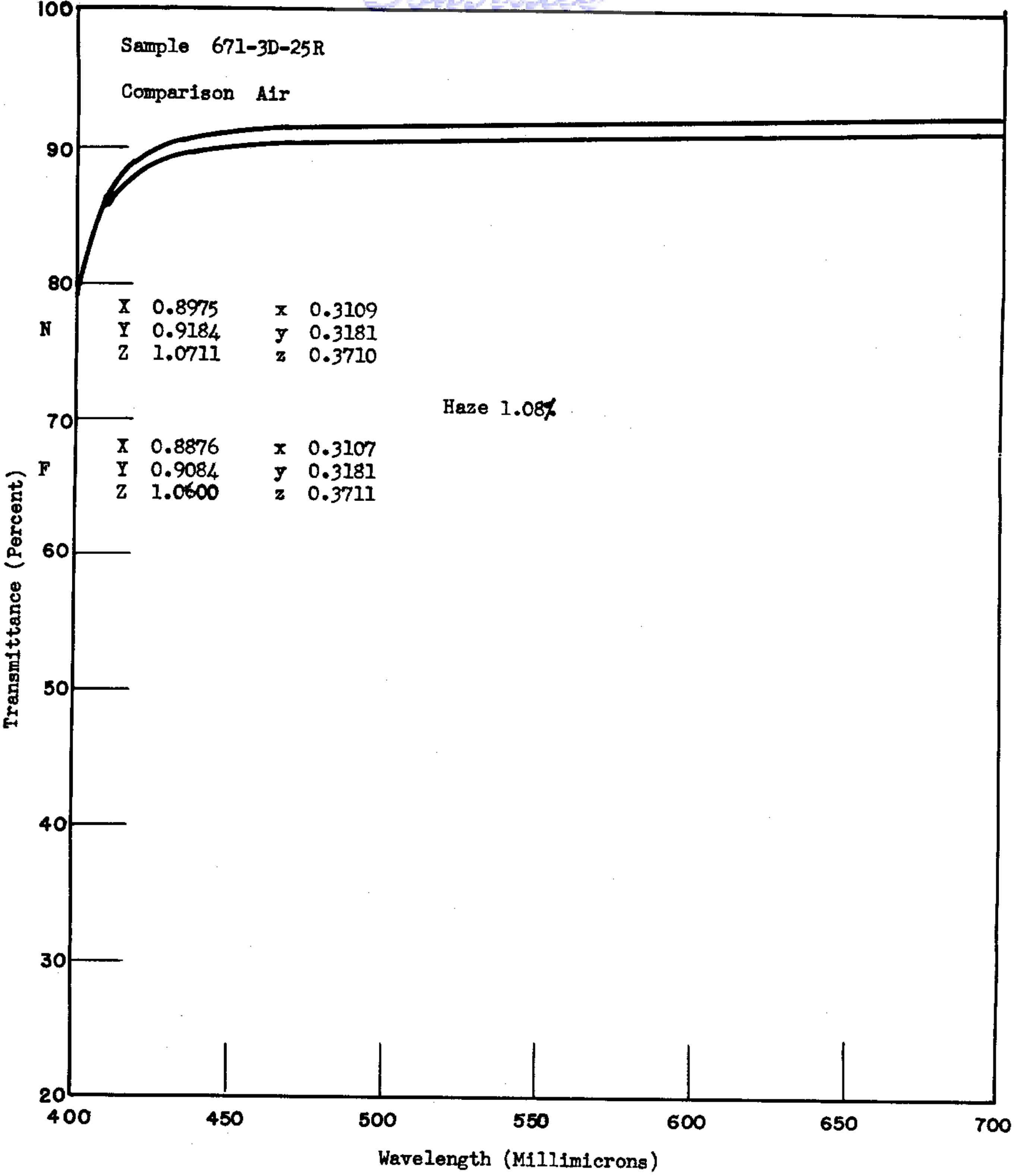


FIGURE 30. TRANSMISSION CURVES FOR "GAFITE" SAMPLE 671-3D-25R

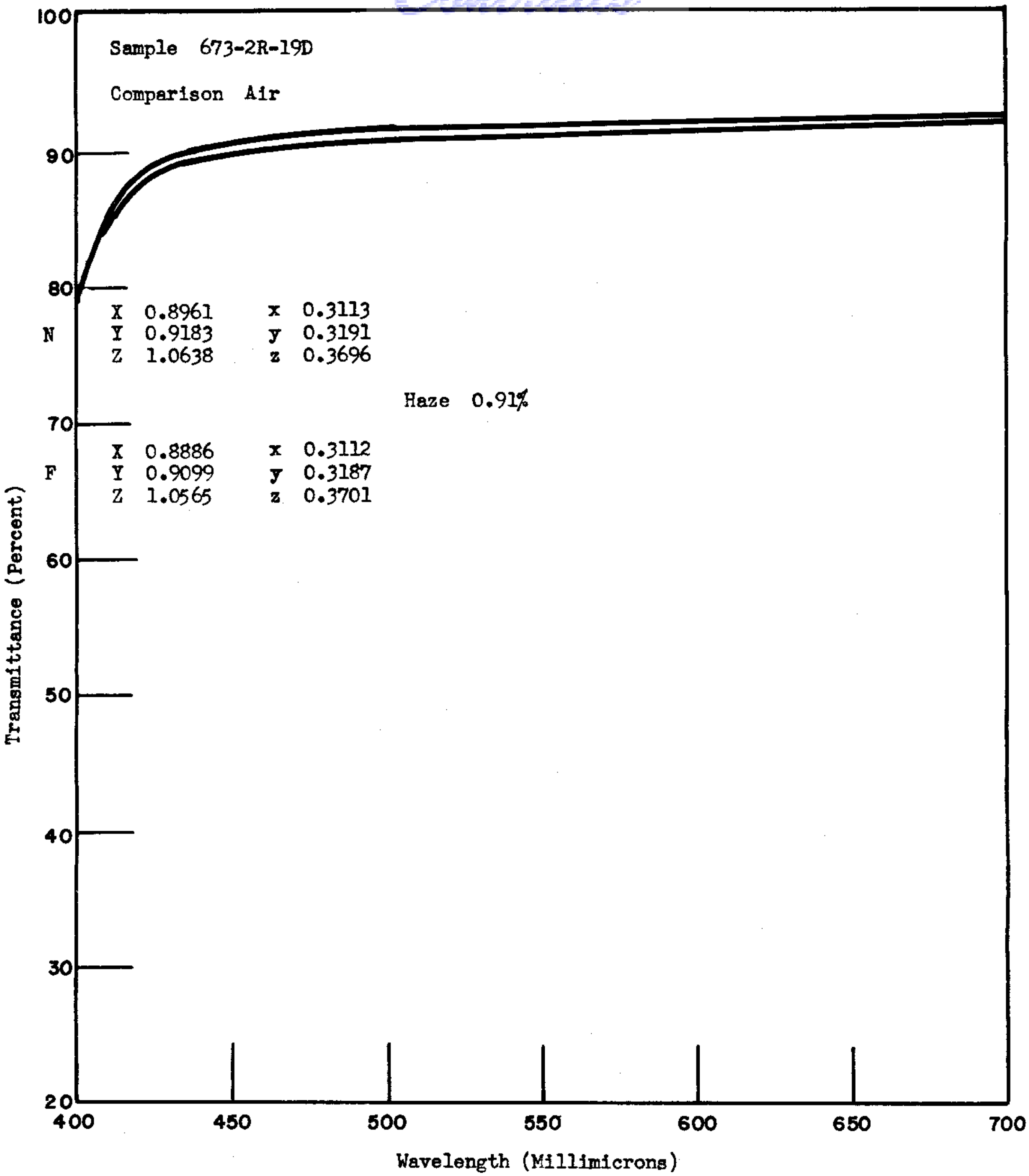


FIGURE 31. TRANSMISSION CURVES FOR "GAFITE" SAMPLE 673-2R-19D

