Interpretation of the thickness resonances in ferroelectret films based on a layered sandwich mesostructure and a cellular microstructure.

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Abstract—Transmission coefficient spectra of two ferroelectret films (showing several thickness resonances) measured with air-coupled ultrasound (0.2–3.5 MHz) are presented and an explanation for the observed behavior is provided by proposing a film layered sandwich mesostructure (skin/core/skin) and by solving the inverse problem, using a simulated annealing algorithm. This permits to extract the value of the ultrasonic parameters of the different layers in the film as well as overall film parameters. It is shown that skin layers are thinner, denser and softer than core layers and also present lower acoustic impedance. Similarly, it is also obtained that the denser film also presents lower overall acoustic impedance. Scanning Electron Microscopy was employed to analyze the films cross-section, revealing that both denser films and film layers present more flattened cells and that close to the surface cells tends to be more flattened (supporting the proposed sandwich model). The fact that more flattened cells contributes to a lower elastic modulus and acoustic impedance can be explained, as it has been made previously by several authors, by the fact that the macroscopic film elastic response is furnished by cell micromechanics which is governed, mainly, by cell wall bending. Consistency of extracted parameters with trends shown by a simple model based on a honeycomb microstructure is discussed as well as the possibilities that this sandwich mesostructure and the associated impedance gradient could offer to improve the performance of FE films in ultrasonic transducers.

Index Terms— Ultrasonic transducers, ferroelectret film, cellular solids, thickness resonances, sandwich structure, inverse problem, mechanical properties.

I. INTRODUCTION

FERROELECTRETS (FE) are permanently space charged cellular polymers that present piezoelectric response [1], [2]. Key to this behavior is the large porosity in the form of large and flattened pores in the film plane. These pores make possible both to entrap the electrical charge that build up macroscopic electric dipoles and to achieve an extremely low elastic modulus in the thickness direction that ease the film deformability in that direction. Both features give rise to the

well-known piezoelectric response of these films. Hence, one of the key issues in the optimization of the film electromechanical conversion capability is to strike the correct balance between cell and cell-wall shape and dimensions [3]–[10]. The other key feature is the efficient polarization of these films [11]–[13]. Another interesting feature is that they present a viscoelastic / poroelastic response [14], [15].

FE films are normally manufactured using a continuous biaxial orientation process (by stretching the film in two perpendicular directions), adding small particulate (to create lens-like voids) and subjecting it to a swelling process using a high-pressure gas injection technology [3], [5], [7], [16]. The swelling process alters the pore shape in a way that is critical to improve the electromechanical conversion capability [3]–[10]. Polarization is achieved by barrier discharge in the pores [17]. Main materials to produce FE are polypropylene (PP), polyethylene terephthalate (PETP) [10], FEP, cellular cyclo olefine (COC), polyethylene-naphthalate (PEN) [18]; main reason to modify the solid material has been to improve charge stability. FE films have been used for pressure sensors, tactile sensors [19], air-coupled transducers [20]–[25], energy harvesting [26]–[28], etc.

Recently it has been shown the possibility to use FE films to produce water immersion transducers for the frequency range 0.1–3.0 MHz [29], [30]. The main advantages are that FE films: i) present a very short response and a very large frequency bandwidth, ii) are flexible and conformable (which could permit to fabricate complex shapes [21], [22]), iii) present a reduced thickness and require thin backings. The main drawback is the reduced sensitivity which is the result of the lack of resonant response and the poor impedance matching of FE films to water. Improving transducer response demands improving sensitivity while keeping the large frequency bandwidth. This requires a better knowledge of the film properties and response in the frequency range of interest (0.1–3.0 MHz), which is much larger compared with conventional air-coupled FE transducers that operate around the first thickness resonance of the FE film (normally: 0.2 – 0.7 MHz).

