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Brownian Movement in Quartz An Attempt at Quantitative Evaluation

By C. G. I. Friedlaender (Halifax, Nova Scotia)*)

With 2 figures in the text

Abstract. Evaluation of a movie film of the movement of a gas bubble in a liquid inclusion in quartz from Dunbrack Prospect, Musquodoboit River, Halifax County, Nova Scotia, resulted in the tentative determination of the viscosity of the liquid.

Résumé. La viscosité du liquide contenu dans une inclusion d'un quartz provenant de Dunbrack Prospect, Musquodoboit River, Halifax County, Nova Scotia, a été tentativement déterminée par l'évaluation de mesures effectuées sur une prise de vue cinématographique.

The Problem

Moving gas bubbles are fairly frequent in liquid inclusions in various minerals.

Two questions arise:

- 1. can the mouvement of the gas bubble be characterized so as to distinguish between Brownian Movement and other types of mobility?
- 2. can quantitative determinations be arrived at?

1. Type of movement

Bubbles in liquid inclusions may display different types of motion, which, however, are frequently lumped together indiscriminately (G. DEICHA, 1955).

There is, firstly, the irregular and uninterrupted motion of very small bubbles with a diameter in the order of the micron: the Brownian Movement. The swarming motion of fine particles dispersed in water described, in 1828, by Robert Brown has subsequently been termed Brownian Movement. An overall account of the Brownian Movement may be found in the publications of J. PERRIN (1909) and G. DE HAAS-LORENTZ (1913), a discussion of the theory of

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the Brownian Motion in G. E. UHLENBECK and L. S. ORNSTEIN (1930, as well as in M. C. WANG and G. E. UHLENBECK (1945).

The Brownian Movement is the more lively the smaller the bubble. If the diameter of the bubble is greater than 0.004 mm, the movement will be hardly perceptible. The movement in neighbouring inclusions is unrelated.

Then there is, secondly, a "Non-Brownian" mobility, in regular dependence of outside factors such as change of temperature or inclination of the inclusion containing the bubble (E. ROEDDER, 1965). The bubbles with this type of mobility are bigger: their diameter may be in the order of the millimetre. The mobility in neighbouring inclusions, even if of different size, will be parallel.

Thirdly, we may observe in liquid inclusions gas bubbles without any apparent mobility. What causes the inhibition may not always be recognized. The most plausible explanation is that the bubble is in contact with, and in some way adheres to, the wall of the inclusion.

2. Quantitative determinations

The formula developed by Einstein (ALB. EINSTEIN, transl. A. D. COWPER, ed. R. FÜRTH, 1956) states that the square of the average mean displacement Δ is

$$\bar{\varDelta}^2 = \frac{R\,T}{N} \frac{1}{3\,\pi\,\eta\,r}\,t\,,$$

where R is the gas constant, 8.31×10^7 ,

- T is the abs. temperature,
- N is the Avogadro number, 6.02×10^{23} ,
- η is the viscosity of the liquid,
- t is the time interval of consecutive displacements,
- r is the radius of the moving particle.

If the diameter of the bubble and the apparent displacement in a known time interval have been ascertained at a given temperature, the equation can be solved for η

$$\eta = \frac{R T}{N \bar{\mathcal{A}^2}} \frac{t}{3 \pi r}$$

and the viscosity of the liquid can be calculated.

With proper equipment, there will not be any difficulty to ascertain the time interval of consecutive displacements and the temperature. On the other hand, a number of difficulties will affect the other observations:

1. ascertaining the diameter of the bubble. Difficulties are caused by the diffraction at the borderline bubble-liquid and by the movement of the bubble.

2. ascertaining the centre of consecutively considered positions of the bubble.

3. the measurement of the distance between the centre of the bubble of these consecutively considered positions.

4. calibrating the length measurements with a standard. Furthermore, the distance measured in the horizontal plane will necessarily be smaller than the true distance between the bubble positions considered as the bubble does not move only in the plane of observation, parallel to the thin section.

The determination of the viscosity of the liquid in inclusions through evaluation of the Brownian Movement can definitely be of interest.

