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An FE model of a cellular polypropylene: 
exploring mechanical properties

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ABSTRACT

Several analytical models have been suggested to describe the changes in the electromechanical properties of Cellular Polypropylene (Cell-PP) due to charging. However, there is a limited number of studies considering the non-linear dependence of the piezoelectric coefficient $d_{33}$ on the mechanical load applied. One of the main reasons for this non-linearity is the stiffness of the film that increases proportionally to the applied mechanical load. Moreover the size and shape distribution of the enclosed voids is an important determinant of the electromechanical properties.

In this work, the geometry of a 3D model of Cell-PP is designed on the basis of analytical Splines. Both the manufacturing procedure of Cell-PP films (bi-axial stretching) and the pressure expansion treatment were simulated in order to account for a realistic void distribution. The FEA is done on a 2D cross-section of the modelled film. The modelled mechanical response is analysed based on increasing mechanical load applied. The load-deflection curves obtained from the analysis are then compared to the experimental results acquired via Dynamical Mechanical Analyzer (DMA) to validate the model. Four types of Cell-PP films, expanded at different pressures, were used in this validation. The aim is to develop a model that describes the effect of morphological parameters on the stiffness of the films by simulating the manufacturing procedure.

Keyword list: Cellular Polypropylene, Mechanical properties, Non-linear, Finite elements, Morphology, Mechanical response, Energy harvesting, Cell-PP

1. INTRODUCTION

There are many applications for sensors (Sonar, Accelerometer etc.) as well as actuators (Stacked actuators, speakers etc.), which utilize the piezoelectric and inverse piezoelectric effect respectively [1,2]. Among other piezoelectric materials, polymers were found to have piezoelectric behaviour when poled under high electric fields. Polymers like Polyvinylidene Fluoride (PVDF) and Cellular Polypropylene (Cell-PP) are cheap, non-dense materials whose acoustic impedance is close to that of the air. This further increases their sensitivity over ceramic materials to inputs like vibrations, sound waves etc. [3,4]. Cellular polymers are non-homogenous as they have voids with a variety of sizes. These voids are created during the manufacturing process. Cell-PP for example is created by heating up and bidirectional stretching of Polypropylene mixed with nucleating agent (CaCO$_3$) [5]. Once the mixture cools down, the material gets strengthened as the molecules are now oriented and stay that way. Polypropylene is commonly used in industries for wrapping or package labelling. As it is a cheap, lightweight and flexible material it might be advantageous to be used as a piezoelectric material in applications like Wearable energy harvesting.

Cell-PP exhibits large piezoelectric coefficient in the third direction $d_{33}$. Usually, this is achieved by reducing the stiffness in the third direction $c_{33}$. This reduction is an outcome of pressure and thermal treatment [6-8] and is usually beneficial as piezoelectricity is strongly related to the elastic properties and to the embedded dipole charges inside these materials [9,10].

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In addition, studies done on polypropylene ferroelectrets have shown that by a second expansion process, which occurs after poling and electrodoping of the film, the piezoelectric coefficient $d_{33}$ could be increased by 40% in some cases [11]. However, there is a limitation while using this procedure, as there is a point where the over-expanded films tend to become stiffer [12-13].

A number of models for Cell-PP piezoelectricity have been proposed. Finite element method was employed by Tuncer et al. in order to compare the numerically calculated response with the analytical expression [14]. Other models treat the film as alternating layers of polypropylene and gas, based on the height of the voids [15]. According to these models, the number of voids is counted on the basis of cross sections of the film and categorised to bins based on their maximum height. It is assumed that the same distribution applies for the whole sample. Harris and Mellinger [16] were able to estimate the final charge density of the voids after poling by utilizing the Paschen’s law of breakdown field in gases. The final charge density is determined by the void height and the pressure of the gas inside the void layer. In order to derive the piezoelectric coefficient of the film, the modulus of elasticity (modulus of axial compression $c_{33}$) is considered to be constant.

