

DEVELOPMENT AND MODELING OF INTERFACIALLY ENGINEERED COMPOSITES WITH DESIGNED FAILURE

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ABSTRACT

There is a growing demand to increase the reliability of polymer composite materials by modifying their failure behavior. In this study, we developed and investigated a method that can be used to influence the damage processes and failure behaviour of composites by locally applying polycaprolactone (PCL) interlayer material. We manufactured locally interfacially engineered UD carbon/epoxy composite plates and conducted short-beam shear tests to investigate the effects related to the application and the geometry of PCL on the interlaminar shear strength, the damage processes and the final failure of the composite in case of interlaminar loading. Furthermore, we carried out finite element modeling using the cohesive zone method (CZM) to analyze the influence of PCL interlayer material on the initiation and propagation of damage. The results show that the application of PCL is suitable for modifying the damage and failure processes of UD carbon/epoxy composites when applied to interlaminar loading.

1 INTRODUCTION

Nowadays, polymer composites are gaining popularity, mainly due to their excellent mechanical properties and low density. With the widespread use of high-performance composite structures, they are emerging in structural components subjected to significant loads. However, in safety-critical applications, their failure process might be disadvantageous. To ensure the further spread of composites in structural applications, their damage and failure processes need to be influenced and controlled [1].

The failure of composites is a complex process. As a result of their complex microstructure and their manufacturing processes, polymer composites contain a wide range of flaws from different sources and in varying sizes. The failure process consists of several concurrent damage processes leading to the final failure of a composite part. With the proper design of the composite system, this failure behavior can be turned to our advantage, contributing to increased reliability of polymer composite materials [2].

Several methods can be used to influence and control the damage processes and failure behavior of polymer composites. One concept is to predefine the location of damage initiation and failure, which can be achieved by introducing artificial defects [3]. This method may simplify structural health monitoring and improve the reparability of the composite part. Another way is to influence damage propagation and the mode of failure, or even achieve a pseudo-ductile behavior with a gradual failure process on the macroscopic level. Several different methods can be related to this concept. Damage propagation can be influenced by modifying at least one phase – the matrix [4], the reinforcement [5], or the interface [6] – or by applying a fibrous [7] or thermoplastic [8] interlayer material. The concepts of predefining the location of the failure and influencing or controlling the failure mode can also be combined, providing a designed failure of the composite part [9].

Our research group developed a novel method to create composites with a designed interface via 3D printing specific patterns of polycaprolactone (PCL) interlayer material directly on the reinforcement

system [10-12]. The low melting temperature of PCL enables simple processability through additive manufacturing, which ensures a great freedom of design of the interlayer. PCL is soluble in epoxy; thereby, in the case of polymer composites with epoxy matrix, it does not create a new phase. PCL can modify the interfacial adhesion and the matrix material locally. Therefore, it can be used to modify the failure of composites by controlling delamination, increasing fracture toughness, and achieving a pseudo-ductile behavior. Furthermore, by applying PCL as clever interfacial patterns in the composite structure, the formation of damage and, finally, the location of the failure can be predefined in terms of mode and position.

The aim of the current research was to influence the failure behavior of UD carbon/epoxy composites by predefined failure location and modifying damage propagation in case of interlaminar loading, and to validate the method of local interface engineering carried out with polycaprolactone (PCL) interlayer material. A further goal of the study was to provide a better understanding of the effects related to the method in terms of damage initiation and propagation through finite element modeling (FEM).

2 EXPERIMENTAL

2.1 Materials

As matrix material of the manufactured composite plates, IPOX MR 3010 (IPOX Chemicals Kft., Budapest, Hungary), DGEBA-based epoxy resin (EP) was applied with IPOX MH 3124 amine type curing agent. The components were mixed in a weight ratio of 100:35 (mixture viscosity 500 mPa·s, pot life 30 min at 25°C). We used PX35FBUD0300 (Zoltek Zrt., Nyergesújfalu, Hungary) stitch-bonded unidirectional carbon fabric (309 g/m² surface weight), consisting of Panex35 50k rovings as reinforcement material. The interlayer material was printed from eMorph175N05 (Shenzhen Esun Industrial Co. Ltd., Shenzhen, China) poly (ϵ -caprolactone) (PCL) filament (1.75 mm diameter, 60°C melting temperature, 180°C printing temperature).

