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Numerical investigation on the mechanical/structural parameters on the acoustic properties of a porous ceramic.

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Abstract

The freeze-casting fabrication technique allows for the achievement of a controlled pore structure for different classes of materials, using different types of solvents, through the criogenic cooling of the slurry. Aiming at the investigation of the influence of phase composition on sound propagation properties, ceramic $\text{Al}_2\text{O}_3/\text{MgO}$ monoliths were fabricated with different starting powders compositions, while maintaining the solid/solvent proportions and cooling parameters, so that, after sintering, the samples pore morphologies remain similar although with different phase compositions. Based on preliminarily measured properties and the expected phase compositions and its resulting properties, the Johnson-Champoux-Allard-Lafarge (JCAL) and the Biot-JCAL models were implemented for the different sets of properties of the samples and the resulting sound absorptions are compared. The observed influence of the different properties of the samples is minimal.

Keywords: Porous Ceramic, Freeze-casting, JCAL, Biot model.

PACS: Acoustics, Acoustic modeling, Noise control.



1. INTRODUCTION

Wave propagation in saturated porous medium is subject of great interest of different areas of research, given the diverse applications of porous materials, such as noise absorption and control, and the their natural occurrence in economically important contexts, such as in geological investigation and oil and gas extraction. Understanding the mechanisms of wave propagation of these media is difficult, though, given its inherent geometrical and mechanical complexity and the multiplicity of existing porous materials configurations. Although there has been a great development in the understanding and modeling of these materials so far, sometimes even the application of well established theories is not simple and its results are not intuitive.

Ever since the release of the fundamental work by Biot with theory on the propagation of waves in fluid saturated poroelastic materials [1] [2] [3], numerous models have been proposed for specific classes of materials, obtaining its acoustic properties (e.g. surface impedance, wave number) from material characteristics (porosity, tortuosity, young modulus). These models are derived based on assumptions of the bulk behavior of these materials, mainly rigid body [4], limp body [5] and poroelastic. Thus, it is interesting to select an appropriate model to study different class of materials, given the different behaviors that may or may not be observed based on the selection.

One class of material which has received attention in recent research is porous ceramics. The combination of many of the ceramic materials qualities with the benefits controlled porosity may bring yields great results in different applications [6]. Among these applications, in various recent studies the sound absorption of the samples has been evaluated for porous ceramics fabricated following different routes and using several ceramic materials. These routes and materials result in different pore morphologies and porosities, consequently resulting in different sound absorption capabilities. Du et al. [7] used direct foaming method to obtain highly porous (more than 80 %) silica ceramic foams reaching upwards of 0.8 absorption coefficient

for given raw material compositions, with mechanical strength and sound absorption characteristics being dependent on such composition. Yan et al. [8] produced ZrC ceramics via evaporating solvent and hot-pressing, developing a method that allowed porosity and pore size control, both characteristics that were then correlated to the sound absorption characteristics. He et al. [9] studied porous ceramic previously produced via twice foaming coated with a graphene oxide/styrene-butadiene rubber composite. They obtained an increase in the low frequency (200 Hz-800 Hz) sound absorption in relation to the uncoated samples, an improvement, according to the authors, probably related to the increased tortuosity, viscoelastic damping and inter-layer dissipation brought by composite coating. Du et al, [10] prepared porous silica samples via freeze-casting method, using TBA as solvent, controlling freezing temperature and solid content to obtain different porosities and pore sizes. The acoustic characterization found that the smaller pore size resulted in overall better absorption, the same for higher porosity.

In similar fashion, we are producing Al_2O_3 (alumina)/MgO (Magnesium oxide, magnesia) porous samples (monoliths) using the freeze-casting method with camphene as solvent. The freeze-casting method allows for the controlled creation of open connected porous structures for various different class materials, with different types of solvents, through the cryogenic freezing of the suspension [11] [12]. Solid concentration and speed of the moving freeze front are examples of preponderant process parameters for the connectivity and geometry of the generated pores [13] [14]. Such porous structures have numerous possible applications, one of which being the acoustic absorption [10]. Camphene has been widely used for freeze-casting fabrication given that it can be frozen and sublimed at room temperature [15] [16], avoiding the need for complex freezing process.

In order to better understand the sound absorption, the sample's structures are being characterized with methods such as archimedes principle and gas adsorption (Brunauer-Emmett-Teller, BET). These methods allow for direct analysis and estimation of the porosity and other im-

portant acoustic absorption model parameters such as thermal characteristic length [17]. Based on preliminary material characterization results obtained from the freeze-cast samples with the above mentioned methods, numerical implementations of the Johnson-Champoux-Allard-Lafarge (JCAL) and Biot model were implemented to check the expected sound absorption properties of the samples and to study the effect of the parameters that cannot be directly obtained with experimental setups available to the authors.

