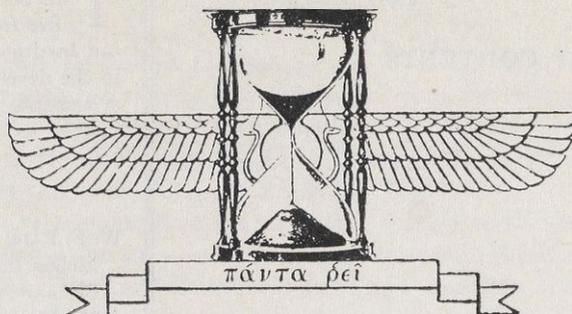


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RHEOLOGY BULLETIN



THE
CONSTITUTION

PUBLICATION OF THE

SOCIETY OF RHEOLOGY

VOLUME 18, NO. 1

FEBRUARY, 1947

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Associate Editors

A. Bondi	J. A. Pask
J. H. Elliott	W. W. Pendleton
R. G. Fordyce	B. R. Roberts
A. Gemant	R. Simha

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Rheology Bulletin

Publication of the Society of Rheology

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The Constitution

THE necessity for certain changes in our constitution in order that it will conform with our new publication policy and the changed status of the members of the "five founder societies," now "member societies," of the American Institute of Physics has been discussed in earlier numbers of the *Bulletin* and at the 1946 annual business meeting.

The committee appointed at that meeting to propose the necessary changes to the membership for its approval or disapproval is W. H. Markwood, Jr., chairman; R. B. Dow; and T. Alfrey, Jr. The committee is actively engaged in the matter at present and will welcome suggestions. The present constitution can be found in the *May, 1946* number of the *Bulletin*.

Since the November meeting, the committee has been considering what articles of the constitution and by-laws will be involved and how they should be changed. That information is presented here and the detailed proposals will appear in the next issue of the *Bulletin*.

Constitution. *Article II* must conform to our policy involving the forthcoming American Institute of Physics news journal and the *Journal of Colloid Science*. *Article IV* may be changed to delete "Publication Editor" when the new A.I.P. journal appears. New articles dealing with meetings of the Society, officer reports, and committee reports may be added.

By-Laws. The changes under discussion may alter the by-laws rather extensively. They will be concerned with the detailed duties and responsibilities of the officers and executive committee; types of membership, dues, and delinquencies; committees. There should also be articles dealing with our representation as members of the American Institute of Physics and with our representation on the Council of the American Association for the Advancement of Science. Further articles should present the details of our publication policies; prepare for affiliated or local sections; set up requirements for future by-law amendments.

Let your ideas and attitudes be known.

Reviews

Rheological Theory

Interpretation of the Internal Viscosity of Chain Molecules. II. W. KUHN AND H. KUHN, *Helv. Chim. Acta* 29, 830 (1946).

The considerations of part one are extended to a computation of the relaxation times associated with the decay of tension in a thread-like molecule held at constant separation of chain ends and caused by the rotational Brownian motion of chain segments. The time constant associated with the rotation around a particular axis depends upon its position within the chain. It is largest for axes situated near the center of the molecule. The resultant over-all relaxation time is proportional to the degree of polymerization, a result similar to that obtained by Frenkel.¹ Here a detailed expression for the proportionality factor as function of parameters characterizing the segment and the geometry of the rotation barrier is obtained.

—ROBERT SIMHA

¹ Frenkel, *Acta Physicochim. URSS* 9, 236 (1938).

Statistical and Energy Contributions to the Total Retractive Force in Highly Extended Chain Molecules. Superposition of Statistical and Energy Effects in Highly Deformed Rubber, I and II. W. KUHN AND H. KUHN, *Helv. Chim. Acta* 29, 1095, 1615, 1634 (1946).

The principal novel feature in these three papers is an attempt to introduce those effects caused by the change of the internal energy upon deformation in addition to the well-known entropy contribution to the retractive force.

The first article is concerned with the energy increment in a single strained chain in which atomic distances and valence angles are deformed and ultimate breakage of a link occurs. On the basis of thermochemical and spectroscopic data on hydrocarbons of low molecular weight it is shown that up to chain-end separations equal to about 95 percent of the total extended length the chain segments may be considered as effectively rigid. Beyond this range the energy of the chain increases with elongation and is proportional to the square of the force applied, while the entropy decreases only as the logarithm of the force. The energy increases approximately with the square of the excess extension over the maximum unstretched length of the molecule.