Reference [31] studied thickness resonances of some FE films and found some anomalies that were explained by assuming that: i) velocity decreases with the frequency and ii) this decrease follows a stepwise shape between the first and the second resonances. The first assumption is quite

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reasonable provided that a quite large attenuation coefficient value and a strong increase with the frequency were observed [31]. Quite on the contrary, the second one is quite an anomalous feature that was not further explained. Nonetheless, obtained fitting of theoretically calculated values into the experimental data was quite good. More recently, [32] has shown that similar distortions are found in the thickness resonance spectra of plant leaves and that this distortion can be explained by the layered structure of the leaves. However, solving this inverse problem (IP) can be extremely challenging. Reference [32] presented a method to solve it for the case of bilayer structures, based on the use of a Simulated Annealing algorithm and on the measurement of the transmission coefficient over a frequency range that fulfills two conditions: i) more than one thickness resonances are observed and ii) thickness resonances are completely damped out in the high frequency limit.

The objective of this paper is to obtain this kind of measurements in FE films, apply a modification of the procedure shown in [32] to solve this IP, analyze the capability of a sandwich model to reproduce the measured response, evaluate the meaning of the obtained parameters for the different layers in the FE film, its consistency with data that can be obtained by alternative methods and discuss the potential practical benefit that can be obtained from a ferroelectret film with a sandwich-like layered structure. Towards this end, the actual film structure is studied by analyzing Scanning Electron Microscopy (SEM) images and a symmetric layered mesostructured (three layers in a sandwich configuration) is proposed. In addition, and as it has been made previously by different authors, a cellular microstructure is assumed to provide an explanation for the extracted film parameters and the variations observed in them when comparing different films and different layers.

II. MATERIALS

Two different types of polypropylene FE films obtained from EMFI have been studied, commercial names: HS03 and HS06. Nominal thicknesses are 70 and 90 μm , for HS03 and HS06, respectively, while densities are 530 and 320 kg/m^3 for HS03 and HS06, respectively.

III. METHODS

A. Scanning Electron Microscopy (SEM) images.

Strips of 20 x 5 mm were cut out of the available sheets, put in liquid nitrogen and broke into two pieces. A thin film of gold was sputtered over the fracture and then examined using a Scanning Electron Microscope (SEM). Image analysis, performed using ImageJ, consisted on determining film thickness and mean value and standard deviation of different cell features. Ten pictures and between 100 and 120 measurements for each parameter were obtained for each film.

B. Ultrasonic Measurements

Pieces of 4 x 4 cm were cut for the measurements: three pieces for each type of film. The ultrasonic set-up is shown in

Fig. 1. The ultrasonic transmission coefficient spectra, both magnitude and phase, at normal incidence were measured using air-coupled ultrasound in the frequency range 0.2-3.2 MHz. Four pairs of high-sensitivity, wideband, air-coupled transducers with center frequency 0.25, 0.65, 1.25 and 2.5 MHz designed and fabricated by our group were used. Transducers are flat, non-focused, and have circular aperture, (diameter of 25, 20, 15 and 15 mm, respectively) [33], [34]. Transmitter transducer was driven by a semicycle of square wave generated by a Olympus 4077 pulser-receiver (PR), with amplitude 400 V and tuned to the transducers centre frequency. Receiver transducer is placed in opposition at a distance between 100 and 10 mm. Receiver transducer is connected to the reception stage of the 4077 PR and the received signal is then transferred to a digital scope (Tektronix 5054), where the signal is digitized and stored. Finally, it is sent to a PC where Fast Fourier Transform (FFT) is extracted. First, signal received without sample in between the transducers and with 0 dB gain in the receiver stage of the 4077PR is stored. This signal is used to generate the reference magnitude and phase spectra. Then, sample is put in between transducers at normal incidence. Gain in the receiver stage of the 4077PR is set to 20 dB. This procedure is repeated with each pair of transducers. Overlap of the spectra is first checked to validate the measurements. Then, all spectra were concatenated and resampled ($\Delta f = 14$ kHz).

Fig. 1. Experimental set-up for ultrasonic measurements: pulser-receiver, oscilloscope and three pairs of transducers (0.25, 0.65 and 1.25 MHz, from left to right, the 2.5 MHz pair is identical to the 1.25 MHz pair and is not shown). Transducers are mounted on an U-shaped holder that keeps them in the right position, a PVC cover protects transducers and measuring volume and provide a slot for the introduction of the sample in the right place and position.

