Experimental and numerical analysis of a LLDPE/HDPE liner for a composite pressure vessel

1. Introduction

Storage cylinders for compressed natural gas (CNG) used in vehicles are pressure vessels that have been traditionally produced using isotropic materials, such as steel and aluminum. Nevertheless, polymer composites have recently been introduced for that purpose [1], usually relying on the composite manufacturing technique of filament winding (FW).

Regarding the FW process, a liner is used as a mandrel, allowing the manufacturing of Type 4 pressure vessels according to ISO 11439 [2]. This inner liner, when polymeric, may be produced via rotational molding, which stands out for its low production and tooling costs and results in flexible and impact resistant parts of various shapes [3].

Polymeric liners may be produced based on polyethylene blends. The physical properties of blends are highly dependent on their morphology [4] and the mechanical properties of homogeneous mixtures differ from those of mixtures comprised of separated phases [5,6]. The use of polymeric blends, for instance the physical mixture of two polyethylenes (PE), aims to improve the performance of homopolymers regarding processability and properties, so that good resistance to hydrostatic pressure and low permeability, which are of interest for CNG pressure vessel liners [7], can be achieved.

The need to increase the minimum required strength (MRS) for polyethylene to conform to the ISO 12162 standard led to this study on better performing high-density polyethylene [8]. The use of polymeric blends, for instance the physical mixture of two polyethylenes (PE), aims to improve the performance of homopolymers regarding processability and properties, so that good resistance to hydrostatic pressure and low permeability, which are of interest for CNG pressure vessel liners [7], can be achieved.

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evaluate if the proposed carbon/epoxy composite cylinder would be able to withstand a maximum internal burst pressure of 40 MPa in order to operate under 20.7 MPa.

2. Experimental

2.1. Materials

A medium density polyethylene (LLDPE) DL1002RM hexene-1 copolymer grade (melt flow index: 3.3 g/10 min @190 °C/2.16 kg; density: 0.939 g/cm³; yield strength: 20.5 MPa) and a grade GM 9450 F high density polyethylene (HDPE) with wide molecular weight distribution (melt flow index: 0.38 g/10 min @190 °C/5.00 kg; density: 0.952 g/cm³; yield strength: 26.0 MPa) were used. Both polymers were supplied by BRASKEM S.A and cryogenically micronized.

The specimens were extruded and injected in a Thermo Scientific Hooke Minijet II injector, with injection pressure of 200 bar and cylinder/mold temperature of 200 °C/60 °C. The extrusion of the samples was performed in a single-screw extruder (18 mm), Ciola/MEP-18 brand with two heating zones. The mixtures were pelletized in an automatic SEIBT shredder with ACS 300 frequency converter. The fusion temperature of the samples was 190 °C. The blend composition was chosen based on previous studies [8] in which density, crystallinity and melting temperature of various blends were evaluated.

2.2. Polymeric liner rotomolding process and characterization

Two types of liners have been produced by rotational molding using the same LLDPE/HDPE blend, the Ø72 × 270 mm (diameter × length) prototype liner with an inner volume of 1 L and the actual-scale liner (Ø224 × 725 mm), with an inner volume of 22 L. Fig. 1 shows schematic drawings of both liners.

To produce the prototype liner, a shuttle type rotomolding equipment with LPG gas oven and twin 0.25 HP engines operating at 11 rpm (primary axis) and 20 rpm (secondary axis was used). The cast steel cylinder mold had inner dimensions of 280 mm (length) and 78 mm (diameter), with 1.33 L inner volume. Mold temperature was controlled with a ST20 MINIPA Rayon brand pyrometer, being pre-set to 250 °C. A larger, homemade, industrial rotomolding equipment was used for the actual-scale liner. The cast steel cylinder mold had inner dimensions of 830 mm (length) and 240 mm (diameter), with an inner volume of 37.5 L. The mold temperature was kept at 240 °C and rotation was pre-set to 18 rpm (primary axis) and 20 rpm (secondary axis was used).

Table 1
Relationship between wall thickness (e) and mass of the prototype and real liners.

<table>
<thead>
<tr>
<th>Polymeric Blend</th>
<th>Prototype liner</th>
<th>Actual liner</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>e (mm)</td>
<td>Mass (g)</td>
</tr>
<tr>
<td>Sample #1</td>
<td>1.0</td>
<td>55</td>
</tr>
<tr>
<td>Sample #2</td>
<td>2.0</td>
<td>110</td>
</tr>
<tr>
<td>Sample #3</td>
<td>3.0</td>
<td>160</td>
</tr>
<tr>
<td>Sample #4</td>
<td>4.0</td>
<td>205</td>
</tr>
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</table>

Table 2
Mechanical properties of aluminum 6061-T6.

