Korea-Australia Rheology Journal
Vol. 14, No. 2, June 2002 pp. 87-90

Note on the beginnings of sinusoidal testing methods

R.I. Tanner*
School of Aerospace, Mechanical and Mechatronic Engineering, University of Sydney, NSW. 2006 Australia

(Received March 6, 2002; final revision received May 9, 2002)

Abstract

The measurement of the storage and loss moduli (G' and G" respectively) of materials as functions of frequency is now commonplace and is of wide utility. Yet it is not easy to trace the history of such measurements, and so this article discusses the genesis of this important experimental technique. We find that the technique grew out of a parallel technique for dielectric measurements (ca. 1900) and was developed in the mid-1930s by Philippoff and others. Important breakthroughs due to digital circuitry have occurred only in the last 20 years or so.

The application of a small-amplitude sinusoidal-in-time strain (or stress) to a material sample and the measurement of the stress (or strain) is now one of the most common methods of rheological characterization. Generally, shear or elongational deformation can be used, depending essentially on the consistency of the sample. In this article we trace the early history of these methods of testing.

If one were concerned only with elastic solids or purely viscous fluids the sinusoidal test would only be of modest interest, since measurements of elastic moduli and Newtonian viscosities, which are the quantities required for these materials, can be made in simpler ways. Thus we are led to viscoelastic materials as candidates for sinusoidal testing.

It seems likely that Weber (1835; 1841) was the first to publish on viscoelastic response. He was interested in using silk threads for electrical galvanometer suspensions, and noticed that the elasticity of silk fibres in tension was imperfect. On loading, he found an initial elastic response, then a slow creep. Removal of the load led to an immediate contraction, and then a slow recovery to the original length. This behaviour, surprising to Weber, is characteristic of viscoelasticity, rather than plasticity, where the original length is not recovered. Weber also stated what would happen in a relaxation test, and found nonlinear responses in some cases.

Some more work by R. Kohlrausch (1847) and Friedrich Kohlrausch, his son (1863; 1866; 1876) addressed the torsional response of fibres for galvanometer work. In his 1863 paper the latter investigated the damping of the fibre and looked at the decrement of the motion in an oscillating system. No attempt to use a constant magnitude sinusoidal test was made.

We can also mention the substantial contribution of Schofield and Scott Blair (1932) who studied the viscoelasticity of bread dough in the linear regime; however, they used creep measurements and did not do sinusoidal testing.

Theoretical aspects of small-strain experiments are well-known (see Tanner and Walters (2000)). We shall note the viscosity of metals concept of Kelvin (1865) seeking to explain the damping of vibrations in metallic objects, the early theories of Maxwell (1867) and Meyer (1874), and the complete formulation of linear viscoelasticity theory by Boltzmann (1874). Several of these papers dealt with the decay of vibrations in materials, which is related, but not too closely, to our subject of constant amplitude, sinusoidal testing. Clearly, the rate of decay of vibration is of prime importance in these nearly elastic materials, and tests are complicated by the inertia of the instrument and/or sample.

Around 1850 the electric telegraph began to use rapidly varying currents. There arose therefore a considerable interest in the dielectric properties of insulators, with a view to avoiding losses in telegraph cables. When a dielectric is subjected to a transient electric field, a current flows. If the dielectric is imperfect, energy losses occur. Mathematically, the relations between electric field and current are exactly analogous to those between stress and rate of deformation, and the same concepts (for example, loss tangent, tan δ) apply. So, much earlier than subsequent constant frequency viscoelastic mechanical measurements, measurements in dielectric materials became of interest. For example, Wagner (1915) produced many curves for dielectric losses introducing the concept of the loss tangent (tan δ) in rubbers over a frequency range of 350 to 4800 Hz and a temperature range of -5°C to 45°C. These measurements also considered the effect of electric field strength, clearly a nonlinear effect. Thus, by analogy, the physical and mathematical basis for mechanical measurements with a constant amplitude and frequency sinusoidal

---

*Corresponding author: rit@mech.eng.usyd.edu.au

input were well-known by about 1912.

