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Numerical simulation of colloidal self-assembly of super-hydrophobic arachnid cerotegument structures

Alexander É. Filippov[a,b,*], Jonas O. Wolff[c], Michael Seiter[d], Stanislav N. Gorb[a]

[a] Department of Functional Morphology and Biomechanics, Kiel University, Am Botanischen Garten 9, D-24118 Kiel, Germany
[b] Donetsk Institute for Physics and Engineering, National Academy of Sciences of Ukraine, Donetsk, Ukraine
[c] Behavioural Ecology, Department of Biological Sciences, Macquarie University, Sydney, NSW 2109, Australia
[d] Department of Integrative Zoology, University of Vienna, Faculty of Life Science, Althanstrasse 14, 1090 Vienna, Austria

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ABSTRACT

Certain arachnids exhibit complex coatings of their exoskeleton, consisting of globular structures with complex surface features. This, so-called, cerotegument is formed by a multi-component colloidal secretion that self-assembles and cures on the body surface, and leads to high water repellency. Previous ultrastructural studies revealed the involvement of different glandular cells that contribute different components to the secretion mixture, but the overall process of self-assembly into the complex regular structures observed remained highly unclear. Here we study this process from a theoretical point of view, starting from the so-called Tammes-problem. We show that slight changes of simple parameters lead to a variety of morphologies that are highly similar to the ones observed in the species specific cerotegument structures of whip-spiders. These results are not only important for our understanding of the formation of globular hierarchical structures in nature, but also for the fabrication of novel surface coatings by colloidal lithography.

1. Background

Surface modification and functionalization through nanostructures is a versatile way of producing desirable properties on various materials. However, it is not a novel innovation, since it is deployed by nature for a long time. Well-known examples are super-hydrophobic surfaces that gain this feat by a nano-structuring of the surface, often by a coating of crystals or particles (Barthlott et al., 1997, 2016; Rakitov and Gorb, 2013). Some arachnids, such as the cryptic, large whip-spiders (Amblypygi) and some mites, exhibit a crust of dried secretion containing globular micro-structures covered with regularly arranged nano-particles (Alberti et al., 1981; Raspotnig and Matischek, 2010; Wolff et al., 2016, 2017). In the whip-spiders these are built from a multi-phasic secretion, emerging from two distinct types of glandular apparatuses (Wolff et al., 2016). Within this relatively small arachnid group (216 known species) a high diversity of structures has evolved, that seems to base on differences in structure and properties of the basic colloidal particles (Wolff et al., 2016, 2017). Wolff et al. (2016) have identified six clusters of related morphotypes, which correlate with the hypothesized inter-familiar relationships of whip-spiders (Weygoldt, 2000).

In material science, surface coatings with regularly arranged bi-hierarchical globular particles have been produced by colloidal lithography, a technique utilizing the self-assembly of nano-particles on a substrate surface (Yang et al., 2006; Badge et al., 2013). However, the range of producible patterns and properties of these coatings are still restricted, as well as their durability. Therefore, the study of the versatile whip-spider cerotegument seems promising for generating new solutions.

Water repellence and ultrastructure of granules are collected in Figs. 1–3. All the figures are organized in the same manner and differ only by in different species. The figures for three different groups of granules are separated for better magnification and readability of the images. In each Fig. 1st row shows tap water droplets, directly ejected on the carapace, having a nearly spherical shape. 2nd row presents fine structure of species-specific granules and 3rd row reproduces ultrastructure of colloid particles. Besides, mutual correspondence with numerically found structures and natural ones are marked by the numbers used in the text below to numerate them in Fig. 4 depending on the interactions responsible for the appearance of these structures.

In order to gain a better understanding of the process of self-assembly and self-arrangement of nano-particles on spherical microstructures, we here apply a theoretical approach. We introduce a numerical model that allows us to test the effect of different interaction properties between the particles on the morphology

* Corresponding author.
E-mail address: filippov_ae@yahoo.com (A.É. Filippov).

