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NATURE-INSPIRED PILLAR-TYPE SMART MULTIPURPOSE COATINGS

NATALYA KIZILOVA[†*], SIGNE KJELSTRUP[†] AND ZHILIANG ZHANG[†]

[†]Norwegian University of Science and Technology (NTNU)
PoreLab, S.P. Andersens vei 15b, 7031 Trondheim, Norway
e-mail: signe.kjelstrup@ntnu.no

[*] V.N. Karazin Kharkiv National University
Svobody sq., 4, 61022 Kharkiv, Ukraine
e-mail: n.kizilova@gmail.com

[†] Norwegian University of Science and Technology (NTNU)
Nanomechanical Lab, Department of Structural Engineering
Richard Birkelands vei 1A, 7034 Trondheim, Norway
email: zhiliang.zhang@ntnu.no

Abstract. Design and composition of smart multipurpose coatings can be taken from Nature. Many plants and animals have very complex structures from macro to micro/nanoscales with superior properties like superhydrophobicity and self-cleaning, high/low adhesion/reflection, drag reduction, air/water accumulation, and others. The topic was intensively studied in the literature. Recent successes in material sciences and technologies give more possibilities for precise small scale engineering. This has motivated us to do a systematic comparative analysis of geometric principles and superior physical and chemical properties of natural surfaces. Here we give a brief review of the variety of superior physical properties, geometric and material design of the most promising natural coatings. A variety of structures with similar physical properties is presented. The mechanisms of the multipurpose function of the same coating are discussed. Special attention is paid to the sophisticated structure of salvinia leaves and coatings manufactured based on this design. The interconnection of physical mechanisms at macro-, micro- and nano-scales is discussed. A general approach to the surface energy as a function of material composition, micro- and nano-scale roughness, is proposed for physical characterization of the hydrophobic and anti-icing abilities of natural and engineered surfaces and coatings.

Key words: Nature-inspired, Smart Surfaces, Pillar-Type Coating, Superhydrophobic, Salvinia Leaf, Surface Energy.

INTRODUCTION

Smart materials and coatings with superior properties like high strength and durability, superhydrophobic and superlyophobic, antiadhesive and antireflective, as well as many others

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are of a significant interest for different applications. During the evolution many plants and animals developed such properties for their better function and survival. Among other physical properties, an ability of natural surfaces to repel water droplets together with dust particles (self-cleaning), and prevent icing at low ambient temperatures (icephobicity) is widespread in nature, and their artificial analogies (biomimetics) are very important for numerous civil, military and industrial applications. The term superhydrophobic (SHb) or ultrahydrophobic is used for surfaces which have contact angle (CA) with water droplets >150°, while superhydrophilic properties (high adsorption towards water) are characterized by CA<5-10°. Similar definitions are accepted for the superlyophilicity/phobicity (low/high adsorption to oils). According to Scopus statistics, only a few papers on superhydrophobicity were published in 1980-ies (Fig.1). A special interest for the topic appeared in connection with the description of the lotus leaf effect and the self-cleaning ability of this plant [1,2]. The SHb properties have been also detected on many natural surfaces like rice leaf [3-5], rose petals [6], peanut leaf [7], insect eyes [8], butterfly wings [9], legs and exoskeleton of spiders [10], gecko feet [11], fish skin [12], and others. Significant interest for the topic is shown by the increasing number of publications dedicated to theoretical and experimental studies of nature-inspired and biomimetic engineered coatings (Fig.1).

Recently, a series of comprehensive reviews and books on properties, manufacturing and applications of bioinspired surfaces have been published [13-19]. Nevertheless, the interest for nature-inspired materials is rising exponentially, as progressive discoveries in material science, - technologies and measurement techniques allow for elaboration of more advanced smart coatings for multipurpose applications.

