Rheological Properties of Some Polymerized Oils[*]

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Introduction

It has long been known that some colloidal or polymeric solutions exhibit the structural viscosity, the thread-forming property or the Weissenberg effect, and various authors have attributed these properties to some "elastic" nature of the liquid.

Some drying oils, e.g., tung oil or linseed oil get rapidly polymerized when heated to about 250°C and become not only viscous but also elastic; they actually turn back elastically when stirred in a vessel; they show a marked thread-forming ability and the liquid-thread springs back elastically when it snaps. These elasto-viscous liquids are still more peculiar in that they show obviously the Weissenberg effect; they "climb up" to a rotating rod which is dipped in them to several centimeters or more. When they undergo still more heating, polymerization proceeds to leave a rubber-like brittle solid.

It is intended, in this study, to obtain some relation between these rheological properties by measuring the dynamic visco-elasticity.

Experimental Procedure

Torsional Oscillation.—(I)[1,2,3].—If a cylinder (moment of inertia I g.·cm²) is suspended from a wire coaxially in the other fixed cylinder which is filled with a visco-elastic liquid (whose resistance R, elastance E), and a torsional oscillation of the amplitude Θ_0

$$\Theta=\Theta_0 e^{i\omega t} \tag{1}$$

is applied at the top of the suspension wire whose torsion constant is k dyn.·cm., the complex amplitude ratio $\boldsymbol{m}$ is given by

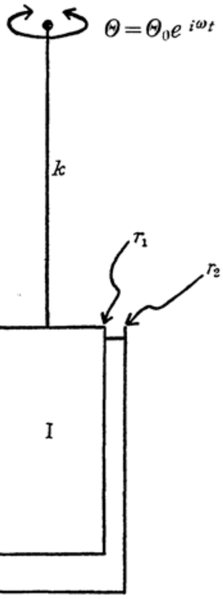

Fig. 1. Top-drive coaxial rheometer.

$$\boldsymbol{m}=\frac{\boldsymbol{A}}{\Theta_0}=\frac{k}{(k+E-\omega^2 I)+i\omega R},$$
$$\theta=\boldsymbol{A}e^{i\omega t}=Ae^{i(\omega t-\phi)} \tag{2}$$

where l, r_1, and r_2 are the length of the immersed portion of the internal cylinder, the radii of the internal and the external cylinder respectively.

So that if the amplitude ratio $m=|\boldsymbol{A}/\Theta_0|=A/\Theta_0$ which is the ratio of the amplitude of the motion θ of the inner cylinder to that of the top end of the suspension wire, and the phase angle $\phi=-\text{Arg}(\boldsymbol{A}/\Theta_0)$ by which the inner cylinder lags the top end of the wire are experimentally observed, the complex quantity $\boldsymbol{m}$ is determined as $\boldsymbol{m}=me^{-i\phi}$, and R and E are known from (2) or from

$$\begin{cases} R=\dfrac{kT\sin\phi}{2\pi m} \\[2ex] E=k\left(\dfrac{\cos\phi}{m}-1\right)+\dfrac{4\pi^2 I}{T^2} \end{cases} \tag{3}$$

where T is the period of oscillation ($\omega=2\pi\nu=2\pi/T$). The dynamic viscosity η and the dynamic rigidity G are calculated by the conventional formula

$$\begin{cases} \eta=\dfrac{R}{4\pi l}\left(\dfrac{1}{r_1^2}-\dfrac{1}{r_2^2}\right) \text{ poises} \\[2ex] G=\dfrac{E}{4\pi l}\left(\dfrac{1}{r_1^2}-\dfrac{1}{r_2^2}\right) \text{ dyn./cm}^2 \end{cases} \tag{4}$$

