Journal of Alloys and Compounds 480 (2009) 629–633

Contents lists available at ScienceDirect

Journal of Alloys and Compounds

journal homepage: www.elsevier.com/locate/jallcom

A statistical approach to estimate the experimental uncertainty of viscosity data obtained by the oscillating cup technique

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ARTICLE INFO

Article history:
Received 18 November 2008
Received in revised form 30 January 2009
Accepted 1 February 2009
Available online 10 February 2009

Keywords:
Liquid alloys
Dynamic viscosity
Experimental error
Oscillating cup viscometer

ABSTRACT

The determination of the dynamic viscosity of metallic alloys at elevated temperatures is a key experiment for materials design and characterisation. To do so, the oscillating cup method is the most suited and commonly applied one. However, critical estimations of the experimental uncertainties are scarce, complicating the comparison of results obtained by different authors as well as with models set up in order to describe the changes of the dynamic viscosity with temperature and the alloy's composition. We present a statistical approach, first to estimate the uncertainties of the actual measured parameters of a torsional oscillation, and second to analyse the propagation of these errors when calculating the dynamic viscosity. Overall relative uncertainties in the range of 5–10% are obtained for selected experiments carried out on pure liquid Al, Sn and Cu. This figure is in good agreement with previous error estimations. However, method is ready to be involved in the usual numeric calculation procedure of the dynamic viscosity in the actual experiment and is thus able to take the individual conditions with a given sample into account.

1. Introduction

Knowing the dynamic viscosity of molten metals and alloys is of great interest for both basic research and industrial production. Mass transport phenomena as the viscous flow need to be controlled in casting and soldering applications and rule the nucleation and growth of crystal nuclei. The dynamic viscosity itself is a materials parameter, reflecting both structural (on either length-scale) and thermodynamic properties of the material.

Obviously, measuring the dynamic viscosity of molten metals and alloys is an important task. Though several methods can be considered, only the oscillating cup technique is applied recently in several groups. The main reason is the high reactivity of the samples with almost all high-temperature stable materials which could be used for constructing parts of the apparatus to be immersed into the liquid in conventional rotational techniques. However, literature surveys [1–4] show large discrepancies in the experimental results obtained with different apparatus even for melts of the pure elements. On the other hand, a variety of model equations for the viscosity of multicomponent liquids—for recent reviews see Refs. [5,6]—have been developed by several authors and even the slightest changes in the temperature dependence of the dynamic viscosity are discussed in very detail in terms of structural changes in the liquid state, e.g. [7,8]. Of course, these model considerations are to be tested against the experimental data. In order to do so properly one should discuss on the experimental uncertainties present in the experiments—but this is hardly done in these studies.

More general discussions on the experimental uncertainty of measured values of the dynamic viscosity have been given by Sato [9] as well as Brooks et al. [10]. In the latter work experimental errors in the range of 9% are suggested.

The present work aims to suggest a method to estimate the experimental errors of experimental viscosity data in conjunction with the regular data treatment. In order to do so we will first briefly describe the typical experimental set-up and the following calculation procedure. To estimate the experimental error we will apply a statistical approach, i.e. we will simulate several experiments with the individual parameters varied within their uncertainties and discuss the influence thereof on the obtained dynamic viscosity.

2. Basics of the experimental procedure

Fig. 1 illustrates the principal design of an oscillating cup viscometer utilized for studies on liquid metals. The upper, water-cooled part contains the oscillation wire. The sample container is connected to the wire using a suitable suspension, which should be constructed to have a low momentum of inertia and provide a good heat shielding. The sample container is situated in the heater zone of the apparatus.

