



**HAL**  
open science

# Experimental identification of pore opening, growth and coalescence in a bonded assembly using X-ray $\mu$ -tomography

Marina Bunea, Malick Diakhate, Jérôme Adrien, Eric Maire, Claudiu Badulescu

## ► To cite this version:

Marina Bunea, Malick Diakhate, Jérôme Adrien, Eric Maire, Claudiu Badulescu. Experimental identification of pore opening, growth and coalescence in a bonded assembly using X-ray  $\mu$ -tomography. 25e Congrès Français de Mécanique, Aug 2022, Nantes, France. hal-04281686

**HAL Id: hal-04281686**

**<https://hal.science/hal-04281686v1>**

Submitted on 13 Nov 2023

**HAL** is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

# Experimental identification of pore opening, growth and coalescence in a bonded assembly using X-ray $\mu$ -tomography

M. BUNEA<sup>a</sup>, M. DIAKHATE<sup>b</sup>, J. ADRIEN<sup>c</sup>, E. MAIRE<sup>c</sup>, C. BADULESCU<sup>a</sup>

<sup>a</sup>ENSTA Bretagne, IRDL-UMR CNRS 6027, Brest, France

email : [marina.schitanu@ensta-bretagne.fr](mailto:marina.schitanu@ensta-bretagne.fr)

<sup>b</sup>Univ. de Bretagne Occidentale, IRDL-UMR CNRS 6027, Morlaix, France

<sup>c</sup>UMR CNRS 5510, Laboratoire MATEIS, Université Lyon, INSA Lyon, Villeurbanne, France

## Résumé:

*Le collage structural est de plus en plus utilisé dans différents secteurs industriels tels : i) le naval, ii) l'automobile ou encore iii) l'aéronautique [1]. Les adhésifs couramment utilisés dans le contexte d'assemblages structuraux sont des colles de type époxy bi-composants (résine et durcisseur). Ainsi, durant la phase de mélange résine – durcisseur, mais également lors de l'étape de polymérisation, on constate l'apparition des pores dans le joint. La taille et la répartition des pores sont fortement influencées par la nature des composants des adhésifs, leur viscosité, leur température, le temps de mélange etc. On peut constater expérimentalement [2], que cette population de porosités influence considérablement la résistance mécanique du joint et par conséquent conduit à la dégradation de l'adhésif. Naturellement, les changements de morphologie du joint pourraient être dus à des mécanismes (d'ouverture, de croissance, de coalescence des pores) se produisant durant le chargement mécanique du joint. Alors, l'objectif de cette étude porte sur la mise en place d'une stratégie robuste d'identification expérimentale de ces mécanismes d'endommagement en utilisant la  $\mu$ -tomographie aux rayons X. La stratégie proposée est construite tout d'abord à partir des données tomographiques synthétiques pour lesquelles l'apparition des différents mécanismes (ouverture, croissance, coalescence) est maîtrisée. L'effet de la voxelisation des porosités sur l'identification des ces mécanismes est étudié et un critère de coalescence est proposé. La deuxième étape a pour but la validation de la stratégie proposée, au moyen de données semi-synthétiques, obtenues à partir des résultats d'un modèle par éléments finis. Finalement, la stratégie est appliquée sur deux cas concrets : un essai de traction monotone sur un échantillon massif et un essai de traction sur un joint collé, sollicité dans la direction hors-plan, en s'appuyant sur un échantillon de type mini-scarf [3].*

## Abstract:

*Structural bonding is increasingly used in various industrial sectors such as i) naval, ii) automotive and iii) aeronautics [1]. The adhesives traditionally used in the context of structural assemblies are two-component epoxy adhesive type (resin and hardener). Thus, during the resin-hardener mixing phase, but also during the polymerization step, it can be observed the appearance of the pores in the*

