VISCOSITY OF MOLTEN LITHIUM-6 AND LITHIUM-7

by

Nguyen Tu Ban

AN ABSTRACT

Submitted to the School for Advanced Graduate Studies of
Michigan State University of Agriculture and
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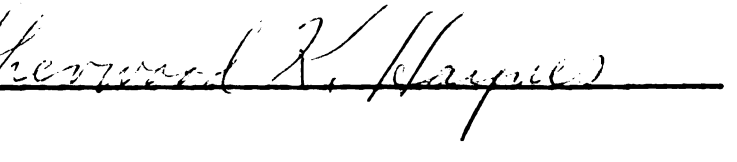
Approved 

ABSTRACT

This study is an extension of a program exploiting isotopic mass as a probe for liquid-state and solid-state investigations to non-equilibrium phenomena of liquids.

The viscosity of molten lithium-6 and lithium-7 was measured by observing the damping of a freely oscillating sphere filled with molten lithium. The viscosity coefficient was found by treating the damping of a slowly oscillating sphere containing a viscous fluid as a problem in the laminar flow of incompressible fluid.

The temperature dependence of the viscosity of lithium-6 was found to be in general agreement with a law stating that the viscosity varies exponentially in the reciprocal of the absolute temperature. This law is also predicted by some existing theories of viscosity. The extrapolated value for the absolute viscosity of lithium-6 at the melting point (180.4 ^{o}C) was found to be 4.50 mp.

The effect of isotopic mass on viscosity was studied by measuring the viscosity coefficient of lithium-7 at a given temperature and compare it with that for lithium-6 at the same temperature. The ratio of the viscosity of lithium-7 to that of lithium-6 at 236.8 ^{o}C was found to be $(4.186\pm0.080/3.831\pm0.016) = 1.093\pm0.025$, in general agreement with the theoretical prediction that it is equal to the

square root ratio of the atomic mass of lithium-7 to that of lithium-6, that is to $(7.018/6.017)^{\frac{1}{2}} = 1.080$.

On the assumption that the viscosity coefficient for a mixture of isotopes of an element, consisting predominantly of one component, is a linear function of the number composition, the absolute viscosity of lithium-natural (92.5% lithium-7, 7.5% lithium-6) at 236.8 ^{o}C is estimated to be 4.16 mp. This value is 24% lower than the corresponding value obtained by Andrade and Dobbs using lithium-natural. A detailed examination of their paper led to the conclusion that their values are significantly in error. This fact is of technical interest.

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To Qúy, Bích

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INTRODUCTION

It was realized by the earliest investigators of isotopes that isotopic mass is of potential value as a means for investigating the behavior of matter in the aggregate[4]. In studies of isotopes of the same element, the atomic mass can be changed while the atomic force field is kept the same. The dependence of physical properties on mass is usually simple, whereas the dependence on force field is very complicated. In the early days, however, low enrichments of isotopes obtainable precluded exploitation of isotopic mass in bulk studies. It was not until the establishment of atomic energy projects that an array of elements became available in substantial amounts[13].

A. Choice ot Topic

This study grew out of a program, supported by the Atomic Energy Commission, exploiting isotopic mass as a probe for liquid-state and solid-state investigations. Earlier studies in this program had shown that such studies were feasible, but most of the topics investigated depended only on higher order effects of isotopic mass[36]. Moreover, these topics had been for the most part deliberately chosen in fields where the phenomena were relatively well understood. It was decided to extend the exploitation of isotopic mass in bulk studies to imperfectly understood properties,

particularly properties involving non-equilibrium phenomena of liquids. Here viscosity suggests itself as a likely candidate.

Technically, viscosity is important because most time dependent processes in liquids - fluid flow, heat transfer, mixing, etc... - depend on it; scientifically, because its study may point the way to a better understanding of the structure and behavior of liquids. Moreover, the existing theories predict that viscosity depends on mass in the first order.

B. Choice of Material

For greatest clarity in interpreting the results, a monatomic substance, that is an element, should be studied first. It is true that the liquids for which the theory is simplest are those for which the alterations in electronic distribution are least when the isolated molecules condense to form a liquid, i.e., the inert gases. Indeed, the properties of helium-3 and helium-4 have been studied extensively. Little attention has been given to neon-20 and neon-22, argon-36 and argon-40, and the others, because of their unavailability in bulk, let alone their small relative mass difference. But there are other reasons for extending the study beyond this type of substance. The experimental difficulties at the low temperatures required for liquefaction, and the narrow temperature range in which the liquid is stable at ordinary pressures, are two shortcomings of the

inert gases. More telling perhaps, is the restricted nature of the interactions in these materials. With metals, one can attain interactions of more general interest, and at the same time retain simplicity, particularly if a simple metal is chosen. Atomically speaking, the simplest metal, at least at ordinary pressures, is lithium, with its low atomic number (Z=3), and its simple structure (bcc) in the solid state. Fortunately, there are two stable isotopes, lithium-6 (7.5% abundance ratio) and lithium-7 (92.5%), occurring in nature with a large relative mass difference (14%). Finally, both isotopes are available in quantity at high enrichment (up to 99.9%) at moderate cost[14].

C. Choice of Method of Measurement

In the theory of hydrodynamics when one goes from ideal fluids to real fluids, it is necessary to introduce a dependence of stresses on relative motion of portions of the fluid. The simplest useful assumption is that the "drag" between adjacent portions of the fluid is proportional to the velocity gradient at the region of interest, that is, for a specific component

$$\text{Lim.}\ \frac{\Delta F}{\Delta S} = \tau_{xy} = \eta \frac{\partial u}{\partial x}$$

where the viscosity η is taken to be independent of the gradient. Although in principle one needs only to produce and measure a velocity gradient and a viscous stress (that is, a force and an area), and thereby use the defining equation directly, in practice it is almost always necessary

to use an integrated form of the equation resulting from the application of the principles of hydrodynamics.

In choosing a method of measurement, it is important to find an experimental system which can be represented adequately by a theoretical model simple enough to be analyzed mathematically. Beyond this, the method must be adapted to the magnitude of the viscosity, the chemical reactivity of the substance, the amount available, the temperature range contemplated, and the precision desired. This last depends on the precision possible in the measurements of dimensions and time, and if it is to be an absolute measurement, on the adequacy of the theoretical representation and solution.

The well-known methods which are susceptible to mathematical treatment so that absolute viscosity may be obtained, may be grouped as follows:

1. The measurement of the resistance offered to a moving body in contact with the viscous fluid. Popular methods are the falling sphere method (Stokes*, 1845), the rising bubble of gas method (Marshall, 1917).

2. The measurement of the rate of flow of a viscous fluid. The single most important representative is the transpiration or Poiseuille method (1842).

3. Methods in which neither the flow nor the resistance

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* Only the name of the investigator who first used the method experimentally is cited. The reader is referred to Barr[5], Bingham[8], and Hatschek[25] for detailed development of these methods and of others not mentioned, and for references to the original papers.

to flow are measured. To cite a few:

a. Concentric cylinders; the outside cylinder is rotated at constant angular velocity, and the torque exerted upon the inner coaxial cylinder which is immersed in the viscous fluid is measured (Stokes, 1845).

b. A horizontal disk supported at its center by a wire and oscillating around the wire as an axis (Coulomb, 1801).

c. An oscillating solid sphere immersed in the viscous fluid and supported by a bifilar suspension (König, 1885).

d. A hollow sphere filled with liquid and oscillating around its vertical axis (Helmholtz and Piotrowski, 1860).

Due to the extreme corrosion of lithium in its molten state on most materials[28,35], its readiness to react with non-inert atmospheres, and the small amounts of the isotopes available, it is necessary to confine a small volume of the sample in contact with non-reacting materials. These requirements eliminate practically all the methods mentioned except those involving observation of the damping forces of a confined liquid. Furthermore, the experimental system must be of a design such that the confined liquid can be maintained at constant temperature with relative ease over the temperature range contemplated.

The method which seems to fulfill all the requirements is that of Helmholtz and Piotrowski[26], to which Barr[5] refers as being "at least of historical interest". The original

paper is certainly not such as to encourage others to use the method*. The calculations given there are very complicated and troublesome, the results obtained, very unsatisfactory. However, subsequent investigators (for example, Ladenburg[32], 1908, Verschaffelt[41], 1915) have modified and refined this method, and shown that it can be a satisfactory one. More recently, Andrade and his group adapted the method to measure successfully the viscosities of liquid sodium and potassium[1], liquid natural lithium, rubidium and caesium[2], liquid indium and tin[16], and liquid magnesium and calcium[17]. Andrade and Chiong[1] also presented a method of calculation, adapted from that used by Verschaffelt[41] for calculating the viscosity of liquefied gases from the damping of the oscillation of a sphere immersed in the liquid, which simplified the calculation greatly. In view of all these considerations, it was decided to adapt the method of observing the damping of free oscillation of a hollow sphere filled with lithium.

D. Statement of Problem

It is proposed to measure the absolute viscosity of molten lithium-6 and lithium-7 as a function of temperature, using a freely oscillating sphere filled with molten lithium,

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* "On voit combien ces relations sont compliquées et quelle incertitude le calcul des expériences laissera planner sur la valeur de μ/δ, qu'elles sont destinées à donner." Brillouin. Viscosité des liquides, part 1, p. 102, 1907.

and to compare the experimental results with the prediction of current theories.

THEORETICAL CONSIDERATIONS ON THE VISCOSITY OF LIQUIDS

Although the theory of the viscosity of dilute gases is highly satisfactory, that of liquids remains confused. This state of affairs is understandable in view of the continuous interactions between the molecules of liquids, as contrasted with the momentary interactions between the molecules of gases. A critical survey of existing theories of liquid viscosity has just been given by Bondi[9]. Other recent papers which include a treatment of viscosity as one of the important transport processes in liquids have been written by Eisenschitz[18], by Longuet-Higgins[33], and by Cole[15]. Much of the following discussion is based on these papers.

From dimensional analysis alone, it is possible to draw useful conclusions about the dependence of the viscosity coefficient η of a hard-sphere fluid on temperature T and molecular mass m, the variables of primary interest in the present study. If $[m]$, $[l]$, $[t]$, and $[T]$, represent the dimensions of mass, length, time and temperature respectively, then the viscosity coefficient has the dimensions

$$[\eta] = [m]\,[l]^{-1}\,[t]^{-1} \qquad (1)$$

Since the temperature may be taken to have the dimensions $[k]^{-1}\,[m]\,[l]^{2}\,[t]^{-2}$, where k is Boltzmann's constant, it is possible to eliminate $[t]$ and thereby to obtain

$$[\eta] = [m]^{\frac{1}{2}}\,[l]^{-2}\,[k]^{\frac{1}{2}}\,[T]^{\frac{1}{2}}$$

It follows then that

$$\eta = \text{constant} \cdot (mkT)^{\frac{1}{2}}/\ell^2 \qquad (2)$$

and that if ℓ does not depend strongly on m or T, the viscosity coefficient increases as the square root of m and T.

This prediction is indeed verified very well experimentally, and moreover appears in the first approximation as a result of the molecular theory of transport properties of dilute gases.

For liquids, in contrast, this happy state does not obtain. Let us consider first the effect of temperature. It is found experimentally that almost always the viscosity coefficient decreases and moreover decreases very rapidly with increasing temperature. It is apparent then that the characteristic length ℓ must be changing with temperature. Now measurements of liquid viscosity are normally made at constant pressure, and hence the mean intermolecular separation is indeed changing. It might be thought then that measurements at constant volume would give agreement for the so-predicted dependence of η on T, with ℓ considered as constant. Indeed, as has been demonstrated by Bridgman[12] with non-polar liquids, the dependence of viscosity on temperature is much less at constant volume than at constant pressure. Nevertheless, the simple law for dilute gases is still far from being fulfilled. In the theories as well as the experiments, no simple and satisfactory relation has been derived for the temperature dependence. Some of the expressions that have been proposed will be shown later in

this section.

We will now turn to the effect of isotopic mass. When substances that differ in chemical composition as well as in molecular mass are studied, the difference in molecular force fields precludes a study of the effect of mass independent of simultaneous effects of changes in molecular diameter and intermolecular spacing. However, substances that differ only in isotopic mass usually have only very slight differences in their atomic geometry, and with these substances it would be possible to study the effect of mass in isolation from the effect of length. The experimental data are scanty - in the literature there seem to be reported[38] measurements on only the pairs H_2O-D_2O, H_2-D_2, CH_4-CD_4, (apart from He^3-He^4, where completely new phenomena come into play). Only for light and heavy methane does the ratio of viscosities follow the square root of the mass dependence[37]. Since the shape of the molecules studied is not simple, it is perhaps not surprising that the simple law is not followed. Accordingly, to minimize the complications arising from shape, spherical molecules are desirable, and the present study was undertaken with the isotopes of lithium, lithium-6 and lithium-7. The experimental results will be given in a later section of the thesis. So far as the predictions of molecular theory are concerned, there is insufficient agreement among the different workers to allow a brief statement to be made.

We shall now summarize the various approaches to the

theory of viscosity of liquids:

1. The modern point of view anchors the theory of viscosity of liquids solidly in the statistical theory of transport processes in liquids. The central problem here is the calculation of the molecular distribution functions, and particularly the pair distribution function. When a pair distribution function, appropriate to a steady non-uniform state has been found, the flow of momentum or energy necessary to establish uniform conditions in a liquid can be calculated, and the transport coefficients are derived by averaging the contributions made by a representative pair to these fluxes.

Born and Green[10], and independently Kirkwood[30] have given methods for doing this. Their theories are very much similar in spirit, but differ in the approximations used to reduce distribution functions of higher order to those of lower order, thus enabling the circumvention of the nearly impossible task of evaluating the complete partition function.

Owing to the proximity of the molecules to each other in a liquid, the distribution function is primarily determined by the magnitude of the intermolecular forces. Born and Green's theory relates viscosity to the intermolecular potential $\phi(r)$ by considering the following physical pictures[23]:

The exchange of momentum in liquids depends on the continuous action of the intermolecular forces, in contrast with the transport of momentum in gases which depends largely on the motion of the molecules themselves. Hence, if two

adjacent layers in the liquid move with different velocities, each will tend to drag the other in such a manner as to dissipate the state of relative motion, in the absence of sustaining external stresses. As a result of this drag, the molecular structure of the fluid is deformed. A study of the deformation of the molecular structure will enable one to establish the quantitative relations which determine the coefficient of viscosity.

