TIME-TEMPERATURE-STRAIN RATE EQUIVALENCE FOR VARIOUS ENGINEERING THERMOPLASTICS

JEROME J. LOHR, Vehicle Environment Division,
National Aeronautics and Space Administration,
Ames Research Center, Moffett Field, California

SYNOPSIS

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The mechanical behavior of an acetal, a polycarbonate, a polyamide, and a fluorocarbon has been studied over the temperature range from -100 to +170°C. Stress relaxation measurements were made over a time period from 0.1 to 1000 sec. and at several levels of strain. Tensile tests were run at strain rates from 0.03 to 30 in./in./min. The concept of the yield stress master curve previously developed for linear amorphous polymeric materials is shown to also apply to these crystalline materials. The materials all exhibit nonlinear behavior as demonstrated by changes in the stress relaxation modulus with increased applied strain. The stress relaxation master curve is fitted with a generalized Maxwell model and integrated to synthesize a stress-strain curve for a linear material loaded at a constant strain rate. The comparison of these predicted stress-strain curves with experimental curves indicates that the polyacetal, polyamide, and polycarbonate may be treated as linear materials up to approximately 2% strain. The fluorocarbon, however, is nonlinear at the lowest measured strains. Author

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INTRODUCTION

An effective strength analysis of plastic materials must consist of a stress analysis with a superposed yield or failure criterion. The strength analysis must include the time- and temperature-dependent mechanical behavior of the plastics in both the stress analysis and the yield or failure criterion. The stress relaxation master curve concept, which includes this time- and temperature-dependent behavior of viscoelastic materials, is now quite well developed.[1,2] In a previous paper[3] a yield stress master curve similar in principle to the stress relaxation master curve was shown to apply to essentially amorphous polymers tested in their glassy temperature range. In the present paper the linearity of mechanical behavior and the applicability of the yield stress master curve concept will be investigated for four crystalline-type polymers, a polyamide (Nylon), a polycarbonate (Lexan), a polyacetal (Delrin), and a fluorocarbon (Teflon).

Linearity, as applied to viscoelastic materials, such as polymers, means that the stress within the material (due to an externally applied stress or strain) is assumed to be a linear function of the strain and strain rate parameters. In other words, the ratio of stress to strain may be a function of time and temperature but not of strain or strain rate. Consequently, measurement of the time-dependent relaxation modulus, $E_r(t)$, defined as the ratio of the time-dependent stress, $\sigma(t)$, to a rapidly applied constant strain, ϵ_O, becomes a straightforward means of verifying the linearity of a material since, for a linear material, the variation of $E_r(t)$ should be the same for various values of the

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strain, ϵ_o. However, in an ordinary tensile test of a viscoelastic material, departure from a constant ratio between stress and strain is not necessarily an indication of nonlinear behavior. This is because time-dependent stress relaxations may also be taking place during the tensile test.

Some knowledge of the linearity displayed by glassy polymers subjected to high stresses would also be important in formulating a theory to explain the yield stress master curve concept. In the temperature range above the glass transition temperature of polymeric materials, the theory of rubberlike elasticity[4] specifies that the relation between force and extension is nonlinear. Smith[5] has derived functional forms of both the strain-dependent and time-dependent terms in the rubbery region. No valid general treatments are known to exist, however, for materials in the glassy state. Some specific data,[6] in the form of values of $E_r(t)$ at various values of ϵ_o for textile filaments, do exist but because of the differences in structure and geometry between filaments and the sheet stock of interest here, direct comparisons probably cannot be made. Other existing data which might indicate linearity are similar to that of Marin et al.,[7] on poly(methyl methacrylate) and polystyrene; however, these data were derived from creep tests which were run before the time-temperature superposition principle was understood and so are lacking in generality.

Although proof exists that both the stress relaxation[1] and yield stress[3] master curve concepts apply to linear amorphous polymers such as poly(methyl methacrylate), polystyrene and polyvinyl chloride, crystalline polymers have received little attention. However, there seems to be agreement[1,2] that nonlinearity occurs at lower strains in crystalline polymers than in amorphous polymers. The essentially crystalline polymers chosen for study herein have found many applications as engineering materials where their mechanical integrity, as influenced by their environment, has been determined empirically or from experience. In the present experiments, stress relaxation and tensile tests have been made to provide data which could be used in a strength analysis to predict mechanical integrity or for use in formulating a theory to explain the parameter dependence of mechanical behavior. Stress relaxation master curves have been constructed from tests made at various strains to determine the linearity, as measured by $E_r(t)$, of the materials tested. Yield stress master curves for these materials also have been constructed for investigating the applicability of the concept and its stress and temperature limits. The stress relaxation master curves then have been included in an expression formulated in the analysis section of this paper for predicting the shape of the stress-strain curves based on the assumption that the materials behave linearly. These predicted stress-strain curves thus have included the effects of the time-dependent stress relaxation occurring during the tensile test and can be compared with the experimental stress-strain curves to give a continuous indication of the nonlinearity exhibited by the materials in the constant-strain-rate tensile test.

ANALYSIS

The stress, σ, at constant strain rate, $\dot{\epsilon}$, for a material with a linear viscoelastic response and time-dependent relaxation modulus, $E_r(t)$, may be found from the expression[2]

$$\sigma = \dot{\epsilon} \int_0^t E_r(z)\,dz \qquad (1)$$

where t is time and z is the variable of integration. To perform the integration, an approximate form of $E_r(t)$ may be taken of the type

$$E_r(t) = E_o + \sum_{i=1}^{N} E_i e^{-t/\tau_i} \qquad (2)$$

where E_o is the rubbery or long time modulus and τ_i is the relaxation time for the modulus increment E_i. In practice, E_i's were taken as constant, ΔE, for a given polymer; thus, the expression becomes

$$E_r(t) = E_o + \Delta E \sum_{i=1}^{N} e^{-t/\tau_i} \qquad (3)$$

In terms of spring and dashpot models, eq. (3) corresponds to a spring of stiffness, E_o, in parallel with N Maxwell elements, each with relaxation time τ_i.

Equation (3) may be substituted into (1) and $\dot{\epsilon}$ replaced with ϵ/t. The resultant computation provides σ as a function of ϵ.

It should be pointed out that this is not an attempt to fit the experimental stress-strain curve as was done in the development of

Knowles and Dietz.[8] A fit of the stress-strain curve would require the addition of nonlinear terms probably in the form of powers of the strain.[8] The present development is intended only to separate the time and nonlinear effects in a stress-strain curve of a viscoelastic material.

MATERIALS AND EXPERIMENTAL EQUIPMENT

The materials used were commercial sheet stock nominally 1/32 in. thick. The polyamide was Dupont Nylon type 101, the polyacetal was Dupont Delrin type 150, the fluorocarbon was Dupont virgin Teflon, and the polycarbonate was General Electric Lexan. The first three materials were opaque, whereas the Lexan was clear. For the Lexan, a comparison of the yield stress at similar temperatures and strain rates with that given by Robertson[9] would indicate that the molecular orientation or crystallinity of the Lexan used herein was low. This is, of course, consistent with the observed optical clarity of this material.

The stress relaxation was measured with the instrument shown in Figure 1. Tests were run for as long as 1000 sec. at various constant temperatures from -50 to +150°C. The specimen is elongated a known and reproducible amount by an essentially instantaneous stroke of the linear actuator and the load is thereafter recorded as a function of time. The instrument was designed for high stiffness, rapid loading, and reproducible displacements. The plunger, which is linked to the wedges, could be activated either electrically or mechanically so that the entire loading process was completed in not over 0.04 sec. Data were taken starting at 0.4 sec. after the initial load application.

Standard ASTM D 638 tensile specimens were used for both stress relaxation and tensile tests. In both cases the distance between grip faces was set at 3.5 in. The elongation of the specimen was measured with a Schaevitz linear variable differential transformer (LVDT) and demodulator. Displacements were measured to an accuracy of 0.0005 in. and were previously[3] found to be reproducible within ±0.0002 in.

The strain in the specimen was calculated by dividing the measured displacement by the 3.5-in. gage length. Different total displacements were attained by varying the position of the wedge stops or changing to a second pair of matched wedges which have a different slope. The Kulite Bytrex JP-100 semiconductor load cell was excited by a Harrison Lab 6226 A power supply. The load cell output was recorded on a Texas Instruments Oscilloriter. The instrument shown in Figure 1 was mounted on the door panel of a Statham SD-8 air bath temperature control chamber which maintained temperature within at least ±1/2°C. for the duration of the test. Liquid nitrogen was used for cooling, but tests at temperatures below -25 to -50°C. were not run because of problems caused by frost buildup, and some stickiness in the loading wedges which prevented attainment of reproducible displacements.

Tensile tests were run on a Plas-Tech model 591 tensile test machine at nominal displacement rates of 0.1, 1, 10, and 100 in./min. and at various constant temperatures from -100 to +170°C. After the test temperature was attained, it was held for 10 min. to allow the sample temperature to equilibrate. The temperature for each test was maintained within ±1°C. by a Missimer furnace modified to use a Research Inc. Thermac temperature control system. Cooling was provided by liquid nitrogen. Heating was provided by quartz infrared lamps. Temperature control below ambient was achieved by bucking the Thermac system against a more than adequate constant supply of liquid nitrogen. Thus, at all temperature levels, the temperature was actually controlled by the heat input from the Thermac system.

