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Review

Coatings preventing insect adhesion: An overview

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ABSTRACT

Insect pests cause considerable damage worldwide to plants, buildings and human health. This review explores how controlling insect adhesion to coatings might mitigate these problems. We summarise the current knowledge of the mechanisms of insect adhesion on natural and synthetic surfaces and natural examples of non-adhesive and slippery surfaces. Biomimetic, multi-scaled rough and particle-transferring surfaces provide an efficient method to reduce adhesion of crawling insects.

1. Introduction

1.1. Coatings: definition and applications

Coatings are materials prepared, generally, from liquids which form a film after drying or curing once applied to substrates. Their use has become ubiquitous in the past decades for decorative, protective and functional applications [1]. Important functional and protective properties include antibacterial, self-healing, self-cleaning, antifouling, anticorrosive, hydrophobic, oleophobic, or ice-repellency and flame-retardancy. Such coatings should be durable, easy-to-apply, inexpensive and environmentally friendly [1,2].

As well as the important aesthetic and barrier properties that coatings deliver, they are being applied to a wide range of substrates for their specialised functional properties, e.g. fruits, textiles and solar cells [3–5]. Nature shows the control of the surfaces of the coatings is important to access these functional effects, as demonstrated in the well-known examples of e.g. self-cleaning lotus leaves, shark skin or butterfly wings [6–8]. For example, lotus leaves (*Nelumbo nucifera*) are highly hierarchical rough structures made of hydrophobic, three dimensional epicuticular wax crystals, allowing for water to roll off the surface and to self-clean [6]. Bio-inspired strategies from the so-called Lotus-effect ® gave rise to extensive research in self-cleaning coatings, such as the self-cleaning outdoor Lotusan paints [9].

Insects interact with the surfaces of coatings when they land, crawl or climb on them. Whilst insects, such as pollinators and seed dispersers, are essential for most land ecosystems, many insects are considered pests, because they pose a serious threat to agriculture, forestry, buildings and human health. Possible strategies to tackle insect pests include insecticides, insect-repellent and low insect adhesion coatings [10–15]. The former however are harmful to the environment and alternative strategies are preferred [10]. This paper reviews the threats caused by insects, their mechanism of adhering to surfaces and the possible formulating strategies related to protective building and agricultural coatings.

1.2. Insect damage to crops

Insect pests in agriculture need to be controlled to avoid crop diseases or damage. Crops can be harmed or contaminated in the field or during storage [16,17].

Insect pests damage plants and crops *via* feeding, sap-sucking or infesting different parts of the plant: leaves, buds, flowers, stems, fruits and seeds, roots, tubers and bulbs; as well as seedlings and sown seeds [16]. Fruits, nectar and sap are rich in sugars, which are of particular importance in some insects' diet, as Hemiptera [18]. Invading pests include gall insects, thrips or aphids. Generally, these attacks will result in notches, irregular margins, scarification and holes in leaves, flowers or other parts of the plants. Damages to plants are not only aesthetic, but can reduce the plant's growth, photosynthesis and disturb the plant's water and nutrient balances [19].

During storage, insects can harm crops by feeding, leading to a population exponential growth, and hence contaminate the products [16,17]. Common storage insect pests include moths, mites, cockroaches and beetles.

Oerke reported the worldwide losses of various crops due to animal

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pests (insects, birds, snails, etc.), which widely depend on the type of crop and region [20,21]. About 8% for wheat, 15% for rice, 10% for maize, 11% for potatoes, 9% for soybeans and 12% for cotton are lost to animal pests. The losses in the Mediterranean basin, where approximately 98% of the world's olive trees are harvested, olive fruit loss to insect pests is at least 15% of production, equivalent to $800 million loss [22–24]. Overall, the global crop production was reduced by about 14% due to insect pests [20,25,26].

Another major economical example of the non-control of pests is the loss of $46 million in 1999 in a Californian vineyard, where the grapes were contaminated with Pierce's disease, which prevents the fruits from growing, and was transmitted by leafhoppers [27].

Many other examples of crop diseases and defects caused by insect pests are reported elsewhere [21,25,28,29].

1.3. Transmission of diseases by insects

Insects transmitting diseases are referred to as either biological or mechanical vectors, depending on where in the insect the pathogen developed, which affect several billions of people every year [30,31].

Biological vectors carry pathogens within their bodies, which are transmitted to humans or animals through bites by blood-feeding insects, such as lice, mosquitoes or fleas. Mechanical vectors, for instance flies, carry infectious agents at the surface of their bodies (legs or mouthparts) and hosts become infected by simple physical contact [30,31]. Although not responsible for transmitting diseases, stings (due to Hymenoptera, e.g. honeybees, hornets or wasps) may also be accountable for allergies, which can range from discomfort and local swelling to life-threatening anaphylaxis [32].

Upon physical contact or blood-feeding on their hosts, insects can transmit pathogens developed within their bodies our mouthparts, and toxins or potential allergens through their saliva [30]. Some species of ticks for instance, carry some toxins in their saliva which can cause paralysis of the host, allergic reactions and transmit a broad range of viral, bacterial and protozoan pathogens [33,34].

Examples of transmitted diseases to humans include malaria, with about 2 billion people at risk and causing approximately one million deaths per year [30,35]; or dengue, which affects 50–100 million people and causing about 20,000 deaths annually [30,36,37].

Insect pests also caused considerable losses to livestock, which are mainly due to reduction of milk and meat production through both transmitted diseases and stress owed to bites [30]. A dramatic example is the tsetse flies: they caused about $4.5 billion losses by infecting cattle in Africa with trypanosomiasis [38]. Ticks, mites, stable and horn flies were reported to cause the loss of approximately cumulative $5 billion on overall US livestock [39–41].