It was not until 1933 that Wladimir Philippoff, in his Berlin PhD dissertation, applied sinusoidal testing to viscoelastic liquids. The Berlin-Dahlem Kaiser Wilhelm Institute für Chimie contained at that time Weissenberg and Rabinowitsch as colleagues for Eisenschitz and Philippoff. Eisenschitz and Philippoff (1933) wrote a preliminary note on their technique, which they thought was new, giving it the title, "A new method for the determination of the mechanical material constants of colloids".

A fuller account of the work was given by Philippoff (1934), following his PhD thesis completion. The paper begins with a discussion of the response of a (linear) Maxwell fluid to a sinusoidal input in complex ($e^{j\omega t}$) form, where ω is the applied frequency in radians/sec, and shows that the time constant can be estimated from the complex viscosity magnitude, and hence the shear elastic modulus could be inferred. All of this mathematics was known from electrical alternating current theory, and Philippoff had been trained as an electrical engineer in Berlin. He opted for a resonance method of measurement, rejecting damped natural vibrations as unsuitable. The arrangement devised is shown in Fig. 1, taken from the Philippoff (1934) article. The essential parts of the machine are the steel band S, which could be set to various tensions and consequently the mechanical vibration system had a variable natural frequency. Attached to the beam is a rod which contacts the sample which was held in a tube (K). The electromagnets W_1 and W_2 were used to keep the beam vibrating and the current in these magnets could be measured, and the force driving the sample could thus be deduced. Note the pentode valve R in the circuit; this device was still relatively novel. The amplitude of vibration was estimated using a contact controlled by a micrometer screw M; F is a telephone which enabled contact between the screw and the vibrating beam to be judged. The frequency was judged subjectively by sound in comparison with the pitch of a tuned pipe. Thus given the force, the amplitude and the frequency the response of the fluid could be found. Note that the phase angle could not be directly measured, and that calibration with a fluid of known viscosity was necessary. Fig. 2, from Philippoff (1934) shows the general appearance of the machine; it weighed about 15 kg.

Frequencies between about 30 and 630 Hz could be investigated, and a series of tests (Fig. 3, from Philippoff (1934)) showed remarkably good results for a series of Newtonian fluids and one non-Newtonian solution (cellulite in dioxane); all of these results are for the dynamic viscosity only (real part of the complex viscosity). The second part of the paper (beginning on page 900) describes some applications to non-Newtonian systems.

This work was a remarkable experimental investigation at the time, and I believe it deserves respect, especially since it appears to be quite novel. However, it was clearly a clumsy instrument and measurement of G' was not possible.

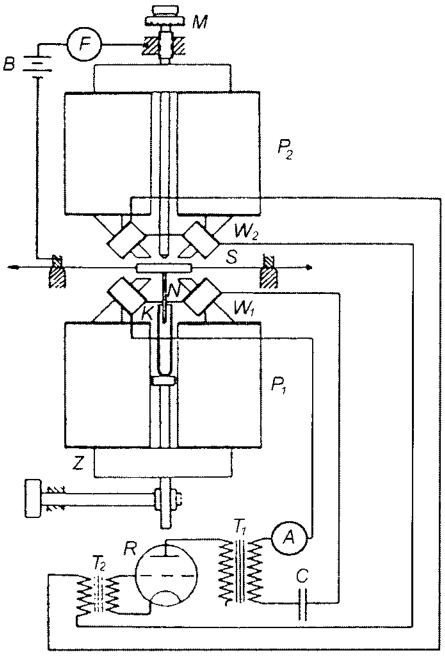

Fig. 1. (Grundsätzliche Anordnung) Basic organization of Philippoff's rheometer. S is the steel vibrating beam to which the needle N was driven in the tube K containing the sample. The screw M enabled the amplitude of vibration to be judged via contact with the beam S; noise in the telephone F indicated contact. (From Physik. Z.)

