THICK COMPOSITE DESIGN FOR HYDROGEN VESSELS: A CONTRIBUTION TO COMPOSITE DESIGN METHOD

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Abstract: Hydrogen stock vessels add new product specifications because of higher pressure use. Today static application leads up to 700 bars. But pressure devices must resist, because of certification, at 3 times pressure (2100 bars). Composite vessels give material-structure potential solutions. But designers face limits of knowledge due to actual good practices of thick composite structures use. This paper presents levels of knowledge enrichment for designers (multi-scale experiments and models), analysis of process influence on models improvement, and a discussion for an efficient testing strategy (meaning virtual testing in order to reduce the number of tests and their costs).

Keywords: Composite, Design, Virtual testing

1. Introduction

Today, composite structure is the solution for high performances structures. Transport industry turned this advantage for lighting structures and optimising specific strength. The researchers focus on thin structures behaviour prediction and sizing. But other applications, such as hydrogen storage for energy use, increase product requirements (maximum pressure) in order to give higher performances (storage capacity). For example, hydrogen pressure vessel leads up to 700 bars in static application. The vessel needs to pass the qualification pressure test at three times to the maximum in-service pressure. Consequently the structure must support 2100 bars. Only thick composite solutions (40mm thickness) can give capable products.

But actual knowledge in composite design is out of this scope, due to the thickness of these structures. Behaviour models should be enriched to overpass the thin layers hypothesis that are no longer relevant (such as out of plane effects neglectable), and to integrate process parameters effects on material - structure [1] (thin structure limits for cylindrical shape: radius/thickness>10). Another challenge exists in the intrinsic composition of composite product, because data and models are available at different scales (from the fibre - matrix micro level to the structural assembly macro level). The design starts from a micro level choice (fibre and matrix choice, and fibre volume fraction) to a structural final design (stacking: number of plies - angles - thickness). Transports of models from one scale to another highlight the needs of exchanges between the design and the material and structure characterization. But this is time consuming and expensive. In order to be cost efficient, it is necessary to enrich the thick composite models and to improve the design. Designers need relevant models to use virtual testing and validation.

The design parameters for vessel optimization (size and cost) are the component selection (fibre and matrix) and the thickness (stacking of wounded layers architecture constrained by the winding process limits). The vessel size and the maximum pressure give the strength limit supported by the structure. Internal nearly circumferential layers must support maximal stress up to 2500 MPa. Today, composite's designers oversize the vessels, with no evaluation of their margin. This paper proposes to focus on the exact needed characterization needed during the design stages. Given the specificity of the filament winding process (fibre tension, curing cycle, winding patterns, shape of parts...) in a thick composite background inducing properties variability and thickness gradient, it is necessary to ask the question of scale and geometry, relevant to the characterization of the mechanical properties of the composite material and structure. Thus, mechanical tests from different samples are performed (coupons coming from plates, small diameter wound tubes, and large diameter wound tubes) based on standardized tests, but also with a specific tests especially developed. The results show that there can be major differences in the mechanical properties obtained depending on the coupon definition and testing scale. Consequently the structure sizing depends on the material variability and the process impact on the structure behaviour. First these physical phenomena should be integrated in the simulation models, then a optimization of the structure for mass/cost reduction could be performed.

After an introduction on the design - sizing method and the physical origin of the differences in section 2, we propose a comprehensive approach to characterize the mechanical properties of composite wound material and structure in section 3. The results show that the process parameters impacts must be integrated, on one hand, in the structure behaviour models, and, on the other hand, in the design criteria base of the design method presented in section 4.

2. Design and sizing of composites pressure vessels

The design of composites wounded pressure vessels relies on:
• A good knowledge of filament winding process. This process is mainly used for the realization of cylindrical or axisymmetric parts, with little adaptation for plate panels. Wound structures are made by superposition of hoop winding at an angle nearly 90°, and helical winding with angle from 5° to 80°. Indeed, the choice of a winding angle induces the geometry, especially in the dome. If openings are required in the dome, designer should fix

winding angle with regards to opening diameter and winding trajectory (geodesic or not, with or without friction). The winding of helical layers also provides a pattern at the crossing of filament [2][3], giving a zone with a slightly different behaviour. The fibre tension applied during winding also modifies the properties of the layer, by compaction of the previously deposited layers [4][5]. Finally the curing of wound structures often differs from other composites processes by the lack of pressure or vacuum (by the way of autoclave or vacuum bag) applied during the cure [6].