For the measurement, the sample container is set into a free but damped oscillatory motion, and the time course of the torsion angle ϕ can be obtained from the

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doi:10.1016/j.jallcom.2009.02.006

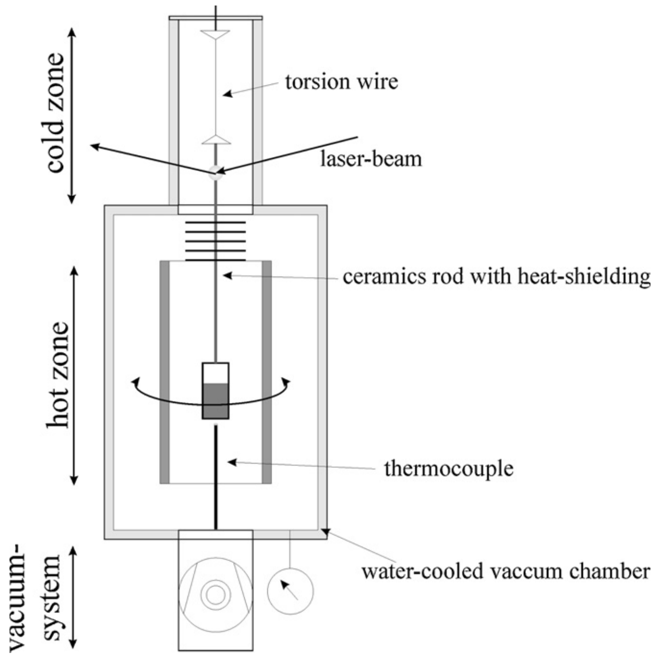

Fig. 1. Schematic view of an oscillating cup viscometer.

equilibria of torques:

$$J\ddot{\phi} + L\dot{\phi} = -D\phi. \tag{1}$$

Therein, J denotes the momentum of inertia of the whole pendulum, and D the restoring moment of the torsion wire. The damping parameter L is a function of the dynamic viscosity. We thus wish to determine the parameters T (time period), Λ (logarithmic decrement) and $\alpha = \arcsin(\phi_0/\phi_{\max})$ of the oscillation described by the function

$$\phi(t) = \phi_{\max} \sin\left(\frac{2\pi t}{T} + \alpha\right) \exp\left(-\Lambda \frac{t}{T}\right) \tag{2}$$

which complies with Eq. (1). Therefore, a laser beam is reflected by a mirror mounted to the pendulum and the reflected beam is monitored. If placed asymmetrically, i.e. at an angle $\phi_0 \neq 0$, a single photo-diode is sufficient to determine all parameters but the initial amplitude $\phi_{\max}$. The start of the actual measurement (corresponding to $t = 0$) is thus given by the initial crossing of the laser beam over the diode. The described situation is depicted in Fig. 2. As the deflected laser beam crosses the photo-diode at the times t_i and thus the relation

$$\hat{\phi} = \sin\left(\frac{2\pi t_i}{T} + \alpha\right) \exp\left(-\Lambda \frac{t_i}{T}\right) - \sin\alpha = 0 \tag{3}$$

holds. Averaging N periods into a single data-set, finding the parameters Λ, T and α can be realized by numerically finding minima of

$$X^2 = \frac{1}{2N+1} \sum_{i=0}^{2N} \hat{\phi}(t_i)^2 \tag{4}$$

and the residual $(X^2)^{1/2}$ can be used as a figure of merit.

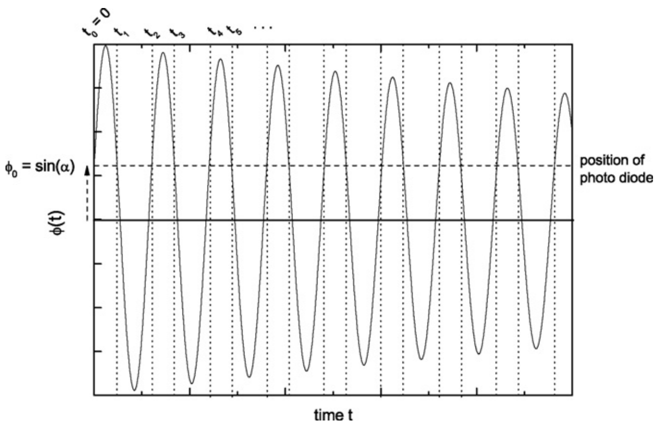

Fig. 2. Recording of the oscillation is done by a photo-diode placed at an elongation $\phi_0 \neq 0$. From a set of times t_i when the deflected laser beam passes the detector the oscillation parameters T, Λ and α can be deduced.