The above models do not explore the mechanical properties and thus do not consider the non-linearity of the film’s piezoelectricity. The stiffness in thickness direction ($c_{33}$) is typically in the order of MPa, ranging from 1 to 10 MPa depending on the frequency of excitation and the inflation of the sample [17]. Wegener et al. [18] demonstrated that the stiffness of the material depends on its relative density ($\rho_{\text{expanded}}/\rho_{\text{uninflated}}$) and the morphology of the voids. In terms of morphology, the voids are considered to be rhomboids with their dimensions defined by the relative density. This stiffness dependence is non-linear as both low inflated and highly inflated samples tend to be stiffer. There is a break point in the relative density where the film is more compliant. Similarly, response wise, when a mechanical load is applied, the structure changes and as a result the stiffness increases [19,20]. This non-linearity is obvious when the film is experiencing high strains. In another study, a two layer sample was modelled and the electromechanical coupling coefficient was derived in terms of relative density and stiffness of the film [21]. In the later study, morphological parameters were also taken into consideration.

The above suggests that the stiffness of the material is not constant and needs to be described in terms of mechanical stress applied to the film. There is a link between the morphological structure of the film and the stiffness, under high strains, which has not been explored yet. The aim of this study is to provide criteria for characterizing the mechanical response of these films based on their morphology. In the present paper, a variety of samples expanded at two different pressures. The stress-strain curves in compression mode are obtained within a range of 10 to 320kPa. An FE model is created for each of the films and the real and modelled responses are compared.

2. MATERIALS

To study the mechanical response of Cellular Polypropylene films, 3D models of the geometry were produced having similar morphological properties with the original films. The morphological properties were obtained from SEM cross sectional images: distribution of void heights (referred as $D_h$ distribution), void aspect ratio (referred as $D_r$ distribution), total thickness $H$ of the film and porosity $P$ (area of gas / total area). As there is a lot of noise at the output of the image processing program, voids with less than 1 μm$^2$ of area were not considered for further analysis. Log-normal curves are fitted to the observed distributions and the parameters $\mu$ and $\sigma$ (Location and Scale respectively) are estimated. Finally, the inflation of the material was measured between the expanded and uninflated state.

Two types (families) of Cellular Polypropylene, provided by Treofan, Germany were used: the EUH family with nominal density of 550 kg/m$^3$ and nominal thickness of 75μm and the LRH family with nominal density of 680 kg/m$^3$ and nominal thicknesses of 60, 70 and 80μm.

Film morphological parameters were estimated from cross section images obtained with a Hitachi TM3030 Scanning Electron Microscopy (SEM) using a custom made image processing software.

The 3D geometry of Cell-PP was designed on the basis of analytical Splines. The film is considered to be made of $N$ layers of polymer. Each layer was defined by two Splines parallel to X axis with the distance between them designating the thickness of the layer. A set of points, equal to the average number of voids observed in the SEM images, were randomly distributed within the 3D space, between the layers, designating the position of each void. In order to mimic the biaxial stretching applied in the manufacture of those films, discs were created in the X-Y plane of the film with their
radius determined by contact with nearest neighboring disks in the same layer. Initially, the film’s porosity is zero, as the layers are in contact with each other. Simulation of the pressure-expansion treatment is done by increasing the height of each void, so as to achieve the observed aspect ratio (void aspect ratio). During this procedure, the void borders push the Splines, which in turn follow the expansion in the Z direction depending on the voids, above or below them. The procedure is shown in figure 1. The morphological parameters of the modeled material were estimated from 2D cross sections with exactly the same procedure used for the real material.

Log-normal distributions were fitted to both the void height ($D_h$) and aspect ratio ($D_r$) empirical distributions for the real and the modeled films. The estimated parameters of the real films were linked to the modeled films parameters. The statistical suite R [22] with the package fitdistrplus [23] were used for all statistical analysis. The comparison of two empirical distributions was done with Kolmogorov-Smirnov test in order to determine whether they come from the same distribution.

