Modélisation multi-échelle des composites en polyamide 6 renforcé de fibres de verre recyclés intégrant la viscoélasticité, la viscoplasticité et l'endommagement anisotrope

Multiscale modeling of recycled glass fiber reinforced polyamide 6 composites incorporating viscoelasticity, viscoplasticity, and anisotropic damage

Saif Eddine Sekkal[1,2], Fodil Meraghni[1], George Chatzigeorgiou[1] et Francis Praud[1]

[1] Arts et Métiers Institute of Technology, CNRS, Université de Lorraine, LEM3-UMR7239
4 rue Augustin Fresnel, Metz, 57078, France
e-mail : saif_eddine.sekkal@ensam.eu, fodil.meraghni@ensam.eu, georges.chatzigeorgiou@ensam.eu et francis.praud@ensam.eu
[2] Centre Technique des Industries Mécaniques (CETIM)
21, rue de Chemnitz, BP 2278 - 68068 Mulhouse Cedex

Résumé

Cette étude traite de la réponse mécanique des composites en polyamide 6 renforcé de fibres de verre recyclées mécaniquement, en mettant l'accent sur leur comportement non linéaire et dépendant du temps sous des conditions de chargement complexe. Le processus de recyclage induit des caractéristiques microstructurales uniques, notamment une certaine anisotropie marquée et une variabilité dans l'orientation des fibres et la distribution des mèches ou paquets de fibres. L'approache proposée combine des essais mécaniques en trajets complexes et une modélisation multi-échelle afin d'établir un lien entre les propriétés microstructurales et la réponse mécanique macroscopique des composites recyclés. Le modèle multi-échelle conçu intègre le comportement viscoélastique et viscoplastique de la matrice polymère, associé à une modélisation de l'endommagement anisotrope dans les mèches. Afin de prendre en compte la variabilité expérimentale, plusieurs Volumes Élémentaires Représentatifs (VER) ont été générés via l'algorithme RSA (Random Sequential Adsorption), permettant d'étudier les effets de la position et de l'orientation des mèches sur la réponse mécanique. Les résultats obtenus démontrent que le modèle prédit efficacement les réponses sous charge monotone ainsi que les tendances générales des comportements en chargement cyclique et en fluage. L'approche multi-échelle a également été utilisée pour alimenter un schéma avancé de réseau neuronal permettant des prédire les réponses de structures en quasi-temps réel.

Abstract

This study investigates the mechanical response of mechanically recycled glass fiber reinforced polyamide 6 composites, emphasizing their nonlinear and time-dependent behavior under complex loading conditions. The recycling process induces unique microstructural features, including significant anisotropy and variability in fiber orientation and strand distribution. The proposed research combines detailed experimental testing and advanced numerical modeling to bridge the gap between the microstructural properties and macroscopic mechanical response of recycled composites. The designed multiscale model incorporates the viscoelastic and viscoplastic behavior of the polymer matrix, coupled with anisotropic damage modeling for the strands. To account for experimental variability, multiple Representative Volume Elements (RVEs) were generated using the RSA algorithm (Random Sequential Adsorption), enabling the study of strand position and orientation effects on the mechanical response. The results demonstrate that the model effectively predicts monotonic loading responses and general trends in cyclic and creep behaviors. The multiscale approach has also been used to generate the input data for an advanced neural network scheme that allows near real time structural computations.

Mots Clés : Composites recyclés mécaniquement, Modélisation non linéaire multi-échelle, Génération de microstructure, Endommagement anisotrope, Réseaux de neurones artificiels.

Keywords : Mechanically recycled composites, Multiscale nonlinear modeling, Microstructure generation, Anisotropic damage, Artificial Neural Network.

1. Introduction

Continuous fiber reinforced thermoplastic composites are increasingly valued in automotive, aerospace, and renewable energy applications for their superior mechanical properties, cost efficiency, and excellent recyclability. However, with growing environmental regulations urging sustainable waste management, the development of effective recycling methods and understanding the mechanical behavior of recycled composites have become critical.