1.4. Insect damage to buildings

About 2,300 termite species have been discovered, of which 183 species are accounted for damaging buildings [42]. Termites are mostly present in Asia (mainly in India, Malaysia, China, Japan), Australia, Africa and in the USA. Termites are however of ecological importance: they raise soil quality by improving pH, organic carbon content, water content and porosity for soil aeration [43–45].

Termites feed primarily on cellulose, present in various sources e.g. wood, lichen, grass or soil, and are considered pests as soon as they start damaging man-made structures [44]. Wood-feeding termites can cause serious damage to buildings, sometimes causing them to collapse or villagers to abandon their houses. The precise economical cost of termite damage is difficult to assess due to the lack of data in underdeveloped countries, but has been estimated to be between and $2 and $40 billion per year, with about $1-1.5 billion p.a. in the USA alone [42,46,47].

When entering buildings, insect pests can further infest or damage objects or furniture made of wood, wool, linen, etc., or even pieces of art or books if entering museums and libraries [48].

In the next sections, we first review the current knowledge about insect biomechanics and insect adhesion to natural surfaces. Methods to repel or reduce insect adhesion to surfaces are discussed. Emphasis is given on paints and coatings which can reduce insect attachment to buildings and plants without using insecticides.

2. Insect adhesion and slippery plant surfaces

2.1. Biomechanics of insects

2.1.1. Insect climbing mechanisms

Adhesion is defined as the force required to dissociate two surfaces from one another. To describe friction, Coulomb distinguished static friction from dynamic friction: static friction is the friction between two objects in contact that are not moving relative to each other; while dynamic friction is the force which is necessary to slide a surface on one another. Insects can climb on a surface by means of interlocking or by adhesion forces. Their pads generate both adhesion and friction forces [49,50]. Surface roughness tends to reduce adhesion as asperities reduce contact area, yet insects and geckos can adhere to smooth as well as rough substrates [49,51,52]. Alternatively, body hairs have also been reported to provide adhesion, as observed in honeybees, which carry lubricated pollen particles from flowers to hives [53,54].

Claws and spines on the tarsus (insect foot) can interlock with substrate asperities [55,56]. The stiffness and morphology of the claws, especially the sharpness of the claw tip, impact their performances to cling to asperities [57]. On a soft surface, a stiff claw may be able to dig into the surface and find grip for locomotion. On a rigid rough surface, a claw can grip on surface protrusions that are larger than approximately the diameter of the claw tip [58,59].

On smooth surfaces with insufficient grip for their claws, insects use their adhesive pads, which fall into two categories: hairy (or fibrillar) and smooth pads. Both types of pads increase the contact area with substrates to improve attachment to rough surfaces [60,61] (Fig. 1).

The movement of insect legs occurs through attachment and detachment of the pads, *via* a peeling mechanism, similar to pressure adhesive tapes [62]. When pulling the pad towards the insect's body, adhesion is enhanced ('attachment'); while moving the pad in a distal direction (away from the insect's body), when no adhesion is required, pulls off the pad from the surface ('detachment') [49,63].

Hairy adhesive pads are composed of densely packed arrays of fine and flexible fibers, the setae, generally tipped with triangular or circular end-plates [62,64,65]. They are found in many insects, such as beetles, bugs and flies, with widths ranging from ca. 100 nm in spiders and geckos to more than 5 μm in beetles [62]. These long and flexible

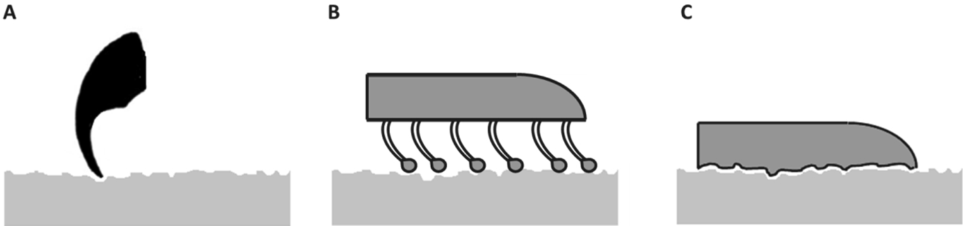

Fig. 1. Schematic representation of (A) claw, (B) hairy pad and (C) smooth pad in contact with a rough substrate.

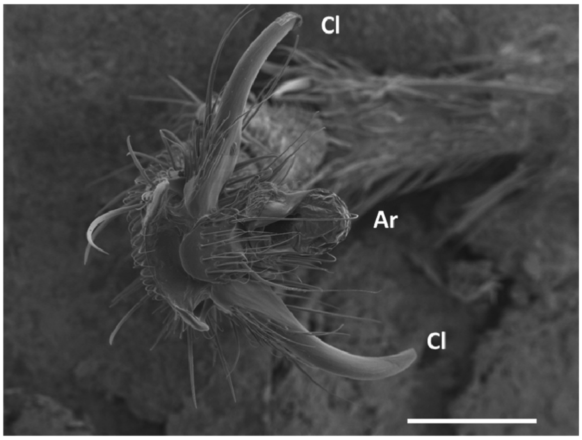

Fig. 2. Scanning electron microscopy image of *Atta cephalotes* ant tarsus showing the claws (Cl) and arolium (Ar). Scale bar: 100 μm.

hair arrays provide low elastic modulus, which are hence able to balance the surface roughness by bending and allow rapid attachment and detachment from a surface by pushing/pulling mechanisms [60]. The latter is aided by the distal orientation of the hairs as well as easy, rapid and simultaneous setae peeling off from the surface [66]. The maximum adhesive force was found to increase with the number of adhesive setae in leaf beetles [67]. Adhesion force differences in males and females have been reported in beetles, due to different seta tips for mating purposes [60,68,69].