(II).—The scheme of analysing the visco-elasticity of liquid outlined above is inexact from the theoretical point of view. Recently several authors[4,5,6] have developed the rigorous treatment of this type rheometer. Among them S. Oka[6] proposed a rigorous method of determining the visco-elastic constants by the same top-drive oscillatory device as that described here. According to the theory the complex dynamic rigidity G^* of the liquid studied is obtained as a root of a quadratic equation

$$(B_0+B_1)G^{*2}-\{\mu B_1-(B_2+B_3)\rho\omega^2 r_1^2\}G^* -B_3\mu\rho\omega^2 r_1^2=0 \tag{5}$$

where ρ is the density and μ is a quantity which involves the observable amplitude ratio and the phase difference

$$\mu=\frac{1}{2\pi l r_1^2}\left(I\omega^2-k+\frac{k}{\boldsymbol{m}}\right), \qquad \boldsymbol{m}=me^{-i\phi} \tag{6}$$

and the coefficients B_0, B_1, B_2,......are the quantities relating only to the geometry of the vessel, viz. $x=(r_2-r_1)/r_1$ (which must be less than 1)

$$\begin{cases} B_0=\dfrac{1+(1+x)^2}{2(1+x)} \\[2ex] B_1=\dfrac{x(2+x)}{2(1+x)} \\[2ex] B_2=\dfrac{1}{16}\left\{-4(1+x)\ln(1+x)+4x-6x^2 +2x^3-\dfrac{3x^4}{1+x}\right\} \\[2ex] B_3=\dfrac{1}{16}\left\{4(1+x)\ln(1+x)-4x-2x^2 -2x^3+\dfrac{x^4}{1+x}\right\} \end{cases} \tag{7}$$

If G^* is known from Eq. (5), its real and imaginary

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* Part of this paper was read before the Third Rheological Meeting at Kyoto, Japan, Nov., 1953.

[1] T. Nakagawa, *J. Chem. Soc. Japan* (in Japanese), 72, 759 (1951).

[2] T. Nakagawa, This Bulletin, 24, 191 (1951).

[3] T. Nakagawa, ibid., 25, 93 (1952).

[4] J.G. Oldroyd et al., *Proc. Phys. Soc.* B, 64, 44 (1951); *Quart. Journ. Mech. and Applied Math.*, 4, 271 (1951).

[5] H. Markovitz et al., *Rev. Sci. Instr.*, 23, 430 (1952); *J. Appl. Phys.*, 23, 1070 (1952).

[6] S. Oka et al., *Bull. Kobayashi Inst. Phys. Res.*, 3, 9, 17 (1953) (in Japanese).

* In our unpublished work on the concentrated carbon-black suspensions, a distorted ellipse which had without doubt resulted from the non-linearity of the material itself was obtained (cf. Fig. 6).

parts give the usual dynamic rigidity G and the dynamic viscosity η by the relation

$$G^*=G+i\omega\eta \tag{8}$$

It goes without saying that the rigorous scheme (II) involves the scheme (I) as the limiting case when $\omega\to 0$.

Lissajous' Figure Method for the Determination of m and ϕ.—Whether the approximate analysis (I) or the rigorous treatment (II) is applied, two experimentally determined quantities m and ϕ need to be measured.

When two simple harmonic motions whose directions of oscillation are at right angles to one another are composed, the resulting Lissajous' figure is an ellipse as long as they are of the equal period (cf. Fig. 2). Let the area of the ellipse be (S) and that of the rectangle tangent to the ellipse be $[S]$. Then the next relation is known to hold.

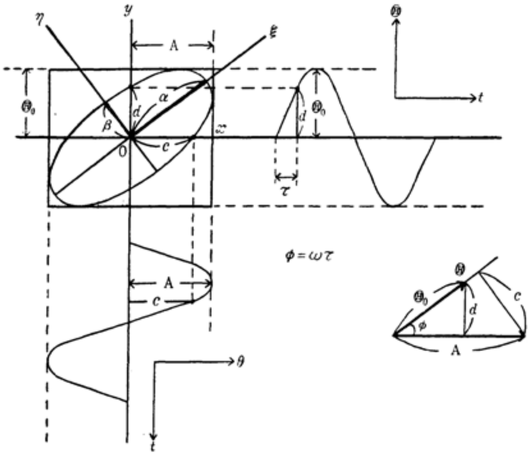

Fig. 2. Lissajous' figure.