2.2 Manufacturing of composite plates

For the experiments, composite plates with [0°₆] layup sequence were manufactured by vacuum-assisted resin transfer molding (VARTM). Besides baseline composites, locally interfacially engineered composites containing PCL interlayer material were manufactured. For these plates, the PCL filament was printed directly on the surface of the carbon fabric by the fused filament fabrication (FFF) method (Fig. 1) using a CraftBot Plus 3D printer (CraftUnique Kft., Budapest, Hungary). In the case of interfacially engineered composites, the interlayer material was applied perpendicular to the fiber direction (Fig. 1) and was placed only between the two central plies. Regarding the geometry of PCL, the thickness of the interlayer material was, based on previous research [9], constantly 0.2 mm in order to avoid fiber waviness and focus on the effects related to other mechanisms of PCL, such as the local modification of the matrix material and the interfacial adhesion. However, the width (w) of the interfacially engineered zone varied, PCL interlayer materials with 1, 2, 4 and 6 mm width were applied.



Figure 1: PCL interlayer material 3D printed directly on the reinforcement material.

Specimens for the material testing were cut from the composite plates with a Mutronic Diadisc (Mutronic, Rieden am Forggensee, Germany) diamond disc cutter. Each interfacially engineered specimen contained one zone modified with PCL.

2.3 Methods

Short-beam shear (SBS) tests were carried out according to ISO 14130 standard using a Zwick Z250 (Zwick GmbH, Ulm, Germany) universal testing machine with a 20 kN load cell to investigate the effect of PCL and its geometry on the failure process. The test setup can be seen in Fig. 2. The measurements were conducted with a 1 mm/min test speed using specimens with a nominal thickness of 3 mm, a width of 15 mm, and a length of 30 mm. The supports had a radius of 2 mm, while the loading member had a radius of 5 mm. The short-beam shear tests were monitored with a 5 MPixel resolution IDS U3-3080CP-P-GL (Imaging Development Systems Inc., Obersulm, Germany) camera. We tested 10 specimens from each type.

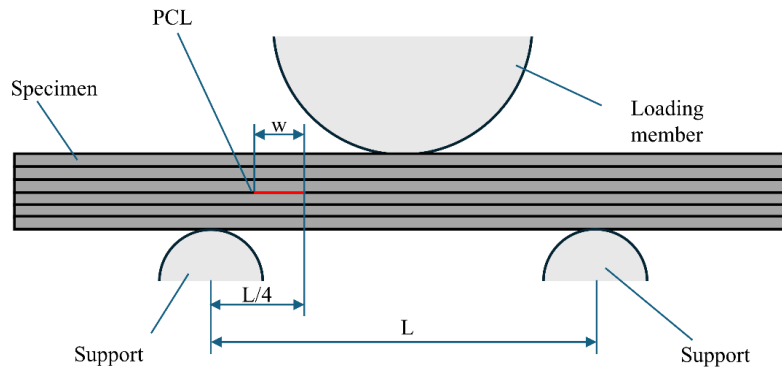


Figure 2: The test setup of the short-beam shear tests

3 FINITE ELEMENT MODELING

For modeling the delamination, we conducted finite element analysis for the loading and boundary conditions of the short-beam shear test with Ansys Workbench 2024R1 FEM software using the cohesive zone method (CZM). The model parameters were specified based on the relevant literature [12] and refined using our own data. To model the delamination process, we applied the fracture energy-based debonding model available in the software. A pure Mode II fracture model was adapted, reflecting the dominant shear-driven delamination mechanism in the short-beam shear test.