These results are used in the subsequent articles to derive the stress-strain curve for simple elongation of a cross-linked rubber-type polymer. Two effects are disregarded in the detailed calculations. First, the migration of points of cross-linkage. In other words, the assumption that the coordinates of such a point are changed on deformation in the same ratio as the macroscopic dimensions of a cube of the material should not be valid at high elongation. Secondly, orientation effects leading ultimately to crystallization in natural rubber are neglected. It is then assumed that each portion of a chain between crossbonds can be extended to a certain fixed (small)

limit beyond its maximum stretched out (undeformed) length, before dissociation is produced. With the harmonic type of expression for the energy of an individual chain and the probability for a given large separation of chain ends which replaces the usual Gaussian distribution, the total energy of the sample is obtained as function of the degree of elongation. The energy first increases steeply above a certain elongation and then decreases indicating that more and more chains are being broken and do not contribute to the stress, since no reforming is assumed to take place during the duration of the experiment. The elongation at the maximum energy is proportional to the square root of the degree of polymerization of the chain portions. The maximum energy itself is independent of this number. The energy contribution to the total retractive force behaves in a similar fashion. The maximum degree of extension attainable turns out to be inversely proportional to the square root of the initial elastic modulus.

Finally the total retractive force calculated is compared with a stress-strain curve of a natural rubber sample given by Treloar¹ for the whole range up to the breaking point which occurs at approximately 650 percent elongation. It can be deduced that the maximum possible extension of an individual chain portion beyond its maximum length amounts to about 7 percent on the average. The theoretical curve is found to be above the experimental one. At roughly 600 percent the latter curve lies about midway between the curve of entropy stress and the total one.

—ROBERT SIMHA

¹ Treloar, *Trans. Faraday Soc.* 60, 59 (1944).

Notes on the Rate Process Theory of Flow. A. BONDI, *J. Chem. Phys.* 14, 591 (1946).

This paper deals with the separation of the contributing terms of the energy and entropy of activation for viscous flow and the identification of some of the molecular constants entering into these terms, as a step toward the prediction of the viscosity of liquids from first principles. The energy of activation for hole formation ΔH_i^h is found to be determined mainly by the magnitude of the dispersion energy and by the extent of displacement of the hole forming molecules from their equilibrium position. The latter, in the form of the viscosity-pressure coefficient ΔV_i , is, at present, not predictable from any molecular constant. The energy of activation for motion into the hole ΔH_i^j was found for many substances to be numerically equal to the excess energy of vaporization ΔE_{vap}^j and is thus an additional measure of restricted external rotation. The cases in which $\Delta H_i^j > \Delta E_{vap}^j$ offer strong evidence for the requirement of deformation of the flowing molecule against internal potential energy barriers. The rotational, translational, and cooperative terms which contribute to the entropy of activation have been separated, but can so far be determined only from viscosity data. Numerical

examples show that the stereometric arrangement of molecular structure determines the magnitude of ΔV^\ddagger and ΔH^\ddagger and thereby the viscosity of liquids to a far greater extent than chemical composition (except for OH groups). The existence of aliphatic fatty acids as double molecules in the liquid state over a wide range of temperatures is confirmed viscosimetrically, while the viscosity data of aliphatic alcohols suggest the presence of distinguishable multiple molecules only at very low temperatures but the existence of a continuous OH-bond network structure at ordinary temperatures. One of the important consequences of the rate process theory of flow is the recognition that the viscosity of a liquid is determined by the (very small) concentration of molecules in relatively shallow potential energy wells from which the activated molecules are preferentially recruited. Viscosity is therefore not a bulk property in the commonly accepted sense and depends only to a minor extent on the structure of a liquid, i.e., on its state of order.

—AUTHOR'S REVIEW

The Relation between the Viscosity of Liquids and Their Pressure. G. M. PANCHENKOV, *Comptes Rendus Acad. Sci. URSS* **51**, 365 (1946).

