**Hydrophilic and hydrophobic materials and their applications**

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# Abstract

Wettability of a material’s surface plays a significant role in how fluids interact with such surfaces. Wetting behaviour is universal but can vary depending on the chemical nature of the solid and liquid phases. Plants and animals adapt to their environment through having evolved special properties. These properties are such as hydrophilic and hydrophobic. Hydrophilic surface has a strong affinity to water and spreading of water on such surface is preferred. The degree of hydrophilicity of the substance can be measured by measuring the contact angle between the liquid and solid phases. Hydrophobic materials are known as non-polar materials with a low affinity to water, which makes them water repelling. A contact angle of less than 90° indicates hydrophilic interaction where as an angle greater than 90° indicates a hydrophobic interaction. More recently, superwetting such as superhydrophilicity has been receiving an increased focus in literature due to its potential significance. Superhydrophilic surface has a contact angle of less than 5°.

The fabrication of hydrophilic materials can be carried out in two main ways: depositing molecules on surfaces or modification of surface chemistry. Both methods have been successful historically in achieving their intended purposes. Hydrophobic and superhydrophobic materials can be produced with many fabrication methods such as layer-by-layer assembly, laser process, the solution-immersion method, sol-gen techniques, chemical etching and Hummer’s method.

The applications of such an important property are significant. For example, hydrophilic surfaces can be used in anti-fogging applications, biomedical, filtration, heat pipes, and many others. Hydrophobic and superhydrophobic materials have been successfully applied in many sectors, such as: (I) the removal of petroleum from aqueous solutions, (II) applied to plastic, ceramics and mesh to contribute to the oil removal from aqueous solutions, (III) hydrophobic layers have a strong self-cleaning effect on plastics, heat pipes, metals, textiles, glass, paints and electronics, (IV) hydrophobic layers improve the anti-freezing behaviour of heat pipes which prevents unwanted build-up and (V) they function as a water and dust protecting coat on electronics

The presence of this property is historic but there is still a huge potential for development for its applications in many sectors such as water treatment, heat transfer applications, biomedical devices, and many more.

*Keywords: Hydrophilic, hydrophobic, superhydrophilic, superhydrophobic, water droplet angle, hysteresis.*

|  |  |  |  |
| --- | --- | --- | --- |
| Nomenclature | |  |  |
| SHO | Super hydrophobic | ED | Electro dialysis |
| LBL | Layer-by-layer | MD | Membrane distillation |
| CAH | Contact angle hysteresis |  |  |
| PU | Polyurethane | *Variable definition* | |
| PE | Polyethylene | θ | Angle |
| TEFLON | Polytetrafluoroethylene | s | Solid |
| RO | Reverse osmosis | l | Liquid |
| MSF | Multi-stage flash distillation | v | Vapour |

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# 1. Introduction

Wettability of a material’s surface plays a vital role in how fluids interact with such surfaces. Wetting behaviour is universal but varies in a rather peculiar nature. Plants and animals adapt to their environment through having evolved special wettability characteristics, showing distinctive wetting and non-wetting properties [1–5]. A Lotus leaf exhibits the “never wetting” or “ultra-ever dry” property. In a non-wetting situation, water can roll around freely on the leaf's outer surface. The water droplet exhibits an almost spherical shape enabling this rolling behaviour. This introduces a self-cleaning property, as the contaminants can be removed by the rolling drop [6]. A similar water rolling behaviour is also observed in rice leaves. They display anisotropic non-wetting state, which enables an easier rolling of the water droplets due to the longitudinal direction rather than the perpendicular direction on the surface [7].

Wetting behaviour is also very important for animals and it has a direct impact on their survival. Some examples of mammals which incorporate this characteristic include desert lizards which make use of scale hinges to transport drinking water [8]. Capillary ratchet plays an important role with the wetting properties of Shorebird’s beaks in order to move water drops from the tip of the beak to the mouth [9]. Water can then be transported in one direction, continuously on the surface of the ‘peristome’ of *Nepenthes alata* with no energy input [10], and certain mammals, such as Namib desert beetles, have adapted themselves to survive in very dry environment primarily due to their ability to collect water from humid air by hydrophobic and hydrophilic phase-to-phase patches on their backs [11]. These animals have only been able to survive due to the unique characteristics of their surfaces. The unique surfaces observed in nature allows scientists to artificially fabricate and investigate such phenomena.

The terms “hydrophilic surface” and “hydrophobic surface” have been used extensively in literature for many years, which describe opposite effects of the behaviour of water on a solid surface. A hydrophilic surface has strong affinity to water molecules, whereas hydrophobic surfaces repel water [12].

A primary indicator that is used to classify if the wetting behaviour of fluid is hydrophilic or hydrophobic is the measurement of the contact angle. The surface contact angle is the angle formed at the intersection of the liquid-solid and liquid-vapour interface. Depending on the water droplet contact angle, the material is classified as either: hydrophilic (θe < 90˚), hydrophobic (90˚< θe < 150˚) or superhydrophobic (θe > 150˚). The higher the contact angle is, the bigger the strength of the liquid-liquid interaction becomes, making the material more hydrophobic [16–18]. The contact angle range for each wetting and non-wetting scenario is shown in Figure 1.

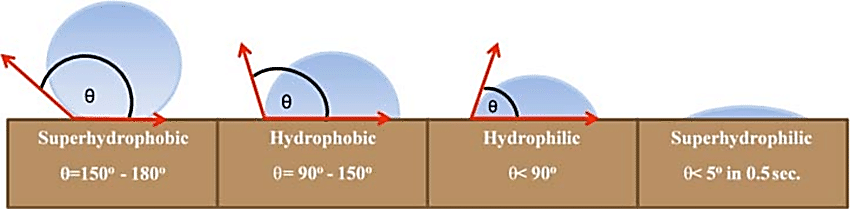


Figure 1. Schematic of water droplet's contact angle with different surface wettability [13].

Surface treatment technology has many different applications as it can be applied in medical, and technological ways [14,15]. To alter the wettability characteristics of any material, certain liquids can be, very simply, applied as coatings to various types of materials, and thereby change the wetting ability these materials. Treated surfaces can be classified into three different categories depending on their water droplet contact angle (θe), as discussed previously.