• The use of specific sizing model. Especially in the cylindrical part, analytical models have been developed [7][8], making the distinction between thin and thick pressure vessel. We enriched the model proposed by Xia [7] with specific developments including modelling of residual stresses and consideration of variable mechanical properties in each layer [9].

In the specific case of pressure vessel, the use of elastic behaviour is classically made. Thus, as the loading is biaxial and as fibres are disposed along the two directions of loading (respectively thanks to hoop winding and helical winding), the damage that can occur in transverse direction or in shear has usually negligible effect in the stress redistribution. In addition, as the shape of pressure vessel has no edge, the risk of failure by initiation and propagation of delamination is clearly limited. The prediction of failure of pressure vessels is then based on a maximum strain or a maximum stress failure criterion in the fibre direction. A first way to introduce damage effect could be done here, by changing the stress/strain to failure according to damage level [10] but it implies specific testing.

As models are described at the layer scale, to perform computations, the minimal requirement is to have all the elastic constants at this scale and fibre resistance. Composite design starts at the micro level (fibre-matrix) to propose a first selection of fibre, matrix and their ratio. Then, the architecture is defined (multi-layers and orientations). This needs micro-macro transformation model and mechanical data from the single constituent to the structural level as illustrated in figure 1. In the next section, several ways to obtain these constants are proposed.
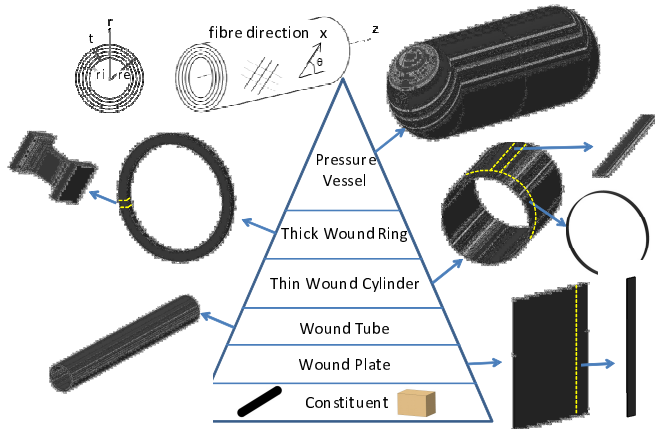

Figure 1. Samples from filament wound structures for mechanical testing

2.1. Overview of available samples for testing

The testing of composite materials could be done at several scales (Figure 1). The first one is the constituent scale, respectively fibre and matrix. By using micromechanics models, one could obtain layer properties. Unfortunately, predictions made by this way lead often to inaccurate results [11]. So the use of constituent properties and micromechanics models will be limited to material screening activities.

As we focus on filament wound structures, the main idea is to test coupons obtained with filament winding process. Thus, it allows including the specificities of this process detailed in the introduction section. In our work, we consider many samples: i)

panels (unidirectional layer or cross layers), ii) tubes with small diameter with hoop layers or 30° and 45° helical layers, iii) thin cylinders with large diameter, that can be used as thin ring or curved tensile coupons and iv) thick cylinders with large diameter that can be used as ring or out of plane coupons.

Classic testing method and coupons do not care of the thickness influence during the identification or validation of the elastic constants used in the design.

2.2. Mechanical properties required

The stress is related to the pressure (p), the internal (r_i) and external (r_e) rays, and the position in the structure (r): hoop stress $\sigma h(r) = \frac{r_i^2 p}{r_e^2 - r_i^2}\left(1 + \frac{r_e^2}{r^2}\right)$. In composite, stress evolves in each layer k due to the fibre orientations ($+/-\theta$ and nearly 0°). The generalised Hook law links stress and displacement in each layer (k) using the stiffness matrix: $[\boldsymbol{\sigma}] = [\boldsymbol{S}][\boldsymbol{\varepsilon}]$. Xia detailed the calculation of the displacements and stress in each ply (k): $u_r^{(k)}(r) = A^{(k)} r^{\beta(k)} + B^{(k)} r^{-\beta(k)} + \alpha_1^{(k)} \varepsilon_0 \cdot r$ (radial displacement at radius r in ply k) [7]. $\beta^{(k)}$ and $\varepsilon^{(k)}$ are related to the stiffness parameters of the ply k (modulus and Poisson ratio). A and B are constant values (in the k ply) coming from the resolution of the equilibrium on the cylindrical part: $2\pi \sum_{k=1}^{N} \int_{r_k}^{r_{k+1}} r^2 \tau_{z\theta}^{(k)}(r) dr = 0$. A full set of mechanical properties is required to make computations. In section 3.2 we will validate the hypothesis of transversely isotropic in order to reduce the independent elastic constants from nine to only five.

