Acoustic modelling in view of a determination of the Boltzmann constant within 1 ppm for the redefinition of the kelvin

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Abstract. iMERA/Euromet Project 885 is co-ordinating European effort towards a new determination of the Boltzmann constant $k_B$ to within 1 ppm with the aim of redefining the unit of thermodynamic temperature. This project will enable the National Physical Laboratory to perform primary thermometry in the region of $\text{–}40$ °C (Hg) to 156 °C (In) with sub-millikelvin uncertainties by 2012. The chosen technique relies on determining the speed of sound in a monatomic gas. Using the radial acoustic modes of a spherical resonator, consisting of a copper shell and filled with argon or helium, the speed of sound can be measured with great precision and from this measurement the Boltzmann constant can be inferred. This project draws on expertise in dimensional, density, microwave and acoustic measurements at the state-of-the-art. In order to gain further understanding of the experimental configuration a vibro-acoustic model has been developed using the finite element method. Initial calculations were carried out to ensure that predictions of the resonant frequency could be made with the required precision by comparing against an analytical model of a spherical shell filled with a gas. A more elaborate model better representing the experimental configuration was then developed. Thermo-viscous effects close to the fluid-structure boundary were accounted for using a linear acoustic formulation, from which a normal incidence admittance boundary condition was derived and imposed on the inner surface of the resonator. Acoustic pressure, particle velocity and temperature variation as a function of position may be obtained within the gas as a function of frequency. It is therefore possible to investigate how changes in the configuration affect the frequency of radial modes. It is hoped that this approach will shed a better understanding of the underlying complex physical phenomena allowing a minimization of the overall uncertainty.

1. Background
One aim of fundamental metrology is to provide a system of measurement in which the units of measurement are stable in time and space. To this end, it is preferable that the definitions of the units of measurement do not refer to any specific artefact or substance. Instead, they should relate the units
to immutable constants of nature. This work was undertaken as part of a project to replace the current definition of the unit of temperature with a definition based on the Boltzmann constant $k_B$.

The previous most accurate determination of the Boltzmann constant was made by Moldover et al [1] at National Institute of Standards and Technology (Gaithersburg, MD, USA) using a spherical acoustic resonator. This yielded an uncertainty in $k_B$ of $\pm 1.8$ ppm with a coverage factor of one. After reviewing possible techniques, the National Physical Laboratory (Teddington, UK) has chosen to again use a spherical acoustic resonator technique, but updating the experiment with 30 years of improved technology. In this experiment, the speed of sound is determined by measuring the resonance frequencies of a dimensionally characterised resonator at the triple point of water (0.01 °C). The frequencies are measured at a number of pressures, and the zero pressure speed of sound inferred. The experiment is conducted using isotopically and chemically analysed pure argon. From this result $k_B$ is inferred using:

$$k_B = \frac{M c_0^2}{\gamma N_A T}$$  \hspace{1cm} (1)

where
- $M$ is the molar mass with isotopic corrections
- $c_0$ is the zero-pressure speed of sound
- $\gamma$ is the ratio of ideal gas heat capacities
- $N_A$ is the Avogadro constant
- $T$ is the thermodynamic temperature.

By examining the large number of factors which must be considered in order to achieve the required level of uncertainty (i.e. within 1 ppm), the interaction of the acoustic modes with the vibrational modes of the shell were identified as being of key importance. An analytical theory exists for the interaction of a uniform spherical shell with a resonating fluid [2],[3], but the effects of shell motion are considered to be independent of thermo-viscous boundary layer corrections. Although efforts are made to include effects due to imperfect resonator geometry using perturbation theory, the experimentally observed vibrational spectra of spheres are actually much more complex than predicted by [2],[3].

Joly et al [4] developed a formulation of linear acoustics in thermo-viscous fluids at rest in the form of a set of two coupled equations for particle velocity and temperature variation. In the case of normal local incidence, which is closely approximated during a breathing mode in a spherical resonator, an expression for the specific acoustic admittance at the fluid/structure interface may be obtained.

As part of the work described in this paper, in order to further understand the complex thermal, acoustical and vibrational interactions which occur when a gas-filled spherical resonator is subjected to an acoustic excitation, the acoustic admittance boundary condition described in [4] was used in an acoustic boundary element formulation coupled to the inner wall of the resonator shell. The latter was modelled using the finite element method in order to allow more complex geometries to be examined. Once the vibro-acoustic finite element analysis is completed with the thermo-viscous boundary condition in place, shell displacements can then serve as input quantities to the coupled set of equations described in [4] and the temperature and particle velocity variations within the resonator may be obtained.