According to the material type and the intended product application, the composite recycling technologies are classified into mechanical, thermal, and chemical methods [2]. This study focuses on the mechanical response of mechanically recycled glass fiber reinforced polyamide 6 composites. The goal is to simulate and predict their nonlinear and time-dependent behavior under complex loading conditions. An important feature of the mechanical recycling is that induces unique microstructural features, such as significant anisotropy and variability in fiber orientation and strand distribution [6]. These characteristics are critical to understanding the material's performance.

In the current study, a detailed experimental testing has been performed, aiming at assisting the proposed advanced multiscale model to bridge the gap between the microstructural properties and macroscopic mechanical response of recycled composites. Cyclic loading-unloading tests, creep recovery experiments, and controlled relative humidity conditions were used to assess the material's damping properties, energy absorption capacities, and inelastic behaviors. A novel multiscale modeling framework was developed to predict the composite's nonlinear response. This approach incorporates the viscoelastic and viscoplastic behavior of the polymer matrix, coupled with anisotropic damage modeling for the strands [3, 4]. To reduce the computational cost and provide a more useful simulation scheme for practical applications, a data-driven type approach has been also proposed in the recently developed framework of Multiscale Thermodynamics-Informed Neural Networks [1].

2. Material description and experimental protocol

The analyzed recycled composites are manufactured and supplied by Cetim, a Research and Technology Organization (RTO) that contributed to the development of mechanical recycling of polymer composites. The production process is briefly summarized in Figure 1. The current study focuses on recycled composites produced from uniformly cut chips sized $20 \times 20 \times 1$ mm^3. The original virgin material is a PA-GF woven composite with a fiber volume fraction of 47%, referenced as Tepex®Dynalite 102-RG600(x)/PA-GF/47%, and supplied by Bond-Laminates GmbH (Am Patbergschen Dorn 11-D-59929 Brilon, Germany).

Optical microscopy lead to the observation that the composite microstructure should be analyzed at both the microscopic and mesoscopic scales. At mesoscopic scale, the fibers form a wave pattern with mostly in-plane orientation, while at the microscopic level, the morphology of the microstructure within the strands is more homogeneous, with multiple fibers having a similar orientation. Moreover, X-ray tomography investigation reveals that the woven structure is maintained at the plate center across its thickness. Notably, in several occasions, the woven structure deviates from the initial yarn orientations, which is probably attributed to the material flow during thermocompression. In addition, the upper and lower surfaces show fewer instances of woven structure, while multiple unidirectional fiber bundles are apparent.

The necessary experiments for the multiscale model calibration and validation include uniaxial tests performed on three types of specimens, having the reference direction oriented at 0° , 45° and 90°. Specimens were cut off from 5 mm thick plates using a water jet hyperbaric machining process. Consistency in testing conditions is maintained by conditioning all specimens at 50% of relative humidity before being tested at room temperature (20±1°C). Monotonic loading, cyclic loading and unloading and creep and recovery tests have been conducted to assess the validity of the proposed multiscale strategy.