Two modeling concepts were used. The first approach involved introducing an artificial defect at the interface in the zone containing PCL. This was achieved by removing the contact definition between the adjacent surfaces of these plies over the width of the interlayer material. In this setup, no contact regions or cohesive separation definitions were assigned to the interfacially engineered zone, simulating an initial delaminated zone. In the subsequent simulations, the locally modified adhesion zone was modeled using the cohesive zone parameters of an interface modified with PCL. Non-modified interfacial regions were modeled with CZM using the parameters related to a baseline interface in each case.

To define the CZM model, interlaminar shear strength values, related to a baseline and a PCL-modified interface, determined during the short-beam shear tests, were used. Further CZM parameters and material properties are shown in Table 1.

Parameter		Value
E_1	[GPa]	123.3
E_2 and E_3	[GPa]	7.78
ν_{12} and ν_{13}	—	0.27
ν_{23}	—	0.42
G_{12} and G_{13}	[GPa]	5.00
G_{23}	[GPa]	3.08

G_{IIC} [12]	[J/m ²]	234.1
$G_{IIC,PCL}$ [12]	[J/m ²]	334.1

Table 1: Material properties and CZM parameters.

4 RESULTS AND DISCUSSION

4.1 Results of the short-beam shear tests

Initially, the force-deflection curves obtained from the measurements should be analyzed. Fig. 3 demonstrates the force-deflection curves of a characteristic specimen for each type.

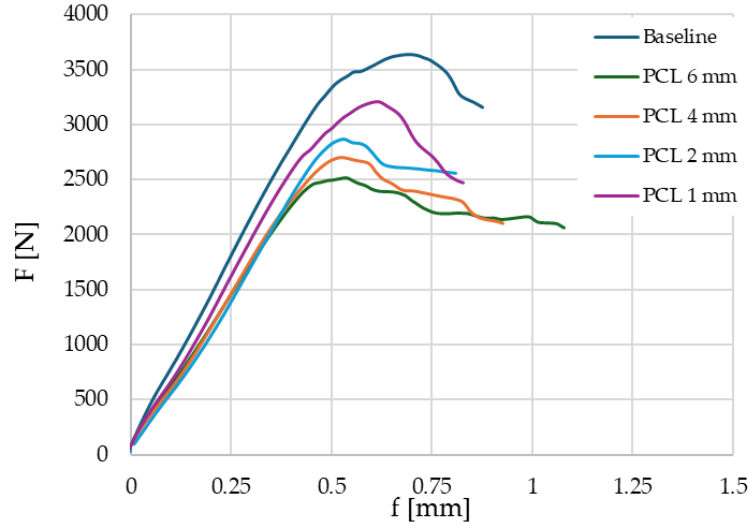


Figure 3: Characteristic force-deflection curves of the short-beam shear tests

The curve of the baseline sample indicates sudden delamination propagation. Compared to the baseline specimens, the specimens with 1 mm and 2 mm wide PCL interlayer material exhibit a lower maximum force. However, the overall shape of the force-deflection curve is similar to that of the baseline samples, indicating relatively rapid crack propagation. In this case, PCL acts as a local defect, and crack initiation is expected to occur near the interfacially engineered region. In contrast, for the specimens containing 4 mm and 6 mm wide PCL interlayer material, a plateau appears after the reduced force maximum. This indicates that the interfacially engineered zone slows down the propagation of the delamination.

The calculated interlaminar shear strength values can be seen in Fig. 4. The results show that increasing the width of the PCL interlayer material reduces the interlaminar shear strength, while the standard deviation of the results decreases significantly as well, indicating a more predictable failure.