*joint. The size and distribution of the pores are greatly influenced by the nature of adhesive components, their viscosity, their temperature, mixing time, etc. It can be observed experimentally [2] that this population of porosity considerably influences the mechanical strength of the seal and consequently leads to the degradation of the adhesive. Naturally, the changes in the morphology of the joint could be due to mechanisms (opening, growth, coalescence of the pores), occurring during the mechanical loading of the joint. So, the objective of this study concerns the establishment of a robust strategy for the experimental identification of these damage mechanisms, using X-ray  $\mu$ -tomography. The proposed strategy is built first from synthetic tomographic data for which the appearance of the different mechanisms (opening, growth, coalescence) is mastered. The effect of voxelization of porosities on the identification of these mechanisms is studied and a coalescence criterion is proposed. The second step aims to validate the proposed strategy, using semi-synthetic data obtained from the results of a finite element model. Finally, the strategy is applied to two concrete cases: a monotonic tensile test on a bulk sample and a tensile test on a bonded joint based on a mini-scarf type sample stressed in the out-of-plane direction [3].*

**Keywords: epoxy adhesive, porosity, growth, coalescence, X-ray  $\mu$ -tomography**

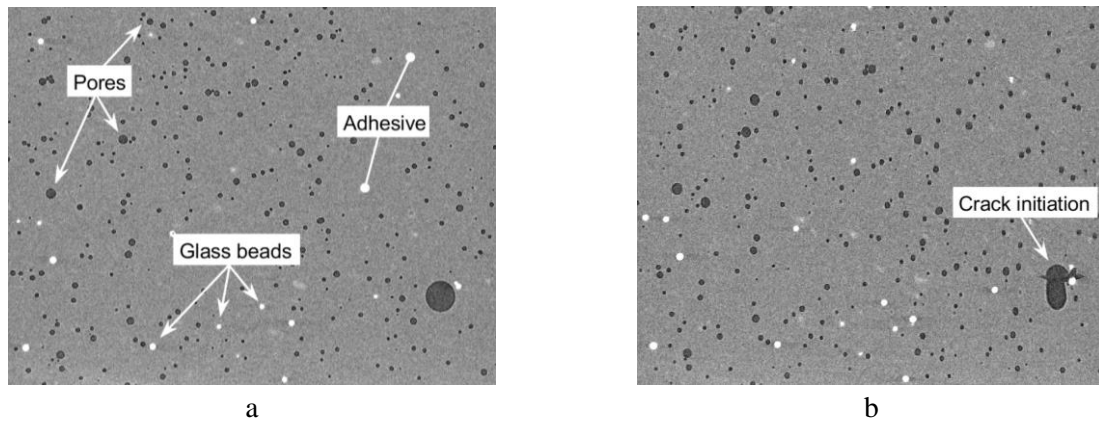
## 1 Introduction

The bonded joints are used in all domains of engineering industries such as automotive, naval, aerospace, structural, and so on. The mechanical performance of adhesive joints depends on the quality of the substrate-adhesive interface and adhesive defects such as poor polymerized regions, impurities, or pores, which are directly linked to mixing time, velocity and technique, components' temperature and viscosity, air bubbles occurrence into prepolymer blend, additives addition, etc. The most common defects of epoxy adhesives are the pores [2,3], which actually exhibit stress concentrations. These stress concentrations exhibit the points of crack initiation and its propagation through the porous medium of the adhesive layer as a function of their size, number, and locations. Thus, in order to investigate the damage mechanisms of the adhesive joints, it is applied X-ray computed tomography inspection technique (CT) at certain steps of mechanical loadings, which permits to make an internal analysis of the bonded joints by identifying the presence of the pores, crack occurrence and its propagation through the adhesive layer. Generally, the damage mechanisms depend on the strain distribution into the adhesive structure, which is influenced by its porosity during the mechanical loading. So, the damage mechanisms of a porous volume usually are described by the opening, growth, and coalescence of the pores. The coalescence of the pores can be determined based on their shape as Dumont et al. [3] investigated the fusion of the pores based on their sphericity ( $\Psi$ ), which in the case of perfect sphere equals 1. It was found that for sphericity  $\Psi < 1$ , the pores exhibit the coalescence phenomena.

This research aims to develop a robust strategy for the identification of damage mechanisms produced by porosity in the case of epoxy adhesive joints, using X-ray  $\mu$ -tomography. First, the proposed strategy is built from synthetic tomographic to investigate the coalescence of pores and the appearance of new pores. Also, in this study, it was analyzed the voxelization effect of porosities on the investigation of these mechanisms and it is proposed a coalescence criterion to identify the true fusion of the pores. Secondly, the proposed strategy was validated by a semi-synthetic data obtained from the results of a finite element model and experimentally: a monotonic tensile test on a bulk sample and a tensile test on a bonded joint.