Biomimetic of such surfaces of animals have always been the way to create new technologies and enhance people's lives. Hydrophilic and hydrophobic materials play a crucial role in oil-water separation in environmental and energy aspects [19]. Active membranes can be used to achieve sea water desalination, energy conversion and fuel cell processes [20]. Wetting and spreading characteristics found in nature is an area of increased interest due to the wide applications that this phenomenon could be applied to. Development opportunities of wide range of products can be exploited at industrial level.

To enable to implement wetting and spreading of liquids onto surfaces, Young’s equation was introduced in 1805 [21]. The initial wetting models, Young's equation, have been altered to describe and explain various wetting conditions. Dynamic and static wetting have both been explored due to their applicability in different settings. Many concepts, such as contact angle hysteresis, tri-phase contact line theory, have been proposed [22–24]. Chemical composition of the surface as well as the micro-scale topography are the two main parameters that determine the liquid’s wettability. Technological development and advances along with scientific research have unitized high speed video imaging and high-resolution microscopes enabling wettability phenomena to be observed. 3D printing and micro-fabrication enabled the design and manufacturing of artificially wet materials.

**Objectives**

Hydrophilic and hydrophobic properties are rather well-known and these are applied in many different industries. This article is a comprehensive state-of-the-art on hydrophilic and hydrophobic materials. The focus of this article will be on the fabrication and characteristics of hydrophilic, hydrophobic and superhydrophilic and superhydrophobic materials and reviewing the applications of these materials in many domains, such as petroleum, plastic, heat pipes, other metal applications, textiles, glass, ceramics, mesh, paint, electronics and miscellaneous materials. The objectives of this article are (I) to compile findings on this subject that are applied in different industries and (II) to draw conclusions as well as giving recommendations for further investigations.

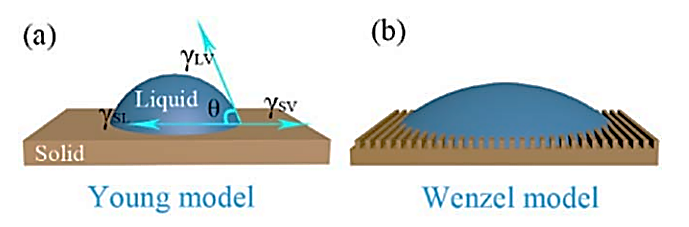
# 2. Theories of wetting and spreading

The importance of hydrophilic and hydrophobic characteristics extend into many disciplines such as chemistry, physics and engineering. Determining the wetting capacity of a liquid onto a surface is determined by the surface and interfacial interactions between the two media, and extensive research have been carried out in this field to examine the forces influencing the wetting and spreading behaviour patterns. Forces such as van der Waals, electrostatic forces and other intermolecular forces dictate the wetting capability of a liquid. Many techniques have been developed to give insights into the wetting and spreading behaviour. Such techniques include atomic force microscopy, surface force apparatus, and molecular dynamics [3,4,25]. In 1805, Thomas Young had established the first laws for flat, solid surface that are chemically homogeneous. Specific energy at the interface between the liquid, gaseous and or solid phases is described as the surface tension. The condensed phase is denoted by γIJ. Subscripts of the γIJ are the interfaces between the solid (s), liquid (l), and vapor (v). Liquid/vapor surface energy can be denoted as γ simply. When liquid drops are placed in contact with a solid surface, they tend to reach equilibrium state with lowest energy. The equilibrium contact angle (θeq) measured at the edge of the drop on the solid surface (shown in Figure 2a) can be described by the Young Eq. (1): d

Fabrication processes may generate undesired characteristics of the surface such as pore, striations, microgrooves or irregular convex. To determine the contact angle of a rough surface, the Wenzel model was developed and used since 1936. The Wenzel model states that the liquid is in contact with the entire solid surface and completely penetrates into cavities (as shown in Figure 2b). To describe this model mathematically Young’s equation was modified as follows [22]:

=

where, r is the roughness factor, which equals the quotient of actual surface and geometric surface. Increasing surface roughness will promotes liquid spreading for hydrophilic surfaces. Considering a realistic viewpoint, solid surfaces are un-homogeneous in nature and have a rough surface, as well as being chemically heterogeneous causes the contact angle to be slightly different than theoretical values [26].

 Figure . (a) Young's model. (b) Wenzel model [22].

Superhydrophobicity and superhydrophilicity (superwetting properties) are topics that have received an increased attention in wetting studies in recent years. Many of the literature carried out in this field aim to understand and explain the physics behind liquid penetrating (or suspending on) the surfaces of certain geometry. These are controlled at sub-microscopic level. The core principles of superhydrophobicity, superhydrophilicity and fabrication of such surfaces and coatings and their applications were reviewed by many research studies [12,27–39]. This paper will discuss many of the research studies carried out in this subject area.

# 3. Characteristics of hydrophilic materials

## 3.1 Solubility criterion

Any substance is categorised as hydrophilic if they readily dissolve in water. This is the opposite for hydrophobic substances, where substances are poorly soluble in water [40]. The ability of hydrophilic solids to pick up water from the air is described as hygroscopic [41]. Table salt and sugar exhibits the characteristics of hydrophilic, as they can be easily dissolved in water. The surface chemistry of such substances is identical to the bulk of their crystal which in-turn makes the entire chemical substance hydrophilic. Halite, which is a natural NaCl salt, has long been observed by the mining and mineral community to have hydrophilic nature [42]. Research studies have shown that finite contact angles were observed for hydrophilic solutions which included KI, KCl and NaHCO3 [43,44]. Many other organic and inorganic substances as well as polymers are known to dissolve in water. Dissolution can tell the extent of the wettability of a substance, however, it could also be misleading. The main reason for this is that solubility of any substance is governed by the balance of intermolecular forces between molecules in liquid and solid phases, together with an entropy change that accompanies the dissolution and solvation [41]. For example, washing up liquid is soluble in water but it is categorised as an amphiphilic molecule due to the presence of a polar and non-polar regions giving rise to hydrophobic properties as well [45]. Complete spreading of water drops on compressed detergents discs is prevented due to the hydrophobic regions on the surfactant molecules [46].

