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TESTING OF SILICONE RUBBER AT ELEVATED TEMPERATURES

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FOREWORD

This report was prepared by The Connecticut Hard Rubber Company, New Haven, Connecticut, under USAF Contract No. AF 33(616)-2542. The contract was initiated under Project No. 7340, "Rubber, Plastic and Composite Materials," Task No. 73405, "Compounding of Elastomers," formerly RDO No. 617-12, "Compounding of Elastomers," and was administered under the direction of the Organic Materials Branch, Materials Laboratory, Directorate of Research, Wright Air Development Center, with Lt J. M. Kelble acting as project engineer.

The period covered by this report is from 1 July to 15 August 1955.

ABSTRACT

An apparatus to measure tensile strength, elongation, and tear strength of silicone rubber at elevated temperatures was designed and constructed. Fifteen different silicone compounds were tested at room temperature, 212°F and 400°F.

Although it is known that physical properties of silicone rubber are only slightly affected after the rubber is aged at high temperatures, it was found that they are greatly reduced when measured at high temperatures.

The best tensile strength, elongation and tear strength at 400°F were displayed by Cohrlastic HT Heat Stable (CHR 5908A), a compound containing Valron (Du Pont) as the filler. CHR Compound 5901, composed of Dow Corning 6-128 with glass wool as an additional filler, also exhibited good tensile and tear strengths at 400°F.

The least reduction in these properties at 400°F, as compared with results obtained on the silicone compounds at room temperature, was displayed by CHR 5901. Dow Corning Silastic 152, postcured at 600°F, showed the least change in elongation.

Many of the materials tested were not developed or intended by the manufacturer for the conditions to which they have been subjected. Any failure or poor performance of a material is therefore not necessarily indicative of the utility of the material under less stringent conditions or for other applications.

PUBLICATION REVIEW

This report has been reviewed and is approved.

FOR THE COMMANDER:

M. R. Whitmore

M. R. WHITMORE
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INTRODUCTION

Very few comprehensive studies have been made of the properties of elastomeric compounds at elevated temperatures. It is true that many laboratories are equipped to measure tensile strength, elongation, modulus and tear-resistance at temperatures up to 212°F. For example, Morron and his co-workers (1)* have reported the effect of temperatures from -60°F to +260°F on the tensile strength of vulcanizates of natural rubber, butyl, neoprene, GR-S and Perbunan. The tensile strength of all of these rubbers decreases with an increase in temperature, but the rate of decrease is different for each rubber. For those rubbers which have been studied, such as the ones named above, tensile strength decrease is already serious at a temperature of 212°F. It is obvious that better rubbers than these are needed for use at temperatures from 300°F to 500°F and above.

There are applications in which rubber is not strained to the breaking point during its use at elevated temperatures. For example, in cases where the rubber is confined, it may still serve a useful purpose even though the ambient temperature may be as high as 200°F to 300°F. Because of this, during the past thirty years the well-known artificial aging tests have been developed in which elastomeric compounds are subjected to high temperatures for various periods of time in the presence of air, oxygen or various fluids. After such aging, the compounds are examined at room temperature for strength, hardness, swelling, etc. Through the use of such tests, outstanding improvements in the aging-resistance of all elastomers have been made possible.

In this new era of supersonic flight, rubber items will soon be required to perform at temperatures of 300°F to 500°F, and probably as high as 1000°F (2). Silicone rubber has been far more successful in resisting aging at elevated temperatures than any other elastomer. This is apparently the natural result of the oxidation-resistance of the dimethylsiloxane chains. Little has been published on the testing of silicone rubber at elevated temperatures. However, Doede and Panagrossi (3) have shown that modulus, hardness and tensile strength decrease less with the silicones, over the temperature range of 0 to 400°F, than with most other rubbers.

More information concerning the properties of silicone compounds measured at high temperature is necessary; there is

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*/Numerals in text refer to Bibliography at end of report

then a need for simple methods of measuring tensile strength, elongation and tear strength at temperatures from room temperature to 400°F, and higher. It is the purpose of this contract to develop a test apparatus in which these measurements can be made and to provide preliminary high-temperature testing data on a few silicone rubber compounds.

I. EXPERIMENTAL WORK

A. Design of a High-Temperature Test Apparatus

Objective:

The objective of this experiment was to design and build an apparatus in which physical properties of rubbers at elevated temperatures could be measured.

Procedure:

The test apparatus consisted of a box made of Marinite, fitted to the front of a Scott Tester, Model L6, as shown in Figures 1 and 2. To the front of this box was fitted a cover containing a window made of two thicknesses of Pyrex glass, separated by an insulating air space. At the sides and in the rear of the box were placed strips of Transite wound with resistance-wire for heating. In order to insure that the test samples were heated by convection alone, Transite, placed between the heating strips and the samples being tested, served as a radiation seal.

The heating system of the box was checked. A pyrometer was used to measure temperature. One thermocouple lead was embedded in the sample, while another was placed outside the sample, two millimeters away. The box was then heated at a rate of 30 degrees per minute. It was found that the difference between the two thermocouples was substantially constant during the heating period. The temperature inside the sample remained from 10 to 15 degrees cooler as the sample was heated. In actual tensile tests, a thermocouple was placed two millimeters away from the sample, which was pulled approximately one minute after the thermocouple recorded the desired test temperature.

In Figure 3 are pictured Z-clamps which hold the rubber sample that is being tested. The lower Z-clamp is fixed. The upper one is attached by means of a braided stainless steel wire, operating through a system of Cenco frictionless pulleys, to the fixture which holds the lower clamp on the unmodified portion of the Scott Tester. Because of the 180-degree change in direction in the pulley system, the reading on the dial is twice the force applied to the sample. The necessary calculation for tensile strength is as follows:

$$\text{Tensile Strength, psi} = \frac{\text{Dial Reading}}{2 \times \text{Cross-Sectional Area}}$$

