Acoustic field in cavities filled with thermo-viscous binary gas mixtures, determination of gas mixtures thermophysical properties

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The acoustic behaviour in thermo-viscous gas mixtures involves deviations from the adiabatic and laminar movement in pure gases, which results from the influence of several diffusive fields, namely shear, entropic, and concentration variation fields, taking energy to the acoustic field itself. Actually, a strong coupling between these fields occurs inside boundary layers while their effects appear to be additive processes in the bulk of the medium.
Although recent literature on this subject leads to results of interest, these results still have limitations because they do not provide complete solutions for the propagation and diffusive fields in and out of the boundary layers.
The aim of this work is to provide such complete solutions in the whole domains considered, in order to get a better analytical understanding of the fields above mentioned in closed cavities and ducts. More particularly here, we consider the possible use of acoustic methods in spherical cavities for the accurate determination of binary gas mixtures thermophysical properties (speed of sound, first acoustic virial coefficient, thermal conductivity, mutual diffusion coefficient), that have been greatly improved during the past two decades.

1 Introduction

Several approaches to describe the behaviour of binary gas mixtures, starting from hydrodynamical and thermodynamical fundamental equations [1] or from the point of view of kinetic theory [2], have been carried out in the past, and emphasize that the thermophysical properties of a gas mixture are not reduced to a simple weighted mean of the single properties of each component, but that they result from the cross-interactions between the different components of the mixture. Particularly, the phenomena involved are mutual diffusions of the components depending on the main parameters involved in an acoustic field, including concentration variations actually due to pressure variations (barodiffusion) and temperature variations (thermal diffusion). Providing some expressions of mutual diffusion effects on the acoustic propagation has been the purpose of several works during the last decades [3, 4].

More recent works [5, 6], which enabled a description of the mutual diffusion effects coupled to the viscous and thermal effects in the boundary layers, were specifically dedicated to applications using gas mixtures or humid air. However, today the analytical procedure whereby a general analytical model of acoustic behaviours of a binary gas mixture, near and far from the boundaries, is still missing.

The aim of the present work is to provide such analytical model for binary gas mixtures, starting from the hydrodynamical and thermodynamical fundamental equations (§ 2), leading to solutions in infinite domain (§ 3) and close to walls together (§ 4). The theoretical results obtained are used to estimate the possibilities of determining some thermophysical properties of binary gas mixtures from the acoustic resonance properties of a gas-filled spherical cavity (§ 5).

2 Linearised fundamental equations of acoustics in binary gas mixtures

The gas mixture considered is composed of two gases named 1 and 2 of respective molar mass M_1 and M_2, molar fractions x and $(1-x)$ and static mass concentrations C_0 and $(1-C_0)$. The static temperature, pressure and density of the gas are denoted T_0, P_0 and ρ_0 respectively, and the total molar mass of the gas mixture is given by

$$M = \frac{M_1 M_2}{M_1 + (M_2 - M_1)C_0} = xM_1 + (1-x)M_2 \,. \tag{1}$$

The acoustic propagation in the gas mixture involves the particle velocity $\vec{v}$, the pressure variation p, the temperature variation τ, the density variation ρ', and the concentration variation c.

Most of the hydrodynamical and thermodynamical equations in binary gas mixtures presented in this section are adapted from the formalism of Landau and Lifshitz [1].

The Navier-Stokes equation

$$\rho_0 \frac{\partial \vec{v}}{\partial t} = -\vec{\nabla}\, p + \left(\eta + \frac{4}{3}\mu\right)\vec{\nabla}\,(\vec{\nabla}\,.\vec{v}) - \mu\vec{\nabla}\,\wedge\vec{\nabla}\,\wedge\vec{v}, \tag{2}$$

ρ_0, η and μ are respectively the mass density, the bulk and shear viscosity coefficients of the mixture.