2. FREEZE-CASTING

The freeze-casting process consists on the preparation of a slurry containing the solid components of the sample dispersed in a solvent. Such slurry is then frozen by an externally applied temperature gradient. The nucleation of solid crystals generates a freezing front that pushes the solid content, creating a structure consisting of connected solid solvent crystals with agglomerated solid content in between such crystals. Then, the solvent is sublimated and the remaining structure, consisting of a replica of the solidified solvent, is subsequently sintered [11].

For the monoliths production, the used starting powders were CT3000SG α -Al₂O₃ (99.8 wt %/Almatis Brazil) and Magnesium Oxide (≥ 99 % trace metals basis, -325 mesh, Sigma Aldrich). The used dispersant was Texaphor 963 (Cognis, Southampton Hampshire, UK) and the solvent was camphene (95 wt%/Sigma Aldrich). Initially, a solution containing camphene and Texaphor was prepared at 60° C inside a closed vessel to keep the camphene loss due to evaporation to a minimum. After verifying full dissolution of the dispersant in the solvent, alumina was added under vigorous stirring. Once the alumina powder was sufficiently dispersed, maintaining the stirring, magnesia was added. To promote adequate dispersion of both solid components in the solvent, the solution was kept under stirring for an additional 15 minutes after the inclusion of both solid components.

After mixing, the suspension was poured inside a pre-heated PTFE mold on the top of a refrigerated copper plate. The plate is U shaped and was partially immersed on liquid nitrogen to maintain

its temperature. This unidirectional cooling has been shown to propitiate a unidirectional solidification, promoting an alignment of pore channels parallel to freezing front [18]. Following complete solidification, the slurry was demolded and left resting for seven days in open air for the complete camphene sublimation. After the sublimation, the green bodies were then sintered at 1300°C for 2 hours, with a heating rate of 5°C.

3. POROUS MATERIAL SOUND PROPAGATION MODELS

For comparison of the effects on the ceramic samples, the normal incidence sound absorption of the samples against an impervious backing will be obtained. Originally proposed to describe the complex equivalent fluid density of a rigid (motionless), isotropic porous material, the model proposed by Johnson et al. [4] was then complemented with the model by Champoux and Allard [19] that further elaborated the equivalent bulk modulus dependence on frequency for the same materials. Finally, Lafarge et al. [20] suggested changes to bulk modulus equation in order to correct lacking description of low-frequency thermal phenomena in the previous models, leading to the JCAL model. A complete description of the model may be found in [21].

The equations for the equivalent density and bulk modulus, both functions of sound frequency ω , are given as:

$$\rho_{eq}(\omega) = \frac{\rho_f \alpha_\infty}{\phi} \left[1 - \frac{v_f \phi}{\omega k_0 \alpha_\infty} \sqrt{1 + i \frac{\omega}{v_f} \left(\frac{2 \alpha_\infty k_0}{\phi \Lambda} \right)^2} \right] \quad (1)$$

$$K_{eq}(\omega) = \frac{\gamma P_0}{\phi} \left\{ \frac{\gamma(\gamma - 1)}{1 - i \frac{\phi v_f'}{\omega k_0'} \sqrt{\frac{i \omega}{v_f'} \left(\frac{2 k_0'}{\Lambda' \phi} \right)^2 + 1}} \right\}^{-1} \quad (2)$$

In equations 1 and 2, P_0 is the atmospheric pressure, ρ_f is the saturating fluid density, γ is its



specific heat ratio, ν_f its kinematic viscosity and ν'_f is defined as ν_f/Pr , where Pr is the Prandtl number. In addition to the saturating fluid properties, the JCAL model is based on 6 geometrical parameters of the porous medium, specific to the pore morphology. These are the open porosity ϕ , the tortuosity α_∞ , static viscous permeability k_0 , static thermal permeability k'_0 and the viscous and thermal characteristic lengths Λ and Λ' , respectively.