Smooth pads are found in many insects, such as ants, bees, stick insects, grasshoppers and cockroaches (Fig. 2). A smooth pad consists of a very soft cuticular sac between the tarsal claws. Surface protrusions causes the soft pad to deform, hence maximising contact area on rough substrates [61,70]. The adhesive pad is referred to as arolium in many insects [55,71]. Arolia can be retractable and fluid-filled (e.g. in Hymenopteran insects) or non-retractable (e.g. in cockroaches) [56,71–73]. A synthetic attachment device inspired from an insect pad/claw system was described in [74]. These artificial adhesive pads and claws were reported to increase adhesion synergistically on steel spheres.

Many insects of orders such as Hymenoptera, Blattodea or Phasmatodea possess several attachment pads on the same leg [75]. The function of each individual pad has been investigated in e.g. stick insects (*Carausius morosus*) [76] and cockroaches (*Gromphadorhina portentosa*) [77] where the results suggested arolia serve as adhesion pads (“toe” pads) and tarsal pads (euplantulae or “heel” pads) are friction pads, hence providing little adhesion, in these types of insects [50]. This indicates the pads can passively and energy-efficiently control the adhesion and friction forces during locomotion.

Adhesion in insect pads is mediated by an adhesive secretion: this fluid maximises adhesion to rough substrates by filling protrusions between the pad and the surface [57,66,70,77]. The composition and nature of this fluid however remain unclear, due to the variations between insect species and the low volume secreted by pads [57,78–80]. For many insects possessing smooth pads (e.g. ants, cockroaches or certain mites), the secretion has been found to be a water-in-oil emulsion mainly containing hydrocarbons, fatty acids, alcohols, amino acids, etc. It was suggested that in smooth pads, the thin films of fluid rheologically behave like shear-thinning Bingham fluids with a yield stress [57,78], while the secretion was found to be a Newtonian fluid in beetles and flies (hairy pads) [79,81].

In insects possessing smooth pads, the thickness of the adhesive fluid was reported to about 100 nm, making its investigation challenging [57,82]. On smooth surfaces, insects should minimise the secretion of fluid to increase capillary adhesion (wet adhesion situation), whose viscosity could also impede locomotion speed and pad re-usability [61,83]. Interestingly, the adhesion in dry and wet conditions (absence and presence of adhesive secretion, respectively) has been reported to be similar by Labonte & Federle [83]. The fluid’s viscous forces were found not to improve adhesion significantly and the pad retraction speed was not correlated to the amount of fluid. The fluid layer may not only help to increase adhesion on rough surfaces, but it could at the same time act as a lubricant to ease fast detachment from surfaces [77,83].

2.1.2. *Effect of surface roughness on insect climbing*

The locomotory behaviour of insects depends on both the insect type and the nature of the surface. Many plants use surface roughness to reduce insect adhesion, as further discussed in the section II.2. Slippery plant surfaces.

Although not demonstrated formally, some insects, such as ants and cockroaches, are suspected to use their antennae to investigate surface asperities before walking on a substrate [12,84]. Antennae can indeed perceive the ant’s environment, such as airflow, chemical signalling, and detect mechanical fragilities; and were suggested to counterbalance poor vision [85].

Both types of pads can comply with surface asperities to maximise attachment forces to surfaces [60,61] (Fig. 1). They secrete adhesive fluid to improve surface contact as it compensates the surface asperities, hence increasing adhesion to rough surfaces [57,66,70,77]. Ants, which possess smooth pads and extensible adhesive pads [72], have been reported to passively deploy their arolia after mechanical claw slipping on the surface [56,72].

Hairy pads, as found in beetles, do not provide sufficient adhesion on micro-rough surfaces. The setae are suspected to make incomplete contact with the surface asperities, leading to a reduction of the contact area [59,68]. Adhesion forces generated by insects were reported to be larger on surfaces displaying surface asperity size smaller than 300 nm (‘smooth’, pad adhesion, Fig. 1B) or larger than 3 μm (‘rough’, claw interlocking, Fig. 1A) [59,68,69,86]. Specifically, surfaces with asperity diameters between 50 nm and 1.0 μm led to the lowest attachment forces, as large seta tips cannot interlock with small surface protrusions [58,59,68]. Similar roughness effects have been described in insects possessing smooth pads [87,88]. In particular, Scholz et al. [88] modelled that the *Nepenthes alata* pitcher plant inner wall should possess a pore size of 1 μm to minimise the adhesion of insects.

The roughness ranges can be defined as (1) ca. 0 nm (‘smooth’, pad adhesion), (2) 50 nm–1.0 μm (‘nano/micro-rough’) and (3) larger than 3 μm (‘rough’, claw interlocking) [59,69]. In the ‘nano/micro-rough’ range (50 nm–1.0 μm), both adhesive pads and claw interlocking are inefficient for climbing (Fig. 3).

Zhou et al. have studied insect adhesion on various rough substrates produced by lithography and displaying different pillar spacings (3–22 μm) and heights (0.5 and 1.4 μm) [89]. They studied cockroaches and beetles (smooth and hairy pads, respectively), which were found to make partial contact on dense array of pillars, while full contact was obtained for large spacing (above 4 μm) and smaller pillars (0.5 μm).

2.1.3. *Self-cleaning mechanisms*

The accumulation of particles on the body or tacky adhesive pads of insects lead to a loss of adhesion [57] and possibly to locomotion problems [90,91]. Fouling particles must be removed from crawling insects’ pads to maintain their adhesive properties, or from the body of flying insects for controlled flight. Fig. 4 shows *Atta cephalotes* ant tarsi after being contaminated with 300 nm titanium dioxide particles.

Fouling particles can be removed through self-cleaning, *via* grooming, brushing using cleaning structures, or when bringing them into contact with a surface with greater attraction forces to these contaminating particles through scratching, rolling and sliding movements [52,92–94]. The adhesive secretion of insects has also been suggested to aid the self-cleaning process by washing off particles [93].