Loads for both the tensile and stress relaxation tests were calibrated with dead weights to be accurate within ±1% of the measured load. Displacements for both tests were calibrated with a micrometer to within 0.0005 in. However, displacements could not be read this accurately for oscilloscope traces in the tensile tests. Therefore, a Moseley 135 X-YY (dual pen) recorder was used to obtain load-time and displacement-time curves for the three lower nominal strain rates. A typical test record is shown in Figure 2. The displacement-time plot gives an indication of the typically high degree of linearity of displacement as a function of time that was noted in all tests. The slope of the displacement-time curve for each specimen was divided by the 3.5-in. gage length to determine the strain rate.

RESULTS AND DISCUSSION

Stress Relaxation Master Curves

Stress relaxation master curves for the four materials at various initial strains are shown in Figures 3-6. The stress relaxation shift factors used to construct each of these curves are shown in Table I. A typical cycle consisted in setting a temperature, allowing the sample to equilibrate for 10 min. after the set temperature was attained, applying a preload of approximately 3 to 4% of the load when extended, straining the sample to a preset displacement, and, finally, allowing the specimen to relax for approximately 15 min. A cycle thus required about 30 min. The measured load was corrected for the effect of the preload. For instance, if the preload were 3 lb. and the initial measured load were 75 lb., each subsequent measured load in the relaxation cycle would be decreased by 4%. The relaxation modulus was then calculated by dividing the corrected load by the product of the initial cross-sectional area and strain. Superposition of the shifted data was generally within ±3%. The stress relaxation shift factors shown in Table I were those measured to effect superposition of the data. (It should be recognized, however, that these factors probably are not reproducible to the three significant figures given in Table I because of possible variations in sample composition, temperature, or displacement.) Although the possibility of changes in crystallinity of these materials as discussed by Tobolsky[6] is admitted, no measurements of changes in crystallinity were made. However, a stress relaxation test on a sample of Nylon taken directly from ambient temperature to 50°C. was compared with the test on a sample started at -25°C. and increased to 50°C. through four temperature steps. The consequent differences in stress relaxation moduli observed at 50°C. corresponded approximately to the change in moduli which would be produced by a 3°C. shift in test temperature, indicating that any differences in crystallinity were small.

The stress relaxation master curves shown in Figures 3-6 have been referenced at the temperatures shown to indicate most clearly the true change in shape and position of the master curves with increasing applied strain, ϵ_0. All of the materials show a measurable nonlinearity, even at the lowest strain level used. This would indicate that if these materials were tested in stress relaxation at even lower strains, a higher value of the modulus would be found for each relaxation time. Thus, from the curves shown in Figures 3-6, it is not possible to say where the behavior departs from linearity. The magnitude of the decrease in modulus as a function of increasing strain can be important from a rheological standpoint. However, with the exception of Teflon, the deviation from linearity in the upper third of each set of master curves is of the order of a 10% change in modulus with each 1% increase in strain. This change, when compared with the three decade change in modulus between the glassy and rubbery regions,[6] is relatively small. On the other hand, from the standpoint of strength analysis of viscoelastic materials, these deviations may be appreciable. For combinations of time and temperature which produce a relaxation modulus in the transition region (approximately below 150,000 psi), the change in modulus is quite appreciable. For Teflon, the deviation is significant even at strains on the order of 1%. Nonlinearity effects thus would be expected to be obvious at lower strains in tensile tests of Teflon than in tensile tests for the other materials. The implications of the nonlinearity demonstrated by the stress relaxation tests will be discussed further in the next two sections.

Yield Stress Master Curves

Yield stress master curves for the four materials are shown in Figures 7-10. With the exception of Lexan, the definition of the load used for calculating yield stress may be seen in Figure 2. That is, the yield stress was calculated from the load at which a line drawn from 2% strain parallel to the initial slope of the load-time curve intersects the curve. The Lexan exhibited a definite yield load over the entire test temperature range; therefore, in this case, yield load was used in the calculation of its yield stress. In all cases, yield loads were divided by the initial cross-sectional area to give yield stress. The yield stresses determined at the various temperatures and strain rates were plotted as a function of strain rate. For each material, the four yield stresses determined at a temperature were shifted laterally to construct yield stress master curves as discussed previously for amorphous polymers.[3] The shift factors used in constructing the master curves are shown in Table II.

The major portion of each master curve shown in Figures 7-10 has been approximated by a straight line which can be described by an equation of the type[3]

$$\sigma_y = K_1 + K_2 \, ln \left(\frac{\dot{\epsilon}}{\dot{\epsilon}_o} a_T \right) \tag{4}$$

where σ is the yield stress in psi, $\dot{\epsilon}$ is the strain rate in in./in./min., and a_T is the temperature shift factor; $\dot{\epsilon}_o$ equals 1 in./in./min., K_1 and K_2 are constants which depend on the reference temperature chosen and the slope of the master curve, respectively. The values of K_1 and K_2 and limits of applicability of eq. (4) are shown in Table III. Equation (4) can be used to predict the temperature and strain rate dependence of yield stresses over a wide range of temperatures or strain rates. The strain rate dependence of the four materials at 25°C. may be seen directly from Figures 7-10. Thus, the yield stress master curve concept can be used for predicting behavior at both high and low strain rates which may not be readily available in the laboratory, just as the stress relaxation master curve allows prediction of the modulus, E, at very long or short times.

In their higher temperature ranges, Nylon (Fig. 7) and Teflon (Fig. 9) deviate from the straight-line approximation in a manner similar to that shown previously for Mylar.[3] This temperature region, in general, lies above the glass transition temperatures of these three materials, but beneath their crystalline melting temperatures; thus, some change in slope might be expected. Because of the equivalence between time and temperature embodied in the master curve concept, this behavior is then observed at either very low strain rates or at high temperatures. The change in slope in the upper portion of the master curve for Delrin (Fig. 8) in the temperature range below -50°C. is not understood at this time.

The a_T shift factors have been referenced so as to have a value of 1 at their glass transition temperatures and are shown in Figure 11. The values of the glass transition temperatures for Lexan, Nylon, and Teflon shown in Table I were taken from Boyer.[10] Boyer gives three values, -97, -50, and +127°C., for glass transition temperatures of Teflon. However, a comparison of the absolute value of the yield stress of Teflon with that of the other materials indicates that the transition at -50°C. should be used as the reference temperature for the present work. This definition is similar to the definition of the glass transition temperature suggested by Tobolsky et al.[11] The value of 170°C.

used for Delrin is its published heat distortion temperature[12] and should be approximately equal to its glass temperature. The data shown in Figure 11 indicate that the temperature dependence of the yield stress shift factors for the various materials in their glassy state is very similar. The solid line is a best fit curve for PMMA, PVC, and Mylar from reference 3. There are not sufficient points below the reference temperature for Teflon to draw definite conclusions about the temperature dependence of its a_T shift factors. However, the similar behavior shown by six different polymers, each of which is from a different family of thermoplastics, encourages one to think that a general theory for describing the temperature-dependent behavior of polymers in the glassy state might be developed. The data shown in Figure 11 indicate that such a theory should be related to the glass transition temperature of each material, just as the WLF equation[13] which holds in the rubbery region is related to the glass transition temperature.

As also noted for amorphous materials,[3] the incremental a_T shift factors for stress relaxation and yield stress of these materials compare favorably in the temperature range below the glass transition temperature. These incremental shift factors can be determined from Tables I and II.

Comparison of Predicted and Experimental Stress-Strain Curves

Stress-strain curves computed from eq. (1), in combination with eq. (3), are compared in Figures 12 and 13 with the experimentally determined stress-strain curves. The curves shown in Figure 12 are from data for Delrin but are typical of the behavior of Nylon and Lexan as well. Figure 13 shows typical curves for Teflon which exhibited behavior quite different from that of the other three materials.

The master curves measured at the lowest strain levels (0.65 or 0.71%) shown in Figures 3-6 were used for determining the parameters in eq. (3). As pointed out earlier, these master curves may include some effects of nonlinearity but for this computation they are an adequate approximation to the zero-strain relaxation modulus. The curve-fitting procedure then consisted in (using Delrin as an example) determining approximately a long-time modulus (E_o = 75,000 psi for Delrin) and assuming an incremental modulus (ΔE = 25,000 psi for Delrin). To cover the range of interest of the modulus, a value of $N = 14$ was required. The relaxation times, τ_i, were picked as the times when the modulus had the value $E_o + (i - 1)\Delta E + 0.368\ \Delta E$ on the stress relaxation master curve. This value was used since at this intersection $t = \tau_i$; therefore, $e^{-t/\tau_i} = 0.368$. The τ_i for temperatures other than the reference temperatures shown in Figures 3-6 were measured by shifting the master curves laterally an amount, $\Delta \log a_T$, given in Table I for the temperature difference between the reference and test temperatures.

In Figure 12, the close comparison up to at least 2% strain between the predicted and experimental curves for the 150°C. test was typical for Delrin, Nylon, and Lexan in the temperature range just below their glass transition temperatures. The comparison for the 50°C. test shown in Figure 12 is also typical for these materials when tested approximately 100°C. below their glass transition temperatures. In both cases the experimental curve may be described as generally linear up to strains of approximately 2%. The stress-strain curve above 2 to 3% strain might best be described as a plasticity or flow curve.

At all of the combinations of temperature and strain rate shown for Teflon in Figure 13, the predicted and experimental curves diverge at low strains. This highly nonlinear behavior agrees with the behavior of Teflon shown by Figure 5. The Teflon curves also show plastic behavior above approximately the 2 to 3% strain range.