## 2 Identification of the new pores

The identification of pore opening and growth in a bonded assembly is achieved by making the correspondence between two tomographic real volumes, one of them showing the initial volume (before a mechanical loading step) and the other one exhibiting the deformed volume. Two tomographic images of initial and deformed configurations of a bulk adhesive sample subjected to tensile loading are shown as an example in figure 1.



**Fig. 1.** Tomographic images of a bulk adhesive sample subjected to tensile loading: a) initial volume configuration and b) deformed volume configuration.

In order to find if the results of the developed method are correct, it was generated two synthetic adhesive volumes with close characteristics to the real tomographic volumes, where newly appeared pores of deformed volume are known. So, it was generated initial volume configuration and final volume configuration (deformed volume) in MATLAB software. A theoretical volume has been generated: a unit right rectangular prism as adhesive matrix and a preestablished arbitrary number of rigid spheres as pores, taking into account the fact that the shape of the pores tends to be more spherical. The generated spheres have various radii, they are unfused and distributed in a uniform random manner.

In order to perform the correspondence between the configurations, it was developed the method of a single pore correspondence. In order to find the correspondence of pores by this method, it is necessary to initially select manually a certain pore  $P'_i$  in the final configuration and its corresponding pore  $P_i$  in the initial configuration. The displacement of the selected pore  $P'_i$  is calculated. Then the developed algorithm searches the nearest neighboring pore  $P'_{i+1}$  of the selected manually pore  $P'_i$  in the final volume configuration. Its corresponding pore  $P_{i+1}$  in the initial volume configuration is found based on the calculated displacement for the selected manually pores, after which it follows the calculation of their displacement. Further, the algorithm searches the nearest neighboring pore  $P'_{i+2}$  of the previously found pore  $P'_{i+1}$  in the final volume configuration and it follows the same corresponding steps and so on until the correspondence of the last pore is performed.

The results of the correspondence method are the displacements in the X, Y, respectively Z-axis directions and the labels of each found corresponding pore. The identification of pores coalescence and new pores are made based on their labels. The strategies for measuring the volume displacement field including the results of this method will be presented in the forthcoming work.

The used condition to identify the new pores was the labelling of all unfound corresponding pores in initial volume configuration by logical 1.

### 3 Identification of fused pores

It was found that the problem of the closest pores appears during the voxelization process, which leads to their fusion due to the voxel shape. Considering that the shape of the pores is spherical, it was found that if one of these length sizes is much larger than the other two sizes, it results that this pore represents the fusion of two or more pores.

In order to find the true pore fusion, it was applied the ellipsoid interpolation. Each fused pore was separated by the watershed transform function of MATLAB. Further, the set of the surface points  $x$ ,  $y$ , and  $z$  for each separated pore was identified by drawing its contour on each XY plane of volume in the Z-axis direction. The fusion of the pores was determined on the calculated roots of the *characteristic polynomial equation* [4] described by

$$f(\lambda) = \det(\lambda S_1 + S_2), \quad (1)$$

where  $\lambda$  exhibits scalar constants as roots of the *characteristic equation*  $\det(\lambda S_1 + S_2) = 0$ ,  $S_1$  and  $S_2$  are the ellipsoids defined by  $X^T S_1 X = 0$  and  $X^T S_2 X = 0$ ,  $X = [x \ y \ z \ 1]$  are the homogeneous coordinates of ellipsoids. Algebraically, an ellipsoid can be represented polynomially [5] as follows

$$Ax^2 + By^2 + Cz^2 + Dxy + Eyz + Fxz + Gx + Hy + Iz + J = 0, \quad (2)$$

where  $A, B, C, D, E, F, G, H, I$  and  $J$  are the parameters describing the ellipsoid, resulting

$$S = \frac{1}{2} \begin{pmatrix} 2A & D & F & G \\ D & 2B & E & H \\ F & E & 2C & I \\ G & H & I & 2J \end{pmatrix}. \quad (3)$$

Wang et al. [4] found that the characteristic polynomial equation  $f(\lambda) = 0$  exhibits always at least two negative roots. If the other ones are two distinct roots, it means that the ellipsoids are separated, but if the equation presents a double root, it indicates that the ellipsoids touch each other.

In the case of generated synthetic volume, it was found that four pores fused due to the occurrence of the new pores in the final configuration.