![Figure 1. 3D geometry illustration. a) Layers are stacked to form a solid polymer b) Points are randomly dispersed within the solid and in-between the layers c) Lines, designating the radius of the disks to be created, expand in X-Y plane. Their radius is determined by contact with neighboring disks d) The final height of each void is determined by the product of its radius and the chosen aspect ratio.](image)

3. METHODS

Cellular Polypropylene samples were inflated with a pressure expansion procedure under 2 and 5 MPa for 20 minutes and decompression for 75 and 90 seconds respectively. The gas used for the expansion was Argon. Subsequently, the samples underwent a heat treatment for 10 seconds at 85 °C to make the inflation of the structure permanent. The inflation procedure is shown in figure 2. The samples were then cut to disks of 5mm diameter. Compressive static mechanical analysis tests were done with the aid of a Perkin Elmer DMA 8000. The static uniaxial stress was increased from zero to 330kPa with steps of 10kPa while the dynamic force was held to zero. Simultaneously, a capacitive sensor (Micro epsilon NCDT 6200) logged the displacement of the moving part of the clamp. The stress-strain curves were obtained and the $c_{33}$ constant was derived.

The 2D cross sections of the modelled films were fed to a drafting program to generate a suitable surface for importing into ANSYS FEA software. The solution selected for this 2D analysis was plain stress. A suitable mesh size was selected and pads were placed on the top and bottom layer to ensure for distribution of the compressive load. The geometry was constrained from displacements in any other direction except the direction of the compression. For this analysis, the
voids are considered to be closed pore. As a result, the pressure inside them is subjected to change proportional to their area. The mechanical response was obtained after applying different mechanical loads. The load-deflection curves obtained were compared to the experimental results acquired via Dynamical Mechanical Analyzer (DMA) to validate the model.

**Film Inflation**

![Film Inflation Diagram](image)

Figure 2. Inflation procedure a) Sample in its initial state b) Voids get compressed due to external pressure applied c) The gas diffuses inside the voids. The pressure inside the voids becomes equal to the external pressure d) External pressure is quickly released and the voids expand.

### 4. EXPERIMENTAL RESULTS

#### 4.1 Distributions

It was observed from cross sectional images that as the inflation ratio increases, previously not expanded voids gain height. This separation of the layers happens mostly where CaCO₃ particles exist. The empirical distributions of void heights (Dh) and aspect ratios (Dr) for all the inflated materials at 2 and 5 MPa are shown in figure 3. The parameters of the log-normal distributions fitted to the empirical data are also provided for each material in each plot.

For low inflations, there are no major differences in the inferred distributions among the different materials. In the case of Dr distributions there were differences by material (fig 3b, d) that became more pronounced at the higher inflation (fig 3d). For each material type the location parameter μ for Dr has a bigger value for the more inflated samples indicating that void lengths increased more than void heights following higher inflation. This is more noticeable in the case of LRH80 and LRH70, where the scale parameter σ also increased after higher inflation (5MPa). On the contrary, the Dr distributions of LRH60 and EUH75 did not change significantly. Two sample Kolmogorov-Smirnov (KS) tests for EUH75 and LRH60 yield similarities among the 2MPa and 5MPa of inflation with p-values of p = 0.3161 and p = 0.04 respectively. Additionally, the Dr distribution for these two materials gets narrower (σ decreased) after high inflation (5MPa in comparison to 2MPa). As μ parameter of Dh distribution suggests, voids of LRH60 at 5MPa (Figure 3c) inflated the most, having a comparable void height distribution to the LRH80 at the same inflation (KS test p-value = 0.08364, both μ, σ comparable).
Figure 3. Histograms (relative frequencies) for the empirical distribution of void heights (a,c) and aspect ratio (b,d) based on 5 SEM cross sections of the samples per each of 4 materials. Polymers expanded at 2 MPa (a,b) and 5 MPa (c,d). Insets: the parameters ($\mu$ Location and $\sigma$ Scale) of the log-normal distribution fitted on the empirical data.