C. Data Extraction from Ultrasonic Transmission Coefficient Spectra.

A 1D layered model for the FE films is assumed with a sandwich structure (1: skin / 2: core / 3: skin), where skin layers are considered identical. The transmission coefficient spectra (magnitude and phase) for this sandwich structure were calculated using the transfer matrix method [35]-[42]. Material parameters are extracted by the solution of the IP as the set of layer parameters that minimize the error (Δ) between

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measured and calculated spectra:

$$\Delta = (\Delta M^2 + \Delta \varphi^2)^{1/2} \quad (1)$$

where ΔM and $\Delta \varphi$ are:

$$\Delta M = \left(\frac{\sum N((M_{exp} - M_{cal})/M_{exp})^2}{N} \right)^{1/2} \times 100 \quad (2.a)$$

$$\Delta \varphi = \left(\frac{\sum N((\varphi_{exp} - \varphi_{cal})/\varphi_{exp})^2}{N} \right)^{1/2} \times 100 \quad (2.b)$$

where M_{exp} and φ_{exp} are measured magnitude and phase spectra and M_{cal} and φ_{cal} are calculated magnitude and phase spectra. As explained in [32], parameters extracted for each layer are:

- $f_{res} = v/t$, where v is the ultrasound velocity and t the layer thickness. For isolated layers under symmetric boundary conditions, thickness resonances appear at $(f_{res}/2) \times m$, where $m = 1, 2, 3, \dots$ For isolated layers under asymmetric boundary conditions, thickness resonances appear at: $(f_{res}/4) \times (2m - 1)$, where $m = 1, 2, 3, \dots$

- Acoustic impedance: Z

- Attenuation coefficient $\times$ thickness: $\alpha \times t$

- Variation of attenuation with frequency: n

Towards this end a power law is assumed [42]:

$$\alpha = \alpha_0 \left(\frac{f}{f_0} \right)^n \quad (3)$$

That is, four parameters are obtained for each layer. As we have three layers, but two of them are identical, the dimension of the space of search is eight. In addition, from these parameters the surface density (ρ_{surf}) can also be obtained:

$$\rho_{surf} = Z / f_{res} \quad (4)$$

Moreover, the high frequency limit of the phase spectra can be used to determine the time of flight and the total thickness t^{Total} [32]. Hence, we can also work out effective velocity (v^{eff}) and the volumetric density (ρ) of the whole film:

$$v^{eff} = t^{Total} / \sum_{m=1}^3 (1/f_{res, m}) \quad (5)$$

$$\rho = \sum_{m=1}^3 \rho_{surf, m} / t^{Total} \quad (6)$$

where m denotes the layers in the sandwich. As t^{Total} and ρ can be determined from independent methods, estimations of these values (with $\pm 10\%$ tolerance) are used to impose constraints to the search algorithm, through an adaptative penalization to the fitting quality or error (Δ), borrowed from evolutionary algorithms [44]. The search algorithm employed is the same as in [32]: modified constrained Simulated Annealing (SA) algorithm [44], [45]. The algorithm is run twenty times for each measured spectra with the purpose of determining: i) the existence of multiple solutions, ii) the variability/accuracy of the extracted parameters [32].

IV. RESULTS

A. SEM analysis.

Fig. 2 shows Scanning Electron Microscopy (SEM) pictures of the films cross-section. SEM images also revealed that film surfaces are non-porous. Results of the image analysis are shown in table I.

TABLE I.
PARAMETERS OF THE FILMS OBTAINED FROM SEM IMAGES. MEAN VALUE AND STANDARD DEVIATION.

Quantity	HS03	HS06
Film thickness (μm)	70.4 ± 1.8	77.0 ± 2.6
Outer layer thickness (μm)	3.51 ± 0.8	2.22 ± 1.2
Cell wall thickness: t (μm)	2.01 ± 0.8	1.96 ± 0.9
Cell aperture: w (μm)	3.62 ± 2.1	5.74 ± 2.4
Cell length: D (μm)	30.1 ± 16.0	36.2 ± 20.1
Vertex angle: φ ($^\circ$)	14.2 ± 6.0	33.0 ± 12.0

Fig. 2. Cross-section SEM images of the PP FE films studied. Left: HS03, right: HS06, showing cellular microstructure of the films composed of PP layers with lens-shaped cells. More flattened cells are observed in HS03, and cells tend to be more flattened in the section close to the film surface.

Fig. 3. Detail of the cross-section area close to the film surface (15 μm depth). White arrows indicate the film surface. Left: HS03; Right: HS06. Figures show the more flattened cell shape in the section close to the film surface.