Born and Green[11] obtained for the viscosity coefficient the expression

$$\eta = \frac{1}{30}\int \nu\, \phi'(r)\, r^3\, d\vec{r} - \frac{1}{15} m \int \omega\, v^4\, d\vec{v} \qquad (3)$$

where ν is a measure of the deformation of the molecular distribution from its equilibrium configuration, and ω a function of the magnitude of the velocity v, n, and T. By assuming that the second term in equation (3), due to thermal motion, can be neglected for the liquid state, and using for $\phi(r)$ the Lennard-Jones 6-12 potential, they derived as a "preliminary" formula for viscosity:

$$\eta \sim \left(\frac{\pi^2}{315}\right)\left(42\pi\right)^{1/2}\left(\frac{r_0}{r_1}\right)^5 \frac{m\nu_0}{r_0}\; e^{-\phi(r_1)/kT} \qquad (4)$$

where r_0 is the equilibrium distance in the molecular field, r_1, distance of nearest neighbors, and ν_0, the frequency of vibration of the molecules about the equilibrium point r_0.

Kirkwood, Buff and Green[31], by introducing a principle of irreversibility which plays for liquids a role equivalent to the hypothesis of molecular chaos in the theory of dilute

gases, obtained for the coefficient of viscosity an exceedingly complex relation which lends itself well to numerical calculation. It would serve little purpose to present it here, since the mass and temperature dependence of this coefficient is not at all apparent. On the other hand, Kirkwood obtained for the first time an expression for the volume viscosity* of liquids, a result hardly accessible to elementary theories.

2. The liquid is considered to behave like a dense gas. The elementary kinetic theory is accordingly adapted without any appreciable change in the fundamental concepts or combined with the elementary notions concerning the Brownian movement. The physical fact here is that in a gas, energy and momentum are transported by diffusion, that is, by bodily movement of individual molecules through relatively large distances, and that the exchange of energy and momentum occurs almost entirely through bimolecular collisions, which are relatively infrequent events for any given molecule. In liquids, on the other hand, a given molecule spends the greater part of the time interacting strongly with the other molecules, and the very idea of a collision loses its significance. A more serious criticism to this approach is perhaps the fact that it is doomed to failure in explaining the

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* The definition of volume viscosity in terms of hydrodynamics is given in Lamb, Hydrodynamics, (Dover Publications, 1945), Article 326.

temperature effect on viscosity of liquids. It may be recalled that it is empirically established that for all except the superfluids, liquid viscosity decreases with increasing temperature in contrast to the viscosity of gases which increases as the temperature rises.

3. The elementary kinetic theory is applied to the interstices between the liquid molecules. This is the hole theory of viscous flow of liquids, first proposed by Fürth[22]. He stated:

> "The holes are considered as the counterparts of the clusters in a dense gas or vapour. They are formed by the action of the irregular thermal movement (or by the statistical fluctuations), and they are destroyed again by the same process; they interact with each other, and they perform a kind of Brownian motion.... Since in our theory the holes in a liquid are the counterparts of the clusters in a gas, we may expect that the phenomenon of viscous flow in a liquid can be explained in a similar way, namely by assuming that the irregular "Brownian" motion of the holes produces a transfer of momentum between adjacent layers in a moving liquid."

From this crude model*, Fürth obtained for the viscosity coefficient:

$$\eta = 0.915 \frac{RT}{V} (m/\sigma)^{\frac{1}{2}} e^{A/RT} \qquad (5)$$

where A is the so-called work function, σ , the surface tension of the liquid, and R, the universal gas constant.

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* "This inner inconsistency of the theory makes it clear from the beginning that it can provide only a rough picture of the real behaviour of matter in the liquid state, ..." Fürth[22].

4. Self diffusion is considered to be a simple phenomenon and accessible to an elementary treatment, from which conclusions regarding viscous flow are drawn:

a. by using Einstein's formula for the diffusion coefficient

$$D = kT / 6\pi a \eta \qquad (6)$$

Frenkel[20] assumes that the equilibrium position of the atoms in a liquid is not absolutely permanent. After performing a more or less large number of oscillations about the same equilibrium position during a certain time τ, each atom can jump into a new equilibrium position, the jump δ being of the same order of magnitude as the mean distance between adjacent atoms. This step by step wandering or self diffusion can be described as a sequence of two processes: the "evaporation" of the atom from its initial equilibrium position into an intermediate one, and the "condensation" from this intermediate position, into a new equilibrium position. Frenkel gives for the diffusion coefficient

$$D = \frac{\delta^2}{6\tau_o} e^{-W/kT} \qquad (7)$$

but gives no theory from which to calculate τ_o or W. Following his earlier argument[21], δ/τ_o would be the thermal velocity $(kT/2\pi m)^{\frac{1}{2}}$ giving for the viscosity

$$\eta = \frac{(2m\pi kT)^{\frac{1}{2}}}{\pi a \delta} e^{W/kT} \qquad (8)$$

where a is the particle radius, and W, the activation energy.

b. by regarding the movement of a molecule in viscous flow as a diffusion under external constraint.

Eyring[19] pictured the process of diffusion as continually taking place in the liquid, with or without an applied force tending to cause viscous flow. The application of such a force simply tends to make a preferred direction in which the molecules move.

> "Viscous flow is assumed to take place by the activated jumping of an aggregate composed of one or more molecules from an initial normal configuration to a second normal configuration[29]."

Eyring does not specify the mechanism of momentum transfer, he simply assumes that the mechanism is such that the theory of absolute reaction rates is applicable. Then the coefficients of viscosity and diffusion are related by the equation

$$kT / D\eta = a_1 \cdot a_2 / a_3 \qquad (9)$$

where a_1, a_2, and a_3 are lengths of molecular dimensions. Kincaid, Eyring and Stearn[29] give for the viscosity coefficient the expression

$$\eta = 7.71 \times 10^{-4} \frac{M^{\frac{1}{2}} T}{V^{\frac{1}{3}} E_v} e^{E_v/nRT} e^{-E_s/R} \qquad (10)$$

where E_v, measured in calories per mole, is the energy of activation for viscous flow. The n appearing in the exponential term was chosen to give the correct temperature coefficient of viscosity by computing values for η neglecting the entropy of activation E_s which is small by comparison with experiment.

For this reason, Eyring's theory does not lend itself to fully absolute calculations, and in Onsager's words, the method "lends itself to a facile interchange of deduction and induction."

5. A mechanism of momentum transfer between contiguous layers is postulated.

It is assumed that the arrangement of molecules in the liquid state is very similar to that in the solid state, and that consequently the average intermolecular force which acts on a given molecule is, on the whole, very little different from the average intermolecular force in the solid state. The essential difference between the two states is not the magnitude of the intermolecular force, under which the molecule vibrates, but the amplitude of motion.

To derive an expression for the coefficient of viscosity of simple liquids in the neighborhood of the freezing point, Andrade appealed to the following physical picture[3]:

a. There is for a liquid molecule, a vibration about a slowly displaced equilibrium position, with a frequency which is the same as that of the molecule in the solid state;

b. At every extreme libration, the molecules of one layer come into contact with those of an adjacent layer. At this contact they will, in general, enter into a temporary union, the duration of which does not exceed the very brief time necessary for the molecules to acquire a common velocity of translation. This sharing of velocities between molecules in adjacent layers introduces viscous forces.

Andrade then postulated that the rate of transfer of momentum between contiguous layers in laminar flow was proportional to the difference in velocity between them, hence

$$\frac{4}{3}\frac{1}{\sigma^2}\nu m \frac{dv}{dz}\sigma = \eta \frac{dv}{dz}$$

or

$$\eta = \frac{4}{3}\frac{\nu m}{\sigma} \qquad (11)$$

where σ is the average distance between the centers of molecules, and ν, the frequency of vibration of the molecules about their slowly diffusing equilibrium position. Using Lindemann's theory of melting, ν can be written as

$$\nu = K\,(T_m / AV^{\frac{2}{3}})^{\frac{1}{2}} \qquad (12)$$

where K is about 3.1×10^{12}, which gives for the viscosity coefficient the relation

$$\eta = 5.7 \times 10^{-4}\,(AT_m)^{\frac{1}{2}}\,V^{-\frac{2}{3}} \qquad (13)$$

where A is the atomic weight, V, the volume of a gram-atom at the melting point T_m, and $V = A/\rho$.

By the assumptions made in its deduction, formula (13) can only be expected to apply where

(1) the molecules are very simple and symmetrical and do not themselves consist of a vibrating system of atoms, that is, to monomolecular liquids;

(2) the atomic arrangement is not much influenced

by melting, that is, to substances that have a simple cubic structure in the solid crystal and which do not experience a large change of volume on melting.

To account for the observed temperature behavior of viscosity, Andrade took into account the effect of expansion on the average atomic distances and the effect on the potential energy, and derived

$$\eta\, v^{1/3} = B\, e^{c/vT} \qquad (14)$$

where v is the specific volume, and B and c, two arbitrary constants.

In so far as the above account is a fair summary of the status of the theory of liquid viscosity, one is led to the conclusion that, at present, there is no statistical theory of transport processes in liquids which is at the same time absolute, practically useful, and free from mathematical uncertainty. On the other hand, while the elementary theories give the temperature effect as observed, the various interpretations given to this effect can hardly be reconciled, and it is difficult to decide between them. However, many of these theories have also the feature that the viscosity coefficient varies as the square root of the isotopic mass, when the other factors are nearly constant as they certainly must be for isotopes of the same element at higher temperatures. Hence measurement of the viscosity of separated

isotopes would further provide a critical check on their adequacy.

THEORY OF THE EXPERIMENT

The damping of a slowly oscillating sphere containing a viscous fluid can be treated as a problem in the laminar flow of incompressible fluids. A very brief summary of the method, originally treated by Helmholtz[26], later modified by Verschaffelt[41], Andrade[1], and others, will be given.

The general equation of motion for a viscous fluid may be written in vector notation as

$$\frac{\partial \vec{v}}{\partial t} + \vec{v}\cdot\nabla\vec{v} = G - \frac{1}{\rho}\nabla p - \nu \nabla\times\nabla\times\vec{v} \qquad (15)$$

where $\vec{v}$ is the velocity, G the body forces (gravitational force in the present case), ρ the density, p the pressure, and $\nu \equiv \eta/\rho$ the kinematic viscosity, where η is the viscosity coefficient.

We make the following assumptions:

1. the liquid moves in concentric spherical shells about the axis of rotation Z, each infinitesimally-thick shell behaving as if it were solid;

2. the components of velocity in the X-Y plane are of the form:

$$v_x = -y\,\psi(r,t) \qquad v_y = x\,\psi(r,t)$$

where r is the distance from the center of the sphere.

Furthermore assume

$$\psi(r,t) = \phi(r)\, e^{\alpha t}$$

where $$\alpha \equiv -\beta + i\gamma$$

with $\beta \equiv \delta/T$ and $\gamma \equiv 2\pi/T$

where δ is the logarithmic decrement and T is the period of oscillation of the sphere filled with the sample in its molten state.

Then equation (15) reduces to

$$\frac{d^2\phi}{dr^2} + \frac{4}{r}\frac{d\phi}{dr} = b^2\phi \qquad (16)$$

where $b \equiv \alpha/\gamma$.

The solution of this equation is

$$\phi(r) = r^{-3}\left[A(br - 1)\, e^{br} + B(br + 1)\, e^{-br}\right] \qquad (17)$$

where A and B are constants to be determined by the boundary conditions.

On the other hand, the equation of motion of the oscillating sphere is given by

$$I\frac{d^2\theta}{dt^2} = -M\theta - P - P' \qquad (18)$$

where θ is the angular displacement, M the restoring force of the suspension per unit angle, P the viscous force due to the sample in its molten state, and P' the external damping, due mainly to the internal friction of the suspension wires.

On the further assumptions that:

1. the external damping is responsible for only a small contribution to the total damping;

2. there is no slip between the liquid and the container wall, in accordance with all modern experiments;

equations (17) and (18) combine to give:

$$(2 - q)\eta - (\pi\rho/T)^{\frac{1}{2}}\, aR\eta^{\frac{1}{2}} - \frac{3I\delta}{8T\pi R^3}\left[\frac{T^2}{T_o^2} + 1\right] = 0 \qquad (19)$$

where $q \equiv \dfrac{gR - 1}{(gR - 1)^2 + (hR)^2}$,

$a \equiv 1 - \delta/4\pi + \delta^2/32\pi^2$, neglecting higher terms,

$g \equiv (\pi\rho/T\eta)^{\frac{1}{2}} \cdot a$,

$h \equiv (\pi\rho/T\eta)^{\frac{1}{2}} \cdot (1 + \delta/4\pi + \delta^2/32\pi^2)$,

T_o = the period of oscillation of the sphere filled with the sample in its solid state,

R = the inner radius of the sphere,

and I = the moment of inertia of the suspended system.

The viscosity coefficient is then approximated very closely by

$$\eta = \frac{a^2R^2}{4(2 - q)^2}\,\frac{\pi\rho}{T}\left\{1 - (1 - \mu)^{\frac{1}{2}}\right\}^2 \qquad (20)$$

where $\mu = \dfrac{3(2 - q)\, I\delta}{2\pi^2 a^2 \rho R^5}\left[\dfrac{T^2}{T_o^2} + 1\right]$

the measured quantities being I, R, T_o, T, δ, and ρ.

APPARATUS AND METHOD

A. Description of the Apparatus

1. Oscillating System

The principal part of the apparatus is the oscillating system shown in Figure 1. The suspension head consisted of a brass cylinder A fitting into a cylindrical brass block B rigidly fastened to a levelling table C, the three levelling screws being of the type used in the standard transit theodolite. Two rubber O-rings were used for sealing the cylinder A to the cylindrical block B for vacuum tightness while still allowing relative rotation. The suspended system was set in oscillation by turning the lever L through a small angle. In order to keep this angle within suitable limits, a stop S, on which was attached a vernier scale with one-tenth degree least count, was mounted on the block B. The stop S could be adjusted to bring the suspended system to any desired equilibrium position.