The author applies the following equation¹ which shows that viscosity is a function of pressure as well as temperature.

$$\eta = 3(6R)^{1/2} \left(\frac{\omega^2}{N_0} \right)^{1/3} \frac{e^{4/3}}{M^{5/6}} T^{1/2} e^{\epsilon/RT} (1 - e^{\epsilon/RT})^2.$$

Here N_0 is Avogadro's number, ω is the actual molecular volume corresponding to one gram molecule of matter, ϵ is the bond energy of the molecules of the liquid, and the remainder of the symbols have their usual significance. The magnitude of the bond energy between the molecules of a given liquid depends upon the distance between them and hence is a function of pressure. The bond energy ϵ_p under the pressure p is given by

$$\epsilon_p = \epsilon_0 + \frac{2}{\gamma} \int_{V_p}^{V_0} l dv$$

where ϵ_0 is the bond energy under atmospheric pressure, V_p and V_0 are the corresponding molecular volumes, l is the molar latent heat of expansion of the liquid, and γ is the coordinational number (undefined).

This integral is expressed in terms of readily measurable thermodynamic quantities and the calculated viscosities of ethyl alcohol and ethyl bromide at 30°C over the pressure range 1–12,000 atmospheres compared to Bridgeman's experimental values.² For ethyl alcohol the calculated values are lower than the experimental, up to 10,000 atmospheres, but are larger at 12,000 atmospheres. In the case of ethyl bromide, the same discrepancy is found except that above 8000 atmospheres the calculated values are larger than the experimental. The differences between calculated and observed values are as great as 25 percent in some cases.

—J. H. ELLIOTT

¹ G. M. Panchenkov, *Comptes Rendus Acad. Sci. URSS* **50** (1945).
² Bridgeman, *Proc. Am. Acad. Sci.* **49**, 3 (1913).

The Viscosity of Liquid Mixtures. G. M. PANCHENKOV, *Comptes Rendus Acad. Sci. URSS* **51**, 457 (1946).

Panchenkov's equation (see previous abstract) may be applied to liquid mixtures in the form

$$\eta_m = A_m \rho_m^{4/3} T^{1/2} e^{\epsilon_m/RT} (1 - e^{-\epsilon_m/RT})^2, \quad (1)$$

where the subscript m denotes mixture. A_m is a function of the composition of the mixture and the other symbols have their usual significance. The bond energies of the molecules of each kind in a binary mixture are calculated as follows:

$$\begin{aligned} \epsilon_1 &= \epsilon_{11}C_1 + \epsilon_{12}C_2, \\ \epsilon_2 &= \epsilon_{21}C_1 + \epsilon_{22}C_2, \end{aligned} \quad (2)$$

where C_1 and C_2 are the mole fractions of the two components and $\epsilon_{12} = \epsilon_{21}$ is the binding energy between molecules of the first kind and those of the second kind. It is assumed that only neighboring molecules interact and that their relation concentrations around any of them are identical. ϵ_m in Eq. (1) is obtained from the equation:

$$\epsilon_m = \epsilon_1C_1 + \epsilon_2C_2 \quad (3)$$

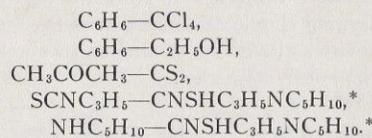
which reduces to:

$$\epsilon_m = \epsilon_{11}C_1^2 + 2\epsilon_{12}C_1C_2 + \epsilon_{22}C_2^2.$$

Depending on the relative magnitudes of ϵ_{11} , ϵ_{22} , and ϵ_{12} the viscosity composition curve may be linear, show a maximum, minimum, or point of inflection.

Values of A_1 , ϵ_{11} , and $A_2\epsilon_{22}$ are obtained from the experimentally determined viscosities as a function of temperature. A_m and ϵ_{12} are similarly determined from data for a known mixture.

Values of ϵ_{11} , ϵ_{22} , and ϵ_{12} are calculated for the individual components and the following mixtures, by using data reported in the literature:



The viscosity-composition curves obtained from these values of ϵ_{11} , ϵ_{22} , and ϵ_{12} are found to be in good agreement with the experimental values.

—J. H. ELLIOTT

* The compound formed between allyl isothiocyanate and piperidine.

On the Application and Derivation of the New Viscosity-Temperature Relationship of Liquids. M. S. TELANG, *J. Phys. Chem.* **50**, 373 (July, 1946).