## 3.2 Polar spreads on polar

‘‘Like dissolves like’’ can be used to determine if a liquid will or not dissolve in another. For example, polar molecules will be able to dissolve in water as water is a polar molecule. This helps in predicting whether or not a substance will dissolve in water. This also applies for liquids spreading on surfaces. Surfaces containing polar groups have an electric dipole or multipole moment. This is a qualitative method to define a hydrophilic surface or ‘‘polar spreads on polar.’’ However, predicting hydrophilicity of metals using this method is more complex. Metal surfaces don’t have any polar chemical groups and if they are not covered with an oxide layer, water spreads almost completely on the surface of the metal. The reason for water spreading on metals surface is due to the dispersion forces, and these are adequate to induce water spreading on clean surfaces of noble metals [47].

## 3.3 Contact angle value criterion

Polar liquids such as water prefers to spread on hydrophilic surfaces. The contact angle of water on a hydrophilic surface is less than 90° in a solid–water– air system. Any contact angle higher than 90° describes the system to have water on a hydrophobic surface [42,48]. Contact angle hysteresis is defined as the difference between the advancing contact angle and the receding contact angle. Its value is dependent on whether the measurements are done under static or dynamic conditions. The rate of liquid movement also influences contact angle hysteresis [49]. Contact angle hysteresis may not always be demonstrated on smooth and homogeneous surfaces [50,51]. To measure static contact angle of bulk materials with smooth surfaces, sessile drop and captive-bubble techniques are often used [52,53]. The sessile-drop technique is a simple technique that is commonly used. However, captive-bubble method has its own advantages. Both solid and gas phases, in the captive-bubble method, are already saturated with water or water vapor, and measurements of contact angle are carried out under more stable and reproducible conditions [48]. Representative contact angle values can be used for to identify the wetting behaviour of the liquid in-terms of hydrophilic and hydrophobic surfaces. Hydrophilicity is found in abundance in nature and is one of many characteristics that animals have to help their survival. Hydrophobicity is also found in nature but not as abundant as hydrophilicity. Artificially synthesised hydrophobic substances are saturated hydrocarbon-based products such as wax, polyethylene, polypropylene, self-assembled monolayers with hydrocarbon functional groups as well as fluorine-based polymers, hydrocarbons, and monolayers. The addition of heteroatoms, such as oxygen, into the hydrocarbon structure, adds a polarity to the molecule enhancing hydrophilicity of the surface [31].

# 4. Characteristics of hydrophobic materials

It is important to ensure that the fabrication methods for hydrophobic coatings result in coatings with the correct characteristics, as these determine the degree of hydrophobicity of the materials. Hydrophobic coatings have specific characteristics and these will be discussed in this section.

Hydrophobicity is commonly known for being water repelling. This characteristic is often linked to the material being non-polar. As water is polar, the non-polar material will repel water [54–57]. Non-polar materials do have an affinity with other non-polar materials, making hydrophobic materials/coatings usable to attract alkanes (fats and oils), noble gasses and other homonuclear diatomic elements [58].

The hydrophobicity is characterised by three properties: the water droplet contact angle (θe), hysteresis and the surface roughness. The contact angle of the water droplet describes the angle between the liquid-solid and liquid-vapour interface. The bigger the angle becomes, the more hydrophobic the material is [59–61]. Figure 3 shows images of water droplets that spread on surface with different characteristics. Figure 3a shows the intersection between the liquid-solid and the liquid-vapour phase. As the water droplet contact angle increases, the material becomes more hydrophobic. This would result in a droplet that can be seen in Figure 3b. Figure 3c shows a water droplet with a decreased contact angle, which results in a less hydrophobic/more hydrophilic material [59].

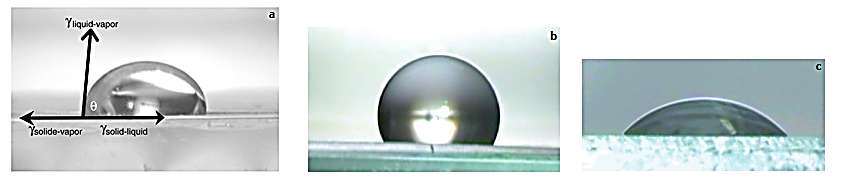


Figure 3. Water droplet spreads onto a surface depending on the interaction of the surface with the water molecules. 3(a) shows the contact angle between liquid-vapour and solid-liquid. The figure also shows a very strong hydrophobic/SH material (b) and a very weak hydrophobic or hydrophilic material (c) [59].

The wetting of rough surfaces can be predicted with Young’s equilibrium contact angle, which can be found with Young’s equation. Young’s equation can be written as equation 1 shown in section 2.

The water droplet contact angle (θe) is determined by the surface tension of the solid-vapour (ɣSV), the solid-liquid (ɣSL) and the vapour-liquid (ɣVL) phase. As the water droplet contact angle is determined, the tested material can be characterized as either hydrophilic (θe < 90˚), hydrophobic (90˚< θe < 150˚) or SHO (θe > 150˚) [59,62–65]. These materials can either present the ‘’lotus effect’’ or the ‘’petal effect’’. The expression of these effects strongly depends on the contact angle hysteresis (CAH). The CAH describes the surface adhesion where a small CAH means weak surface adhesion. This can be translated to the lotus and petal effect, as they both are SHO but the petal effect has a very strong adhesion to water droplets and the lotus effect hasn’t. Therefore, CAH is bigger in materials with the petal effect than in materials with the lotus effect [17,66–69]. The CAH can be determined by measuring the advancing angle θa and the receding angle θr. With a small CAH (θa > θr)the water droplet slides down the inclined platform (lotus effect). In contrast is the high CAH (θa < θr) which results in the water droplet being unable to slide down the inclined platform (petal effect). Figure 4 shows the water droplet with the advancing and receding angle on an inclined platform [17,60,70].

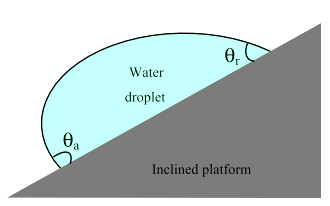


Figure 4. An experimental set-up of how the CAH can be measured. The figure shows the slope, the advancing angle θa and the receding angle of the water droplet θr.