While the suspension head was being machined in the lathe, the end carrying the lever L was faced so that the circular table D to which the lever L was attached was accurately perpendicular to its axis. Thus adjustment of the circular table D to be truly horizontal, by means of a precision Wyler level with sensitivity 0.013 m/m per 1000 m/m, automatically set the suspension head with its axis vertical.

Scale: 1/4

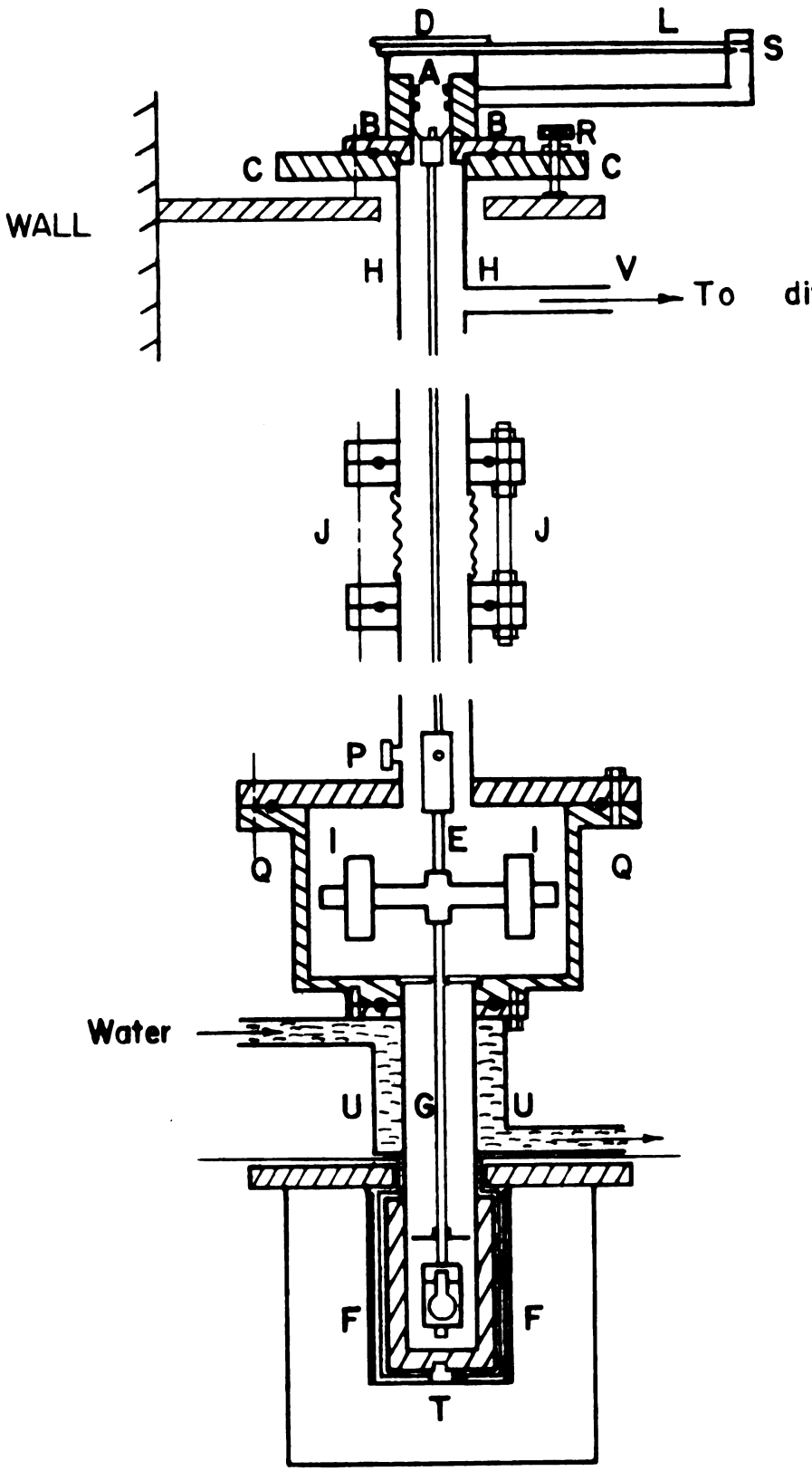

FIGURE 1

GENERAL VIEW OF APPARATUS

A threaded hole was chased into the other end of the suspension, to which a special clamp holding the suspension wires could be screwed. When the suspension wire clamp was in place, the two wires which are held fixed at opposite ends of a diameter, were to be equidistant from the axis of the suspension head. To ensure this, each half of the clamp shown in Figure 2, was machined separately from a length of cylindrical brass stock. Two holes of diameter slightly less than 0.0048 inch were bored in the brass stock, the holes being equidistant from the axis of the cylindrical brass stock, which was then milled to obtain exactly a half-cylinder. In this way, it was possible to obtain two perfectly-matched halves for the cylindrical plug. It may be mentioned that the wires were only clamped tightly along a length of one-eighth inch, thus insuring that they were held rigidly up to the point where they leave the clamp. The necessity for clamping a suspension wire correctly has been emphasized by Bearden[6].

The bifilar suspension was of 40 s.w.g. tungsten wire*. The virtues of tungsten have been stressed by Heyl and Chrzanowski[27]. To obtain pure rotation of the sphere, it is critical that the center of mass of the suspended system be on the truly vertical axis of the suspension head.

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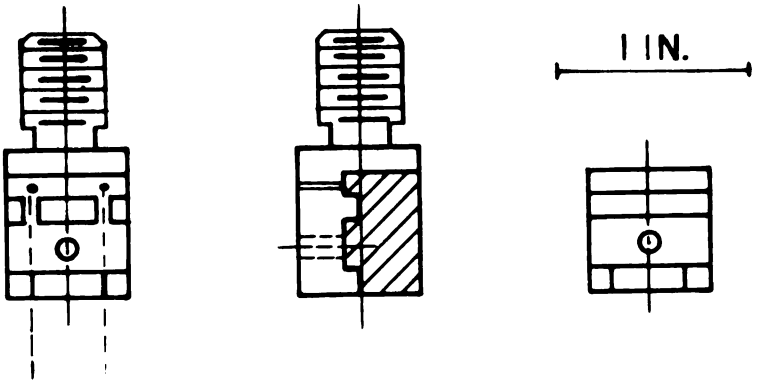
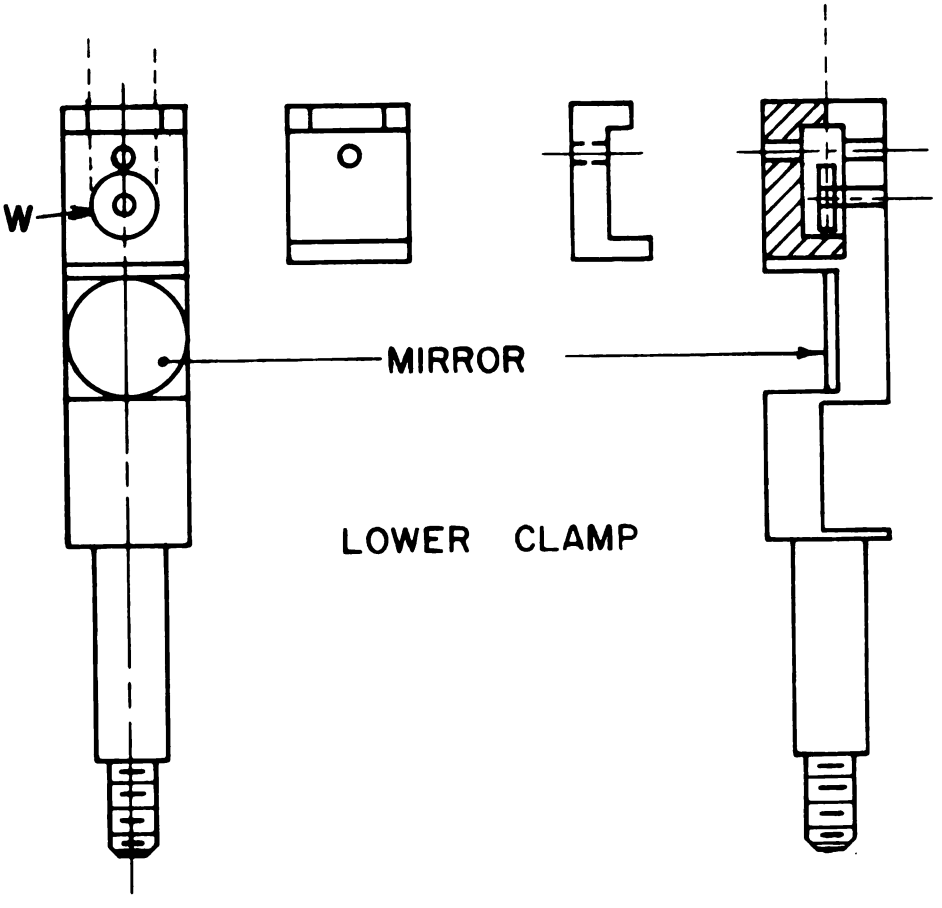
* The tungsten wire was obtained from the General Electric Company through the fine cooperation of Mr. Dallas T. Hurd of the Advanced Development Engineering Section. It was kept straight during shipping, in length of 2.5 meters, through special arrangement with Mr. Hurd.



UPPER CLAMP



SUSPENSION WIRE CLAMPS

FIGURE 2

To ensure this, the bifilar suspension was made of a single wire looped around a pulley. The half loop of the wire carried a small wheel W provided with a fine circumferential groove, the wheel itself being part of the lower clamp shown in Figure 2. This clamp was very similar to the one described above. The wheel was added to allow the suspended system to settle with its center of mass in the vertical axis of the suspension head, and to equalize the tension in the bifilar suspension. After the suspension was in position, and the center of mass of the whole suspended system had settled, the two wires were clamped tightly at the lower end by screwing the two half-cylinders together. In this way, the suspension wires were held rigidly by the lower clamp jaws without the wheel having to bear any weight.

The inertia rod I was connected to the lower clamp through a brass shaft E. The sphere* itself was connected to this shaft through a long thin-walled stainless steel tubing G. Details of the brass shaft E and its connection to the stainless steel tubing G are shown in Figure 3.

2. Vacuum Enclosure

The vacuum enclosure consisted of a brass tube H, furnished with two glass windows, one shown at P, the other parallel to the plane of the paper, a cylindrical brass box Q housing the inertia rod I, and another brass tube, the

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* The term "sphere" is used throughout this thesis to denote the sample holder.

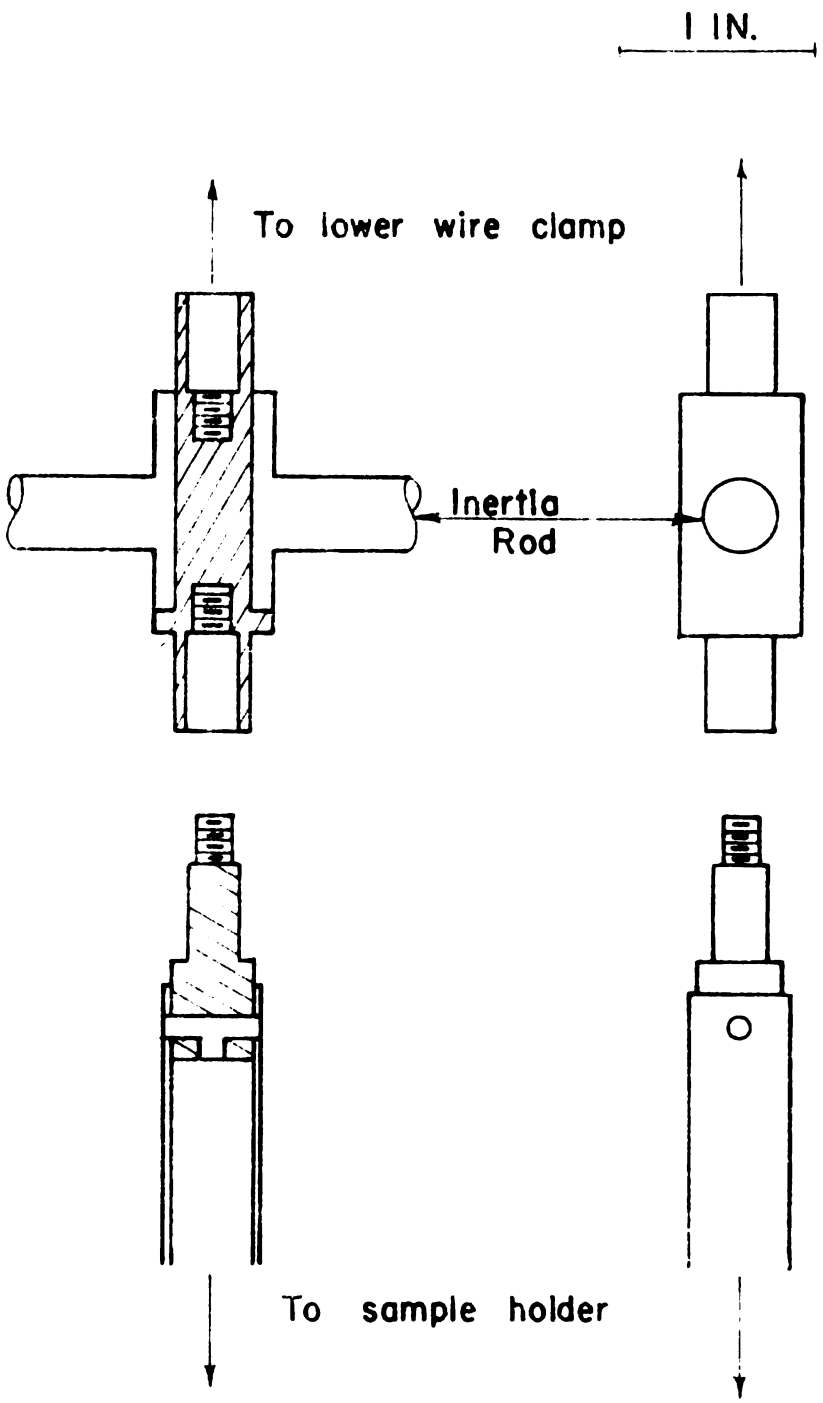

FIGURE 3

DETAILS OF CONNECTING SHAFT E

lower end of which carried a stainless steel tube, to which was soldered a copper cylinder. The stainless steel tube minimized heat conduction to the upper part of the apparatus. The copper cylinder, machined from a length of copper stock, had one-half inch thick wall and base. The upper brass tube H was actually two tubes connected by a flexible copper bellows J. This arrangement allowed adjustment of the lower part of the vacuum enclosure so that the sphere hung centrally in the copper cylinder. Evacuation was made through the side tube V. The vacuum was maintained at 0.01 micron or less during an experiment.