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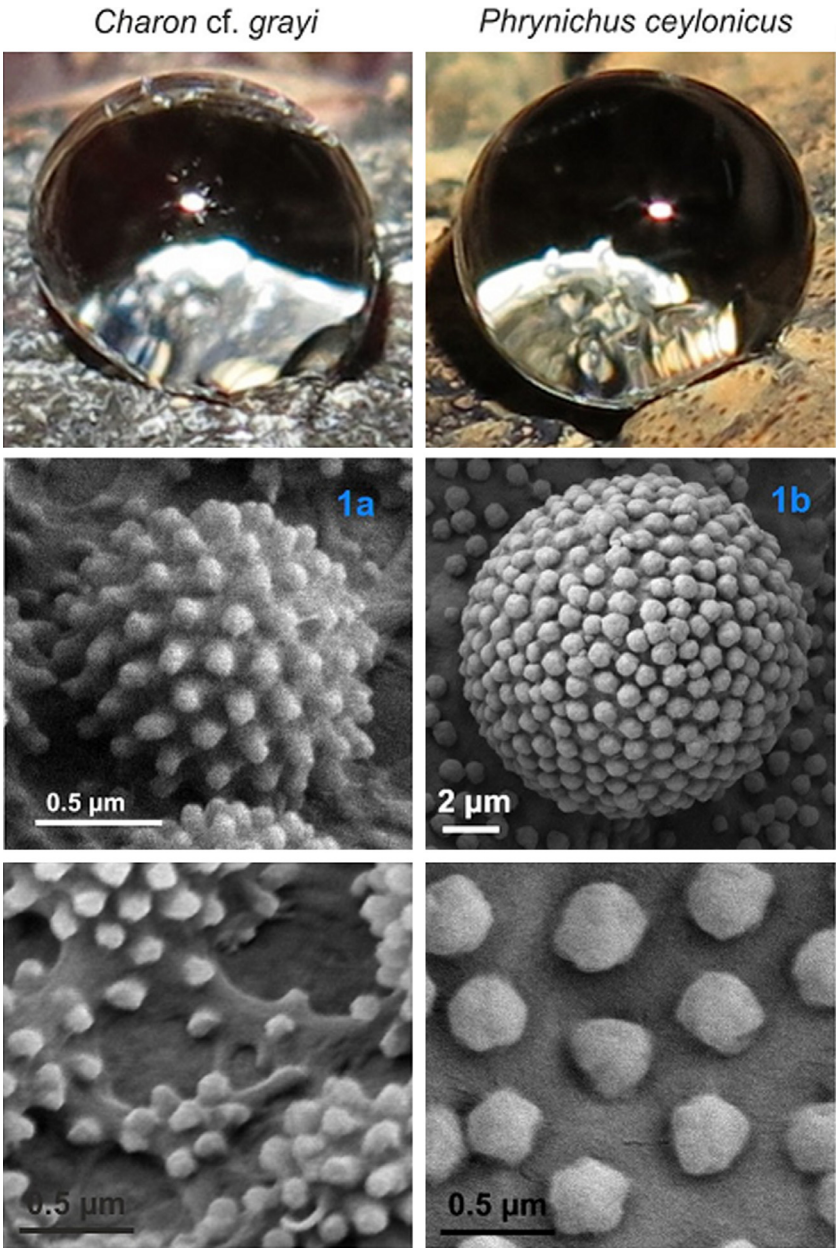

Fig. 1. Water repellence and ultrastructure of granules in *Charon cf. grayi* and *Phrynichus ceylonics*. 1st row: Tap water droplets, directly ejected on the carapace, form a nearly spherical shape. 2nd row: Fine structure of species-specific granules. Mutual correspondence with numerically found structures and natural ones are marked by the numbers used below to numerate them in Fig. 4 depending on the interactions responsible for the appearance of these structures. 3rd row: Ultrastructure of colloid particles.

of the eventual structure. This issue is related to the so-called Tammes problem, seeking for the most optimal packing of a given number of pores or particles on a sphere with maximized distance between them (Tammes, 1930; Tarnai and Gáspár, 1987; Erber and Hockney, 1991).

The aim was to alter the model parameters in a way that structures similar to those observed in whip-spiders could be reproduced. This way we wanted to test which alterations are necessary to transform the structures within a possible evolutionary scenario, and likewise how colloids can be tuned to tailor functionalized surfaces by colloidal lithography.

2. Numerical simulation

The model of the systems under consideration is organized as a combination of discrete and continuous approaches. From a general (physical, chemical and biological) point of view one can expect that the observed structures appear in some kinetic process during which continuously an initially more or less uniformly distributed substance redistributes and solidifies in a 3-dimensional space.

Such a process can be observed in many different chemical and physical systems (including such underlying processes in biological systems). For example, it can happen during superconducting (Geim et al., 1997) or magnetic ordering (Filippov, 1997) in quasi 2-dimensional systems, growth of surface structures (Filippov, 1998; Kovalev et al., 2016), and so on. In all the cases, the process involves an interaction of many spatially distributed densities and its direct simulation can cause extremely time consuming calculations.

In the particular case of the cerotegument, the problem is complicated by the specific topology of the surface, on which the process runs, because the redistribution of the densities and their further solidification take place on spheres. Here, one has to deal with a kind of phase transition (or phase separation) inside a