It should be mentioned that multi-diode arrangements [10] as well as position sensitive detectors [11] can be applied for the detection of the oscillation. Obviously this requires adapted fitting algorithms, however the further discussion can easily be transferred to these set-ups.

The calculation of the dynamic viscosity is preferably done by an absolute method, i.e. calibration with a reference liquid is not necessary. Suitable equations have been proposed by Roscoe and Bainbridge [12,13] as well as Beckwith, Newell and Kestin [14,15]. Though the resulting relations and their respective theoretical background are rather different, the results yielded in an experiment are nearly the same. For that reason we will stick to the Roscoe equation in the following. In an amended form given by Brooks et al. [16] the dynamic viscosity is given by

$$\eta = \left(\frac{J\Lambda}{\pi R^3 H Z}\right)^2 \frac{1}{\pi \rho T} \tag{5}$$

with

$$Z = \left(1 + \frac{R}{4H}\right) a_0 - \left(\frac{3}{2} + \frac{4R}{\pi H}\right) \frac{1}{p} + \left(\frac{3}{8} + \frac{9R}{4H}\right) \frac{a_2}{p^2}, \quad p = \left(\frac{\pi \rho}{\eta T}\right)^{1/2} R. \tag{6}$$

$$a_0 = 1 - \frac{3}{2}\frac{\Lambda}{2\pi} - \frac{3}{8}\frac{\Lambda^2}{4\pi^2}, \qquad a_2 = 1 + \frac{1}{2}\frac{\Lambda}{2\pi} + \frac{1}{8}\frac{\Lambda^2}{4\pi^2}$$

Therein, R denotes the crucible's inner radius and the height H of the samples within the cup is calculated from the sample's mass taking the temperature dependency of the mass density ρ into account. This equation has to be solved numerically using the experimentally determined oscillation parameters (Λ, T, α). However, the oscillation of the empty pendulum is damped too, and the respective logarithmic decrement α_0 has to be subtracted prior to the viscosity determination.

3. Uncertainty of the oscillation parameters Λ, T, α

The actual measured quantities are the times t_i containing experimental errors which lead to the residual in X^2 in Eq. (4). It is reasonable to assume that the errors occurring in the computer-based data acquisition are of statistical nature only and mathematical statistics is to be applied.

Generally speaking, any measured value $Y^{(k)}$ differs from its true value Y following a Gaussian distribution function with the full width at half maximum (FWHM) σ. Repeated (N fold) sampling of the same quantity yields an average value $\langle Y \rangle$ approaching the true one and an empirical variance:

$$s^2 = \frac{1}{N-1} \sum_{k=1}^{N} \left(Y^{(k)} - \langle Y \rangle\right)^2 \to \sigma. \tag{7}$$

Hence, following [17] the individual experimental value $Y^{(k)}$ is within an interval $Y \pm 2\,s$ around the true value with a probability of about 95.5%. This level of confidence will be applied in all further estimations.

On that basis, in order to estimate the uncertainties of the oscillation parameters the experiment can be modelled in the following way: an ideal set of t_i^0 (i.e. without experimental errors) is calculated following Eq. (3) for a given set (Λ, T, α). Disturbances $\Delta t \cdot N_{\mathrm{R}}$, with $-1 \leq N_{\mathrm{R}} \leq 1$ being random numbers following a Gaussian distribution, are added to the t_i^0 giving modelled experimental data including their uncertainties. In the experiments, a figure of merit $(X^2)^{1/2} \leq 10^{-3}$ is observed and we consequently choose $\Delta t = 1$ ms to obtain larger residuals in the following procedure for a pessimistic estimation. Finally, the oscillation function is fitted to that modified data. Multiple repetition of this procedure yields distributions of Λ, T, α around the chosen initial values following Gaussian functions and their widths can be addressed as half of the uncertainties of the respective parameters. An example is given in Fig. 3 for a common