From the estimated parameters $\mu$ and $\sigma$, the mean, median, mode, variance, skewness and kurtosis of the distributions were calculated and their statistical differences in terms of inflation factor and/or the material were evaluated (Table 1). Considering the void height distribution, significant increases of almost all parameter values were only due to higher inflation. For the Dr distribution, the only significant differences were found for the location and the mode parameter and were limited to EUH75 samples having significantly lower values compared to LRH70 or LRH80.

Table 1. Analysis of Variance results for the factors (material or inflation) affecting the parameters of a) void height and b) void aspect ratio distribution. ns = no significant effect, * significant difference, p<0.05 ** significant difference p<0.01. The factor level differences, if significant, are indicated.

| Parameter   | Void Height | Void Aspect Ratio |
|-------------|-------------|-------------------|
|             | Material    | Inflation (MPa)   | Material | Inflation |
| location    | ns          | 5>2 *             | LRH70>EUH75 | ns        |
| scale       | ns          | 5>2 **            | ns       | ns        |
| mean        | ns          | 5>2 *             | ns       | ns        |
| median      | ns          | 5>2 *             | ns       | ns        |
| mode        | ns          | ns                | LRH80>EUH75 | ns        |
| variance    | ns          | 5>2 *             | ns       | ns        |
| skewness    | ns          | 5>2 **            | ns       | ns        |
| kurtosis    | ns          | 5>2 *             | ns       | ns        |

It is also evident that materials with higher initial thickness $H$ have higher aspect ratios at high inflations ($\mu$ of Dr, figure 3, increases proportional to nominal $H$ of the film). An exception is EUH75 as its high thickness is due to its external glossy layer which does not inflate.

4.2 Mechanical response

The mechanical response of the materials is described in figure 4. For every stress increment, the new thickness is calculated based on the displacement measured by the capacitive sensor. The strain ($x$-axis) is then given as the True strain:

$$\varepsilon = \int_{l_0}^{l} \frac{dl}{l_0}$$ (1)
Figure 4. Stress over true strain response of all samples. The second part of the names indicate the expansion pressure (5 hollow symbols, 2 filled symbols). The stress increment is 10kPa.

Exponential relationships of the form \( y = a \exp(bx) \), were fitted to the compressive stress (\( y \)) over true strain (\( x \)) data of figure 4. The rate of change of stress with strain, captured by the constant (\( b \)) of the exponent, decreases linearly with the median of the aspect ratio distribution (figure 5). The relationship appears to be linear mostly due to the constraining response of materials inflated at 5MPa (hollow symbols in figure 5). LRH70 and LRH80 differ from LRH60 and EUH75 in both the median void aspect ratio (high values for the former group) and the rate of compressive stress change to true strain (lower \( b \) for the former group). The materials inflated at 2MPa showed intermediate values for both properties with rather small differences among different materials (filled symbols in figure 5).

Table 2. Parameters of the quadratic polynomials \( (c_{33} = a + bX + cX^2) \), X=pressure (Pa), fitted to the data presented in figure 6. The second part of the sample name indicate the expansion pressure Sig.=significance level, ns = no significant difference from zero, * significant difference, \( p<0.05 \) ** significant difference \( p<0.01 \) and *** significant difference \( p<0.001 \).