The general conclusion may be drawn that Nylon, Delrin, and Lexan may be approximated as linear materials up to about 2% strain. To utilize these data in a strength analysis, it would be necessary to apply the yield or failure criteria established in Figures 7-10, in combination with a linear stress analysis to obtain the strain at which failure occurs. For uniaxial tests where the materials do not fail until the strain is substantially greater than 2%, a strength analysis of the type described here would be inapplicable. However, available data[14,15] on biaxial tests of poly(methyl methacrylate) and Nylon, respectively, indicate that under biaxial loading, yield and failure occur at strains of 1 to 2%. Thus, in this case, a linear stress analysis which includes the yield stress master curve concept might also become an effective strength analysis.

Another interesting observation from Figures 12 and 13 is that very little stress relaxation takes place during a tensile test. This is shown by the nearly straight stress-strain curves predicted by eq. (1). These curves cover approximately one decade of time; consequently, from Figures 3-6 it may be seen that in this relatively short time (in comparison with the time span covered by the complete stress relaxation master curve), the modulus does not decrease significantly. However, as may be seen from a comparison of the predicted curves for Delrin at 50 and 150°C. shown in Figure 12, the change in temperature causes a large change in slope. Thus, from observation of Figures 12 and 13, one might conclude that temperature and linearity effects are just as important in a strength analysis of these polymers as time-dependent effects.

CONCLUDING REMARKS

The yield stress master curve concept has been shown to apply to four crystalline thermoplastics: a polycarbonate (Lexan), polyamide (Nylon), polyacetal (Delrin) and a fluorocarbon (Teflon). Using this concept allows prediction of tensile behavior at strain rates which are not readily available in the laboratory and over a wide range of temperatures. The temperature dependence of the shift factors used in constructing the master curves for these four materials, and for PMMA, PVC, and Mylar which had been studied previously, are very nearly the same. This indicates the possibility of formulating a general theory for describing the mechanical behavior of thermoplastic materials below their glass-transition temperatures.

The stress-strain behavior of Delrin, Nylon, and Lexan was shown to be somewhat nonlinear below approximately 2% strain and highly nonlinear above approximately 2 to 3% strain. Teflon exhibited highly nonlinear behavior over the range of strains studied (0 to 6%). A strength analysis consisting of a linear viscoelastic stress analysis and the yield stress master curves should be approximately correct for

Nylon, Delrin, and Lexan if failure occurred below approximately 2% strain. For strains above 2 to 3%, a strength analysis based on the principles of plastic or nonlinear behavior, but still including the temperature-dependent yield criteria, would probably be required to describe adequately the mechanical behavior of the material.

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TABLE I

Measured Stress Relaxation Shift Factors

T, °C.	Nylon				Teflon			
	E_o = 0.71% log a_T	1.30% log a_T	2.14% log a_T	3.69% log a_T	E_o = 0.65% log a_T	1.34% log a_T	2.14% log a_T	3.69% log a_T
-50					7.17	7.15	7.73	7.27
-25	6.88	6.66	6.36	7.39	5.29	4.88	4.90	4.31
0	4.40	4.25	3.90	3.94	3.24	2.97	3.19	2.39
15			1.64	1.44	2.04	1.76	1.82	0.74
25	0	0	0	0	0	0	0	0
35					-1.92	-2.30		
40	-3.63	-2.53						
50	-4.68	-3.19			-3.12	-4.12		
60	-5.68	-4.31						
70	-6.39	-4.53						
	Delrin				Lexan			
25		8.09	5.78	5.66	7.71	7.10	6.80	11.25
50	5.35	4.79	4.24	3.92	5.73	4.89	4.56	9.02
75	2.21	1.78	1.95	1.80	3.08	3.27	3.20	4.56
100	0	0	0	0	1.60	1.85	1.59	1.70
115							0.65	0.60
125	-2.17	-2.37	-2.11	-1.83	0	0	0	0
135			-2.80	-2.52	-1.02	-1.64	-1.00	
145					-3.81	-4.12		
150	-5.15	-5.49	-4.95	-4.36				

TABLE II

Measured Yield Stress Shift Factors Referenced at 25°C.

T, °C.	Nylon	Delrin	Teflon	Lexan
	log a_T	log a_T	log a_T	log a_T
-100	12.88	14.17	10.27	9.83
-75	11.04	10.93	7.64	7.44
-50	8.42	7.73	6.11	5.73
-25	6.24	5.17	4.71	3.60
0	3.50	2.15	3.32	1.92
25	0	0	0	0
50	-2.50	-2.41	-2.30	-2.23
75		-5.58	-3.34	-4.33
100	-4.32	-7.56	-4.52	-6.09
125		-9.51	-5.80	-7.60
140				-8.78
145				-10.74
150		-11.34	-7.02	-11.44
170		-14.34		

TABLE III

Characteristic Parameters and Limits of Applicability of Equation (4)

Material	T_g, °C.	K_1,[a] psi	K_2,[a] psi	Yield stress range, psi	Temperature range, °C.
Nylon	50	6000	634	$3{,}000 < \sigma_y < 26{,}000$	-100 to 25
Delrin	170	9100	256	$2{,}000 < \sigma_y < 16{,}000$	-50 to 150
Teflon	-97, -50, 127	1360	201	$1{,}000 < \sigma_y < 7{,}000$	-100 to 25
Lexan	150	9880	287	$3{,}000 < \sigma_y < 18{,}000$	-100 to 125

[a] From eq. (4) K_1 referenced at 25°C.

FIGURE LEGENDS

Figure 1.- Schematic diagram of stress relaxation instrument.

Figure 2.- Typical load-time and displacement-time curves showing definition of yield stress.

Figure 3.- Stress relaxation master curves as a function of strain for Nylon.

Figure 4.- Stress relaxation master curves as a function of strain for Delrin.

Figure 5.- Stress relaxation master curves as a function of strain for Teflon.

Figure 6.- Stress relaxation master curves as a function of strain for Lexan.

Figure 7.- Yield stress master curve for Nylon.

Figure 8.- Yield stress master curve for Delrin.

Figure 9.- Yield stress master curve for Teflon.

Figure 10.- Yield stress master curve for Lexan.

Figure 11.- Comparison of yield stress shift factors for the various polymers as functions of temperature difference between test temperature and reference temperature.

Figure 12.- Comparison of experimental and predicted stress-strain curves for Delrin.

Figure 13.- Comparison of experimental and predicted stress-strain curves for Teflon.

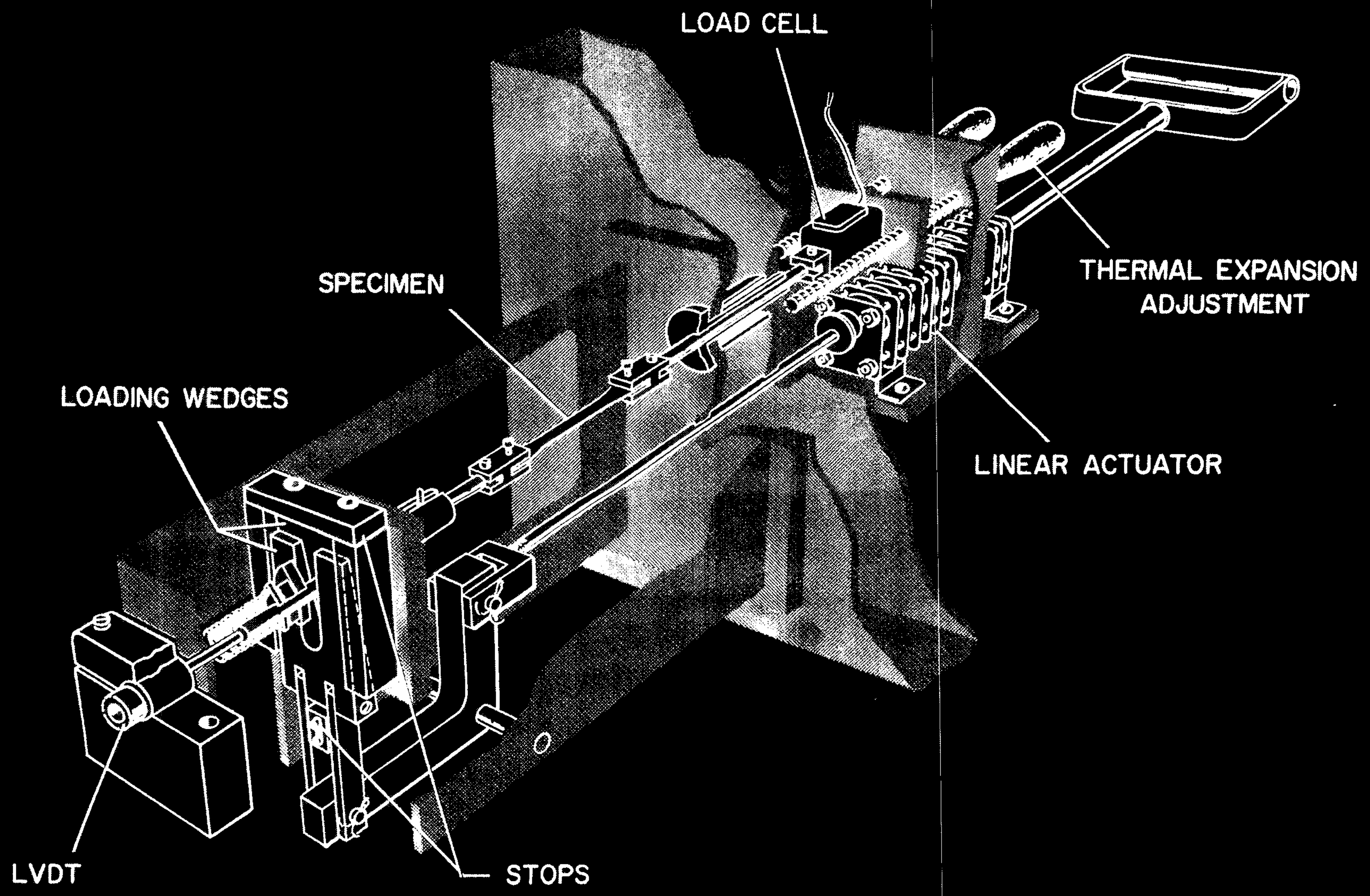

Figure 1.