In practice, self-cleaning is achieved in just a few steps on smooth

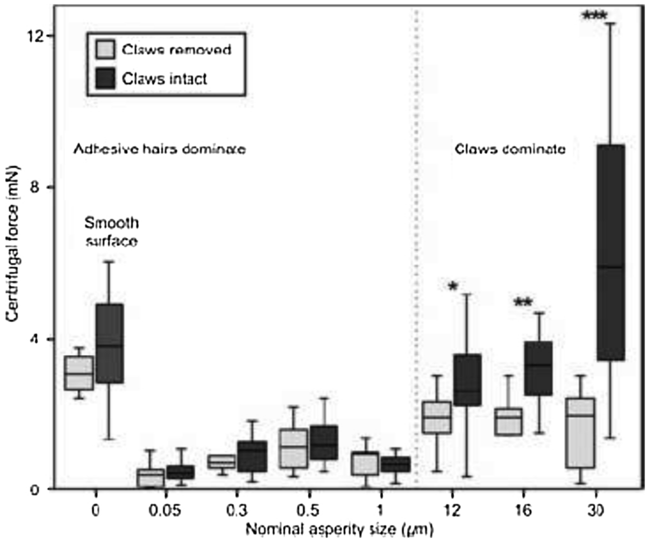

Fig. 3. Centrifuge measurements of whole-body detachment forces of male *G. viridula* on epoxy substrates of varying roughness. Asperity size is approximate and corresponds to the average nominal particle size of the original sandpaper. Reproduced with permission from [59].

surfaces, as observed in ants [92], stick insects and beetles [93], or even geckos [95]. On rough surfaces, on which claws are mainly used, the pad/surface contact area is assumed to be too low to efficiently remove particles from the pad [93,96]. Particles were found to be removed more easily from hairy pads: in smooth pads, the presence of shear was necessary, while pull-off movements only could efficiently remove particles of hairy pads [92,93]. The low surface energy of the setae might also aid the self-cleaning process [93].

The fouling particle size has been demonstrated to significantly impact self-cleaning of the pads of ants, Coccinellids and dock beetles, but surprisingly did not significantly affect stick insects [92,93]. 1 μm and larger than 45 μm particles were easily removed from insect pads, whereas 10–20 μm particles needed more steps to be removed through self-cleaning [92,93,96]. For hairy pads, large particles cannot fit in between fibrillar setae, leading to rapid particle removal [93,94,96]. Interestingly, no pad fouling has been observed on ants' smooth pads using contaminating particles larger than 100 μm, suggesting the particles need to be smaller than the claw basal spacing to adhere to the arolium [84].

2.2. *Slippery plant surfaces*

Pitcher plants families Cephalotaceae, Nepenthaceae, and Sarraceniaceae have been long known for capturing and digesting insects, mainly ants, which are attracted by the nectar the plants secrete [88,97,98]. They then fall into the pitcher, with barely any chance to escape, and get digested by pitcher fluid. Some species of the genus *Nepenthes* possess an insect-slippery waxy surface, like the widely studied *N. alata* [88,98,99], while some others (e.g. N. *bicalcarata*) only become slippery when wet [97].

Nepenthes pitchers consist of several zones: (1) the lid and peristome, (2) the slippery zone, (3) the transitional zone, and (4) the digestive zone [100] (Fig. 5A). The sections below describe these two main slippery mechanisms in species of the genus *Nepenthes*.

2.2.1. *Slippery wax-covered plant surfaces*

Many plants are known to possess a superhydrophobic surface, such as the lotus leaf, rose petals or the inner pitcher walls of *Nepenthes* pitcher plants [6,98,101]. The latter is superhydrophobic due to the combination of roughness and hydrophobicity of its epicuticular crystals present at its top surface. Superhydrophobic substrates exhibit water contact angles greater than 150° and contact angle hysteresis lower than 10° [102]. The combination of cutin biopolymer and lipids protects plants from water loss [9]. Climbing of insects on epicuticular wax-covered pitcher plant surfaces has been widely studied, see e.g. [88,98,99].

Gorb et al. studied the epicuticular wax coverage in *N. alata*, which consists of two different layers (Fig. 5B/C) and display different structures, chemical compositions, mechanical properties and different insect anti-adhesion mechanisms [99]. The lower wax layer shows a foam-like structure and is composed of platelet-shaped wax crystals coming off the surface at sharp angles [99]. The upper layer wax displays closed-packed platelets perpendicularly oriented to the lower layer. Both waxy layers are mainly composed of alkanes, aldehydes, primary alcohols, free fatty acids, esters and triterpenoids expressed in different proportions in the two layers, with predominant aldehydes and alcohols, which are likely to co-crystallise [99,103].

Insects slip into the pitcher *via* a two-fold mechanism relying on (1) pad contamination and (2) surface roughness. (1) Upon contact with insect feet, crystals of the upper wax layer break off and contaminate the adhesive pads of insects. They are also too small and too fragile for claw interlocking [99]. (2) In laboratory conditions where the upper epicuticular lipid layer was removed, the rough lower wax layer reduces the real contact area of insects' feet with the plant surface as pads cannot comply well with the surface protrusions [88,99]. In similar experimental conditions, the upper wax layer has interestingly been found to be non-recoverable, suggesting that when all crystals have detached from the upper layer, the micro-roughness of the lower wax layer would be efficient at preventing insects from climbing the walls of the inner pitcher's surface [100].

