In Situ Estimation of the Thickness of Oxidized Layer in Composite Structures by Means of Ultrasonic Guided Waves

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ABSTRACT

Under the exposition to air rich in oxygen, oxidation leads to a non-negligible thickness of oxidized layer (TOL) which appears initially on the free surfaces of composite structures. This oxidized layer has modified mechanical properties in comparison with the virgin ones and is progressing during time within the composite thickness as evidence of diffusion-limited oxidation. Being able to estimate *in situ* the TOL of composite structures under service is thus of great importance for safety and maintenance purposes. Ultrasonic guided waves (UGW) propagating in aged composites will have different velocities in comparison with UGW propagating in virgin composites. Taking benefits of this physical properties linking TOL thickness to UGW velocities, a strategy allowing to estimate *in situ* the TOL of composite materials is here proposed. As a first step towards its implementation, this strategy is here validated numerically using semi-analytical models allowing to predict UGW velocities in composites as a function of TOL. This model allows to demonstrate the sensitivity of the various UGW propagating modes to TOL thickness for signals representative of SHM applications. As a next step, the proposed approach will be used experimentally for the evaluation and tracking of the *in situ* growth of TOL. Such a tool can be deployed *in situ* and help to follow the health state of composite structures in practical applications.

KEYWORDS: Thickness of oxidized layer, Ultrasonic guided waves, Structural health monitoring

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INTRODUCTION

Ageing of composite materials under exposure to temperature in air due to oxidation is an important issue for transport applications relying on composite structures [1, 2, 3]. Indeed, under the exposition to air rich in oxygen, diffusion-limited oxidation leads to a non-negligible thickness of oxidized layer (TOL) which appears initially on the free surfaces of composite structures. This oxidized layer is progressing during time within the composite thickness as oxygen go deeper into the material and it is consumed when reacting with the polymer network. The TOL has modified mechanical properties in comparison with the virgin composite materials. Consequently, being able to estimate *in situ* the TOL of composite structures under service (*e.g.*, in parts exposed to temperature) is of great importance for safety and maintenance purposes.

Ultrasonic guided waves (UGW) [4] propagating in composite structures are however sensitive to the material properties of the composite material in which they propagate. More precisely, their velocities directly depend on their Young moduli and density. Consequently, UWG propagating in aged composites will have different velocities in comparison with UGW propagating in virgin composites. Taking benefits of this physical properties linking TOL thickness to UGW velocities, an UGW-based strategy allowing to estimate *in situ* the TOL of composite materials is here proposed. Such ideas have already been explored in the literature [5, 6, 7, 8, 9, 10], but never *in-situ* nor in the realm of structural health monitoring which is the purpose here.

This strategy is here validated numerically using semi-analytical models allowing to predict UGW velocities in composites as a function of TOL. This model allows to study the sensitivity of the various UGW propagating modes to TOL and to select the most sensitive propagating mode along with the most sensitive frequencies. Guidelines for a future step where the proposed approach will experimentally validated on composite samples equipped with piezoelectric elements and aged under controlled conditions of exposure time at constant temperature are also provided. The proposed approach is expected to be used for the evaluation and tracking of the *in situ* growth of TOL. Such a tool can be deployed *in situ* and help to follow the health state of composite structures in practical applications.

LAMB WAVES IN AGED COMPOSITES

MODELING OXIDIZED COMPOSITES

Lamb waves are guided waves that can propagate in structures that can be qualified as "*thin*", meaning that two of their dimensions are large with respect to the third one. Typically, the length and width are much larger than the thickness in composite structures dedicated to aeronautical or maritime applications thus making them belong to the class of thin structures in which Lamb waves can propagate. Lamb waves are guided by their host structure and consequently their propagation characteristics (wave speed among others, also denoted as phase velocity) depends on the material properties over the whole thickness of the material composing the host structure [4].

Composite materials are by essence made of several superimposed plies having different materials properties and different orientations. When the composite is exposed to temperature in air, it will thus endure oxidation due to oxidation mechanisms (i.e., oxygen diffusion and oxygen reaction). Consequently, both outer sides of the material will degrade over a certain thickness (denoted as the thickness of oxidation layer, TOL) leading to modified mechanical properties of those plies within the TOL. As oxidation progresses, this TOL with increase with time, and consequently, the mechanical response of the whole material will be affected.