Cell wall thickness is similar for both films and is slightly smaller than the thickness of the outer film layer. Cell length distribution is quite wide with comparable mean values in HS03 and HS06. In spite of the large variability, cell aperture and vertex angle are clearly larger in HS06. Fig. 2 also reveals that cells close to the surface tend to be more flattened. Fig. 3 shows some details of the section of the film close to the surface. Most likely, these features are specific of these films and the way they were fabricated and cannot be considered features to be found in all kinds of FE films. However, the fact that they are present here permits us to study if this subtle variation does have any measurable impact on the

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macroscopic response. If so, it could be possible to consider the optimization of the macroscopic response of these films by the proper modification of their layered mesostructure.

B. Ultrasonic Measurements

Figs. 4 and 5 show magnitude and phase spectra in HS03 and HS06 films, respectively. Dots represent measurements and solid line is the calculated spectra using the layers parameters extracted from the solution of the IP and assuming a symmetrical sandwich structure. As required by the method, two orders of the thickness resonances are observed, and the thickness resonances are completely damped out in the high frequency range (>1.8 MHz and >2.8 MHz for HS03 and HS06, respectively).

Fig. 4. Magnitude (up) and phase (down) spectra of the ultrasonic transmission coefficient at normal incidence for HS03 film in air. Dots: measurements. Solid line: calculated values using the sandwich model and the layer parameters obtained from the solution of the IP.

Fig. 5. Magnitude (up) and phase (down) spectra of the ultrasonic transmission coefficient at normal incidence for HS06 film in air. Dots: measurements. Solid line: calculated values using the sandwich model and the layer parameters obtained from the solution of the IP.

Table II lists some parameters that are obtained directly from the measured spectra. Multiple runs of the SA algorithm to solve the IP produced different results for HS03 and HS06. For HS03 two distinct solutions were found. However, error (1) of one of these solutions is clearly smaller ($\Delta \sim 0.17$ and 0.6). Therefore, the solution with the best error was taken and the other disregarded as a local minimum. On the contrary, no

multiple solutions were found for HS06, and all extracted values gather around one single point in the space of search. In this case: $\Delta \sim 0.4$. Tables III and IV show the layer parameters extracted for one of the samples of the HS03 and HS06 films, respectively: mean values and standard deviation corresponding to run the SA algorithm 20 times for each measurement. Table V summarizes the overall film properties.

TABLE II.

EXPERIMENTAL VALUES OF RESONANCE (f_{res}^n) AND ANTI-RESONANCE (f_{a-res}^n) FREQUENCIES AND THE VALUE OF THE MAGNITUDE SPECTRUM FOR HS03 AND HS06 FILMS, OBTAINED FROM FIGS. 2 AND 3.

Order of resonance (n)	1	2	3
HS03			
f_{res}^n (MHz)	0.638	1.190	--
Amplitude at f_{res}^n (dB)	-20.1	-30.3	--
f_{a-res}^n (MHz)	0.323	0.944	--
Amplitude at f_{a-res}^n (dB)	-35.0	-33.7	--
HS06			
f_{res}^n (MHz)	1.120	2.096	~ 2.87
Amplitude at f_{res}^n (dB)	-20.9	-30.1	-36
f_{a-res}^n (MHz)	0.550	1.610	2.48
Amplitude at f_{a-res}^n (dB)	-38.5	-35.5	-35.8

TABLE III.

MEAN VALUE AND STANDARD DEVIATION FOR THE LAYER PARAMETERS OF HS03. SA ALGORITHM IS RUN TEN TIMES

f_{res} (MHz)	Z (MRayl)	α_{xt} (Np)	n	ρ_{surf} (kg/m ²)
Skin layer				
3.77 ± 0.14	0.044 ± 0.0003	(2.07 $\pm$ 0.03) $\times 10^{-2}$	2.0 ± 0.01	(1.16 $\pm$ 0.04) $\times 10^{-2}$
Core layer				
3.77 ± 0.30	0.050 ± 0.0004	(2.33 $\pm$ 0.18) $\times 10^{-2}$	2.1 ± 0.04	(1.35 $\pm$ 0.1) $\times 10^{-2}$

TABLE IV.