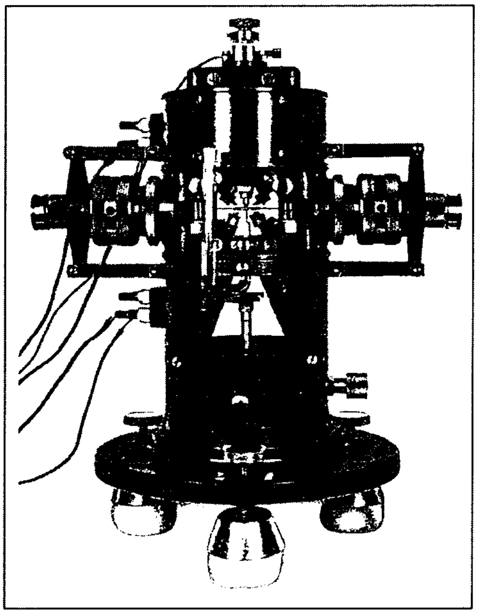

Fig. 2. (Aussenansicht des Apparates) This view of the apparatus shows it a sturdy instrument weighing about 15 kg. The transverse screws served to tighten the vibrating band. (From Physik. Z.)

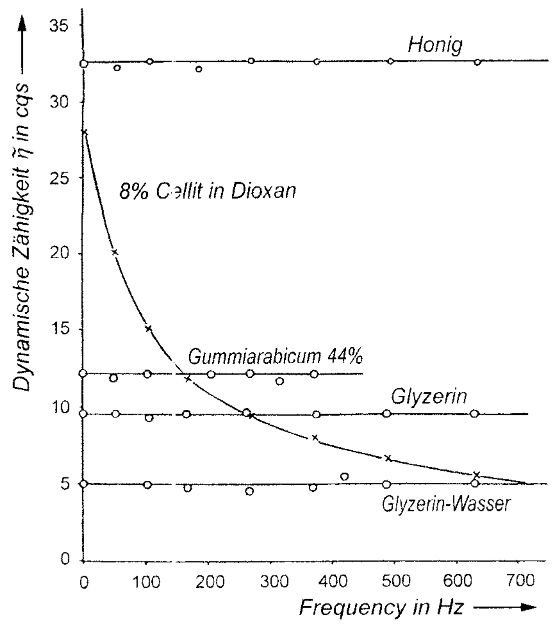

Fig. 3. Some dynamical viscosity results for various Newtonian fluids and one non-Newtonian fluid (cellulite in dioxane). The results are remarkable and extend over a wide frequency range. (Linearity of response was also tested, and was excellent.) (From Physik. Z.)

As a further short note about Wladimir Philippoff, he was born in Peterhof, Russia in 1907 and he moved to Germany in 1924. He got his Dr. Eng (Electrical Engineering) in 1934 in Berlin. From 1932-45 he worked at the Kaiser Wilhelm Institute of Chemistry, and his subsequent career was devoted to rheology. In 1942 his excellent book, "Viskosität der Kolloide", was translated into English by the American authorities as part of the war effort. After the end of the World War he worked for the Allied Military Government for three years, and in 1948 he moved permanently to the U.S.A. In 1959 he joined Exxon Research (Esso in those days) at Linden, New Jersey, where he was until his retirement. He investigated normal stresses, birefringence and other rheological topics; he was awarded the Bingham Medal in 1962.

In 1941 John Douglass Ferry (born 1912) began his lifelong interest in sinusoidal testing as a probe for molecular structure. He began (Ferry, 1941) with transverse wave propagation in a sample and obtained some rigidity values but later (Smith, *et al.*, 1949) he abandoned this process in favour of an electromagnetic transducer drive. Although the machines used could be tuned to resonance, as in Philippoffs work, Smith et al preferred to work just away from the resonant frequency, where it was easier to measure G' as well as G''. Their method of measurement was to find the changes in the electrical impedance as seen from the driving coil using classical alternating current bridge methods. Clearly this technique was vastly more accurate than Philippoffs measurement techniques, and set a new standard for other workers. They were able to cover the frequency range 30 Hz 200 Hz in one machine, and 200 - 500 Hz in a second one.

John Ferry set a new standard for the subject. (Tanner and Walters, 1998). He was born on May 4th, 1912 in Dawson, Yukon Territory, Canada, where his father was a mining engineer. He obtained his Bachelors degree from Stanford University in 1932 and his PhD from the same University in 1935. His doctoral work involved a protracted stay at the University of London in England.