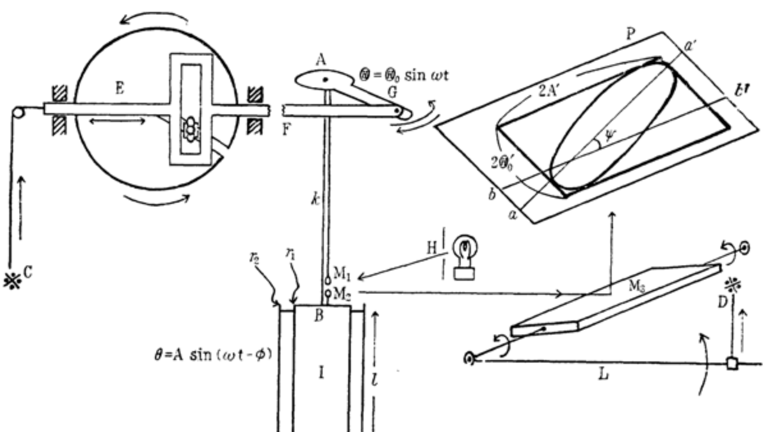

Fig. 3. General assembly of the apparatus.

$$\sin\phi=\frac{4}{\pi}\cdot\frac{(S)}{[S]}=1.273\times\frac{(S)}{[S]} \tag{9}$$

Eq. (9) gives a means of computing the phase angle ϕ by the planimetry. Since ϕ is an angle which satisfies

$$0\leqq\phi\leqq\pi,$$

two values of ϕ are generally obtained from Eq. (9), and they are discriminated as follows from the simple consideration.

1°. When the major axis of the ellipse finds itself in the first quadrant of the θ-Θ plane as in Fig. 2, ϕ is an angle of the first quadrant, that is

$$0\leqq\phi\leqq\frac{\pi}{2}$$

2°. When the major axis is in the second quadrant, ϕ is

$$\frac{\pi}{2}\leqq\phi\leqq\pi$$

and if ϕ' satisfying Eq. (9) and being smaller than $\pi/2$ is determined, the true phase angle ϕ is obtained from $\phi=\pi-\phi'$. From 1° and 2° it is possible to determine to what quadrant the angle ϕ belongs by inspecting the direction of the Lissajous' ellipse.

As is known from Fig. 2, it is also possible to determine ϕ from

$$\sin\phi=c/A=d/\Theta_0$$

The apparatus for practising the above-stated is sketched in Fig. 3. The whole apparatus consists of (1) the driving device and (2) the measuring system.

(1) A driving motor (1/16–1/20 HP, 1500 rpm) and a reduction gear train can produce several low speed rotations ranging from 20 rpm to 0.05 rpm by changing the combination of the gears. Use of a friction table and a movable friction wheel makes it possible to change the motion speed continuously. Rotational motion from the friction desk is changed into the oscillatory motion at the top A of the suspension wire by the eccentric coupling mechanism E and levers F (33 cm.) and G (28.5 cm.). Eccentricity on E is adjustable along its radius between 0 and 5 cm. It is thus possible to obtain the driving oscillation whose period can be changed continuously from about 0.5 seconds to about thirty minutes and whose

amplitude Θ_0 is changeable between 0–10 degrees. It was, of course, confirmed beforehand that the motion is harmonic. The observed Θ-t curve is correctly sinusoidal. (The oscillation of the internal cylinder, on the other hand, is not always harmonic. In the case of Fig. 6, it is known from the Fourier analysis that the contribution from the over-tones reaches several per cent.)

(2) The measuring system is constructed as follows. In order to observe the motion of the internal cylinder, a small galvanometer mirror M_2 is attached to a chuck B by which the lower end of the suspension wire is connected to the inner cylinder; and in front of the mirror M_2 a strip mirror M_3 (3 cm. × 15 cm. × 3 mm.) is placed at the angle 45° to the beam of light from M_2, and is oscillated in phase of the driving motion $\Theta=\Theta_0 \sin \omega t$ by a lever L (10–40 cm., adjustable) which is connected to the oscillation mechanism E by a cord C-D. A Lissajous' figure is thus traced on a photographic plate or paper P; a proper lens system should be used in order to focus the image of a pin-hole H onto the plate P.

Experimental determination of the amplitude ratio m is a little complicated. It is incorrect to compute m directly as A'/Θ_0' from the observed Lissajous' figure, because the transverse deflection A' and the vertical deflection Θ_0' on the plate are of different scale by reason of the different paths as is plain from Fig. 3. One way for evaluating m is to use another mirror M_1 which is attached to the lower end of a long rod connected to the upper end of the suspension wire. As M_1 moves in phase of the driving oscillation $\Theta_0 \sin \omega t$, the Lissajous' figure obtained from M_1 is an oblique line aa′. If then the strip mirror M_3 is kept in position, a horizontal line bb′ is obtained and m is determined as follows

$$m=\frac{A}{\Theta_0}=\frac{A'}{\Theta_0'\cdot f}=\frac{A'}{\Theta_0'}\cdot \tan \phi \tag{11}$$

$$f=\cot \phi \tag{12}$$

where A', Θ_0', and ϕ are the quantities illustrated in Fig. 3. A' and Θ_0' are respectively side lengths of the rectangle in contact with the ellipse which is easily drawn if the base line bb′ is obtained.