3. Furnace and Temperature Control

For maintaining the sphere at constant temperature, a non-inductively wound electric furnace F, deep enough to heat the whole length of the copper cylinder, was used. An iron shield was placed between the copper cylinder and the wall of the furnace to minimize spurious damping of the oscillating liquid metal due to stray magnetic field from the heating coils. A water-cooling jacket U, separated from the top of the furnace by a thick insulating board, was used to keep the inertia rod and the suspension wires from getting warm.

The temperature of the furnace as indicated by a copper-constantan thermocouple T shown in Figure 1 was measured on a 10-mv Speedomax recorder, and controlled by a Series 60, 2-Action Duration Adjusting Type, Leeds and Northrup con-

trol unit. To maintain the thermocouple voltage within the range of the recorder at high temperature, a simple potentiometer was constructed. The settings on the potentiometer were calibrated against a K-3 Leeds and Northrup Universal Potentiometer (stated accuracy 0.02% of voltage). The control unit held the temperature at any setting very constant, as indicated by a perfect straight line on the recorder.

The whole apparatus, except the furnace, was supported by the three levelling screws R, resting on small brass cups which were placed on a thick wooden platform. The wooden platform was in turn bolted to the platform of a scaffold, a welded construction of 2 x 2 inches steel angles. The scaffold, shown in Figure 4, measuring nine feet, was bolted solidly to the wall of the building.

4. Sample Holder

The sample holder or sphere was made in two halves, from lengths of stainless steel type 446 1 1/4-inch rod. Each half of the sphere was first roughed out using a one-inch ball end mill, then hand ground to a hemisphere with a one-inch steel ball and jeweler's rouge to obtain a satin-smooth finish. The upper hemisphere was provided with a neck through which the molten lithium could be poured. The two hemispheres were fastened together inside a tight-fitting cylindrical sleeve, and hard silver soldered as shown in Figure 5. It was critical that the two hemispheres did not overlap where they joined.

A matching cap was made for each sphere using gold O-ring

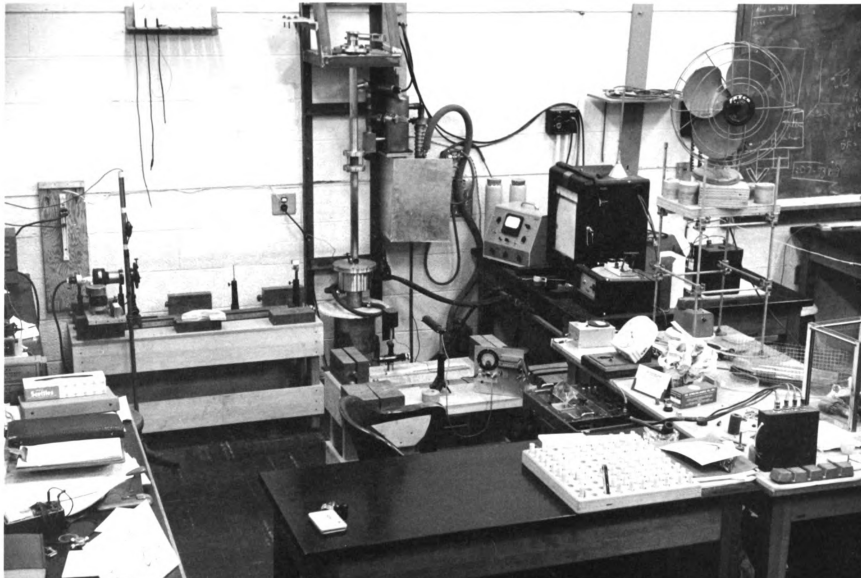

Figure 4

General View of Experimental Setup

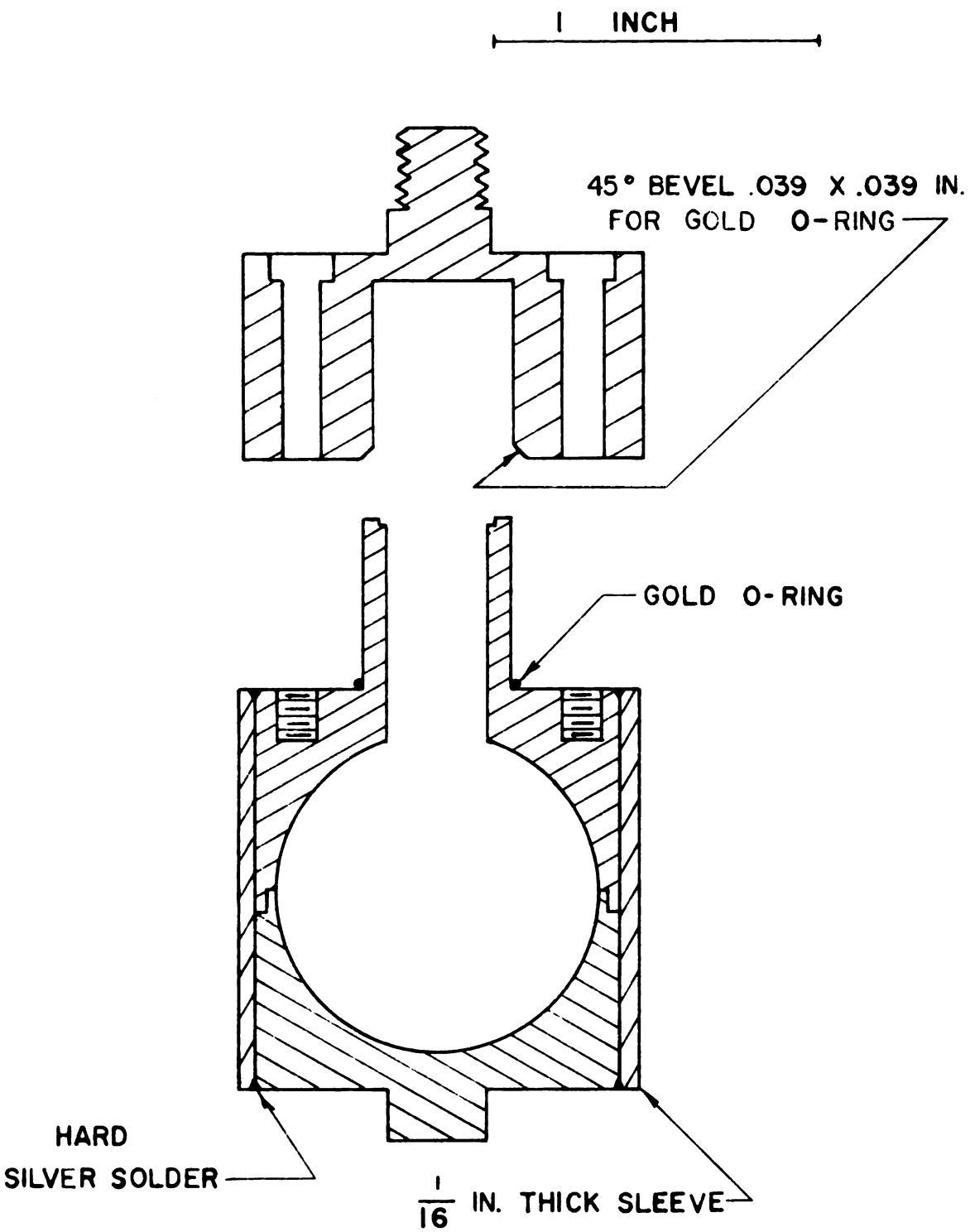

FIGURE 5

LI SAMPLE HOLDER

MATERIAL: STAINLESS STEEL TYPE 446

for vacuum seal. A stainless steel type 446 vane, in the form of a cross, was used to prevent rotation of the liquid in the neck of the sphere during an experiment.

The inner radius was calculated from the volume of distilled water needed to fill the sphere, the volume of the neck being known accurately from measurements with a depth micrometer. The calibration was done using a certified 10-ml burette. The error on volume measurement was estimated to be 1 part in 1000, with a resulting error in the radius of 1 part in 3000*. The data are shown in Appendix A.

B. Methods of Measurement

1. Preparation of the Samples

The apparatus shown in Figure 6 was used for transferring lithium to the sphere without contamination#. A lump of lithium, larger than necessary to fill the sphere, was put in the stainless steel funnel with the freshly cut surface down. The stem of the funnel, slightly flared at the end

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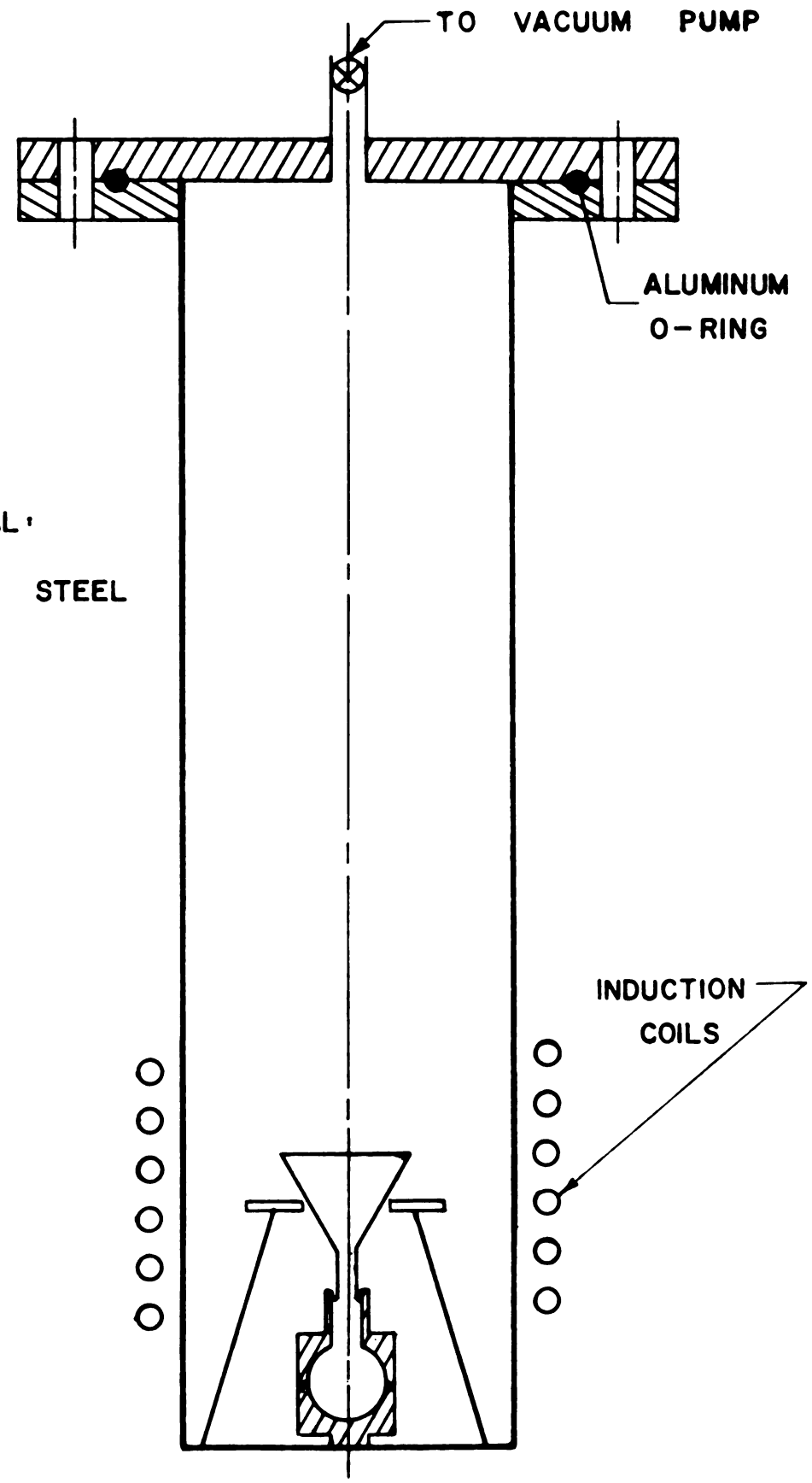
* It was to the credit of the machinist, Mr. Solonika that when the first sphere made was cut in half to check, the line where the halves joined could hardly be detected visually. In matter of fact, the fitting was so good that only careful inspection showed that the sphere was constructed from two hemispheres. The sphericity of the cavity, as checked with a one-inch steel ball covered with jeweler's rouge, was judged excellent.

This work was conducted at the Oak Ridge National Laboratory. The author assisted in the transferring of lithium-6; the filling with lithium-7 was done by Mr. E. B. Olszewski, chemist at the laboratory.



MATERIAL:

STAINLESS STEEL

FIGURE 6

APPARATUS FOR TRANSFERRING LITHIUM TO THE SAMPLE HOLDER

to prevent overflow, extended into the neck of the sphere to an appropriate depth. The chamber was then evacuated and the lithium melted by induction heating. The oxide-nitride coating on the lithium rested intact on the funnel and pure lithium ran down into the sphere. The lithium-filled sphere was allowed to cool under vacuum, then the chamber flooded with argon gas before it was opened. Upon cooling, the lithium in the stem of the funnel shrank and pulled up, breaking away from the solidified lithium inside the sphere. A jet of argon gas was directed on the openning of the sphere while the vane was inserted and the cap attached.