<table>
<thead>
<tr>
<th>Property</th>
<th>Unit</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Elastic modulus</td>
<td>GPa</td>
<td>63</td>
</tr>
<tr>
<td>Yield strength</td>
<td>MPa</td>
<td>280</td>
</tr>
<tr>
<td>Transversal tensile strength</td>
<td>MPa</td>
<td>330</td>
</tr>
<tr>
<td>Poisson’s ratio</td>
<td>–</td>
<td>0.30</td>
</tr>
<tr>
<td>Shear Modulus</td>
<td>MPa</td>
<td>357</td>
</tr>
<tr>
<td>Specific density</td>
<td>g/cm³</td>
<td>2.70</td>
</tr>
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</table>

Table 3
Mechanical properties of an unidirectional lamina of carbon/epoxy.

<table>
<thead>
<tr>
<th>Property</th>
<th>Unit</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fiber volume fraction</td>
<td>%</td>
<td>70.00</td>
</tr>
<tr>
<td>Longitudinal elastic modulus</td>
<td>GPa</td>
<td>181</td>
</tr>
<tr>
<td>Transverse elastic modulus</td>
<td>GPa</td>
<td>10.3</td>
</tr>
<tr>
<td>Major Poisson’s ratio</td>
<td>–</td>
<td>0.28</td>
</tr>
<tr>
<td>Shear Modulus</td>
<td>GPa</td>
<td>7.17</td>
</tr>
<tr>
<td>Ultimate longitudinal tensile strength</td>
<td>MPa</td>
<td>1500</td>
</tr>
<tr>
<td>Ultimate longitudinal compressive strength</td>
<td>MPa</td>
<td>1130</td>
</tr>
<tr>
<td>Ultimate transversal tensile strength</td>
<td>MPa</td>
<td>48.9</td>
</tr>
<tr>
<td>Ultimate transversal compressive strength</td>
<td>MPa</td>
<td>250</td>
</tr>
<tr>
<td>Ultimate in-plane shear strength</td>
<td>MPa</td>
<td>90.5</td>
</tr>
<tr>
<td>Specific density</td>
<td>g/cm³</td>
<td>1.62</td>
</tr>
</tbody>
</table>
Table 1 shows wall thickness and weight of the obtained liners: sample #1 to #4.

Tensile tests were performed on an EMIC DL 2000 machine with a 2000 kgf (20 kN) load cell operating at 10 mm/min (ASTM D638-00, type IV), and Izod impact tests were performed in an EMIC machine with a 2.7 J pendulum (ASTM D256-00). Five extruded and injected specimens were tested for each sample.

Hydrostatic burst pressure testing of the liners was performed according to ISO 11439 for pressure vessels using two specimens for each sample. Flutrol (150 psi) equipment was used with a HBM 50 MPa load cell and an interface Spindler 8, 60 Hz from HBM to collect the data. The specimens were pressurized at a rate of 0.1 MPa/s and data was processed using Catman 4.0 Professional software.

2.3. Theoretical determination of minimum thickness and ratio of burst pressure for distinct size liners

According to the ASME Division 1 (Section VIII), the minimum required thickness \( e \) for a thin cylindrical vessel (i.e. the liner) to resist a particular hydrostatic pressure \( P \) may be calculated using Eq. (1) [9]:

\[
e = \frac{P \times R}{\sigma_y - 0.6P}
\]

where \( R \) is the inner radius (mm) and \( \sigma_y \) is the yield strength of the polymeric material (MPa). The yield strength provided by the manufacturer and the experimentally measured yield strength were termed \( \sigma_{y_{inf}} \) and \( \sigma_{y_{exp}} \), respectively.
The parameter \( a \), defined in Eq. (2), may be used to compare the burst pressure performance of cylinders of distinct sizes, in this case, the prototype and the actual cylinders [10]:

\[
a = \left( \frac{D^2 - d^2}{1.3D^2 + 0.4d^2} \right)
\]

where \( D \) and \( d \) are the external and the internal diameters of the polymeric liner, respectively.