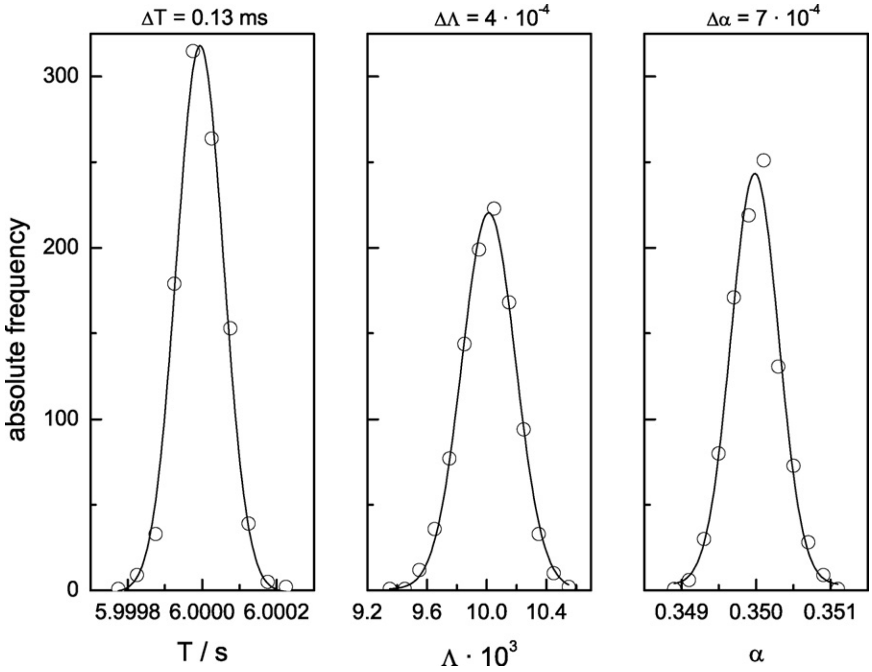

Fig. 3. Estimation of the uncertainties of the oscillation parameters (Λ, T, α). Repetitive modelling of the experimental situation yields distributions of the respective quantities with the FWHMs correlated to their experimental errors.

set of oscillation parameters: (Λ, T, α) = (10×10^{-3}, 6 s, 0.35) and 1000 repetitions of the modelling cycle described above.

The phase angle α is not necessary for the viscosity calculation, but for the control of the experiment. It shall therefore not be discussed further. The uncertainty of the period of the oscillation is rather small and its distribution function in Fig. 3 is narrow. In contrary, the logarithmic decrement is affected strongly: the distribution is broadened and the resulting relative error is in the order of 4%.

Applying the described procedure it is now possible to study the influence of the oscillation parameters as well as the number of oscillations on their errors. The following conclusions can be drawn: (i) the relative uncertainty of the period is negligible (i.e. less than 0.01%) for all combinations of parameters; (ii) the relative error of the logarithmic decrement only weakly depends on the number of oscillations if at least 10 oscillations are used for the calculation of an individual set of parameters and (iii) the relative error of the logarithmic decrement strongly depends on the logarithmic decrement itself, i.e. a weak damping cannot be measured to a high accuracy.