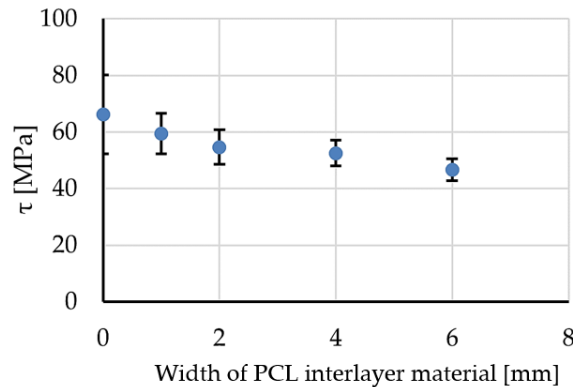


Figure 4: Interlaminar shear strength determined with the short-beam shear tests.

4.2 Results of the finite element analysis

The finite element analysis resulted in the delamination failure of the specimens similar to the short-beam shear tests (Fig. 5).

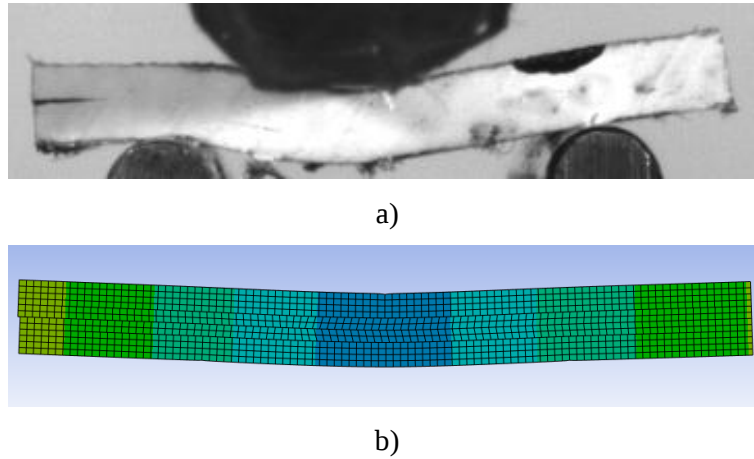


Figure 5: Delamination failure during a) a conducted short-beam shear test, b) finite element analysis of a short-beam shear test

As the contact status in Fig. 6 indicates, compared to the baseline model, the modified zone pre-defines the position of delamination initiation for both modeling concepts. This means that even in the case of the lowest width, the method is suitable for influencing the position of interlaminar crack initiation.

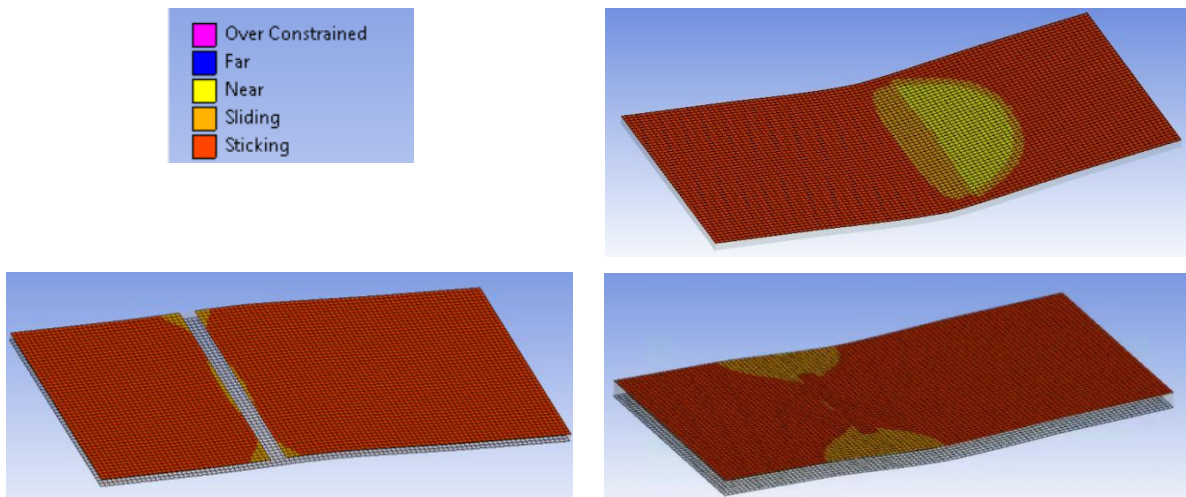


Figure 6: Contact status between the two central plies a) Status types, b) Baseline, c) PCL as initial delamination in 1 mm width, d) PCL interlayer material with 1 mm width