Both, the water droplet contact angle and the CAH are caused by surface heterogeneity and surface roughness. This surface roughness can be divided into either micro or nanoscale roughness and both help to increase the water droplet contact angle [18,56,71]. Different fabrication methods can improve the macro, micro and nano-roughness of the material roughness, which results in a more hydrophobic material [64,72–74]. Nanoscale roughness is often modified with techniques as etching, which adapts particle size, coverage and etch time. These can provide the material with the required roughness parameters [62,65,75–77]. Macro- and micro-roughness can be achieved with other techniques such as laser processing and Hummers’ method. Figure 5 gives an overview of surface roughness and the hydrophobic effect. It can be seen that larger pillars(c) result in a low water droplet contact angle, making the affinity of water to the material larger than in the other two (a, b) materials. The finer the pillars and structures become, the more the higher the hydrophobic effect is achieved [77,78].

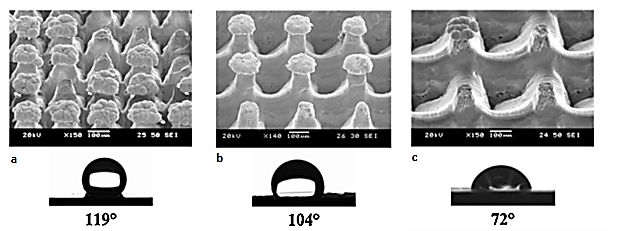


Figure 5. Three different structured materials and their hydrophobic effect. It can be seen that finer pores and more and smaller micro-pillars have the biggest hydrophobic effect [77].

# 5. Fabrication of hydrophilic materials

Generally, enhancing hydrophilicity of surfaces can be performed in two ways: either deposition of a molecular film of a new material that is more hydrophilic than the substrate, or by modification of the substrate’s surface chemistry. Deposition of coatings onto surfaces is the more common method for inorganic substrates. However, modification of surface chemistry is used in the case of polymeric materials. In this section, the most commonly used methods for making surfaces hydrophilic are briefly reviewed.

## 5.1 Deposited molecular structures

Monolayers can be formed from organic molecules which adsorb onto the solid surface. The organic molecules can be sourced from a solution or a vapour phase. This ultimately changes the wetting characteristics of the surface [79]. Many research studies have been conducted on densely pack molecular structures that are used on metals. Alkanethiols have been generally used on gold [80,81], silver [82–84], copper [82–84], platinum [85,86], and palladium [87]. Chlorosilanes have been used on silicon oxide [88–91], aluminium [92,93], and titanium [94]. Phosphoric acids can also be used on titanium [95,96], and aluminium [97,98].

Langmuir-Blodgett film technique is used to mechanically deposit mono and multi-layers. However, one major disadvantage of using this method is that it suffers from poor stability when the multi-layers are contacted by liquids [79].

Hydrophilic surface is achieved if the end of the deposited organic layers are polar. If saturated hydrocarbon-based group or fluorinated group are at the end of the layer, then water will not be attracted to the surface giving rise to hydrophobic conditions. Hydrogen bonding is the way in which water molecules will be attracted to the surface if the surface has chemical groups such as –OH, –COOH and POOH [80,81,99]. A contact angle of zero has never been recorded. Beside arranging self-assembled monolayers of chemically bonded short functional molecules on inorganic surfaces, increased research recently has focused on the coating of materials using macromolecules and biomacromolecules. These are popular in altering polymers contacting the biofluids, such as blood [100]. Biomacromolecules, such as Albumin [101–103] and heparin [104–106], have been widely used in the health sector to provide the hydrophilic characteristics which complement the body’s needs. Synthetic polymers such as polyethylene glycol [100,107,108] and phospholipid [107,109–112] macromolecules have been investigated extensively. The presence of hydrophilic layer in bioengineering applications may lead to proteins being adsorbed onto the surface, which is highly undesirable. Therefore having such protective coatings prevents protein adsorption when materials come into contact with biological fluids [113].

## 5.2 Modification of surface chemistry

Modifying surface chemistry has been investigated for the past few decades. Plasma, corona, flame, photons, electrons, ions, X-rays, g-rays, and ozone are methods that were investigated to alter the chemistry of polymer surfaces without affecting their bulk properties [114,115]. Oxidation of polymer surfaces can be carried out using plasma treatment, in air or oxygen environment [116,117] corona [118,119] and flame [118,120,121].

In both plasma and corona treatments, the electrons are accelerated which then bombard the polymer with energies 2–3 times that necessary to break the molecular bonds, producing free radicals. This generates cross-linking and react with surrounding oxygen to produce oxygen-based functionalities [116]. Hydroxyl, peroxy, carbonyl, ether and ester groups are the typical polar groups which are created at the surface [119]. In the flame treatment, surface combustion of the polymer takes place with formation of radicals such as hydroperoxide and hydroxyl [120,122]. An oxidation depth through flame treatment is approximately 5–10 nm. This increases to over 10 nm for air plasma treatment [123]. Plasma, corona and flame treatments result in extensive surface oxidation and ultimately highly wettable surfaces. Due to the chemistry of the environment, polar groups produced during surface oxidation have a tendency to be buried away in the bulk when in contact with air for extended period of time. However, their presence remains on the surface when in contact with water or any other polar environment [124]. Many polymers undergo oxidation and degradation under ultraviolet (UV) light. For example, outdoor consumer products made from polymers need the incorporation of UV absorbers when exposed to the sunlight to inhibit discoloration, cracking, and fading [125,126]. Wavelength of light ranges from 10 nm to 400 nm, the incident photons have sufficient energy to break the intermolecular bonds of most of the polymers. This enables structural and chemical changes of the macromolecules [127]. The exposure of the polymer to UV radiation ultimately results in making the surface more hydrophilic due to the chain scission, crosslinking, and increases the density of oxygen-based polar groups [128–131]. Surface hydrophilicity can be enhanced by alkali treatment of polymers at elevated temperature [132,133]. Polymer surface such as polyethylene terephthalate contain groups such as hydroxyl and carboxyl groups which contricute to the hydrophilicity of the interaction during etching with concentrated bases [134,135]. Conductive oxide surfaces can be electrochemically treated using anodic potential to control its wetting characteristics [136,137].