The latter dependence, calculated for $T = 6$ s and $\alpha = 0.35$ is shown in Fig. 4. In an experiment the logarithmic decrement is in the region of $(15–50) \times 10^{-3}$. However, as mentioned previously, the basic damping—usually is $\Lambda_0 \leq 2 \times 10^{-3}$—arising from the empty pendulum has to be measured in an individual experiment and subtracted for viscosity calculation. On the one hand, a small Λ_0 yields a high signal-to-background ratio, but considering Fig. 4 it will also introduce a rather high uncertainty. In order to take that into account, we shall represent the dependence $\Delta\Lambda/\Lambda(\Lambda)$ by means of the empiric relation

$$\frac{\Delta\Lambda}{\Lambda} = 0.34\Lambda^{-1.05}, \tag{8}$$

with the coefficient and exponent being determined by a least square fitting procedure. The result is represented by the solid line in Fig. 4 and is ready for use in the further estimation.

4. Uncertainty of the dynamic viscosity

The oscillation parameters, the sample's density and mass as well as the momentum of inertia and the crucible's inner radius are necessary quantities to calculate the dynamic viscosity following Roscoe's equation (5) and (6). As mentioned previously, the calculation is necessarily performed in a numerical way, i.e. from a starting guess of η a new approximate value is calculated, which again is used as a starting value in the next iteration step. In order to study the propagation of the experimental uncertainties, we make use of the same modelling procedure utilized in Section 3.

The following assumptions on the error limits of the input data appear to be reasonable: (i) the inner radius of the sample crucible can be easily measured to an accuracy of 0.3%. Allowing for thermal expansion as well as material changes during the heating cycle, we may thus estimate $\Delta R/R = 0.005$; (ii) the accuracy

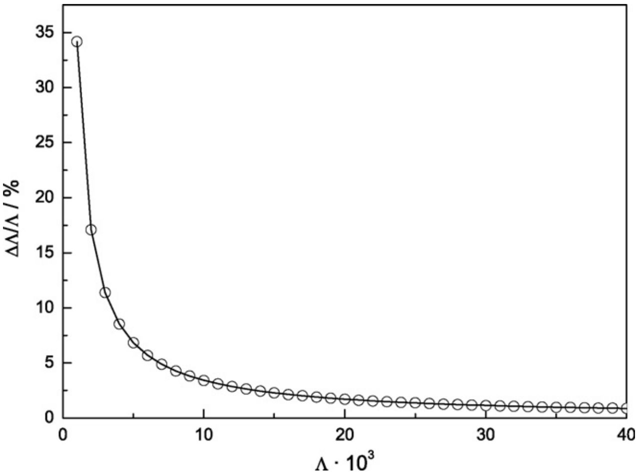

Fig. 4. The relative error of the logarithmic decrement depends strongly on the logarithmic decrement itself (circles). For the purpose of the further estimation the dependency is expressed by the empirical relation given in Eq. (8) (solid line).

