Plasma-deposited interlayers with controlled adhesion to glass fibres

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Abstract: Functional interlayers were deposited on planar glass and glass fibres from pure tetravinylsilane and its mixtures with oxygen gas by plasma-enhanced chemical vapour deposition. The interfacial adhesion of the deposited films was evaluated by nanoscratch test. We demonstrate that the interlayer adhesion is controlled by power (0.1-10 W) and oxygen fraction (0-0.71) in the gas mixture. The feasibility of the nanoscratch test was examined for single glass fibres with a diameter of 19 microns.

Keywords: PECVD, glass fibres, nanoscratch test, adhesion.

All reinforcing fibres have to be coated with a suitable interlayer (thin film) to ensure their effective functionality in fibre-reinforced polymer composites. The interlayer adhesion on reinforcing fibres is most critical and decisive for composite performance. In this study, we demonstrate that interlayer adhesion can be evaluated directly on single glass fibres with a diameter of 19 microns using the nanoscratch test. The feasibility of the nanoscratch test, commonly used for planar specimens, is examined for glass fibres coated by plasma polymer film with a thickness of 0.10 microns.

Functional interlayers of pure tetravinylsilane (power 0.1-10 W) and its mixtures with oxygen gas (oxygen fraction 0-0.71) in a form of a-CSi:H and a-CSiO:H films with a thickness of 0.10 microns were deposited on planar glass substrates and single glass fibres (GF) using an RF (13.56 MHz) helical coupling system operated in a pulsed regime. The interlayer was damaged at the fibre surface to demonstrate its presence on the fibre (Fig. 1). Nanoscale tribological properties of the interlayers were investigated using a 2D TriboScope (Hysitron) attached to an NTegra Prima Scanning Probe Microscope, SPM (NT-MDT). A conical, 90°, diamond indenter (TI-0040, Hysitron) with a tip radius of 1 µm was used to make scratches on interlayers under normal loading, which linearly increased with time from 1 μN to 3 mN. The scratch length was 10 µm and was reached in 30 s.

During the nanoscratch test, the value of the normal load at which adhesion failure is detected is known as the critical load, which is used as a measure of the film adhesion. The critical load for a-CSi:H interlayers as a function of effective power is given in Fig. 2. The adhesion increased significantly with enhanced power and was 50% higher for the film deposited at 10 W than that deposited at 0.1 W. We expect that chemical bonding at the interlayer/glass interface is realized through Si-O-C and Si-O-Si bonding species due to recombination of free radicals at Si-OH groups on glass surface. An increased density of interfacial bonds with enhanced power is responsible for elevated adhesion. However, the adhesion can be even improved if oxygen atoms are incorporated into a-CSiO:H interlayers deposited at a power of 2.5 W (Fig. 3), where the critical load as a function of oxygen fraction increased for oxidized films up to the maximum at 0.33-0.46 due to raised concentration of Si-O-C and Si-O-Si bonding species at the film/glass interface. Therefore, oxidized interlayers can be efficiently employed as the functional interlayer in GF/polyester composites, where the vinyl groups at the interlayer surface are important for chemical bonding with polyester matrix.

![Fig. 1. Scanning electron micrograph of plasma coated glass fibres.](image1)

![Fig. 2. Critical load characterizing adhesion of a-CSi:H interlayer on planar glass substrate dependent on the effective power.](image2)
We explored the feasibility of the nanoscratch test for interlayers on single glass fibres. Adjusting the position of the fibre so that its axis coincides with the axis of movement of the indenter during nanoscratch tests was extremely demanding with respect to time and experience experimenter. Therefore, only two interlayers of significantly different adhesion (critical load) were selected and deposited on single glass fibres at an oxygen fraction of 0 and 0.45 (2.5 W). The lateral force depending on the normal force for fibrous and planar samples is compared in Fig. 4 for films deposited from pure TVS. The critical load corresponding to the first delamination of the film, evidenced by an abrupt decrease of the lateral force, is marked as $L_{cF}$ and $L_{cP}$ for the fibrous and planar specimens, respectively.

Both scratches corresponding to the planar and fibrous samples had similar character; the scratch pattern was observed by AFM (Fig. 5). Although the critical loads for individual scratches on planar and fibrous samples vary somewhat (Fig. 4), the mean values are both consistent for the interlayer on planar and fibrous substrates (Fig. 6).

The differences between the critical loads for planar glass and single glass fibres are insignificant with respect to the magnitude of critical loads for both types of interlayers. Thus, we can summarize that the nanoscratch test on fibrous samples used in this study can give reliable data to distinguish at least greater changes in interfacial adhesion for different interlayers.

![Surface topography of scratch pattern characterized by atomic force microscopy.](image)

**Fig. 5.** Surface topography of scratch pattern characterized by atomic force microscopy.

![Comparison of critical loads for two selected interlayers deposited on planar glass and glass fibre. The critical loads corresponding to the same film agree for both planar and glass substrates.](image)

**Fig. 6.** Comparison of critical loads for two selected interlayers deposited on planar glass and glass fibre. The critical loads corresponding to the same film agree for both planar and glass substrates.

Our study confirmed the feasibility of the nanoscratch test for interlayers (thin films) deposited on single glass fibres. However, the scratch measurements on glass fibres with a diameter of 19 microns are much more difficult than those on planar substrates due to experimental arrangements. Interlayer adhesion is one of the key parameters controlling composite performance. Thus, the nanoscratch test for interlayers on planar substrates is sufficient as an effective tool to develop novel and functional interlayers with controlled adhesion for high-performance fibre-reinforced polymer composites.

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