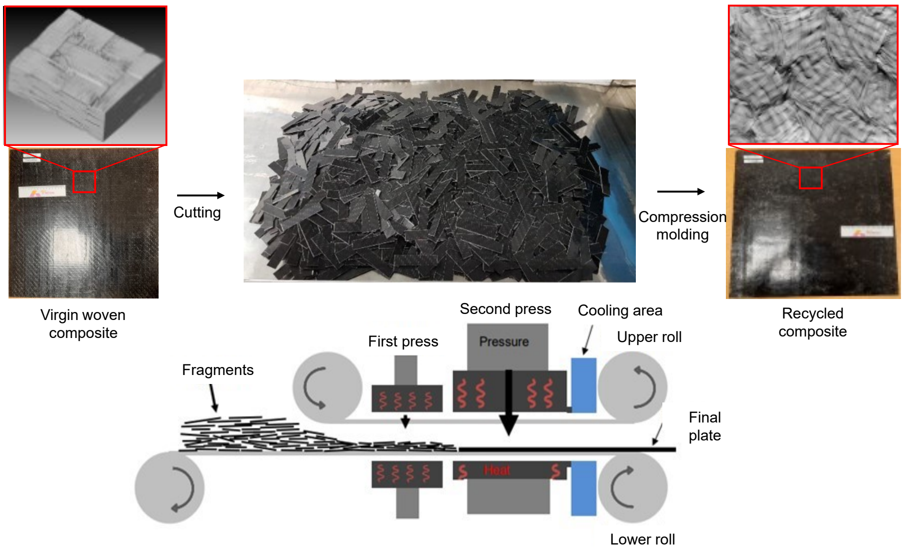

Fig. 1. – Schematic of the Thermosaïc® production line showing the processing steps for mechanical recycling of thermoplastic composites [6].

3. Multiscale modeling strategy

To account for experimental variability, multiple Representative Volume Elements (RVEs) were generated, enabling the study of strand position and orientation effects on the mechanical response. A key challenge in Recycled ThermoPlastic Composite RVE generation is achieving the high strand volume fraction necessary to accurately represent their real microstructures. To attain this value, the RVE is constructed, using the Random Sequential Adsorption (RSA) approach [8], in multiple layers rather than by placing inclusions directly in the 3-D RVE. Strands are sequentially placed in each layer of the RVE according to a user-defined Orientation Distribution Function (ODF) and this process is repeated until the required strand ratio is reached. The produced RVEs are periodic by construction. This strategy has been implemented in Matlab, using the polyshape and intersect functions to detect intersections between the generated strands.

The constitutive model for the thermoplastic PA6 matrix considers three nonlinear mechanisms, namely viscoelasticity, viscoplasticity and ductile damage [3]. On the other hand, the strands are regarded as equivalent to a unidirectional composite. For those, a hybrid micromechanical-phenomenological model accounting for discrete damage and inelastic strains is chosen [4].

Figure 2 provides a comprehensive depiction of the RVE generation, the constitutive law of the individual phases and certain key outcomes. The analysis of the monotonic and cyclic loading-unloading tests reveals a significant stiffness discrepancy between the various specimen orientations. with the 0° exhibiting the highest stiffness, the 90° the lowest, and the 45° specimens showing intermediate stiffness. Moreover, the 90° orientation shows increased energy dissipation per cycle, with notable hysteresis loops. With regard to the creep and strain recovery tests, the 90° oriented specimens exhibit more pronounced creep response than those at 0°, with significant strain accumulation and wider hysteresis loops, suggesting greater energy dissipation. The 45° oriented specimens demonstrate consistent results for all specimens, with creep behavior similar to the 90° orientation. In all types of tests, the proposed multiscale model is capable of describing both qualitatively and quntitatively the mechanical response. The average curves obtained from the simulations of the multiply generated RVE are within the range observed by the experimental variability [7].