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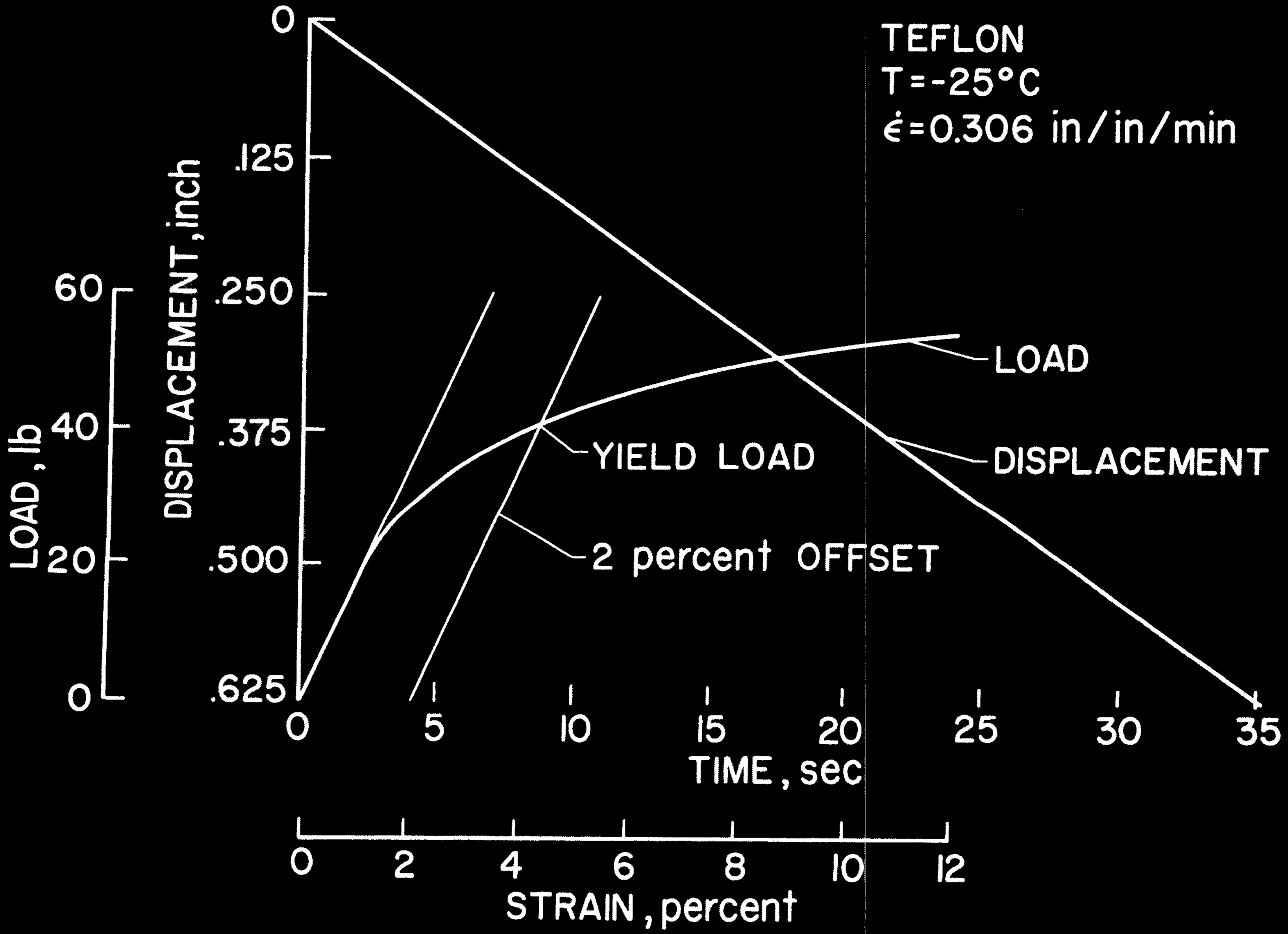

Figure 2.

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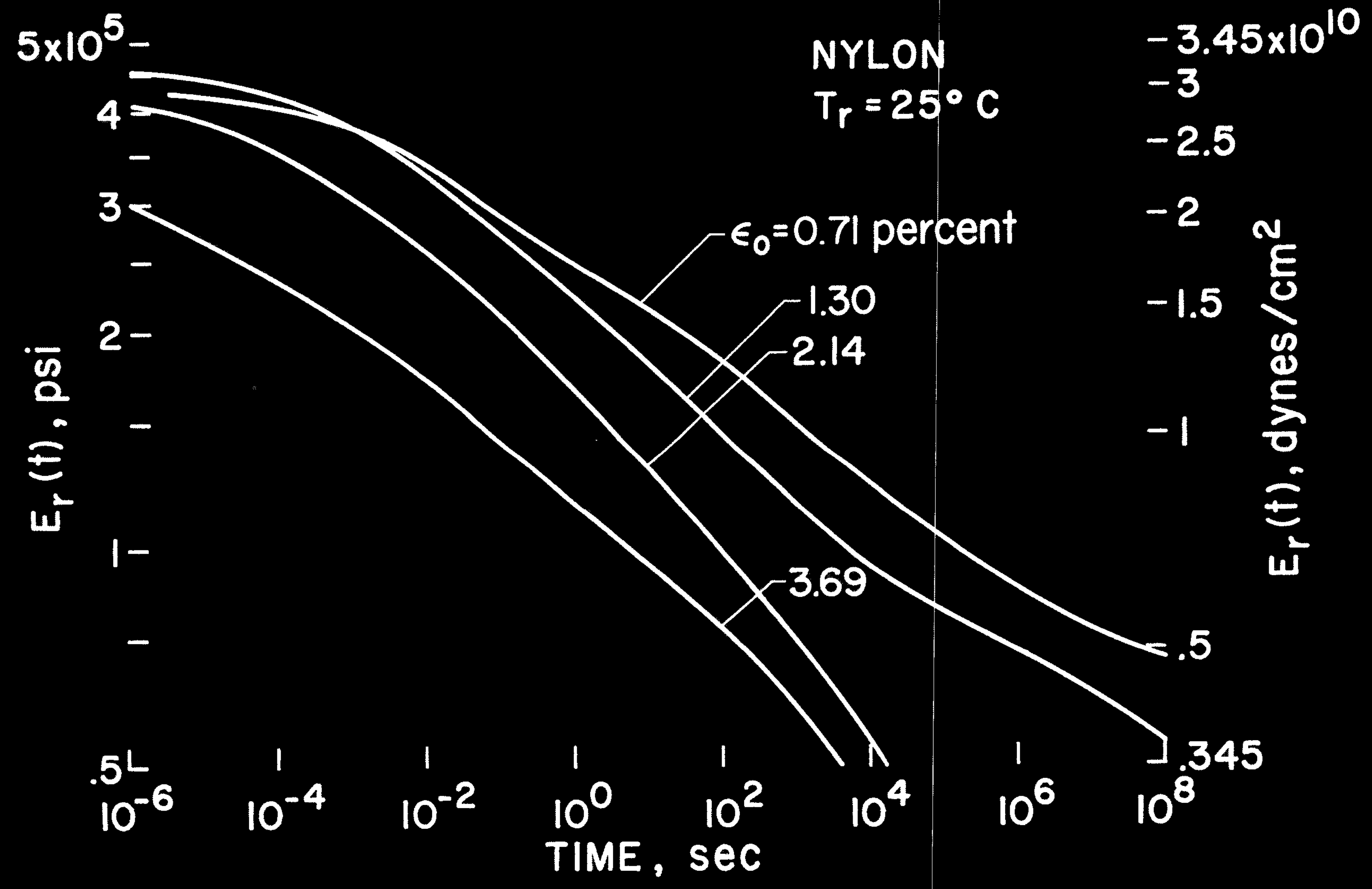

Figure 3.