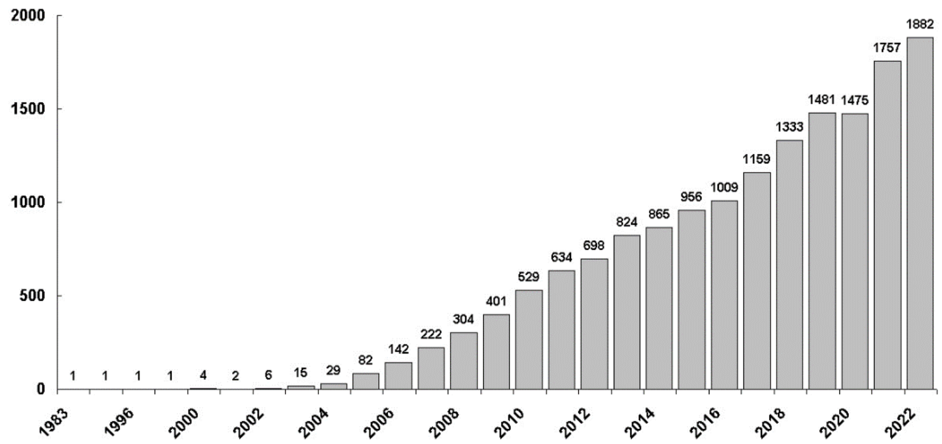


Figure 1: The number of papers published in Scopus with keywords “superhydrophobic surface”

A common feature of the natural surfaces with superior properties, is the combination of geometric complexity at different scales (macro- to micro- and nano-scales) with the use of different materials at the corresponding scales. The most typical examples of combinations of different physical properties in natural surfaces are summarized in Table 1. The self-cleaning properties are determined by superhydrophobicity and a very low tilt angle, needed for rolling away the water droplets together with dirt particles (low rolling angle). This evolutionary trait is important for efficient photosynthesis in muddy ponds and swamps. The branched wax nanocrystals on the lotus leaf serve as an air-retaining layer between a droplet and a structure that provides its superior physical properties.

A combination of SHb and high adhesion is important for survival in arid and semi-arid regions characterized by high temperatures and low rainfall. The sliding behavior of water droplets on SHb surfaces depends on the length and continuity of the three-phase (solid/liquid/gas) contact line (CL) [9]. On the peanut leaf surface the CL exhibits a quasi-continuous behavior at the micro-scale, and discontinuous characteristics at the nano-scale, while on the lotus leaf surface the CL is discontinuous. One may speculate that lotus and peanut have evolved different optimized solutions to keep wetting for higher efficiency of breathing and photosynthesis and survival.

An ability to repel water and retain a thin film of air along the leaf surface or accumulate it in the air-pockets seems very important for underwater photosynthesis in rice, salvinia and other plants inhabiting ponds, lakes, marshes or water covered soils. Salvinia leaves are covered by elastic trichomes with a spherical crown with a complex nanostructure of the hydrophobic wax nanocrystals that are produced by the epithelial cells. The very tips of the pillars are formed by dead cells that do not produce wax and remain hydrophilic [20,21].

A combination of SHb and hydrophilic surfaces, used for water collection from fog-laden wind, can be found in the desert beetles [22] and spider silk webs [23]. The overwings of such spiders has peaks and troughs covered by wax, while the tops of peaks remain wax-free [22]. A similar ability by spider webs are provided by the specific structure of the spider silk, which is composed by periodic spindle-knots made of random nanofibers with high water adhesion properties and separated by joints made of aligned nanofibers [23]. This structure results in a surface energy gradient *($\nabla\gamma$)* [24,25] between the knots and the joints and in a difference in Laplace pressure (δP) [26-28], and interaction of both factors results in continuous condensation and directional collection of water droplets of submillimetre-size around the knots.

The cacti have evolved some complex structures for high efficient water absorption from air in the arid zones. Their stems are covered by well-distributed clusters of spines and trichomes, and each spine growing from the trichomes has an acute angle at its tip and oriented barbs followed by narrower and wider gradient grooves and a base with belt-structured trichomes. The geometry of the tip and the barbs provide a gradient in $\nabla\gamma$ from the tip to the base along the spine. The gradient in roughness of the microgrooves contributes to a gradient in the surface-free energy along the spine. That amplifies the driving force for water collection and movement towards the cacti blade [29]. Again, the spines are multifunctional and provide also light interception and small water loss [30,31].

