Manufacturing Silk/Epoxy Composite Laminates: Challenges and Opportunities

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Abstract: Application of natural fibers in polymer composites has been gaining popularity in several industries pursuing environmentally friendly products. Among the natural fibers with proven potential applications, silk fibers have recently received considerable attention from researchers. Silk fibers provide higher mechanical properties compared to other commonly used natural fibers such as sisal, jute, and hemp. Silk may also exhibit comparable specific mechanical properties to glass fibers. However, silk composite laminates are rarely used in commercial products due to a number of fabrication challenges. This paper investigates such challenges for silk/epoxy laminates, especially issues related to manufacturing and preform architecture. First, challenges arising from preform architecture (i.e., random and woven preforms) are presented. Unlike glass fibers for which random mats are easier to manipulate, handling random silk preform proves to be more challenging, particularly compared to woven silk fabrics. The random silk/epoxy laminates show higher thickness variation and lower compaction, yielding lower fiber content. Second, fabrication of laminates by vacuum bag/wet lay-up and vacuum assisted resin transfer molding (VARTM) processes are presented. VARTM is found to be more appropriate for silk/epoxy laminate fabrication, as it allows a uniform impregnation of the silk preform, yielding higher part quality and limited void formation. Moreover, applying 0.21 MPa (30 psi) external pressure to the VARTM laminates allows to increase the fiber content of both random and woven silk/epoxy laminates from ~17 and ~30% to ~21 and ~33%, respectively. In contrast, wetting of silk preform during wet lay-up process, which is operator dependent, is difficult to achieve; and the produced laminates have high void content. Furthermore, SEM images show a weak silk/epoxy adhesion in laminates fabricated without external pressure. Finally, the mechanical performance of these laminates is assessed. The woven silk/epoxy laminates fabricated by pressurized VARTM exhibits the highest improvement in the specific flexural strength and modulus over pristine epoxy with 30 and 65% increase, respectively.

Keywords: Silk fibers, natural fiber composites, fabrication, mechanical properties.
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INTRODUCTION

Over the past few years, environmental awareness and stricter environmental policies have forced industries, including automotive and construction, to pursue alternative eco-friendly materials, thus leading to increased interest in natural materials [1-3]. Driven by their promising biodegradability, natural fiber reinforced composites have attracted wide attention. For instance, the low cost, low density, and sustainable nature of plant fibers, such as sisal, jute, and hemp, made them attractive in comparison to the commonly used reinforcing fiber, E-glass, despite their lower properties. Therefore, a significant growth in the commercial use of plant fiber reinforced composites has been recently observed, mainly in the automotive industry [4].

In contrast, silk fibers have received limited scientific interest as a reinforcement for composites. Yet, silk filaments exhibit higher mechanical performance than plant fibers and offer naturally continuous length. In addition to a considerably higher toughness, its lower density makes silk a potential reinforcement to produce composites with specific mechanical properties comparable to those reinforced with glass fibers, as suggested in recent studies [4-5]. For example, Yang et al. [5] fabricated woven silk/epoxy composites using a wet lay-up process followed by vacuum bagging and hot pressing at high pressure levels, yielding laminates with fiber volume fractions as high as 70%. The authors investigated the tensile, flexural, interlaminar shear, impact, dynamic and thermal properties of the silk/epoxy composites and reported a linear increase of almost all properties with increasing fiber content between 30 and 60%. The reported specific mechanical properties compared well with those of glass/epoxy composites [5]. Similarly, Shah et al. [4] prepared nonwoven and woven silk/epoxy composites using vacuum assisted resin transfer molding (VARTM), with fiber volume fractions of 36 and 45%, respectively. The authors reported the tensile and flexural specific strengths (~90 MPa/g cm^{-3} and ~170 MPa/g cm^{-3}) to be comparable, although not necessarily superior, to those of glass/epoxy laminates. These findings propose silk fibers as potential sustainable alternative reinforcement materials to glass fibers in structural applications.