The mass conservation equation for the gas mixture

$$\frac{\partial \rho'}{\partial t} + \rho_0 \vec{\nabla}\,.\vec{v} = 0 \,. \tag{3}$$

The mass conservation equation for the component "1" of the gas mixture

$$C_0 \frac{\partial \rho'}{\partial t} + \rho_0 \frac{\partial c}{\partial t} + C_0 \rho_0 \vec{\nabla}\,.\vec{v} + \vec{\nabla}\,.\vec{\imath} = 0 \,, \tag{4}$$

where the concentration flux density is written as

$$\vec{\imath} = -\rho_0 D \frac{M_1 M_2}{M^2}\left[\frac{M^2}{M_1 M_2}\vec{\nabla}\,c + \frac{k_P}{P_0}\vec{\nabla}\,p + \frac{k_T}{T_0}\vec{\nabla}\,\tau\right], \tag{5}$$

D, k_T, and k_P being respectively the diffusion coefficient, and the thermal and baro diffusion ratios, the barodiffusion ratio being exactly determined by

$$k_P = x(1-x)\frac{M_2 - M_1}{M} \,. \tag{6}$$

The Fourier's equation in the gas mixture

$$\left(\frac{\partial}{\partial t} - \frac{\lambda}{\rho_0 C_P}\Delta\right)\tau = \frac{1}{\rho_0 C_P}\frac{\partial p}{\partial t} + \frac{P_0}{\rho_0 C_P}k_T \frac{\partial c}{\partial t} \,, \tag{7}$$

where λ is the thermal conductivity of the gas mixture, and C_P its isobaric specific heat capacity.

Then, using the following differential state equation for the mixture (ideal gas),

$$\frac{\partial \rho'}{\partial t} = \rho_0\left[\frac{1}{P_0}\frac{\partial p}{\partial t} - \frac{1}{T_0}\frac{\partial \tau}{\partial t} - \frac{k_P}{x(1-x)}\frac{M^2}{M_1 M_2}\frac{\partial c}{\partial t}\right], \tag{8}$$

and the following identities and notations (a_0 being the adiabatic speed of sound in the gas mixture)

$$T_0/P_0 = 1/\hat{\beta}, \quad \rho_0 C_P = \frac{\gamma\hat{\beta}}{\gamma - 1} \,, \quad \gamma P_0 = \rho_0 a_0^2 \,, \tag{9a}$$

$$b = \frac{M^2}{M_1 M_2}c \,, \quad \zeta_P = \frac{k_P}{x(1-x)} \,, \quad \zeta_T = \frac{k_T}{x(1-x)} \,, \tag{9b}$$

$$\ell_D = \frac{D}{a_0} \,, \quad \ell_h = \frac{\lambda}{\rho_0 a_0 C_P} \,,$$

$$\ell'_v = \frac{\mu}{\rho_0 a_0} \,, \quad \ell_v = \frac{1}{\rho_0 a_0}\left(\eta + \frac{4}{3}\mu\right), \tag{9c}$$

the linearised fundamental set of acoustic equations in binary gas mixtures is given by

$$\frac{1}{a_0}\frac{\partial \vec{v}}{\partial t} = -\frac{1}{\rho_0 a_0}\vec{\nabla}\, p + \ell_v \vec{\nabla}\,(\vec{\nabla}\,.\vec{v}) - \ell'_v \vec{\nabla}\, \wedge \vec{\nabla}\, \wedge \vec{v}, \quad (10)$$

$$\vec{\nabla}\,.\vec{v} = \frac{\gamma\hat{\beta}}{\rho_0 a_0^2}\frac{\partial \tau}{\partial t} - \frac{\gamma}{\rho_0 a_0^2}\frac{\partial p}{\partial t} + \zeta_P \frac{\partial b}{\partial t}\,, \quad (11)$$

$$\left(\frac{1}{a_0}\frac{\partial}{\partial t} - \ell_h \Delta\right)\tau = \frac{\gamma - 1}{\gamma\hat{\beta}}\frac{1}{a_0}\frac{\partial}{\partial t}(p + P_0\zeta_T b)\,, \quad (12)$$

$$\left(\frac{1}{a_0}\frac{\partial}{\partial t} - \ell_D \Delta\right)b = \frac{\ell_D \gamma}{\rho_0 a_0^2}\left[k_P \Delta p + k_T \hat{\beta}\Delta\tau\right]. \quad (13)$$

In equations (12) and (13), the pressure variation p can be expressed as the sum of a perturbed acoustic pressure p_a and an excess of pressure variation $p_h + p_b$ associated to the heat (index h) and the concentration (index b) diffusion movements. It is the same for the temperature variation $\tau = \tau_a + \tau_h + \tau_b$.