This paper is a continuation of the author's studies on correlation of viscosity with temperature.¹ Data from the *International Critical Tables*, Landolt-Boernstein, and other sources, are used to test the applicability of the author's findings. Four groups of liquids, of various degrees of association, are listed. Arrhenius', Porter's, Guzmán's, Raman's, and other authors' equations are interpreted in their relationship to the problem. Eyring's "theory of holes" and his derivation of Batschinski's equation regarding viscosity as a rate process are discussed. A bibliography of 72 references completes this paper.

—B. R. ROBERTS

¹ Cf. *Rheol. Bull.* **17**, No. 2, 10 (Aug. 1946).

Macromolecules

Studies on Siloxanes I. The Specific Volume and Viscosity in Relation to Temperature and Constitution. C. B. HURD, *J. Am. Chem. Soc.* **68**, 364-370 (1946).

The specific volumes and viscosities of eleven siloxanes are reported. Four of these samples were ring compounds and seven were chain type. Ten contained only methyl groups, one contained two phenyl and six methyl groups. Coefficients for the equation $V = V_0(1 + at + bt^2)$ are tabulated. Molar volumes, MV_{20} , were calculated for the compounds, also for the groups $\text{Si}(\text{CH}_3)_2\text{-O-}$, for $[\text{CH}_3 + \text{Si}(\text{CH}_3)_2]$ and for the contribution from the ring structure. A linear relation between $\log \eta$ and $1/T^\circ\text{K}$ is reported. Values are tabulated for $Q\eta$ and A in the formula $\log \eta = Q\eta/2.30RT + A$. The ratios of $Q\eta$ to ΔH and to $\Delta \epsilon$ of vaporization are reported. The relations between η and n , and also between $Q\eta$ and n are shown graphically. By using Batschinski's equation, $\eta = C/(V-w)$, values for C and w were calculated as well as a value $\epsilon_{\text{Si}} = 22.3$ in Batschinski's additive relationship. There is slight evidence of association in these liquids.

—R. G. FORDYCE

Vapor Pressure-Viscosity Relations in Methylpolysiloxanes. D. F. WILCOCK, *J. Am. Chem. Soc.* **68**, 691-696 (1946).

The isolation of linear methylpolysiloxanes containing 7, 8, 9, 10, and 11 silicon atoms is reported. Their vapor pressure curves and viscosities were reported. The vapor pressure curves of shorter linear compounds and of the cyclic compounds up to 8 were also determined.

The following simple relations expressing viscosity and vapor pressure as functions of temperature were found to hold for the linear molecules:

$$\log p_2 = 6.28 - \frac{1030}{T} + \left(0.443 - \frac{360}{T}\right)x \quad (x \geq 5),$$

$$\log \eta_2 = -2.04 + \frac{380}{T} + \left(0.37 + \frac{326}{T}\right) \log x;$$

and for cyclic molecules:

$$\log p_R = 7.07 - \frac{1190}{T} + \left(0.265 - \frac{294}{T}\right)x,$$

$$\log \eta_R = -2.13 + \frac{214}{T} - \left(0.04 - \frac{866}{T}\right) \log x,$$

where x is the number of silicon containing units.

A comparison of the energy of vaporization with the activation energy for viscous flow indicates that the unit of flow in high molecular weight silicone polymer fluid is $(\text{CH}_3)_{14}\text{Si}_7\text{O}_7$. This is larger than the unit of flow in hydrocarbons as estimated by Eyring and co-workers. Since a smaller activation energy for viscous flow was found it was concluded that intermolecular attractive forces are much smaller in liquid linear polysiloxanes than those in hydrocarbons of comparable molecular weight.

—R. G. FORDYCE

The Viscosity-Molecular Weight Relation for Natural Rubber. W. C. CARTER, R. L. SCOTT, AND M. MAGAT, *J. Am. Chem. Soc.* **68**, 1480-1483 (1946).

Results are presented showing the measurement of osmotic pressure molecular weights and intrinsic viscosity determinations for twenty-two different fractions of hevea rubber ranging in molecular weight from 40,000 to 1,500,000. These data were found to correspond to the relation: $[\eta] = 5.02 \times 10^{-4} M^{0.667}$ for hevea.