Ferry moved to Harvard University in 1936 and he spent nine years there in various capacities. In 1946 he was appointed Assistant Professor in the Chemistry Department of the University of Wisconsin at Madison. His association with the University of Wisconsin, Madison was to be a long and highly successful one. He very quickly moved through the ranks and he had been appointed to a full Professorship by 1947. Later, between 1959 and 1967, Ferry became Chairman of the Chemistry Department and, from 1973 until his official retirement in 1982, he held the position of Farrington Daniels Research Professor.

During an illustrious academic career, Ferry received numerous honours. For example, in 1953 he was awarded the Bingham medal by the Society of Rheology: he was Honorary President of the International Congress on Rheology in Kyoto, Japan, in 1958; he was President of the American Society of Rheology between 1961 and 1963 and, from 1961 to 1963; he also served as Chairman of the International Committee on Rheology.

Madison became an international centre of excellence in rheology and, during his tenure there, Ferry was to supervise over 50 research students, two of whom (T.L. Smith and D.J. Plazek) were later to receive the Bingham medal of the Society of Rheology.

In 1943 Herbert Leaderman (1943 a,b) investigated the response of textile fibres. His report (1943a) is still remarkable for its excellent literature survey. It does not encompass sinusoidal testing, but some excursion into nonlinear creep response was made.

Mason (1947) demonstrated the use of piezo-electric crystals as drivers of the motion. This paper was a virtuoso performance from Bell Laboratories and it covered frequencies in the kilohertz range, again obviously relying on electrical impedance change measurements.

Further notable papers are those of Andrews, Hofman-Bang and Tobolsky (1948) and Cox and Merz (1958). Andrews *et al.* (1948) contributed to linear creep and relaxation studies, without doing sinusoidal tests. Cox and Merz (1958) noticed that for some materials the magnitude of the complex viscosity $|\eta^*|$, as a function of ω in sinusoidal shearing, followed the same curve as the steady shear viscosity η as a function of the steady shear rate. Although it does not always hold, this (empirical) Cox-Merz rule is often useful; the simple fluid requirement $|\eta^*(0)| = \eta(0)$ always holds and is independent of the Cox-Merz rule.

Following these papers a good deal of ingenuity was deployed to cover the range of materials of interest and the relevant frequency ranges, especially by John Ferrys group at the University of Wisconsin in Madison. Eventually, he produced his classic book entitled "Viscoelastic Properties of Polymers". This volume makes it unnecessary to continue a detailed history of the subject; it gives a very full coverage of the literature. The first edition was produced in 1961, with a third edition in 1980.

In the early 1960s it was possible to derive phase angles by using analog computers to correlate the input and output signals (Simmons, 1968) but at low frequencies (< 1 Hz) the results were not easy to obtain accurately. By 1970 one could do digital correlations (Tanner and Williams, 1971) which gave a great improvement in accuracy especially at low frequencies (< 3 Hz). However, in this paper the data were recorded and then analysed in a separate computer which was a time-consuming procedure. See also Ferry (1980) for developments in this period. From the mid-1970s one begins to see the appearance of stress-controlled equipment, where a known torque can be applied electromagnetically, and the output to be measured is the displacement. For some systems, for example those with a yield stress, this arrangement gives an advantage over constant amplitude measurements, because the stress level is capped. More recently (>1980) machines have become more and more computer-controlled and amenable to automatic data reduction, graphical presentation, and report printing. Most of these programs use the ideas presented above, with the need for an external computer to run the software but often the operation manuals do not describe the algorithms in detail. The new output flexibility contributes to the popularity of sinusoidal testing.

Thus the main changes in the subject since the third edition of Ferry perhaps lie in the exponential improvement in measurements and recording methods, progressing from analogue computers in the 1960s to digital methods in the 1980s. This has enabled much better phase angle resolution and recording of data, so that the measurement of G' and G" is now routine.

References

Andrews, R. D., N. Hofman-Bang and A. V. Tobolsky, 1948, Elastoviscous Properties of Polyisobutylene. I. Relaxation of Stress in Whole Polymer of Different Molecular Weights at Elevated Temperatures, *J. Poly. Sci.* **3**, 669-692.

