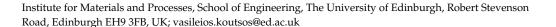




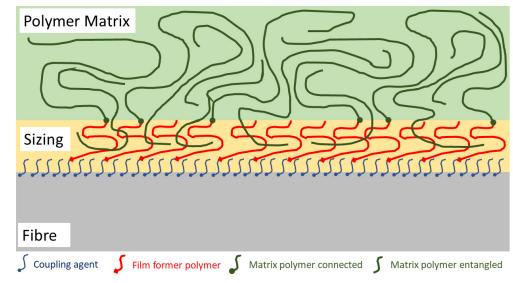
Editorial

## Interfacial Adhesion Between Fibres and Polymers in Fibre-Reinforced Polymer Composites

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The interfacial adhesion of fibres with the polymer matrix plays a major role in the mechanical properties of fibre-reinforced polymers (FRPs). The adhesive properties of the fibres are typically enhanced by the application of a thin coating, known as sizing (Figure 1). The fibre sizing protects the fibres from abrasion during processing and facilitates handling during compounding and manufacturing; moreover, its most important property is related to enhancing the bonding between the fibres and the polymer matrix, thus ensuring a successful stress transfer that inhibits delamination.



**Figure 1.** Schematic drawing of a simplified sizing located at the polymer matrix–fibre interface. The coupling agent molecules and film-former polymers are chemically linked to the fibre surface and coupling agent molecules, respectively. The matrix polymers can be either chemically linked (connected) or entangled with film-former polymers.

The sizing is formulated appropriately to wet the fibres well [1] and protect them throughout the lifespan of the FRP composite material. It is an important component of FRPs, not only for mechanical reasons, but also for chemical and thermal stability, as well as corrosion resistance. The critical role of sizing is achieved by incorporating several components, including coupling agents, the film former, antistatic agents, emulsifier agents, antifoam agents, lubricants, plasticisers, rheology modifiers, and antioxidants. Most FRP composites contain glass fibres [2], which have excellent mechanical properties and are inexpensive; however, for more demanding engineering applications, carbon fibres [3–7] are preferred due to their superior mechanical properties. Additionally, natural fibres are being investigated for more extensive use in engineering applications [8]. Primarily,



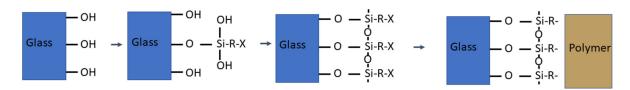
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thermosetting polymer matrices [9] are used (such as epoxies); increasingly, thermoplastic polymer [9] systems are being considered as they can be recycled, in principle [10]. The specific sizing formulation is determined by the physicochemical properties of both fibres and the polymer matrix.

A critical component of the sizing is the coupling agent (see Figure 1), which includes chemical groups with an affinity to the fibre surface. In glass fibres, this is accomplished by the incorporation of silane molecules, which, through hydrolysis and condensation, are chemically bonded to the glass fibre surface (see Figure 2). The silane molecule also incorporates a suitable chemical group that reacts favourably with the film-former molecules (which are usually polymeric) that are physicochemically compatible with the bulk matrix polymer. These include vinyl, amino, methacryloxy, or epoxy groups depending on the polymer matrix [11]. The coupling of the film former and the bulk polymer matrix is achieved by chemical bonding (incorporating suitable chemical groups) and/or interfacial diffusion and entanglements, which can be achieved by using a polymeric film former (see Figure 1). For these reasons, the film former usually constitutes the largest component (up to ~80%) of the sizing weight and is critical in facilitating effective stress transfer between the fibre and the polymer matrix, resulting in sufficiently high interfacial tensile strength, fatigue resistance, and impact resistance to prevent delamination.



**Figure 2.** Schematic drawing of the hydrolysis, condensation, and chemical bonding to the film-former polymer of a silane coupling agent used in connection with glass fibres. R denotes an alkyl group; X denotes a chemical group with a chemical affinity to the film-former polymers.

Carbon fibres are less reactive than glass fibres; the chemical bonding to appropriate coupling agents is achieved by the surface treatment of the carbon fibres using suitable physicochemical means (e.g., heat, oxidative, or plasma treatment) [12,13]. These methods induce the formation of functional groups (e.g., carboxyls, hydroxyls, carbonyls, amino, and amide groups) on the carbon fibre surface that can react with the coupling agent molecules. Similarly to glass fibres, the film-former polymers combine chemical groups that can react with the coupling agents and the polymer matrix, forming the necessary strong and tough coupling between the fibres and the polymer matrix.

Despite these general principles, several outstanding issues and questions remain that require further research. First, the sizing formulations are not known in detail, as they constitute a critical component of the FRPs that composite companies are reluctant to reveal. It is not well known how the many components of the sizing layer interact with each other and influence the overall properties of the interfacial coating. For example, viscosity modifiers can enhance wetting during the application of sizing, but they can also negatively impact mechanical performance. More studies are needed for optimising the sizing formulations. In addition, the molecular-scale structuring is speculative and has not been thoroughly investigated. The molecular structure will certainly impact the interfacial adhesion properties. Furthermore, the interfacial adhesion mechanisms of the sizing are still a subject of intensive research; they include direct chemical bonding, hydrogen bonding, van der Waals forces, electrostatic interactions, and interfacial polymer diffusion, which induces physical bonding via entanglements. The fibre roughness and/or the sizing agent-induced roughness also play a critical role, as they can enhance the effective area of the interface and can induce mechanical interlocking. This important mechanism is ubiquitous

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and applies to all fibre–polymer systems; the polymer matrix penetrates and mechanically locks to surface irregularities, increasing the interfacial bonding [14]. However, similarly to the formulation, the relative importance of the different interfacial adhesion mechanisms is largely unknown.

There is great scope for both experimental and modelling work, including artificial intelligence (AI) methodologies, to optimise all the parameters of the complex system of fibre, sizing, and polymer matrix in order to achieve superior mechanical adhesion and robustness without compromising processing and manufacturability. To this end, experimental studies should combine surface-sensitive techniques such as X-ray photoelectron spectroscopy (XPS) [15], energy-dispersive X-ray spectroscopy (EDX) [16], infrared spectroscopy [17,18], Raman scattering [5], scanning electron microscopy (SEM) [19], atomic force microscopy (AFM) [5,19,20], advanced optical microscopy [21], and micromechanical techniques such as interlaminar fracture toughness [5] and interfacial shear strength testing [3]. There is also a need for industry partnerships to apply these techniques not only to model sizings but also to relevant industrial-type sizings that can be used in practical applications.

Conflicts of Interest: The author declares no conflicts of interest.

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