One of the filled spheres was immersed in mineral oil and the lithium was melted out to check visually whether any contamination had been introduced by this method of filling. Hardly any oxide or nitride of lithium was detected, the molten lithium floating to the top of the oil with a silvery brilliance.

The amount of lithium needed for each isotope to fill the sphere while still leaving ample room for thermal expansion in the temperature range contemplated was calculated, using the densities given by Snyder and Montgomery[39]. As a check the completed sphere was weighed. If the proper amount of lithium was not present, the transferring process was repeated. Two trials were necessary for lithium-6.

The lithium samples used were purchased from the Stable Isotopes Division of the Oak Ridge National Laboratory.

The following analyses were given by the supplier along with these clarifications:

1. The spectrographic results reported are semi-quantitative estimates and should not be interpreted or construed to be precise quantitative determinations;

2. <-No spectrum line visible. Probably absent, definitely less than value given;

3. <T -Present, but less than value given.

Lithium-6 Lot No. SS 5(b)

Isotopic Analysis:

Lithium-6, 99.3 ± 0.2%; Lithium-7, 0.7 ± 0.1%, (atomic percent)

Spectrographic Analysis (element and weight percent):

Al	<.01T	Fe	.05	Pb	<.01
Ba	.01	K	<.01	Sn	<.01
Be	<.001	Mg	.01	Sr	.01
Ca	.25	Mn	<.01	V	<.01
Cr	<.01FT	Na	.02	Zn	<.25
Cu	.02	Ni	<.01		

The lithium-6 sample with the above analysis was vacuum distilled for further purification of the metallic lithium, at the suggestion of Dr. P. S. Baker of ORNL, before being used to fill the sphere. The analysis of the final product is not available from ORNL at this time. (See Appendix G).

Lithium-7 Lot No. SS 7(b)

Isotopic Analysis:

Lithium-7, 99.99%; Lithium-6, 0.01%, (atomic percent)

Spectrograpnic Analysis (element and weight percent)

Al	<.05	K	.01	Rb	<.01

Ba	<.02	Mg	<.01T	Si	<.05
Ca	.02	Mn	<.02	Sn	<.05
Cr	<.05	Mo	<.05	Sr	<.2
Cu	<.01T	Na	.01	V	<.02
Fe	<.05	Ni	<.05		

2. Determination of the Moment of Inertia of the Suspended System

The moment of inertia of the suspended system I was made up of a fixed part I_o due to the sphere, the inertia rod, and various connecting rods, and a variable part I_n, due to the two movable disks K in position d_n which can be calculated. This arrangement, shown in Figure 7, was adopted so that the total moment of inertia I could be changed while keeping the load constant. Thus, the tension in the suspension wires, and hence the restoring force, would remain constant. It may be mentioned that this load was kept the same for the whole period during which the experiments were conducted, a sphere taken off the suspended system being immediately replaced by a dummy.

To ensure uniformity in density and true geometrical form, the disks K were cut from the same stock of brass and carefully turned in the lathe. The inertia rod was mounted on the suspended system in the manner shown in Figure 3, with a close sliding fit between the disks and the inertia rod.

A Sheffield Master Gagemakers Comparator, sensitive to variation of twenty millionth inch (20×10^{-6} inch), and a set of Johansson Gage Blocks were used to check the uni-

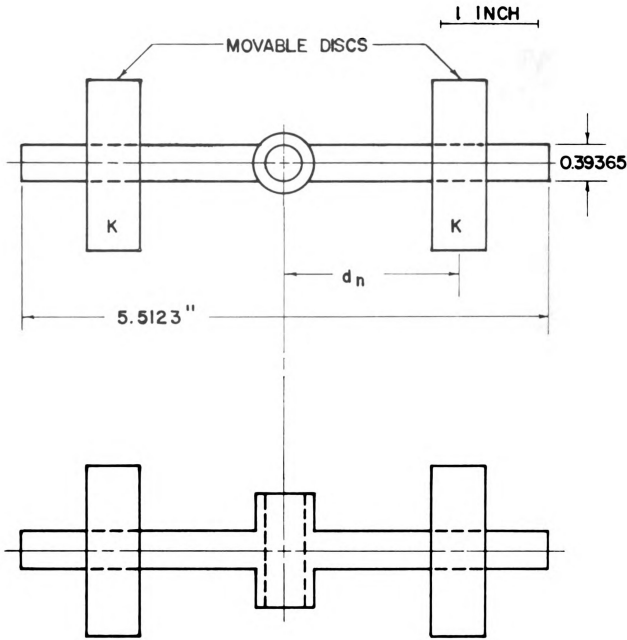


FIGURE 7

ADJUSTABLE MOMENT OF INERTIA SYSTEM

formity of the disks and the inertia rod. The maximum variation in the thickness of either disk was 1 part in 1500, and only 1 part in 6000 for the diameter. The mean thickness of the two disks did not differ by more than 1 part in 25,000, and the mean diameter, by more than 1 part in 40,000. The excellent match in geometrical dimensions was exceeded by that for the weights of the two disks, which differed only by 1 part in 100,000. The results are tabulated in Appendix B. The inertia rod showed a maximum variation of 1 part in 4,000.

A special gauge, shown in Figure 8, was made for setting the disks on the inertia rod to obtain different values for the moment of inertia I_n. It consisted of a brass cylinder of diameter slightly larger than that of the inertia rod, and two "standard" cylindrical blocks Y and Z. The dimensions shown on Figure 8, measured with the Sheffield comparator, were chosen to obtain large variation in the period of oscillation. Three different moments I_n could be set with the gauge. A groove was cut in the cylinder X to give an outlet for the trapped air when the gauge was pushed over the inertia rod to displace the disks K.

The moment of inertia I is related to the period of oscillation by

$$T_n = 2\pi (I/M)^{\frac{1}{2}}$$

where $I = I_o + I_n$, and M being the restoring couple. Although in principle M may be calculated from a knowledge of dimensions and elastic constant, it is simpler and more

ALL UNITS IN CM.

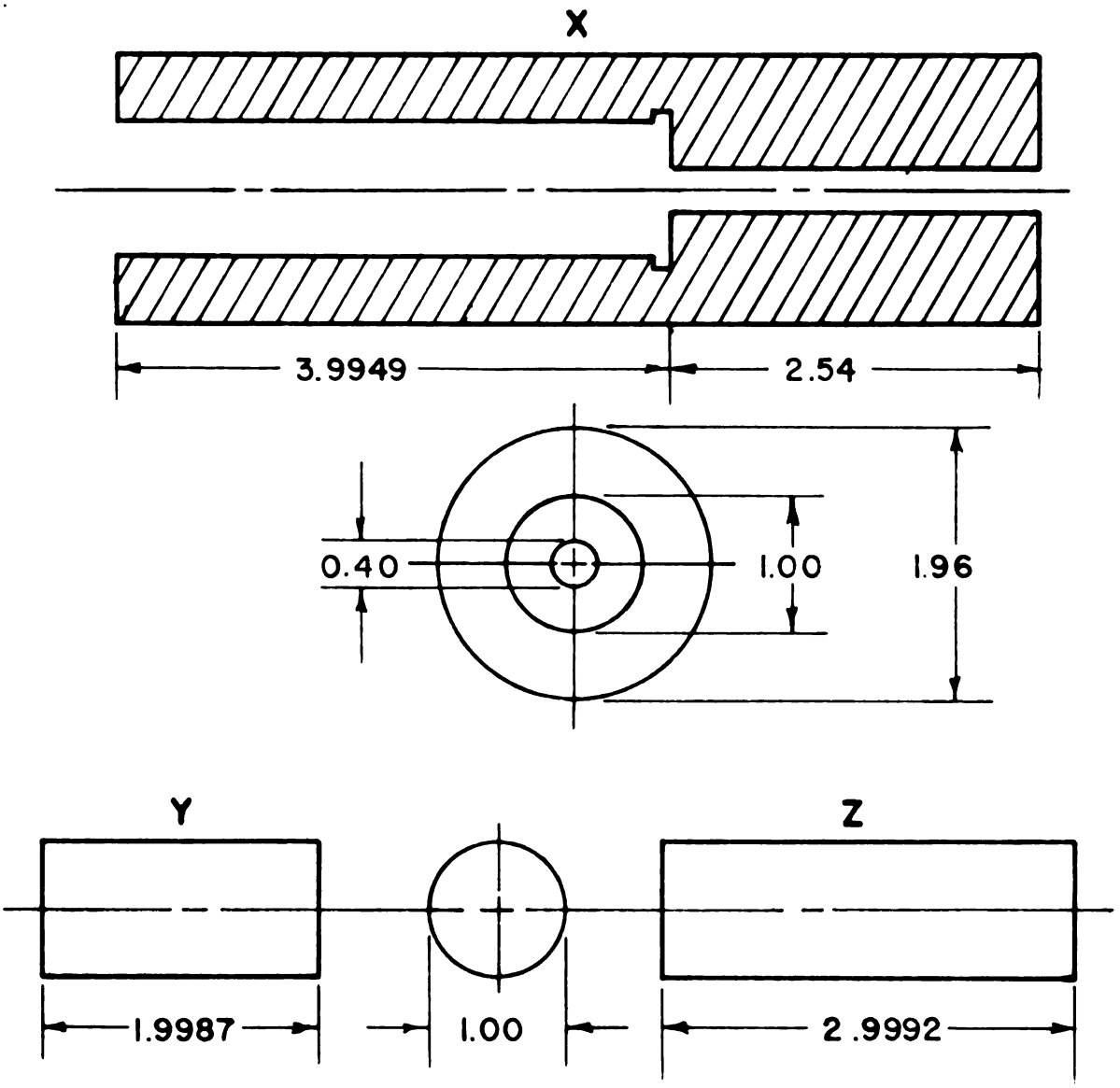


FIGURE 8

GAUGES FOR SETTING MOMENT OF INERTIA

accurate to eliminate it by measuring the period for different moments of inertia I. Thus, any two values of n enable I_o, hence I to be computed, that is,

$$I_o = \frac{I_2T_1^2 - I_1T_2^2}{T_2^2 - T_1^2}$$

Furthermore, the moment of inertia I_n is given by:

$$I_n = I_{1Z'} + I_{2Z'} + (m_1 + m_2)\, d_n^2$$

where $I_{1Z'}$, and $I_{2Z'}$ are the moment of inertia about the vertical axis passing through the center of mass of the two disks respectively; d_n, the distance between the center of mass of each disk and the center of the inertia rod; and m_1 and m_2, the mass of the two disks. Hence,

$$I_o = -(I_{1Z'} + I_{2Z'}) + (m_1 + m_2)\frac{d_2^2T_1^2 - d_1^2T_2^2}{T_2^2 - T_1^2} \qquad (21)$$

and the total moment of inertia I is given by the measured quantities m_1, m_2, d_n, d_1, d_2, T_1, and T_2 through:

$$I = I_o + I_n = (m_1 + m_2)\left[d_n^2 + \frac{d_2^2T_1^2 - d_1^2T_2^2}{T_2^2 - T_1^2}\right] \qquad (22)$$

Three different values of I_n were used, giving two independent values of I_o. The period of oscillation varied from about 8 seconds to about 15 seconds in the different cases. The results will be given in a later section of the thesis.

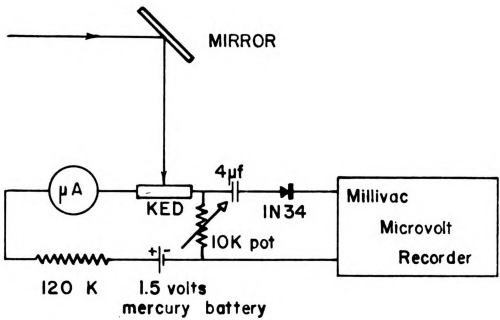
3. Measurement of Amplitude and Period of Oscillation

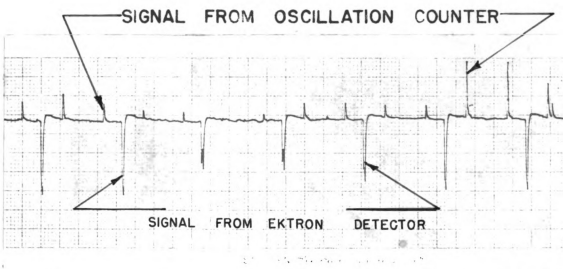
Since the logarithmic decrement was not very large, of the order of 500×10^{-6}, it would have been cumbersome to

record the amplitude photographically. Instead, the amplitude decrease was detected electrically using a Kodak Ektron cell, a lead-sulfide photoconductor, measuring 0.02 x 2 mm. A beam of light of suitable strength from a zirconium-arc lamp, admitted at window P of Figure 1, was reflected by a front-silvered mirror mounted on the lower wire clamp of the suspended system, and emerged horizontally from a second window, the plane of which was perpendicular to that of the first. The oscillating system was adjusted so that at rest the mirror was at 45° to both light beams.

The Kodak Ektron detector, provided with a shield against stray illumination, was mounted on a Spindler and Hoyer precision cross carriage with micrometer screw permitting a lateral movement of 25 millimeters. The screw allowed precise positioning of the detector to 0.01 millimeter. The screw carriage was in turn mounted on a sliding carriage provided with a vernier scale with a least count of 0.01 millimeter. The sliding carriage was used for coarse displacement of the detector in excess of 25 millimeters, and the screw carriage for fine adjustment. The sliding carriage was initially positioned so that the light beam with the mirror at rest lay along the perpendicular bisector of the carriage axis.

The electrical signal from the Ektron detector was recorded on a Millivac microvolt recorder, model MR-65A. The circuit used is shown diagrammatically in Figure 9. For any one run, the detector was initially positioned so that





SAMPLE STRIP CHART RECORD

FIGURE 9

CIRCUIT FOR AMPLITUDE MEASUREMENT

the light beam slightly swung past it at the extremity of a swing, the signal then appearing as a double pulse. As the amplitude decreased the overshoot of the light beam also decreased, bringing the two peaks of the signal closer to each other. Finally, when the light beam just reached the detector at the extremity of its swing, the signal became a single pulse. A sample strip chart record is shown in Figure 9.