Boltzmann, L., 1874, Zur Theorie der elastichen Nachwirkung, *Sitz. Kgl. Akad. Wiss. Wien (Math-Naturwiss Klasse)* **70**, 275-306.

Cox, W. P. and E. H. Merz, 1958, Correlation of Dynamic and Steady Flow Viscosities, *J. Poly. Sci.* **28**, 619-622.

Eisenschitz, R. and W. Philippoff, 1933, Eine neue Methode zur Bestimmung mechanischer Materialkonstanten von Kolloiden, *Die Naturwissenschaften* **28**, 527-528.

Ferry, J. D., 1941, Studies of the Mechanical Properties of Substances of High Molecular Weight. I. A Photoelastic Method for Study of Transverse Vibrations in Gels, *Rev. Sci. Instr.* **12**, 79-82.

Ferry, J. D., 1961-1980, Viscoelastic Properties of Polymers, Wiley, New York.

Kohlrausch, R., 1847, Nachtrag über die elastische Nachwirkung beim Cocon und Glasladen, *Pogg Ann. Physik* (3) **72**, 353 -405.

Kohlrausch, F. W., 1863, Ueber die elastische Nachwirkung bei der Torsion, *Pogg. Ann. Physik* (4) **119**, 337-368.

Kohlrausch, F. W., 1866, Beiträge zur Kenntnis der elastischen Nachwirkung, *Pogg. Ann. Phys.* (5) **128**, 1-19, 207-227, 399-419.

Kohlrausch, F. W., 1876, Experimental-Untersuching über die elastische Nachwirkung bei der Torsion, Ausdehnung, und Beigung, *Pogg. Ann. Physik* (6) **158**, 337-375.

Leaderman, H., 1943a, Elastic and creep properties of filamentous materials and other high polymers, Textile Foundation, Washington D.C.

Leaderman, H., 1943b, Creep and creep recovery in plasticized polyvinyl chloride, *Ind. Eng. Chem.* **35**, 374-378.

Mason, W. P., 1947, Measurement of the viscosity and shear elasticity of liquids by means of a torsionally vibrating crystal, *Trans. Am. Soc. Mech. Engrs.* **69**, 359-370.

Maxwell, J. C., 1867, On the dynamical theory of gases, *Phil. Trans. Roy. Soc. Lond.* **157**, 49-88.

Meyer, O. E., 1874, Theorie der elastichen Nachwirkung, *Pogg. Ann. Physik* (6) **151**, 108-119.

Philippoff, W., 1934, *Dynamische Untersuchungen an Kolloiden Systemen*, Parts I and II **35**, 884-900, 900 910.

Schofield, R. K. and G. W. Scott Blair, 1932, Relationship between viscosity, elasticity and plastic strength of soft materials as illustrated by some mechanical tests, **A138**, 707-718

Simmons, J. M., 1968, Dynamic modulus of polyisobutylene solutions in superposed steady shear flow, *Rheol. Acta* **7**, 184-188.

Smith, T. L., J. D. Ferry and F. W. Schremp. 1949, Measurements of the mechanical properties of polymer solutions by electromagnetic transducers, *J. Appl. Phys.* **20**, 144-153.

Tanner, R. I. and G. Williams, 1971 On the orthogonal superposition of simple shearing and small-strain oscillatory motions, *Rheol. Acta* **10**, 528-538.

Tanner, R. I. and K. Walters, 1998, Rheology: An Historical Perspective, Elsevier, Amsterdam.

(Kelvin) Thomson, W., 1865, On the elasticity and viscosity of metals, *Proc. Roy. Soc.* **14**, 289-297.

Wagner, K. W., 1915, Dielektrische Eigenschaften von verschiedenen Isolierstoffen, *Elektrotechnische Zeitschrift* **12**, 120-123, 135-137, 163-165.

Weber, W., 1835, Ueber die Elasticität der Seidenfäden, *Ann. Phys. Chem.* **34**, 247-257.

Weber, W., 1841, Ueber die Elasticität fester Körper, *Ann. Phys. Chem.* **54**, 1-18.