Random silk/epoxy laminates showed no improvement in flexural strength over the neat epoxy. While practically no change is observed for in absolute numbers for both laminates fabricated with wet lay-up and VARTM without external pressure, a slight decrease is registered for the specific flexural strength as the density of the laminates increases. A net increase of 26-48% in flexural modulus is observed over neat epoxy, with a concurrent decrease (~50%) in strain to failure. These results are attributed to the observed weak silk/epoxy interface (Figure 4), as well as excessive voidage [9]. Applying a 0.21 MPa external pressure, however, yields a significant increase in mechanical performance. For instance, flexural strength increases by 26% to reach 150.7 MPa. Woven silk fabric/epoxy laminates, on the other hand, showed improved mechanical performance, which is expected given their higher fiber content. At 30% fiber content, the woven silk/epoxy laminate fabricated using VARTM without external pressure displayed a 23% increase over neat epoxy, reaching a comparable performance to the pressurized random silk/epoxy laminate. The woven silk/epoxy laminates fabricated by pressurized VARTM exhibited the highest improvement in the specific flexural strength and modulus over pristine epoxy with 30 and 65% increase, respectively. Comparing the obtained properties of both silk preforms further confirms the superiority of woven silk as a reinforcement for epoxy composites. Furthermore, the fractured surfaces of these laminates, depicted in Figure 5, show mostly pulled silk fibers, which implies that an appropriate surface of silk fibers may significantly improve silk/epoxy adhesion, yielding further improvement of the composite performance.

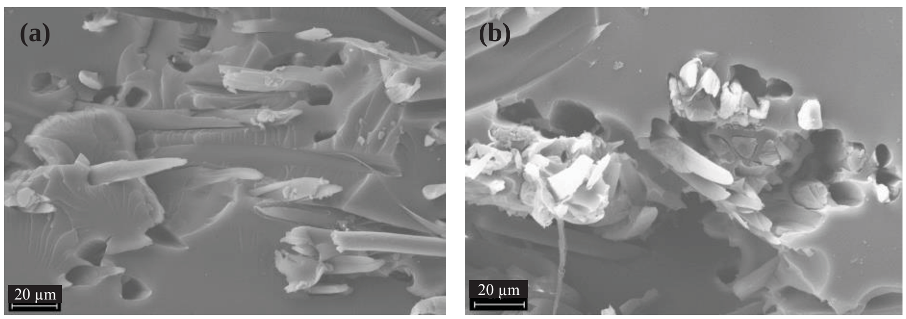

FIGURE 5. Representative SEM images obtained at 500X magnification from fractured surfaces of silk/epoxy laminates manufactured with: (a) random silk mats without external pressure and (b) woven silk fabric pressurized at 30 psi.

CONCLUSION

This paper investigated fabrication challenges for silk/epoxy laminates, especially issues related to manufacturing and preform architecture. Woven preform architecture was observed to be more convenient for structural silk composites, as handling random silk preform proved to be more challenging. The random silk/epoxy laminates showed higher thickness variation and lower compaction, yielding lower fiber content. Furthermore, vacuum assisted resin transfer molding (VARTM) is found to be more appropriate for silk/epoxy laminate fabrication, as it allows a uniform impregnation of the silk preform, yielding higher part quality and limited void formation. Silk preform wetting in wet lay-up, on the other hand, is difficult to achieve; and the produced laminates contain excessive voids. In addition, applying an external pressure as low as 0.21 MPa (30 psi) to the VARTM laminates yielded an increase in the fiber content of both random and woven silk/epoxy laminates from ~17 and ~30% to ~21 and ~33%, respectively. Furthermore, SEM images showed weak silk/epoxy adhesion in laminates fabricated without external pressure. Regarding the mechanical performance, the woven silk/epoxy laminates fabricated by pressurized VARTM exhibited the highest improvement in the specific flexural strength and modulus over pristine epoxy with 30 and 65% increase, respectively. These performances can be further enhanced using a suitable surface treatment or a sizing applied to silk fibers at higher fiber contents.

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