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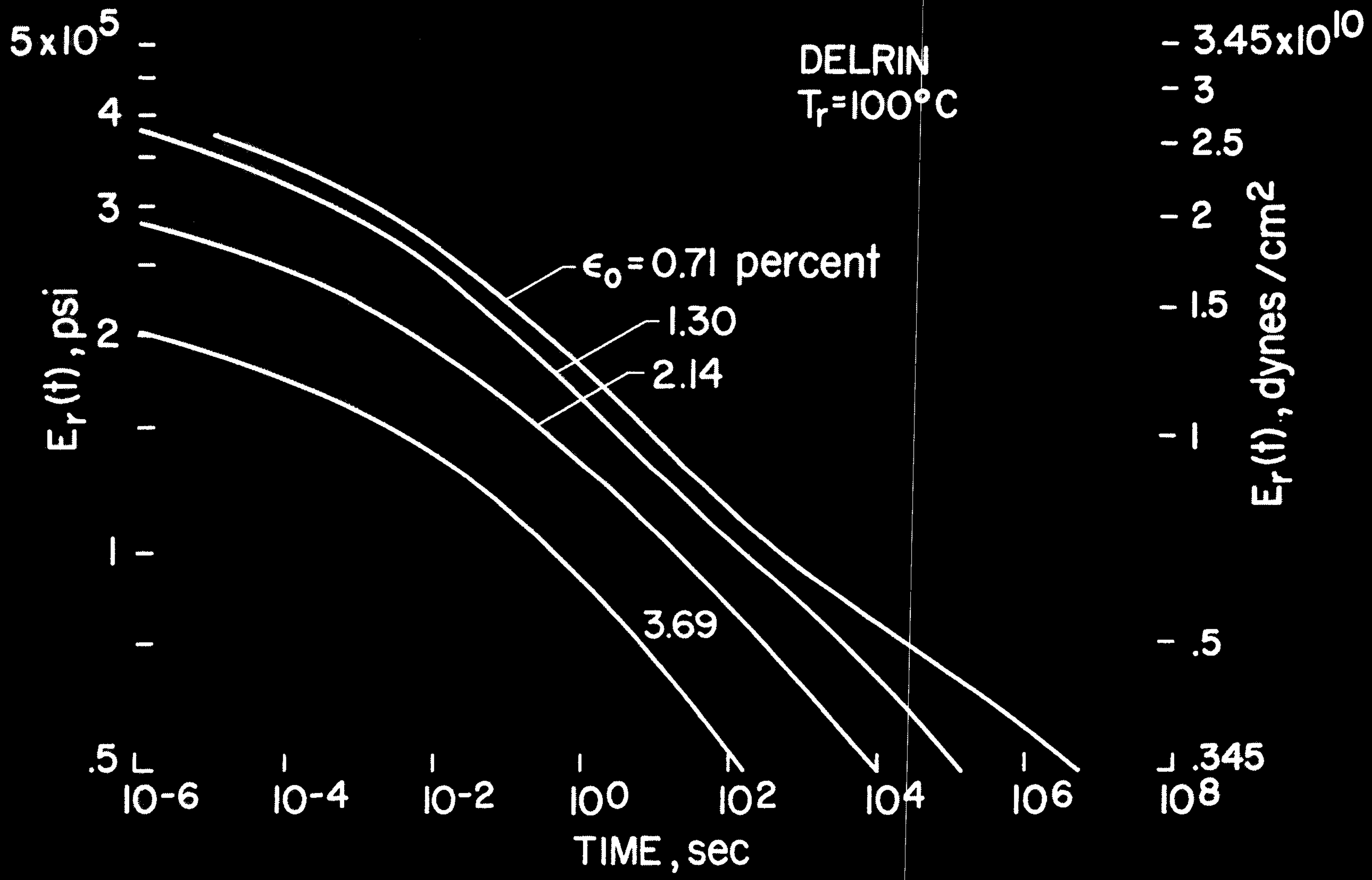

Figure 4.

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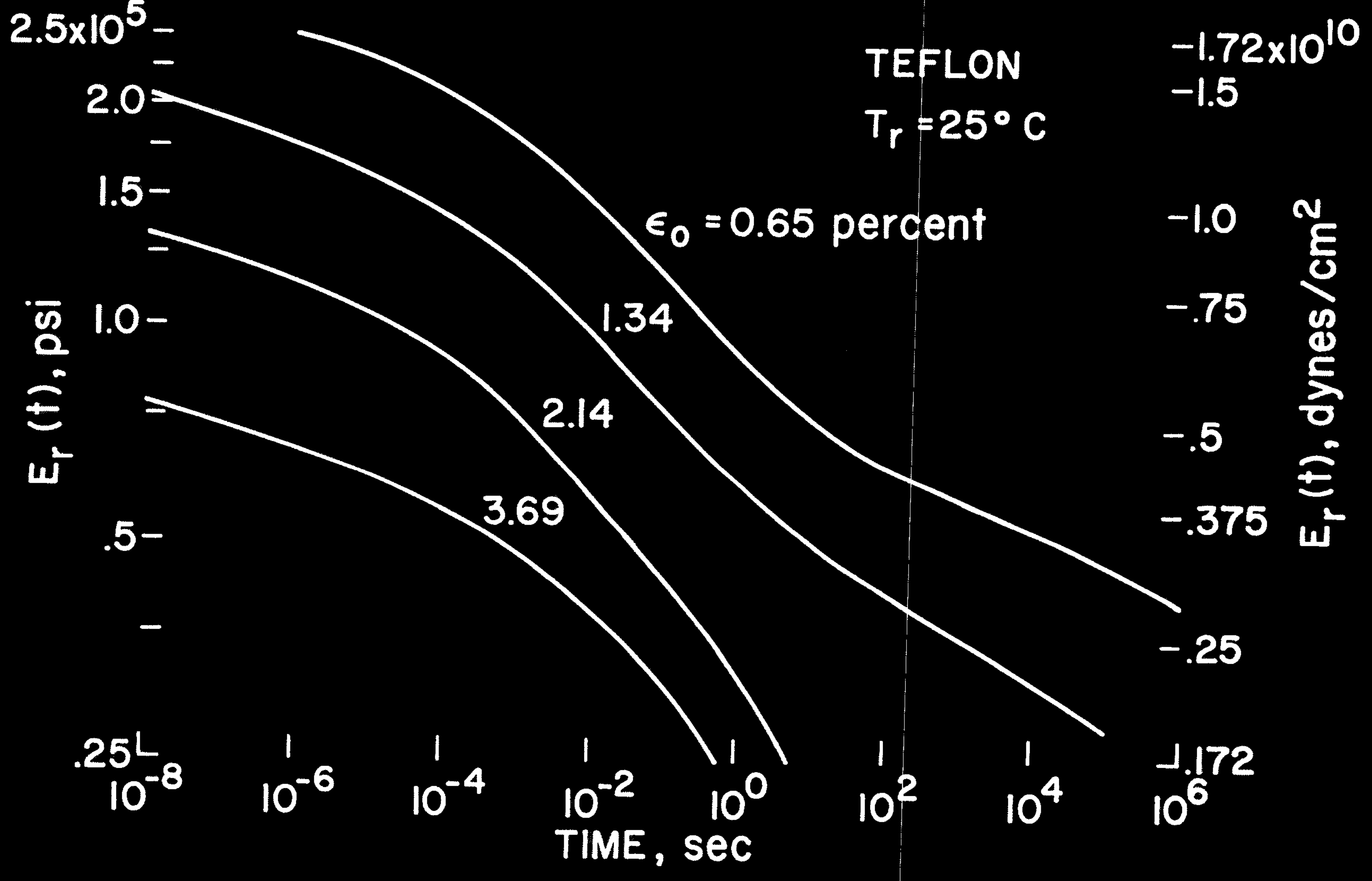

Figure 5.

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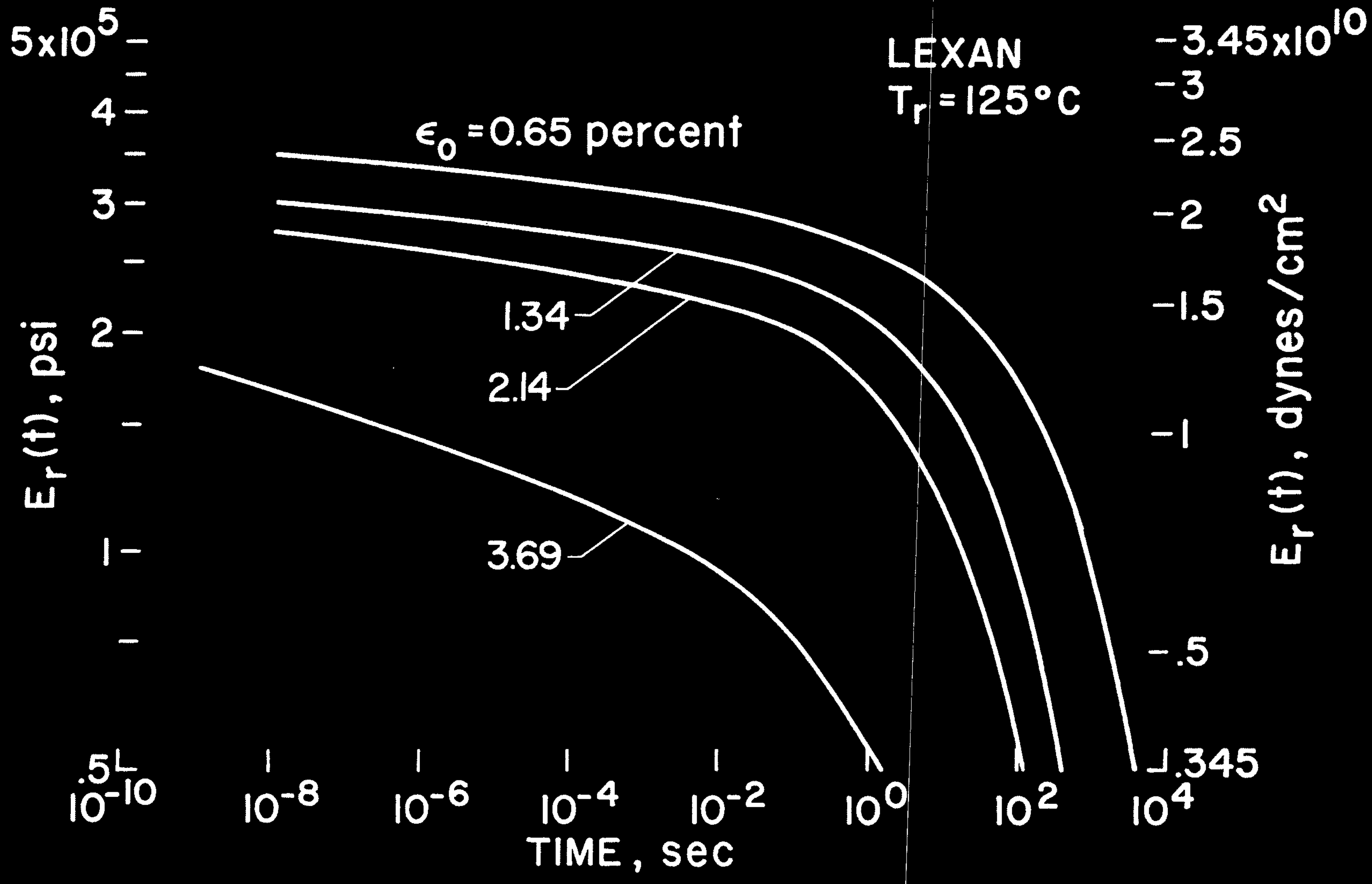

Figure 6.