—R. G. FORDYCE

Shape and Size of Pectinic Acid Molecules Deduced from Viscometric Measurements. H. S. OWENS, H. LOTZKAR, T. H. SCHULTZ, AND W. D. MACLAY, *J. Am. Chem. Soc.* **68**, 1628-1632 (1946).

The intrinsic viscosities of various pectins measured in 0.155M sodium chloride solution were found to decrease with rise in temperature and to be essentially independent of pH. The length-to-diameter ratios calculated by means of Simha's equation varied from 53 to 165 with molecular weights ranging from 2.3×10^4 to 7.1×10^4 . The results indicate that pectin has a rigid rod-like structure in aqueous salt solution. Osmotic pressure measurements indicated number average molecular weights varying from 1.8×10^4 to 3.9×10^4 . The discrepancy between these figures and the viscosity values is ascribed to heterogeneity of the samples.

—R. G. FORDYCE

Effect of Molecular Weight and Method of Deesterification of the Gelling Behavior of Pectin. R. SPEISER AND C. R. EDDY, *J. Am. Chem. Soc.* **68**, 287-293 (1946).

The molecular weight and molecular weight distribution are reported for pectins deesterified by acid and by enzyme catalysis. The acetone solution viscosity of nitrated pectin was used to determine molecular weight. Close agreement between observed viscosity-average molecular weight and the weight average calculated by combining fractions implies that the exponent a in Flory's equation is unity. Molecular weight distribution for pectin nitrated was determined by fractional precipitation of the acetone solution with toluene.

The gelling behavior of the pectin samples prior to nitration is correlated with molecular weight, extent of esterification, and the deesterification technique. Gel strength is shown to increase with increasing molecular weight but is substantially independent of the esterification degree and of the method of deesterification.

The strengths of ionic-bonded gels are less affected by molecular weight than are hydrogen-bonded gels. Ionic-bonded gel strengths however are strongly dependent on the degree of esterification.

—R. G. FORDYCE

Effects of Salts on the Viscosity of Pectinic Acid Solutions. H. LOTZKAR, T. H. SCHULTZ, H. S. OWENS, AND W. D. MACKLAY, *J. Phys. Chem.* **50**, 200 (May, 1946).

Viscosity values of pectinic acids in solution of salts, typical for various phases of pectin technology, were determined at $25 \pm 0.003^\circ\text{C}$. Alkali metal salts (sodium chloride, potassium chloride, sodium hexametaphosphate), ammonium chloride, and various alkaline earth and other polyvalent metallic salts were studied in this connection. In dilute pectinic acid solutions the salts depress the electroviscous effect of the ionized pectinic acid molecule. The

relative viscosity η_r satisfies the equation

$$\log \eta_r = \log \eta_{r_0} - \kappa \mu,$$

(μ being the ionic strength of salt, η_{r_0} the relative viscosity of ash free pectinic acid solution, η_r the relative viscosity with salt, κ the slope which varies with pH and kind of pectinic acid). In several diagrams the relative viscosity is plotted against pH and (molal salt concentration)¹. Viscometric details are identical with those of earlier papers of the same authors, abstracted previously.

—B. R. ROBERTS

Engineering Applications

Internal Friction in Engineering Materials. J. M. ROBERTSON AND A. J. YORGIADIS, *J. App. Mech.* **13**, A173 (1946).

Two types of apparatus were used: one for torsional tests, and another for tension-compression tests. The frequency range covered 9 from 68 c.p.s. Data refer to room temperature. Stresses covered up to 50,000 lb./sq.in. Materials include: lucite, Bakelite, plywood, magnesium alloys, monel metal, and steels. The results are the following:

Specific damping capacity is independent from the frequency in the narrow range covered. Specific damping capacity increases linearly with stress up to a certain limit corresponding approximately to the yield point. For higher stresses, damping increases at a rate higher than with the first power of the stress. The dynamic modulus, both of elasticity and rigidity, slowly decreases with increasing stress. Damping is of the same order of magnitude for shear and tension-compression; they are strictly the same for stresses that produce equal distortion in the two types of deformation. From this it appears that, in the frequency range covered, no thermal loss mechanism caused by changes in volume was operative; damping in this range is caused by plastic flow.

—ANDREW GEMANT

Determination of Plastic and Brittle States of "Pool" Bitumens. W. LETHERSICH, *J. Soc. Chem. Ind.* **65**, No. 7, 190-196 (July, 1946).