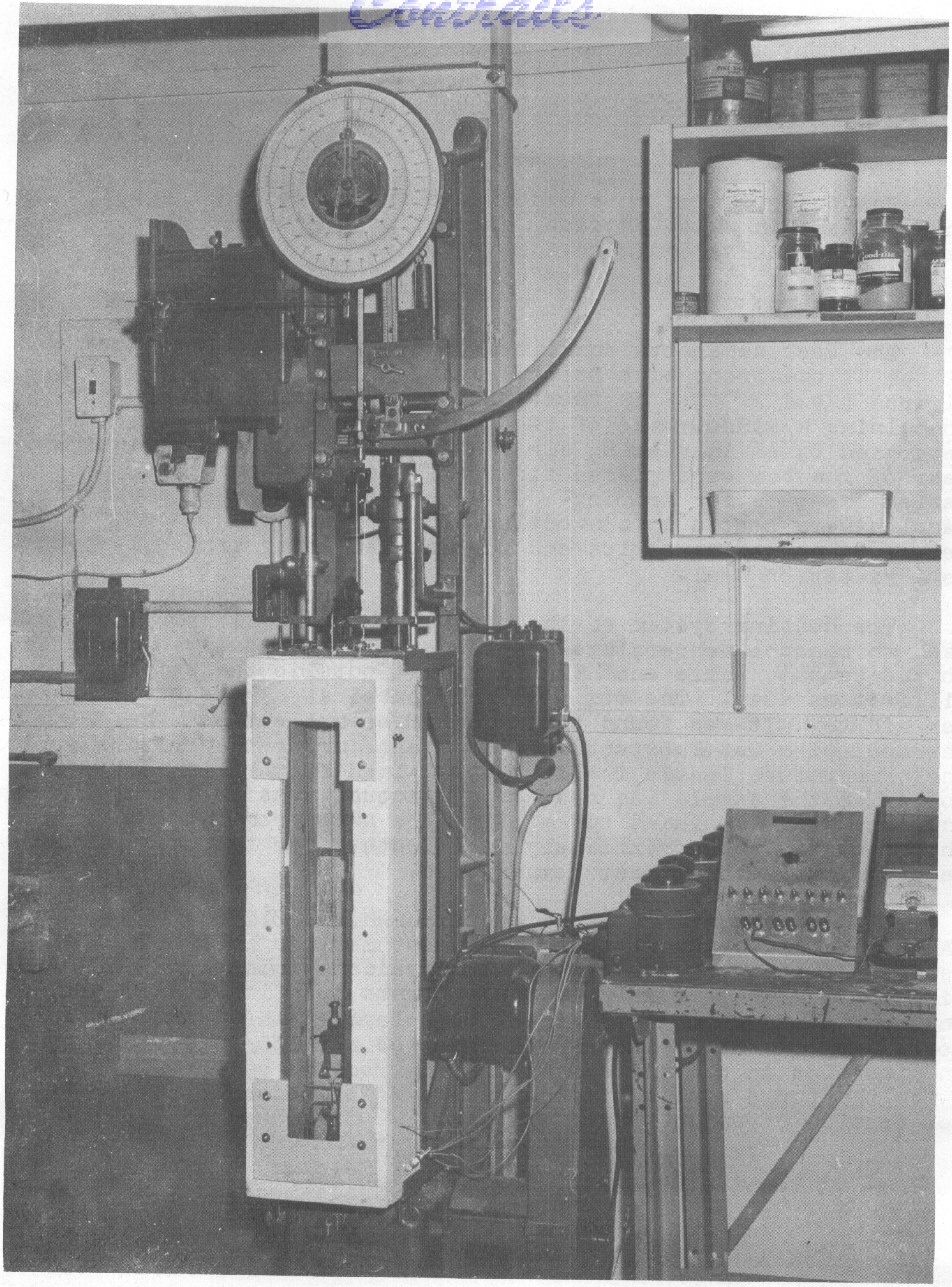

FIG. 1: HIGH-TEMPERATURE TEST APPARATUS

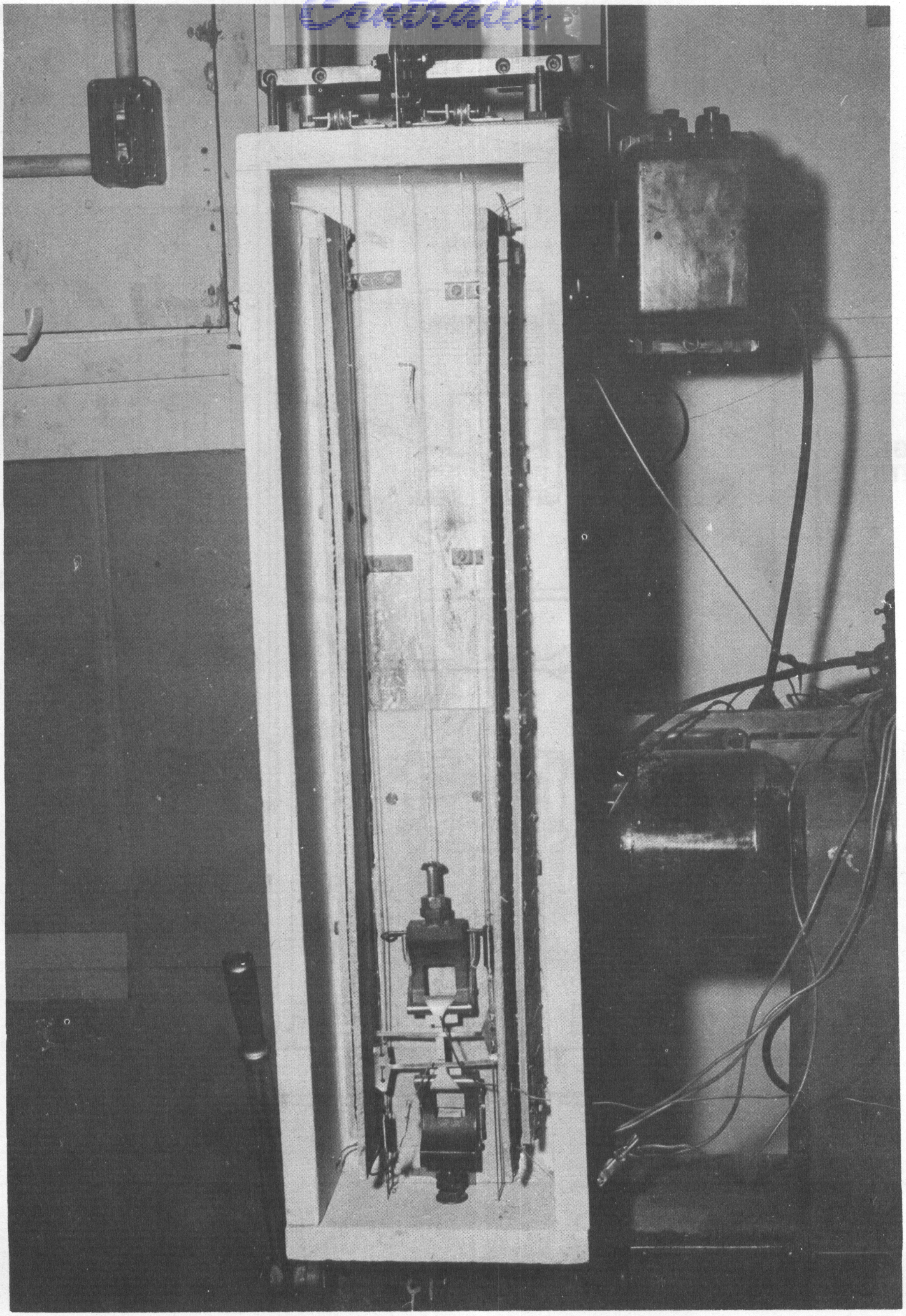

FIG. 2: HEATER BOX ASSEMBLY

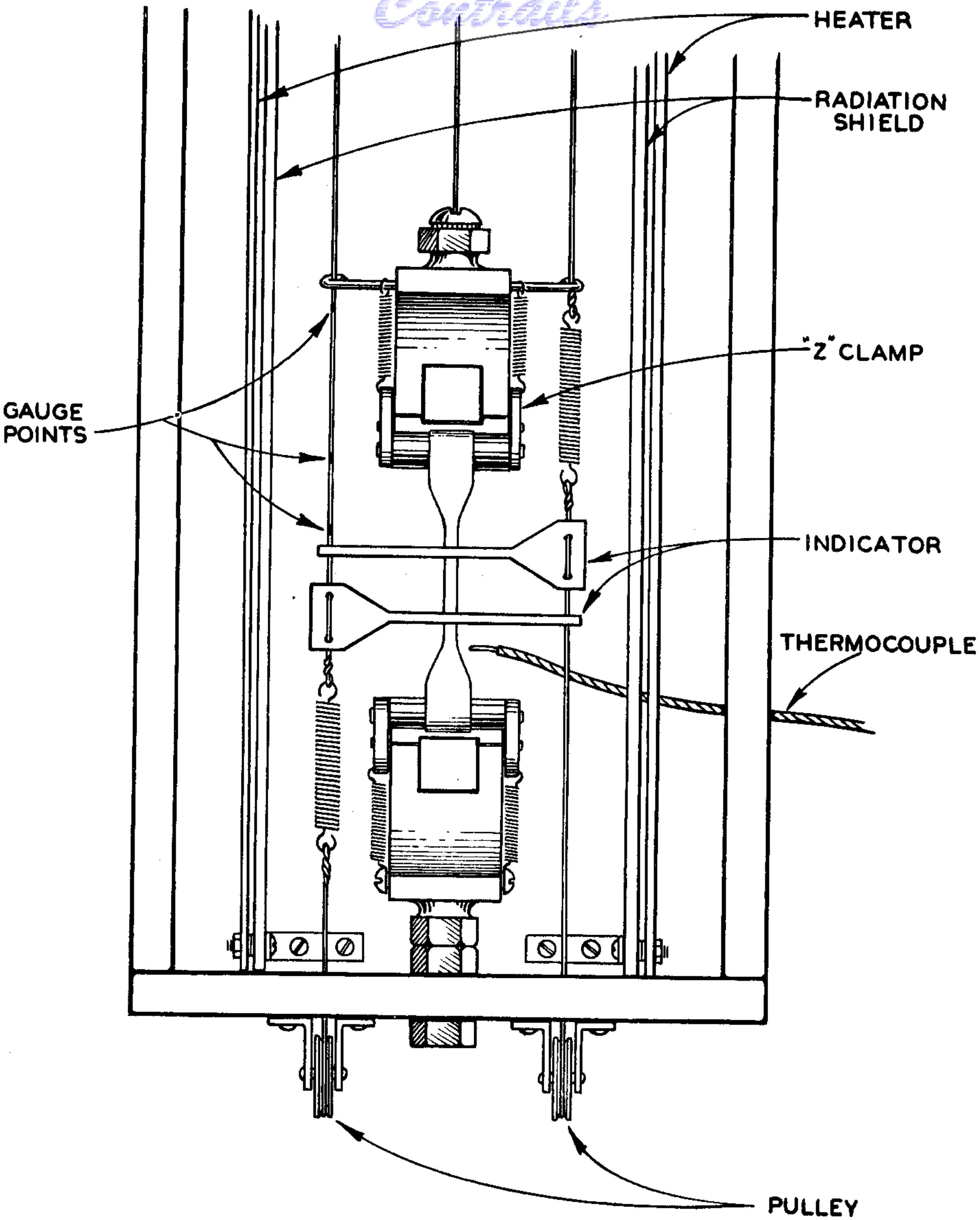

FIG.3: DETAIL OF TEST APPARATUS.

In this diagram are also pictured the indicators used for measuring the elongation of the rubber sample. These indicators ride on spring-loaded, braided stainless steel wire which can be moved by means of pulleys. On the wire to which the lower indicator is attached, there are three beads of silicone resin which serve as gauge points for measuring modulus at specific elongations at high temperature.

B. Calibration of the High-Temperature Test Apparatus

Objective:

The objective of this experiment was to determine the difference in measurements obtained in the test apparatus and those obtained with an unmodified Scott Tester.

Procedure:

The test apparatus was calibrated by comparing measurements (tensile strengths, elongations and tear strengths) obtained in the test apparatus with those obtained with the unmodified Scott Tester. Tensile strength and elongation were determined according to ASTM Specification D412-49T; the tear strength according to ASTM Specification D624-48, Method B; and the hardness according to ASTM Specification D676-49T. Tests were conducted at room temperature only. The results of these comparative tests are given in Table I.

Results:

The median result of three test samples is reported in Table I. An examination of the results of this calibration showed little systematic variation between the two series of measurements. The average deviations between the two sets of measurements were as follows: tensile strength, 10.2 percent; elongation, 7.7 percent; and tear strength, 29.7 percent. The agreement was satisfactory enough to justify the use of the test apparatus for relative measurements at various temperatures.

TABLE I

CALIBRATION OF THE TEST APPARATUS[1]

Compound[2]	Hardness, Shore A	Tensile Strength, psi			Elongation, %			Tear Strength, lbs/in.		
		Scott Tester	Test Apparatus	Percent Deviation	Scott Tester	Test Apparatus	Percent Deviation	Scott Tester	Test Apparatus	Percent Deviation
Connecticut Hard Rubber:										
5908A	56	1650	>1540	-6.7[3]	875	900	+2.9	195.0	206.0	+5.7
R-11438	53	780	745	-4.5	360	340	-5.6	38.5	48.5	+26.0
5910	58	680	770	+13.2	290	320	+10.3	46.5	56.0	+20.4
5912	57	630	575	+7.1	240	270	+12.5	36.5	48.0	+31.5
5904	57	470	510	+8.5	225	240	+6.7	37.0	48.5	+31.1
5901	85	670	710	+6.0	90	100	+11.1	69.0	59.0	-14.5
Dow Corning:										
6-128	71	590	640	+8.5	100	110	+10.0	15.5	32.5	+110.
152	49	520	580	+11.5	310	315	+1.6	50.0	50.5	+1.0
152 (600)	55	490	470	-4.1	270	230	-14.8	64.0	60.5	+5.5
675	61	555	625	+12.6	280	290	+3.6	64.0	81.5	+27.4
6-526	48	685	650	-5.1	280	260	-7.1	35.5	43.0	+21.1
250	47	420	495	+17.9	200	215	+7.5	56.5	63.5	+12.4
General Electric:										
551A	43	470	490	+4.3	265	280	+5.7	25.0	37.0	+48.0
750A	57	620	690	+11.3	180	185	+2.8	23.0	31.5	+36.9
100	45	205	270	+31.8	110	125	+13.6	8.1	12.5	+54.3
Average Deviation				10.2			7.7			29.7

1. Readings taken at room temperature
2. Formulas and cures for these compounds are listed on page 10.
3. Since the tensile strength value was higher than that measurable in the test apparatus, this figure is only approximate.

C. Physical Properties of Silicone Rubbers at Elevated Temperatures

Objective:

It was the objective of this experiment to determine in the test apparatus, the modulus, tensile strength, elongation and tear strength of various compounds at room temperature, 212°F and 400°F. The formulations of the compounds tested are listed, when possible, as follows:

1. CHR 5908A, Cohrlastic HT Heat Stable, containing Valron filler, (Du Pont) 50 parts PHR

2. CHR R-11438

Linde W-96	100.0	parts
Linde W-95	36.0	"
Hi Sil X-303	46.0	"
Zinc Oxide	5.0	"
Di-tertiary-butyl peroxide	0.8	"

3. CHR 5910

Linde W-96	100.0	parts
Linde W-95	36.0	"
Hi Sil X-303	50.0	"
Zinc Oxide	5.0	"
Iron Oxide	2.0	"
Di-tertiary-butyl peroxide	0.8	"

4. CHR 5912

General Electric SE-52	100.0	parts
Hi Sil X-303	50.0	"
Benzoyl peroxide	1.5	"

5. CHR 5904

Dow Corning Silastic 152	50.0	parts
Dow Corning Silastic 675	50.0	"

6. CHR 5901

Dow Corning Silastic 6-128	100.0	parts
Pyrex Glass Wool	10.0	"

7. Dow Corning Silastic 6-128

8. Dow Corning Silastic 152
9. " " " 152 (600)
10. " " " 675
11. " " " 6-526
12. " " " 250
13. General Electric SE-551A
14. " " SE-750A
15. " " SE-100

The above compounds received oven cures of 1 hour at 300°F followed by 24 hours at 480°F, with the exception of the compound designated Silastic 152 (600) and Cohrlastic HT Heat Stable. The former compound was cured for 3.5 hours at 600°F after its normal cure of 1 hour at 300°F and 24 hours at 480°F. The recipe and cure of Cohrlastic HT Heat Stable is proprietary information of The Connecticut Hard Rubber Company which does not wish to reveal the details.

Procedure:

Modulus, tensile strength and elongation were determined according to ASTM Specification D412-49T, and the tear strength according to ASTM Specification D624-48, Method B. All measurements were made at three different temperatures, room temperature, 212°F and 400°F. As stated in Experiment A, all samples were pulled after soaking approximately one minute at the desired temperature.