The same light source used for recording the amplitude decrease was also used for counting the oscillations. A cadmium-selenide photoconductive cell, Clairex-3, placed as shown in Figure 4, was used in connection with a thyratron phase-control circuit to activate a counter. The circuit used is shown in Figure 10. The signal from this circuit was also recorded on the Millivac recorder at the same time that the amplitude decrease was being measured, and thus acted as a time marker. The Clairex cell was placed at the center of and immediately behind the sliding carriage to give two counts for each complete oscillation of the system. The period of oscillation was measured by timing 500 counts (250 complete oscillations) with a stop watch reading to 0.01 second. The watch was calibrated repeatedly for the same amount of time taken to measure 500 counts using the time signal broadcasted by the National Bureau of Standards. In this way the period was measured with an absolute accuracy of one millisecond. The period was measured at least three times during an experiment, and it was found to be

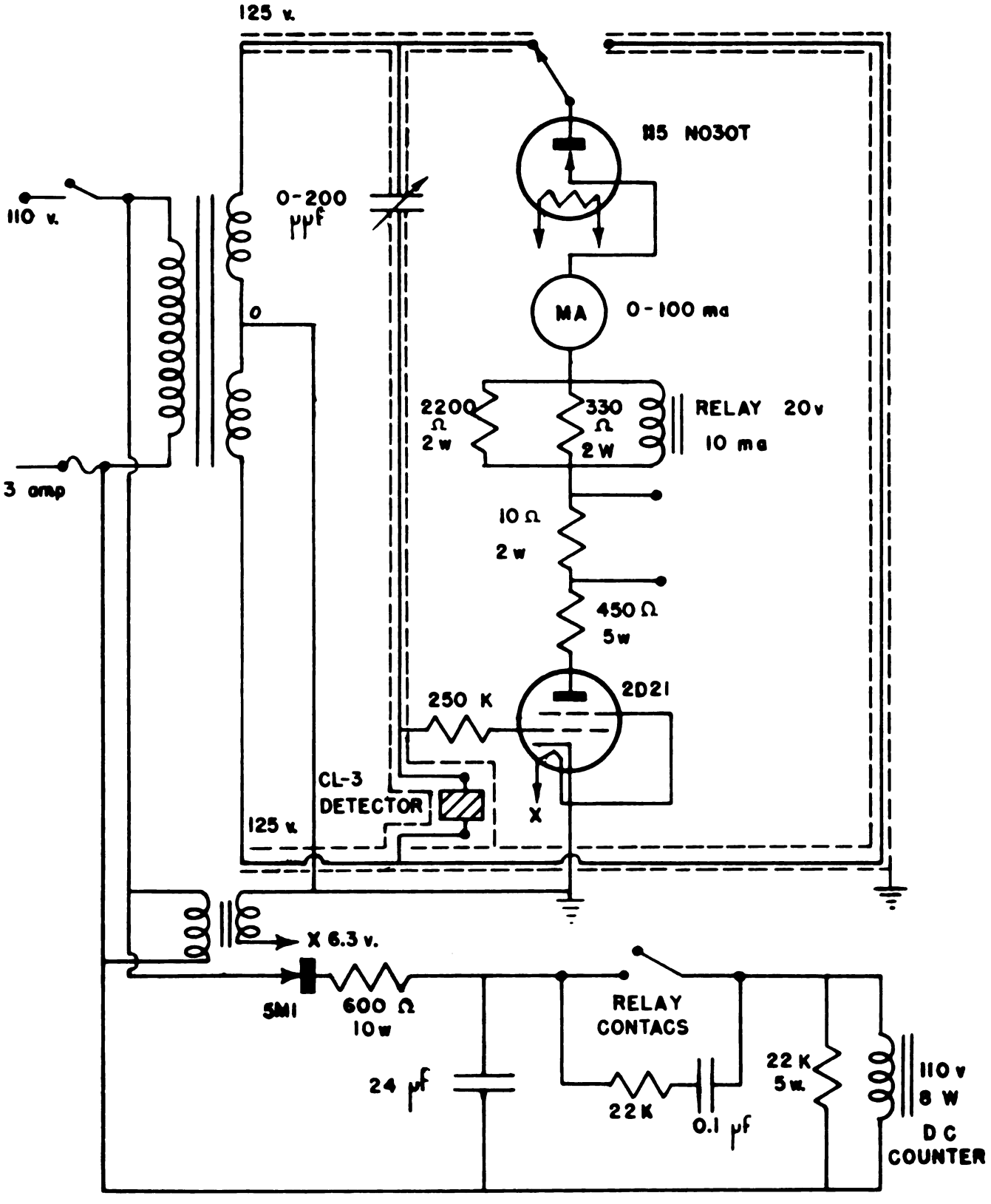


FIGURE 10

THYRATRON PHASE — CONTROL CIRCUIT

constant to within ± 0.5 millisecond.

For any one experiment, the sphere was set oscillating about the position of equilibrium by turning the lever arm 1 1/2 degree away from the position of equilibrium and back, and the counting circuit energized. The screw carriage was then moved on the sliding carriage to obtain a slight overshoot of the light beam past the Ektron detector, and the signal was recorded. When the signal changed as discussed in the above, the detector was advanced a fixed amount using the micrometer screw. This process was repeated fifteen times for each run. The lateral displacement of the Ektron cell for each advance was 0.50 millimeter during the lithium runs, and 0.20 millimeter during the runs to find the residual damping with the lithium solid. Each time the sphere is set into oscillation from rest, at least three runs were taken over a period of about four hours, giving three values for the logarithmic decrement δ.

At the end of each run, the position of equilibrium was determined by measuring the left and right excursions of the beam, thus allowing the computation of the amplitude of swing. The equilibrium position was found in this way to stay constant to within 0.05 millimeter.

Both the temperature of the furnace and the pressure in the vacuum enclosure were monitored during the entire period taken for each run. The pressure did not exceed 0.01 micron, ensuring that the residual damping due to air was negligible. The temperature recorded on the Speedomax remained

constant as discussed earlier.

4. Temperature Measurement of the Lithium-filled sphere

After the experiments with lithium-6 were completed, a copper-constantan thermocouple, insulated by a thin sheet of mica, was directly attached to the outside of the sphere on the horizontal equator. The sphere temperature thus measured was determined as a function of the furnace setting as indicated by the thermocouple T of Figure 1. During this calibration, the conditions under which the experiments were run were strictly observed. The copper-constantan thermocouple attached to the sphere were taken from the same spools used for making the thermocouple T. The temperature measurement was carried out using a Leeds and Northrup Universal potentiometer, type K-3.

The sphere itself was found to reach an equilibrium temperature about ten hours after the furnace was started at room temperature, and about six hours after a change in controller setting. For the experiments, the apparatus was generally left for twenty-four hours at a particular temperature before the decrement was measured.

C. Calculation of the Viscosity Coefficient

The viscosity coefficient η was calculated by the relation

$$\eta = \frac{a^2 R^2}{4\,(2-q)^2}\,\frac{\pi\rho}{T}\left\{1-(1-\mu)^{\frac{1}{2}}\right\}^2 \qquad (23)$$

the various symbols having been defined earlier. The quantities determined experimentally were δ, ρ, R, I, T_o, and T. The temperature dependence of these quantities will be now discussed.

1. Logarithmic Decrement δ

The plot of the logarithm of the amplitude as a function of the cumulative number of oscillations at any given temperature yielded a straight line, the slope of which was the logarithmic decrement δ. The logarithmic decrement was calculated by the least-squares method, the variables being the logarithm of the amplitude and the number of oscillations.

2. Density ρ

Precise determinations of the density of liquid natural lithium apparently have not been reported, let alone those of the separated isotopes. However the densities of the solid isotopes have been measured by Snyder and Montgomery[39]. The coefficients of thermal expansion in the solid and liquid state, as well as the volume change upon melting of the above isotopes, are still lacking, but they should vary little with isotopic mass. Consequently, the values given by Bernini and Cantoni[7] for the coefficients of thermal expansion, and that of Losana[34] for the volume change upon melting for natural lithium have been adopted.

We may then write:

$$\rho_t^{Li^6} = \frac{\rho_{om}^{Li^6}}{1 + 174 \times 10^{-6} (T - 180.4) + 106 \times 10^{-9} (T - 180.4)^2}$$

$$\rho_t^{Li^7} = \frac{\rho_{om}^{Li^7}}{1 + 174 \times 10^{-6} (T - 180.7) + 106 \times 10^{-9} (T - 180.7)^2}$$

where $\rho_{om}^{Li^6}$ and $\rho_{om}^{Li^7}$ are the densities of liquid lithium-6 and lithium-7 respectively at 180 C-deg., and

$$\rho_{om}^{Li^6} = \frac{0.460 \left[1 + 153.5 \times 10^{-6} \times 20 + 92 \times 10^{-9} (20)^2\right]}{1.0157 \left[1 + 153.5 \times 10^{-6} \times 180 + 92 \times 10^{-9} (180)^2\right]}$$

$$\rho_{om}^{Li^7} = \frac{0.537 \left[1 + 153.5 \times 10^{-6} \times 20 + 92 \times 10^{-9} (20)^2\right]}{1.0157 \left[1 + 153.5 \times 10^{-6} \times 180 + 92 \times 10^{-9} (180)^2\right]}$$

3. Inner Radius of the Sphere R

The change in the inner radius of the sphere with temperature was calculated, using as the linear coefficient of thermal expansion for stainless steel, type 446, the value

$$\alpha = 9.9 \times 10^{-6} \text{ / C-deg.}$$

4. Moment of Inertia I

No correction was made for the change in the moment of inertia I due to the expansion of the sphere, since this change amounted to only 5 parts in 10,000 when the temperature rose from 20 to 300 C-deg.

5. Method of Calculation

The actual calculation for the viscosity coefficient was carried out by programming equation (23) for MISTIC, a high speed digital computer. The degree of approximation used and the results obtained will be given in the following section of the thesis.

EXPERIMENTAL RESULTS

A. Verification of Method

The earlier history of the oscillating-sphere method for the determination of viscosity would give one little confidence in it. Later investigators have said that it is reliable, but certain details in their presentation lead to the suspicion that the precision may not be as high as claimed. Hence it was deemed essential to verify the validity of the method with use of some standard substance. Water seems suitable, since its viscosity is extremely well established, and is of the same order of magnitude as that expected for the lithium isotopes.

The sample of water was maintained at a fixed temperature by a rudimentary system consisting of a water bath in which was immersed a copper tube helix carrying tap water as a refrigerant. The experiment was carried out in accordance with the procedure described earlier. The results are shown in Table I. The values found experimentally are a few per cent lower than the accepted ones[24]. The explanation may lie in the fact that the temperature given in the table is the temperature of the copper jacket forming the bottom part of the chamber, whereas the temperature of the sphere may be a little larger, say a degree or two. It is possible to get better agreement by adjusting the temperature, but such procedure seems unwarranted. The chief point to be estab-

lished was the absence of gross effects that might induce error.

B. Effect of Temperature on Viscosity of Lithium-6

For the study of the effect of temperature on viscosity, only the lithium-6 sample proved to be useful. It was hoped to determine the effect of temperature with both isotopes, but with lithium-7 difficulties in the sealing of the sphere prevented an extensive study.

To make sure that the spherical portion of the sample holder was completely filled, the amount of lithium-6 that it contained was determined by weighing the sample holder before and after pouring the sample. The weight was 4.074 grams, corresponding to a volume large enough to fill the spherical cavity and a portion of the neck, while leaving ample room for thermal expansion over the temperature range used.

The moment of inertia of the suspended system, with the lithium-6 in the solid state, was then measured, with the results shown in Table II. The error is believed not to exceed 1 part in 6,000.

The residual damping, with the sample in the solid state, was measured according to the procedure outlined earlier, with the results shown in Table III. It is satisfying that the decrement is very small, and that it does not increase with temperature, a finding which shows that any damping due to the magnetic field of the furnace current is negligible.

Table I

Viscosity of Water

Temp. °C	η (expt.) mp.	η (std.) mp.	per cent deviation
16.6	10.48	10.94	- 4.2
17.1	10.42	10.80	- 3.5
18.6	9.87	10.40	- 5.1

Table II

Moment of Inertia for Li-6 Sphere

T_{10} = 8.795 sec.	d_1^2 = 5.13204 cm^2
T_{20} = 15.048 "	d_2^2 = 27.71601 "
T_{30} = 12.787 "	d_3^2 = 18.18255 "

With d_1 and d_2	I = 4,443.79 $gm\text{-}cm^2$
With d_1 and d_3	I = 4,443.75 "
With d_2 and d_3	I = 4,444.01 "
Average value	I = 4,443.86 "

Table III

Residual Damping for Li-6 Sphere

Temp. °C	Residual logarithmic decrement δ_o
22	41.79×10^{-6}
26	41.90 "
152	41.21 "
178	38.63 "

The value of the residual logarithmic decrement adopted was the one obtained at the temperature closest to the melting point of lithium-6.

The viscosity was then determined by observing the viscous damping at various temperatures. The first measurement was made at the highest temperature contemplated (266.8 °C). At the completion of this measurement, the temperature was decreased in steps until the lowest temperature used (188.3 °C) was reached, then brought up stepwise to the maximum again, and finally decreased once more to the lowest temperature. No effect due to a possible thermal hysteresis was noticed. The detailed sequence of measurements is given in Appendix C. At each temperature setting, the experiments were run at least once on each of two consecutive days. During each day of data-taking, three measurements of the logarithmic decrement were made as the amplitude of swing decreased. The decrements measured during any one day were found in most cases to be constant within 1/2 per cent, the largest deviation between extreme values never exceeding 1 per cent. The decrements obtained for each day were averaged, and the resulting value was used in the calculation after the residual decrement had been subtracted from it.

The temperature of the sphere could not be measured directly while a determination of damping was being carried out. It was determined indirectly by measuring the furnace temperature, which was found in a subsidiary investigation to be a linear function of sphere temperature, as shown in

Figure 11. The sphere temperature was measured according to the procedure described earlier. The effect of temperature on the radius R, the density, and the periods T_o and T is given in Appendix C.