Additional anti-adhesive properties hypotheses to (1) and (2) were proposed by Gorb and Gorb [104]: (3) the wax-dissolution hypothesis: the insects' adhesive fluid may cause epicuticular wax crystals to

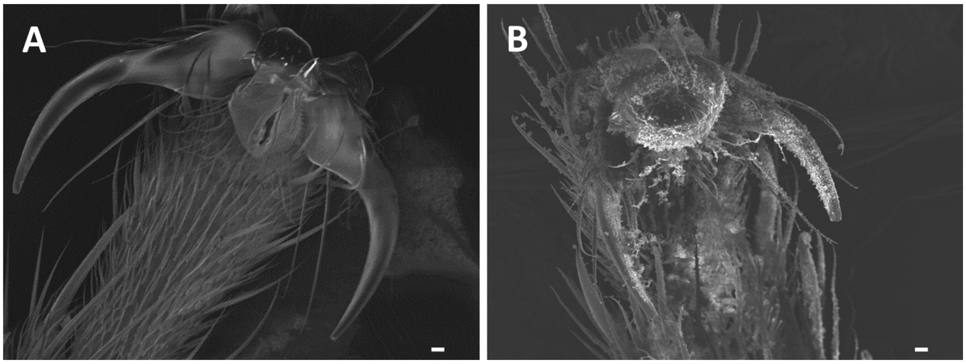

Fig. 4. Scanning electron microscopy images of (A) uncontaminated and (B) contaminated tarsi of *Atta cephalotes* ants after free roaming on 300 nm titanium dioxide particles. Scale bars: 10 μm.

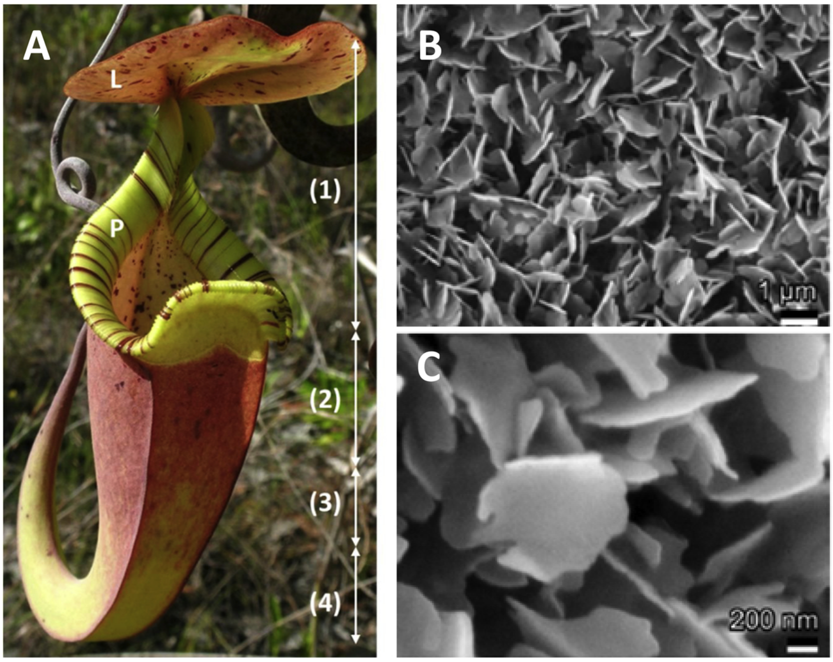

Fig. 5. (A) *Nepenthes* pitcher plant morphology with four functional zones: (1) the lid (L) and peristome (P), (2) the slippery zone, (3) the transitional zone, and (4) the digestive zone. Scanning electron microscopy of the waxy zone: (B) the upper wax layer and (C) wax crystals of the upper wax layer. Adapted with kind permission from [99]. Scale bars: (B) 1 μm and (C) 200 nm.

dissolve, covering the surface in a thick layer of lubricating, slippery fluid; and (4) the epicuticular wax crystals may absorb the secretion, hence reducing the attachment forces of insects (fluid absorption hypothesis).

2.2.2. Slippery when wet plant surfaces

Unlike epicuticular wax-covered surfaces, some other plant surfaces, such as the peristomes of *Nepenthes bicalcarata* pitcher plants, only become slippery to insects when wet, in the presence of dew or rain [97]. The pitcher rim, or peristome, possesses ridges oriented toward the inside of the pitcher [97,105] (Fig. 5A). These surfaces are superhydrophilic (fully wettable), thereby stabilising thin lubricating water films between the insect foot and the surface so that insects slip *via* aquaplaning [97].

When insects climb on the wet peristome, they fall into the pitcher with barely any chance to escape. In contrast to this, dry peristomes are not slippery to insects [98]. In the lower part of the pitcher plant, specialised glands secrete a digestive fluid to absorb nutrients obtained from the insects [88]. In some species, this fluid has non-Newtonian properties and particularly a high extensional viscosity, so that it sticks to the insect's legs, which helps to retain them in the pitcher [106,107].

Due to surface tension forces, getting a grip on the pitcher wall becomes difficult. The pitcher fluid indeed needs to be removed from the insects' adhesive pads to make full contact with the wall, which may be a long process if the fluid is viscous [97]. Friction forces of weaver ants (*Oecophylla smaragdina*) were measured on the peristome surface [97]. The slipperiness of peristomes is a combination between water lubrication and surface topography, which is effective against different attachment structures of the insect tarsus [97,105]. Water films on pitcher rims only prevent adhesion for soft adhesive pads but not for claws, while surface roughness creates friction only for the claws but not for pads [97].

Interestingly, some insects have evolved counteradaptations to climb on slippery plant surfaces, including ants capable of overcoming aquaplaning in *Nepenthes* pitcher plants and "wax-running" ant partners of *Macaranga* ant-plants with slippery waxy stems [87,97,107–109]. In these obligate ant-plant mutualisms, it is both in the ants' and the host plants' interest to isolate the ant partners from predators and competitors, promoting the development of such specific adaptations [107,109].

The knowledge of insect locomotion and naturally slippery surfaces could be used to formulate products preventing crawling insects from entering buildings or adhering to plant surfaces, a reduction in the use of insecticides and the issues they can cause.