From the equivalent fluid properties, its characteristic impedance Z_c and the its complex wave number k are obtained:

$$Z_c = \sqrt{(K_{eq}\rho_{eq})}, \quad (3)$$

$$k = \sqrt{\omega(\rho_{eq}/K_{eq})}. \quad (4)$$

Finally, from equations 3 and 4 it is possible to determine the normal incidence surface impedance of a sample with thickness d :

$$Z = -i\frac{Z_c}{\phi} \cotg(kd). \quad (5)$$

The Biot model and its different formulations considers a two phase system of wave propagation, i.e. the wave propagates in the solid and fluid phases of a saturated porous material. Two types of waves propagate in poroelastic materials according to the model: two compressional (called fast and slow waves [21]) and one shear wave. In the solid medium, the compressional and the shear waves can propagate, while only the compressional waves propagate in the fluid medium.

For the normal incidence case, there is no shear wave propagating, given there is no oblique excitation of the frame. In this case, the sample surface impedance may be obtained from the characteristic impedance for the compressional waves in both media, given in Equations 6 and 7, where $i = 1, 2$ and indicates the slow and fast wave related impedances and f and s indicate the medium (fluid and solid):

$$Z_i^f = (R + Q/\mu_i) \frac{\delta_i}{\phi \omega}; \quad (6)$$

$$Z^s = (P + Q\mu_i) \frac{\delta_i}{\omega}, \quad (7)$$

where P, Q and R are elasticity coefficients necessary to the stress-strain relations proposed by Biot [1] [21], μ_i is the ratio of the fluid's velocity over the frame's velocity and δ_i are the wave numbers. Given the specific impedances of each propagation medium, the surface impedance of a sample with thickness d may be calculated by Equation 8 [21]:

$$Z = -i \frac{(Z_1^s Z_2^f \mu_2 - Z_2^s Z_1^f \mu_1)}{D}, \quad (8)$$

where D is shown in Equation 9:

$$D = (1 - \phi + \phi \mu_2 [Z_1^s - (1 - \phi) Z_1^f \mu_1] \text{tg}(\delta_2 d) + (1 - \phi + \phi_1) [Z_2^f \mu_2 (1 - \phi) - Z_2^s] \text{tg}(\delta_1 d). \quad (9)$$

In possession of the surface impedances given by both models, it is now possible to determine the acoustic absorption of the samples. The reflection coefficient r of the sample is determined and the sample acoustic absorption α can be acquired:

$$r(\omega) = \frac{Z(\omega) - Z_f}{Z(\omega) + Z_f}, \quad (10)$$

$$\alpha = 1 - |r|^2. \quad (11)$$

With the models set, it is possible to implement their solutions computationally. The numerical implementation of the JCAL model was done with the direct coding of the model equations. A routine based on the one made available in [22], which is itself based in [23], was implemented for the Biot model.

4. RESULTS AND DISCUSSION

Archimedes principle was used to obtain the total, open and closed porosities of the sample. Shortly, the archimedes principle method consists on the measurement of the dry, water imersed and water saturated sample masses. With these three values, the densities of water and of the solid content, it is possible to obtain the open, total and closed pore volume as well as the saturating water volume and the sample approximate density.

Three samples were produced for testing of the sound absorption, each with a different concentration of the magnesia and alumina starting powders. This choice was done in order to evaluate the possible influence of phase composition on the sample's acoustic properties and given that the alumina/magnesia phase diagram is well known [24]. The sintering temperature choice was also done with the purpose of promoting the growth of the spinel phase (MgAl_2O_4 , magnesium aluminate) and have a final phase composition proportional to the starting powders, thus mechanical properties equivalent to that of the predominant phase (Al_2O_3 , MgO or MgAl_2O_4).

Additionally, for the description of the sample's elasticity coefficients, two main considerations are made:

- the N coefficient (used to calculate the P parameter) will be considered to be equal to the shear modulus of the ceramic porous material, with a complex loss factor of $\eta = 0,01$;
- the Young (E) and shear modulus (G) are calculated based on the predominant phase mechanical properties and the equations proposed for open cell foams in [25], presented in Equations 12 and 13:

$$E^* \approx E_s \left(\frac{\rho^*}{\rho_s} \right)^2, \quad (12)$$

$$G^* \approx E_s \frac{3}{8} \left(\frac{\rho^*}{\rho_s} \right)^2, \quad (13)$$

where ρ^* is the foam density, E_s and ρ_s are the solid material Young modulus and density, respectively and E^* and G^* indicate the equivalent foam elastic and shear modulus, respectively. Besides these shown properties, [25] points out that the Poisson Ratio of porous materials is difficult to obtain and the average value of the obtained ratios studied foams is $\nu \approx 1/3$.

The found porosities and densities of the samples are shown in Table 1, jointly with the found approximate mechanical properties.