The composite material considered here is build up with six 2D woven composite plies having a thickness of 350 µm for a total thickness of 2.1 mm. The material properties of one ply, either virgin or oxidized are provided in Table 1. It is then assumed that the effect of gradients of oxidation are negligible and they can be translated directly by the finite TOL. From this table it can be seen that the plies under study are orthotopic transverse isotropic, due to the fact that they are 2D woven. It is assumed also that oxidation only affect the matrix degradation, not affecting the fibers neither the interface between matrix and fibers which are not considered here. Under these assumptions, it can be noticed that the mechanical effects of oxidation are rather small: increase of material properties and reduction of the density.

	$E_1 = E_2$	E_3	$G_{12} = G_{13} = G_{23}$	$\nu_{12} = \nu_{13} = \nu_{23}$	ρ
Virgin	70 GPa	6.34 GPa	2.88 GPa	0.3	1530 kg/m^3
Oxidized	70.7 GPa	8.71 GPa	4.25 GPa	0.3	1470 kg/m^3

Table 1: Material properties of one composite ply

In terms of model, it is assumed that the oxidation process will divide outer plies in two with part of it having a thickness equal to TOL possessing the mechanical properties of the oxidized material while the remaining part stays virgin. The oxidized composite is thus no more a 6 plies composite material but a 8 plies one, with the two outer plies being divided in an oxidized part of thickness TOL and a remaining virgin one of thickness $h_{ply} - TOL$, as illustrated on Figure 1.

Figure 1: Illustration of the modelling assumptions for aged composite material

DISPERSION CURVES OF OXIDIZIED COMPOSITES

The underlying idea of the proposed approach is that due to these material properties changes in the TOL, the resulting propagating properties of Lamb waves will change with time. It is thus expected that by tracking those changes, it will be possible to follow in situ the TOL evolution along the life span of composite structures.

The dispersion curves of an arbitrary composite material can be computed using several methods among which the semi-analytical ones are quite efficient. Methods like the transfer-matrix method (TMM), allowing to consider material properties of each ply and the interface conditions between the inner plies and the upper and lower free surfaces are suited to perform such computations [11, 12]. Using the TMM method, the phase velocities of the waves propagating in the virgin composite and in oxidized ones are being computed for TOL corresponding to various percentage of the thickness of the outer plies ranging from 0% (virgin composite) to 90% (outer plies almost fully oxidized).

Dispersion curves for the virgin composite material are for example presented in Figure 2. From this figure, it can be seen that in the chosen composite material, only two Lamb waves mode are coexisting in the frequency range below 400 kHz. The S_0 mode is corresponding to longitudinal waves, is propagating around 7 km/s and is almost non-dispersive in that frequency range. The A_0 mode, corresponding to flexural waves propagates much more slowly (around 1 km/s) and is much more dispersive.