2.4. Finite element simulation procedure

The wound composite modeler (WCM) Abaqus plug-in was used to model the carbon/epoxy layers, which generates an output subroutine comprised of structural geometry and winding layout parameters, stress and strain along the fiber direction and also transverse to the fiber direction. For the purpose of failure analysis, a subroutine was written in Python to enable evaluation of the change of mechanical properties due to failure criteria.

FEA simulations of the composite pressure vessel were conducted using ABAQUS/CAE 6.8 considering linear elastic behavior of the polymeric liner and the carbon/epoxy laminate. The mechanical behavior of the component was estimated based on: micromechanical models (strength of material and elasticity approach) to predict the lamina properties, classical laminate theory to predict laminate properties; elasto-plastic behavior of the liner, von Mises stress criterion for the liner and Tsai–Wu failure criterion for the composite laminate [11].

Fig. 4. Hydrostatic burst pressure for the prototype liner as a function of the wall thickness: Theoretical calculation using supplier data (●) or experimental data (■), and comparison with actual burst pressure testing (▲).

Fig. 5. Typical results for the actual liner: (a) Curve obtained during hydrostatic pressure testing, and (b) Visual aspect of the fracture region.
The shell was modeled as an axisymmetric continuum with a number of sub-layers, each of them with orthotropic material properties. A multi-layered filament-wound vessel wall was considered in the analysis based on the elasticity solution. The CAX4R element type was used and the mesh model of the pressure vessel included 108,436 elements and 109,018 nodes. Reduced integration was applied to the stiffness matrix shear components and full integration to the other matrix elements. The winding path was calculated using the established non-geodesic fiber trajectories, according to the shape of the dome of the liner designed and built for this study. It is important to point out that the top and bottom domes of the pressure vessel presented different curvatures because of the cast steel cylinder mold project.

The carbon/epoxy laminate was built using angle-ply layers with fibers oriented at positive and negative directions of the same angle. Six different preliminary FEA simulations were carried out using sub-laminates with layers oriented at ±10°, ±20°, ±30°, ±40°, ±50° and ±60° and, according to these simulations, the ±40° laminate showed the best performance as regards strength.

Aluminum, for the metallic boss, and carbon/epoxy characteristics, input data in the FEA simulation, were taken from literature [12–14] and are listed in Tables 2 and 3, respectively. The properties of the polymeric liner used in the simulation were experimentally determined in this work. The cylinder wall consisted of the polymeric liner and five carbon/epoxy sub-laminates. Each hoop (90°) and helical (±40°) sub-laminate consisted of many laminas.

Fig. 2 shows the ply stacking sequence and the material orientation angles used in the simulation. Each helical layer is 0.5–0.75 mm thick and each hoop layer is 1.00 mm thick. The overall lateral thickness of the cylinder was 37 mm.

3. Results and discussion

3.1. Experimental and theoretical results

Table 4 presents the physical and mechanical results obtained for the 95% of LLDPE/5% of HDPE blend used. All results were in the expected range for this type of blend.

Fig. 3(a) presents the response of the 4 mm-thick prototype (liner only) to the hydrostatic test, where a burst pressure of 2.0 MPa was found. Fig. 3(b) shows longitudinal (brittle) fracture at the center and transverse (ductile) fracture at the center and the left end. The brittle rupture occurred first, yielding microcracks parallel to the longitudinal axis of the pressurized cylinder. The ductile rupture occurred during the final fracture, propagating perpendicular to the longitudinal axis of the cylinder. The brittle behavior may have occurred due to disruption of the amorphous regions of the polymer, whereas in the ductile region fracture leads to higher elongation which may have occurred after the deformation of the polymer crystallites [15].

Fig. 4 shows the theoretical calculation (using Eq. (1)) of the maximum pressure for the prototype liner as a function of the wall thickness along with the experimental hydrostatic burst pressure test result. Eq. (1) was applied using both the yield strength reported by the manufacturer (σ_{yил} = 21.25 MPa) and the measured value (σ_{yexp} = 15.35 MPa).
in Table 4). It can be noted that the measured burst pressure lay within the range defined using $\sigma_{y_{exp}}$ and $\sigma_{v_{exp}}$ values.

Fig. 5 shows the typical response of the actual liner (wall thickness $= 12.5$ mm) submitted to the hydrostatic pressure test. Macroscopic analysis of the specimen showed initial longitudinal (brittle) rupture at the center, followed by transverse (ductile) rupture at the center and at the far left, reaching a maximum pressure of 1.63 MPa. Thus, the fracture behavior of the actual liner was similar to that found for the prototype.