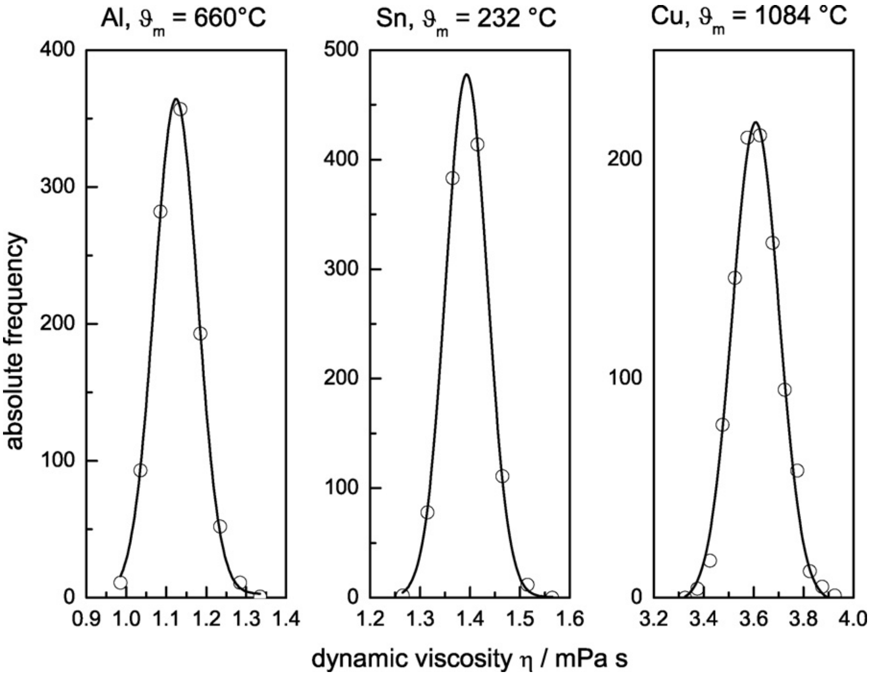

Fig. 5. The variation of the input parameters for Roscoe's equation within their respective range of uncertainty yields a distribution of viscosity values whose width is related to the overall uncertainty of the dynamic viscosity. The distributions are shown for liquid Al, Sn and Cu close to their respective melting temperature.

of weighing is rather high, too. However, during an experiment there might be minor evaporation of the sample material, which we take into account by estimating $\Delta m/m = 0.005$. For the investigation of materials with a high vapour-pressure suitable sealed ampoules are to be utilized; (iii) the momentum of inertia of the whole pendulum can be determined in calibration experiments to an accuracy of 0.4%; (iv) in most cases no information on experimentally determined mass densities of liquid alloys is at hand and the alloy's density is to be estimated from those of the pure liquid constituents. We estimate $\Delta\rho/\rho = 1\%$; (v) finally, the relative uncertainty of the logarithmic decrement is given by Eq. (8).

Modifications again following a Gaussian distribution are applied to the input data for Eqs. (5) and (6) within these limits. The dynamic viscosity is then calculated from the modified values. Consequently, multiple repetition of this procedure yields a Gaussian distribution of values of the dynamic viscosity with its maximum equalling to the viscosity obtained from the undisturbed, real experimental data. Its width can again be addressed as half of the experimental uncertainty. This procedure is easily to be adopted for each recorded value of the dynamic viscosity. In Section 5, we shall apply this approach for representative experiments carried out for pure liquid elements.

5. Selected examples: pure liquid Al, Sn and Cu.

For the demonstration of the error estimation procedure, we select representative experiments carried out for pure liquid Al (purity 99.9%), Sn (purity 99.999%) as well as Cu (purity 99.99%) in the temperature range between 1200 °C and solidification at a cooling rate of 1 K/min. The samples have been kept in boron nitride crucibles with an internal diameter of 22 mm and throughout the experiments a pressure of less than 5×10^{-5} mbar has been obtained. Furthermore, remaining oxides on the surface of the sample material are usually reduced by the presence of graphite in the hot zone of the apparatus.

The volume of the sample in the solid state was about 6 cm^3 in each case. The necessary mass density has been taken from the compilation in Ref. [18].

Fig. 5 depicts the distribution of the viscosity values obtained by multiple repetition of the calculation procedure outlined above for all three elements close to their respective melting temperature. Again, the distribution curves obtained follow the Gaussian function and thus the application of the statistical approach seems plausible. The peak positions correspond to the results of solving Roscoe's equation with the undisturbed data and the uncertainty of the individual value is to be taken from the full width at half maximum s_η applying the common relation $\Delta\eta = 2\,s_\eta$. A digest on the obtained figures for the examples is given in Table 1.

Going further, the uncertainties for all measured values of the dynamic viscosity with respect to the temperature can be calculated. This is done repeatedly in order to analyse the reproducibility of the approach. Three runs each experiment are depicted in Fig. 6 by different symbols and it can be seen that the estimated relative errors of the dynamic viscosity coincide well and are of the order of 5% in the case of liquid Cu and rise up to 10% for liquid Al. Higher dynamic viscosities are obviously measured to a higher accuracy than lower ones. The same relation holds for the dependence of the experimental uncertainty on the liquid's density, as can be seen from the comparison of liquid Al and Sn. Both liquids show a comparable dynamic viscosity, however the density of liquid Al is reduced by almost a factor 3 compared to that of liquid Sn. Consequently, the resulting lower logarithmic decrement caused by the Al melt cannot be measured to the same accuracy.