Tensile strength, elongation and tear strength data are listed in Tables II, III and IV, and stress-strain curves are shown in Figures 4 through 18. It should be noted that the scale for Figure 4 (Cohrlastic HT) has been reduced so that the data may be completely represented.

Results:

The results show that the best tensile properties at 400°F were obtained with CHR Compound 5908A (Cohrlastic HT Heat Stable) and CHR Compound 5901. The best elongation properties at 400°F were obtained with CHR Compound 5908A, Silastic 152 and Silastic 152 (600). The best tear strengths at 400°F were exhibited by CHR Compound 5908A and CHR Compound 5901.

The least reduction in tensile strength at 400°F, as compared with the room-temperature results, was exhibited by CHR Compound 5901 and Silastic 152 (600). The least reduction in elongation at 400°F was displayed by Silastic 152 (600), CHR Compound 5908A, and SE-100. The least reduction in tear strength at 400°F was exhibited by CHR Compounds 5901 and 5912.

TABLE II

TENSILE STRENGTH OF SILICONE RUBBERS AT ELEVATED TEMPERATURES

Compound	Room Temperature	212°F		400°F	
	Tensile Strength, psi	Tensile Strength, psi	Percent Change	Tensile Strength, psi	Percent Change
Connecticut Hard Rubber:					
5908A	>1540	980	-36	405	-74
R-11438	745	355	-52	180	-76
5910	770	415	-46	215	-72
5912	675	390	-42	195	-71
5904	510	395	-23	155	-70
5901	710	595	-16	390	-45
Dow Corning:					
6-128	640	445	-30	290	-55
152	580	455	-22	205	-65
152 (600)	470	405	-14	235	-50
675	625	450	-28	200	-68
6-526	650	420	-35	235	-64
250	495	265	-46	180	-64
General Electric:					
551A	490	385	-21	165	-66
750A	690	555	-20	140	-80
100	270	205	-24	125	-54

TABLE III

ELONGATION OF SILICONE RUBBERS AT ELEVATED TEMPERATURES

Compound	Room Temperature	212°F		400°F	
	Elongation, %	Elongation, %	Percent Change	Elongation, %	Percent Change
Connecticut Hard Rubber:					
5908A	>900	700	-22	510	-33
R-11438	340	235	-31	115	-66
5910	320	215	-33	100	-69
5912	270	220	-18	140	-48
5904	240	200	-17	95	-60
5901	100	15	-85	20	-80
Dow Corning:					
6-128	110	95	-14	40	-64
152	315	300	-5	160	-49
152 (600)	230	250	+9	160	-30
675	290	260	-10	105	-64
6-526	260	195	-25	110	-58
250	215	150	-30	130	-40
General Electric:					
551A	280	215	-23	105	-63
750A	185	190	+3	90	-51
100	125	100	-20	85	-32

TABLE IV

TEAR STRENGTH OF SILICONE RUBBERS AT ELEVATED TEMPERATURES

Compound	Room Temperature Tear Strength, lbs/in.	212°F Tear Strength, lbs/in.	212°F Percent Change	400°F Tear Strength, lbs/in.	400°F Percent Change
Connecticut Hard Rubber:					
5908A	206.0	115.0	-44	96.5	-53
R-11438	48.5	45.0	-7	27.5	-43
5910	56.0	58.0	+4	33.0	-41
5912	48.0	54.0	+12	32.0	-33
5904	48.5	37.0	-24	16.5	-66
5901	59.0	77.0	+31	48.0	-19
Dow Corning:					
6-128	32.5	31.5	-3	15.0	-54
152	50.5	46.0	-9	24.0	-52
152 (600)	60.5	48.5	-20	16.5	-73
675	81.5	67.0	-18	24.5	-70
6-526	43.0	41.0	-5	9.3	-78
250	63.5	42.5	-33	13.0	-80
General Electric:					
551A	37.0	34.5	-7	21.0	-43
750A	31.5	33.0	+5	12.5	-60
100	12.5	12.5	0	8.5	-32

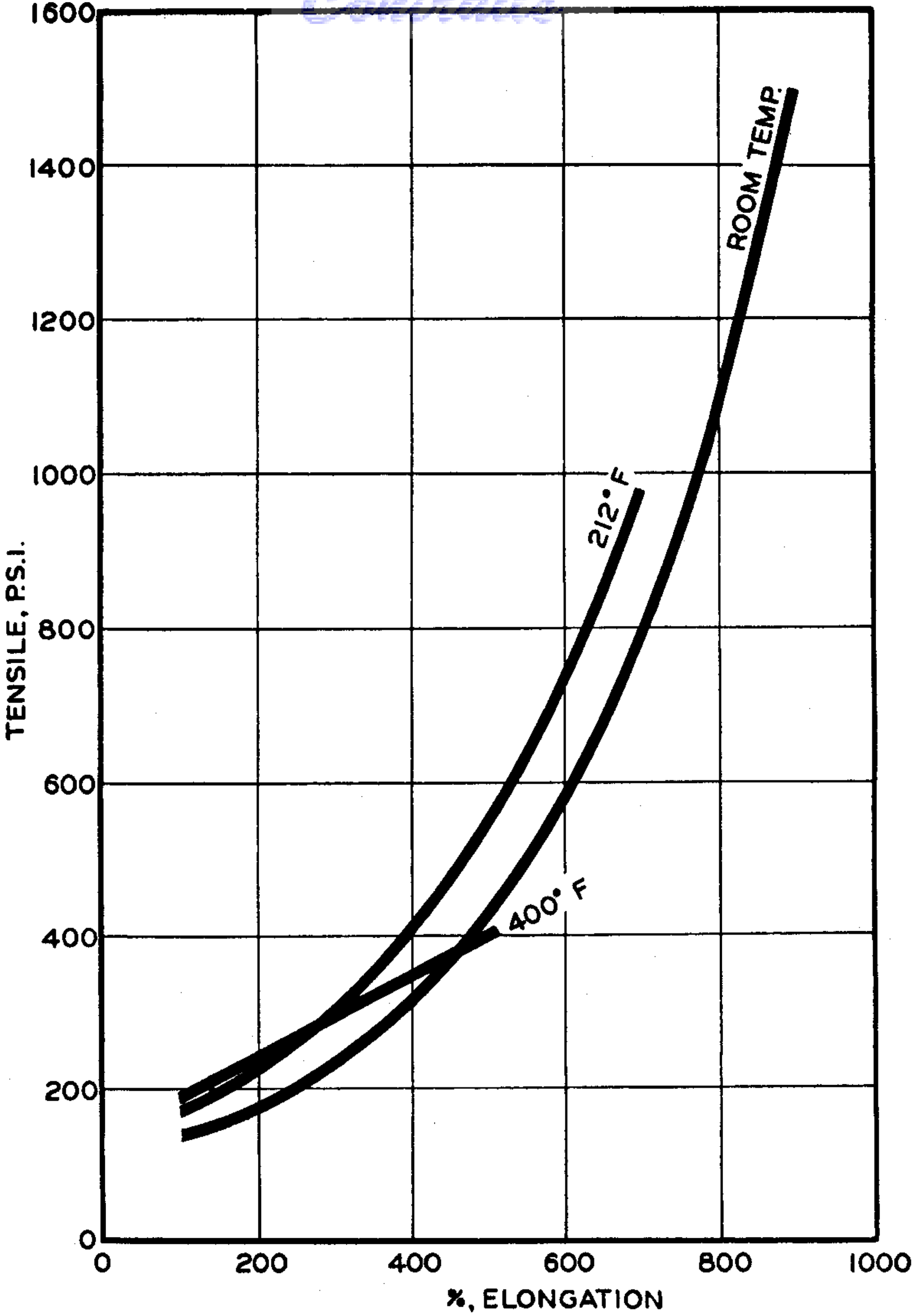

FIG. 4 : STRESS-STRAIN CURVES FOR CHR 5908 A AT VARIOUS TEMPERATURES.

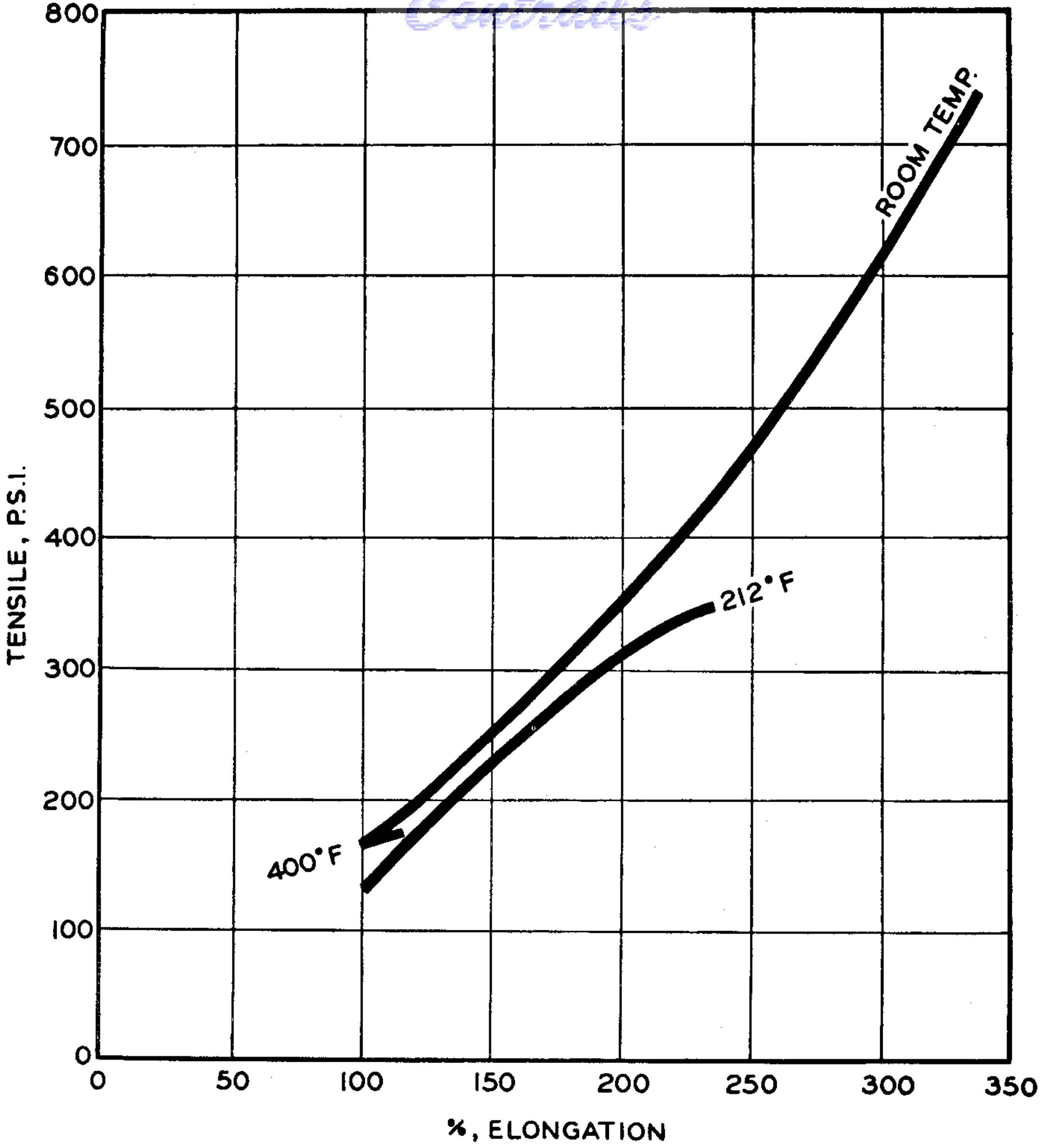

FIG. 5: STRESS-STRAIN CURVES FOR CHR R-11438 AT VARIOUS TEMPERATURES.