# 6. Fabrication of hydrophobic materials

Fabrication of hydrophobic materials has been an active research topic since many researchers have tried to recreate the ‘’lotus’’ and ‘’petal’’ hydrophobic characteristics. This research included expensive and toxic processes, making them unattractive to apply on an industry-scale. Since the production of most hydrophobic and SHO coatings are costly, environmentally harmful and are very time-consuming, researchers are now looking for a cheaper, sustainable way to produce hydrophobic and SHO coatings [62,138].

*Y. Zhang et al.* [139] described the fabrication of hydrophobic surfaces as a two-step fabrication where (I) the rough material is constructed and (II) the material is chemically modified to meet the desired characteristics. A great number of fabrication techniques have been reported, such as layer-by-layer assembly, laser process, the solution-immersion method, sol-gel techniques, chemical etching and Hummer’s method. This chapter shortly reviews all the named fabrication techniques.

## 6.1 Layer-by-layer assembly

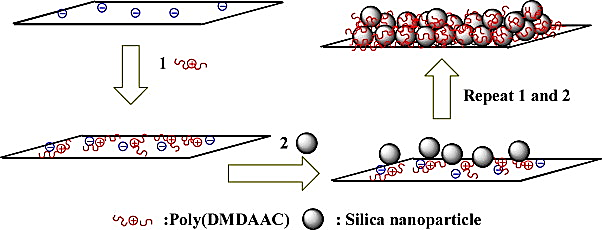
Layer-by-layer (LBL) assembly is a rather attractive fabrication method because of its ability to create multifunctional films on materials while maintaining bulk properties [140,141]. LBL can be applied on a wide variety of substrates such as noble metals, oxides and synthetic polymers. Applying LBL layers on synthetic polymers has been found to be rather challenging as the polymers often require aggressive pre-treatment such as plasma treatment, oxidative chemical reactions or polymeric adsorption. LBL deposition typically consists of four steps: adsorption of a positive component, washing, adsorption of a negative component and final washing. These steps can be repeated multiple times to build a thicker film [140,142]. The LBL deposition can be seen in Figure 6 where the first arrow shows the adsorption of positive components, the second arrow shows the washing and adsorption of negative components and the repeat 1 and 2 arrow shows the end result [78,143,144].

Figure 6. An overview of the layer-by-layer assembly method where it can be seen that a thick layer of silica nanoparticles is built on the material by multiple steps [144].

## 6.2 Laser process

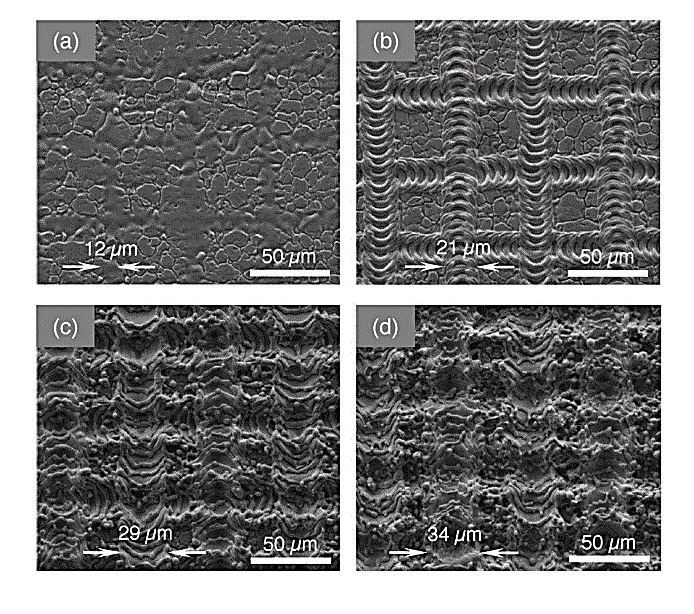
Laser processing has found to be a promising fabrication technique due to three main factors: (I) its excellent control of surface roughness from nano to microscale, (II) it’s a single-step processing under the right conditions and (III) the ability to work with various types of materials [77,145,146]. Laser treatment can be used to increase the static water droplet contact angle by increasing the material’s surface roughness [147]. Lasers are usually used to apply complex structures to the material, where the material is often coated with a fluorine-based material afterwards to create an SHO surface. The fluorine-based material and the surface roughness result in a very hydrophobic material [14,62,147–150]. Figure 7 shows different SEM images of laser-textured surfaces with various laser powers. Figure 7a has the lowest laser power, resulting in a less defined grid compared with b, c and d [77].

Figure . SEM pictures of laser textures after treatment with different laser powers [77].

## 6.3 Solution-immersion method

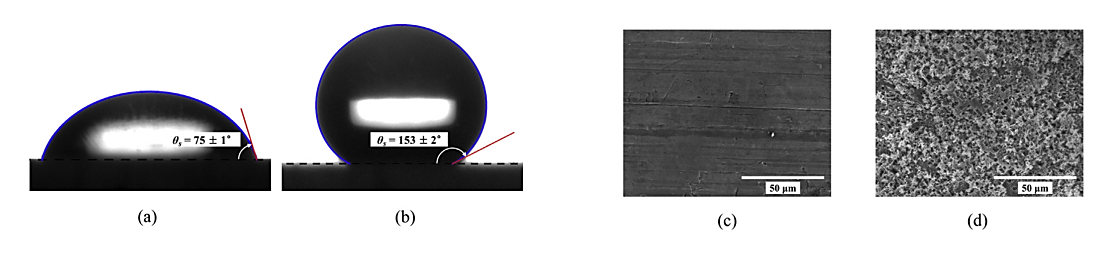
The solution-immersion method can be applied to coat multiple materials with a SHO layer. The solution-immersion method makes it very easy to fabricate hydrophobic coatings. Many research studies [73,76,151–153] have been able to apply a hydrophobic coat to multiple materials such as wood and metals. The solution-immersion method often contains multiple steps. Research by *M. Kim et al.* [73] applied a hydrophobic layer to wood by immersing the sample in 1M alkaline solution, followed by a 0.3M acidic solution and finished by coating it with a 2H-perflurodecyltrichlorosilane solution. Figure 8 shows the hydrophobic effect before and after the solution-immersion method. It can clearly be seen that the smooth surface (8c) became very rough (8d). This results in a SHO surface, which is shown (8b) as the water droplet contact angle is bigger than 150˚.

