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Two-matrix composites: Carbon fiber micropultrusions embedded in flexible epoxy matrices

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ABSTRACT

Two-matrix composites combine fibers with two distinct matrices. This is achieved by impregnating fiber bundles with a high-stiffness matrix and embedding the cured bundles in a flexible matrix. Two-matrix composites have been shown to offer unprecedented combinations of transverse flexibility and longitudinal tensile strength, and could offer improved fiber alignment and manufacturability. Here, we explore this concept further by embedding carbon fiber micropultrusions in flexibilized epoxy matrices and examining the longitudinal compression behavior. Our results on thin-walled rings reveal that the failure mode depends on micropultrusion diameter, with small diameters resulting in micropultrusion kinking and larger diameters in splitting and crushing. Additionally, we find that two-matrix composites can offer higher compression strength than conventional composites with the same flexible matrix, despite a lower fiber volume fraction. The inherent manufacturing advantages and high anisotropy could make two-matrix composites interesting candidates for specific applications, such as morphing wings or additively manufactured composites.

1. Introduction

Several high-performance industries have adopted the use of continuous fiber-reinforced polymer composites (FRPCs) for structural components in recent decades. Typically, the fundamental building block of these composite structures is the unidirectional (UD) composite layer. A classical UD layer consists of many fibers, typically carbon, that are arranged in one single direction and are embedded in a polymer matrix, most commonly a thermoset. One particularly limiting characteristic of a typical UD layer is the low tensile failure strain in transverse direction $\bar{\epsilon}_{t,2}$, as compared to that in longitudinal direction $\bar{\epsilon}_{t,1}$. For a typical UD carbon fiber epoxy composite, the longitudinal tensile failure strain would be approximately 1.5%, while the transverse tensile failure strain would remain below 1% [1,2]. This anisotropy in terms of tensile failure strain is problematic for composite laminates, in which all layers are expected to strain by the same amount when the laminate is uniaxially loaded. Tensile loading of a laminate along one direction could result in matrix cracks in the off-axis plies, due to the mismatch between $\bar{\epsilon}_{t,1}$ and $\bar{\epsilon}_{t,2}$ (see Fig. 1a). Such transverse matrix cracks negatively affect the performance of the composite laminate as they could degrade the thermomechanical properties of the laminate, initiate delamination damage, and facilitate moisture ingress [3–5]. In attempt to solve this problem, Vasiliev and Salov proposed a radically different type of unidirectional composite, in which fibers are combined with two distinct matrix materials, instead of only one matrix [6]. In their “two-matrix” composites, glass fiber bundles were first impregnated with a high-stiffness epoxy matrix and cured, after which the composite bundles were embedded in a secondary, flexible epoxy matrix that would provide the composite with high transverse flexibility (Fig. 1b). The “direct” solution of embedding standard fibers in a flexible matrix was found to be unfeasible since the increase in transverse flexibility, achieved by selecting a highly flexible matrix, would come at the unacceptable cost of a significant reduction in longitudinal tensile strength (Table 1). This is because the flexible matrix is less efficient at transferring stress between fibers around fiber breaks. Two-matrix composites offer a better route, as they separate the two conflicting functions of the matrix between two different matrix materials. The stiff matrix provides efficient stress transfer around fiber breaks and results in a high longitudinal strength, while the flexible matrix enables a high transverse tensile failure strain.

While Vasiliev and Salov were the first to propose the two-matrix concept for combating low transverse flexibility of UD composites, other researchers have investigated similar ideas. In the “Design and Manufacture of Low-Cost Composite-Bonded Wing” program, an improved and cost-efficient stiffening approach for hat stiffeners was sought [7]. The proposed solution consisted of embedding pultruded

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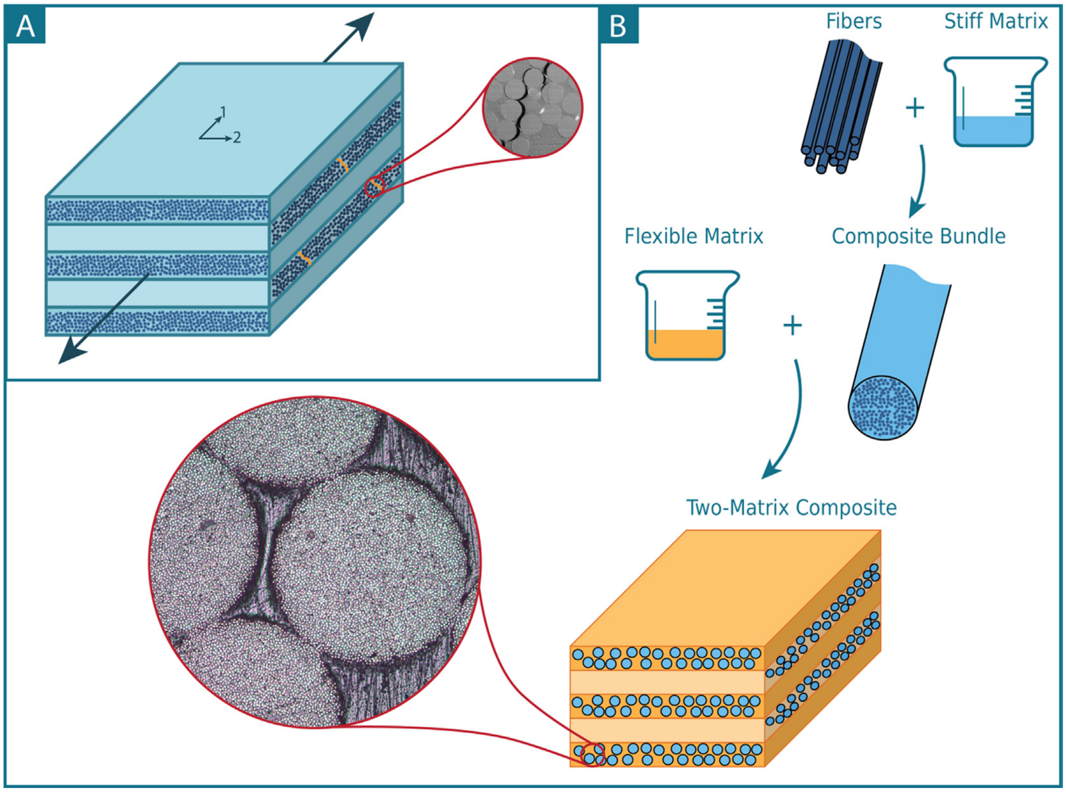

