BIOINSPIRED HOOK SURFACES BASED UPON A UBQUITOUS WEED (GALIUM APARINE) FOR DRY ADHESION

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Galium aparine
ABSTRACT

*Galium aparine* is a plant noted for its ability to attach to solid objects through its microscopic hooked trichomes via a hook and loop mechanism. This dry adhesive behaviour has been mimicked using a simple and inexpensive epoxy resin replica moulding technique. Enhancement of adhesion strength is achieved by varying the epoxy resin replicas’ Young’s modulus in order to provide strong hook and loop interlocking, whilst retaining sufficient bulk material flexibility for subsequent detachment via a hook uncurling mechanism.
1. INTRODUCTION

Surfaces of natural living species have evolved over time to enable them to thrive in their local environment. One such example is dry adhesion (based solely on interfacial physical interactions), and has been found for geckos, lizards, spiders, and plants. The replication of such phenomena (biomimetics) is of potential interest for applications in the biomedical and robotics industries. Hook and loop attachment (which was identified when a dog owner observed a seed from the burdock plant entrapped in his pet’s fur) is a promising variant amongst the range of bioinspired dry adhesion mechanisms. Other examples of hook attachment mechanisms exist in the plant world, including the branches of climbing plants such as Artabotrys spp., the leaves of Desmoncus spp., and the fruit of Agrimonia eupatoria, Circaea lutetiana and Geum urbanum. In the case of leaves and stems, the hooks are mainly used for climbing, whilst hooks present on their diaspores facilitate their dispersal via humans and animals. For instance, the ubiquitous, scrambling weed Galium aparine, (a herbaceous annual found throughout Eurasia and North America), consists of tiny hooked trichomes which cover virtually every exposed surface of the plant, giving rise to its ability to strongly adhere to a wide range of objects. The hooked trichomes enable Galium aparine to compete with other plants for sunlight, which includes the leaf trichomes of Galium aparine attaching to neighbouring plants, usually via hooks situated on the abaxial leaf surface, so that it is positioned on top of the other plant, and thus situated advantageously for efficient photosynthesis. In addition, trichomes present on the stems of the plant create friction with adjacent plants leading to a physical interlocking. Finally, the plant disperses its diaspores by epizoochory (dispersal via attachment to animal fur and feathers); this is enabled by the presence of hooked trichomes present on the surface of the mericarps (the plant produces fruit in the form of schizocarps that mature and then split into two one-seeded mericarps), Figure 1. Given that Galium aparine is known to grow in different environments ranging from hedgerows to wastelands to arable fields to forests, it has been postulated that the plant’s ability to readily adhere to different surfaces may in part be responsible for its proliferation across such diverse habitats.
In this article, we describe the replication of the hooked trichomes of *Galium aparine* using soft moulding, Scheme 1. This technique is widely used for preparing 3-dimensional structures and entails preparing a negative mould from which the positive replica is made.\textsuperscript{27,28} A key feature is that the negative mould can be repetitively used, and thereby provides a major benefit compared to bottom-up approaches. Epoxy resin replicas of the leaf, stem, and mericarp surfaces of *Galium aparine* have been successfully fabricated, and their mechanical properties optimised so as to achieve comparable adhesive strengths to those measured for the native plant.
2. EXPERIMENTAL

2.1 Replica Fabrication

*Galium aparine* plant leaves, stems, and mericarps were rinsed with water to remove any surface debris and allowed to dry in air. Negative moulds from each of these surfaces were prepared by application of a polyvinylsiloxane base and cure mixture (President Plus Jet Light Body, Coltene/Whaledent AG) to the substrate, and then immediately pressing down on top using a glass slide for the duration of the 10 min cure period. Once the negative mould had hardened, it was carefully peeled away from the natural substrate surface, rinsed with water, and left to dry. A mixture of epoxy resins (D.E.R. 331 (high stiffness) and D.E.R. 732p (low stiffness), Univar Ltd.) and curing agent (Amicure PACM, Air Products Chemicals Europe B.V.) were used to prepare the corresponding positive replicas with varying stiffness. The epoxy resins, D.E.R. 331 and D.E.R. 732p, are chemically similar and differ only in the percentage content of epoxide groups (approximately 23% and 13.5% respectively), which means that the stoichiometric amount of curing agent required increases in proportion to D.E.R. 331 epoxy resin content in the mixture. The quantity of curing agent needed for each formulation was calculated by correlating its amine hydrogen equivalent weight...
(AHEW = 53) to the epoxy equivalent weight of the epoxy resins (EEW_{D.E.R.331} = 187; EEW_{D.E.R.732p} = 320), \(^{32}\) Table 1. For each calculated epoxy resin formulation, the components were thoroughly mixed before being poured onto the negative moulds. Any trapped air bubbles were removed by placement under vacuum. Subsequently, the mixture was cured in an oven at 80 °C for 2 h and then immediately post-cured at 140 °C for 2 h. Finally, the negative moulds were gently peeled away to reveal the positive replica of the original plant substrate.

Table 1: Varying stiffness epoxy resin formulations used to fabricate *Galium aparine* replicas.