Figure 8. Before the solution-immersion method, the material is smooth and not hydrophobic (θ < 90˚), after the solution-immersion method the material became very rough which, in combination with the polymer layer applied on top, creates a very SH surface [73].

## 6.4 Sol-gel technique

The sol-gel technique can be applied to various types of materials. The sol-gen technique can be divided into multiple steps where (I) the sample will be washed, (II) the cleaned sample will be immersed into a solution, (III) the coated sample will be washed again and (IV) the sample will be dried [56,138,154–156]. The research of *S. Wang et al.* [72] used the sol-gen technique to apply a wood surface with silica particles. Afterwards, the silica got a fluorination treatment with a surface modifying agent of 1H, 1H, 2H, 2H-perfluoroalkyltriethoxysilanes which resulted in a SHO coating on the wood [157,158].

## 6.5 Chemical etching

Chemical etching is a type of surface treatment that is commonly applied to metals. Many researchers have conducted experiments with applying hydrophobic layers onto metals, and most researchers preferred using magnesium alloy [61,159–162]. The etching process consists of multiple steps where (I) the sample is cleaned, often ultrasonically, and dried, (II) the material is etched in a aqueous solution, often containing copper, (III) the material is rinsed with water and often with ethanol, (IV) the material will be dried and finally (V) the material will be modified with a silica-containing solution, making the end product SHO [61,159–163].

## 6.6 Hummers’ method

The Hummers’ method is an alternative oxidation method developed by Hummers’ and Offeman in 1958. The method used a solution of NaNO3 and KMnO4 dissolved in H2SO4 to oxidize graphite into graphite oxide. The Hummers’ method was a very fast way of oxidizing. As the Hummer’ method is very easy and quick to perform, it is now a widely used method [164–166]. *N. Cao et al.* [167]used Hummer’s method to produce a hydrophobic aerogel. After the Hummers’ method, various applications of solutions and drying processes have been applied, finishing with a reaction with 1H, 1H, 2H, 2H-perfluorodecanethiol. Although the product is highly hydrophobic, the reaction takes up much more effort and time than the other methods previously mentioned [168,169].

Besides the six named fabrication techniques, there are plenty of other techniques that can be applied to fabricate a hydrophobic coating. Each described method has its pros and cons, where some methods are very easy and fast but can only be applied to specific types of materials and others are complex but achieve high-quality results on many types of materials. Table 1 is an overview of each technique with the advantages and disadvantages for each of the techniques.

Table 1. Advantages and disadvantages of each fabrication method.

|  |  |  |
| --- | --- | --- |
| Technique | Advantages | Disadvantages |
| Layer-by-layer assembly | * Can be applied to various materials * Bulk properties remain | * Requires many steps, slow technique * Often very challenging to apply to certain materials |
| Laser processing | * Can be applied to various materials * Excellent control on surface roughness * Can be done in 1 step * Fast technique | * The technique is very expensive |
| Solution-immersion method | * Can be applied to various materials * Simplistic and easy technique | * Requires many steps, slow technique |
| Sol-gen technique | * Can be applied to various materials | * Requires many steps, slow technique |
| Chemical etching | * Many researches available about this technique | * Only really applied to metal materials * Requires many steps, slow technique |
| The Hummers’ method | * Fast technique | * Can’t be applied to many materials |

# 7. Applications of hydrophilic materials

As discussed previously, the hydrophilic property has many uses in the natural and artificial environments. It has highly been investigated in various animals and this information is being used to produce synthetic materials that have a wide range of uses. This section will discuss specific uses of hydrophilic materials.

## 7.1 Glass/ Anti-fogging surfaces

High humidity reduces visibility drastically, which can be dangerous depending on the setting. Anti-fogging surfaces can be used to reduce fogging. Swimming goggles is a scenario where fogging occurs on the inner surface of the goggles due to the humidity build up in the enclosed space. Relative humidity is a function of temperature, allowing water to reach its dew point when encountering a cold surface. This results in the formation of water droplets due to the condensation taking place. Optical clarity of the transparent surface will be reduced significantly, shown in Figure 9 left. Recently, anti-fogging surfaces has been becoming a necessity and it is highlighted by micro- and nanofluidic applications. Visualization of two phase flow in the cathode micro-channels of proton electrolyte membrane fuel cells [170].

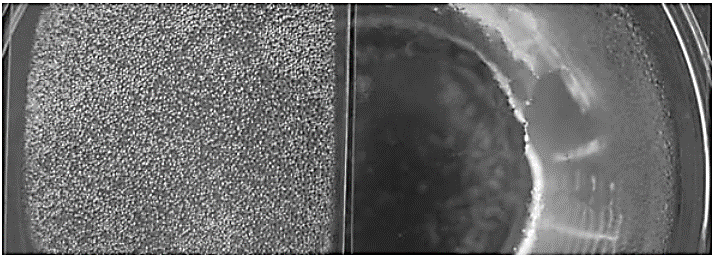


Figure 9. Left: untreated polyester film showing low optical clarity. Right: plasma-treated superhydrophilic polyester with high optical clarity [2].

Cell cultivation has similar challenges in-terms of fogging. This has been resolved due to the innovation of hydrophilic materials [137]. Humidity inside food packaging increases when they are refrigerated. This occurs due to the decrease in temperature. Consequently, condensation occurs on the inner surface of packaging. A superhydrophilic surface can be anti-fog because water spreads on the rough hydrophilic surface to form a thin film instead of droplets. Superhydrophilic treatment is different from traditional anti-fogging coating that is widely used for swimming goggles and eyeglasses. The later usually employs various surface coatings to treat the surface hydrophobic, which tends to have low adhesion with the tiny water droplet formed on it. Hydrophobic anti-fog surfaces are usually more effective than the superhydrophilic surfaces [40]. However, a coating approach might be undesirable in many conditions, such as inside a microchannel. Safety concerns arise due to chemical agencies used for biomedical sample and food, especially when the surface is subjected to environments of high temperature and high humidity.