| SAMPLE    | a     | Sig. | b    | Sig. | c     | Sig. | \( R^2 \) |
|-----------|-------|------|------|------|-------|------|----------|
| LRH60_2   | -7.8e4| ns   | 14.3 | ***  | -3.7e-6| ns   | 0.98     |
| LRH70_2   | 2.6e5 | *    | 13.7 | ***  | 4.6e-6 | ns   | 0.99     |
| LRH80_2   | 2.2e5 | *    | 10.3 | ***  | 6.2e-6 | ns   | 0.98     |
| EUH75_2   | 3.9e4 | *    | 13.9 | ***  | 8.0e-8 | ns   | 0.98     |
| LRH60_5   | 1.9e5 | ns   | 18.3 | ***  | -1.9e-5| ***  | 0.97     |
| LRH70_5   | 4.7e5 | ***  | -1.8 | **   | 1.6e-5 | ***  | 0.95     |
| LRH80_5   | 7.1e5 | ***  | -2.6 | ***  | 2.1e-5 | ***  | 0.97     |
| EUH75_5   | 2.2e5 | ns   | 19.1 | ***  | -1.4e-05| *     | 0.98     |
Figure 5. Relationship between the Median of the Dr and the constant b of the exponent of the compressive stress by the true strain exponential relationship for each material. Open dots: expansion at 5MPa, solid dots: expansion at 2MPa.

A similar pattern of constraining response of the materials inflated at 5 MPa is shown in figure 6 where stiffness constant $c_{33}$ is plotted against compressive stress. For LRH70 and LRH80, $c_{33}$ constant increases slowly; the increase is accelerating but $c_{33}$ remains at low levels up to 350 kPa of compressive pressure. For LRH60 and EUH75 the $c_{33}$ constant increases sharply and decelerates with increasing compressive pressure. In all cases the response is quadratic (table 2) with positive quadratic terms for the former group and negative for the latter. For materials inflated at 2 MPa the response is linear (the quadratic term is not significantly different from zero) and the rate of increase is slightly lower than that of the LRH60 and EUH75 inflated at 5MPa.

Figure 6. Quadratic polynomial fits (lines) of $c_{33}$ depending on pressure for the four materials at two different inflations (2 and 5MPa). Second part of the names indicate the expansion pressure in MPa. Hollow symbols: materials expanded at 5 MPa, solid symbols: materials expanded at 2MPa. The stress increment is 10kPa. The parameters for each curve are given in table 2.
There are uncertainties occurring due to the method of measuring the total thickness of the films and the thickness of the external layers. At high inflations, measurements of the film thickness induce high uncertainties as the films have rough surface due to the expanded voids. Despite that, for the range of interest the difference between LRH70 and LRH80 in terms of their response remains statistically significant. On the contrary, the thickness of LRH60 at 5MPa could not be measured with equal accuracy. There is an uncertainty of 5% affecting the thickness making the strain responses of LRH60 and EUH75 (figure 4) indistinguishable at a 90% confidence interval.

5. MODEL RESULTS

Only two independent parameters are used for the creation of the 3D geometry: the total thickness H of the uninflated material and the Inflation. The number of voids, and their ratio are determined by these two parameters. Since the purpose of this model is to predict the morphological parameters by simulating the biaxial stretching and the expansion procedure, it was calibrated based on one sample. The sample chosen was the LRH80 at 5 MPa of inflation. The model was not re-calibrated for the other films. Cross sections of the real and modelled material at 2 and 5 MPa are shown in figure 7.

![Cross sections of real and modelled LRH60 film inflated at 2MPa top and 5MPa bottom](image)

Figure 7. Cross sections of real and modelled LRH60 film inflated at 2MPa top and 5MPa bottom