It was little trouble to make successive approximations to an exceedingly high accuracy in the calculation of the viscosity coefficient with MISTIC. The calculation was continued until the relative difference between successive results was less than 10^{-7}. (For example, for lithium-6 at 188.3 C-deg., the final approximation was taken as 4.389 740 32 mp., the preceding one being 4.389 740 28 mp.; physically, the value is meaningful only to three or four significant figures).

The values found for the viscosity coefficient of lithium-6 as a function of temperature are summarized in Table IV and shown (together with one value for lithium-7) in Figure 12. The detailed results are tabulated in Appendixes C, D, and E.

C. Effect of Isotopic Mass on Viscosity of Lithium

To study the effect of isotopic mass on viscosity, the value for the viscosity coefficient of lithium-7 was measured at a given temperature and compared with that for lithium-6 at the same temperature. A defect in the construction of the lithium-7 sphere resulted in a leak which developed after the sphere had been held at high temperature for a time, and consequently the comparison can be made at only

one temperature.

The weight of the sample was 4.660 grams, corresponding to the proper volume of material. Because of the destruction of the sample, the moment of inertia and the residual damping could not be determined directly. However, it appears that use of the corresponding values for the lithium-6 sphere can not introduce appreciable error, in as much as the sample holders for lithium-6 and for lithium-7 are nearly identical, and the difference in mass and in moment of inertia for the actual isotope samples are negligible in comparison with the corresponding values for the whole system. The residual damping, therefore, should be very nearly the same, since the tension on the suspension wires and the geometry of the system are almost unchanged.

The viscosity coefficient of lithium-7 at 236.8 C-deg. was found then to be

$$\eta_{\text{Li-7}} = 4.19 \pm 0.08 \text{ mp.}$$

This value is also included in Figure 12.

D. Estimation of the Absolute Value for Natural Lithium

On the assumption that the viscosity coefficient for a mixture of isotopes of the same element is a linear function of the number composition, the absolute viscosity of natural lithium (92.5% lithium-7, 7.5% lithium-6) at 236.8 C-deg. is estimated to be

$$\eta_{\text{Li-nat.}} = 4.16 \text{ mp.}$$

Although the above assumption might be debatable in the

general case, it appears safe for a mixture consisting predominantly of one component, such as natural lithium is.

Table IV

Viscosity of lithium-6

Temperature °C	Temperature °K	Density gm/cm^3	Viscosity η mp	$\text{Log}_{10}\eta$	$10^3/T$ °K
188.3	461.5	0.44019	4.39	0.642	2.17
198.0	471.2	.43944	4.28	.631	2.12
217.4	490.6	.43792	4.00	.602	2.04
236.2	509.4	.43642	3.85	.585	1.96
236.8	510.0	.43637	3.83	.583	1.96
266.6	539.8	.43395	3.69	.567	1.85
266.8	540.0	.43390	3.67	.565	1.85

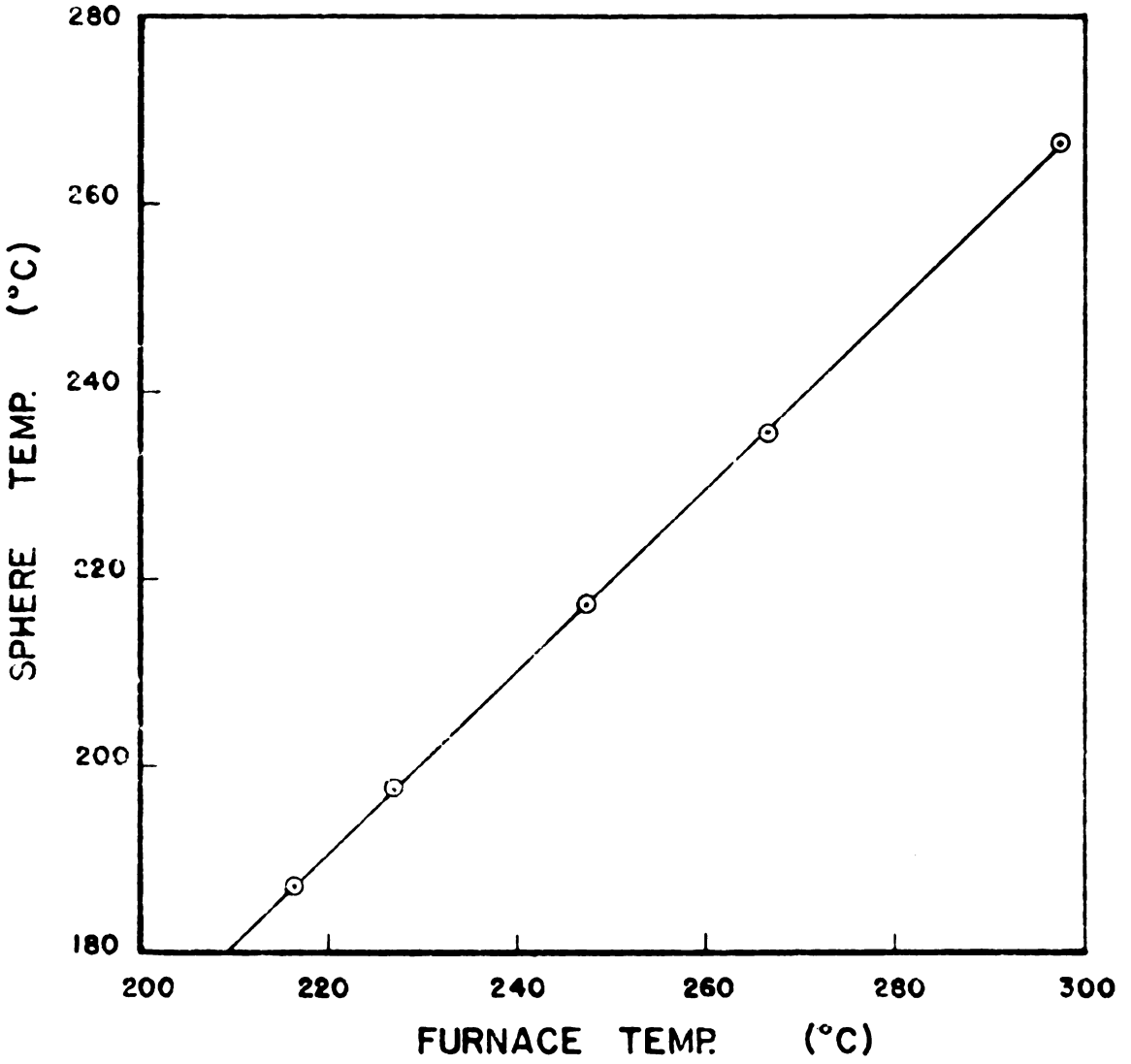


FIGURE 11

SPHERE TEMPERATURE AS A FUNCTION OF FURNACE TEMPERATURE

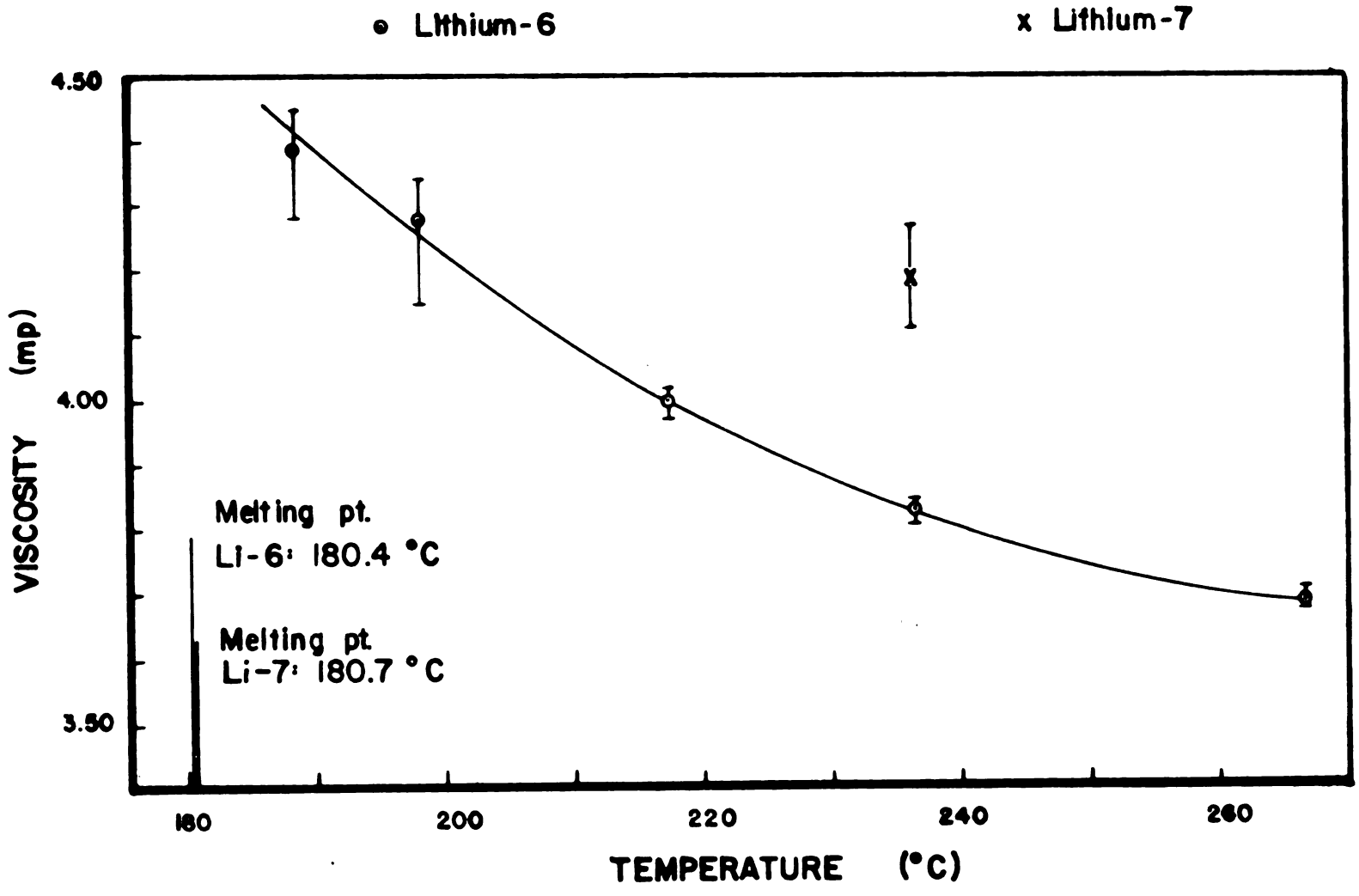


FIGURE 12

VISCOSITY OF LITHIUM-6 AS A FUNCTION OF TEMPERATURE

DISCUSSION OF RESULTS

A. Verification of Method

The results with water give confidence that the method is basically sound. The experiments reported, which were performed at the beginning of the experimental study, were not carried out carefully enough to allow a thorough evaluation of the limits of precision of the method. Nevertheless, it appears that the precision claimed by earlier workers may be illusory, since the final value calculated for viscosity is far more sensitive to inaccuracies in sphere diameter than had apparently been realized*. Moreover, the measurement of the temperature of the actual sample appears to be more difficult than had been considered by earlier investigators. With the present apparatus and technique, it is likely that the absolute value obtained for a viscosity coefficient can not be claimed to be accurate to within less than 3-5 per cent. The relative accuracy is considerably better, probably within 2 per cent or better.

B. Effect of Temperature on Viscosity

Previous experimental work for a variety of liquids has established a law that the logarithm of the viscosity is a

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* See Appendix F

linear function of the reciprocal of the absolute temperature. Moreover, some theories claim to give this sort of dependence. The present data, when plotted in the form log.η against 1/T, shows a general agreement with this law, as may be seen in Figure 13. The only serious deviation occurs at 266.8 C-deg., which may be due to either the temperature measurement or a slight increase in residual damping at high furnace current or to both. Proposed variants of this law, with some dependence on specific volume, do not give significantly different fits. The temperature range is sufficiently limited that a severe test of the proposed law is not possible.

The curve of Figure 13, plotted from Table IV, gives for the absolute viscosity of lithium-6 at the melting point (180.4 ^{o}C) the value

$$\eta = 4.50 \text{ mp.}$$

C. Effect of Isotopic Mass on Viscosity

According to the data previously cited, the ratio of the viscosity of lithium-7 to that of lithium-6 at 236.8 C-deg. is approximately (4.19±0.08)/(3.83±0.02) = 1.09±0.02, or when more exact figures are used to avoid rounding errors, 1.093±0.025. The square root ratio of the atomic mass of lithium-7 to that of lithium-6 is $(7.018/6.017)^{\frac{1}{2}} = 1.080$. Thus it appears that the experimental results are consistent with the theoretical predictions discussed earlier. Data at other temperatures are desirable in order to reduce the uncertainty in the above figure, if indeed the dependence

of viscosity coefficient on isotopic mass is independent of temperature. Data on "isotopic alloys" would be valuable, but caution must be exercised in the interpretation of the results, as the phenomena for mixtures of isotopes of a given average atomic weight are intrinsically more complicated than would be phenomena for an isotopically pure substance of the same atomic weight.

D. Accuracy of the Estimated Absolute Values for the Viscosity of Lithium

The absolute value for the viscosity coefficient of lithium-6, lithium-7, and lithium-natural are of theoretical interest with respect to the verification of the molecular theory of liquids, and of practical interest because of potential applications of lithium in nuclear-reactor technology. Consequently it is important to assess the accuracy of the absolute values reported herein, and to search for explanations of discrepancies with previous work.