The compound eyes of mosquitos, flies, moths and other insects exhibit excellent water-repelling ability and high SHb stability. This is attributed to a hierarchical structure at the micro- and nano-scales with the use of the low-surface-energy natural material chitin [32,33]. The eye surface consists of an array of 200-300 νm sized pillars forming a sub-wavelength system, that simultaneously reduce the reflection of light and enhance night vision capability. Its combination with salvinia-type structure produces a wider set of superior properties [34].

The butterfly wings have different complex structures on their surfaces including the rough micro-scale structures of ribs and grooves, hairs, and cloth-like micro-sculpture [9,35,36]. The complex fractal-type cross-section of the ribs provides efficient adsorption of sunlight, while hairs and surface nano-sculpture provides water repelling properties with directional adhesion. Water droplets easily roll off the surface of the wings along the radial outward direction of the central axis of the body [35]. This complex structure is also multifunctional with high icephobic ability [37]. Besides, the space between the upper and lower laminae of the microstructure provides additional ventilation and drag reduction during the flight. The translucent wings without scales are also SHb due to other types of micro and nanoscale structures [38]. Therefore, the same set of physical properties in the wings of insects can be provided by fractal, setae or layered cuticle structures with $CA=54\text{-}168°$ [39].

The scales of fish provide a variety of physical properties from SHb to lyophilicity in air and superlyophobicity in water. This can be attributed to the riblets with micro- and nano-structures on their surfaces and to liquid mucus [40,41].

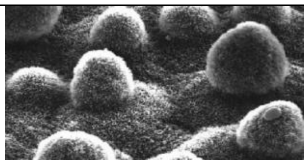
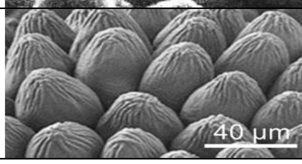
Therefore, different superior physical properties of the surfaces of many plant leaves and petals, exoskeletons and eyes of insects are provided by a combination of natural materials (wax, chitin, chitosan, fat), macrostructures (ribs, grooves, wrinkles) of the surface, its microstructure (riblets, barbs, haits, trichomes) with covering nanostructure (nanocristals, papillas, branshed structures). The resulting surface is a thin multiscale porous structure with lightweight design and a non-uniform distribution of different, sometimes opposite physical properties like hydrophilicity and hydrophobicity. Some examples of the type and dimensions of the corresponding micro and nano structures and their influence of the surface energy γ and the CA with water droplets are presented in Table 2.

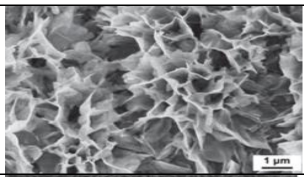
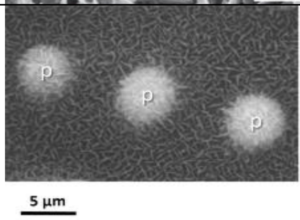
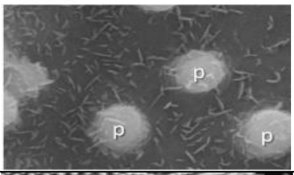
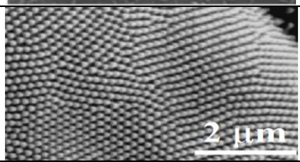
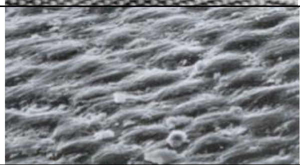
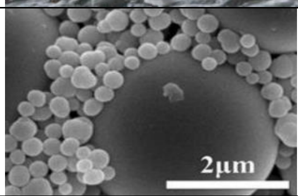
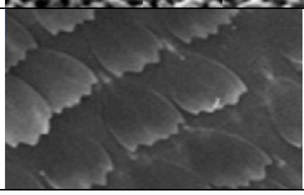
Table 1: Superior physical properties of natural surfaces