3. Coating strategies to minimise insect adhesion

Possible strategies to repel insects or reduce the adhesion of crawling insects to coatings (applied to e.g. buildings or plants) are summarised herein, special attention has been given to the development of substrates bio-inspired by plant surfaces. These approaches might not be suitable for all insects due to the attachment and friction forces discrepancies observed across gender [60,69], pad type and species [12,87,89,97].

3.1. Insecticides and insecticidal coatings

Insect pests are currently controlled mainly by insecticides, which present health and environmental issues, and are summarised in this section.

In the United States only, insects destroy 13% of crops [10]. Hence, one can see the need to eliminate them. Between 2006 and 2007, more than 400 million kilograms insecticides were produced worldwide; while 70% of insecticides were used in agriculture only in the United States [110]. Over the 2008–2012 time period, this number however decreased to 57%, due to an increase of use by private individuals (home and garden) at a similar production level [111].

Since the introduction of synthetic organic insecticides in the 1940s, four major classes of 'conventional' insecticides have emerged: (1) organophosphates, (2) carbamates, (3) synthetic pyrethroids and (4) neonicotinoids [112,113]. [98,99]. Neurochemical insecticides affect the insects' nerve–muscle system and account for 85% of sales [114]. Otherwise, insecticides target respiratory organs (4%) and limit the growth and development of insects (9%), for instance by inhibiting the biosynthesis of chitin or by mimicking juvenile hormones [113,114].

Although beneficial to agriculture, conventional insecticides and pesticides are detrimental to the environment and may affect human and animal health [10,115]. When sprayed, most of the insecticide is lost to the air during application and depending on its persistence and volatility, disperses globally or bioaccumulates in food chains [115–118].

According to Pimentel [119], pesticide use in the United States returns about $4 per $1 invested for pest control. These costs however do not take into account the side economic consequences on health and environment [10]. They were estimated in the USA to be: public health, $1.1 billion per year; pesticide resistance in pests, $1.5 billion; crop losses caused by pesticides, $1.4 billion; bird losses due to pesticides, $2.2 billion; and groundwater contamination, $2.0 billion [10].

Every year, the use of pesticides, including insecticides, leads to 26 million cases of non-fatal poisonings, of which 3 million cases are hospitalised and cause approximately 220 000 deaths [10,120,121]. Insecticides are indeed well-known carcinogen compounds and can cause chronic illnesses, such as neurological damage [10,122]. In addition to humans, insecticides also harm domestic animals.

Upon exposure to insecticides, mutations in insecticide target sites or detoxification processes may occur, resulting in increased resistance to the chemicals [123]. This has been for instance observed in the Australian sheep blowfly, which developed an insecticide-resistant allele [124]. Some insect pests such as the diamondback moth, Colorado potato beetle and cotton aphid have developed resistance to at least 50 insecticides [114].

Insecticides may harm, or even kill non-target organisms (such as organisms that recycle soil nutrients, pollinate crops, and prey on pest species) and reduce and/or contaminate food supplies for animals which feed on them due to bioaccumulation in the food chain [117,118,125]. Using the example of Brazilian tomatoes, insecticides were reported to eliminate more parasitoid natural predators than the targeted tomato pests, due to an increase of their chemical resistance [126], which hence led to a pest population increase. Reduction of pollination also occurs due to honeybee colonies loss [10,125].

It should however be noted that new insecticides with lower health and ecological impact are being developed [125,127], as well as alternatives to lower the amount of insecticides used, e.g. biological control as part of integrated pest management, see e.g. [29].

Novel insecticides based on nanotechnologies and inert materials have arisen to replace the aforementioned conventional insecticides [127,128]. Nanoparticles are particles having at least one dimension smaller than 100 nm; they have grown in attention for the past two decades as their use as encapsulants allows the controlled release of insecticides or pesticides [128,129]. They offer advantages including a higher surface area, higher solubility, higher mobility and lower toxicity due to elimination of organic solvents [116].

Inert materials include diatomaceous earths, zeolites and kaolin, and present a lower environmental impact than conventional insecticides due to their inert nature [127]. Their size is generally comprised between 0.5 μm and 100 μm, improved insecticidal effect has been obtained for particles smaller than 45 μm [127,130,131]. The death of insects is caused by desiccation as these abrasive particles adhere to their cuticle, which normally protects them from water loss [127,130,132]. The particle film technology is a hydrophilic kaolin particle-based coating applied to plants and trees. Particle films were shown to reduce oviposition and adult settling of various pest species on pears and apples [133–135], potatoes [136], olives [137,138] and others [139]. They are however to be used in sunny, dry weather as the hydrophilic and porous particle films are easily washed off by rain [137].

Including insecticides in interior and exterior coatings of buildings (houses, hospitals, restaurants, etc.) can be effective to avoid the presence of insects by repelling, killing or preventing infestation [140–142]. As previously described, the use of insecticides should be limited due to health and environmental concerns [10], although their environmental impact once incorporated in coatings hasn't been assessed in the authors' knowledge. The rest of this section will focus on alternatives to insecticides to reduce the presence of insects in buildings by repelling them or minimising their adhesion to walls using functional coatings.

3.2. *Insect-repellent coatings using natural products*

Essential oils (e.g. eucalyptus or citronella oils) and plant extracts (branches or leaves) consist in an effective natural method to repel insects and present the advantage that they can readily be added to formulated paints [143,144]. Many plants contain chemicals to prevent insects feeding on them, which can be classified in five groups: (1) nitrogen compounds (primarily alkaloids), (2) terpenoids, (3) phenolics, (4) proteinase inhibitors and (5) growth regulators [145]. Most essential oils contain terpenoids, which affect insects in many ways: repellency, acute toxicity, fumigant activity, reproductive toxicity, and neurotoxicity depending on the target site in the insect [146,147]. The efficiency of many essential oils in laboratory conditions, however, lasts only for a few hours [148,149]. Encapsulation allows the slow release of active ingredients and once incorporated into coatings, essential oils repel insects for at least a year [143].