The stress in each ply is compared to the failure criterion in the fibres direction. As the key design issue is the burst pressure of the vessel, it is important to focus on properties that modify the prediction of this pressure. A sensitivity analysis has been made in order to show the properties that have to be measured with accuracy. A wide range of values has been given to each parameter and introduced to the analytical model. Pressure prediction shows important sensibility to: **a)** Modulus in fibre direction E_X, **b)** Out-of plane transverse modulus E_T, **c)** Strength in fibre direction X.

In addition, thick structures mean process defects and material quality and properties variations, such as fibre volume fraction and void content. As far as thin pressures vessels are concerned, typical values of fibre volume fraction and void content obtained on wound structures are close to these obtained on elementary samples shown in figure 1. For the thick vessels, a physical study [9], performed on thick pressure vessel, was made to determine layer by layer thickness, fibre volume fraction and void content. It exhibits that these properties vary both through the thickness and in function of type of layers. Then, one should take into account this variability when designing a pressure vessel. The design method proposed to incorporate material variability is developed in section 4 of this paper.

3. Mechanical testing results

For composite materials, numerous mechanical tests exist in the literature [12]. We focus on tests relevant to our samples, for in-plane properties and also for out-of plane properties.

3.1. In-plane properties characterization

In addition to the mechanical properties introduced in 2.2, the Poisson ratios (v_X) and shear properties (modulus and strength) complete the set of data needed to validate the structure mechanical behaviour. Table 1 shows the different mechanical properties needed for vessel design and the list of coupons and wounded structures dimensions tested (results see table 2).

Coupons drawn from wound plates were tested using normalized tests [13][14]. Plates with 1.2 mm, 2 mm and 6 mm

thicknesses present large differences on void content. ISO Standard proposes some adjustments to improve the compaction during the curing process [13], but it makes the manufacturing method differ from classical filament winding process with a remaining higher level of voids. Thus, winding is not adapted to manufacture plate panels. Consequently the first level of material characterisation (for micro level model) does not give relevant data for starting the design process

Table 1 Sample dimensions / Mechanical Properties measurements

Table column headings	E_X	v_X	X	E_T	T	E_S	S
Plate 1,2 and 2mm – UD0°	x	x	x				
Plate 1,2 and 2mm – UD10°						x	x
Plate 2 and 6mm – CrossPly ±45°						x	x
Plate 6 mm – UD90°				x	x		
Ø310 cylinder, 2 mm thick, ±15°	x*						
Ø310 cylinder, 6 mm thick, ±45°						x	x
Ø310 cylinder, 6 mm thick, 90°				x	x		
Ø32 tube, 2 mm thick, ±45°						x	x
Ø32 tube, 2 mm thick, 90°				x	x		

*Back-calculated property, using other measured properties
Notation : Fibre direction X: E_X: Modulus, v_X: Poisson ratio, X: Strength
Transverse direction T: E_T: Modulus, T: Strength
Shear: E_S: Shear Modulus, S: Shear Strength

Wound cylinders with 310 mm diameter were also used for in-plane properties characterization. Tensile coupons were made by using specifically shaped tabs [15]. The main interest with this type of specimen is that the sample is really representative from the process used for structures. One should also be careful when cutting the coupons from the main cylinder. Its main disadvantage lies in the fact that it is impossible to produce samples with 0° direction. In our study, ±15° coupons were used to estimate the longitudinal modulus by back-computation using laminate plate theory and properties obtained with other coupons (mainly E_T and E_S modulus).

32 mm diameter tubes were used with ±45° and 90° orientations [15]. The coupon presents no free edge, that avoids damage or failure starting from the edge, but the high curvature introduces a bias in the analysis. One can notice also that the fixture used [16] for this test is quite complex. Moreover, the load transfer must stay reasonable.