## 7.2 Bio-fouling and its prevention

The continuous water thin film formed on a hydrophilic or superhydophilic surface has a profound impact on their interactions with molecules and micro-organisms, including biofouling and biocompatibility. In marine engineering, the growth of microorganisms such as algae on a surface of a ship immersed in the sea can cause fouling. Fouling is significant in biomedical devices because they are subject to fouling of cells and biomolecules, such as proteins. Fouling negatively impacts the performance of such devices. It is desired to reduce or avoid or reverse biofouling using anti-fouling and fouling-release [171]. When observing the hydrophilic parts of the surface, a trend is clearly seen that for high-surface energy materials, the degree of fouling is reduced with surface energy. This is explained by the strong attraction between surface and water molecules, establishing a barrier to prevent interaction between fouling agent and surface and thus delaying the fouling [172–174]. Research studies have shown significant decrease of fouling by using fluorescein and fluorescent proteins after the surfaces being treated to be superhydrophilic [172]. However, these results have been obtained in a relatively short period of 30 minutes with static liquid. Therefore, they are not a true indicator for long-term prevention and release of biofouling [172]. The main difference between short-term and longer-term fouling of hydrophilic and superhydrophilic surfaces can be attributed to the quick degradation of hydrophilicity, which tends to be unstable [175].

## 7.3 Enhanced boiling heat transfer

Enhanced boiling is the most efficient mode of heat transfer. It is being implemented in a broad range of power generation and thermal management devices, such as nuclear power plants, refrigeration, cooling of electronics and chemical reactors [176–179]. Surface wettability has a significant effect on boiling heat transfer. Figure 10 shows a boiling curve which correlate the heat flux with wall superheat. Point A is the beginning of nucleate boiling, where vapour bubbles form on the overheated surface. The nucleate boiling continues to fully develop from B to C. At point C, the het flux eventually reaches its maximum value, known as critical heat flux (CHF). This is due to the continuous vapour film forming as an effective thermal insulation layer. Further heating beyond point C will lead to dramatic increase of wall temperature and ultimately result in device failure. Therefore, CHF marks the maximum heat flux that can be provided by a boiling-based cooler.

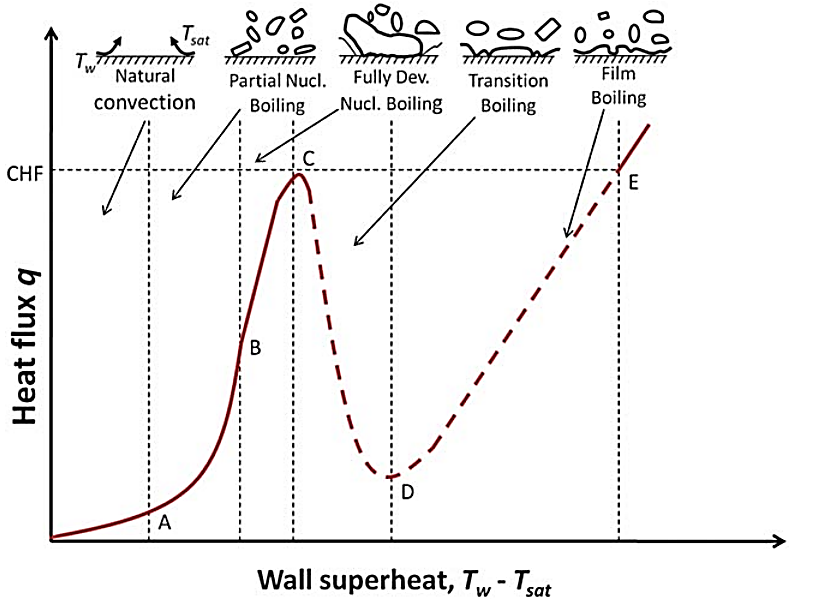


Figure 10. Propagation of boiling stages [114].

However, having a hydrophilic or superhydrophilic surface forms a continuous water film which delays the formation of vapour film in boiling and thus improve CHF. Many studies on vertically aligned nanoforests of hydrophilic/superhydrophilic nanorods, nanowires and CNTs have shown a significant improvement in boiling heat transfer [180–183]. For example, both CHF and heat transfer coefficient (HTC) have been improved by more than 100%. This improvement has been primarily due to the increased density of nucleation sites, high surface tension force of superhydrophilic nanostructures. This allows the liquid to be pumped and the cavity stability provided by the nanopores [180,184]. Furthermore, it has been shown that a surface with mixed hydrophilic and hydrophobic micro-patterns result in enhanced pool boiling to almost the same degree. For example, 65% and 100% improvements were achieved on CHF and HTC, respectively, with a hydrophilic and hydrophobic network [183]. The combination of both wettabilities prevent the formation of vapour film by attracting liquid onto the hydrophilic region, while nucleation is promoted at the hydrophobic regions. This help to remove gas bubble efficiently [183].

## 7.4 Other Applications in Biomedical Field

Hydrophilic coatings have been widely used in the medical field. The main components used in hydrophilic coatings are polyvinylpyrolidone, polyurethanes, polyacrylic acid, polyethylene oxide, and polysaccharides. The development of these types of coatings have more recently moved towards anti-fouling, antimicrobial and biologically active surfaces. Biomedical applications of polymers include vascular heart valves, artificial hearts, catheters, contact lenses, intraocular lenses, and plasmapheresis units. The pharmaceutical industry widely applies coatings for tablets and capsules, adhesives, and blood substitutes [12]. The major advantage of using hydrophilic coatings is that they provide better lubricity compared to hydrophobic coatings [185,186]. Contact lenses must be wetted by tear fluid to move relatively freely on the eye. This provides comfort to the wearer [187]. The applied research on surface modification of contact lenses is substantial [122-124] and mostly deals with making surface of polymer hydrophilic. However, due to hydrophobicity of silicone hygrogels, they require hydrophilic coatings for improved wettability with tear fluid, wearing comfort and biocompatibility.

Surface coatings included neutral hydrophilic polymers such as polyacrylamide and polyethylene oxide (PEO), phospholipids, dextran, pullulan, and others [188–191].

## 7.5 Heat pipes

An experiment conducted by Hao *et al.* [192] which investigated the effects of hydrophilic surface on heat transfer performance for an oscillating heat pipe (OHP). An image of the oscillating heat pipe is shown in Figure 11.