### 5 Validation by ABAQUS simulation

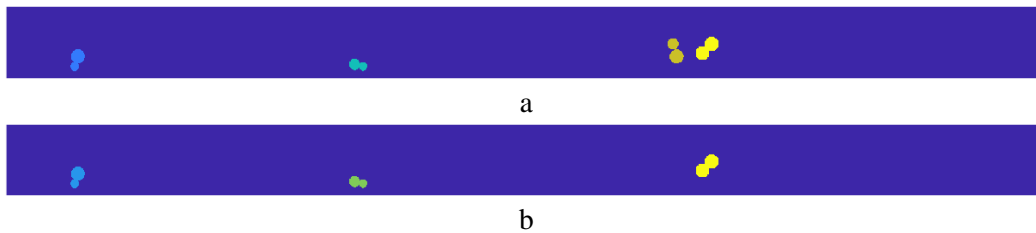
In order to validate the obtained results, it was created a volume with rigid pores by extended finite element method (xFEM) calculation in ABAQUS. To generate the volume by xFEM calculation in ABAQUS, it was selected a transversal image of the tomographic volume of a mini-scarf type sample. The radii and centers of the pores were found in MATLAB by function “*imfindcircles*”. Based on the found radii and centers, it was generated a 2D volume in ABAQUS. The dimensions of generated volume were  $6 \times 0.4$  mm. The properties of the initial volume configuration are given in table 1.

**Table 1.** Elastic parameters of initial volume configurations' components.

Parameter	Epoxy adhesive matrix	Rigid pores
Modulus of elasticity $E$ [MPa]	2,000	50,000
Poisson's ratio $\nu$ [-]	0.4	0.18

The parameters of the rigid pores were given as in the case of glass beads, considering that the pores are rigid. The initial volume configuration was subjected to tensile loading with a displacement of 0.1 mm. In order to regenerate the initial configuration of ABAQUS volume in MATLAB, it was applied the function “*alphaShape*”, to create the bounding surface based on the  $x$  and  $y$  coordinates of a set of 2D points. Further, it was defined as a 2D grid with dimensions equal to those of the model built in ABAQUS and it was determined by the function “*inShape*”, if each point of the 2D grid belongs to alpha shape, so as it was obtained the volume, where the value of logical 1 indicates the matrix and value of logical 0 indicates the presence of pores. In order to define the configuration of the volume, it was determined the complement of the last achieved volume. Similarly, it was defined as the deformed configuration after tensile loading.

The fused pores of volume configurations created by xFEM calculation were found by using the method presented above (fig. 2).



**Fig. 2.** Identified fused pores of initial volume configuration before ellipsoid interpolation (a) and after ellipsoid interpolation (b).

The results of ellipsoid interpolation presented four fused pores in initial volume configuration (fig. 2.a) of which three pores are true fused and one of them occurred due to the voxelization process (fig. 2.b). In the case of final volume configuration, it resulted in the fused pores which were truly fused in the initial configuration and the effects of voxelization disappear due to the increase in distance between the pores during tensile loading. The obtained results prove that the problem of the closest pores appears during the voxelization process.

## 6 Experimental validations

### 6.1 Monotonic tensile test

Also, in order to prove the effectiveness of the proposed strategies, the developed methods were applied to the real adhesive volumes obtained before and after a certain step of tensile loading performed inside of *in situ* X-ray tomographic equipment. It has been used a bulk sample made of epoxy adhesive Huntsman<sup>TM</sup> Araldite 420 A/B. The CT investigation was made at several steps of applied tensile load till specimen failure (35.3 N). The presented real volumes were generated in MATLAB software based on X-ray CT image stacks following several processing steps: median filtering, segmentation, and volumes creation [3]. The size of the resulted adhesive volume was  $1300 \times 1295 \times 125$  voxels. So, in order to validate experimentally the results of the proposed strategies, it was analyzed the tomographic volumes: V1 - initial volume configuration (1146 pores) at tensile loading of 2.58 N and V5 - final volume configuration (1046 pores) at tensile loading of 17.1 N.