It goes without saying that the determination of ϕ is not influenced by the difference of the scale unit of the two axes of coordinates as is understood from Eqs. (9) and (10) and Fig. 2. This is the same in the case of the discrimination 1° and 2°.

Torsion constant k and the moment of inertia I are determined from the observation of the period of the free torsional oscillation[1,2,3]. It should of course be kept in mind that k changes with the length of a wire.

The experimental condition is as follows.

Internal cylinder (made of brass) $\begin{cases} r_1=2.02\text{ cm.,} \\ \text{Height: } 15.0\text{ cm.} \\ I=3.28\times10^3\text{ g.·cm.}^2 \\ \text{(mass: } 1608.6\text{ g.)} \end{cases}$

External cylinder $\begin{cases} r_2=2.51\text{ cm.} \\ \text{Height: } 18.14\text{ cm.} \end{cases}$

Immersion depth l: 14.5–15.0 cm. (to be observed in each run of measurement). End-effect of the internal cylinder was neglected.

Torsion constant k: one wire is properly selected from the piano wires or drill rods tabulated below (Table I) according to the consistency of the liquid being tested. Wire length A-B (about 67 cm, cf. Fig. 3) is measured in each run of the experiment and k is calibrated.

TABLE I

SUSPENSION WIRES USED AND THEIR TORSION CONSTANT

Wire or rod	Length (cm.)	k (dyn.·cm.)
0.30 mm. dia. piano-wire	67.40	9.96×10^2
0.44 mm. dia. piano-wire	67.53	4.44×10^3
0.60 mm. dia. piano-wire	67.29	1.475×10^4
0.80 mm. dia. piano-wire	67.11	4.745×10^4
1.0 mm. dia. drill-rod	67.27	1.258×10^5

Θ_0, amplitude of the driving oscillation: 4–6 degrees of angle. A, amplitude of the forced oscillation of the internal cylinder is designed to be less than 2 degrees of angle, so that the maximum angle of shear γ (cf. Fig. 4) of the sample itself at the wall of the internal cylinder is known to be less than about 11 degrees (cf. Eq. (23)). It may therefore be possible to say that the measurement is done under the condition of small deformation. Temperature control is made by a water thermostat in which the external cup is immersed. For the sake of securing the uniform temperature of the whole measuring vessel, the thermostat is covered by a plate from which only the suspen-

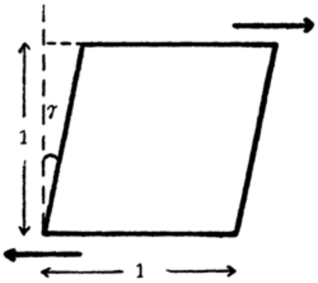

Fig. 4. Shear straining of a unit cube.

TABLE II

SAMPLES TESTED AND THEIR GENERAL PROPERTIES

Sample	Remark
No. 0	The most highly polymerized, sticky, visco-elastic matter with strong thread-forming ability.
No. 1	Moderately polymerized, thread forming, highly viscous liquid.
No. 2	〃
No. 3	Less thread-forming liquid of lower viscosity without elastic nature

sion wire and the mirror M_2 are led outwards. Measurement was done by changing temperature and frequency as follows.

Temperature: 20°–60°C

Period of oscillation: 10–300 seconds

Samples.—Four sample oils listed below (Table II) were made from the thermal polymerization (above 200°C) of linseed oil, and subjected to test.

Results and Discussion

The rigorous scheme of analysis (II) of the observed results, of which description was made in the previous section, was compared with the simple method (I) which has since been used by the author, and it proved that, in such low frequencies as were used here, the difference between the two methods was negligible. In the present study all the data were therefore analysed along the simple scheme (I).