MEAN VALUE AND STANDARD DEVIATION FOR THE LAYER PARAMETERS OF HS06 PROBLEM. SA ALGORITHM IS RUN TEN TIMES.

f_{res} (MHz)	Z (MRayl)	α_{xt} (Np)	n	ρ_{surf} (kg/m ²)
Skin layer				
10.8 ± 0.86	0.049 ± 0.004	(1.99 $\pm$ 1.8) $\times 10^{-3}$	2.0 ± 0.01	(4.63 $\pm$ 0.7) $\times 10^{-3}$
Core layer				
3.25 ± 0.21	0.075 ± 0.0014	(5.29 $\pm$ 1.9) $\times 10^{-3}$	2.8 ± 0.2	(2.31 $\pm$ 0.11) $\times 10^{-2}$

TABLE V.

OVERALL FILM PROPERTIES EXTRACTED FROM THE MEASURED SPECTRA AND (5) AND (6). MEAN VALUE AND STANDARD DEVIATION

Quantity	Film	
	HS03	HS06
Total thickness (μ m)	69.3 $\pm$ 0.7	87.4 $\pm$ 0.8
Effective density (kg/m ³)	530.0 $\pm$ 4.0	370.0 $\pm$ 7.8
Effective ultrasound velocity (m/s)	86.8 $\pm$ 0.7	177.0 $\pm$ 1
Effective elastic modulus (MPa)	3.98 $\pm$ 0.05	11.6 $\pm$ 0.25

Effective impedance (MRayl)	0.046±0.0005	0.065 ± 0.0013
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V. DISCUSSION

SEM images in Fig. 2 show the inner cellular microstructure of the studied FE films and provide a clear representation of how the deformation in the film thickness direction is expected to take place: through the bending of the cell walls, mainly at the cell vertexes and at the curved section of the cell wall around the vertexes. In addition, the fact that cell aperture and vertex angle is not homogeneous along the film thickness can also be seen in Fig. 2. It is more likely to find less opened cells with very small vertex angle close to the film surface. This feature is surely the result of the fabrication route followed in these particular cases and cannot be considered a general property of all FE films; however it is interesting to study how this subtle modification may affect the macroscopy film response. The zoomed images in the section adjacent to the film surface (see Fig. 3) reveal cells with very reduced apertures and vertex angles (HS03: down to 0.5 μm and 5°; HS06: down to 1 μm and 9°, respectively), that rather than cells look more like slits or cracks. How deep does this feature penetrate into the film thickness is not easy to determine and can be expected to be variable, but it can be estimated to expand over a length of about 5-15 μm . Given the fabrication procedure, this kind of anisotropy is not fully unexpected as this can be the result of the presence of a rather stiff boundary condition (non-porous surface layer) that restricts the deformability of adjacent cells during the process of stretching and inflating.

Thickness resonances (Figs. 4 and 5) appear in the transmission coefficient as a local maximum in the magnitude spectrum and as a step in the phase spectrum. The higher the frequency, the more reduced these effects are. This is due to the increase in the ultrasonic attenuation with the frequency. Phase spectra present negative values that correspond to the fact that ultrasound velocity in the film is lower than the ultrasound velocity in the air (~ 345 m/s). For high frequencies, phase spectra can be approximated to a straight line. This indicates that variation of velocity with the frequency is negligible, at least in this frequency range (>1.5 MHz for HS03 and >2.5 MHz for HS06).

Frequency location of the observed first two orders of the thickness resonances (Table II) does not follow the pure geometrical condition observed in homogeneous layers under symmetrical boundary conditions:

$$f_{res}^n = \frac{v}{2t} n, \quad n = 1, 2, 3, \dots \quad (7)$$

This deviation away from the expected response of a homogeneous film is small but clearly measurable. This was accounted for in a previous work [31], by assuming a variation in the velocity with the frequency. However, the high frequency phase spectrum measurement reveals that velocity does not change with the frequency. In addition, as $f_{a-res}^1 = \frac{1}{2} f_{res}^1$, holds for both kinds of films (see Table II), velocity

must be constant, also between f_{a-res}^1 and f_{res}^1 .