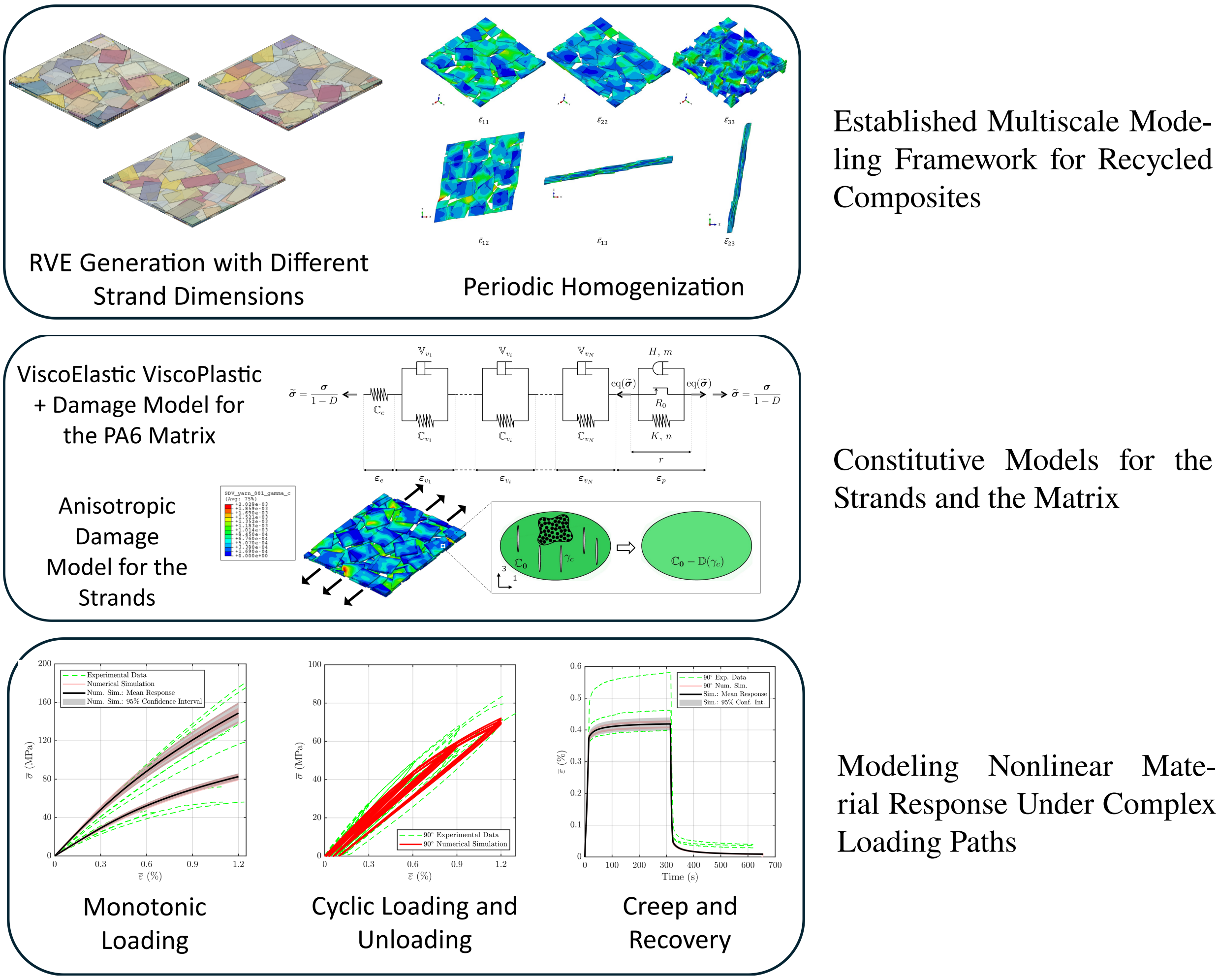

Fig. 2. – Framework for nonlinear multiscale modeling of recycled composites under complex loading paths.

4. Multiscale Thermodynamics-Informed Neural Networks (MuTINN)

Unlike Recurrent Neural Networks, which rely on previous inputs to predict future behavior, the Multiscale Thermodynamics-Informed Neural Networks (MuTINN) model [1, 5] eliminates this dependency by introducing a phenomenological framework that accurately describes the inelastic behavior of RVEs. To achieve this goal, this new approach requires additional variables to effectively account for the history-dependent nature of nonlinear material behavior. In classical homogenization theories, the origin of these variables lies on the microscale and they are traditionally called Internal State Variables (ISV). Including the entire ISVs in a neural network is impractical due to significant computational demands during model training due to the larger input dimensions. Thus, the MuTINN model introduces specialized variables called ”quantities of interest” (QOI), which are defined at the macroscopic level. They are derived by averaging specific ISVs across the composite components.