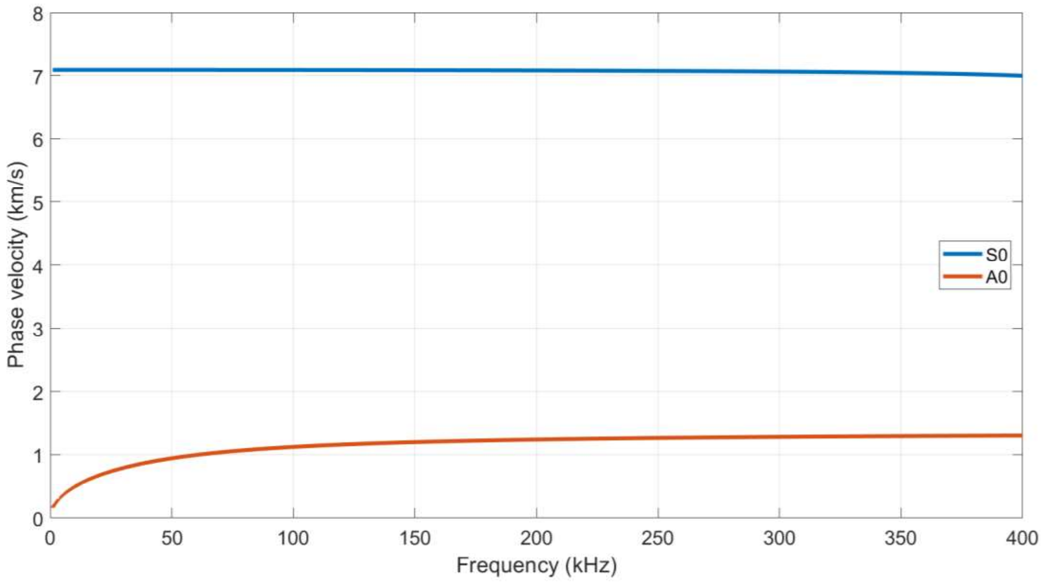

Figure 2: Dispersion curves for the virgin composite material

SENSITIVITY OF WAVES TO THE THICKNESS OF OXYDIZED LAYERS

LAMB WAVES MODES SENSITIVITY TO TOL

Now that the tools allowing to estimate waves velocities as a function of the TOL are available, it is possible to perform a sensitivity analysis in order to know which mode is the most sensitive to TOL and in which proportion. In order to achieve that goal, the variation of the phase velocities associated with each mode with a TOL varying as a percentage of the thickness of the first ply have been computed. For each frequency and for each mode, the reference phase velocity is considered to be the one corresponding to the virgin case.

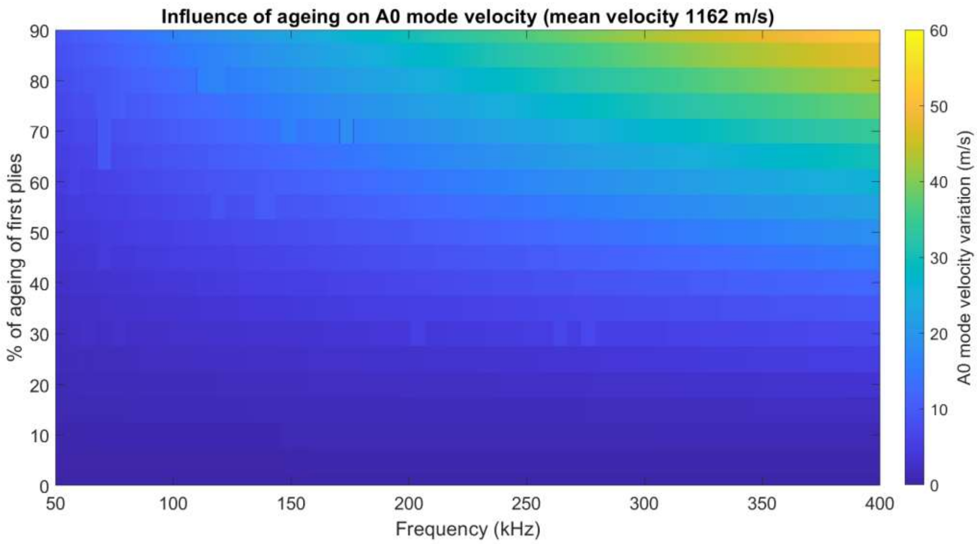

Figure 3: Variation of phase velocity for the A_0 mode as a function of TOL

The variation of the phase velocity for the A_0 mode as a function of TOL is presented in Figure 3. From this figure, it can be observed that the velocity variation is monotonic with respect to the TOL, as intuitively expected and also with the frequency. This monotonicity with frequency can be attributed to the dispersive behavior of A_0 which makes phase velocity very sensitive to the frequency. Consequently, the maximal velocity variation is obtained for large frequencies and large TOL and is around 60 m/s here which corresponds to a 5% variation. This not large, but can be expected to be measurable, as will be discussed later.