Table 2 contains the calculated interlaminar shear strength values, while the force-deflection curves of the simulations are shown in Fig. 7. Besides the baseline simulations, the curves include the results of the 1 mm and 4 mm wide simulations. In the experimental results, these two specimen types demonstrated completely different behavior. The 1 mm wide zone had an effect similar to an initial defect. Meanwhile, the 4 mm wide zone was the first to result in an extended plateau.

Type	Interlaminar shear strength [MPa]		
	Experimental	FEA	
		Initial delam.	PCL
<i>Baseline</i>	66.2		67.9
<i>1 mm</i>	59.4	48.6	65.0
<i>4 mm</i>	52.5	39.6	56.3

Table 2: Interlaminar shear strength values

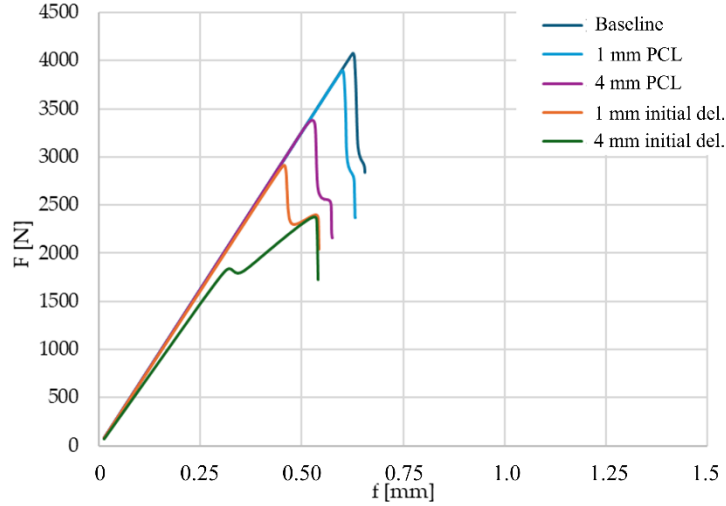


Figure 7: Force-deflection curves of the different simulations.

Based on the results of Fig. 7 and Table 2, the simulations including an initial delaminated zone, where contact was removed at the PCL location, better approximate the gradual failure process. However, in the simulations where a contact region with a cohesive zone was defined at the interfacially engineered zone, the interlaminar shear strength values more accurately reflect the experimentally measured values. Furthermore, similarly to the experimental results, in the case of the simulations, the increase of the width of the modified zone results in a more gradual failure process as well.

5 CONCLUSIONS

In our research, we manufactured locally interfacially engineered composites containing PCL interlayer material with different geometries. We carried out short-beam shear tests to investigate the effect of PCL and its geometry on the failure process. The results indicate that PCL can influence the initiation and propagation of delamination. With increasing width, the modified zone might be able to slow down crack propagation.

Finite element analysis with cohesive zone modeling was carried out for the loading conditions of the short-beam shear test. The simulations demonstrated similar results to the short-beam shear test, validating our method.

To further improve the modeling of the gradual failure process and the effect of PCL interlayer material, the introduction of a gradient region at the edges of the interfacially engineered zone is recommended. This could model the interphase created by the partial dissolution of PCL in the epoxy resin system.