There is a conspicuous lack of published data of measurements of the apparent displacement of gas bubbles in liquid inclusions.

This is probably due, at least in part, to the difficulties I have just mentioned, but in many cases it may prove possible to arrive at a satisfactory quantitative evaluation of the movement of gas bubbles in liquid inclusions.

Measuring Procedures

Moving gas bubbles have been observed in liquid inclusions in quartz from specimens taken at the Dunbrack Prospect, Musquodoboit River, Halifax County, Nova Scotia (C. FRIEDLAENDER, 1968).

With a movie film, the track of such a bubble has been recorded¹). The film made it possible to measure the apparent displacement of the moving bubble.

The speed of the film was 24 frames per second and the exposure of a single frame was $1/_{60}$ second.

Two approaches have been followed to measure the displacement of the bubble. One made use of enlarged prints. Prints of frames spaced at known intervals were successively projected with an epidiascope and the position of the bubble relative to the outline of the inclusion was recorded. The other approach was based on an animation film camera which was used as a projector; at controlled spacing, and therefore known time interval, the position of the moving bubble was recorded.

Both approaches are essentially similar and both contain some inevitable errors. The use of a travelling microscope, directly on the film, would eliminate one reversal and matching operation and should therefore result in smaller measuring error.

The calibration was carried out with a stage micrometer.

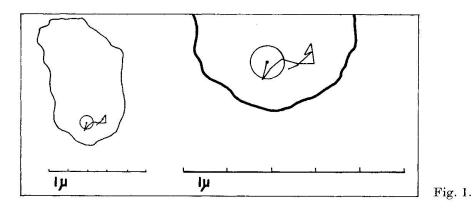
The measuring of the diameter of the bubble was first attempted in a direct way, using the eyepiece micrometer. It is difficult to focus the moving bubble and to match its diameter with the eyepiece micrometer. The error in the

¹) I would like to thank Mr. D. Pike and Mr. D. A. Gibson, Audio Visual Department, Dalhousie University, for their co-operation in the preparation of this film.

determination of the diameter of the bubble may be substantial. The diameter of the bubble was, in a different attempt, measured by means of photographs taken, in an alternating way, of the inclusion with the moving bubble and of a stage micrometer. The film was then measured on a device as used for evaluating X-ray films. The radius of the bubble was determined in this way:

$$r = 0.756 \times 10^{-4}$$
 cm.

The measurements made with enlarged prints and an epidiascope comprised 10 positions; these were spaced at a distance of 20 frames, corresponding to a time interval of ${}^{20}/_{24}$ second (Fig. 1).



The average mean displacement found in this set of measurements was

$$\sqrt{\bar{\mathcal{A}}^2} = 0.390\,\mu.$$

The second set of measurements, made with an animation camera, covered 45 consecutive positions of the bubble at a spacing of 12 frames, that is at a time interval of 0.5 sec.^2).

²) The measurements were as follows:

time	Series				
(in frames)	1	2	3	4	5
12	2.8	1.7	1.4	6.1	1.3
24	6.5	5.5	6.6	5.9	4.3
36	2.7	3.2	3.7	1.8	4.5
48	3.2	2.9	3.9	2.3	10.0
60	2.6	2.2	1.4	3.1	2.9
72	1.9	4.1	3.4	4.8	
84	5.1	3.7	2.2	2.4	
96	2.1	6.8	3.3	3.3	
108	2.5	6.7	1.4	4.2	
120	5.4	6.8	3.9	4.6	

The sum of the squares of the measured displacements is 806.72. The number of steps was 45.

 Δ^2 is therefore 806.72/45 = 17.92

and $\vec{\Delta}$ is $\sqrt{17.92} = 4.23$.

The calibration showed that 1 mm in the graph = $0.109 \ \mu$.

 $\varDelta = 4.23 \times 0.109 = 0.461 \ \mu.$

The average mean displacement in this set of measurements was

$$\sqrt[4]{\Delta^2} = 0.461\,\mu.$$

The displacement of $\Delta = 0.390 \ \mu$ for t = 0.834 sec. corresponds to $\mu = 0.304$ for t = 0.5 sec. The results of the two sets of measurements do not match too well: the ratio of the results is $\frac{\text{series } 2}{\text{series } 1} = 1.52$.