While essential oils and plant extracts can cause contact and airborne allergies (provoking e.g. eczema and asthma) [150,151], their effect once incorporated in paint coatings hasn't been tested to the best of our knowledge, but is expected to be negligible due to encapsulation.

3.3. *Biomimetic strategies*

In this section, we first review the general methods to produce plant bio-inspired properties using synthetic materials, such as superhydrophobicity, like the well-known lotus effect [6], or slipperiness as observed in *Nepenthes* [97,99] and how they can be used to reduce insect adhesion to surface coatings.

3.3.1. *Surface functionalisation and asperities*

A superhydrophobic surface displays a water contact angle greater than 150° and contact angle hysteresis lower than 10° [102]. It is commonly accepted that superhydrophobicity in synthetic surfaces can be achieved by a combination of (1) hydrophobic treatment and (2) surface roughness according to the well-known Wenzel and Cassie models [152,153]. In the Wenzel model (homogeneous wetting), the surface roughness increases the available surface area of the solid [152]. The Cassie model (composite wetting) states that the

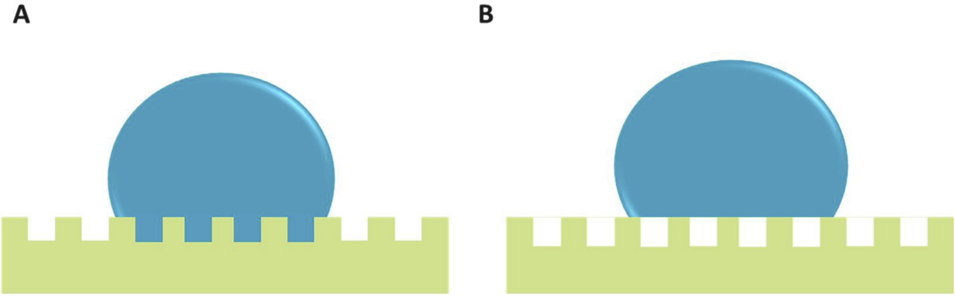

Fig. 6. Schematic (A) Wenzel and (B) Cassie-Baxter wetting models.

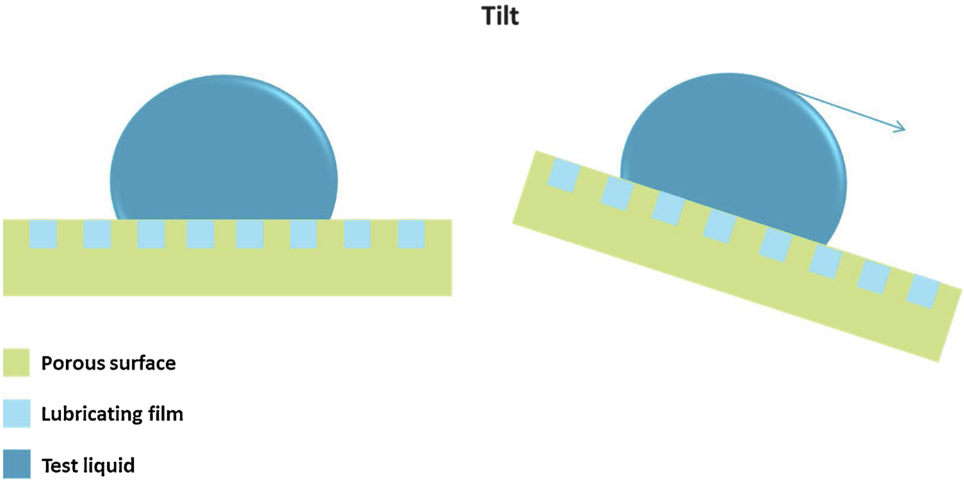

Fig. 7. Schematic Slippery Liquid-Infused Porous Surface (SLIPS) fabrication.

superhydrophobic nature of a rough surface is caused by microscopic pockets of air remaining trapped underneath a liquid droplet, leading to a composite interface (solid-air-liquid) (Fig. 6) [153].

It has been known for decades that surfaces can be rendered hydrophobic by coating them with techniques such as perfluorination or silanisation, and such surfaces have been extensively described in literature [102,154]. The corresponding chemicals indeed bring low surface energy to the top coating. High water contact angles (about 160°) have been measured on functionalised, roughened surfaces [155].

Hydrophobicity properties can be further enhanced by roughening the surface. Methods include nanoparticle functionalisation, which can be e.g. nanosilicas, silicone nanofilaments, carbon nanotubes, plasma treatment, etching, and by producing hierarchical surface structures similar to the asperities found in plants [102,154,156]. The latter can be done using (1) top-down and (2) bottom-up techniques [98]. In (1), the surface to reproduce is replicated by moulding. In (2), structured materials are obtained by chemical self-assembly, from molecular level up to micron scale. These approaches have been extensively described in the literature, see e.g. [154,157,158].

For example, Meredith and coworkers have developed an easy process to create micropatterns bio-inspired from pollen receptive surfaces using blends of commercial polymers [53]. They are formed through demixing of the polymers, and the surface roughness simply increases with the polymer molecular weight. The authors suggest that pollen and stigma surfaces could be used as biomimetic model systems for the design of microparticles [159,160].

3.3.2. Slippery surfaces

Given the importance of surface roughness on insect locomotion (see section II.1 Biomechanics of insects), nano/micro-rough surfaces may provide a mechanical way to control insects [68,99], rather than using potentially harmful chemicals to kill or repel crawling insects.