In equations (11-13), the thermal diffusion and barodiffusion ratios both vanish in pure gases, leading to the well-known equations of acoustics in thermo-visous fluid.

3 Acoustic field in infinite domain

Far from boundaries, the shear viscosity can be neglected in equation (10). Then, applying the operator $\left(\frac{1}{a_0}\frac{\partial}{\partial t} - \ell_h \Delta\right)$ on equations (10) and (11), accounting for equations (12) to remove the variable τ, identifying the two resulting equations to remove the variable $\vec{v}$ leads to an equation involving the pressure and concentration variations p and b. Then making use of equation (13) to remove the variable b, and finally employing the factorization and approximations suggested in reference [7] assuming that $p = p_a + p_h + p_b$, and $\tau = \tau_a + \tau_h + \tau_b$ and $\ell_{D,h,v}\Delta p_a << 1/a_0 \partial p_a/\partial t$ leads lengthy but straightforwardly to

$$\frac{1}{a_0^2}\frac{\partial^2}{\partial t^2}p_a - \left[1 + \left(\ell_{vh} + \gamma x(1-x)\zeta^2 \ell_D\right)\frac{1}{a_0}\frac{\partial}{\partial t}\right]\Delta p_a \approx 0\,, \quad (14)$$

$$\left(\frac{1}{a_0}\frac{\partial}{\partial t} - \ell_h \Delta\right)p_h \approx 0\,, \quad \left(\frac{1}{a_0}\frac{\partial}{\partial t} - \ell_D \Delta\right)p_b \approx 0\,, \quad (15)$$

where

$$\ell_{vh} = \ell_v + (\gamma - 1)\ell_h\,, \quad \zeta = \zeta_P + \frac{\gamma - 1}{\gamma}\zeta_T\,. \quad (16)$$

Considering an harmonic motion of angular frequency ω, equation (14) can be written as

$$(\Delta + k_a^2)p_a \approx 0\,, \quad (17a)$$

the acoustic wavenumber k_a being given by

$$k_a^2 = k_0^2[1 - \mathrm{j}\,k_0(\ell_{vh} + \gamma x(1-x)\zeta^2\ell_D)]\,. \quad (17b)$$

The term $\ell_D\,\gamma x(1-x)\zeta^2$ related to the mutual diffusion phenomena vanishes in a pure gas, leading to the well-known results for the wavenumber k_a in a thermo-viscous gas [7].

4 Acoustic field close to the boundaries

The domain considered here is close to a rigid wall. The coordinate normal to the wall inwardly directed is denoted u, the origin (denoted s) being on the wall, and the coordinates tangent to the wall are denoted (w_1, w_2) or $\vec{w}$ for the both of them. The following appropriate approximations are assumed on the fundamental linear equations (10–13), giving an accurate description of the small amplitude disturbances inside the boundary layers: *i*- the component normal to the wall of the acoustic velocity v_u is much lower than the tangential components v_{w_i}, *ii*- the spatial variation of the acoustic velocity is much higher in the normal direction u than in the tangential directions w_i.

According to these assumptions, for an harmonic motion (angular frequency ω), the particle velocity and the pressure, temperature and concentration variations are described by the following set of equations

$$\left(1 + \frac{1}{k_v^2}\frac{\partial^2}{\partial u^2}\right)\vec{v}_w \approx -\frac{1}{\mathrm{j}\,\omega\rho_0}(1 + \mathrm{j}\,k_0\ell_v)\vec{\nabla}_w p + a_0\ell_v\,\zeta_P\vec{\nabla}_w b\,, \quad (18)$$

$$\frac{\partial}{\partial u}v_u + \vec{\nabla}_w.\vec{v}_w = -\mathrm{j}\,\omega\frac{\gamma}{\rho_0 a_0^2}(p - \hat{\beta}\tau) + \mathrm{j}\,\omega\,\zeta_P b\,, \quad (19)$$