It was found that 113 pores (9.86 % of the pores of the volume) fused in the case of the initial configuration and 88 pores (8.41 % of the pores of the volume) fused in the case of the final configuration, by applying the strategies of identification of pore coalescence. Using ellipsoid interpolation, resulted in 16 truly fused pores (14.16 % of initially fused pores) for initial configuration

and 17 truly fused pores (19.32 % of initially fused pores) for final configuration. So, the difference between the number of the coalesced pores before and after ellipsoid interpolation appeared due to the effects of the voxelization process. Also, the obtained results of the correspondence method showed that almost 3 new pores appeared in the final volume configuration.

## 6.2 Adhesive joints

For the second case of experimental validation, the two real volumes of an adhesive joint obtained at tensile loading of 200 N and 600 N performed inside of *in situ* X-ray tomographic equipment were used. These adhesive volumes were generated by Dumont et al. [6]. The used specimen presented two Al substrates bonded by epoxy adhesive Huntsman<sup>TM</sup> Araldite 420 A/B. The tensile tests were performed perpendicular to the adhesive layer (in the out-of-plane direction). The volumes were also generated in MATLAB software and their size was  $941 \times 944 \times 65$  voxels. The volume resulting in tensile loading of 200 N (V1) was used as the initial configuration (2463 pores) and the volume achieved at 600 N tensile loading (V3) was used as the final configuration (2444 pores).

It was determined that the initial volume configuration contained 461 fused pores (18.72 % of the pores of the volume) and the final volume configuration included 468 fused pores (19.15 % of the pores of the volume). By ellipsoid interpolation, it was found that 234 truly fused pores (50.76 % of initially fused pores) for initial configuration and 238 truly fused pores (50.85 % of initially fused pores) for final configuration. The correspondence results showed that almost 21 new pores occurred in the final volume configuration.

## 7 Conclusions

In order to achieve the proposed objectives for this research, it was developed a correspondence method (a single pore correspondence) to identify the newly occurred pores after a mechanical loading, a method to detect the fused pores, and coalescence criteria to find the truly fused pores. To show the effectiveness of these strategies, it was generated a synthetic adhesive volume with characteristics close to those of real volumes, whose newly occurred pores were known. The results of developed strategies were validated by the results of a finite element model calculation and experimentally using a monotonic tensile test on a bulk sample and a tensile test on an adhesive joint, based on a mini-scarf type sample. It was found that by ellipsoid interpolation, it can be seen if the pores were true fused, or their fusion resulted due to voxelization effects.

## Acknowledgment

This work was supported by project "CARactérisation et Modélisation des mécanismes d'ENdommagement dans des assemblages collés à l'aide d'un essai mini-Scarf (CARMEN)" (in English: "Characterization and modeling of damage mechanisms in bonded assemblies using a mini-scarf test (CARMEN)", Contract no. SAD19004/00046003/13.12.2019 funded by Region Bretagne (75 %) and ENSTA Bretagne (25 %).

## References

- [1] J. Destouesse, M. Diakhate, C. Badulescu, D. Thévenet, M. Stackler, W. Albouy, N. Carrere, Cluster analysis of acoustic emission data to investigate the damage evolution in modified scarf joint under bi-axial loading, *The Journal of Adhesion*. 96 (2020) 969–987.
- [2] V. Dumont, C. Badulescu, G. Stamoulis, J. Adrien, E. Maire, A. Lefèvre, D. Thévenet, On the influence of mechanical loadings on the porosities of structural epoxy adhesives joints by means of in-situ X-ray microtomography, *International Journal of Adhesion and Adhesives*. 99 (2020) 102568.
- [3] V. Dumont, C. Badulescu, G. Stamoulis, J. Adrien, E. Maire, A. Lefèvre, D. Thévenet, On the effect of the curing cycle on the creation of pores in structural adhesive joints by means of X-ray microtomography, *The Journal of Adhesion*. 97 (2021) 1073–1106.
- [4] W. Wang, J. Wang, M.-S. Kim, An algebraic condition for the separation of two ellipsoids, *Computer Aided Geometric Design*. 18 (2001) 531–539.
- [5] S. Alfano, M.L. Greer, Determining If Two Solid Ellipsoids Intersect, *Journal of Guidance, Control, and Dynamics*. 26 (2003) 106–110.
- [6] V. Dumont, C. Badulescu, J. Adrien, N. Carrere, D. Thévenet, E. Maire, Experimental investigation of porosities evolution in a bonded assembly by means of X-ray tomography, *The Journal of Adhesion*. 97 (2021) 528–552.