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REFERENCES

- [1] G.Z. Marton and G. Szebényi, Influencing the damage process and failure behaviour of polymer composites - A short review, *Express Polymer Letters*, **19**, 2025, pp. 140-160 (doi: [10.3144/expresspolymlett.2025.11](https://doi.org/10.3144/expresspolymlett.2025.11)).
- [2] R.B. Heslehurst, *Defects and in Composite Materials and Structures*, CRC Press, Boca Raton, 2014.
- [3] A. Melaibari A., A. Wagih, M. Basha, A.M. Kabeel, G. Lubineau and M.A. Eltaher, Bio-inspired composite laminate design with improved out-of-plane strength and ductility, *Composites Part A: Applied Science and Manufacturing*, **144**, 2021, 106362 (doi: [10.1016/j.compositesa.2021.106362](https://doi.org/10.1016/j.compositesa.2021.106362)).
- [4] C. Zhang, G. Zhang, X. Shi and X. Wang, Effects of carbon nanotubes on the interlaminar shear strength and fracture toughness of carbon fiber composite laminates: A review, *Journal of Materials Science*, **57**, 2022, pp. 2388-2410 (doi: [10.1007/s10853-021-06734-z](https://doi.org/10.1007/s10853-021-06734-z)).
- [5] J. Henry and S. Pimenta, Bio-inspired non-self-similar hierarchical microstructures for damage tolerance, *Composites Science and Technology*, **201**, 2021, 108374 (doi: [10.1016/j.compscitech.2020.108374](https://doi.org/10.1016/j.compscitech.2020.108374)).
- [6] B. Magyar B., G. Szebényi G and T. Czigány, Comparison of different interfacial engineering methods to achieve pseudo-ductile behaviour of carbon fibre reinforced polymer composites, *Proceedings of the 22nd International Conference on Composite Materials* (Eds. A. Mouritz, C. Wang, B. Fox) Melbourne, Australia, August 11-16, 2019, RMIT University, Melbourne, 2019, pp. 472–480.
- [7] H. Wang, Y. Lin, H. Jiang and Z. Liu, Inter-layer failure and toughening mechanisms of carbon/aramid hybrid fiber composites interleaved with micro/nano pulps under low-velocity impact load, *Thin-Walled Structures*, **202**, 2024, 112086 (doi: [10.1016/j.tws.2024.112086](https://doi.org/10.1016/j.tws.2024.112086)).
- [8] S.G. Marino and G. Czél, Development and characterisation of reparable, film-interleaved, pseudo-ductile hybrid composites, *Composites Part A: Applied Science and Manufacturing*, **169**, 2023, 107496 (doi: [10.1016/j.compositesa.2023.107496](https://doi.org/10.1016/j.compositesa.2023.107496)).
- [9] G.Z. Marton, Á. Fendrik and G. Szebényi, Manufacturing of composites with designed failure, *IOP Conference Series: Materials Science and Engineering*, **1313**, 2024, 012014 (doi: [10.1088/1757-899X/1313/1/012014](https://doi.org/10.1088/1757-899X/1313/1/012014)).
- [10] G. Szebényi, T. Czigány, B. Magyar and J. Karger-Kocsis, 3D printing-assisted interphase engineering of polymer composites: Concept and feasibility, *Express Polymer Letters*, **11**, 2017, pp. 525-530 (doi: [10.3144/expresspolymlett.2017.50](https://doi.org/10.3144/expresspolymlett.2017.50)).
- [11] G. Szebényi, B. Magyar and T. Czigány, Achieving Pseudo-Ductile Behavior of Carbon Fiber Reinforced Polymer Composites via Interfacial Engineering, *Advanced Engineering Materials*, **23**, 2020, 2000822 (doi: [10.1002/adem.202000822](https://doi.org/10.1002/adem.202000822)).
- [12] B. Magyar, T. Czigány and G. Szebényi, Metal-alike polymer composites: The effect of inter-layer content on the pseudo-ductile behaviour of carbon fibre/epoxy resin materials, *Composites Science and Technology*, **215**, 2021, 109002 (doi: [10.1016/j.compscitech.2021.109002](https://doi.org/10.1016/j.compscitech.2021.109002)).