Amplitude Effect.—Our whole procedure of analysing the observed quantities, including the scheme (I) and (II), is based on the assumption that the shear stress S and the shear strain γ satisfy the generalized Hooke's law

$$S=G^*\gamma \tag{13}$$

in which complex rigidity G^* should not depend upon S and γ, while it is a function of frequency. If the linearity relation (13) does not hold, various non-linear behaviors are expected to be observed; e.g., Lissajous' figure will probably deviate from ellipse, or the amplitude dependence of the dynamic visco-elasticity will be notable.

In all the Lissajous' figures obtained, deviation from ellipticity was not perceptible (cf. Fig. 5)*.

Moreover, it is possible to test experimentally the influence of amplitude on the dynamic properties by keeping the period T constant and by changing the eccentricity at the cam E. As to the amplitude effect the experimental result is summarized in Fig. 7 and Fig. 8, in which $\gamma_{\max.r=r_1}$ is the amplitude of the shear strain at the wall of the internal cylinder[7] (cf. Fig. 4)

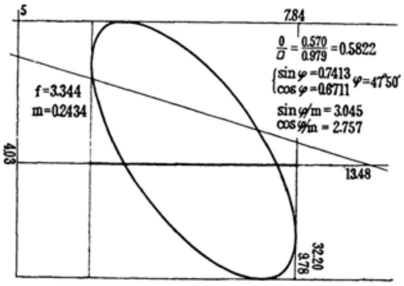

Fig. 5. A sample photograph of the observed Lissajous' figure. (linear visco-elasticity)

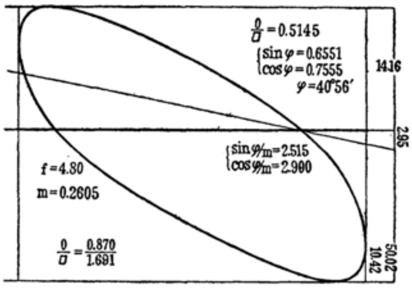

Fig. 6. A sample photograph of the distorted Lissajous' figure. (non-linear visco-elasticity)

$$\gamma_{\max.r=r_1}=\left[\frac{2Ar_1^2r_2^2}{(r_2^2-r_1^2)r^2}\right]_{r=r_1}=\frac{2Ar_2^2}{(r_2^2-r_1^2)}. \tag{14}$$

A is the amplitude of the oscillation of the internal cylinder, and in the present case $\gamma_{\max.r=r_1}=5.67\cdot A$ ($r_1=2.02$ cm., $r_2=2.51$ cm.). $\sin\phi/m$ and $\cos\phi/m$ are the quantities corresponding to viscosity and elasticity respectively, if the period T is kept constant (cf. Eqs. (5), (6)). Three series I, II and III were tested under the following condition using the most sticky varnish No. 0 whose rheological details are shown in Fig 9 (cf. Table III).

TABLE III

EXPERIMENTAL CONDITION ADOPTED FOR THE TEST OF THE AMPLITUDE EFFECT

Oil	Series	Temperature (°C)	Period of oscillation (sec.)
No. 0	I	20	50
	II	30	20
	III	20	20

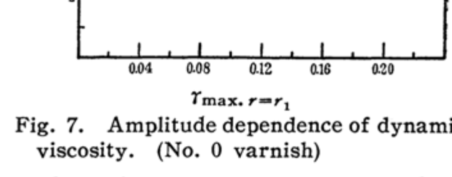

Fig. 7. Amplitude dependence of dynamic viscosity. (No. 0 varnish)

In each series, measurement was done in the order a → b → c → d → e in about six hours, that is, in II the sample first undergoes the largest straining and the other measurements are all carried out within a smaller shear strain; in III, on the other hand, the sample is subjected to a new and larger deformation in each run.

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[7] Th. Schwedoff, *J. de Phys.*, [2], 8, 341 (1889).

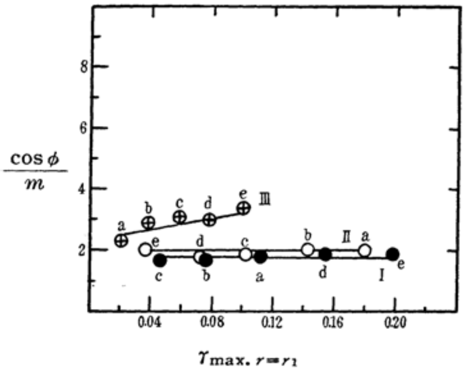

Fig. 8. Amplitude dependence of dynamic elasticity. (No. 0 varnish)

As is known from Figs. 7 and 8, effect of the amplitude on the dynamic viscosity and elasticity is null or slight within the amplitude region tested.