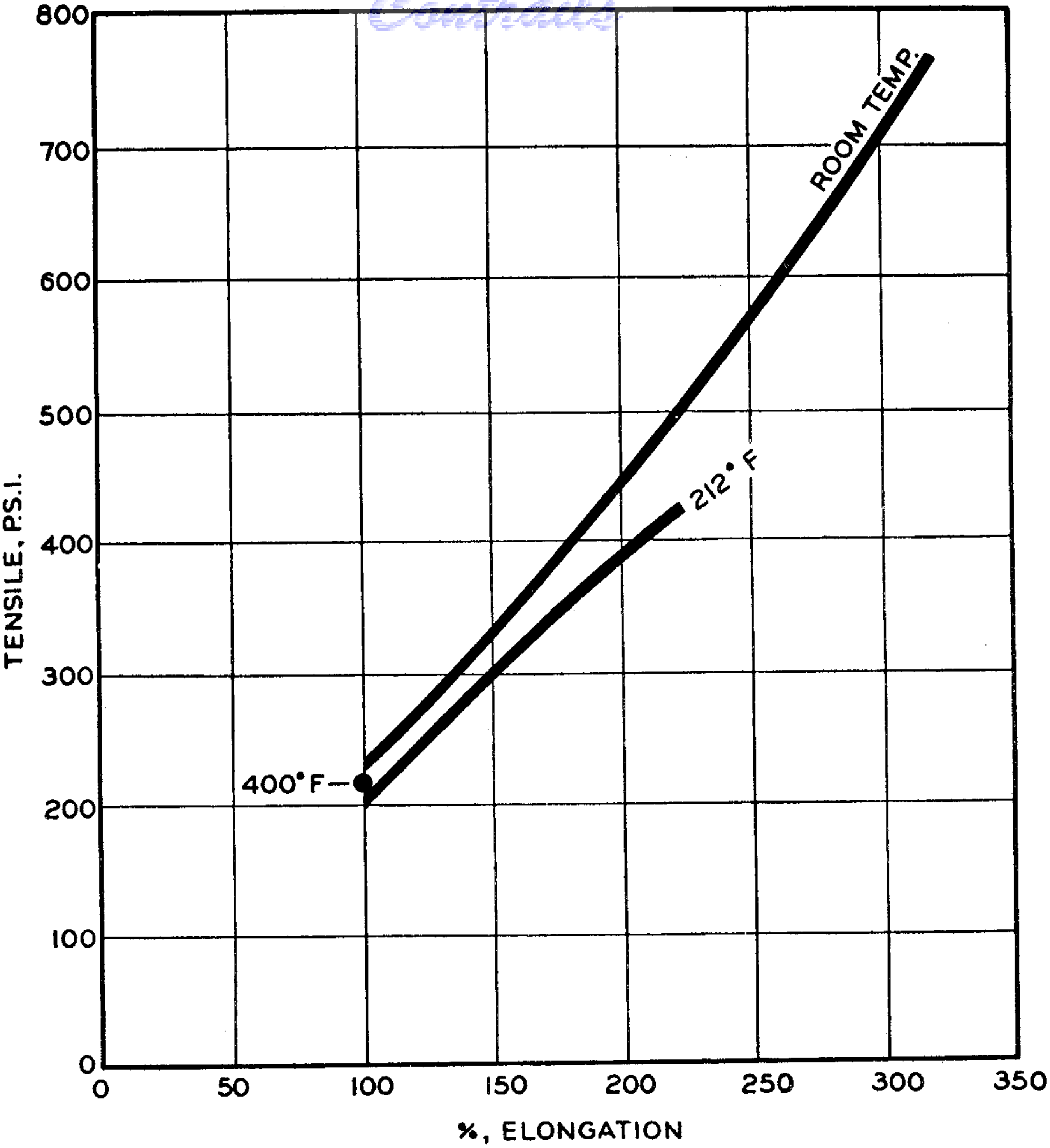

FIG.6: STRESS-STRAIN CURVES FOR CHR 5910 AT VARIOUS TEMPERATURES.

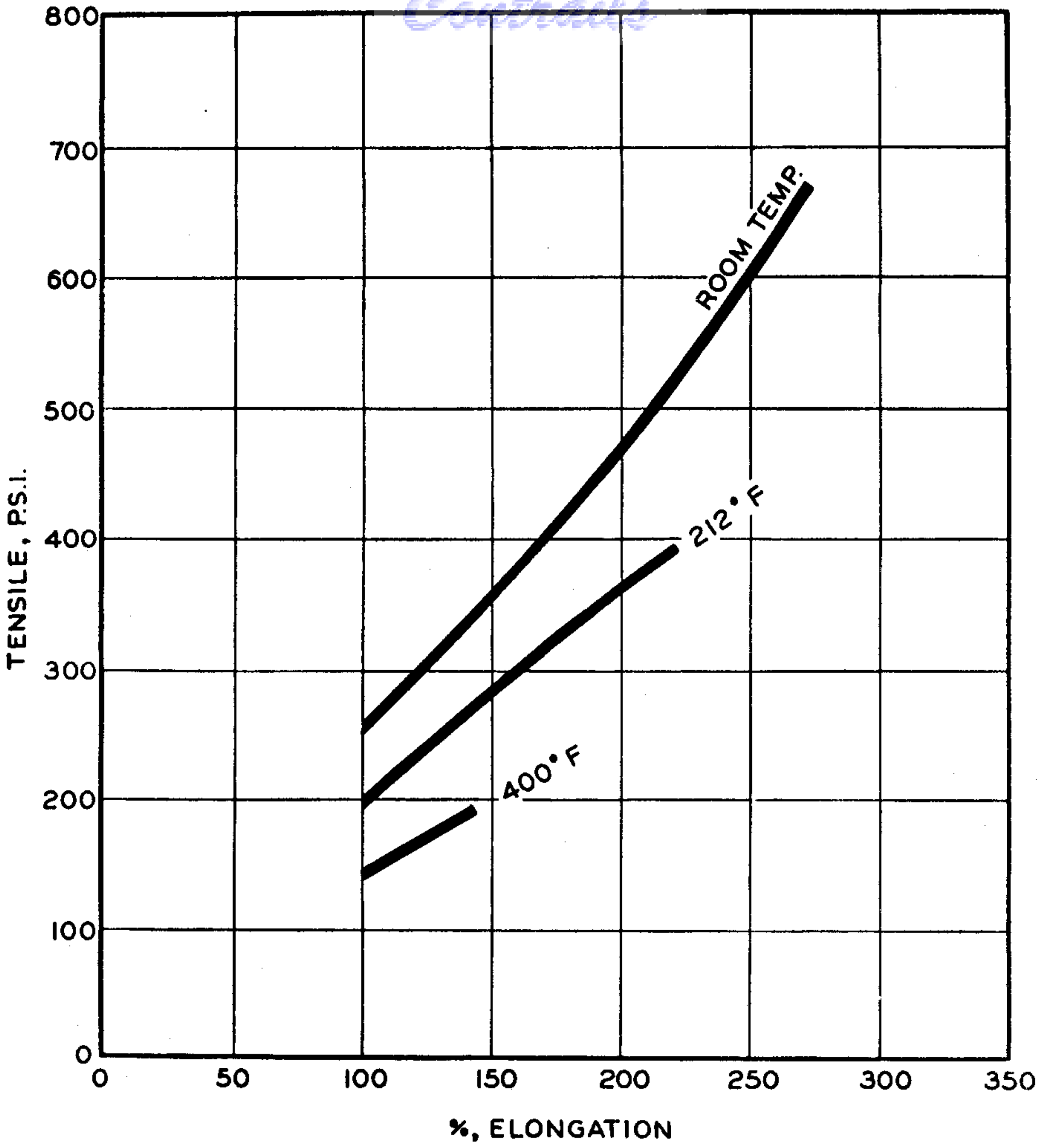

FIG. 7: STRESS-STRAIN CURVES FOR CHR 5912 AT VARIOUS TEMPERATURES

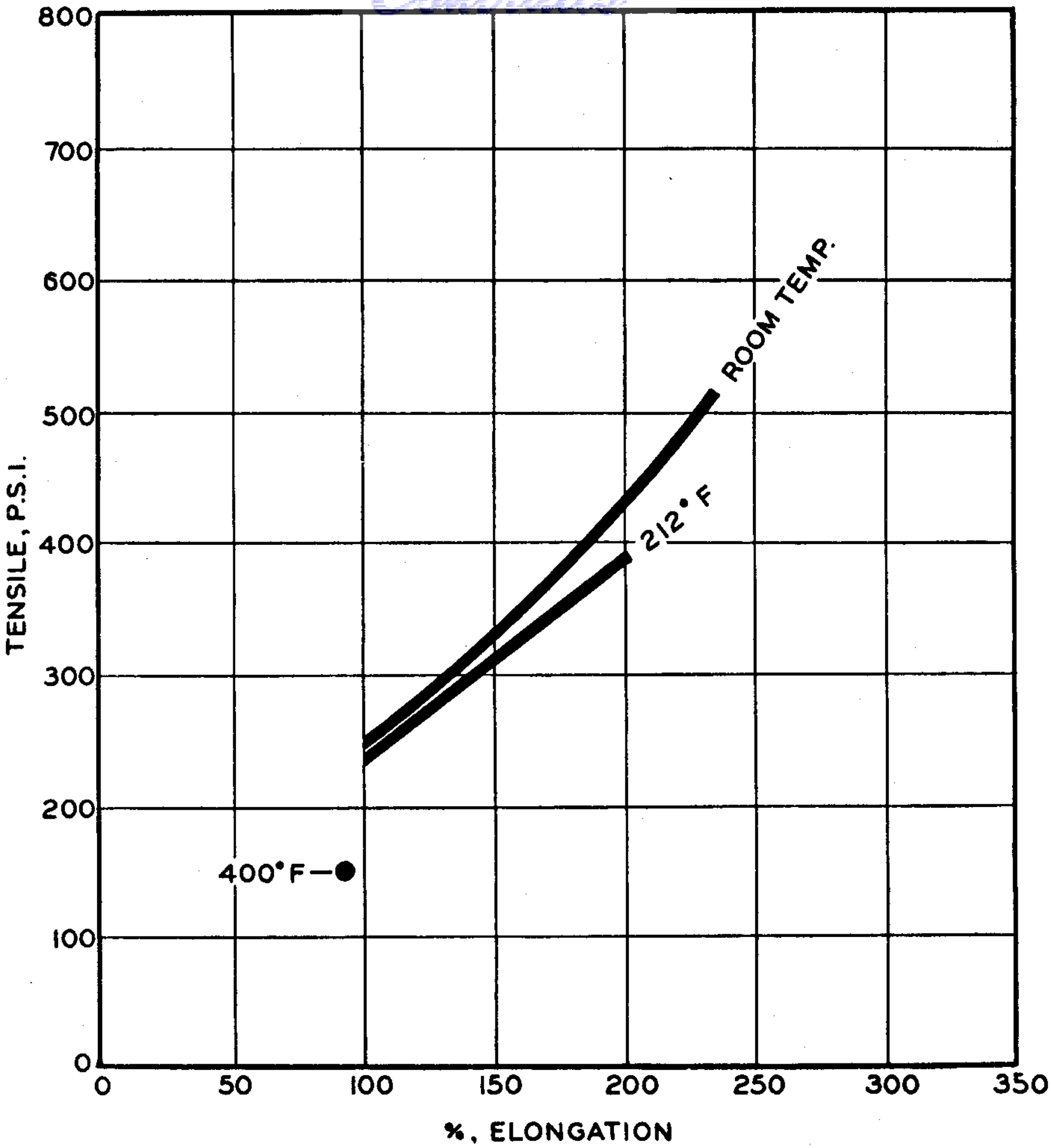

FIG. 8: STRESS-STRAIN CURVES FOR CHR 5904 AT VARIOUS TEMPERATURES.