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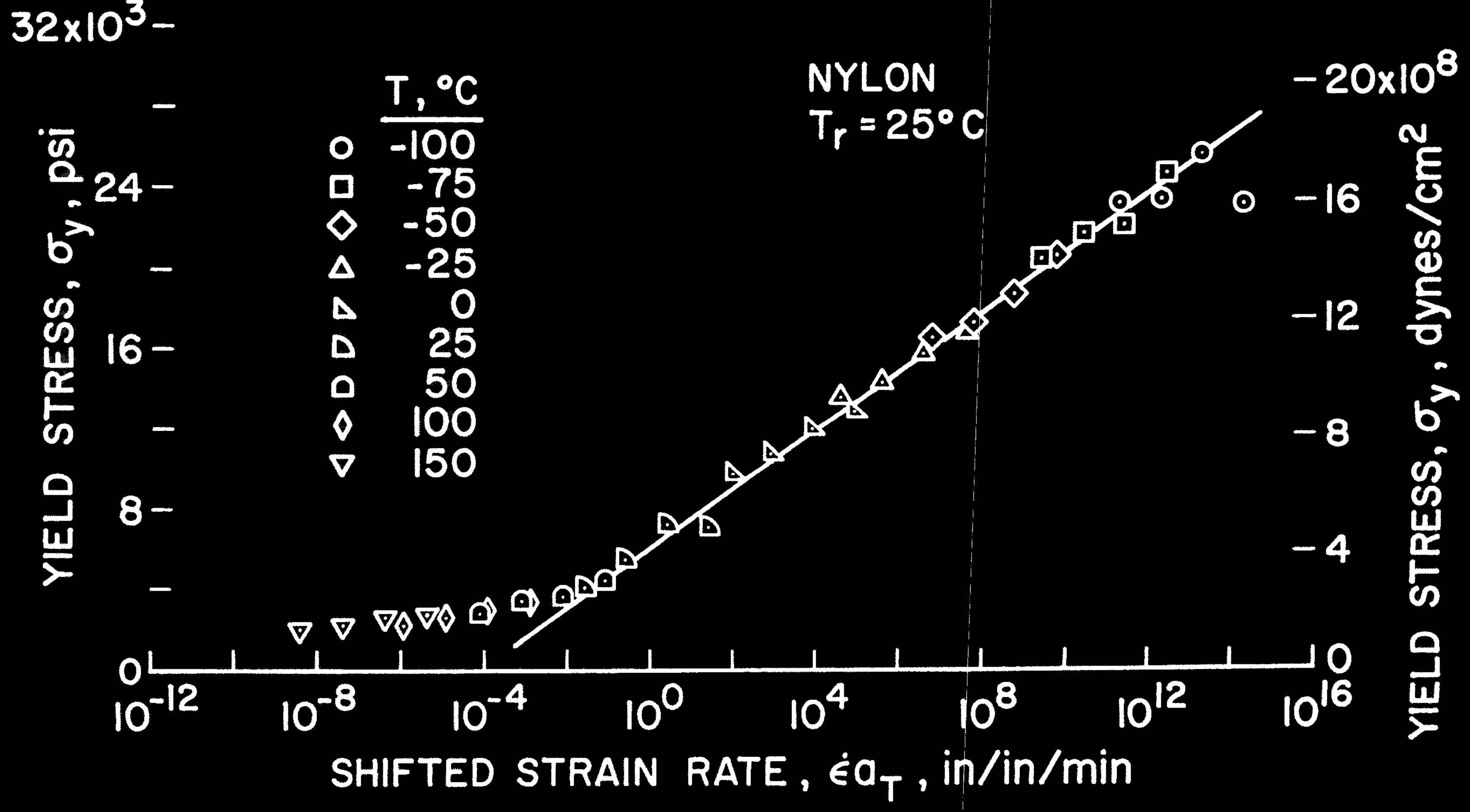

Figure 7.

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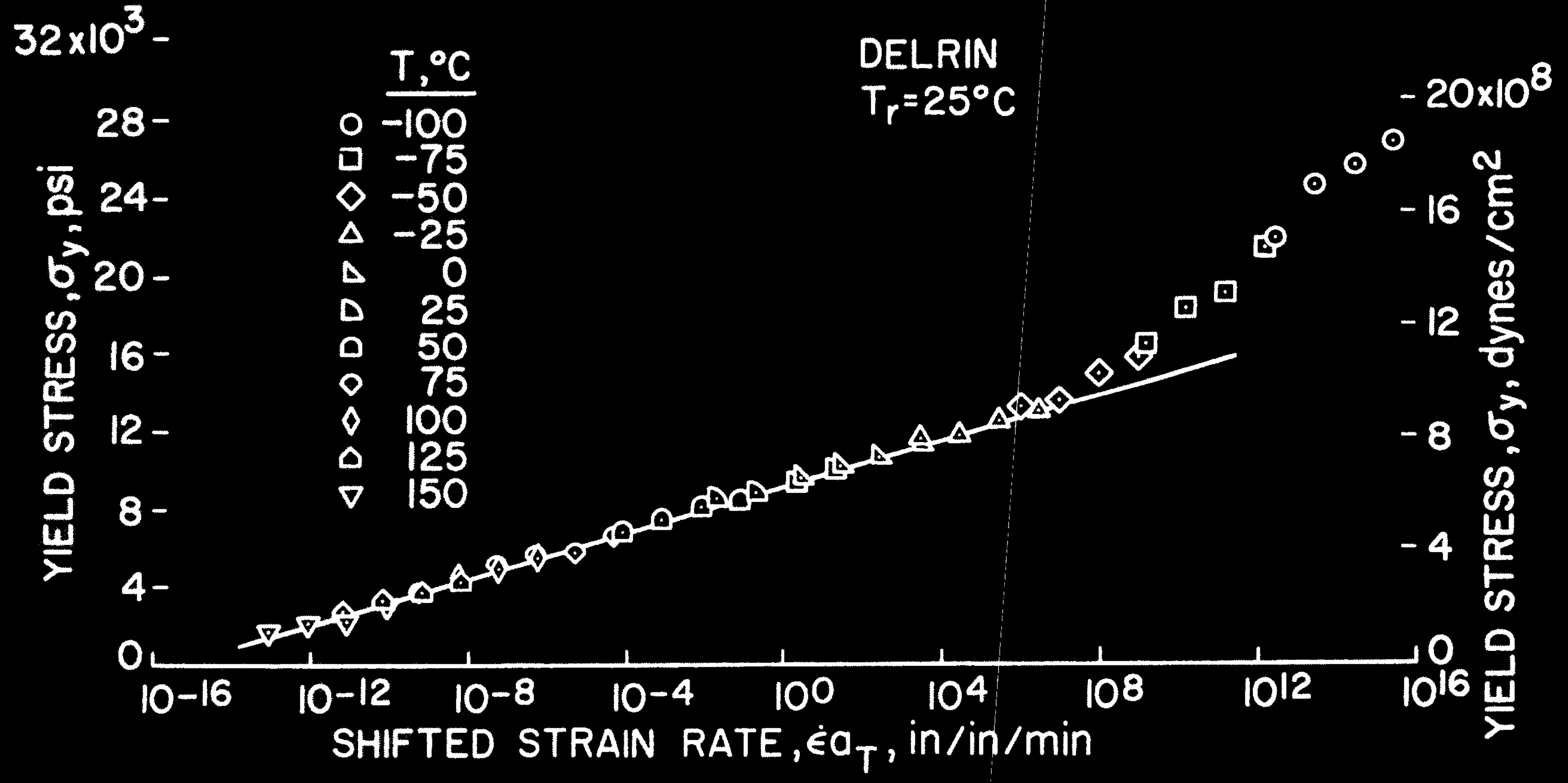

Figure [illegible]