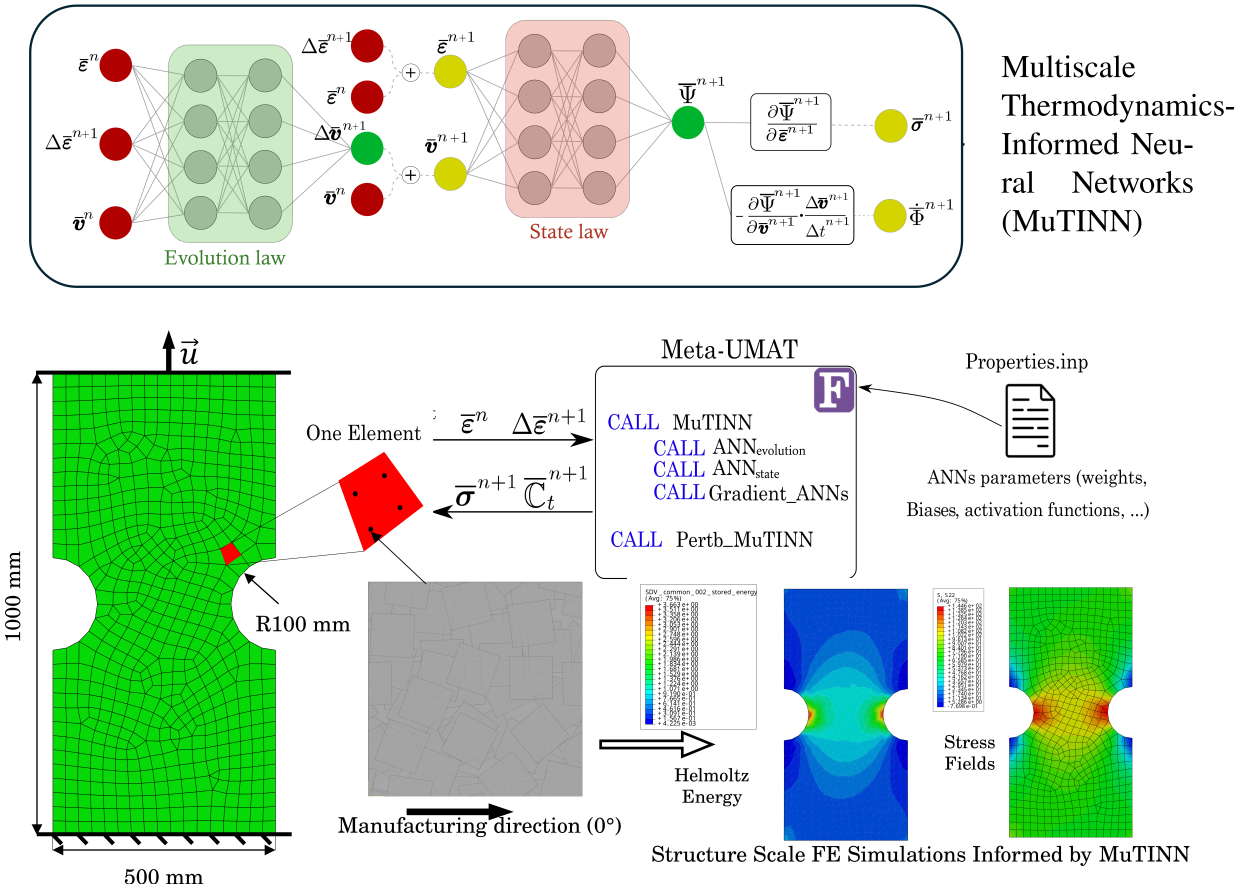

Fig. 3. – General structure of MuTINN and implementation in Finite Element analyses.

The architecture of MuTINN features two interconnected artificial neural networks or ANNs (Figure 3), each serving a distinct function. The first ANN defines the evolution law, processing inputs such as the current macroscopic strain components, the current quantities of interest and the macroscopic strain increment and calculates the corresponding increment of QOI. The second ANN, known as the state law model, considers as input the updated macroscopic strain and the revised quantities of interest, and exports as output the Helmholtz free energy. Automatic differentiation with respect to the inputs allows to calculate the macroscopic stress components and the produced dissipation. To ensure thermodynamic consistency, the loss function of the combination of the two ANNs includes a penalty term ensuring the positivity of the dissipation.