Several studies suggest that insect attachment on surfaces can be reduced by structuring surfaces with either peaks or valleys [68,89]. Graf et al. [12] designed insect-repellent surfaces by tailoring their topographies following the work of Zhou et al. (presented in the II.1.b Effect of surface roughness on insect climbing section) [89]. The surfaces could reduce the escape rate of cockroach (smooth pads) from a cage by 44%. Interestingly, the adhesion of beetles (hairy pads) was barely affected, probably as the roughness (ca. 45 μm) was in the 'rough' domain, where claw interlocking predominates (Fig. 3). As observed in pitcher plants, a low 'capture rate' could be beneficial to some applications, as scouts recruit more ants to the pitcher [161].

Prototypes of paints made slippery through surface asperities have been studied by Zhou [162]. By tailoring the quantity and size of solid particles at the paint surface, called pigment and extender, paints were produced where insects were unable to find grip. As the solid particles were loosely bound to the paint surfaces (low polymer binder amount), it is also possible that contamination of the pads occurs through transfer of the loose particles to the insect feet. This however hasn't been studied by the author. Another study showed the importance of pigment particle size to render surfaces slippery: when the particles were smaller than 500 nm, the coatings were very slippery to fire ants as their adhesion was reduced [163].

Addition of an extra coating layer (overcoat or overlayer) allows the reduction of properties such as adhesion, fouling or wettability. This is for example done on ship hulls or aircrafts to minimise adhesion of marine organisms and insects, respectively, allowing to reduce drag, and hence, fuel consumption [7,164,165]. The addition of silicones and waxes has been found to improve surface slip by reducing the friction on coating surfaces [166], hence one could imagine this strategy can be efficient to reduce insect adhesion to coatings. The use of low surface energy particles is extremely efficient to do so: in insect colonies, waterborne polytetrafluoroethylene (PTFE, Fluon or Teflon) coatings are used to prevent insects from escaping their cages as Fluon-coated walls are very slippery to insects [13,167]. The surface roughness has however been found to predominantly affect insect adhesion rather than surface energy [86].

Slippery liquid infused porous surfaces (SLIPS) have been developed by Aizenberg and coworkers (see e.g. [14,168,169]) and is a similar approach to the overcoating one. Inspired by the combination of the liquid film found on the *Nepenthes* pitcher plant peristome and its multi-scaled structure, the SLIPS technology involves a porous solid infused by a lubricant (Fig. 7). This lubricating film fills the voids of the substrate, is incompressible and must be immiscible with the test liquid in order to repel it.

The porous substrate is nanostructured, for example made of nanofibres of e.g. epoxy or PTFE. To prepare the SLIPS, the surfaces are first rendered more hydrophobic by silanisation. The perfluorinated lubricant is then added to the substrates to form a liquid overcoat [14]. SLIPS surfaces are stable under high pressure, omniphobic, mechanically robust and self-repairing. Besides repelling many liquids and ice, they can interestingly prevent the adhesion of many biological objects, such as bacteria, fungi and insects.

3.3.3. Particle transfer strategies

Similarly to epicuticular wax crystals detaching from *Nepenthes* plant surfaces to make insects slip, another strategy to minimise insect adhesion to coatings could include the transfer of particles as contaminating particles present on insects' pads or body impede their locomotion [57,90,91].

As discussed in the II.1.c Self-cleaning mechanisms section, the fouling particle size has been demonstrated to significantly impact self-cleaning of the pads of ants, Coccinellids and dock beetles, but did not affect stick insects [92,93]. Particle diameters of 1 μm and larger than 45 μm were easily cleaned from insect pads, but particle diameters of 10–20 μm needed more time to be removed [92,93,96]. The surface energy of the contaminating particles was found to have little influence on loss of adhesion [84]. Interestingly, the recovery of adhesion was shown to be influenced by the fouling particles' surface energy in ants (smooth pads), but not in Coccinellids (hairy pads) [92]. Low surface energy substrates also slowed down the regain of adhesion of contaminated hairy pads [92].

Hackmann et al. [90] have studied the removal of fouling particles by antenna cleaner in *Camponotus rufifemur* ants. They report that small particles (< 25 μm) are harder to clean for insects, as they can interlock and remain longer on their antennae, hence impeding their locomotion.

Anyon et al. approximated the arolium-particle adhesive force, F_{pa}, to be [84,170,171]:

$$F_{pa} = 4\pi\gamma R$$

With γ the surface tension of the fluid secretion, R the radius of the particle asperity in contact with the arolium and corresponding to the particle diameter for spherical particles. For spherical particles typically sized between 1 μm–100 μm and considering $\gamma \approx 30\,\mathrm{mN.m^{-1}}$ [82], insect pads would approximately need to generate adhesion forces in the range of 0.38 μN-38 μN to detach them from a loose particle coating, in line with the attachment forces measured for climbing insects [49,58].

This suggests that coatings made of loose particles, formulated with different particle sizes and surface energies, could for instance only repel one type of insect. In practise, this could be achieved in e.g. paints containing low binder amount (matte paints) or no solvent (powder coatings) [166]. Loose particle barriers have for instance been shown to be effective both in the lab and field conditions to protect potatoes from Colorado potato beetles [15].

4. Concluding remarks

We have reviewed the problems caused by insect pests and strategies to control them. The use of insecticides has detrimental effects, not only for the environment, but also for human and animal health. Informed by the mechanisms of insect adhesion to various surfaces, surfaces are being developed that are slippery for insects, providing an alternative strategy to control pest insects in an environmentally friendlier way. Several parameters have been identified as critical for minimising insect adhesion, including surface energy, surface roughness, and the force required to detach particles from the coatings.

Coatings inspired by slippery plant surfaces may provide an alternative to toxic insecticides. To become commercially feasible, the coating should be inexpensive, easy to make and apply.

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