N	Name	Properties	Mechanism	Useful properties for applications
1	Lotus leaf	SHb and super low adhesion to water with very low sliding angle	Plicate leaf surface covered by hydrophobic epicuticular wax and multiscale structures with randomly distributed micropapillae covered by branched nanostructures [1,2]	Self-cleaning, anti-corrosion, anti-icing
2	Rice leaf	SHb and gas film retaining	Surface papillae and epicuticular wax platelets on the adaxial surface of leaf blades [5]	Gas retention and accumulation under water
3	Rose petal	SHb and high adhesivity to water	SHb leaf covered by periodic array of micropapillae with nanofolds on their tops for better water impregnation [6]	Functional materials with unique dynamic wetting behavior; patterned paper surface with anisotropic or tunable adhesion
4	Peanut leaf	SHb and high adhesion towards water	Hill-like microstructures with papillae nanostructures and quasi-square grids with	Fog accumulation, water collection, self-irrigation

			separated ridges [7]	
5	Salvinia leaf	SHb and hydrophilic	SHb surface papillae with a specific geometry and hydrophilic crown at the top [20,21]	Gas retention and accumulation under water; water-oil separation; cleanup oil spills in oceans; drag reduction; water microdroplet manipulation
6	Desert beetles	SHb and superwettable	SHb surface peaks and troughs covered by wax with wax-free tops of the peaks [22]	Vapor extraction from air and drinking water collection
7	Spider silk	High water absorption and directional movement	A thread composed as an array of loose knots of hygroscopic nanofibers connected by aligned nanofibers [23]	Vapor extraction from air and drinking water collection
8	Cactus spines	High water absorption and directional movement	Clusters of spines and trichomes with oriented barbs, gradient grooves and the base with belt-structured trichomes [28]	Fog collection, water loss prevention
9	Compound eye of insects	SHb and high water adhesion	An array of regularly-spaced nanoscale bumps covers the domed surface of the hexagonally-shaped microscale [8,32,33]	Water repelling, anti-fogging, antireflective, self-cleaning, antimicrobal
10	Butterfly wing	SHb, icephobic,	Scales with upper and lower laminae, trabeculae and cross ribs [9]	Water repelling, icephobic, light absorption, drag reduction, ventilation
11	Fish skin	SHb, superlyo-philic/phobic in air/water	Scales with macro/microscopic riblets with nanostructure and viscous liquid mucus [12]	Oil repelling in water, icephobic, drag reduction

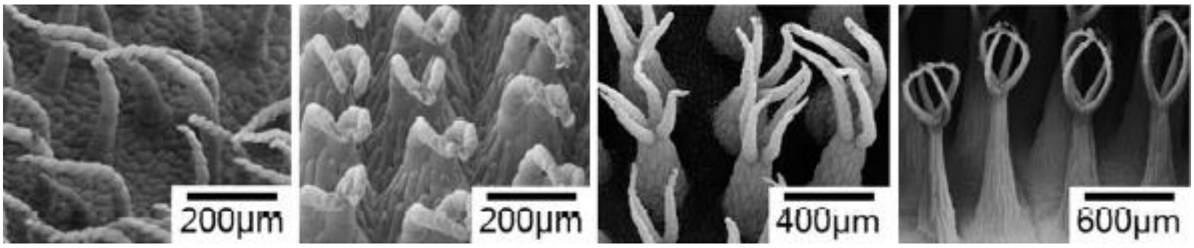
Table 2: Micro- and nanostructure of natural hydrophobic surfaces

N		Microstructure image	CA (°)	Microstructure size (μm)	Nanostructure size (vm)	Ref.
1	Lotus leaf		161	5-9	120-130	[42]
2	Rose petals		150-167	35-42	250-300	[43]

3	Peanut leaf		>150	25-75	100-500	[7]
4	Rice leaf a) normal; b) mutant	 	143-144 62-133	4-5	100-350	[5]
5	Butterfly compound eye		3-7	20-40	100-200	[8]
6	Desert beetle over wings		70-92	500	400-900	[44]
7	Mosquito eye		>160	5-6	200-350	[32]
8	Cicada wing		165	80-120	50-75	[39]
9	Butterfly wing		165	50-75	40-350	[39]