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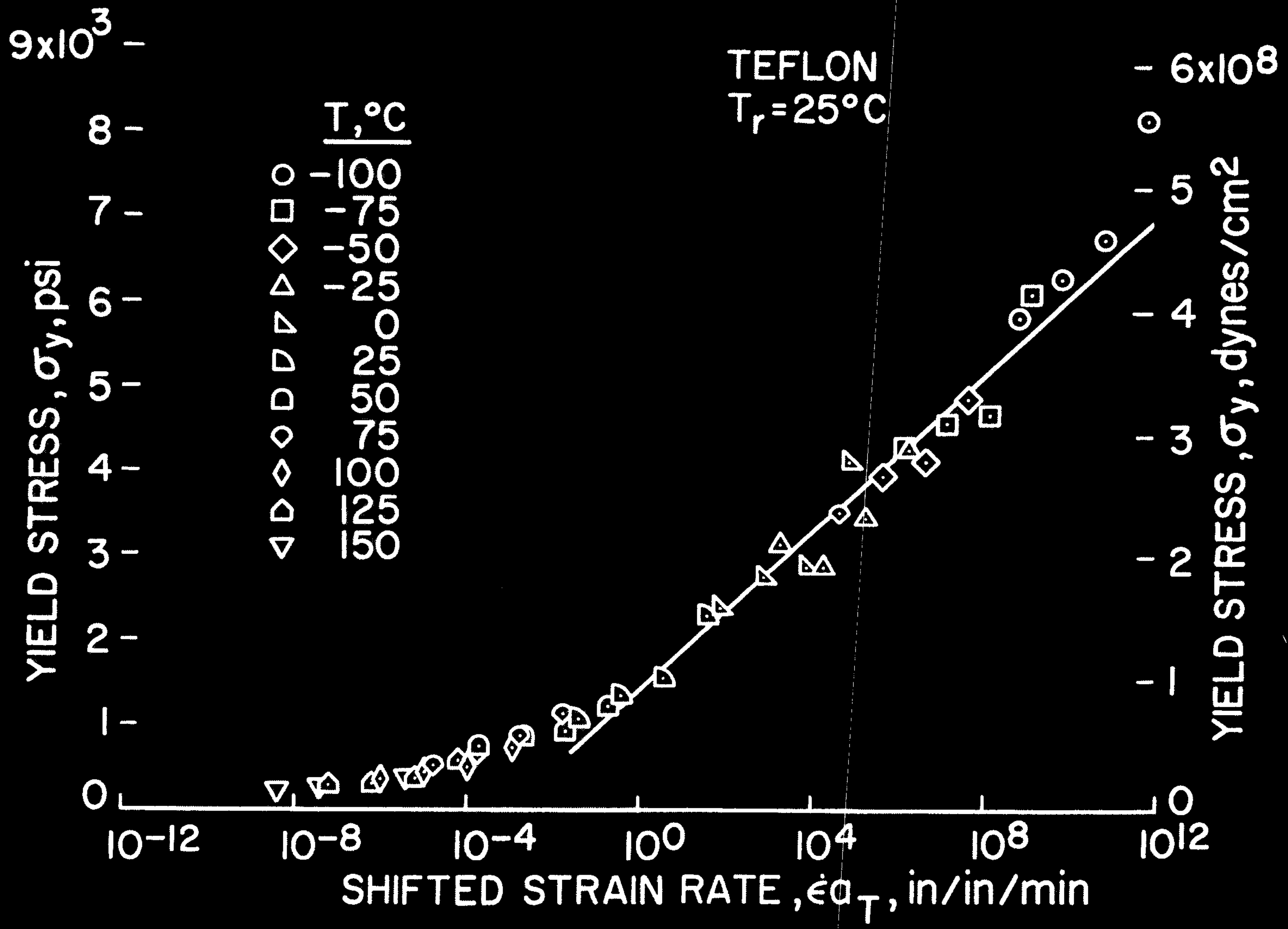

Figure 9.

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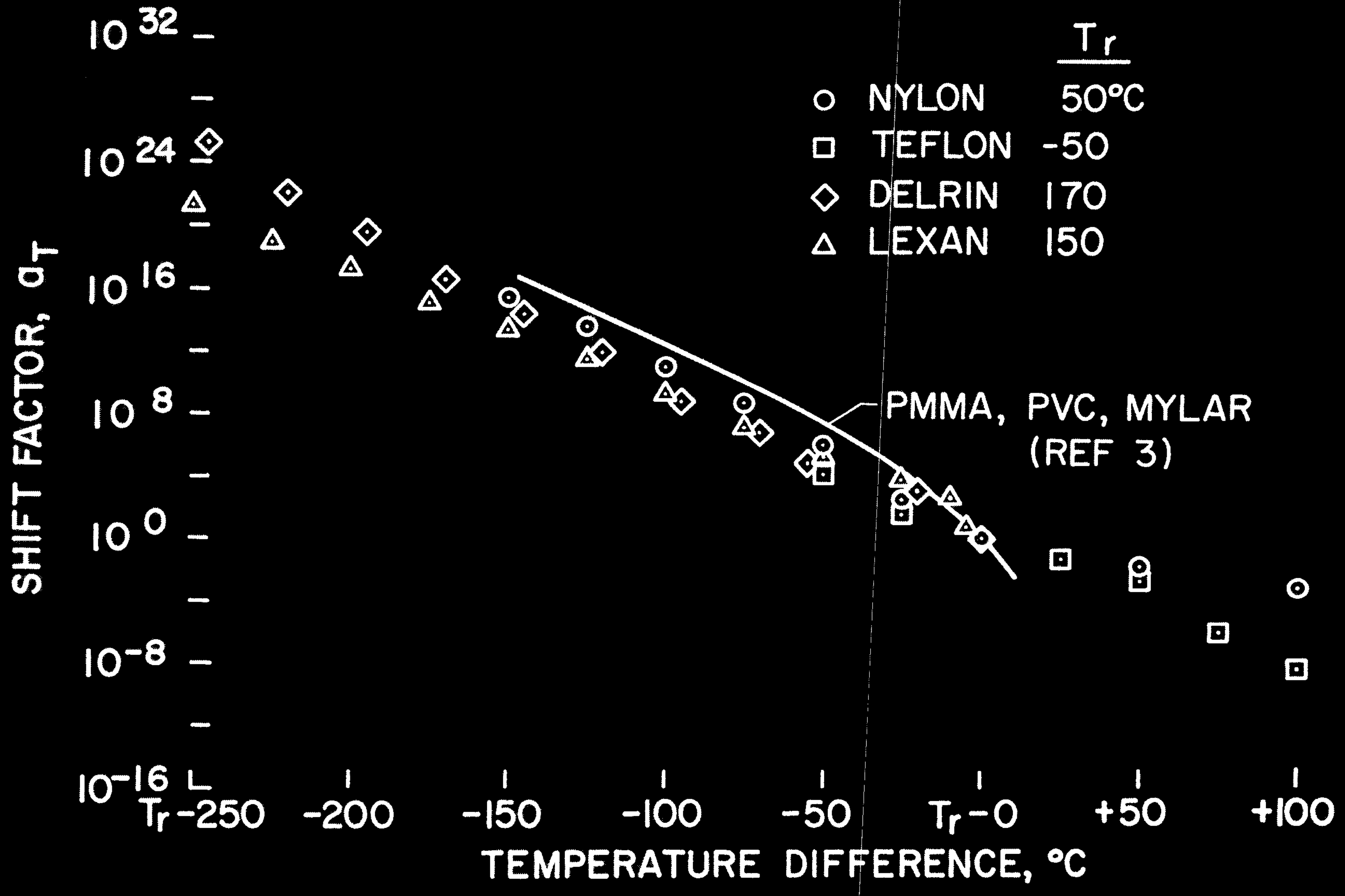

Figure 11.

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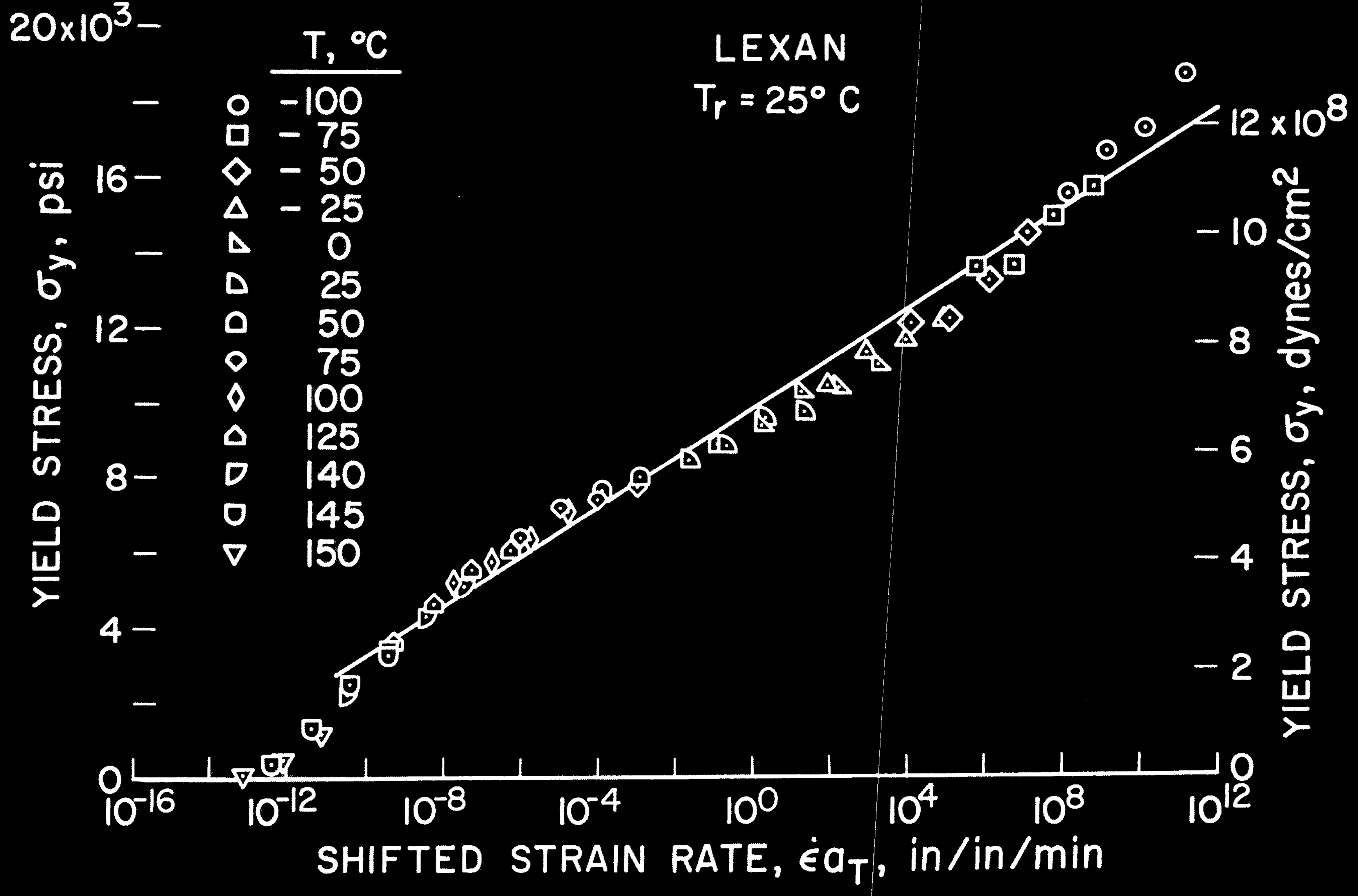