3.2. In-plane results synthesis

Table 2 gives the main results of mechanical characterization. For each sample the average fibre volume fraction (Vf) is given. We can notice important differences on values obtained on modulus (5% in fibre direction, 27 % in transverse direction, 26% in shear) depending on the type of sample used. As a set of properties is required, we have to choose a mean value for each property and associate it to a nominal fibre volume fraction fixed at 65%. As transverse strength Y and shear strength S are not used in the analytical model, no standard values are proposed.

Table 2 Mechanical Characterization results and relevant data selection

	E_X (GPa)	v_X	X (MPa)	E_T (GPa)	T (MPa)	E_S (GPa)	S (MPa)
Plate Vf	118^{+13}_{-11} 66,5 %	$0,32^{+0,01}_{-0,01}$ 66,5 %	2389^{+123}_{-147} 66,5 %	10° Coupons → $6,2^{+0,3}_{-0,3}$ 66,5 % ±45° Coupons →	$17^{+1,6}_{-1,6}$ 66,5 %	$7,6^{+0,6}_{-0,6}$ 66,5 % $7^{+1,2}_{-1,2}$ 66,5 %	$39,8^{+1,9}_{-2}$ 66,5 % $42,7^{+2,7}_{-2,7}$ 66,5 %
Cyl. Ø310 Vf	$124,1^{+1,4}_{-1,4}$ 65 %			$8,5^{+0,6}_{-0,3}$ 65 %		$8,0^{+0,2}_{-0,2}$ 65 %	
Ø32 tube Vf				$6,2^{+0,1}_{-0,2}$ 65 %	$30,3^{+0,1}_{-0,2}$ 65 %	$7,0^{+0,4}_{-0,4}$ 65 %	$63,8^{+6,5}_{-2,3}$ 65 %
Std Value Vf =	124,1 65 %	0,32 65 %	2335 65 %	8,5 65 %		8 65 %	

We retained the values obtained with the large cylinders due to their representativeness filament wound structures. But it gives

macro information on the aggregation of multiple wounded layers. There is a macro to a micro decomposition to go to ply properties. The single ply properties are compared when available with the results on plates.

The tensile strength in fibre strength direction X obtained is low in regards of classical strength for this type of composite material. As it appears to the most critical property concerning the sizing of pressure vessel, we have to investigate alternative testing.

3.3. Characterization in fibre direction

Numerous studies have been led to define a testing method relevant for wound composite characterisation. Most of them use rings drawn from cylinders or thin pressure vessels. Internal pressure is realised with mechanical internal disks [17][18]. The experimentation is easy, but the loading is not uniform in the ring. Hydraulic device to pressurize the ring is an alternative [19][20]. It ensures a uniform loading with uniform internal pressure loading, but the use of the device is quite tedious to obtain. High pressure hydraulic energy is necessary to obtain the failure of the ring and Hwang notices that the pressure applied to the composite ring is difficult to estimate [21].

Consequently, we developed of a new ring test using mechanical device and reproducing internal pressure [9][22]. The aim is testing thin rings for mechanical characterization in fibre direction, as well as testing thick rings to study more widely the behaviour evolution of the pressure vessel through the thickness using multi measurement testing device.

3.4. Out of plane properties

As far as thick structures are concerned, the knowledge of out-of plane modulus is required. Usually, the assumption of transverse isotropy is made, considering the same value for in plane transverse modulus and out-of plane modulus. We have made the characterization of this modulus to ensure the assumption is true. Tensile and compressive tests were led, using RARDE sample geometry [23] and measured both modulus and strength. Coupons were machined by water jet cutting from a thick ring (inner diameter 310 mm, thickness 38 mm). Results are mean out of plane modulus of 7,47 GPa in tension and 8,02 GPa in compression. These values are inside the interval of values [6,2 - 8,5 GPa] obtained on in-plane transverse modulus characterization, and quite close when looking the retained value of E_T (8,5 GPa). The transverse isotropy assumption could then be made.

4. Integration of variability

In thick pressure vessels, the physical properties of the layers are not constant. Thickness of each layer, as well as fibre fraction differs from one layer to another. It is necessary to be able to provide mechanical properties for each layer.