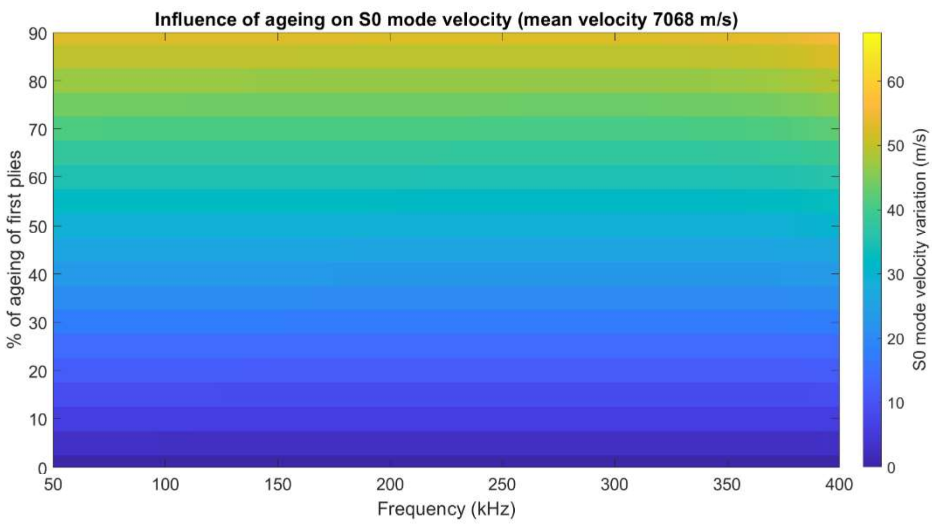

Figure 4: Variation of phase velocity for the S_0 mode as a function of the TOL

The variation of the phase velocity for the S_0 mode as a function of the TOL is presented in Figure 4. From this figure, it can be observed again that the velocity variation is monotonic with respect to the TOL, as intuitively expected. However, almost no variation is observed with the frequency. This can again be attributed to the non-dispersive behavior of S_0 which makes phase velocity unsensitive to the frequency. Consequently, the maximal velocity variation is obtained for every frequency and large

TOL and is around 60 m/s here which corresponds to a 0.8% variation. This is extremely small here and presumably hard to catch experimentally.

NUMERICAL SIGNALS FOR VIRGIN AND AGED CASES

The question arises now whether such phase velocities variations can be in practice measured using the experimental setup typically found in SHM. For SHM purposes, one typically sends 5 cycles tone bursts centered around a central frequency around 200 kHz using piezoelectric transducers separated by a distance chosen here as 12 cm. The piezoelectric transducers are here not modelled and the composite plate is supposed to be infinite to begin with. The resulting signals are computed and plotted for central frequencies equal to 150 kHz, 200 kHz, 250 kHz and 300 kHz as a function the TOL.

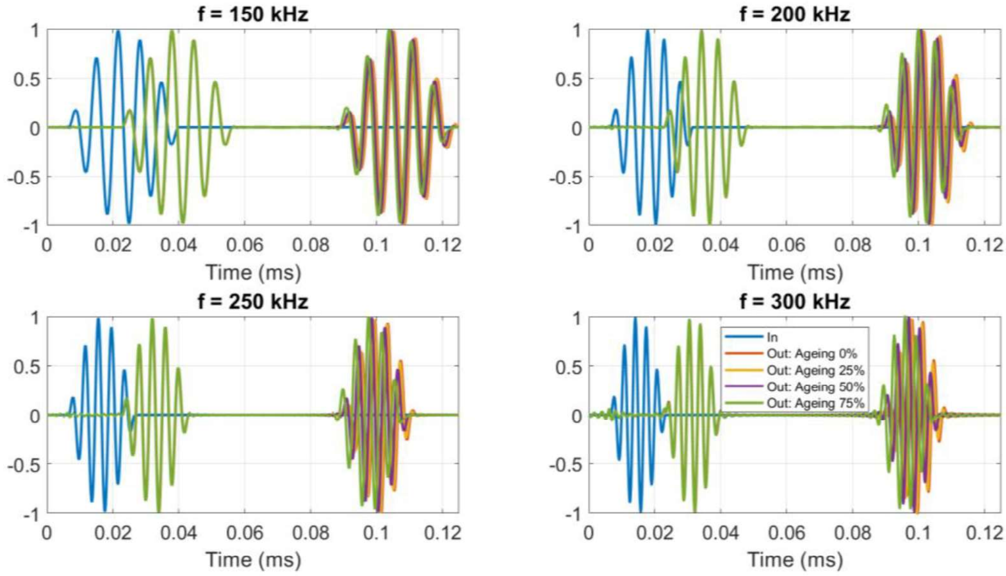

Figure 5: Typical SHM signals computed as a function of TOL for various center frequencies. The input signal is plotted in blue and the propagated ones in colors.