$$\left(1 + \frac{1}{k_h^2}\Delta\right)\tau \approx \frac{\gamma - 1}{\gamma\hat{\beta}}(p + P_0\zeta_T b)\,, \quad (20)$$

$$\left(1 + \frac{1}{k_D^2}\Delta\right)b \approx -\frac{\gamma}{\rho_0 a_0^2}\frac{1}{k_D^2}\left[k_P\Delta p + \hat{\beta}k_T\Delta\tau\right], \quad (21)$$

where

$$k_v^2 = -\frac{\mathrm{j}\,k_0}{\ell'_v}\,, \quad k_h^2 = -\frac{\mathrm{j}\,k_0}{\ell_h}\,, \quad k_D^2 = -\frac{\mathrm{j}\,k_0}{\ell_D}\,, \quad (22)$$

and are subjected to the following boundary conditions

$$\vec{v}_w(u, \vec{w}) = 0 \qquad u = s \qquad \forall\vec{w}\,, \quad (23)$$

$$\tau(u, \vec{w}) = 0 \qquad u = s \qquad \forall\vec{w}\,, \quad (24)$$

$$b(u, \vec{w}) = 0 \qquad u = s \qquad \forall\vec{w}\,, \quad (25)$$

$$i_u(u, \vec{w}) = 0 \qquad u = s \qquad \forall\vec{w}\,, \quad (26)$$

where the expression of the concentration flux density i_u (5), assuming that the gradient of the acoustic pressure with respect to the coordinate u in the direction normal to the wall is negligible, gives

$$\frac{\partial b}{\partial u} + \frac{\gamma\hat{\beta}}{\rho_0 a_0^2}k_T\frac{\partial \tau}{\partial u} \approx 0\,. \quad (27)$$

If a component of the mixture can condensate on the boundary, the boundary conditions (25 to 27) are not valid any more. Indeed, if the component 2 is subjected to condensation and evaporation phenomena on the wall, the pressure variation of component 2 and the normal particle velocity of component 1 are zero (the latent heat in the liquid having to be accounted for) [5].

For the purpose of the present work, the pressure and temperature conditions of the gas are supposed to avoid condensation/evaporation phenomena on the wall, and then the boundary conditions used in the following are eq. (25, 27).

4.1 Temperature variation

Applying the operator $\left(1 + \frac{1}{k_D^2}\Delta\right)$ on equation (20), making use of equation (21), expressing the operator Δ as the sum $\Delta = \frac{\partial^2}{\partial u^2} + \Delta_w$ and assuming that the movement tangent to the wall is almost the acoustic movement, which implies $\Delta_w \tau \approx \frac{\gamma-1}{\hat{\beta}\gamma}\Delta_w p$ leads to the following equation for τ

$$\left(1 + \frac{1}{k_{hD}^2}\frac{\partial^2}{\partial u^2}\right)\left(1 + \frac{1}{k_{Dh}^2}\frac{\partial^2}{\partial u^2}\right)\tau \approx \frac{\gamma - 1}{\gamma\hat{\beta}}\, p_\tau\,, \tag{28a}$$

where

$$p_\tau(\vec{w}) \approx \left[1 - \frac{k_a^2}{k_D^2}(1 - \zeta_T k_P) - \left(\frac{1}{k_{hD}^2} + \frac{1}{k_{Dh}^2}\right)\Delta_w\right] p\,. \tag{28b}$$

and the complex wavenumbers k_{hD} and k_{Dh} satisfie the following relations

$$\frac{1}{k_{hD}^2} + \frac{1}{k_{Dh}^2} = \frac{1}{k_h^2} + \frac{1+\epsilon}{k_D^2}\,, \qquad k_{hD}^2 k_{Dh}^2 = k_h^2 k_D^2\,. \tag{29}$$

The form of this equation illustrates that the temperature variation field τ is a superposition of two diffusion processes, labelled *hD* and *Dh*, where the concentration and thermal fields of the gas mixture are strongly intricate, pointing out the important coupling between the two phenomena.