This paper outlines the initial stages of this analysis of spherical gas-filled resonators. It is understood that this work will be expanded upon and will form the basis for a more in-depth paper.
2. Experimental configuration
Practically it is impossible to create a perfect spherical resonator and real resonators used experimentally are far from spherical. It is the need to extend our understanding to real resonators which motivates this work. For the radial acoustic modes (which are the main focus of our work) the perturbations from ideal spherical resonator values are affected by two types of shell interaction: accidental interactions with complex shell modes which cause anomalous readings at one particular frequency (corresponding to one particular temperature); and interactions with the breathing mode of the shell, which will systematically affect all data, even data taken at frequencies far from the breathing mode. It is this systematic effect that we are seeking to evaluate.

Externally resonators differ from perfect spherical shells in three ways. Firstly they must be held in some way, typically by a neck which additionally provides a thermally conducting path to a temperature bath. Secondly the resonators are manufactured from two hemispheres and then (usually) bolted together resulting in a potential change in elastic parameters at the equatorial plane. Finally the thickness of the shell is not uniform, there being typically some kind of cylindrical flange around the equatorial seam. All of these deviations from sphericity mean that a pure radial acoustic mode within the cavity gives rise to non-uniform shell modes, and so couples not just to the breathing mode, but also to other (generally lower frequency) shell modes.

In the experiments we are seeking to model, the sphere is held tightly by its neck in contact with a long steel post holder and secured inside a cylindrical stainless steel pressure vessel. The entire vessel is immersed inside a bath of anti-freeze at approximately the triple point of water. The arrangement is sufficient to keep the temperature within 10 mK of the triple point of water, with drift rates less than 0.1 mK per minute. The configuration is shown in Figure 1 (a) and details of the resonator dimensions are shown in Figure 1 (b).

3. Method
In order to accurately compute the coupled vibration of the shell and acoustic field inside the spherical cavity filled with a thermo-viscous fluid, the system is modelled in two steps. In a first vibroacoustic model (Section 3.1), the vibration of the shell is computed whilst accounting for the acoustic coupling, by using a potential pressure formulation for acoustics and where thermo-viscous boundary effects are accounted for by an admittance-like condition. In a second model (Section 3.2), the full solution for thermo-viscous acoustics is computed in the fluid domain, according to the vibrating motion of the shell boundaries of the cavity.

3.1. Vibroacoustic modelling
The vibroacoustic modelling described in this section was carried out using the PAFEC (Program for Automatic Finite Element Calculations) finite element software. The shell was modelled using axisymmetric isoparametric quadratic elements. The gas inside the resonator was modelled using an interior boundary element formulation. The fluid inside the cavity was excited using a point source placed on the axis of symmetry 3.24 cm from the centre of the sphere on the neck side of the resonator. It should be noted that this position was arbitrarily chosen pending a more realistic description of the acoustic source. Harmonic excitation is assumed. The solution for the acoustical analysis was obtained using a surface Helmholtz formulation using the Burton-Miller method [5]. A specific acoustic admittance condition is imposed on the boundary element which takes into account the thermo-viscous boundary layer effects. This expression is described in further detail in [4]. The shell properties are those of copper and the gas properties are those of Argon. The fluid loading at the resonator exterior wall has been neglected in this analysis, as initial modelling has suggested that accounting for this does not affect the pressures inside the cavity by an appreciable amount in the context of the uncertainties required here.
Figure 2 shows the finite element mesh generated to carry out the finite element analysis. The resonator also comprises an equatorial belt as well as a “neck”, which are straightforward to include into the analysis, and which will cause the shell to displace in ways that differs from a uniform thickness resonator. At this stage, no mechanical restraints have been implemented.

The properties of argon at the triple point of water are displayed in table 1. These have been obtained from [6].

| Property                                      | Value                      |
|-----------------------------------------------|----------------------------|
| Equilibrium density $\rho_0$                  | 1.783890 kg m$^{-3}$       |
| Speed of sound $c_0$                          | 307.8623 m s$^{-1}$       |
| Shear viscosity $\mu$                         | $2.0984 \times 10^{-5}$ Pa s |
| Bulk viscosity $\eta$                         | 0                         |
| Thermal conductivity $\lambda$                | 0.01641 W m$^{-1}$ K$^{-1}$ |
| Specific heat capacity at constant pressure $C_p$ | 521.845 J kg$^{-1}$ K$^{-1}$ |

The properties of the copper wall are displayed in table 2.