Since the dynamic viscosity is independent on the strain amplitude, it again ought to be independent on the amplitude of the strain velocity from the relations $\theta=Ae^{i\omega t}$, $\dot{\theta}=i\omega A^{i\omega t}$, and

$$\dot{\gamma}_{\max.r=r_1}=\frac{2\omega A r_2^2}{(r_2^2-r_2^2)}. \tag{15}$$

Visco-elasticity of these varnishes is therefore supposed to be nearly linear.

Series III shows a slight non-linearity, especially in the elastic property. Measurement is, in this series, started from the smallest strain, and the sample suffers a new larger deformation in each run of measurement. No. 0 varnish has, as will be seen later, a viscosity of 10^3 poises and an elastic constant of 10^2 dyn./cm² at 20°C of temperature. And such a sticky sol as this generally has a tendency to solidify into a gel-like structure in the course of settling. It will be attributed to this gel-like nature that the elastic constant d(stress)/d(strain) increases with the increase of strain. Since it is expected that this structure breaks down by agitation or straining, the non-linear property will be reduced when the measurement is done in the opposite direction (cf. Series II).

Frequency Dependence.—Frequency dependence of the dynamic viscosity and rigidity was tested under various conditions. The results are summarized in Figs. 9–12. Since it is confirmed in the previous section that no amplitude effect is observed, it will hereafter be possible to consider these data as those of Newtonian and Hookean nature.

Oil No. 0 (Fig. 9) is typically visco-elastic. Dynamic property-frequency curves show a marked dispersion; the dynamic viscosity decreases with the increase of frequency, and the dynamic rigidity increases markedly on the other hand. The lower the temperature, the stronger is the dispersion effect. These materials appear as if they are an elastic mass when handled by a sudden shock; they contract and turn back when freed after being agitated. The external work can, in spite of their fluid nature, be stored up as the elastic potential for a short time. This elastic potential energy gradually dissipates away as a viscous loss; they behave like a viscous fluid when they are treated gently. Varnish No. 0 actually exhibits this property, and Fig. 9 reflects this situation.

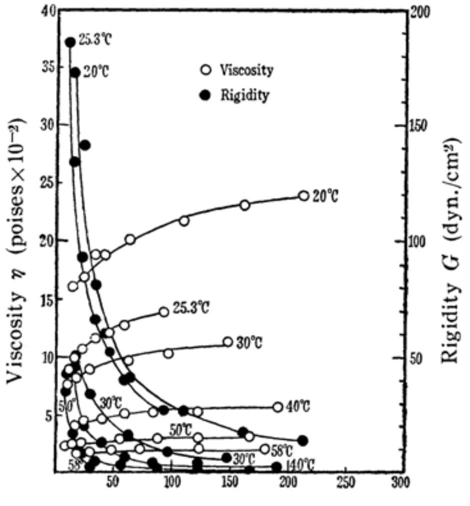

Fig. 9. Dynamic properties of varnish No. 0.

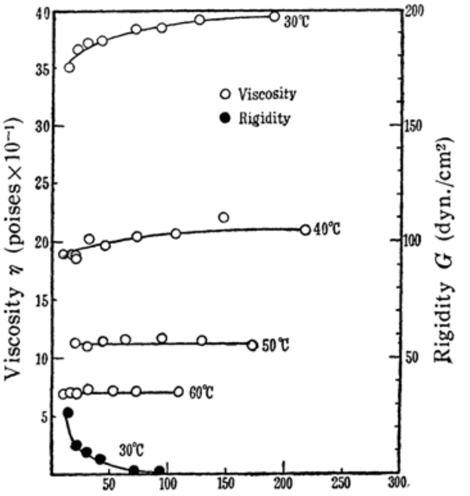

Fig. 10. Dynamic properties of varnish No. 1.

The varnish No. 0 is used as an adding agent in the printing ink manufacturing industry in order to control and improve the "printability" of inks. The peculiar rheological property of the oil No. 0 is supposed to have some influence upon printability.