Assuming that p is quasi-uniform in the u direction inside the boundary layers, the solution for the temperature variation τ, subjected to the boundary conditions (24) and (27), is expressed as the sum of two functions, solutions of the two homogeneous equations included in eq. (28a), as follows:

$$\begin{aligned}\tau(u,\vec{w}) &= \frac{\gamma-1}{\hat{\beta}\gamma}\, p_\tau(\vec{w})\left[1 - \frac{1}{1-\alpha_\tau}\left(\frac{\phi_{hD}(u)}{\phi_{hD}(s)} - \alpha_\tau\frac{\phi_{Dh}(u)}{\phi_{Dh}(s)}\right)\right],\\ &= \frac{\gamma-1}{\hat{\beta}\gamma}\, p_\tau(\vec{w})[1 - \psi_\tau(u)]\,, \end{aligned}\tag{30}$$

α_τ being determined by the boundary condition (27) where the variable b is eliminated making use of equations (20, 21), and is zero in pure gases (i.e. for $x = 0$ or $x = 1$).

4.2 Concentration variation

Applying the operator $\left(1 + \frac{1}{k_h^2}\Delta\right)$ on equation (21), making use of equation (20), and applying a procedure similar to the one used above to express the equation which governs the behaviour of the concentration variation b inside the boundary layers, yields

$$\left(1 + \frac{1}{k_{hD}^2}\frac{\partial^2}{\partial u^2}\right)\left(1 + \frac{1}{k_{Dh}^2}\frac{\partial^2}{\partial u^2}\right) b \approx \frac{\gamma}{\rho_0 a_0^2}\, p_b\,. \tag{31a}$$

where

$$p_b(\vec{w}) \approx x(1-x)\frac{k_a^2}{k_D^2}\,\zeta p\,. \tag{31b}$$

As previously for the temperature variation, the solution for the concentration variation then takes the following form, making use of the functions $\phi_{hD}(u)$ and $\phi_{Dh}(u)$, and accounting for the boundary condition (25) and (27),

$$\begin{aligned} b(u,\vec{w}) &= \frac{\gamma}{\rho_0 a_0^2}\, p_b(\vec{w})\left[1 - \frac{1}{1-\beta_b}\left(\frac{\phi_{hD}(u)}{\phi_{hD}(s)} - \beta_b\frac{\phi_{Dh}(u)}{\phi_{Dh}(s)}\right)\right],\\ &= \frac{\gamma}{\rho_0 a_0^2}\, p_b(\vec{w})[1 - \psi_b(u)]\,, \end{aligned}\tag{32}$$

the constant parameter β_b being determined by the boundary condition (27) where the variable τ is eliminated making use of equations (20, 21).

As expected, in pure gases, the concentration variation b vanishes. Moreover, the factor $k_a^2/k_D^2 \approx \mathrm{j}\,\ell_D\,k_0$ implies that b is a first order term of the characteristic length ℓ_D .

4.3 Tangential particle velocity

The solution for $\vec{v}_w$ of equation (18) at the order one of the characteristic lengths, is written as, accounting for the boundary condition (23),

$$\begin{aligned}\vec{v}_w(u,\vec{w}) &\approx -\frac{1}{\mathrm{j}\,\omega\rho_0}(1 + \mathrm{j}\,k_0\ell_v)\vec{\nabla}_w.p(\vec{w})\left[1 - \frac{\phi_v(u)}{\phi_v(s)}\right]\\ &= -\frac{1}{\mathrm{j}\,\omega\rho_0}(1 + \mathrm{j}\,k_0\ell_v)\vec{\nabla}_w.p(\vec{w})\,[1 - \psi_v(u)]\,, \end{aligned}\tag{33}$$

where $\phi_v(u)$ is solution of the homogeneous equation (18).

The mutual diffusion in the mixture has no effect on the particle velocity at the order one of the characteristic lengths.