| Property                                      | Value                      |
|-----------------------------------------------|----------------------------|
| Density $\rho_w$                              | 8933 kg m$^{-3}$ [7]       |
| Poisson’s ratio $\nu_w$                       | 0.343 [8]                  |
| Young’s modulus $E_w$                         | 129.8 GPa [8]              |
| Thermal conductivity $\lambda_w$              | 402.93 W m$^{-1}$ K$^{-1}$ [9] |
| Heat capacity per unit mass $C_w$             | 379.20 J kg$^{-1}$ K$^{-1}$ [9] |

From the solution described in [2],[3], it can be found that, for a constant shell thickness gas-filled spherical resonator with the properties outlined in figure 1 and in tables 1 and 2, a radial mode exists around 4380 Hz. By sweeping the frequency of excitation around an interval centred on 4380 Hz, a local maximum for the spatial average of the RMS acoustic pressure at the resonator wall can be found, as shown in figure 3. This maximum is found to occur at 4379.523 Hz.

A plot of the maximum structural deformations is shown in figure 4. It should be noted that the values obtained for the acoustic pressures inside the cavity inside the acoustic display element mesh region are approximate. Although the acoustic loading on the resonator wall resulting from thermo-viscous boundary layer effects is accounted for, this section of the analysis only aims to predict the shell displacements and the acoustic pressures at the resonator wall. Nevertheless, this provides some guidance as to how the fluid inside the cavity behaves, as thermo-viscous effects further away from the wall are likely to be negligible.

3.2. Thermo-acoustic modelling
This subsection contains a brief overview of the formulation described in [4]. The fluid is assumed to be Stokesian, which implies that the stress is proportional to the rate of strain and the heat flux is proportional to the temperature gradient. The fluid is assumed to be homogeneous and at rest, with a linear behaviour for the differential form of its equation of state. When accounting for the phenomenological complementary relationships of the fluid, the fundamental laws of mechanics take the following linearised form.

(i) Linearised Navier-Stokes equation:

\[ \rho_0 \frac{\partial \tilde{u}}{\partial t} + \tilde{\nabla} p - \rho_0 c_0 l_v \tilde{\nabla} (\tilde{\nabla} \cdot \tilde{u}) + \rho_0 \omega^l \tilde{\nabla} \times \tilde{\nabla} \times \tilde{u} = 0 \]  \hspace{1cm} (2)

where the viscous characteristic lengths are given by:

\[ l_v = \frac{1}{\rho_0 c_0} \left( \eta + \frac{4}{3} \mu \right) \]  \hspace{1cm} (3)

\[ l_v^l = \frac{\mu}{\rho_0 c_0} \]  \hspace{1cm} (4)

(ii) Conservation of mass:

\[ \frac{\partial \rho^l}{\partial t} + \rho_0 \nabla \cdot \tilde{u} = 0 \]  \hspace{1cm} (5)

(iii) Conservation of energy:

\[ \rho_0 T_0 \frac{\partial s}{\partial t} - \rho_0 c_0 C_p l_h \nabla \cdot (\tilde{\nabla} \tau) = 0 \]  \hspace{1cm} (6)

where the thermal characteristic length is given by:

\[ l_h = \frac{\lambda}{\rho_0 c_0 C_p} \]  \hspace{1cm} (7)

The symbols have the following meaning.
- \( \rho^l \) is the density variation
- \( p \) is the acoustic pressure
- \( \tilde{u} \) is the particle velocity vector
- \( s \) is the entropy variation per unit mass
- \( \tau \) is the temperature variation
- \( T_0 \) is the ambient temperature

The thermodynamic state laws of the fluid can be used to express thermodynamic quantities with two independent variables: \( p \) and \( \tau \). The equations of state of the gas then become

\[ \rho^l = \frac{\rho_0 c_0^2}{1 + \beta \tau} \]  \hspace{1cm} (8)
\[ s = \frac{C_p}{T_0} \left( \tau - \frac{\gamma - 1}{\beta^\gamma} p \right) \]  

(9)

where

\[ \hat{\beta} = \left( \frac{\partial P}{\partial T} \right)_v = \sqrt{\frac{(\gamma - 1)\rho_0 c_0^2 C_p}{\gamma^2 T_0}} \]  

(10)

and \( \gamma \) is the ratio of specific heats.