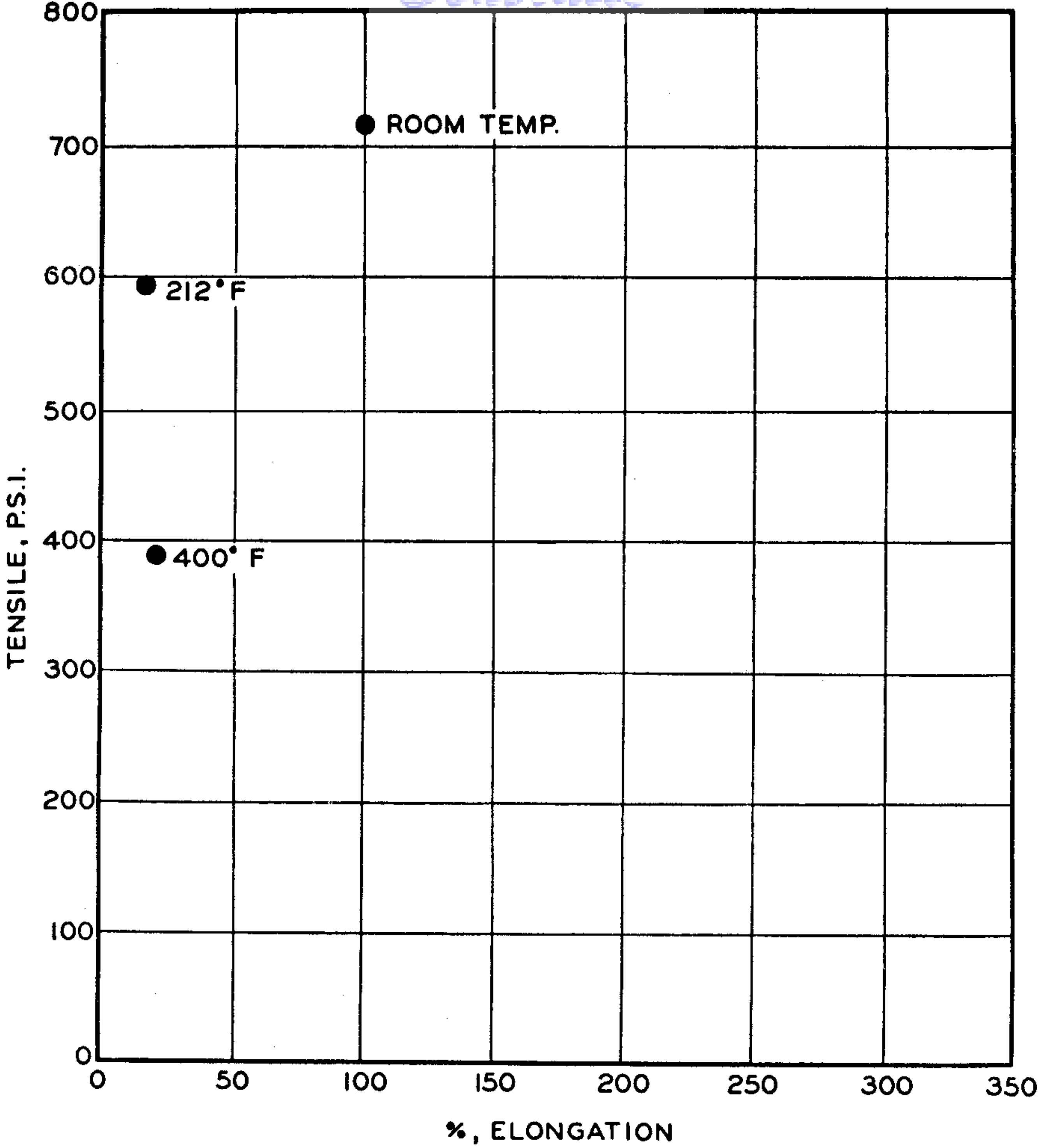

FIG. 9 : STRESS-STRAIN CURVES FOR CHR 5901 AT VARIOUS TEMPERATURES.

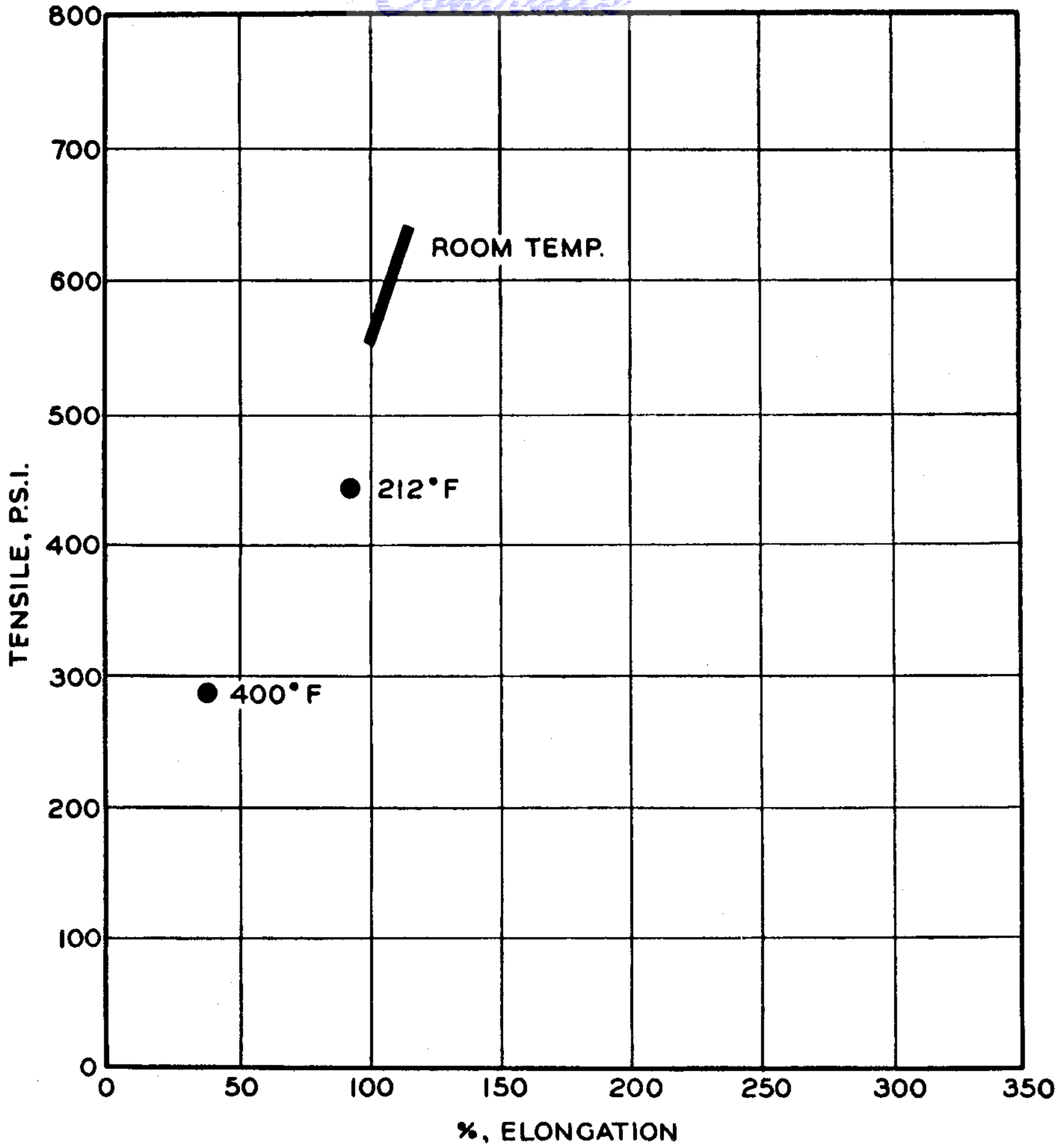

FIG. 10: STRESS-STRAIN CURVES FOR SILASTIC 6-128 AT VARIOUS TEMPERATURES.

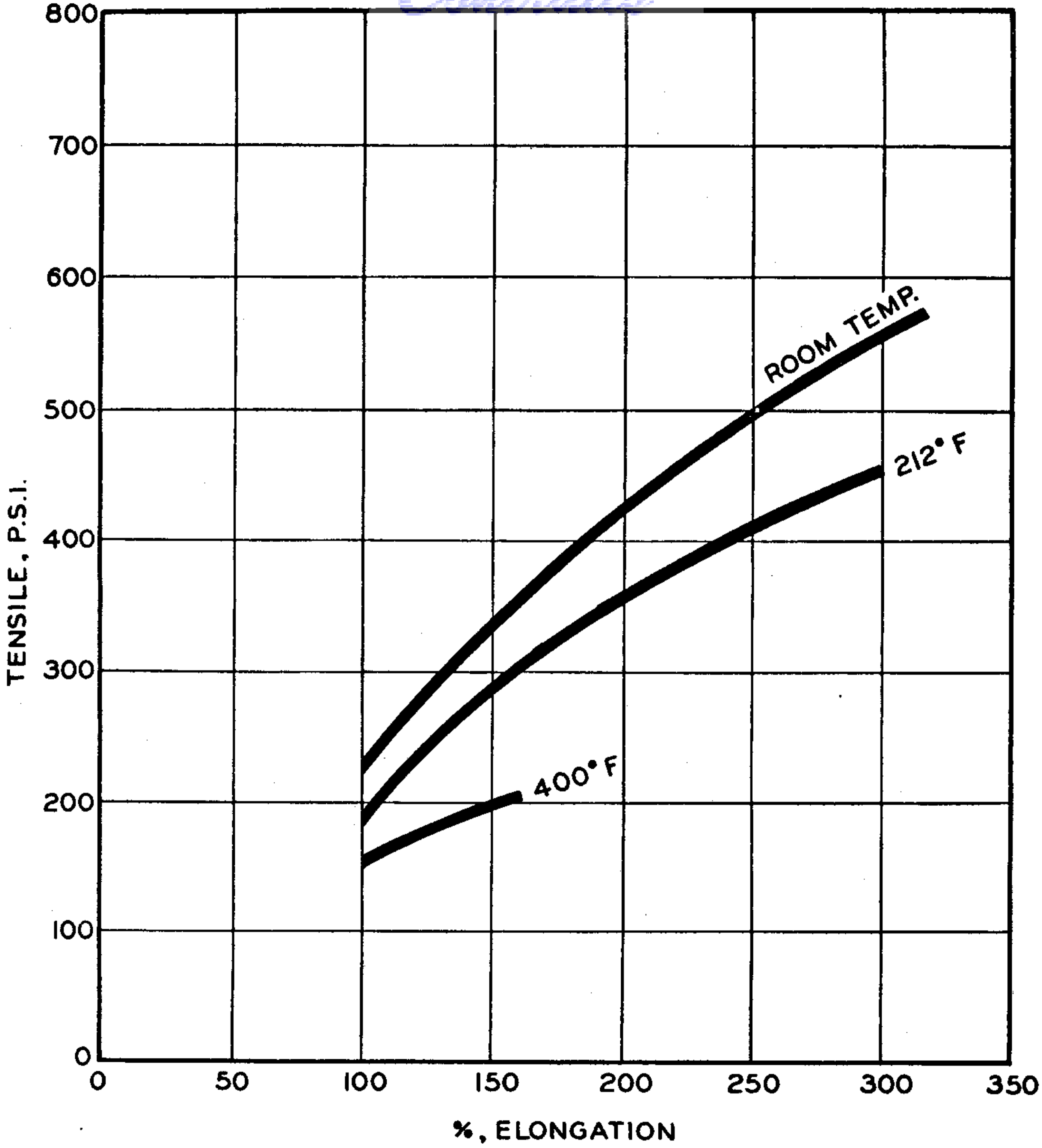

FIG. 11: STRESS-STRAIN CURVES FOR SILASTIC 152 AT VARIOUS TEMPERATURES.