4.4 Reflection on a quasi-plane wall

The acoustic pressure p being quasi-uniform over the depth of the boundary layers, and the wall being characterized by its specific admittance y_s (which vanishes when the wall is perfectly rigid and without condensation-evaporation process), the mean values of the terms of equation (19) over a distance from the wall lower than or equal to these depths, making use of the solutions (30, 32, 33), can be expressed easily at the lowest order of the characteristic lengths as given below:

$$\int_s^u \frac{\partial}{\partial u} v_u\,\mathrm{d}u = v_u(u) - v_u(s) = v_u(u) + \frac{y_s}{\rho_0 a_0}\,p\,, \tag{34a}$$

$$\int_s^u (1 - \psi_X)p\,\mathrm{d}u \approx p\int_s^u \psi_X\,\mathrm{d}u\,, \tag{34b}$$

$$\int_s^u (1 - \psi_X)\Delta_w p\,\mathrm{d}u \approx -k_w^2 p\int_s^u \psi_X\,\mathrm{d}u\,, \tag{34c}$$

$(u - s)$ being of the same order of magnitude as the boundary layer thickness, and the variations of the functions ψ_X in the boundary layers being much higher than those of p (subscript X stands for subscripts v, τ or b indifferently).

Considering the reflection on a wall in a semi-infinite domain, $\phi_X(u) = \mathrm{e}^{-\mathrm{j}k_X u}$, leading then to

$$\int_s^u \frac{\phi_X(u)}{\phi_X(s)}\,\mathrm{d}u \approx \frac{1}{\mathrm{j}\,k_X} = \frac{1-\mathrm{j}}{\sqrt{2}}\sqrt{\frac{\ell_X}{k_0}}\,,$$

the exponential function $\mathrm{e}^{-\mathrm{j}k_X u}$ vanishing rapidly when $\delta \approx \delta_X$ (evanescent waves associated to diffusion processes from the wall). Then, integrating equation (19) from s to u, gives the following impedance-like expression

$$\begin{aligned} y_{vthd} = -\rho_0 a_0\frac{v_u}{p} \approx \frac{1+\mathrm{j}}{\sqrt{2}}\sqrt{k_0}\Bigg[&\frac{k_w^2}{k_0^2}\sqrt{\ell'_v}\\ &+(\gamma-1)\sqrt{\ell_h}\,\frac{\sqrt{\ell_{hD}} + \sqrt{\ell_{Dh}}}{\sqrt{\ell_h} + \sqrt{\ell_D}}\Bigg] + y_s\,. \end{aligned}\tag{35}$$

In pure gases, $\ell_{hD} = \ell_h$ and $\ell_{Dh} = \ell_D$, leading to the well-known expression of the viscous and thermal effects on the reflection of an acoustic wave on a quasi-plane wall [7].

5 Determination of gas mixtures thermophysical properties

The methods for speed of sound measurement based on the use of gas-filled cavities are well-known and have been constantly improved for decades. In particular, they are used for primary acoustic thermometry and for the accurate determination of the Boltzmann constant [8, 9, 10], where the speed of sound is derived from the accurate measurement of the acoustic resonance frequencies of a pure gas-filled cavity.

The resonance frequencies and the quality factors of the acoustic field within the cavity depending strongly on the thermophysical properties of the gas, the high accuracy achievable on these experiments makes the method suitable for an accurate and relatively simple determination of these properties from acoustic measurements. For mixtures, this approach looks promising in particular for determining the **thermal conductivity** λ and the **diffusion coefficient** D, parameters of interest for a several metrological and fundamental purposes such as the validation of kinetic theory based on gas mixtures models [11] or determining the collision diameters and intermolecular force-law [2], or for industrial applications (thermoacoustic engines [6]). Indeed, a method to measure these parameters with a high accuracy that would be easy to carry out in various gas conditions of static pressure, temperature and composition is still missing nowadays.

5.1 Acoustic resonance properties of a gas-filled spherical resonator

The acoustic pressure p in a spherical cavity satisfies the propagation equation (17a), where the dissipative effects in the bulk are expressed in the wavenumber k_a (eq. 17b), associated to the boundary condition (out of sound source):

$$\frac{\partial p}{\partial u} + \mathrm{j}\, k_0 y_{vthd}\, p = 0\,, \tag{36a}$$

y_{vthd} being the specific admittance expressing the viscous, thermal and diffusive effects in the boundary layers (35) written as follows, considering an acoustic resonant radial mode,

$$y_{vthd} = \frac{1+\mathrm{j}}{\sqrt{2}}\,\sqrt{k_0}(\gamma-1)\,\sqrt{\ell_h}\,\frac{\sqrt{\ell_{hD}}+\sqrt{\ell_{Dh}}}{\sqrt{\ell_h}+\sqrt{\ell_D}}\,. \tag{36b}$$