2 SALVINIA LEAF MORPHOLOGY AND PHYSICAL PROPERTIES

Salvinia is a group of floating aquatic ferns inhabiting tropical and subtropical lakes and ponds with fresh water. This plant has become very popular in biomimetic applications due to the superhydrophobic, but not self-cleaning, properties of its leaves [20]. The leaf surface of salvinia is covered by small hydrophobic trichomes which could have a single curved pillar shape (*Salvinia Cucullata*), a stalk with two curved pillars with a hydrophilic crown (*Salvinia Oblongifolia*), a stalk with four curved pillars with unlocked tips (*Salvinia Natans*) or locked by a crown of common dead cells (*Salvinia Molesta*) (Fig.2 a-d accordingly).



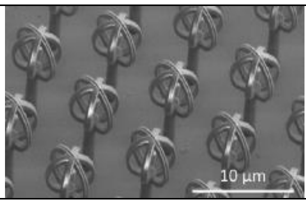
a b c d

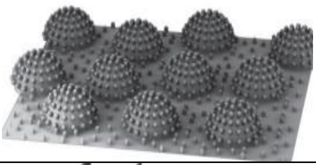
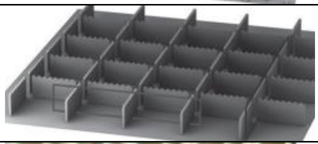
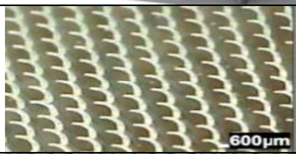
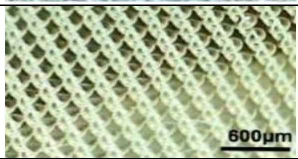
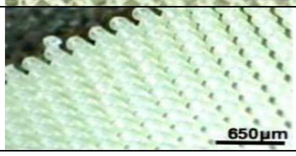
Figure 2: SEM images of microstructure of *Salvinia Cucullata* (a), *Salvinia Oblongifolia* (b), *Salvinia Natans* (c), *and Salvinia Molesta* (d) leaves (adapted from [45]).

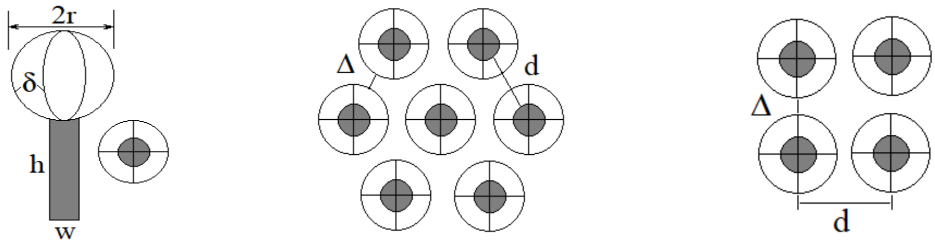
The surface of Salvinia leaves is superhydrophobic, but not self-cleaning; the water droplet does not roll off the leaf; rather it is attached to the hydrophilic crowns. The trichomes form hydrophobic air-pockets that can stay underwater filled with air for several days [46]. The elasticity of the trichomes is of great importance for the stability of the air layer under fluctuating pressure situations. These produce variations in the air layer thickness to which the eggbeater-shaped hairs can adjust, either by bending or by compression and expansion. The layer of trichomes provides the air-layer stability against external pressure variations by using the entrapped air volume as an elastic spring.

A series of bioinspired coatings has been engineered and tested for surface energy values. Some results are summarized in Table 3. Fur-type (Fig.2a) polymer structures were fabricated by hot pulling [47]. The egg-beater structures were fabricated by laser lithography, 2-photon polymerization, and 3D printing [48,49]. The measured CA values depended on the material, dimensions (Fig.3a) and location (Fig.3b) of the trichomes on the surface. Both Wenzel (Fig.4a), Cassie-Baxter (Fig.4b) and mixed W/CB (Fig.4c) and CB/W (Fig.4d) states were observed depending on the dimensions *{r,δ,d}* [48].