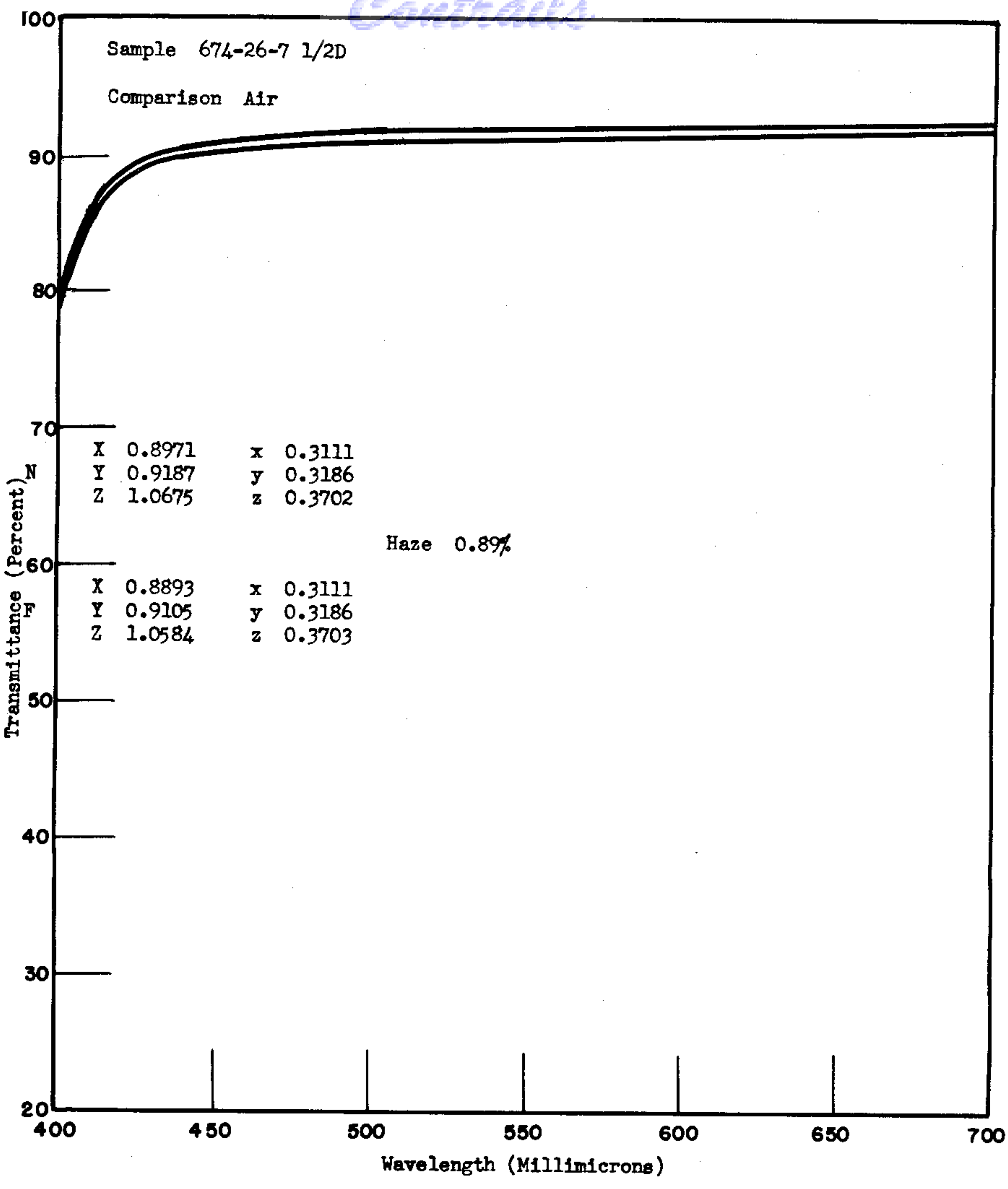


FIGURE 32. TRANSMISSION CURVES FOR "GAFITE" SAMPLE 674-26-7 1/2D

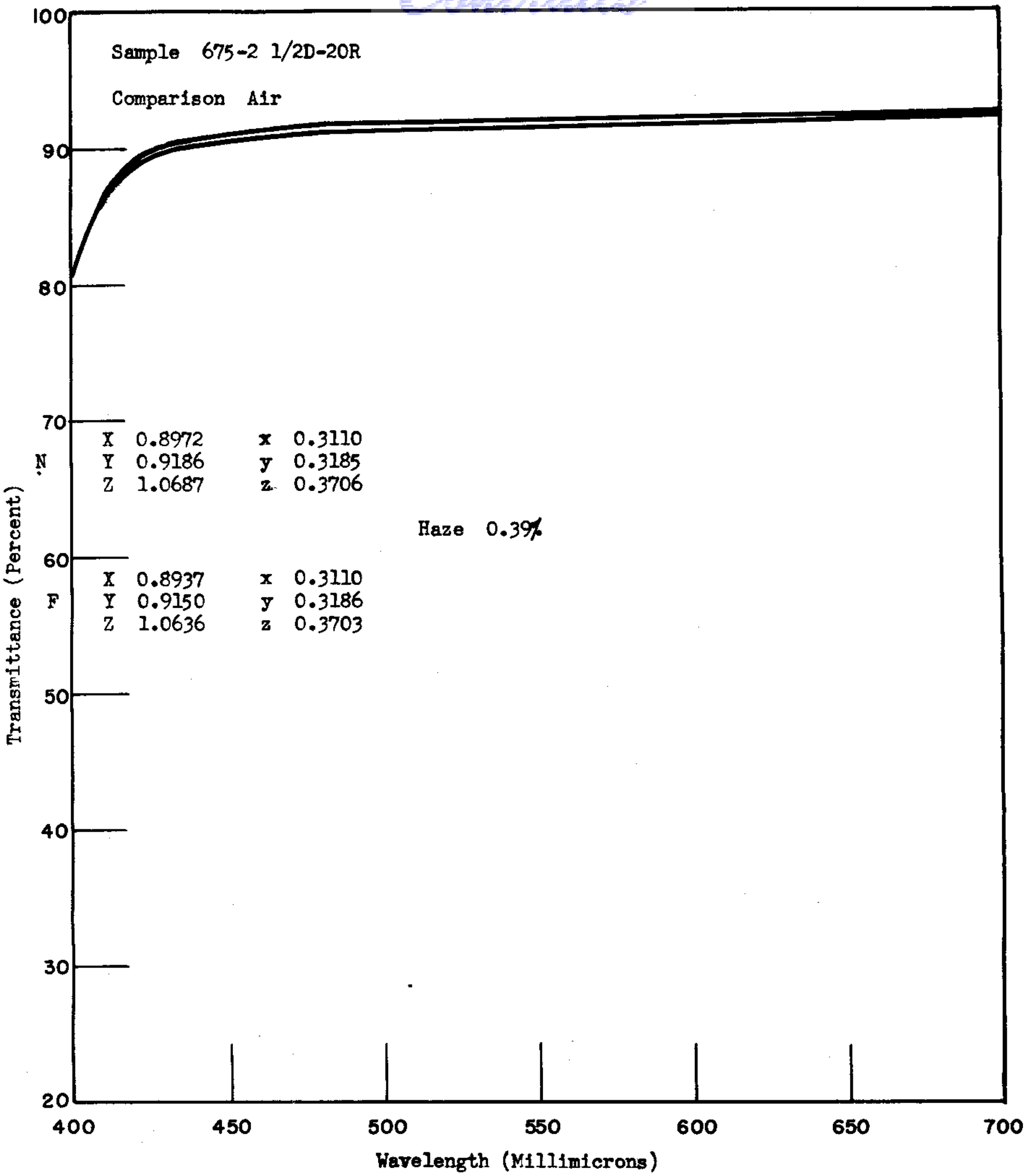


FIGURE 33. TRANSMISSION CURVES FOR "GAFITE" SAMPLE 675-2 1/2D-20R