The perturbing viscous, thermal and diffusive effects on the mode (m,n) resonance properties [9, 12] are given by

$$\frac{\Delta f_{0n} + \mathrm{j}\, g_{0n}}{f_{0n}} = \frac{\mathrm{j}\, y_{vthd}}{z_{0n}} + \frac{\mathrm{j}\, k_0}{2}\left[\ell_{vh} + \ell_D\, \gamma x(1-x)\zeta^2\right], \tag{37}$$

z_{0n} being the eigenvalue, f_{0n} the resonance frequency without perturbing effects, Δf_{0n} the deviation from f_{0n} due to the viscous, thermal and diffusive effects, and g_{0n} the corresponding halfwidth of the radial mode (0,n).

The resonance frequencies f_{0n} depend strongly on the values of the speed of sound and of the first acoustic virial coefficient, on which there are high uncertainties in gas mixtures, then a measurement of f_{0n} cannot lead to accurate values of λ and D. Nevertheless, the losses in the cavity do not depend on the speed of sound and and first virial coefficient in a significant way. Then the measurement of the halfwidths g_{0n} of the radial modes, the only quantities of expression (37) that can be measured directly with a high accuracy (relative uncertainties less than 1×10^{-6}) can provide useful results for λ and D in gas mixtures.

5.2 Thermal conductivity and diffusion coefficient

A qualitative approach shows that: *i*- the thermal conductivity λ depends on both the composition and the static temperature of the mixture [11, 13], but not on the static pressure; *ii*- the diffusion coefficients D depends on the gas composition, the temperature, but it is inversely proportional to the static pressure [2].

The determination of λ and D can then been done at a single static pressure, chosen in a low pressure range allowing however a sufficiently high signal-to-noise ratio, in order to maximize the thermal and diffusive effects, and then to increase the accuracy of the determination method.

Considering a cavity of 80 mm radius filled with a helium-xenon mixture of helium molar concentration x varying from 0 to 1 for P_0=100 kPa and T_0=273.16 K, the losses (g_{0n}/f_{0n}) due to the viscous, thermal and diffusive effects on three radial modes ($n = 2, 6, 8$), are represented in Figure 1. The isolated contribution of the mutual diffusion effects (($g_{0n} - g_{0n\,\text{no diff}})/f_{0n}$) is showed on Figure 2.

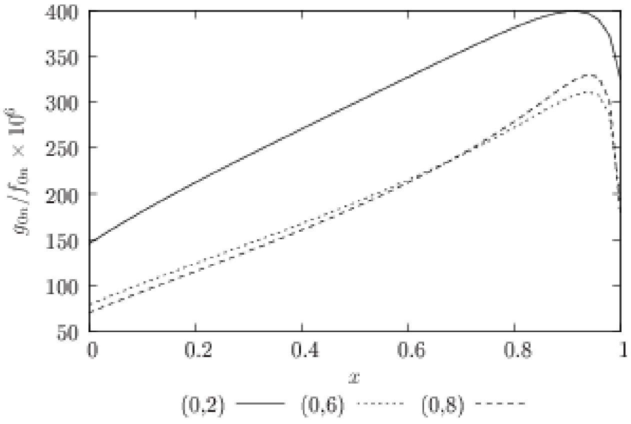

Figure 1: Viscous thermal and diffusive effects on the halfwidths of radial modes of a spherical cavity.

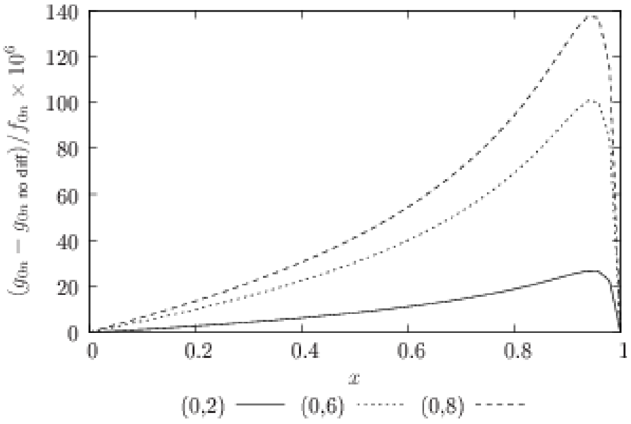

Figure 2: Effect of the mutual diffusion on the halfwidths of radial modes of a spherical cavity.