Figure 10.

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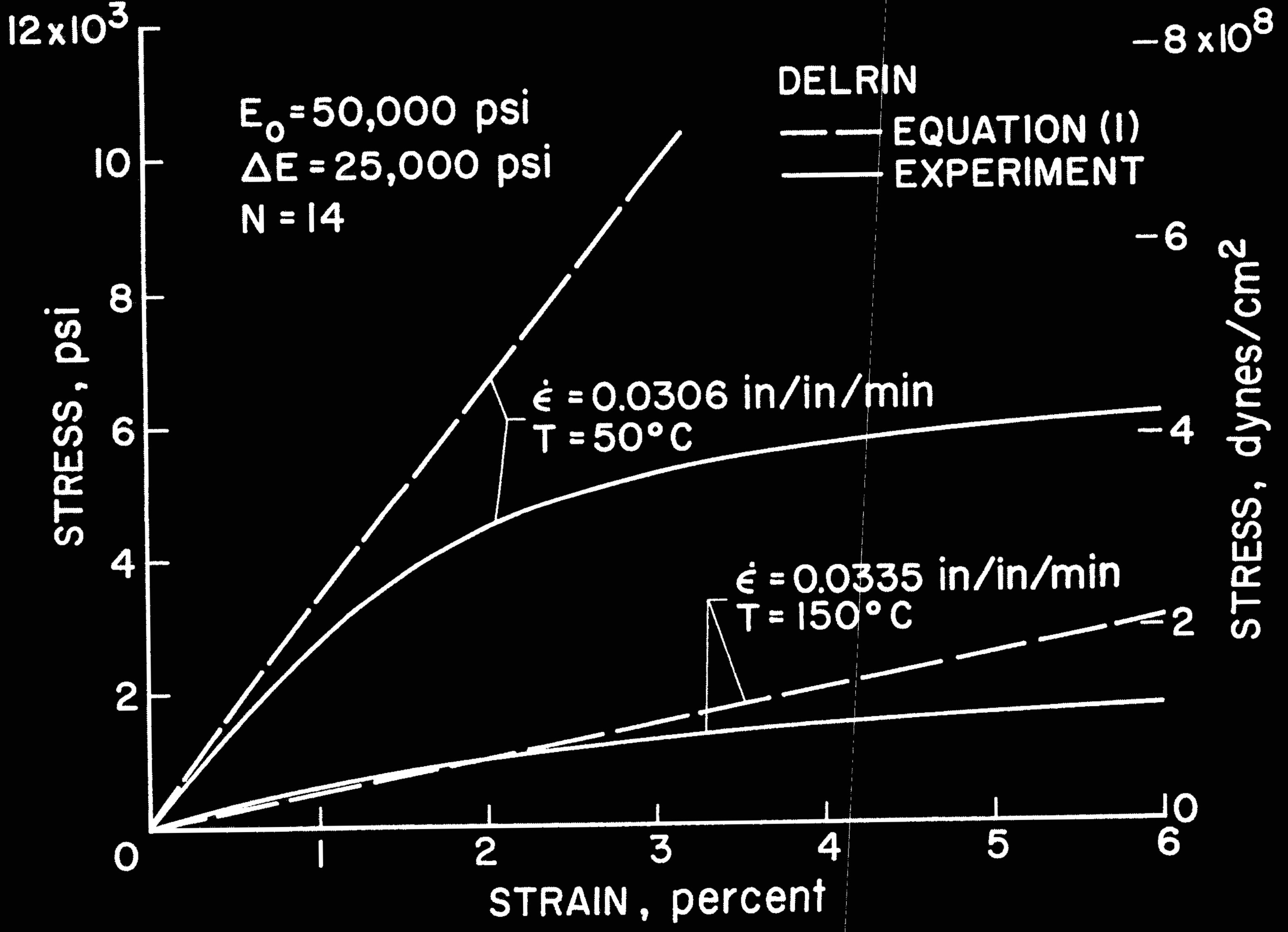

Figure 12.

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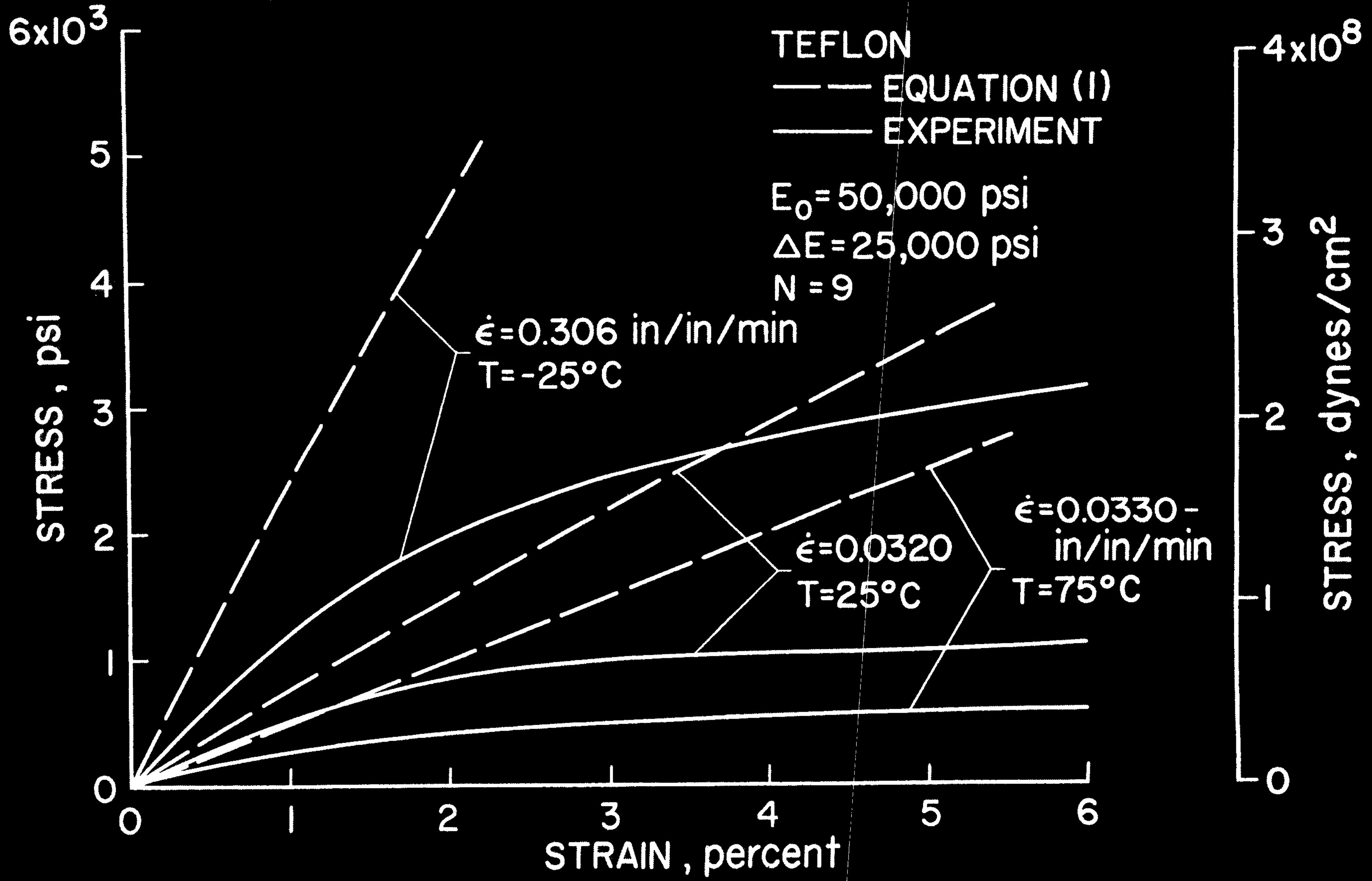

Figure 13.

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