

Adhesion enhancement in a biomimetic fibrillar interface

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Abstract

Two important putative functions of the fibrillar contact interfaces commonly found in lizards and insects are to provide contact compliance and enhanced adhesion. To explore the question of whether a fibrillar architecture inherently enhances adhesion, we constructed model structures consisting of thin sheets of poly(vinyl butyral) (PVB) bonded on one of their thin sides to glass plates. The PVB samples had two flat, unstructured regions interrupted by a central fibrillar region along the bonded interface. The effect of the fibrillar geometry on the performance of the adhesive bond was tested using a tensile pull-off test, in which failure occurred by interfacial crack propagation, starting at an end where a crack initiator was introduced. We observed that fibrils in all samples fail consistently at the same critical stress, which is consistent with a previous theoretical result we have determined for flaw insensitivity during fibrillar pull-off. In addition, we measured the energy release rate required to fail the interface in the fibrillar region to be about an order of magnitude greater than that in the non-fibrillar region. We present experimental evidence demonstrating that this increase results partly from dissipation of strain energy stored in the fibrils.

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1. Introduction

Recent studies on adhesion in several biological systems and their mimics have suggested that a fibrillar surface geometry may be advantageous when exploiting non-covalent surface interactions to attain adhesion. It has been suggested that such surfaces arrest interfacial cracking [1–3], require larger stress and/or more energy per unit contact area to fail [3–6], and have increased interfacial contact compliance [7,8] compared to non-fibrillar counterparts. In addition, it is thought that these surfaces are more capable of adhering to substrates with varying degrees of roughness [8–11]. Finally, theoretical treatments show that fibrillar interfaces can be designed such that failure of a single fibril becomes

insensitive to crack propagation and occurs only when the pull-off stress reaches the theoretical interfacial strength [3,12,13]. Experimentally, this property is manifest in fibrillar samples showing larger pull-off stress per unit contact area than non-fibrillar controls [3,4,6,14].

In many of the studies so far [3,9,14], substantial experimental effort has been dedicated to fabricating very small length scale fibrils (less than 5 μm in diameter) in order to replicate the dimensions of fibrils found on the adhesive surfaces of lizards, spiders, and insects. (A rather incomplete but representative list of the literature discussing structure and function of fibrillar surfaces in biological systems is given by Refs. [15–22].) However, because of difficulties either in constructing a large, uniform array of fibrils at such small length scales or in performing adhesion experiments on these structures, which to date have proven quite fragile, only a handful of experimental results have appeared [1–4,7,14]. Furthermore, the results on crack arrest [1,2] have been obtained

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from surface geometries that are like fibrillar geometries only in that the bond line is periodically interrupted.

Broadly, there are two important questions relating to the performance of a fibrillar interface: (a) Can a significant fraction of fibrils in a large surface array make contact with a possibly rough substrate? and (b) does a contacting fibrillar interface enhance adhesion compared to a flat control? Because in most experimental realizations (a) precedes (b), the ability to answer question (b) is usually compromised by the inability to satisfy (a). This study, by the use of a larger scale model experimental system, obviates the need to attain uniform contact and attempts to answer the question: does a fibrillar interface enhance adhesion, once strong, uniform, contact is established?

Although a framework of ideas and evidence supporting an affirmative answer to this question have been set forth in the experimental work cited [1–4,7,14], the results of those works are largely qualitative. Here, we provide further experimental support for the qualitative conclusions already known and make quantitative statements about the work and stress needed to fail a fibrillar adhesion surface. All of this is done using a macro-scale physical system designed to be mechanically equivalent to the much smaller biological fibrillar surfaces.

Specifically, we fabricated fibrillar interfaces with millimeter scale dimensions by bonding notched poly(vinyl butyral) (PVB) sheets to glass plates. PVB was chosen for the material because its mechanical properties are well known. In addition, PVB adheres to glass very well, and the strength of the bond can be controlled easily by heat treatment. Note that it was necessary to choose a system with fairly strong adhesion to observe the advantages of a fibrillar architecture at the millimeter scale. If we had instead chosen a material system with weaker intrinsic adhesion, it would have been necessary to reduce the fibrillar dimensions, as in biological systems. This is discussed further by Hui et al. [3], Gao et al. [12,13] and in the discussion section.

Pull-off experiments were performed on the PVB/glass samples. These experiments provided data to allow the extraction of an effective energy release rate for non-fibrillar and fibrillar samples having fibrils of various lengths. In addition, the data provided the means of calculating the pull-off stress of a single fibril, and observations of interfacial crack propagation allowed confirmation of the crack arrest results observed for other geometries [1,2].

2. Materials and methods

2.1. Sample preparation

Sheets composed of plasticized poly(vinyl butyral) (PVB), commonly known by the trade name Butacite®

(DuPont), were cut into squares 30 mm by 30 mm, with sheet thickness 0.76 mm. Each square sample was adhered along one of its thin sides to a clean glass slide¹ by heating the slide while the desired side of the sample was maintained in contact with the glass. Heating was executed by setting the glass and contacting polymer on a hot plate maintained at a constant temperature of 108 °C for 60 s. Afterward, the sample was cooled in ambient laboratory conditions until room temperature was reached. A typical bonded sample is shown in Fig. 1.

Care was taken to choose the hot plate temperature so that a repeatable bond width equal to the sheet thickness was obtained. (That is, all of the polymer initially in contact with the plate before heating became intimately bonded, but no melt or flow of the polymer occurred outside of this region.) See Fig. 2. A razor blade was used to de-bond one end of the interface for a small distance. This procedure provided a stress concentrator in each sample, from which interfacial crack propagation began when a vertical tensile force was applied to the top of the sample. The initial length of the region de-bonded in this manner was not precisely controlled, but was usually between 1 and 2 mm.

Opposite the bonded region, a grid was attached to the glass slide to allow measurement of the crack length as the samples were pulled off. The grid was prepared using a simple ink jet printer and transparency film and lines were drawn every 300 μm .

In order to study the effects of fibrillar geometry, both solid square PVB samples and samples having fibrils cut at the bonded interface were prepared. Fibrils were cut using a jig made of eleven equally spaced razor blades. When the jig was applied to a PVB sample, four fibrils resulted, centered along the bond interface. Each fibril had a rectangular cross-section, 1.23 by 0.76 mm. Samples were prepared with fibrils of length 2, 5, 10, and 15 mm.

2.2. Material behavior

Plasticized PVB is widely used as an interlayer material for glass laminates in both architectural and automotive applications. At small strains, PVB is accurately modeled as a linear viscoelastic material. Its mechanical properties are well characterized, and data on the shear relaxation modulus, as well as the storage and loss moduli have been reported elsewhere in the literature [23]. The glass transition temperature T_G of PVB

¹ New glass slides were taken directly from a newly opened box, thoroughly rinsed with acetone and de-ionized water, and dried with N_2 . Dimensions of glass slides: 75 × 50 × 0.96 mm.

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