The second series is to be considered more reliable because 1. the procedure involved less manipulations and fewer steps and 2. the number of measurements was higher.

The approach using photographic prints may however be considered if no animation camera nor any other more suitable instrumentation is available. It should prove adequate, provided a sufficient number of measurements are taken.

Results

With the measured values

$$r = 0.756 \times 10^{-4}$$
 cm,
 $\overline{\varDelta} = 0.461 \times 10^{-4}$ cm,
 $t = 0.5$ sec, $T = 290^{\circ}$ K

and

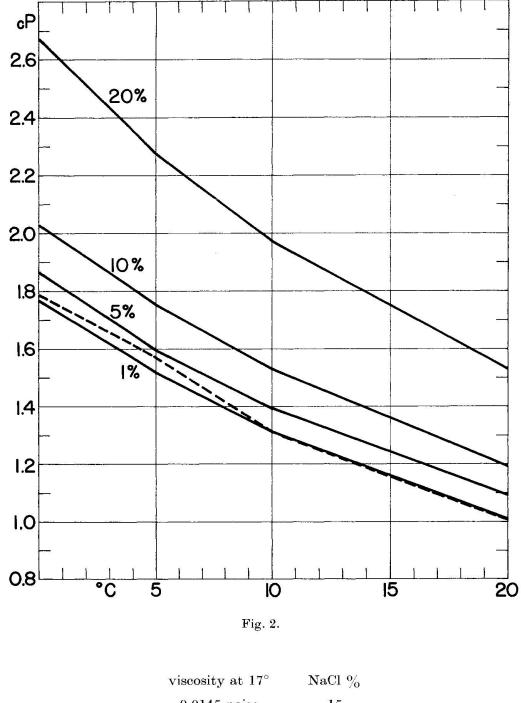
we calculate the viscosity

$$\eta = \frac{R T}{N \bar{\Delta}^2} \frac{t}{3 \pi r} = 0.0133$$
 poise.

Fig. 2 gives the indications of the viscosity of water and that of aqueous solutions of NaCl of 1%, 5%, 10% and 20% at atm. pressure and temperature between 0° and 20° C. (The values have been taken from Landolt-Börnstein and from the International Critical Tables.) At the temperature of 17° C, the viscosity of water may be given as 1.1 centipoise. If we consider, somewhat arbitrarily, the difference (1.33 - 1.1), i.e. 0.23, as maximum error in the present determination, we might indicate $\eta = 0.0133 \pm 0.0012$. This would indicate an error of $\pm 9\%$. It is a rough procedure but under the circumstances it does not appear necessary, nor possible, to go any further.

The composition of the solution itself cannot be extrapolated from the viscosity. Chemical determinations indicate that NaCl is frequently the preponderant constituent (F. G. SMITH, 1954; R. GOGUEL, 1963; I. N. MASLOVA, 1961).

Under the assumption that in the present case NaCl is indeed the preponderant constituent and, further, that other substances do not significantly influence the viscosity, we may tentatively infer the NaCl content from the viscosity value of an aqueous solution of NaCl.



0.0145 poise	15
0.0133	12
0.0121	9

These results show at the same time

- 1. that the movement of the gas bubble in a liquid inclusion in quartz from Dunbrack Prospect is to be considered as Brownian Movement, and
- 2. that quantitative determinations are feasible.

Discussion

Different methods have been devised for the determination of the temperature of formation of the host minerals of liquid inclusions (C. W. CORRENS, 1953; G. DEICHA, 1955; E. INGERSON, 1947; F. G. SMITH, 1953, 1954; N. P. YERMAKOV, 1965).

On the other hand, the measurement of the apparent displacement of bubbles in liquid inclusions has so far not been used for any determination.

In favourable cases, such measurements may afford, however, a possibility to arrive at an indirect determination of the viscosity.

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