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2.2 Surface Characterisation

Scanning electron microscopy (SEM) analysis was undertaken by fixing the plant samples overnight using 2% vol. gluteraldehyde in phosphate buffer solution (pH 7.4, Sigma Ltd.). These were then rinsed twice with buffer solution before undergoing dehydration using a graded series of ethanol solutions. The drying process was completed with a critical point dryer (Samdri 780, Tousimis Research Corp.). Dried plant and epoxy resin replica samples were mounted onto aluminium stubs using adhesive carbon discs and coated with a 15 nm gold layer using an SEM Coating Unit (Model No. E5000, Polaron Equipment Ltd.). Surface topography images were taken on a scanning electron microscope (Stereoscan 240, Cambridge Instruments Ltd.).
2.3 Adhesion Measurements

Young’s modulus and contact separation force were measured using a tensile tester (Model No. 5543, Instron Ltd.). For the Young’s modulus values, a cured flat epoxy resin sample of known size was secured onto the base plate of the instrument, whilst the top plate remained uncovered. The top plate was lowered at a speed of 5 mm min\(^{-1}\) until a preload of 2 N was reached and then retracted to its original position. This procedure was repeated 5 times for each epoxy resin composition. The Young’s modulus, \(E\), was calculated using Equation 1, where \(F\) is the force exerted on the epoxy resin under compression, \(L_0\) is equal to the thickness of the epoxy resin, \(A_0\) is the cross-sectional area through which the force is applied, and \(\Delta L\) is the change in thickness of the epoxy resin under compression.\(^{33}\)

\[
E = \frac{FL_0}{A_0\Delta L}
\]  
(Equation 1)

For contact separation force\(^{19}\) experiments, a plant replica was placed on the base plate and a piece of looped fabric fastener (Velcro\textsuperscript{®} Stick On Tape, Velcro Ltd.) was affixed to the top plate. The top plate was lowered, at a speed of 5 mm min\(^{-1}\), towards the replica until a preload of 2 N was reached. Subsequently, the top plate was retracted until the final detachment event occurred yielding the contact separation force. The contact separation force was determined to be the maximum force measured before the final detachment event.\(^{19}\) This was repeated 5 times for each epoxy resin composition.

3. RESULTS

3.1 Surface Characterisation of Galium aparine

*Galium aparine* has long and narrow leaves measuring up to 8 cm in length which are arranged in whorls of 6-8 around a square-shaped cross-section stem,\(^{34}\) Figure 1(a-c). The plant produces fruit in the form of schizocarps that mature and then split into two one-seeded mericarps,\(^{22}\) Figure 1(d). The entire surface of the plant above the ground (including the leaves, stem, and fruit) is covered with hooked trichomes that can ‘stick’ to a variety of surfaces\(^{19,23,26,35}\), Figure 2. The adaxial surface of *Galium aparine* leaves consists of irregularly shaped convex epidermal cells (the
outermost layer of cells) distributed evenly across the entire surface with hooked trichomes curved towards the leaf base, Figure 2(a-b). Whilst the stem is covered by rod-shaped epidermal cells (the outermost layer of cells), and the trichomes (curved towards the roots of the plant) occur only along the four corners of the square-shaped stem, Figure 2(c-d). Interestingly, the hooked trichomes present on the stem appear to be shorter with a flatter curvature than those found on the leaf surface. In the case of the mericarps, the surface is densely covered with hooked trichomes that exhibit a high degree of curvature and all tend to point in the same direction, Figure 2(e-f). The bases of all the hooks are significantly thicker than those of the tips.

Figure 2. Low and high magnification SEM micrographs of Galium aparine: (a-b) the adaxial leaf surface; (c-d) the stem surface; and (e-f) the mericarp surface.
3.2 Epoxy Resin Replica Surfaces

Soft moulding was used to fabricate epoxy resin replicas of the leaf, stem, and mericarps of *Galium aparine*, Scheme 1. A negative polyvinylsiloxane mould was prepared from the natural template and it then itself moulded using epoxy resin to create a positive replica of the original substrate. Electron microscopy verified excellent replication of the structural features of the *Galium aparine* plant parts, Figure 3. Both the epidermal cellular surface and the hooked trichomes are clearly discernable. The quality of replication was found to remain constant across the range of epoxy resin formulations employed.
Figure 3. SEM micrographs of *Galium aparine*: (a, c) the native leaf surface; (b, d) the epoxy resin replica of the leaf surface; (e, g) the native stem surface; (f, h) the epoxy resin replica of the stem surface; (i, k) the native mericarp surface; and (j, l) the epoxy resin replica of the mericarp surface.
3.3 Contact Separation Force Measurement

Epoxy resin replicas were fabricated using a range of different epoxy resin compositions, Table 1. The Young’s modulus (stiffness) of each epoxy resin formulation was measured using a tensile tester, Figure 4. The general trend shows that there is an increase in Young’s modulus as the percentage weight of the component D.E.R. 331 is increased (greater percentage of crosslinking epoxide groups present). The maximum stiffness value obtained is approximately 0.6 GPa at approximately 70-80 % wt. D.E.R. 331.