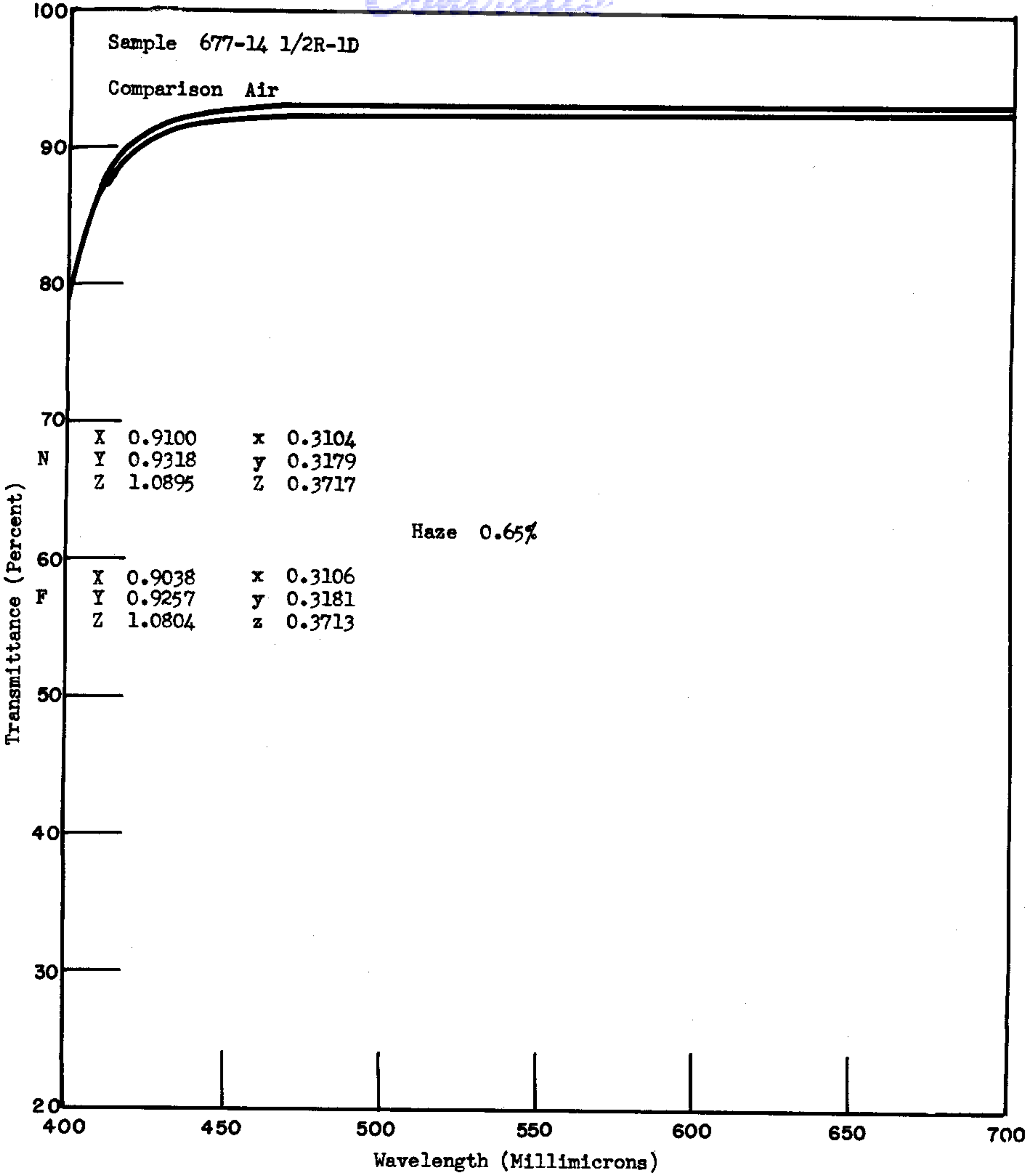


FIGURE 34. TRANSMISSION CURVES FOR "GAFITE" SAMPLE 677-14 1/2R - 1D

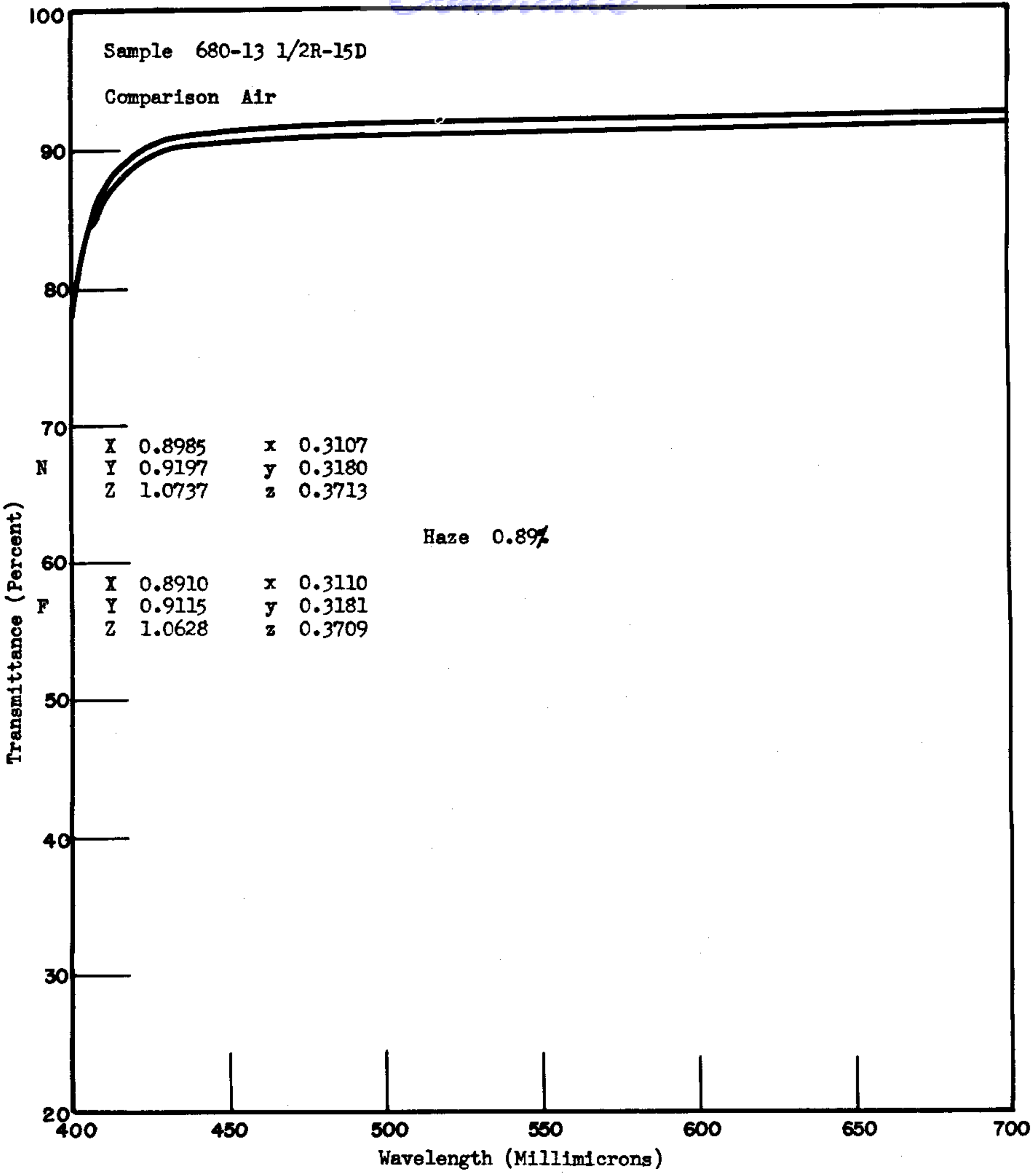


FIGURE 35. TRANSMISSION CURVES FOR "GAFITE" SAMPLE 680-13 1/2R-15D