The dataset (3300 loading paths) used for training, evaluating, and testing the data-driven model is derived from finite element computations performed in a multiscale full-field scheme on the recycled composite RVE. The unit cell is subjected to random in-plane loading in the longitudinal and transverse directions, as well as simultaneous in-plane shear. To evaluate the data-driven model's performance, the latter is first tested in the online phase by predicting mechanical responses to individual, random strain increments. In the next step, the MuTINN results are evaluated for entire complex loading paths. In all studied cases an excellent match with the full-field results has been observed. The integration of MuTINN into a finite element (FE) code is achieved through a Meta-UMAT implemented in Fortran, including a special computational step for obtaining the necessary tangent modulus operator. Figure 3 demonstrates a numerical study on a double-notched structure made of Recycled ThermoPlastic Composite using the developed Meta-UMAT.

5. Conclusions

The proposed multiscale model successfully captures key aspects of the material's behavior, including high dissipation levels observed in hysteresis loops and the anisotropic response attributed to recycling-induced fiber orientation. The results demonstrate that while the model effectively predicts monotonic loading responses and general trends in cyclic and creep behaviors, challenges remain in fully capturing the extent of viscous effects and damage accumulation in certain orientations. Experimental observations revealed variability in mechanical performance, with some specimens showing strain accumulation and reduced recovery during creep recovery tests, underscoring the influence of the complex microstructure.

Moreover, the MuTINN framework demonstrates strong predictive accuracy across various loading conditions with siginficantly reduced computational cost, validated through comparisons with FE periodic homogenization and experimental data. In addition, at the structural level, MuTINN has been efficiently applied for FEA analysis of complex geometries.

Références

[1] Mohammed El Fallaki Idrissi, Francis Praud, Fodil Meraghni, Francisco Chinesta, and George Chatzigeorgiou. Multiscale Thermodynamics-Informed Neural Networks (MuTINN) towards fast and frugal inelastic computation of woven composite structures. *Journal of the Mechanics and Physics of Solids*, 186 :105604, 2024.

[2] Andrey E. Krauklis, Christian W. Karl, Abedin I. Gagani, and Jens K. Jørgensen. Composite material recycling technology—state-of-the-art and sustainable development for the 2020s. *Journal of Composites Science*, 5(1), 2021.

[3] Francis Praud, George Chatzigeorgiou, Jérôme Bikard, and Fodil Meraghni. Phenomenological multi-mechanisms constitutive modelling for thermoplastic polymers, implicit implementation and experimental validation. *Mechanics of Materials*, 114 :9–29, 2017.

[4] Francis Praud, George Chatzigeorgiou, Yves Chemisky, and Fodil Meraghni. Hybrid micromechanical-phenomenological modelling of anisotropic damage and anelasticity induced by micro-cracks in unidirectional composites. *Composite Structures*, 182 :223–236, 2017.

[5] Saïf Eddine Sekkal. *Multiscale modeling of the overall behavior of recycled thermoplastic composites : microstructural investigation and generation, and data-driven homogenization.* PhD thesis, Arts et Métiers Sciences et Technologies, Metz, France, 2025.

[6] Saïf Eddine Sekkal, Fodil Meraghni, George Chatzigeorgiou, Laurent Peltier, and Nelly Durand. Experimental and multi-scale investigation of the mechanical behavior of mechanically recycled glass fiber reinforced thermoplastic composites. *Composites Part B : Engineering*, 264 :110925, 2023.

[7] Saïf Eddine Sekkal, Fodil Meraghni, George Chatzigeorgiou, Francis Praud, and Nelly Durand. Multiscale modeling of mechanically recycled glass fiber reinforced polyamide 6 composites accounting for viscoelasticity, viscoplasticity, and anisotropic damage. *Composite Structures*, page 119016, 2025.

[8] Benjamin Widom. Random sequential addition of hard spheres to a volume. *The Journal of Chemical Physics*, 44(10) :3888–3894, 1966.