The only work that need be considered is Andrade and Dobbs[2]. Their value for lithium-natural at 236.8 C-deg. is about 24 per cent higher than the present result. In order to carry out the comparison on a more extensive scale, we assumed that the viscosity coefficient varies as the square root of isotopic mass, and calculated the viscosities for lithium-natural from the results obtained for lithium-6. It is very significant to note then that the per cent deviation between Andrade and Dobbs results and the present results is very constant over the whole range of temperature

used, as shown in Table V. The only exception occurs at 266.3 C-deg., for the reasons given in a previous section. This constancy strongly suggests that the difference in the results may be due to a systematic error in Andrade and Dobbs technique. A detailed examination of their paper led to the following observations:

1. The sphericity of their sample holder, which influenced the radius calculation, is very questionable, especially after a close study of their calibration data for the sphere; we have demonstrated earlier the extreme sensitivity of the viscosity coefficient η with respect to inaccuracies in sphere diameter.

2. No data were given either for the logarithmic decrements or for the residual damping, even though they constitute one of the most significant results directly obtained from the experiments.

3. These authors claim that there exists no temperature differential between the jacket and the sample holder, this is very questionable, especially since we have found a temperature difference of about 30 C-deg. in our work. However this alone is not sufficient to explain the difference in the results.

Even though we only claim that our absolute values are accurate to within 5 per cent, and our relative values to 2 per cent or better, we have to conclude from the above observations that Andrade and Dobbs results seem to be significantly in error. This fact is of technological

interest.

Table V

Calculated Viscosity of Lithium-natural

Temp. °C	η_{Li-6} mp	η_{Li-nat} (mp)		per cent difference
		Ban*	Andrade	
188.3	4.39	4.71	5.85	24
198.0	4.28	4.60	5.71	24
217.4	4.00	4.30	5.40	25
236.8	3.83	4.11	5.12	25
266.8	3.67	3.94	4.73	20
180.4	4.50	4.83	6.02	25

* The viscosity coefficient of lithium-natural was calculated as follows:

$$\eta_{Li-nat} = \eta_{Li-6} \cdot \left[(1.080 \times 0.925) + 0.075\right] = \eta_{Li-6} \times 1.074$$

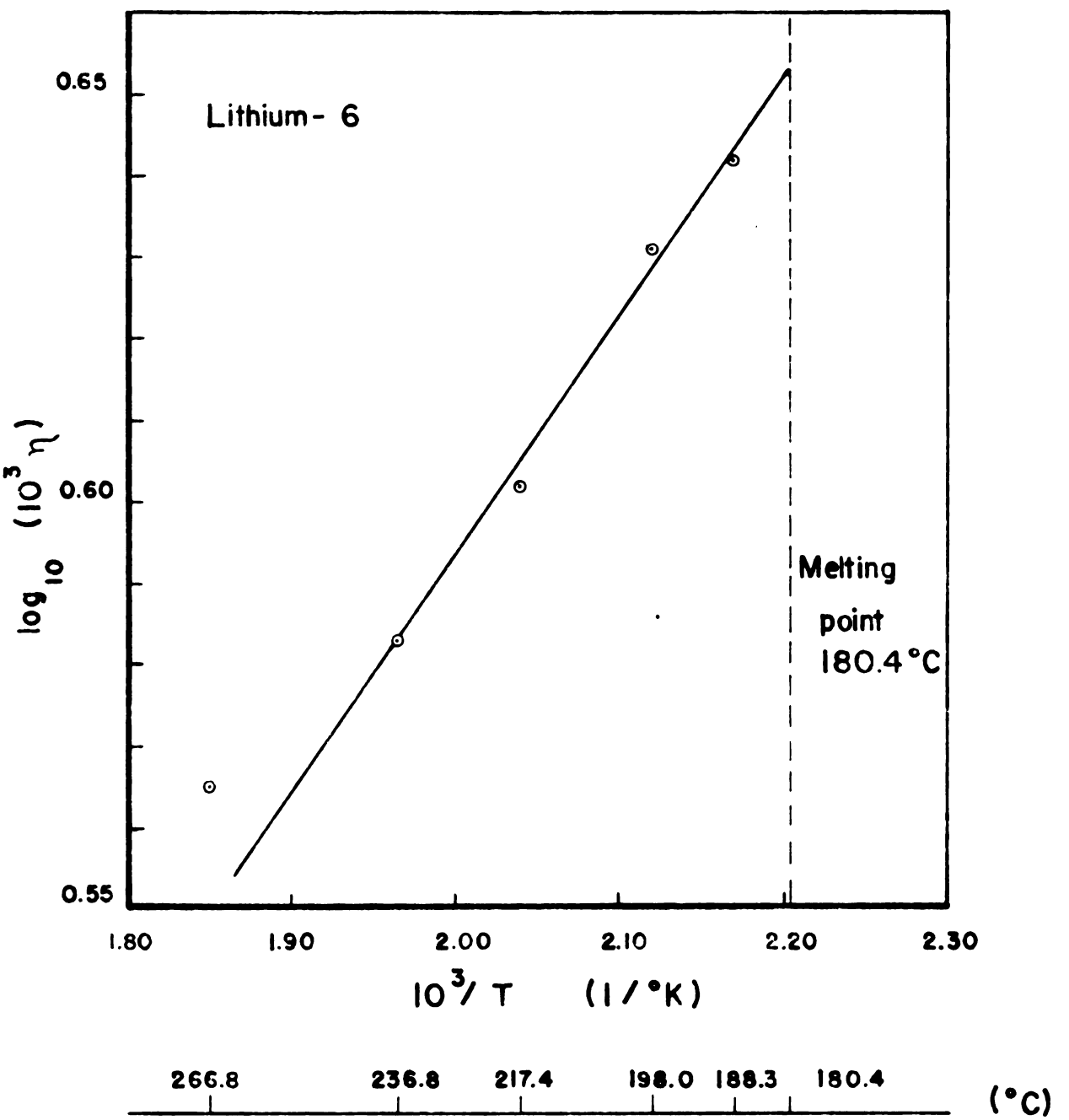

FIGURE 13

PLOT OF $\log_{10}(10^3\eta)$ AGAINST $10^3/T$

APPENDIX A

INNER DIAMETER OF SPHERE

Volume of spherical cavity V_s = Total volume of water used V_t - Volume of neck V_n.

$$V_s = \frac{1}{6}\pi D^3$$

Volume of neck (Cf. Figure 5):

$$V_n = \frac{\pi}{4}\left[(0.350)^2 \times 0.060\right] + \frac{\pi}{4}\left[(0.3125)^2 \times 0.752\right] \text{ in}^3$$

$$= 6.345 \times 10^{-2} \text{ in}^3 = 1.04 \text{ cm}^3$$

The base of the cylindrical neck joining the top of the spherical cavity is not exactly flat, the error introduced by assuming that it is flat was computed to be 7.69×10^{-4} cm^3, i.e., completely negligible.

Measurement No.	Sphere No. 2 (Lithium-6) V_t cm^3	Sphere No. 3 (Lithium-7) V_t cm^3
1	9.88	9.91
2	9.88	9.92
3	9.89	9.91
4	9.88	9.91
5	9.88	9.90
6	9.87	9.91
Average V_t	9.88	9.91
Average V_s	8.84	8.87
Diameter D	2.5654 cm	2.5683 cm

APPENDIX B

DIMENSIONS OF INERTIA DISKS

Note: All dimensions given are measured in inch, unless otherwise indicated.

	Disk No. 1	Disk No. 2
Thickness	.5828 .5829 .5830 .5830 .5830 .5828 .5830 .5830 .5831 .5830 .5828 .5827	.5827 .5829 .5830 .5831 .5830 .5830 .5828 .5830 .5830 .5830 .5829 .5829
Average	.58292	.58294
Diameter	1.7718 1.7717 1.7718 1.7719 1.7717	1.7719 1.7717 1.7717 1.7720 1.7718
Average	1.77178	1.77132
Weight	189.6274 grams	189.6290 grams

APPENDIX C

DATA FOR VISCOSITY OF WATER

	Temperature (^{o}C)		
	16.5	17.1	18.6
R (cm)	1.2793	1.2793	1.2827
T_o (sec)	12.317	8.812	8.812
T (sec)	12.758	8.773	8.768
Density (gm/cm^3)	0.998843	0.998757	0.998482
I ($gm-cm^2$)	9,396.91167	4,447.42603	4,447.42603
$\delta_o \times 10^6$	44.5562	45.4292	45.4292
$\delta_1 \times 10^6$	580.7933	1,052.1470	1,045.6000
$(\delta_1 - \delta_\omega) \times 10^6$	536.1421	1,006.7173	1,000.1708
Viscosity η (mp)	10.4799	10.4177	9.8666

APPENDIX C (Cont.)

DATA FOR VISCOSITY OF LITHIUM-6 AND LITHIUM-7

Order	Temp. °C	R cm	T_o sec	T sec	ρ gm/cm^3	δ x (10^6)	η mp
				Lithium-6			
6	188.3	1.23484	8.795	8.794	.44019	441.8940	4.3239
7	"	"	"	8.793	"	440.2847	4.2768
16	"	"	"	8.794	"	446.6999	4.4649
17	"	"	"	"	"	447.7455	4.4962
Ave.	"	"	"	"	"	444.1560	4.3897
5	193.0	1.28496	8.795	8.794	.43944	435.5604	4.1533
14	"	"	"	8.793	"	442.2466	4.3425
15	"	"	"	"	"	442.3003	4.3440
Ave.	"	"	"	"	"	440.0358	4.2787
4	217.4	1.28521	8.795	8.795	.43792	430.1638	4.0231
8	"	"	"	8.794	"	428.1079	3.9670
9	"	"	"	"	"	429.5190	4.0049
Ave.	"	"	"	"	"	429.2636	3.9980
3	236.2	1.28545	8.795	8.794	.43642	422.5769	3.8374
10	"	"	"	"	"	424.8746	3.8975
11	"	"	"	"	"	421.5107	3.8098
Ave.	"	"	"	"	"	422.9874	3.8480
18	236.8	1.28545	8.796	8.798	.43637	421.5840	3.8144
19	"	"	"	"	"	422.8673	3.8477
Ave.	"	"	"	"	"	422.2257	3.8311
1	266.6	1.28583	8.795	8.795	.43395	415.4996	3.6832
2	"	"	"	"	"	416.4326	3.7067
Ave.	"	"	"	"	"	415.9661	3.6949
12	266.8	1.28583	8.795	8.795	.43390	414.8041	3.6665
13	"	"	"	"	"	415.3273	3.6797
Ave.	"	"	"	"	"	415.0657	3.6731
				Lithium-7			
	236.3	1.28696	8.796	8.797	.50945	484.0310	4.1864

APPENDIX D

LOGARITHMIC DECREMENT DATA

Order	Temp. °C	Log. decr. $\delta_1 \times 10^6$
	Lithium-6	
6	188.3	482.2725
		478.3121
		480.9846
7	188.3	478.0755
		479.6906
		478.9752
16	188.3	486.1153
		484.5164
		485.3552
17	188.3	485.9834
		486.7659
5	198.0	475.4092
		475.0178
		472.1414
14	198.0	482.2980
		482.1750
		478.1542
15	198.0	481.3653
		479.8177
		481.6051
4	217.4	470.8463
		467.6874
		467.8449
8	217.4	467.0125
		466.6431
		466.5554
9	217.4	467.3154
		470.8317
		466.2972
3	236.2	460.2000
		461.5133
		461.9048
10	236.2	463.2557
		465.3280
		461.9273
11	236.2	460.7277
		460.3303
		459.3115
18	236.8	458.1642
		462.2620
19	236.8	461.5875
		461.4054
1	266.6	455.0963
		454.6390
		452.6509
2	266.6	456.3262
		454.9471
		453.9118
12	266.8	453.8711
		452.1115
		454.3171
13	266.8	455.9060
		454.7515
		451.2116
	Lithium-7	
	236.8	523.8293
		522.3651
		521.7860

APPENDIX D (Cont.)

Residual damping for lithium-6 sphere

Temperature °C	Logarithmic decrement $\delta_o \times 10^6$
22	41.4377
	42.1339
Ave.	41.7358
26	43.0992
	40.3912
	42.2050
Ave.	41.8985
152	40.6121
	41.2272
	41.2378
Ave.	41.21422
178	39.0468
	38.7209
	38.1197
Ave.	38.6291.3

APPENDIX E

SPHERE TEMPERATURE AS A FUNCTION OF FURNACE TEMPERATURE

Order	Furnace temp.*		Sphere temp.#		Temperature difference
	mv	°C	mv	°C	
1	10.17	216.4	8.515	187.3	29.1
9	10.21	217.2	8.659	188.1	29.1
2	10.71	226.4	9.123	196.9	29.5
8	10.74	227.0	9.161	197.6	29.4
3	11.87	247.4	10.212	217.2	30.2
7	11.88	247.6	10.213	217.2	30.4
4	12.95	266.7	11.226	235.2	31.5
6	12.96	266.8	11.234	235.9	30.9
5	14.73	297.7	12.945	266.6	31.1

* as measured by thermocouple T of Figure 1.

\# as measured by a thermocouple attached directly to the outside of the sphere on its horizontal equator.

APPENDIX F

DEPENDENCE OF η ON R

The following table will clearly demonstrate the extreme sensitivity of the viscosity coefficient η to inaccuracies in the sphere radius. The data for lithium-6 at 236.8 C-deg. were used; keeping I, T_o, T, δ , and ρ fixed and varying only R, the following results were obtained:

R (cm)	η (mp)
1.283	3.922
1.284	3.885
1.285	3.848
1.28545	3.831
1.287	3.775
1.289	3.704

APPENDIX G

SPECTROGRAPHIC ANALYSIS OF LITHIUM-6 AFTER VACUUM DISTILLATION

The spectrographic analysis of the lithium-6 sample which was used in this investigation has been made by ORNL. Vacuum distillation has in general improved the purity of the sample with respect to impurity contents, with a marked decrease in the calcium and zinc impurities, the only two elements present in relatively substantial amount before distillation. The results are given as follows:

Lithium-6 Lot No. SS 5(b)

Spectrographic Analysis (element and weight percent):

Ag	<.01T	Fe	.03	Pb	<.01T
Al	.01	K	.02	Si	.05
Ba	<.01T	Mg	<.02	Sn	<.02
Ca	.05	Mn	<.01T	Sr	<.01
Co	<.05	Mo	<.01	Ti	<.02
Cr	.01	Na	.05	V	<.01
Cu	.05	Ni	<.01T		

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