Fig. 1. Single- and two-matrix composites. (A) Schematic illustration of a 0/90° single-matrix composite laminate. Uniaxial tensile loading could result in transverse matrix cracking in the cross-plies, due to the anisotropy in failure strain of UD composite layers. Inset image was obtained from Ref. [33] with permission from Elsevier (B) Schematic illustration of the synthesis of a two-matrix composite: fibers are first embedded in a stiff matrix and cured, resulting in composite bundles. These bundles are then embedded in a secondary, flexible matrix to create a two-matrix composite.(For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Table 1
Tensile testing results obtained by Vasiliev and Salov [1], for single-and two-matrix composites.

Composite type	Fiber volume fraction	Tensile strength (MPa)	Tensile failure strain (%)
Single-matrix: glass fibers + stiff matrix	0.67	1470	0.2
Single-matrix: glass fibers + flexible matrix	0.65	1100	1.2
Two-matrix: glass fibers + flexible and stiff matrix	0.51	1420	3.0

rod packs in a syntactic adhesive, which was shown to offer reduced manufacturing cost and complexity without suffering reductions in structural efficiency, in part due to the low fiber waviness [8]. Potter and Wisnom [9] proposed “composites of extreme anisotropy” for applications requiring both a high bending stiffness and a low torsional rigidity. Similarly to the work of Vasiliev and Salov, the researchers embedded carbon fiber pultruded rods (1.7 mm diameter) in a low-stiffness matrix and performed mechanical tests. The researchers could successfully achieve high bending-to-shear stiffness ratios and showed that a demonstrator beam could withstand twist angles up to 20° without signs of permanent damage. Cairns and Bundy [10] suggested the use of carbon fiber pultruded rods (1.2 mm diameter) embedded in a secondary (non-flexible) epoxy matrix to reduce carbon fiber waviness in wind turbine blade applications. The researchers experimentally investigated the effect of surface treatments on the interfacial shear strength between the rods and the surrounding epoxy matrix, and found the highest strength values for media blast erosion. A final comparable concept was presented by Schmitz and Horst [11], who embedded composite bundles in an elastomeric foundation to develop a morphing wing skin with adequate span-wise bending stiffness. The researchers performed compression experiments and FEA, and observed buckling of the bundles inside the compliant foundation. However, the bundles used by Schmitz and Horst [11] had an elliptical cross section with a major axis of 2 mm and were made by stacking strips of carbon fiber prepreg. Additionally, the compliant foundation was supported on one side by a composite laminate, which would not be the case in a general two-matrix composite. An important difference of all these examples with the two-matrix composites of Vasiliev and Salov [6], is the considerably larger bundle diameter: 1.2–2 mm as opposed to approximately 0.5 mm. The use of small diameter bundles enabled the researchers to directly swap the fiber tows with the composite bundles in their manufacturing process. Nonetheless, these examples indicate other potential advantages of the use of pre-cured bundles embedded in a secondary matrix, such as reduced manufacturing costs or increased fiber alignment. As such, it is interesting to explore the two-matrix concept further and to investigate whether it could lead to an alternative building block for the design of composite structures. Here, we present our own type of two-matrix composite, consisting of carbon fiber micropultrusions embedded in a flexibilized epoxy matrix. Our work is the first (to our knowledge) to combine such small diameter pultrusions (280 μm–700 μm diameter) in a secondary, flexible matrix, synthesized using only epoxy resins and appropriate hardener. We discuss the selection and synthesis of the constituent materials, as well as the manufacturing method to create two-matrix composites. In the same spirit as the work of Vasiliev and Salov [6], we use a manufacturing set-up where the input material could easily switch between fiber tows and pre-cured composite bundles. Moreover, we build upon the earlier foundations in terms of longitudinal and transverse tensile