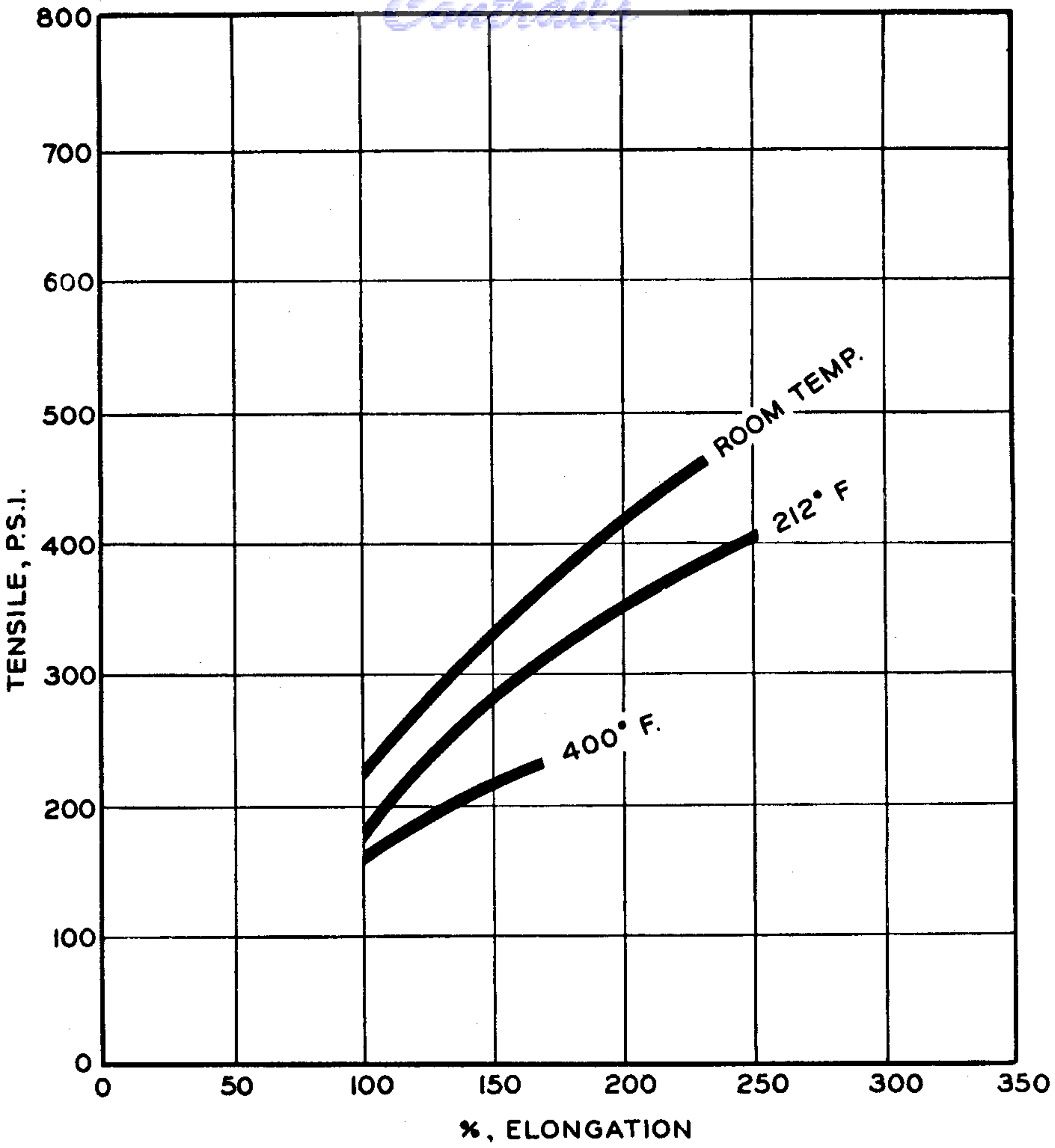

FIG. 12: STRESS-STRAIN CURVES AT VARIOUS TEMPERATURES FOR SILASTIC 152 AGED AT 600° F.

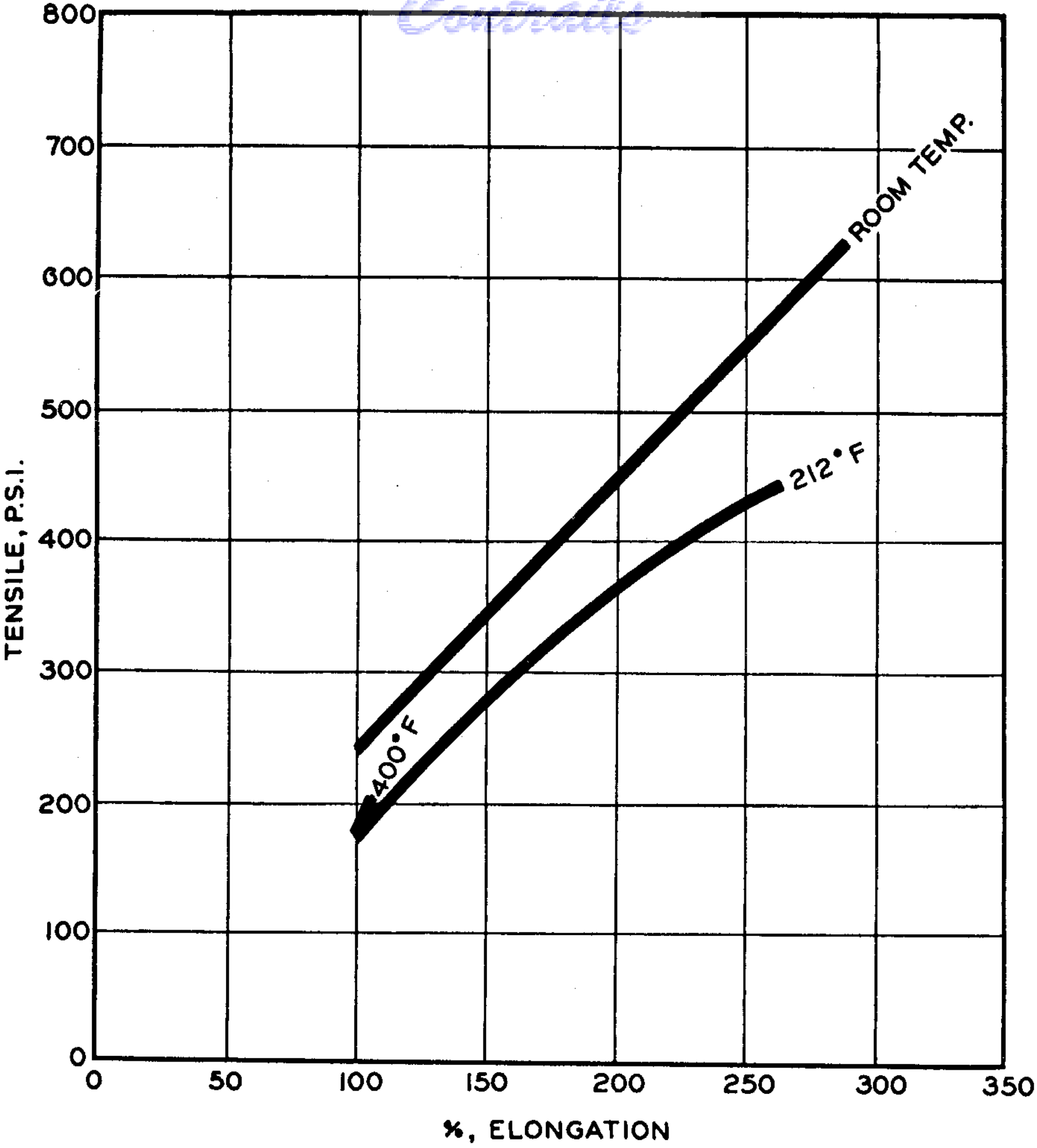

FIG. 13: STRESS-STRAIN CURVES FOR SILASTIC 675 AT VARIOUS TEMPERATURES.

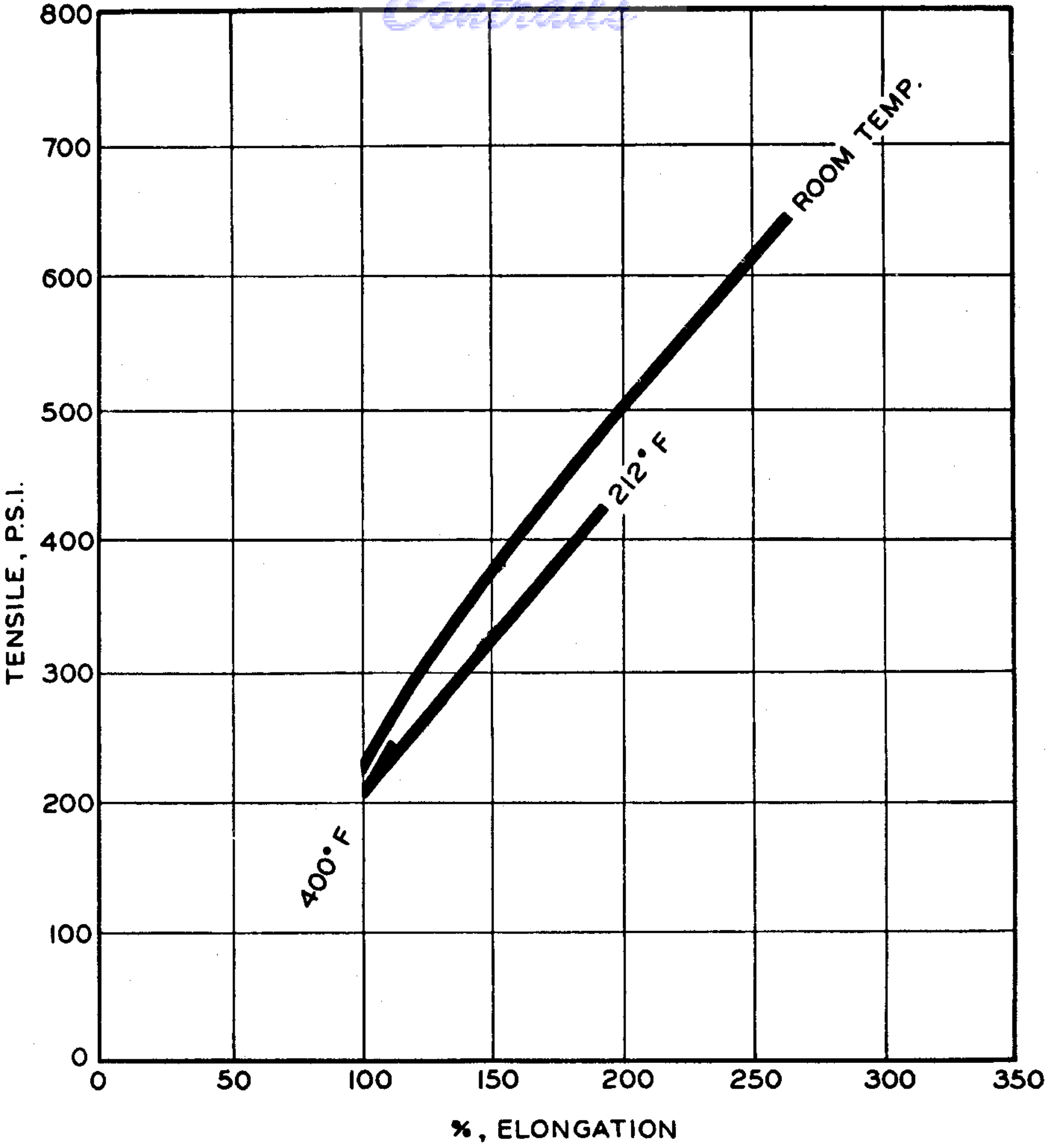

FIG. 14: STRESS-STRAIN CURVES FOR SILASTIC 6-526 AT VARIOUS TEMPERATURES.