Table 1
Dynamic viscosity measured for the three liquid metals Al, Sn and Cu close to their solidification temperature together with the calculated empirical variance and relative uncertainties.

Element	ϑ_m (°C)	$\eta(\vartheta_\mathrm{m})$ (mPa s)	$s_\eta(\vartheta_\mathrm{m})$ (mPa s)	$\Delta\eta/\eta(\vartheta_\mathrm{m})$ (%)
Al	660	1.12	0.05	9.6
Sn	232	1.39	0.04	6.0
Cu	1084	3.61	0.09	5.1

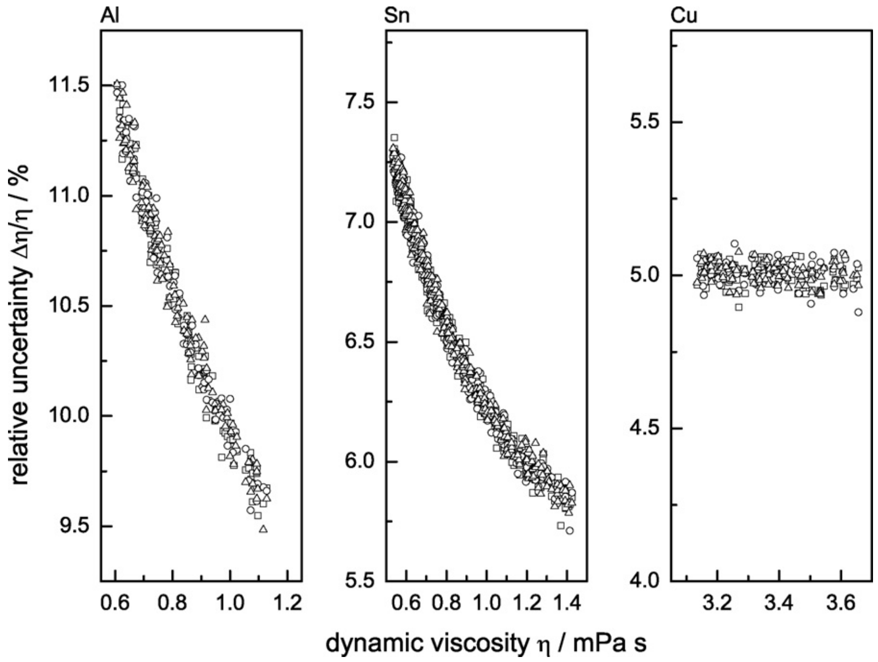

Fig. 6. Calculated relative uncertainty for liquid Al, Cu and Sn. The three runs of the modelling procedure carried out are depicted by different symbols and coincide well.

6. Conclusions

The aim of the present work is to present a statistical approach to estimate the experimental errors occurring in measurements of the dynamic viscosity by means of the oscillating cup technique. With respect to its uncertainty, the most important of the actual measured quantities is the logarithmic decrement of the torsional oscillation caused by the liquid's internal friction. An empirical relation $\Delta\Lambda/\Lambda(\Lambda)$ has been obtained and the error propagation during the calculation of the dynamic viscosity has been investigated.

It is to be stated that the relative uncertainties are in the range 5–10% and are to be reduced by ensuring a sufficiently large logarithmic decrement. The range of the experimental errors obtained with our method corresponds well to earlier error estimations, e.g. those performed by Kehr et al. [11] as well as Brooks et al. [10].

The presented approach takes into account both the statistical nature of the errors occurring during the determination of the oscillation parameters and their systematic propagation during the further data treatment. It is readily to be involved in the real-time calculation procedure including fitting of the oscillation parameters and solving of Eqs. (5) and (6) and thus may provide a basis for a realistic discussion of changes depending on temperature, composition as well as the thermal history of the alloy under investigation.

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