![Graph](image_url)

**Figure 4.** Contact separation force between the epoxy resin replica of the *Galium aparine* leaf adaxial surface and a piece of looped fabric fastener (Velcro). The contact separation force measured using the adaxial surface of the native leaf is 9 ± 1 mN. The Young’s modulus for each epoxy resin formulation is also shown.

The corresponding contact separation force was measured for the epoxy resin replicas of the adaxial leaf surface of *Galium aparine* by pressing a piece of looped Velcro fabric down onto the positive epoxy replica and then measuring the contact separation force by pulling the two opposing surfaces apart, Figure 4. The contact
separation force increases from approximately 5 to 30 mN in correlation to the stiffness of the epoxy resin replica material. This compares favourably with respect to a contact separation force of $9 \pm 1$ mN measured using the adaxial surface of the native leaf. SEM microscopy showed that little, if any, damage was done to the replicas during testing, and that the hooked trichomes remain largely intact, Figure 5. Plastic deformation is only observed for the low stiffness trichome hooks.
Figure 5. SEM micrographs of epoxy resin replicas of the adaxial surface of *Galium aparine* leaves prepared using different formulations, before and after contact separation force testing: (a, c) EP-0 replicas before testing; (b, d) EP-0 replicas after testing; (e, g) EP-70 replicas before testing; (f, h) EP-70 replicas after testing; (i, k) EP-100 replicas before testing; and (j, l) EP-100 replicas after testing.
4. DISCUSSION

Prior examples of replica moulding using natural templates have primarily focused on the use of relatively flat substrates, for example leaves, petals and fish skin. In this study, the 3-dimensional structures of the leaf, stem, and mericarps of *Galium aparine* have all been accurately duplicated. Fabrication of the negative mould within 10 minutes at room temperature represents a key advantage of this technique because it minimises the risk of dehydration of the natural template (shrinkage of the surface replica features is averted).

The epoxy resin leaf replicas successfully mimic the adhesive behaviour of the natural plant as demonstrated by contact separation force measurements, Figure 4. The observed trend of improving contact separation force with increasing stiffness can be easily explained by considering the ease of movement of the hooked trichome. In order to detach from a loop, the hook must either be broken off, uncurl, or the loop itself must snap. SEM microscopy shows that there is no damage to any of the trichomes after testing. Therefore, the hook must uncurl to allow for the separation of the two surfaces. The contact separation force generated by this uncurling process is governed by: the extent of interlocking between the hook and loop, the rigidity of the hook, and the interfacial contact. When the stiffer D.E.R. 331 epoxy resin is present in low concentrations (0-20% wt.) the trichome hooks are flexible and can therefore detach easily from the loops present on the fabric fastener, resulting in lower contact separation forces (as seen by the plastic deformation for the less stiff trichome hooks, Figure 5). Raising the percentage weight of the D.E.R. 331 epoxy component from 30 to 80% wt. makes the hooks less flexible indicating that a greater effort is needed to promote detachment (i.e. higher contact separation forces). The slight decrease in the contact separation force beyond 80% wt. D.E.R. 331 epoxy resin coincides with a similar drop in the Young’s modulus. This confirms that there is a strong correlation between the contact separation force and the stiffness of the epoxy resin hooks. The overall scatter of data points along this general trend can be explained by considering that even small changes to the orientation of both the hooks and the loops during the testing of the different samples can give rise to differences in the contact separation force. The spread of contact separation forces measured for a specific epoxy resin replica can be attributed to the range of trichome hooks lengths for *Galium aparine*. The previously reported
contact separation force of 9.37 to 44.89 mN for a single *Galium aparine* plant mericarp hooked trichome\textsuperscript{26} obtained using a steel wire loop is comparable to the values measured in this work, Figure 4. It is of interest to note that the contact separation forces for a single mericarp trichome and the adaxial leaf surface are approximately equal (which is consistent with the similar physical appearance of the respective surface hooks, Figure 3). In a related study, the frictional forces exerted by the adaxial surface of a *Galium aparine* leaf whilst in motion relative to a piece of looped fabric fastener were measured to be approximately 20 mN,\textsuperscript{23} which is comparable to the contact separation values obtained in the present study. Overall, the outlined soft moulding approach in combination with the hook and loop mechanism appears promising compared to many earlier bioinspired dry adhesion designs which have suffered from factors such as fiber collapse and too high material stiffness.\textsuperscript{47}

5. CONCLUSIONS

Epoxy resin duplicates of the stem, leaf and mericarp surfaces of the *Galium aparine* plant have been prepared using soft moulding. These have been shown to mimic the native plant's ability to adhere strongly to surfaces through the use of its hooked trichomes. The contact separation force between the replica surfaces and pieces of looped fabric fastener (Velcro) is found to correlate to the Young's modulus (stiffness) of the epoxy resin replica material. The accompanying detachment process entails uncurling of the replica trichome hooks.

6. ACKNOWLEDGEMENTS

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7. REFERENCES


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