Tensile strength, ductility, and impact tests are used to determine the plastic and brittle state of bitumens. Brittleness is defined as failure caused by the hydrostatic component. Thus, rupture occurs when the hydrostatic tension reaches the true molecular cohesion of the material. Plastic failure is attributed to the shear component only. Therefore, since the ductility test is applicable to the determination of the stress-strain characteristic, it is useful to investigate the slow plastic flow and failure of a bitumen. On the other hand, since the rate of loading can be controlled in the tensile strength tests and since brittleness depends on the rate of loading, this test can be used to study the brittle state. Forces caused by temperature

cycles on short-circuited apparatus would be an example of the type of problem best studied by means of the tensile strength test. All bitumens are brittle under impact tests and, therefore, a comparison of the impact energy is needed to classify the materials according to brittleness. This type of test is useful for investigating the effects of mechanical shock in handling or in associated apparatus. Reference is made to mechanical models representing the plastic and elastic components of bitumens.

—W. W. PENDLETON

The Measurement of Fluidity and Viscosity of Metal Alloys. N. N. KURNAKOV AND M. J. TRONEVA, *Comptes Rendus Acad. Sci. URSS* **51**, 381 (1946).

"Fluidity" is here defined as the unhindered flow of a molten alloy until it solidifies and is not to be confused with the reciprocal of viscosity in the molten state. The authors poured 80 g of Fe-Si and Fe-P alloys at 1500°C onto an inclined steel angle (4°) and measured the solidified length. Maxima of "fluidity" are found at eutectics and minima with boundary solid solutions.

—J. H. ELLIOTT

Flow Rate Metering Enters a New Technical Era. KERMIT FISCHER, *Instruments* **19**, 331 (June, 1946).

New flow meters for industrial applications, of the variable area type (Rotameter), make use of the fact that pressure and temperature changes affect flow rate mainly by affecting fluid density. By making the float density of the flow meter twice the fluid density, considerable simplifications can be obtained for the measurement of flow of gases as well as of liquids. The variable-area flow rate meter can be made viscosity-sensitive or viscosity-immune, or two combined floats can be used to measure the viscous drag. A continuous record of viscosity changes may thus be obtained.

Industrial applications of this mechanism include measurement of flow of milk and other liquid food products. Wide dimensional ranges may be obtained. Very small instruments are built, as well as units for 20" diameter pipes.

—B. R. ROBERTS

Correction

Mechanism of Fracture of Brittle Solids.* NELSON W. TAYLOR, Research Department, Minnesota Mining & Mfg. Company, St. Paul, Minnesota.

An abstract under the above title appears in the *Rheology Bulletin* for November, 1946. *The present note is to amend the theory by elimination of the heat capacity as an essential factor in the fracture process.* This eliminates the quantity 2θ . It should be replaced by α/k where k is Boltzmann's constant and α is an activation energy, derived from experiment. α depends both on the solid and on its chemical environment. The revised equations read:

$t = 1e^{E\alpha/fkT}/k_0$ and $\log t = -\log k_0 + E\alpha/2.3fkT$ where E is Young's modulus, T the absolute temperature, and k_0 an empirical constant. The second equation has the same form as the Glathart-Preston¹ empirical equation $\log t = -a/m + 1/fm$.

When a solid is placed under a tension, f , an immediate elongation occurs, and after a time, t , there is fracture.

During this period there may be no increase in length, but it is a mistake to assume that no internal rearrangement is occurring. Actually such an atomic rearrangement does take place, and $E\alpha/f$ measures its activation energy requirement. In glass, moisture promotes this re-orientation, in some metals moisture plus oxygen does so; in rubber oxygen is effective, thus lowering the activation energy. Stress enters the $\log t$ equation as the reciprocal, $1/f$, because $r = \lambda_0 E/f$ is the length of the unit prism whose atoms must cooperate to produce the critical elongation λ_0 .

An abstract replacing that which appears in the November *Rheology Bulletin* arrived too late, but is published in the forthcoming *Journal of Colloid Science*, as part of the Bingham Memorial Symposium.

—N. W. TAYLOR

* Presented at the Annual Meeting, Society of Rheology, New York, November 2, 1946.

¹ Glathart and Preston, *J. App. Phys.* **17**, 189 (1946).