

DAMAGE HEALING IN COMPOSITE MATERIALS EMPLOYING SUPRAMOLECULAR POLYMERS

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ABSTRACT

Continuous fibre reinforced composite materials are susceptible to delamination damage. Healing of such damage can be done employing a thermal approach for thermoplastic composite materials. Upon heating to a sufficiently high temperature the matrix becomes soft and ductile allowing the matrix material to plastically deform and/or flow into the damaged regions to close cracks. Then, polymer molecules of the matrix material will be able to diffuse to either side of the damaged regions to actually heal/weld it.

A new and promising class of matrix materials is based on supramolecular polymers [1]. This class of polymers uses non-covalent bonds to hold the repeating units together. Upon heating, supramolecular polymers can show a dramatic decrease of the viscosity that is much larger than possible with currently used thermoplastic matrix materials. Upon cooling the viscosity regains its original value. In this research supramolecular polymers on the basis of quadruple hydrogen bonding with ureido-pyrimidones (UPy) groups are employed.

As a start readily available supramolecular polymers, supplied by SupraPolix BV, were used as an adhesive layer between Uni-Directional (UD) Carbon (C) fibre reinforced / PolyEtherImide composite materials (UD-C/PEI) [2]. The UD-C/PEI composite material without supramolecular adhesive was prepared by pressing prepregs under controlled conditions in a heated press. The quality of the healing process was determined by means of a mode I fracture toughness test using Double Cantilever Beam (DCB) specimens. After DCB tests and separation of sample sides, a thin layer of a supramolecular polymer, named SP1 here, was applied to bond both sample sides. After a healing cycle at specific conditions (pressure 4 bar at 140°C for 30 min) the DCB-test was repeated. On average a value of $G_{Ic} = 0.6 \text{ kJ/m}^2$ was determined as compared to a value of $G_{Ic} = 1.2 \text{ kJ/m}^2$ in the case of UD-C/PEI samples without supramolecular adhesive layer. Performing the healing cycle again under the same conditions, without adding more supramolecular polymer as adhesive at the crack interface, shows that the same G_{Ic} (0.6 kJ/m^2) values can be reached. These results indicate that the supramolecular polymers have the potential to heal delamination type of damage in continuous fibre reinforced composite materials.

In this work the same procedure as mentioned above was followed for a different type of composite material. In this case pressed plates of Woven Carbon Fabric (WCF), impregnated with PolyPhenylene Sulfide (PPS), were used. The samples were manufactured by placing two 2 cm × 20 cm strips on top

of each other in a dedicated oven under a mild pressure of 0.15 bar at 140 °C for 30 min. A thin layer of the SP1 supramolecular polymer was used. Subsequently, the fracture toughness values of the so-prepared samples were determined employing an in-house developed automated DCB-testing device. The average G_{Ic} value of two samples was approximately 0.51 kJ/m². Subsequently, the samples were healed under identical pressure and temperature as used for manufacturing for 6 min. The average fracture toughness value of both samples dropped to approximately 0.4 kJ/m².

Comparing the fracture toughness of the WCF/PPS samples manufactured here with the value of the UD-C/PEI samples manufactured in the past it can be concluded that the fracture toughness of the SP1 adhesive layer of the previously manufactured samples is higher than that of the WCF/PPS samples. However, it is interesting to see that although a higher healing pressure, more time, a different manufacturing route, different matrix materials and different fibre orientations were employed, the difference between the fracture toughness values of both composite systems is relatively modest. This seems to indicate that the fracture behavior is mainly determined by the behavior of the supramolecular adhesive layer itself and not so much by the strength of the bonding between the adhesive layer and the PEI-based or PPS-based composite materials. Moreover, the adhesive layer was deposited on a relatively rough fracture surface on the UD-C/PEI samples which is beneficial to the fracture strength whereas the WCF/PPS samples were manufactured from flat and smooth pressed plates.

Healing of cracks and delaminations employing this type of supramolecular polymer is clearly possible on relatively short time scales and with a low amount of additional pressure. Next research efforts are directed to further understanding of the bonding kinetics and to the synthesis and mechanical characterization of polymers that could be used as matrix material for fibre reinforced structural composites with sufficiently high modulus and toughness. First results of healing and DCB testing of these newly developed supramolecular polymers will be shown.

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