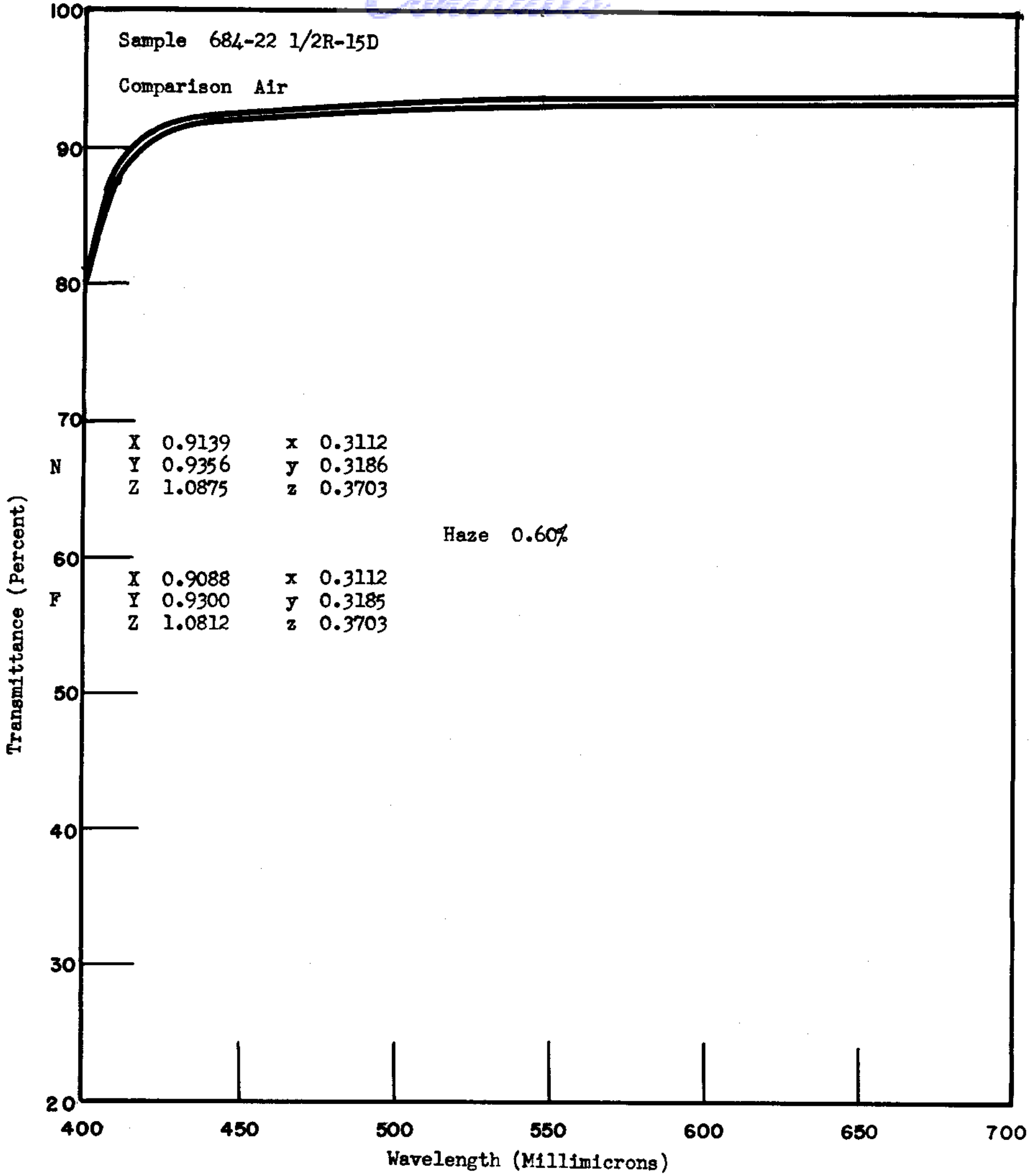


FIGURE 36. TRANSMISSION CURVES FOR "GAFITE" SAMPLE 684-22 1/2R-15D

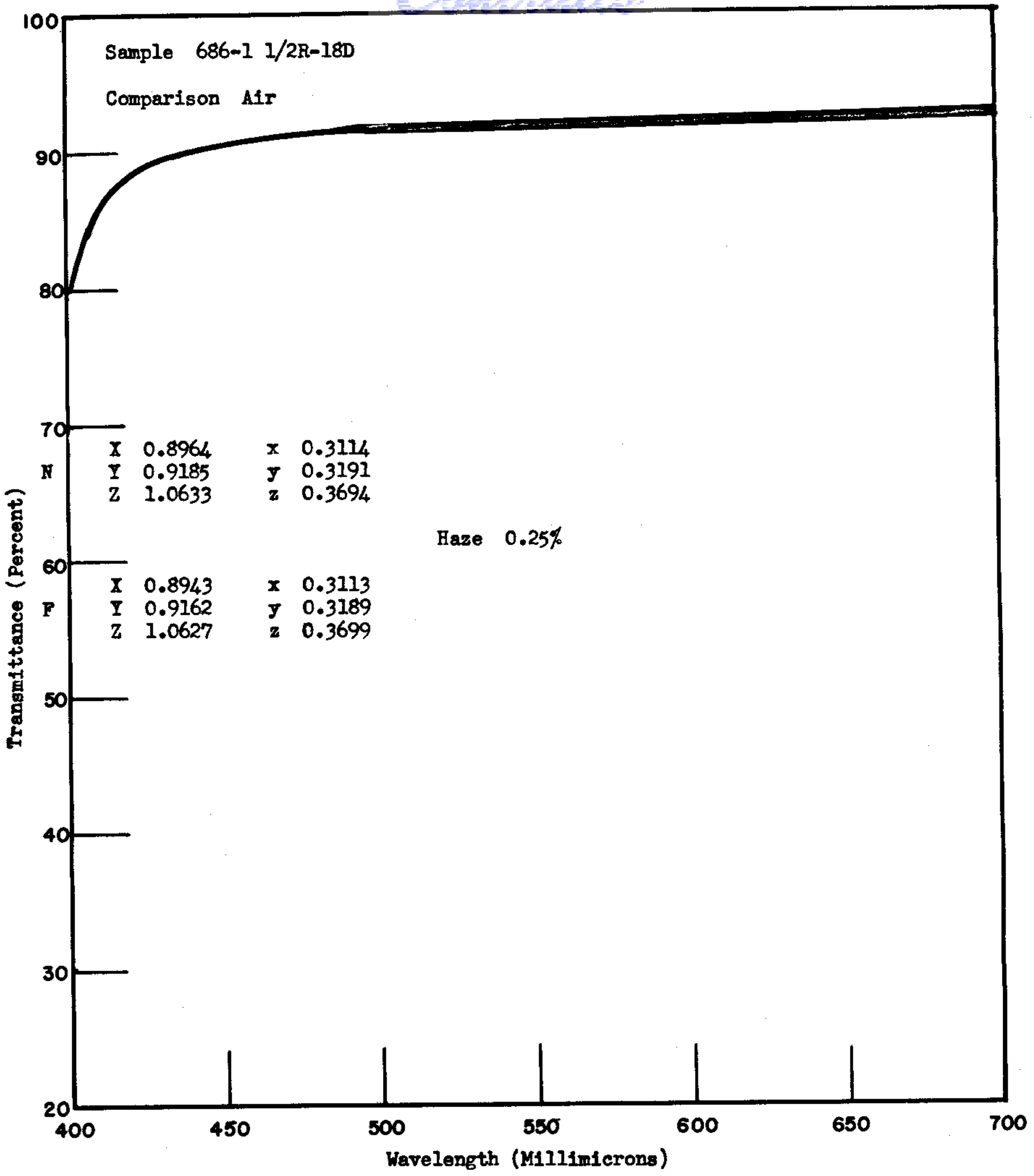


FIGURE 37. TRANSMISSION CURVES FOR "GAFITE" SAMPLE 686-1 1/2R-18D