Fig. 6 shows the theoretical calculations of the maximum pressure for the actual liner as a function of the wall thickness, along with the experimental hydrostatic burst pressure test result. Eq. (1) was again applied using both the yield strength reported by the manufacturer and the measured value. However, in this case, the pressure test data showed better agreement with the theoretical values obtained with the measured yield strength.

In order to compare the strength values obtained for the prototype liner with those obtained for the actual size liner, which belong to distinct size scales, the proportionality relation given by Eq. (2) was used [12]. Table 5 shows this equivalence relation of pressures, allowing comparison of the tensile strength of the prototype and the actual liners. It can be seen that the pressures are very close when analyzing similar ratios, i.e. 3.1:1 to 3.2:1 thickness ratio of the prototype and the actual liners, referring to the 3.0/9.5 and 4.0/12.5 mm wall thickness samples.

### 3.2. FE liner analysis

In a preliminary study, the numerical simulation of a tensile test was conducted to validate the physical material model chosen for the simulation. Fig. 7 shows the results of the numerical simulation and the experimental stress-strain curves, for engineering and true stress, obtained during actual tensile testing. The numerical simulation results were closer to the true stress curve and the maximum axial true stress was 21.1 MPa. Thus, comparison of the actual elastoplastic deformation curve and the results of the numerical simulation allowed validation of the numerical FEA model using the experimentally determined $\sigma_{v_{exp}}$ value.

Table 6 shows the variation of the numerical von Mises stress in the central region of the actual liner, for distinct thicknesses, when the simulated maximum hydrostatic pressure is reached. It can be observed that, in all cases, von Mises stress varies within 20–21 MPa. Fig. 8 shows the distribution of von Mises stress for the actual liner (thickness: 13.9 mm) and it is possible to verify that the central section of the liner is the weakest region. Also, the burst pressure (1.84 MPa) was very close to the experimental burst pressure obtained (1.86 MPa), as shown in Table 6. The same was found for the 12.5 mm-thick liner, but the 9.5 mm and the 15.3 mm-thick liners showed poorer agreement (18% and 17% error, respectively).

### Table 6

<table>
<thead>
<tr>
<th>e (mm)</th>
<th>Numerical Burst pressure (MPa)</th>
<th>von Mises stress (MPa)</th>
<th>Experimental Burst pressure (MPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>9.5</td>
<td>1.23</td>
<td>20.8</td>
<td>1.50</td>
</tr>
<tr>
<td>12.5</td>
<td>1.62</td>
<td>21.0</td>
<td>1.64</td>
</tr>
<tr>
<td>13.9</td>
<td>1.84</td>
<td>20.2</td>
<td>1.86</td>
</tr>
<tr>
<td>15.3</td>
<td>2.00</td>
<td>20.9</td>
<td>2.40</td>
</tr>
</tbody>
</table>
Fig. 8. Von Mises stress range found for the actual liner (13.9 mm thick).

Fig. 9. Composite carbon/epoxy simulation: (a) Tsai-Wu failure analysis - ply#2, sub laminate 1, and (b) deformation at the bottom dome.
3.3. FE composite shell analysis

The 40° filament wound composite shell for the 15.3 mm-thick liner was modeled according to the stacking sequence previously shown in Fig. 2. Analysis was carried out considering the Tsai-Wu failure criterion and the chosen inner pressure was 20.7 MPa, with a safety factor of 2.0.

Fig. 9a and b present the results of the FEA simulation for 40.0 MPa inner liner pressure. The lowest strength ratio was 1.039. This critical condition occurred at the ply#2 of sub laminate 1, and the largest elongation was found at the bottom dome (3.053 mm).

4. Conclusions

Analyzing the results, the ideal thickness of the actual liner to withstand a maximum pressure 2.0–2.2 MPa was found to lie within the 15–16 mm range. The elasto-plastic model used for the polymeric blend liner material showed agreement with the experimental pressure tests and the ASME procedure. In order to account for manufacturing tolerances, a 15.3 mm nominal thickness was chosen for the liner. The hydrostatic pressure limit of the actual liner was 2.0 MPa and the characteristic fracture behavior changed from brittle to ductile.

To withstand the 20.7 MPa operating pressure commonly applied to CNG cylinders, the liner must be used as a mandrel on which carbon/epoxy layers are wound using the filament winding process following, for instance, the lay-up design (thickness and stacking angles and sequence) proposed in this study.

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References