To summarize both the results of the modelled and the experimental material, the exponent constant b values of compressive stress to true strain relationship is plotted in the morphological space of the materials defined by the location parameter μ of the void Dr distribution (x axis) and the location parameter μ of the void height distribution (y axis) (Figure 8). Across the x axis the increased aspect ratio implies a weaker structure of the material (i.e. elongated voids that collapse under moderate stress). When a high aspect ratio value combines with a small height, as in LRH60 inflated at 2 MPa, the material exhibits a high stiffness because voids with a small height may collapse and eventually what is compressed under a certain force is the bulk material. This means that in order to have a compliant material with a weak structure, the void height should be high as in the case of LRH80 and LRH70 inflated at 5 MPa. A low aspect ratio value implies a strong material structure with a relatively high stiffness. The stiffness of such materials, like LRH60 and EUH75, with more spherical voids, increases at higher inflations. This is because the average void height is increasing under high inflation while the structure of the material remains strong. The model describes accurately the high inflated materials but the modelled films tend to be stiffer than the real material (figure 8). A reason for this discrepancy may be the difference between the
distributions of morphological parameters of the 3D voids in the real material and the corresponding distributions imposed in the model, which were as observed in 2D cross-sections.

Figure 8. Location parameter $\mu$ of the height distribution ($D_h$) against the location parameter $\mu$ of the aspect ratio distribution ($D_r$). The constant (b) of the exponent of the strain to stress relationship is noted below each point. Empirical data in triangles, model results in dots. Solid symbols are 2 MPa inflation and hollow symbols 5MPa inflation.

6. DISCUSSION AND CONCLUSION

The stiffness of the low inflated materials is determined by their void height distribution. At higher inflations however, the void height distribution and as a result the porosity $P$ do not necessarily decrease stiffness. As an example, LRH60 and LRH80 inflated at 5MPa with porosities of 0.5 and 0.47 respectively share similar $D_h$ distributions but their mechanical responses differ. The major difference among these two samples is the $D_r$ distribution. Our findings are in agreement with Wegener et al. [18], suggesting that the link between the stiffness of a material and its morphology is the aspect ratio of the voids. It is an open question whether the aspect ratio can be a universal parameter for categorising Cell-PP films in terms of stiffness irrespective of the manufacturing procedure.

The mechanism of expansion, on the other hand, is not simple. With the expansion process the voids grow in both width and height direction. Of the films used in this study, those with higher nominal thickness have their voids grow more in length than in height. This has not been observed in previous studies [13,21] where voids obtained a round shape after inflation. However these studies used thinner films.

Films with high void aspect ratio have a weak structure. As a consequence, the majority of the voids with low height collapse under moderate stress levels resulting in a high stiffness value attributed to both the mechanical properties of the bulk material and the increment of stiffness of the voids due to high gas pressure arising in them. When the aspect ratio becomes lower, voids obtain more of a round shape and the structure gets stronger. High stiffness in this case can be attributed to the morphological characteristics of the structure exhibiting truss-like mechanical performance [18].

The same principle applies when the film is under different levels of compression. For low inflated materials and for the given range of stress, voids of successively larger heights collapse progressively, leading to a linear increase of stiffness. This does not hold for highly inflated materials at 5MPa. The rate of change of stiffness $c_{33}$ increases for LRH70 and
LRH80 which share a high aspect ratio (figure 6). On the contrary, there is a decrease in the rate of change of stiffness for LRH60 and EUH75 which share a narrow Dr distribution (Figure 3). This suggests that the aspect ratio gets higher during compression, making the structure weaker.

The FE results showed the weakness of the structure for the highly inflated materials, where the structure differences are more evident (figure 3 c,d). However, the model does not predict the difference, in terms of response, for most of the low inflated materials. This behaviour is confirmed by experimental data: there are minimal differences among low inflated materials in terms of their morphology (figure 3 a, b) and mechanical response (figure 4).

This paper shows that a 2D cross-section can give valuable information about the material response despite the fact that the porosity of the material can be underestimated [24]. It is yet to be shown how the true morphology of the sample compares to the cross sections emerging from them. In any case, 2D sections can be safely used for the comparison of sample morphology. Furthermore, 2D cross-sections provide good representations of the material structure, as excellently shown by LRH80 inflated at 5MPa for which the model has been calibrated, and to a lesser extend for the other materials (where the model was extrapolated).

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