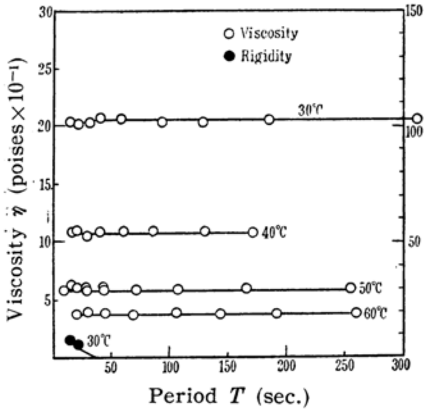

Fig. 11. Dynamic properties of varnish No. 2.

Varnishes No. 1 (Fig. 10) and No. 2 (Fig. 11) are viscous liquids, showing a slight elastic nature at a low temperature and high frequency; towards a very gentle handling they behave as simply viscous liquids. The oil No. 3 (Fig. 12) is a purely viscous liquid and shows no elasticity at any temperature and frequency tested. It is of course possible for this varnish to appear as elastic at a lower temperature and higher frequency than this. In fact, it may occur that these oils act not as simply viscous matter but as a visco-elastic medium between the rollers of a high-speed printing machine when they are used as a vehicle.

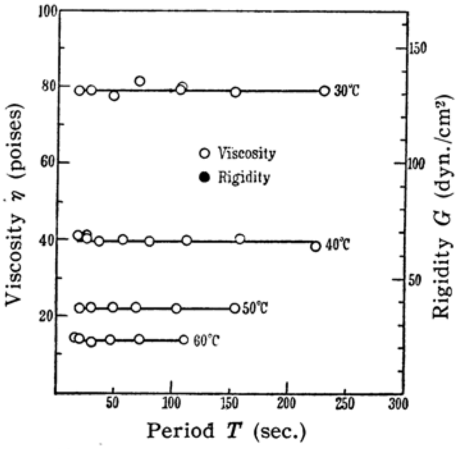

Fig. 12. Dynamic properties of varnish No. 3.

Returning to the varnish No. 0 (Fig. 9), the visco-elastic behavior at 25°C is a little peculiar. The elasticity at 25°C has a value nearly equal to that at 20°C, though the viscosity at 25°C is decreased in an ordinary manner. The fact is that the measurement at 25°C was carried out on the sample which was left and settled in the measuring vessel (between the inner and the external cylinder) for about a month. It is therefore possible for the oil to cause a gel-formation as was stated above, and this gel-formation may be ascribed to a weak network structure of the solute molecules formed in the sol by the secondary forces. On account of the elasticity from this network, the observed dynamic rigidity does not so markedly decrease by raising temperature from 20°C to 25°C. The dynamic viscosity, on the other hand, is supposed to come, in its considerable part, from the flow of the low molecular weight solvent molecules spread in such structure, and therefore it decreases considerably when temperature is raised.

Temperature Dependence.—When the frequency independent viscosities of the varnishes No. 1—No. 3 are plotted against the reciprocal absolute temperature, Fig. 13 is

$$\eta = Ae^{B/T} = Ae^{\Delta E/RT} \qquad (16)$$

obtained. These curves conform to the Andrade formula T is here the absolute temperature (not the period), and R the gas constant. The constants A and B or the apparent activation energy ΔE are calculated and tabulated in Table IV.

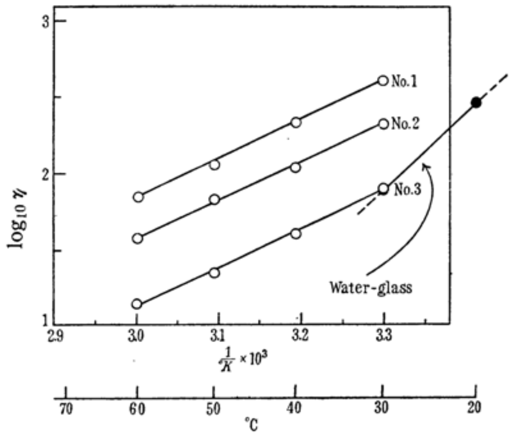

Fig. 13. Temperature dependence of viscosity.