The sandwich model here proposed provides an excellent fitting between calculated and measured spectra, even better than the one obtained before [31]. Two conclusions can be drawn: i) the sandwich model is able to reproduce very well the observed behavior and ii) the SA algorithm is able to solve this IP. Moreover, the multiple runs of the SA algorithm reveal that: i) no multiple solutions were found for HS06 and, although two solutions were found for HS03, it is possible to disregard one of them because it presents a significantly larger error; ii) the dispersion of the extracted material parameters is very low for all parameters (standard deviation below 10%). The main exception is $\alpha \times t$ values for HS06, revealing a very low accuracy in this case; standard deviation of ρ_{surf} in the skin layer of HS06 is also slightly over the 10%.

Overall FE film thickness and density listed in table V are in good agreement with data obtained from conventional means, other authors and manufacturer data. Thickness data obtained from the ultrasonic technique also agrees with estimations obtained from SEM images (Table I and V), though HS06 thickness obtained from SEM images is smaller compared with both ultrasonic and micrometer measurements. In addition, effective ultrasonic velocity and elastic modulus also agree well with data published before. The very low value of the elastic modulus is the result of the porous structure and the fact that deformation of the film is expected to take place through bending of the pore walls. The lower elastic modulus of HS03 is then the result of the more flattened pore shape in HS03, that give rise to cells more deformable in the film thickness direction (Fig. 2). This can be explained by using different models that relate cell geometry and micromechanics with effective macroscopic elastic modulus. For example, the tetrakaidecahedron unit cell has been widely used for these purposes [46]. Later, [47], an elongated tetrakaidecahedron unit cell, more similar to the FE structure observed in Fig. 2, was proposed to study cellular solids. One of the simplest models is based on a hexagonal honeycomb structure [48]. This one and other similar structures have also been used to model FE films [8], [9], [49] and to study the relationship between fabrication route, film microstructure and macroscopic properties.

Fig. 6 shows the basic cell geometry for the elongated tetrakaidecahedron and for the hexagonal unit cells and an illustration of the cell deformation under uniaxial load. In spite of its radical simplicity, the hexagonal honeycomb model can be used to gain a basic understanding of the influence of the microstructure of the cellular solid in the macroscopic elastic response of the FE films. Equivalence between magnitudes in Table I and Fig. 6 are given by: $\varphi = 180 - 2 \times \theta$, $w = 2 \times l \times \cos \theta$, and $D = 2 \times l \times \sin \theta + h$.

Fig. 6. Illustration of the unit cell and the deformation under uniaxial load of: a) hexagonal honeycomb cellular solid [39], b) tetraikadecahedron-cellular solid [38].

Elastic modulus and density of the honeycomb are obtained from [48]:

$$\frac{E}{E_{solid}} = \left(\frac{t^w}{l}\right)^3 \frac{\cos\theta}{(h/l + \sin\theta)\sin^2\theta} \quad (8)$$

$$\frac{\rho}{\rho_{solid}} = \left(\frac{t^w}{l}\right) \frac{h/l + 2}{2\cos\theta(h/l + \sin\theta)} \quad (9)$$

where E_{solid} is the Young modulus and ρ_{solid} the density of the cell wall material. It is clear that both the hexagon angle, θ , and the ratio t^w/l play a significant role in the elastic modulus of the honeycomb. Hence, even this extremely simplified model can explain that denser films may also present a lower elastic modulus, due to the larger value of θ . In general, this feature could appear counter-intuitive if the cellular microstructure is not taken into account.

As it is well known, this cellular microstructure of the FE films can also explain the differences observed in overall acoustic impedance of HS03 and HS06 films (table V). The larger density in HS03 (Table V) can be explained by this more flattened cell structure (Fig. 2), as this reduces the porosity. The lower impedance measured in HS03 is also explained in this way as the more flattened cell shape also significantly reduces the elastic modulus (Table V and (8)). The decrease in the elastic constant in the thickness direction with the increase of film density has already been reported in previous works (e.g. [5]-[9]) and explained in terms of the cellular microstructure. A similar effect was observed in [14].

As an example, Fig. 7 shows some calculations for E and ρ according to (8) and (9). Both the smaller density and the larger elastic modulus observed in HS06 can be explained by the fact that cells in HS06 are less flattened. This complex feature can be accounted for by the model in an extremely simple way: by assuming a smaller value of θ .