It should be noticed that using radial modes, the viscous effects are reduced to those in the bulk, which relative contribution to g_{0n} here is less than 1×10^{-6}, and is not a significant uncertainty source for the determination of λ and D.

The effects on the halfwidths of 1% variation in the values of λ and D are showed on Figures 3 and 4 respectively, and give us an estimation of the accuracy of the acoustic method to determine the thermal conduction and the diffusion coefficient of a helium-xenon mixture.

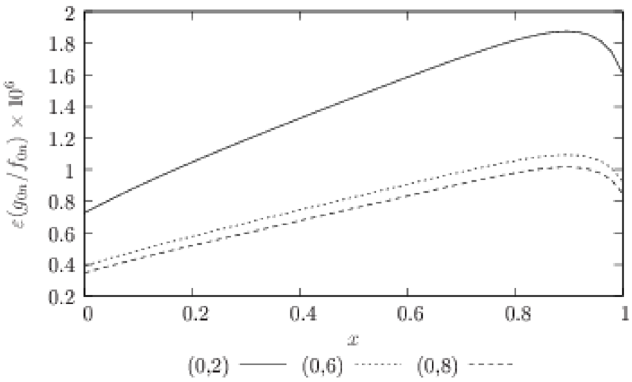

Figure 3: Relative errors on the halfwidths of radial modes of a spherical cavity due to an error of 1% on the value of λ.

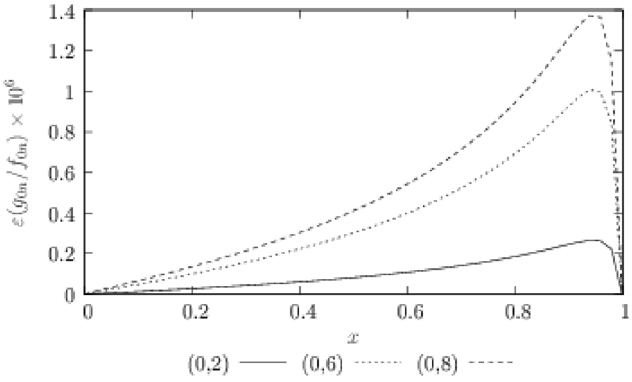

Figure 4: Relative errors on the halfwidths of radial modes of a spherical cavity due to an error of 1% on the value of D.

The thermal effects have a high influence on the halfwidths of low resonance frequencies, whereas the diffusive effects on these low acoustic modes remain negligible. A determination of the thermal conductivity λ of a helium-xenon mixture with a relative uncertainty of 1% would then be achievable by comparing the experimental and theoretical results for the halfwidths of the lowest acoustic radial mode.

Therefore, Figure 4 shows that the diffusion coefficient D could be determined with a relative uncertainty of 1% by comparing the experimental and theoretical results for the halfwidths of the high acoustic radial modes (above (0,8)).

6 Conclusion

The mutual diffusion phenomena have a small effect on the acoustic propagation but a significant effect, in particular, for metrological applications such as the determination of gas mixtures thermophysical properties using acoustical methods in cavity, that seem very promising in term of accuracy and relative simplicity to carry out at several gas conditions (composition, static pressure, temperature).

On the other hand, the analytic modelling described here is suitable for other purposes involving gas mixtures, such as acoustic propagation and gas mixture separation along the stack of thermoacoustic engines [6], or acoustic propagation in porous media filled with humid air, in which occur water condensation and evaporation on the boundaries [5]. For this last case, the modified boundary conditions to be taken into account are those presented ad the end of section 4. The first results of this work still in progress show that the condensation-evaporation of one mixtures component on the wall modify in a significant way the behaviour of the coupled temperature and concentration variations in the boundary layers respect to the model presented here.

References

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