TABLE IV

CONSTANTS OF THE ANDRADE FORMULA

$\eta = Ae^{B/T} = Ae^{\Delta E/RT}$

Oil	A	$B \times 10^{-3}$	ΔE (kcal.)
No. 1	2.33×10^{-6}	5.72	11.4
No. 2	1.03×10^{-6}	5.78	11.5
No. 3	2.12×10^{-7}	5.94	11.8

The value of B (or ΔE) is, when compared with those of the other materials[2], larger than those of the low molecular weight liquids (mercury, toluene, water), smaller than those of the highly associated liquids (glass, rosin, tar-pitch, glucose melt), and of the same order as those of asphalts and the glucose-melt at higher temperature. This is possible from the fact that these oils are polymerized to a certain degree.

It is noted that the apparent activation energy ΔE for the viscous flow has almost the same value as that of others in spite of the great difference of the absolute value of viscosity. One of the possible interpretations may be that at a certain stage of the thermal polymerization the growth of the polymer molecule itself does not occur, but the number of polymers simply increases.

Weissenberg Effect

In 1946–1948 K. Weissenberg[8,9,10] noted that some elastic liquids show a peculiar property to climb up to a rotating rod or ascend in a rotating pipe, and the various authors[8–14] have since developed the theoretical and experimental study of this phenomenon.

Our varnishes No. 0–No. 2, as seen in Figs. 9–11, show evidently an elastic property even in the very low frequencies like 0.1 cycles per second. It is therefore expected that the Weissenberg effect may appear even when a rod is slowly rotated in these oils. The result is illustrated in Fig. 14. A rod of 10 mm. dia., sunk to about 3 cm. depth, is rotated at the rate of two and three rotations per second, and the behavior of the liquids was observed. The experiment was made at the room temperature of about 20°C; the visco-elastic constants of the varnishes at 20°C can be estimated from Figs. 9–13.

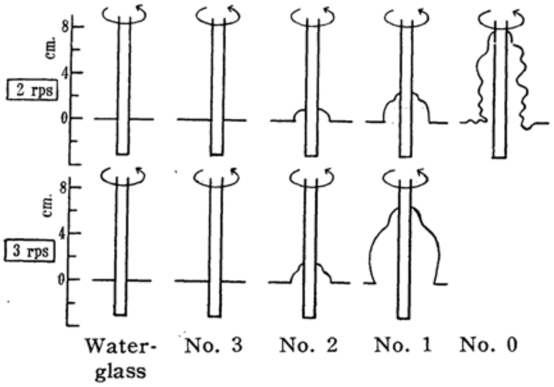

Fig. 14. Weissenberg effect.

In the varnish No. 3 no effect was observed at this temperature and revolution rate, and this situation corresponds to Fig. 12 where no elastic property is shown. The oils No. 2 and No. 1 evidently exhibit the Weissenberg effect, and the varnish No. 1 which is more sticky than No. 2 climbs up higher than No. 2. The effect is stronger when the rotation rate is increased; this is natural from the fact that the Weissenberg effect is concerned with the relaxation phenomena of liquid. In the oil No. 0 which is the most sticky and elastic of all, the effect is so strong that even at 2 rps the oil climbs up to 8 cm. or more in a very irregular manner like a towel which is squeezed.

For the sake of comparison, we tested a water-glass sample, which was previously confirmed to lack an elastic nature in these low frequencies[3]; and no effect was observed. From the falling-sphere method, its viscosity was estimated to be 290 poises at 20°C and it is the similar value to that of No. 3 or No. 2 at 20°C. It is therefore argued that the liquid is too fluid to climb up and that if the viscosity were increased the Weissenberg effect might appear. Water-glass was then cooled to 5°C of temperature in order to make it be more than 1000 poises of viscosity which is the same order of that of No. 1 varnish. The effect was still nothing. As has been discussed above, viscosity alone is not enough to cause the Weissenberg effect however viscous a liquid may be.

Summary

(1) A method for measuring the dynamic visco-elasticity of liquid by the torsional oscillation is described.

(2) The visco-elastic oils produced by the thermal polymerization of linseed oil to various degrees were studied by this rheometer.

(3) The amplitude effect of the dynamic properties was negligible; so that these materials are supposed to be linear in their visco-elastic behavior.

(4) The dynamic visco-elasticity of these oils was measured in the very low frequency region (period: 10 seconds–5 minutes).

(5) The temperature dependence of the dynamic viscosity was discussed.

(6) Weissenberg effect of these oils was tested in a qualitative way.

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