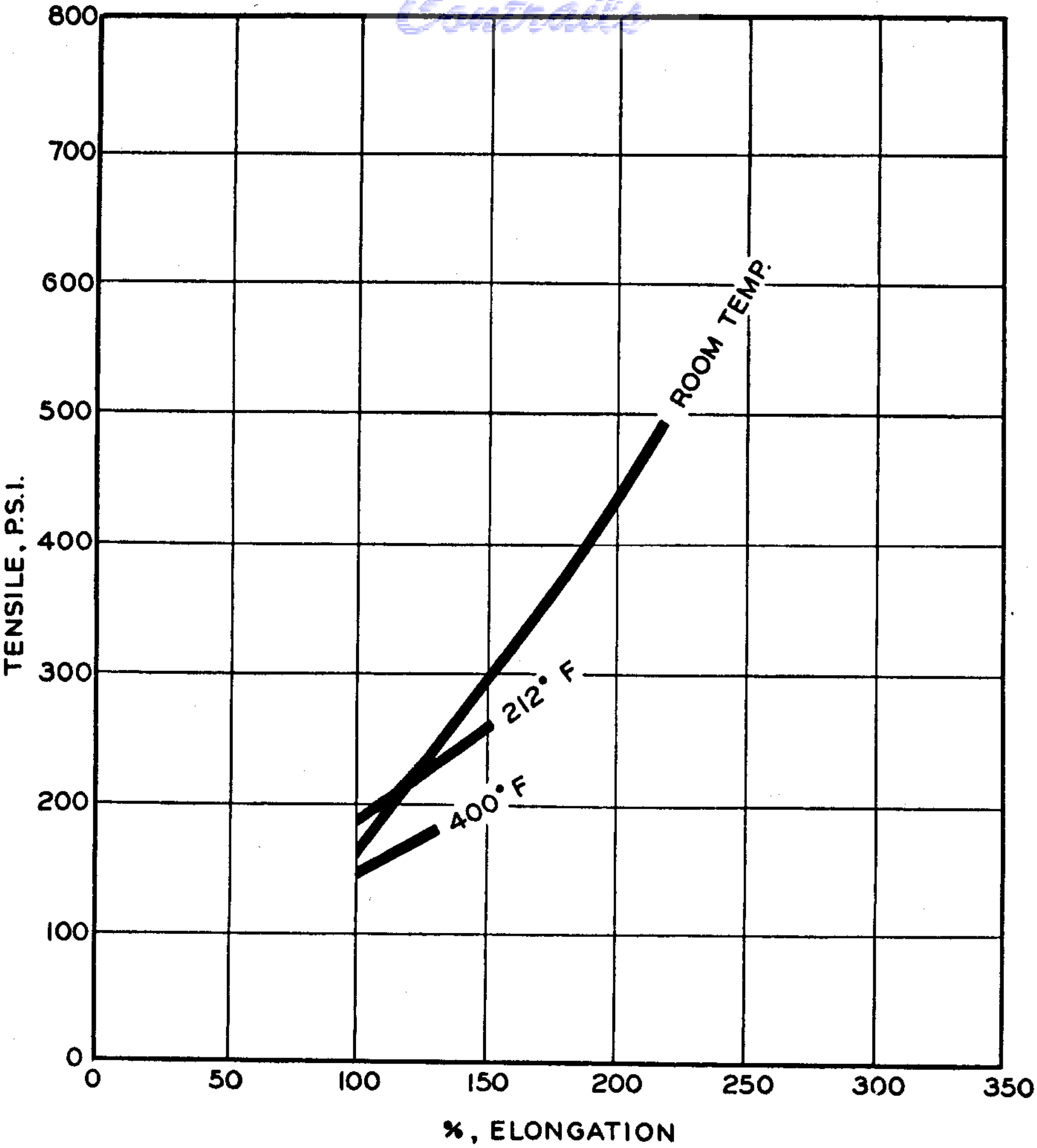

FIG. 15: STRESS-STRAIN CURVES FOR SILASTIC 250 AT VARIOUS TEMPERATURES.

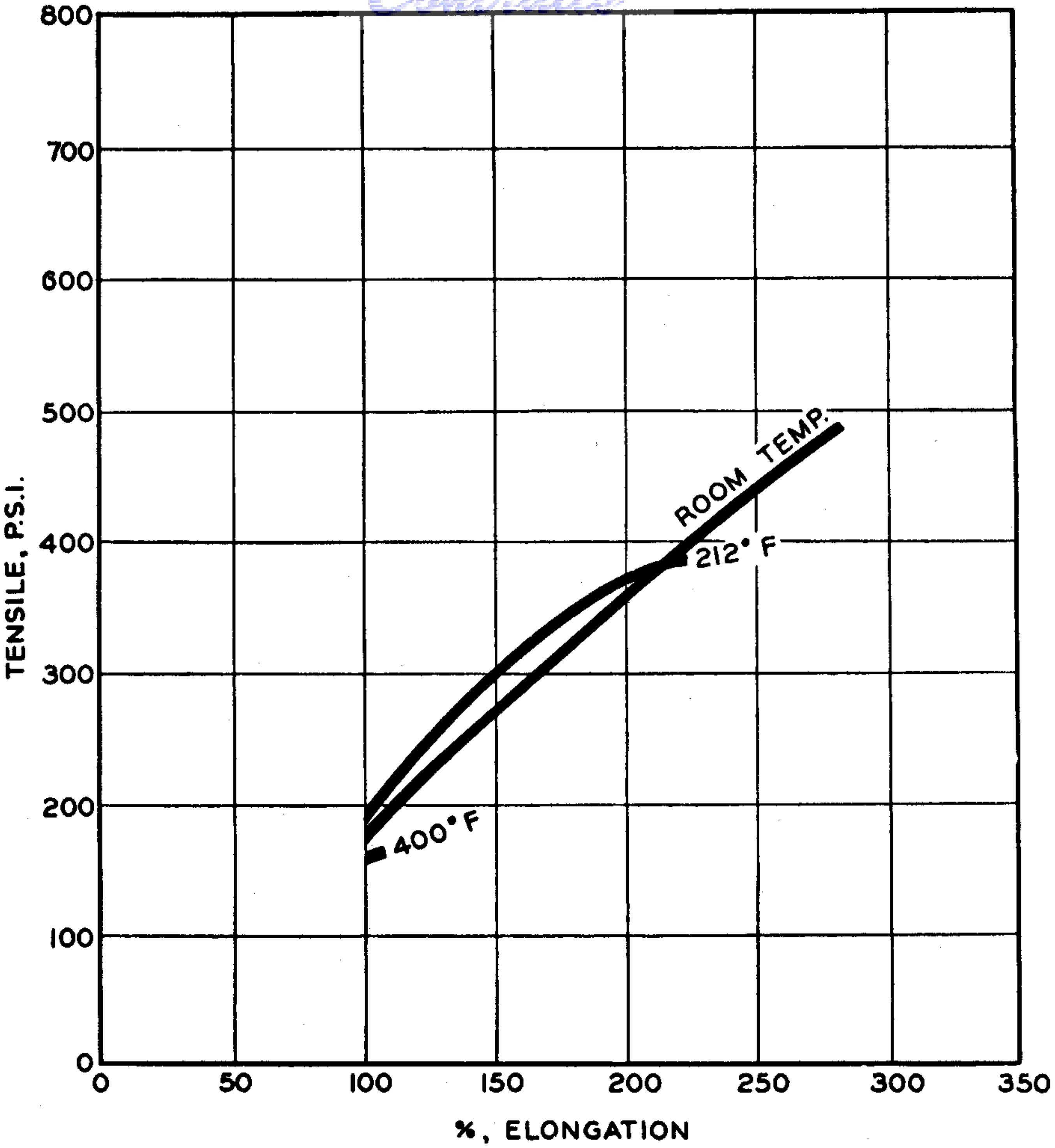

FIG. 16: STRESS-STRAIN CURVES FOR SE-551-A AT VARIOUS TEMPERATURES.