In addition to the properties shown in Table 1, the JCAL parameters shown in Table 2 were used to obtain the sound absorption of the modeled samples using Biot-JCAL model and JCAL (rigid body) model. The sound absorption of the samples obtained using the Biot-JCAL model are shown in Figure 1.

To evaluate the influence of the elastic skeleton condition for the porous ceramic model, the Biot-JCAL and rigid body JCAL obtained sound absorptions are compared in Figure 2.

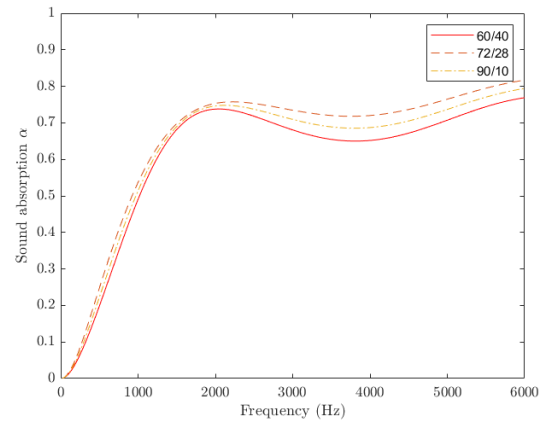


Figure 1: Sound absorption of samples using Biot-JCAL.

It can be seen from Figure 2, even though there is a difference in the calculation route for the impedance and thus sound absorption of all samples when using both models, the obtained results are very similar (virtually the same). Although the Biot-JCAL considers an elastic frame, the elasticity (Young) modulus is very high, given the mechanical properties of the base ceramic powders used for the manufacturing of the samples. Both alumina and Magnesia and the resulting spinel phase resulting from sinter-

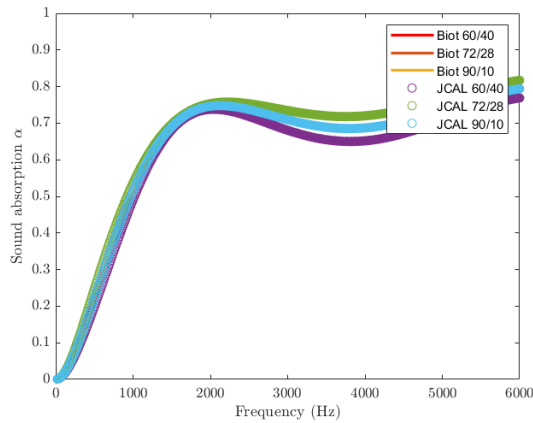


Figure 2: Sound absorption of samples using Biot-JCAL and JCAL.

ing of these materials have very high elasticity modulus, and even with the consideration of a “lowered” modulus due to porous structure and frame energy dissipation (dissipation not related solely to the propagation of waves within the pores), these effects are not sufficient to make a large difference in the sound absorption of the materials. It is possible, then, to say that, for this class of materials, the rigid body model is an adequate research framework for the evaluation of its acoustic properties.

5. CONCLUSION

A comparison between the use of the Biot-JCAL and JCAL model for modelling of the acoustic properties of porous ceramics built via freeze-casting method was performed. This comparison

had the objective of verifying one of the possible influences of the ceramic chemical composition on the acoustic properties of the samples, i.e. if the elasticity of the ceramic material frame was “sufficient” to exert a sensible effect on the sound absorption, and if so, if so, to what extent did the different elasticity of a sample affect these properties. The comparison of the rigid and elastic frame models for the same parameters, though, shows that the effects for rigidities in the range of the analysed ceramics is overly high for the presence of elasticity effects to be noticeable.

For future research, the influence of ceramic base powders and final phase composition will be evaluated within the framework of rigid frame models, then analyzing the effects of the obtained geometric parameters, such as tortuosity and characteristic lengths.

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Table 1: Obtained porosities and assumed mechanical properties

Samples / Solid conc.	ϕ	E_s GPa	ρ_s Kg/m ³	ρ^* Kg/m ³	E^* GPa	G^* GPa
60 wt% Al ₂ O ₃ /40 wt% MgO	64,9 %	320 [26]	3580	2824,4	171,1	64,2
72 wt% Al ₂ O ₃ /28 wt%	76,9 %	273 [27]	3580 [27]	2158,9	99,3	37,2
90 wt% Al ₂ O ₃ /10 wt% MgO	70,7 %	275 [28]	3600 [28]	2734,1	151,8	56,9

Table 2: Obtained porosities and assumed mechanical properties

h (mm)	σ (Nsm ⁻⁴)	α_∞	λ (μm)	λ' (μm)	k_0 (m ²)
20	120000	2	30	80	23e-10

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