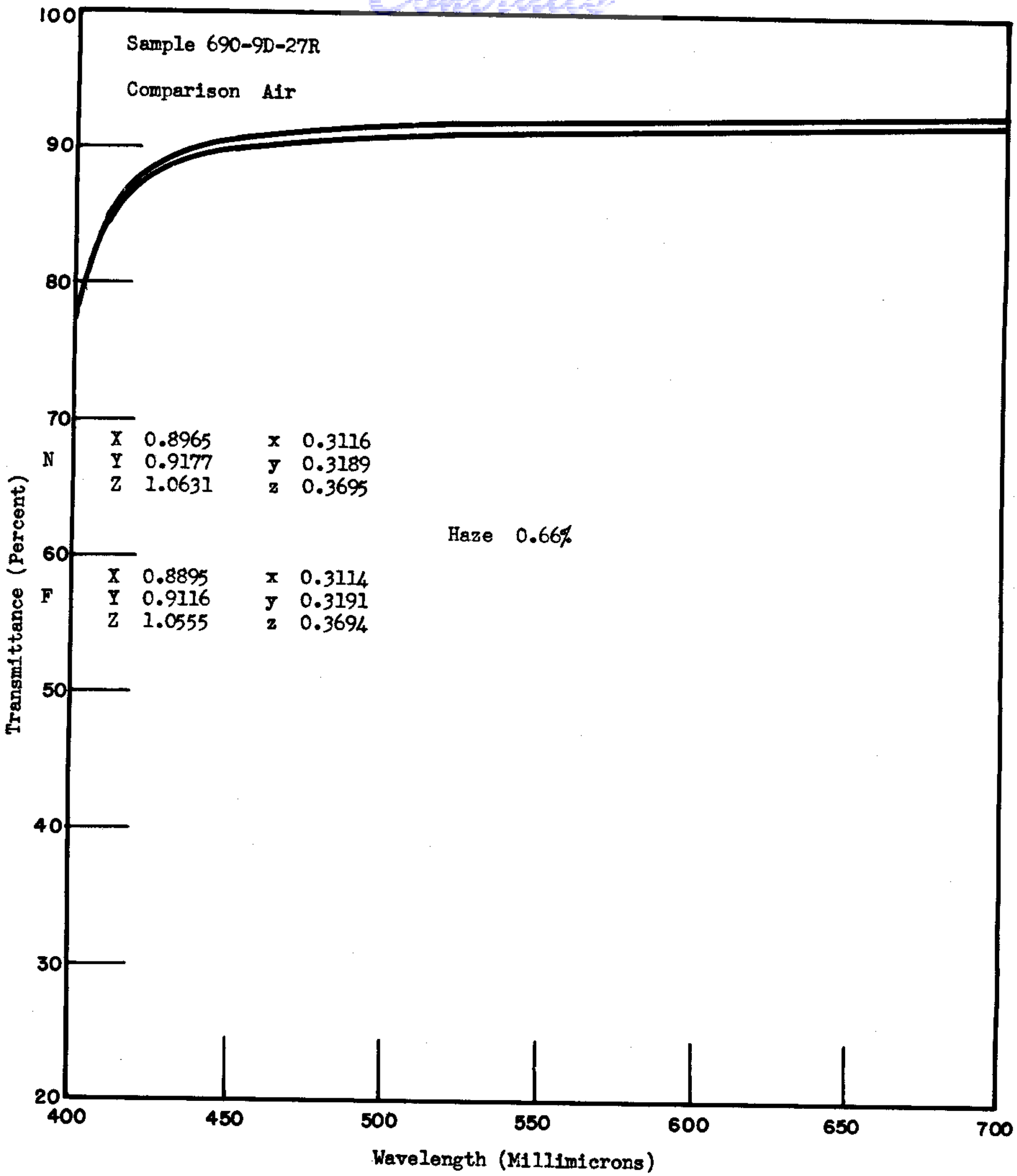


FIGURE 38. TRANSMISSION CURVES FOR "GAFITE" SAMPLE 690-9D-27R

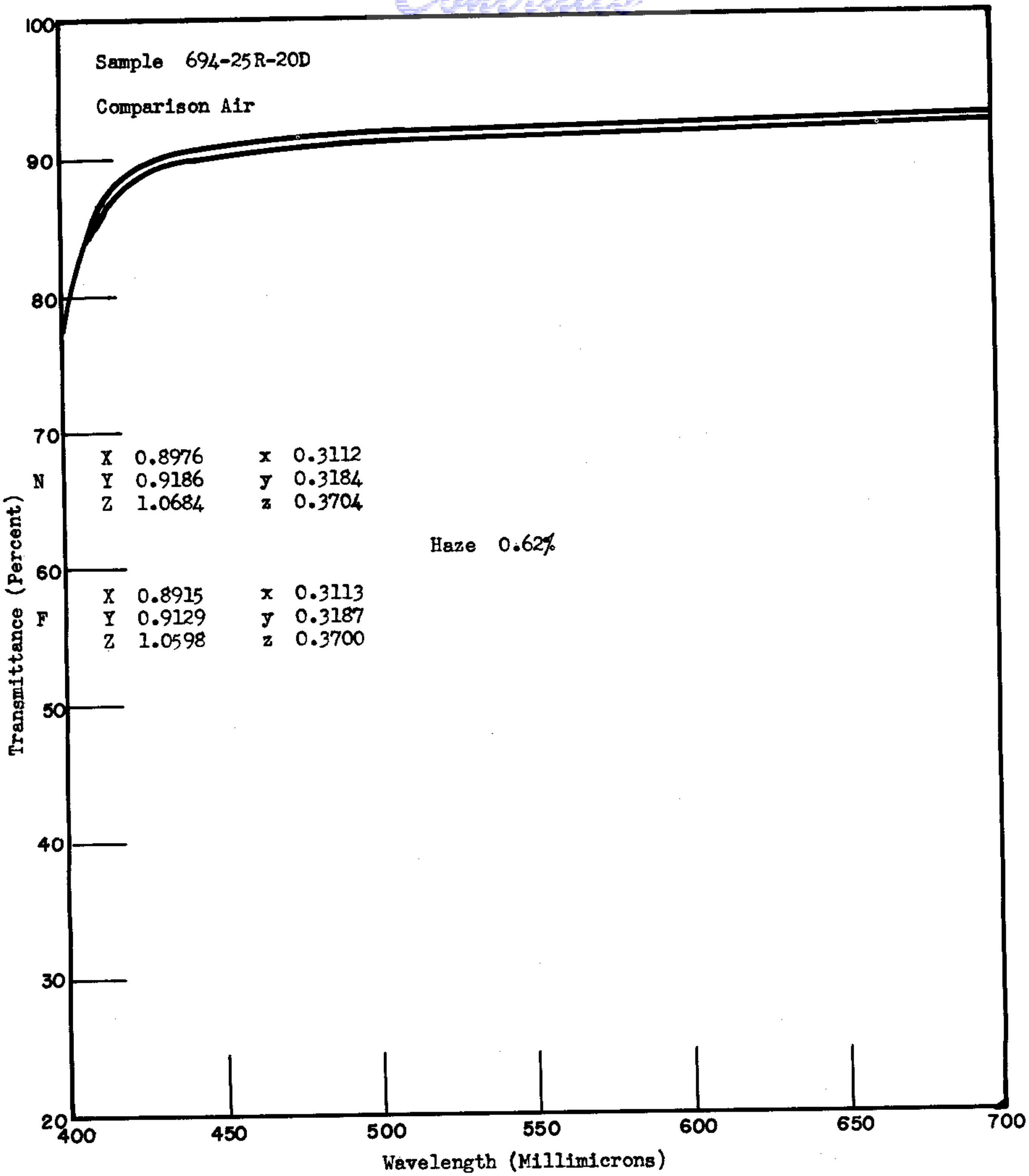


FIGURE 39. TRANSMISSION CURVES FOR "GAFITE" SAMPLE 694-25R-20D