Typical SHM signals computed as a function of TOL for various center frequencies are plotted in Figure 5. The input signal is plotted in blue and the propagated ones in various colors depending on the TOL. As expected, for one input burst signal, two propagated modes are measured after 12 cm. The most rapid one corresponds to the S_0 mode and the influence of TOL can hardly be seen. The slower one is corresponding to the A_0 mode and is much more affected by TOL. Such differences in signals are significant and may surely be measured experimentally. The only issue is to be able to separate the A_0 mode from the S_0 mode and its reflections on boundaries. This can be achieved using for example specially tuned transducers or adequate signal processing tools and remains the objective of an upcoming experimental campaign.

CONCLUSION

Under the exposition of composite materials to temperature in air, oxidation leads to a non-negligible TOL which appears initially on the free surfaces of composite structures and increases within the composite thickness over exposure time. This oxidized layer has modified mechanical properties in comparison with the virgin composite materials. Being able to estimate *in situ* the TOL of composite structures under service is thus of great importance for safety and maintenance purposes.

Ultrasonic guided waves (UGW) propagating in composite structures are sensitive to the material properties of the composite material in which they propagate. UWG propagating in aged composites will have different velocities in comparison with UGW propagating in virgin composites. Taking benefits of this physical properties linking TOL thickness to UGW velocities, a strategy allowing to estimate *in situ* the TOL of composite materials is here proposed. As a first step towards its implementation, this strategy is here validated numerically using semi-analytical models allowing to predict UGW velocities in composites as a function of the number of plies, their orientation and material. This model allows to demonstrate that the A_0 mode at high frequency is the most sensitive to TOL thickness. As a next future step, the proposed approach will be used experimentally for the evaluation and tracking of the *in situ* growth of TOL. Such a tool can be deployed *in situ* and help to follow the health state of composite structures in practical applications.

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REFERENCES

[1] X. Colin, A. Mavel, C. Marais and J. Verdu, "Interaction between Cracking and Oxidation in Organic Matrix Composites," *Journal of Composite Materials,* vol. 39, p. 1371–1389, August 2005.

[2] J. P. M. Costa et X. Colin, «Finite element modelling of the oxidation gradients of epoxy-diamine matrices below and above their glass transition temperature,» *Polymer Degradation and Stability,* vol. 234, p. 111194, April 2025.

[3] K. Pochiraju and G. Tandon, "Modeling Thermo-Oxidative Layer Growth in High-Temperature Resins," *Journal of Engineering Materials and Technology-transactions of The Asme,* 2006.

[4] M. Mitra et S. Gopalakrishnan, «Guided wave based structural health monitoring: A review,» *Smart Materials and Structures,* vol. 25, p. 053001, 2016.

[5] K. Aoujdad, M. E.-C. El-Kettani, D. Leduc et P. Marechal, «Determination of ageing indicators on glass-fiber polyester composite skins using Lamb guided waves,» chez *Journal of Physics: Conference Series*, 2024.

[6] M. Ducousso, A. Dalodière et A. Baillard, «Evaluation of the thermal aging of aeronautical composite materials using Lamb waves,» *Ultrasonics,* vol. 94, p. 174–182, 2019.

[7] Y. Gélébart, H. Duflo et J. Duclos, «Air coupled Lamb waves evaluation of the long-term thermo-oxidative ageing of carbon-epoxy plates,» *NDT & E International,* vol. 40, p. 29–34, 2007.

[8] M. Huger, D. Fargeot et C. Gault, «Ultrasonic characterization of oxidation mechanisms in Nicalon/C/SiC composites,» *Journal of the American Ceramic Society,* vol. 77, p. 2554–2560, 1994.

[9] N. Korde et T. Kundu, «Material hardness and ageing measurement using guided ultrasonic waves,» *Ultrasonics,* vol. 53, p. 506–510, 2013.

[10] W. Li, C. Xu et Y. Cho, «Characterization of degradation progressive in composite laminates subjected to thermal fatigue and moisture diffusion by lamb waves,» *Sensors,* vol. 16, p. 260, 2016.

[11] S. Guo, M. Rébillat et N. Mechbal, «Dichotomy property of dispersion equation of guided waves propagating in anisotropic composite plates,» *Mechanical Systems and Signal Processing,* vol. 164, 2022.

[12] S. Guo, M. Rébillat et N. Mechbal, «Prediction of frequency and spatially dependent attenuation of guided waves propagating in mounted and unmounted A380 parts made up of anisotropic viscoelastic composite laminates,» *Structural Health Monitoring,* 2022.