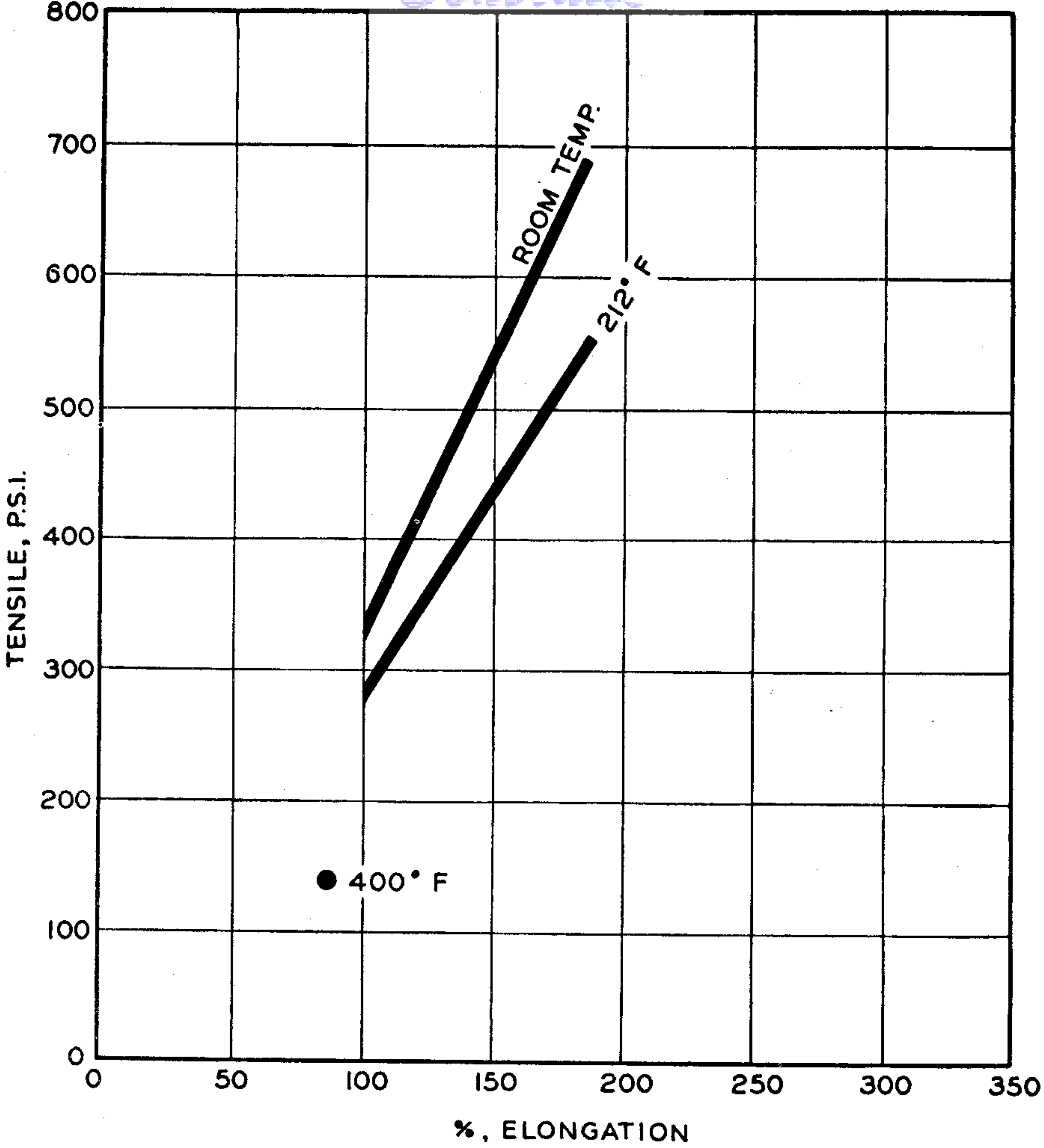

FIG. 17: STRESS-STRAIN CURVES FOR SE-750-A AT VARIOUS TEMPERATURES.

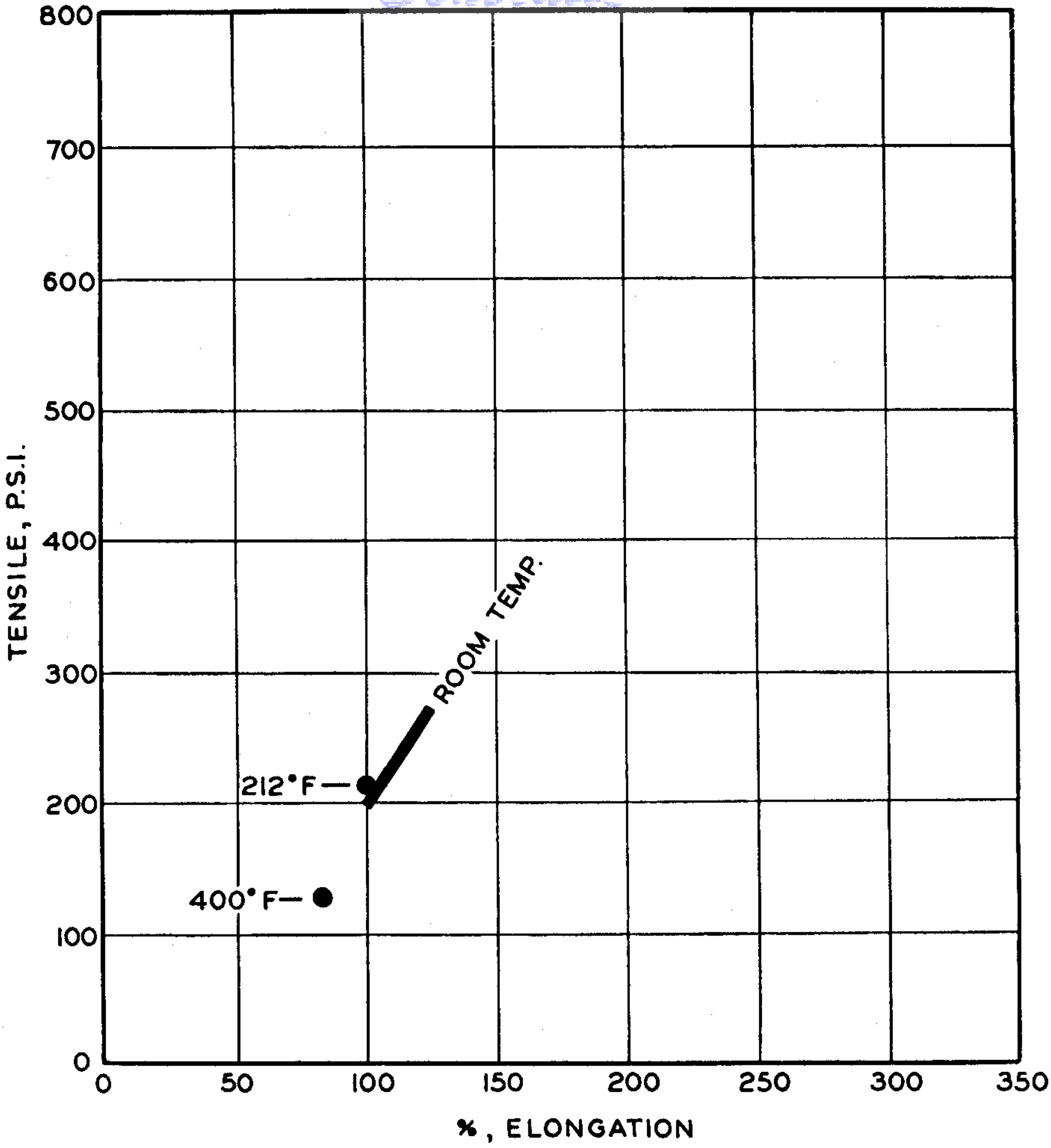

FIG. 18: STRESS-STRAIN CURVES FOR SE-100 AT VARIOUS TEMPERATURES.

II. DISCUSSION AND CONCLUSIONS

Design of a High-Temperature Test Apparatus

An apparatus for measuring tensile strength, tear strength and elongation of rubber compounds at elevated temperatures was designed and constructed. Emphasis was placed on simplicity of construction and on validity of comparative test results.

The test apparatus was built around a Scott Tensile Tester, Model L6. A permanent framework for the high-temperature apparatus was attached to the machine. The heated test box (made of Marinite) and the pulley system were designed as removable equipment, time and ease of assembly of these parts being an important consideration in their design.

The heating system in the box was planned so that heating would be done by convection. The heaters, consisting of resistance-wire wrapped around strips of Transite, were attached along the sides of the box. Another strip of Transite was placed beside each heater to encourage convection heating and to eliminate any radiation effect. Heating was controlled by varying the voltage applied to each heater, the amount of heat used depending on the temperature recorded by a thermocouple placed near the rubber sample. It was found expedient to use an auxiliary heater in the back of the box to increase the heat quickly. At an appropriate time, the auxiliary heater was turned off, the temperature then being controlled by the two heaters, and a series of samples were tested.

Although a great deal of effort was made to maintain uniform heat throughout the box, there still was a large temperature-gradient between the bottom of the box (where the test sample was clamped) and the top. At times, the differential was as much as 125°F. This would indicate that the temperature of a rubber test sample at break may also be a function of its elongation. The problem will be corrected in future work by installing a blower in the apparatus.

The pulley system of the apparatus was designed to integrate the testing of the rubber samples with the Scott Tester. That is, the test dumbbells were pulled in the box, while the readings were taken from the dial of the Scott Tester. This necessitated a 180-degree change in direction in the pulley system, which resulted in a dial reading of twice the actual force.

Calibration of the Test Apparatus

In order to determine the accuracy of measurements in the new apparatus, the physical properties of rubber samples were determined on the unmodified Scott Tester and in the unheated box (room temperature).

It was found that the physical properties measured in the box were usually greater than those measured on the unmodified Scott Tester. This positive difference indicates an additional force, most likely some friction in the pulley system. Friction could be caused (1) by the wire rubbing on the pulley wheels, (2) by the wire being slightly out of line, or (3) by rubbing of the wire at the aperture of the box.

The average deviation between the two methods of measurement was calculated for tensile strength, ultimate elongation and tear strength, these being 10.2 percent, 7.7 percent and 29.7 percent, respectively. However, a closer look at the results gives a slightly different picture. For example, in the case of tensile strength, the highest deviation was exhibited by SE-100 (a casting compound), this material displaying a low tensile strength of 205 psi. With such low tensile strength, it is not too difficult to conceive of a 31 percent deviation between different sheets of the same material. The same is true in the case of tear strength determinations, the four compounds of lowest tear strength showing the four highest deviations.

The agreement between the two methods was satisfactory enough to warrant the use of the test apparatus in determining comparative measurements at various temperatures. Tests were conducted at room temperature, 212°F and 400°F.

Physical Properties of Silicone Rubbers at Elevated Temperatures

Cohrlastic HT Heat Stable (CHR 5908A), a high-strength silicone stock containing Valron filler, exhibited the best physical properties at 212°F and 400°F. Its tensile strength at 400°F was 405 psi, its elongation, 510 percent, and its tear strength, 96 pounds per inch. These results are exceptional when compared with the rest of the data. Outstanding values were obtained in spite of the fact that the temperature at the top of the box was considerably higher than the desired test temperature. As previously mentioned, this effect would be of some disadvantage with rubbers having high elongation. The results obtained with Cohrlastic HT Heat Stable definitely indicate that the higher the room-temperature properties, the higher the properties will be at an elevated temperature.

Another compound which showed good heat-resistance was CHR-5901. This was made from Silastic 6-128 with 10 percent of Pyrex Glass Wool as an additional filler. The incorporation of glass wool into the silicone stock greatly improved physical properties at elevated temperatures, CHR-5901 showing the lowest percent change in tensile and tear strengths. As might be expected, the elongation of this compound was very low.

Other materials which showed good results were Silastic 152 and Silastic 152 (600). The difference in the two compounds becomes prominent in the 400°F tests, the 400°F temperatures having less deleterious effect on the postcured stock.

Conclusions

It is known that silicone rubber, in comparison with other rubbers, has by far the best resistance to high-temperature aging. From the information obtained in this research, however, it can be said that the properties of silicone rubber are seriously affected at the high temperature. Tensile strength, elongation and tear strength all decrease markedly at elevated temperatures. One clue appearing in the data to explain this poor behavior is a general tendency toward a decrease in modulus as temperature is increased. This may mean loosening of rubber-to-filler bonds, since the success in resisting aging would indicate the inherent stability of the vulcanized silicone molecule itself.

In Compound 5908A, a stock containing Valron, the modulus increases with increased temperature. Valron is known to produce strong filler-to-silicone polymer bonds, since peroxides can be omitted in Valron compounds (4). Insufficient time to accumulate more data prevents further speculation at this point on whether fillers such as Valron are to be preferred for use in silicone rubber for high-temperature service.

Several promising approaches for the improvement of properties of silicone rubber at high temperature are:

1. Use of a filler which shows the highest reinforcing qualities at room temperature; such as Valron.
2. Further investigation of the use of glass fibers or similar materials as fillers.
3. Study of highly loaded compounds. Inert fillers generally increase the elongation of silicone rubber as much as three or four times, and it would be imteresting to see if heavily loaded compounds might not cohere under tensile stresses better than lightly loaded recipes.

4. Evaluation of additives. The introduction of an additive might counteract the decreasing modulus at higher test temperatures, experienced with the conventional silicone compounds. Valron, itself, might serve as such as additive.

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