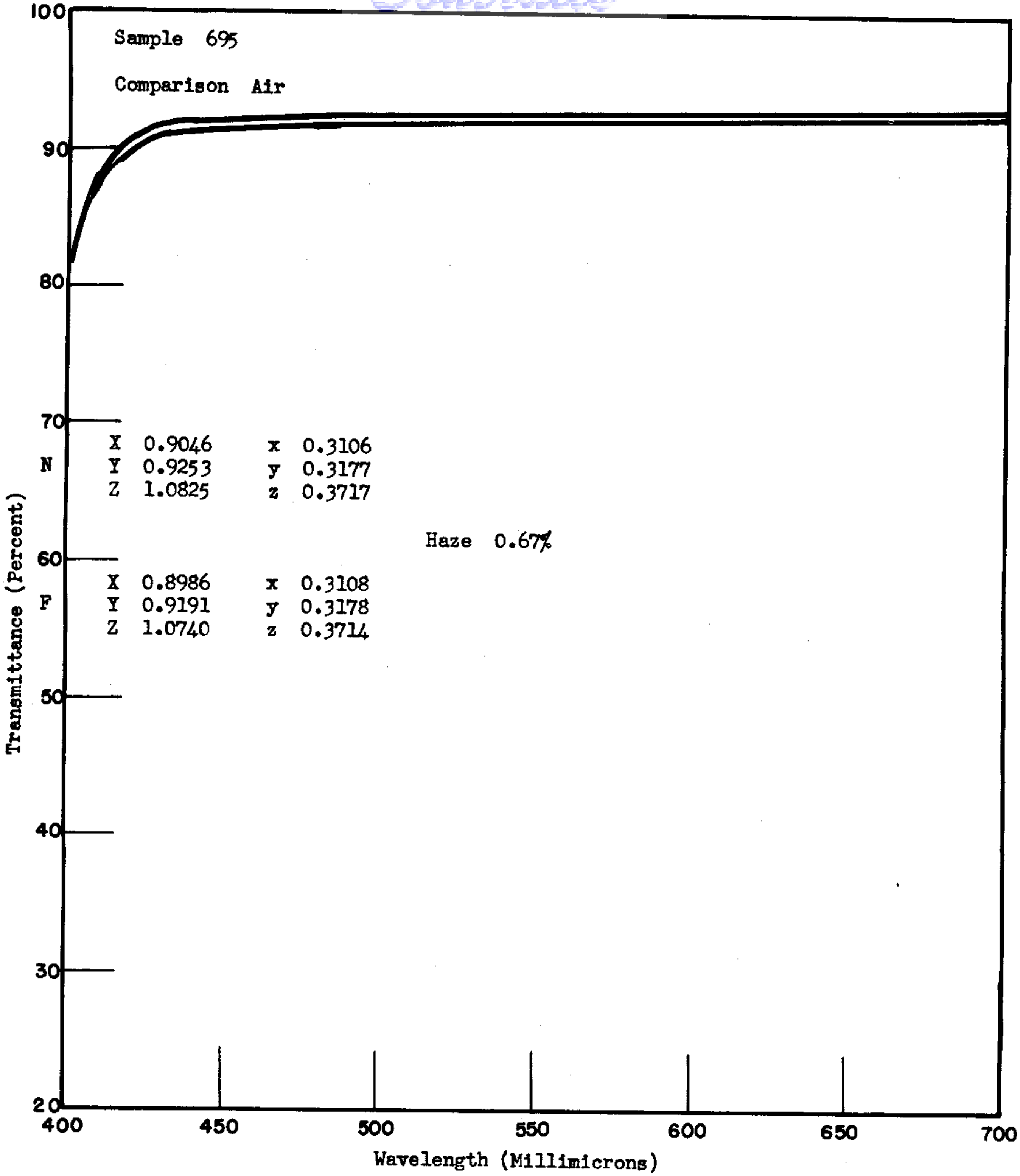


FIGURE 40. TRANSMISSION CURVES FOR "GAFITE" SAMPLE 695

SECTION IV

SUMMARY AND CONCLUSIONS

The outstanding physical properties of polymethyl α-chloroacrylate - among which are its high heat distortion temperature, its high tensile and flexural strengths, its excellent craze resistance, its low notch sensitivity, and its complete formability - are summarized in several reports (6, 7, 8 and 9).