By combining (2), (5), (6), (8) and (9) and eliminating \( p \), \( s \), and \( \rho' \) and expressing the result in terms of \( \tilde{u} \) and \( \tau \), a system of two coupled equations involving \( \tilde{u} \) and \( \tau \) is obtained. This system can be solved numerically by analogy with elastodynamic and thermal problems.

\[ -\frac{\partial^2 \tilde{u}}{\partial t^2} + \left( \frac{c_0^2}{\gamma} + c_0 l_\gamma \frac{\partial}{\partial t} \right) \nabla (\nabla \cdot \tilde{u}) - c_0 l_\gamma \frac{\partial}{\partial t} (\tilde{\nabla} \times \tilde{\nabla} \times \tilde{u}) - \frac{\hat{\beta}}{\rho_0} \frac{\partial}{\partial t} (\bar{\nabla} \tau) = 0 \]  

(11)

\[ \frac{\partial \tau}{\partial t} - \gamma l_h c_0 \nabla \cdot \tilde{\nabla} \tau + \frac{\gamma - 1}{\gamma \hat{\beta}} \rho_0 c_0^2 \nabla \cdot \tilde{u} = 0 \]  

(12)

Using the shell axial and radial velocities of the inner wall as input quantities to the thermo-acoustic code at the Laboratoire d’Acoustique de l’Université du Maine (Le Mans, France) based on equations (11) and (12), temperature and particle velocity variations inside the resonator were obtained during the course of one acoustic cycle. Figure 5 shows the axisymmetric temperature variation inside the cavity. Figure 6 shows the particle velocity vector variation.

4. Discussion

A comprehensive model which aims to include vibrational, acoustical and thermal phenomena in spherical gas-shaped resonators, has been developed and one particular configuration has been investigated. Of all the assumptions that have been made that of axisymmetry of the resonator is perhaps the most controversial. The internal surface is in fact not spherical, but carefully manufactured in the shape of a triaxial ellipsoid, a shape with no rotational symmetry about any axis. Additionally the sphere is manufactured as two hemispheres which are held together by steel bolts. This combined with acoustic excitation which is, in practice, potentially non-axisymmetric, implies that many modes which exists in the experimental device will not be predicted by the model here. A full three-dimensional analysis would be required for this, but this is beyond the scope of this work.

The vibroacoustic analysis centred around 4380 Hz shows that, despite having selected a frequency of excitation corresponding to a radial mode, the shell displacements do not exhibit a breathing mode-like behaviour. Nevertheless, the acoustic display elements in figure 4 as well as the temperature and particle velocity variations in figures 5 and 6 show that the fluid is clearly displaying radial mode behaviour inside the cavity. There is therefore clearly modal interaction between the (0,2) radial mode and the (3,1) non-radial mode, as shown in figure 3.
At this stage, a point source of unit source strength along the axis of symmetry has been used to generate the acoustic excitation. This does not detract from predicting features of how the resonator behaves, given that linearity is assumed. This methodology however needs to be refined to include some features of the emitting transducer, perhaps initially in the form of a rigidly vibrating plane piston, so as to ensure that the acoustic pressures and resulting temperature variations are of the order of what can be measured experimentally.

It should be noted that the admittance boundary condition described in [4] was implemented in the vibroacoustic model (Section 3.1) for local normal incidence, because of the shape of breathing modes in spherical resonators. It accurately accounts for the thermo-viscous acoustic boundary layers effects in the first numerical vibroacoustic model.

5. Conclusions
A fully coupled axisymmetric vibrational, acoustical and thermo-viscous model of a spherical resonator was described in this paper. This model represents an improvement on prior ones given that it enables the shell dynamics to be simulated using the finite element method and therefore allows for more complex geometries to be investigated and to determine their impact on thermo-acoustic behaviour inside the cavity. This model is intended to serve as a design and analysis tool to gain further insight into the complex multi-physics of the behaviour of gas-filled resonators used for the measurement of thermophysical properties. It is hoped that it will help assist further measurements and aid the design of an improved resonator.

References
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Coolant at approximately 0 °C

Figure 1 (a) Schematic arrangement of the resonator within its pressure vessel. (b) Details of the resonators dimensions
Figure 2 – Finite element mesh used for vibroacoustic analysis.
Figure 3 – Spatial RMS value of acoustic pressure per unit source strength at resonator wall as a function of frequency.
Figure 4 – Maximum structural deformations resulting from acoustic excitation at 4379.523 Hz
Figure 5 – Acoustic perturbation due to the shell motion: temperature variation (colour map: temperature variation)
http://www.univ-lemans.fr/~nJoly/sphere_Ar/particle_V.gif

Figure 6 – Acoustic perturbation due to the shell motion: particle velocity (colour map: modulus, arrows: orientation)