Fig. 7. Variation in the elastic modulus (left) and the thickness (right) of a polypropylene hexagonal honeycomb ($L = 5.5\mu\text{m}$, $h = 30\mu\text{m}$) with t and ϕ . Assuming for the propylene: $E = 1.33\text{ GPa}$ and $\rho = 900\text{ kg/m}^3$. The black lines represent obtained values for HS03 and HS06, line thickness correspond to the standard deviation (Table V).

Tables II and III reveal that a common feature in both types of films is that the acoustic impedance of the core layer is slightly larger. In parallel to what we considered before, we can now assume that this lower acoustic impedance in the skin layers is produced by a more flattened pore shape, which agrees with features revealed by SEM images (Figs. 2 and 3).

Unfortunately, and unlike the case of a one layer film [50], the ultrasonic method does not provide the thicknesses of each layer. Nonetheless, imposing some a priori conditions we can obtain an estimation of the possible range of variation. As a matter of fact we know that:

$$\rho_{PP} > \rho_{skin} > \rho_{core} \quad (10)$$

where ρ_{PP} is the density of the polypropylene. As $\rho = \rho_{SUP}/t$, then using the value of ρ_{SUP} from Tables II and III, and the limits in (10), we can obtain that the thickness of the skin layer must be within the range 11-21 μm , and 5-12 μm , for HS03 and HS06, respectively. These values are consistent with features revealed by SEM images. If we assume a mean value of 15 μm for the skin layer in HS03, then density and elastic modulus are: 730 kg/m^3 and 9.9 MPa, for the skin layers, and 344 kg/m^3 and 30.0 MPa, for the core layer, respectively. For HS06, if we assume a mean value of 8 μm for the skin layer, then density and elastic modulus of skin and core layers are: 618 kg/m^3 ; 4.3 MPa, and 317 kg/m^3 ; 18.0 MPa, respectively.

Finally, it is worth noting that none of the observed resonances correspond to resonances of the individual layers (Tables II, III and IV). Quite on the contrary, the explanation of the observed resonances is that they are produced by the whole cavity. We can make an approximate calculation of the frequency location of the first resonance of the whole cavity (f_{res}^{WC}). This is given by:

$$f_{res}^{WC} = 1/(2\sum_{n=1}^3 1/f_{res, n}) \quad (11)$$

where n denotes the layers in the sandwich: 1 and 3 for the skins and 2 for the core. Obtained f_{res}^{WC} from (11) is 630 kHz and 1.02 MHz for HS03 and HS06, respectively, very close to observed first order resonances (see Table II).

VI. CONCLUSION

Inspired by the study of the thickness resonances in plant leaves [32], a layered sandwich mesostructure has been proposed to explain thickness resonances in FE films. This proposed mesostructure is consistent with the analysis of SEM images. It is shown that the IP can be solved using a SA algorithm and that this sandwich model provides an excellent representation of the measured spectra. The two studied films present skin layers thinner, denser and having lower acoustic impedance, compared with the core layers. These features can be explained by a cellular model assuming that cells in the skin layer are more elongated.

The fact that the observed subtle anisotropy in the film thickness direction induces a mesostructure that does have a measurable impact on the film macroscopic response can be of interest in the optimization of FE films for different applications. For example, provided that the flattened cells are more compressible, it could be of interest to prioritize the polarization of these cells. Alternatively, if for whatever reason, these parts of the film are more difficult to pole, then it would be convenient to minimize their presence. In addition, when considering the problem of matching the acoustic impedance of FE films to water or air, to take into account this sandwich structure and its impedance profile can also be beneficial. In particular, for skin layers that present lower acoustic impedance this can be exploited to improve the performance of air-coupled transducers by taking advantage of this extra capability to match impedances. Another possibility could be the increase of the frequency bandwidth by the convenient design of the thickness resonances of the different layers involved and their potential coupling. Finally, as in many cases FE films in transducers are operated in the non-linear regime [51], the areas of the film with more flattened cells may become more important as they are more likely to respond in a non-linear way. Future work will focus on the analysis of the electromechanical response of FE films to determine if it is possible to extract any information about the distribution of the d_{33} value across the film thickness and to study the possibility of improving FE transducers (air- and water- coupled) by engineering this layered structure.

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