Up to March, 1953 General Aniline and Film Corporation had manufactured two types of "Gafite" (polymethyl α-chloroacrylate) which were designated types A and B. "Gafite" A was the material supplied to the Air Force for the original evaluation tests on polymethyl α-chloroacrylate and it was used by the Bureau of Standards in obtaining early data for the Air Force on this plastic (6). "Gafite" A was subject to gross surface distortions appearing on heating for forming.

"Gafite" B was a further development of this plastic which could be heated for forming without gross distortions appearing in the sheet material. "Gafite" B was distributed to the Aircraft Industries Association and was used by them to obtain the data listed in their report (8). "Gafite" B also was employed by the Air Force to form half-scale canopies with good optics (9).

Both "Gafite" A and B possessed limited heat and light stability. These limited heat and light stabilities were the chief restraints to the use of "Gafite" as an aircraft glazing when work on this contract was begun.

The objective of this contract was the preparation of "Gafite" sheets possessing excellent physical properties which in addition were stabilized against bubbling and yellowing. The tests for the latter properties included nonbubbling on exposure to heat in an air oven at 375°F (191°C) for 45 minutes and nonyellowing on exposure to light for six months in south Florida.

Improved heat stability was desired in order to provide a canopy fabricator more times in which a plastic sheet of "Gafite" could be heated and formed without bubbles appearing in the sheet and destroying its transparency.

Improved light stability was desired so that a canopy of "Gafite" could be exposed to strong sunlight without yellowing causing reduced visibility through the canopy particularly under "twilight" lighting conditions.

Plastic sheets of "Gafite" were developed by the Central Research Laboratory of General Aniline and Film Corporation meeting the heat stability requirements and possessing light stability on exposure in a modified weatherometer for the estimated equivalent of six months in south Florida.

These improvements were obtained by addition of minor amounts of stabilizers (0.1% by weight or less) to methyl α-chloroacrylate monomer of higher purity than that previously obtained, coupled with more complete polymerization of this composition to leave less residual monomer in the polymer. These improvements in heat and light stability are expected to entail little or no sacrifice in the physical properties of "Gafite" plastic sheet. "Gafite" possessing improved heat and light stabilities will be manufactured by General Aniline and Film Corporation.

The approaches taken in this research leading to the development of improved "Gafite" were as follows:

1. All the available samples of "Gafite" A and B as well as certain experimental types of "Gafite" were examined for color changes on roof exposure.

2. The heat stabilities of all available samples of "Gafite" were tabulated.

3. The above results were studied for leads in developing an improved "Gafite". These leads included reduction of residual monomer content, incorporation of hydroxyl containing compounds, and incorporation of ultraviolet absorbers as means of improving "Gafite".

4. A number of agents which were selected as possibly conferring stabilizing effects on "Gafite" were incorporated in "Gafite" to empirically determine whether they affected the stabilities of "Gafite" samples.

Along with the general approach outlined above, incidental experimental work on several specific problems was undertaken to learn how to best carry out certain tests or to establish quality standards for methyl α-chloroacrylate monomer.

Experiments were carried out to determine the most effective method of running screening tests for light stability. These experiments indicated that for screening tests, the use of polished surface samples for light exposure was preferable for ease of differentiation by eye; whereas, for measurement of optical changes occurring during exposure tests, the use of samples possessing ground surfaces would yield greater measurable changes in optical density under the same light exposure than polished surface specimens.

The heat stability test also was investigated. The importance of heat transfer conditions in determining the heat stability results was demonstrated. The best method of determining heat stability was deemed to be by continuous heating in an oil bath.

Since methyl α-chloroacrylate is susceptible to changes in color and in heat stability caused by contact of the monomer with air, a simplified method of casting this monomer and avoiding unpredictable air contact was developed utilizing polyvinyl alcohol tubing for sealing the ends of casting tubes.

Experiments on drying and purification of methyl α-chloroacrylate were carried out to learn the physical constants for high purity methyl α-chloroacrylate and to learn the best methods of preparing high purity methyl α-chloroacrylate. The properties of polymer prepared from such monomer were determined.

A study of inhibitors for the polymerization of methyl α-chloroacrylate was undertaken so that this very reactive monomer could be stabilized throughout the necessary purification steps. Three outstanding inhibitors were found: dihydro-1,4,5,8-tetrahydroxyanthraquinone, copper oxide, and chloranil.

Curing conditions were studied to establish the curing cycle for reducing the residual monomer content to a minimum without causing chemical changes in the polymer. Optimum curing conditions were found by infrared analyses of thin film castings of "Gafite".

Three types of heat and light stabilized "Gafite" sheets were supplied to Wright Air Development Center in June, 1954 for extensive testing. These sheets were prepared utilizing the results of the research outlined.

In two of these types triethyl phosphite and ethylene sulfite were employed as additives to further reduce the initial slight color of "Gafite". By use of proper purification and casting techniques "Gafite" possessing little initial color could be obtained without the presence of these agents. "Gafite" with improved heat and light stabilities was obtained without addition of any stabilizer except "Uvinul 400" (0.05% by weight) in company with dibutyl tin diacetate (0.05% by weight) as catalyst.

Programs for extensive evaluation of improved "Gafite" as an aircraft glazing material are being conducted.

Much of this work is being coordinated by Wright Air Development Center who also have a program under a current contract with Rohm and Haas Company for the evaluation of the physical properties of stretched "Gafite".

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