In this way we use a set of micromechanics relations proposed by Christensen [24] to determine fibre and matrix components properties from properties measured at the layer scale. These approach inverses the classical approach of composite design starting with the rule of mixture (ROM). Values obtained differ slightly from the ones that could be measured directly on constituents (e.g. value of matrix modulus that exhibits a lower modulus value). These calculated properties are then called "apparent" constituents properties, and ensure correct mechanical properties value at the layer scale. This is a direct feedback for the designer on the product effective mechanical performances because these measures intrinsically integrate process impacts and thickness influence. This reverse identification introduces variability into the properties coming from the pressure vessel scale.

Figure 2 sets up the principle of modelling and designing the pressure vessel. The approach combines micromechanics rule of mixture for a pre sizing structure (bottom up on the figure 2). This is combined with the structural reverse identification on a scale one ring from initial design, to optimise the material data (apparent constituent properties) and then the design parameters. Designer can then adjust layers number and thickness, ply angle and sequence or question the choice of fibres grade and the matrix selection. This gives the evaluation of the margin on vessels sizing. Today designers choose in a limited selection of winding angle and stacking, based on experience. This calculation environment gives the possibility to explore alternatives under winding process and vessel geometry constrains.
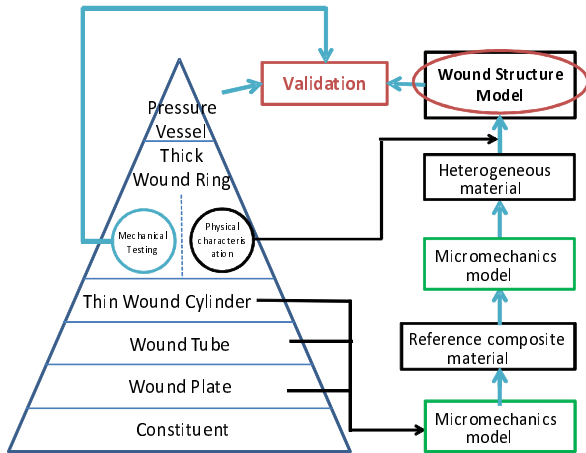

Figure 2. Introduction of variability in the design process

5. Conclusion

The design of composites vessels is based on the good knowledge of the filament winding process and on the use of specific sizing model. It also needs a set of composite material properties at different scales of the structure. To characterize these properties, numerous test methods are available in standards or in the literature. Testing made on different kinds of sample exhibits mismatch in values obtained. We promote reliable results available with samples with shape close from the shape of classical wound structures (large diameter cylinder). As thick structures are concerned, the design method used should include the variability of properties through the thickness. A method to derive each layer properties from a reference set of mechanical data and given physical properties, based on the use of micromechanics model, is proposed. The calculations give the burst pressure results to the designer based on its choices (material selection and ply architecture and number) or can be used as an optimisation tool for an expected burst pressure to optimise the structure definition
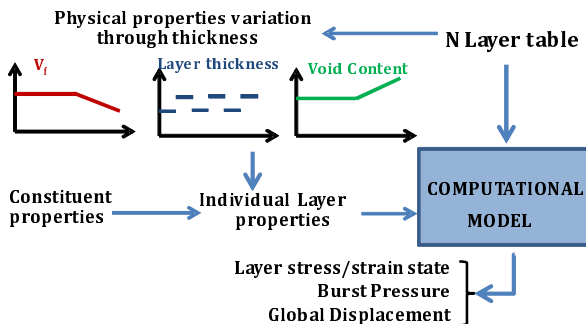

Figure 3. Thick composites pressure vessel design model

Figure 3 sums-up the integration of the variability in the design approach. It combines the constituent data with the apparent properties. Variation should be introduced by the designer using models that describe the change of properties such as ply thickness, void content or fibre volume fraction through the thickness. These rules come from experience feedback on similar structures or from physical analysis of the vessel at the fibre-matrix level. This study integrates the process impacts into thick wounded vessels model. For a 39 mm structure (57 layers: [(±15/±25/902/±35/±45/902)7/90]), it gives for the ultimate pressure (calculated for Vf=65%): 1834bars with standard value, 2443bars with a standard Rule of Mixture. It can reach (depending to the failure criteria FC) 2488bars (Halpin Tsai FC +ROM) or 2527bars (Puck FC + ROM).

These material data, with process impact integration, and the calculus models, give to the industrial partner their margin (20% on the maximum pressure). It opens the optimization design step. This method extends a set of composite sizing tools developed to enhance eco-design of composite structures for optimisation of both mechanical and environmental performances [25].

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