Table 3: Artificial microstructures based on salvinia-inspired, lotus and peanut leaf design

N	Microstructure image	Matertial and manufacture	CA (°)	Microstructure geometry (µm)	Ref.
1	 	Epoxy-based photoresist (HPl); 3D laser lithorgaphy	122	h=7;w=1.5; r=3; δ=1; d=9; Δ=3; n=3	[48]

2		Lotus leaf-based	>150		[7]
3		Peanut-based	>150		[7]
4		ultra high resolution resin with a layer of self-	Before 72.3 After 142.7		[49]
5		assembled monolayer	Before 93.1 After 146.4		[49]
6		Digital light projection 3D printing	Before 109.7 After 151.1		[49]
7		The same	Before 120.8 After 171.3		[49]



a b c

Figure 3: Side and top view of a trichoma (a) with hexagonal (b) and square (c) location on the surface

a b

c d

Figure 4: Water droplet on a salvinia-type surface in Wenzel (a), Cassie-Baxter (b), Wenzel/Cassie-Baxter (c) and Cassie-Baxter/Wenzel (d) states

3 MECHANISMS OF SUPERHYDROPHOBICITY OF NATURAL SURFACES

Experimental studies of the water CA on natural and artificial coatings of different types revealed a significant influence of height and width of the macroscopic structures, as well as size and distribution density of the nanostructures over them. The distance between the pillars was a critical parameter in the determination of a transition from W to CB states. According to Young's formula, the value of the CA, θ, is determined by the force balance that gives the relation between surface energies of the solid-vapor (γ_{sv}), the solid-liquid (γ_{sl}), and the liquid-vapor (γ_{lv}) interfaces as

$$cos(\theta)= r(\gamma_{sv} - \gamma_{sl})/ \gamma_{lv}, \tag{1}$$

where r is the surface roughness;

In the CB state eq.(1) is used with $r=1$, but according to the thermodynamic theory of small systems [50], the function $r=r(p_f,<d>,D_s)$ is determined by the surface packing factor p_f, mean distance $<d>$ and shape factor D_s. These properties will change eq. (1). In the presence of the nanostructure, Young's law (1) can be generalized, using the grand potential formulation based on nanothermodynamic theory [50]

$$\Xi = \hat{\pi}\varsigma = \sum_{\varphi}\Pi^{\varphi}\varsigma^{\varphi} - \sum_{\varphi}\gamma^{\varphi}\Sigma^{\varphi} - \sum_{\varphi}\Gamma^{\varphi}\Lambda^{\varphi} - \sum_{\varphi}\nu^{\varphi}v^{\varphi} , \tag{2}$$

where $\hat{p}$ is the effective pressure, and $\Pi, \varsigma, \gamma, \Sigma, \Gamma, \Lambda, \nu, v$ are the pressure, volume, surface tension and area, line tension and length point tension for the number of corners, $j=\{s,l,v\}$.

Theoretical results have been validated using the empirical relationship

$$cos(\theta)=F(p_f, <d>, D_s, \gamma_{lv} ,\gamma_{sl}), \tag{3}$$

computed for materials and geometrical variables of the engineered system (cf. Table 3) and of natural surfaces (Table 2) with known CA, as measured for different droplet volumes. The results allow generalization of the Young and Laplace law for surfaces that possess micro and nanostructures.

11 CONCLUSIONS

A detailed review on the superior physical properties of natural surfaces is given and the main factors determining the multifunctional properties are summarized. It is shown, that a combination of a specific microstructure with a nanostructure is used in natural materials to amplify hydrophobic properties of uniform materials and make them SHb and ice-phobic. Engineered surfaces, based on a combination of the lotus-leaf, insect eye, and salvinia leaf design seem beneficial when compared to surfaces based on a single design.

Using a nanothermodynamics description, we may optimize pillar-type surfaces to give the highest CA. The surface energy of a complex surface can then be expressed as a function of material (i.e. CA of a smooth uniform surface) and its structure in the terms of the surface packing and shape factors, and surface porosity.

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