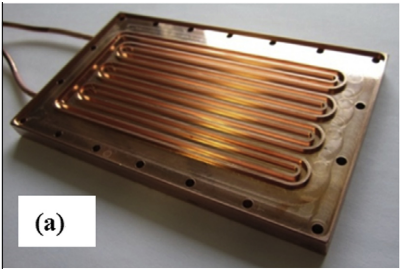


Figure 11. Photo of a four turn copper plate OHP [192].

The experimental results showed that the surface wettability remarkably influences slug motion and thermal performance of OHPs. A stronger liquid slug movement was observed in both superhydrophilic and hydrophilic OHPs in comparison with copper OHP. The global heat transfer performance of the superhydrophilic and hydrophilic OHPs increased in the sixth turn OHP. For six-turn OHPs, the maximum displacement of the liquid slug in the hydrophilic OHPs and superhydrophilic OHPs increased by 5–60% and 25–60%, respectively, relative with that of copper OHPs. An increase by 5–15% and 15–25% of the heat transfer performance of superhydrophilic OHPs and hydrophilic OHPs, respectively, has been observed, in comparison with that of copper OHPs. This investigation has clearly shown that hydrophilic surface improved the heat transport capability of OHPs.

## 7.6 Hydrophilic property for filtration

Commercial hydrophilic membranes used to filter aqueous and organic solvents used in HPLC offer maximum chemical and pH resistance. The membranes are a thin, unsupported, highly porous films. High flow rates can be permeated through this type of membrane with minimal aqueous extractables [193].

Since the filter is optically clear when wet it is ideally suited for culture and microscope examination of cells/particles captured on the filter surface without requiring a separate step to render the filter optically clear. This area requires further research for potential use of hydrophilic membranes to filter pathogens from water.

## 7.7 Road marking paint

Due to local climatic conditions such as variations in temperature, the structure of asphalt, vehicles, drivers’ behaviour, exposure to ultraviolet radiation (UV) and to pollutants road marking paint loses its fundamental properties. Therefore, superhydrophilic materials used within the paint enables self-cleaning” paint using photocatalytic titanium dioxide (TiO2). Super-hydrophilic property allows dirt to be easily washed off with water or rain [194–196].

The superhydrophilic property is capable to prevent pollutants from adhering to the substrate through the formation of a uniform water film over the solid surface. The synergism of hydrophilic and photocatalytic results could maintain aesthetical properties; improve surface maintenance and reduce degrading processes [195].

## 7.8 Mesh

Liu *et al.* [197] have demonstrated facile fabrication method to synthesize aluminium-introduced MFI-type zeolite-coated mesh to separate oily water. The membrane’s hydrophilicity was controlled by adjusting the ratio of Al/Si in precursor solution. Their results have shown that increasing the aluminium content in the zeolite has led to a larger contact angle. Oil was the non-wetting phase in this experiment. Oil rejection rate was over 99% at flux of >80000 L m-2 h-1 for hexane water mixture. Other mixtures of oil with water such as n-hexane, cyclohexane, mineral oil, and vegetable oil have all shown rejection rates of over 96%. The exact results are shown in Figure 12.

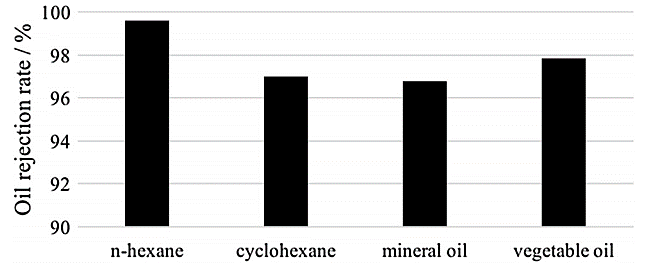


Figure 12. Oil rejection rates of the hydrophilic zeolite mesh for various oils [197].

The results shown in Figure 12 indicate a promising method for oil/water separation that could potentially be used at industrial level. To show the experiment at laboratory scale, Figure 13 shows the before and after of the oil water mixture.

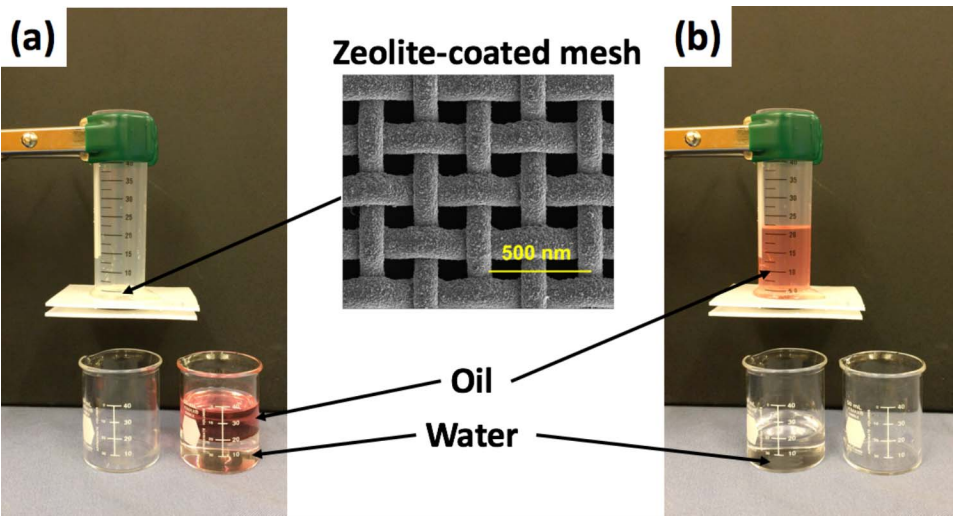


Figure 13. Oil/water separation using zeolite-coated mesh (a) before separation; (b) after separation [197].

The mechanism in which the separation takes place is that the hydrophilic mesh has a very high affinity to water which allows it to pass through easily. However, the oil which is the non-wetting phase is repelled away from the mesh and is rejected above it.

## 7.9 Sample pre-treatment

Sample pre-treatment is a fundamental and essential step in almost all chemical analytical techniques, especially for the analysis of biological and environmental samples with complex matrix. The development of hydrophilic interaction liquid chromatography (HILIC) in the separation of polar compounds, hydrophilic materials have been extensively implemented in sample pre-treatment in various disciplines, such as pharmaceutical, clinical, toxicological, food and environmental analysis [198].

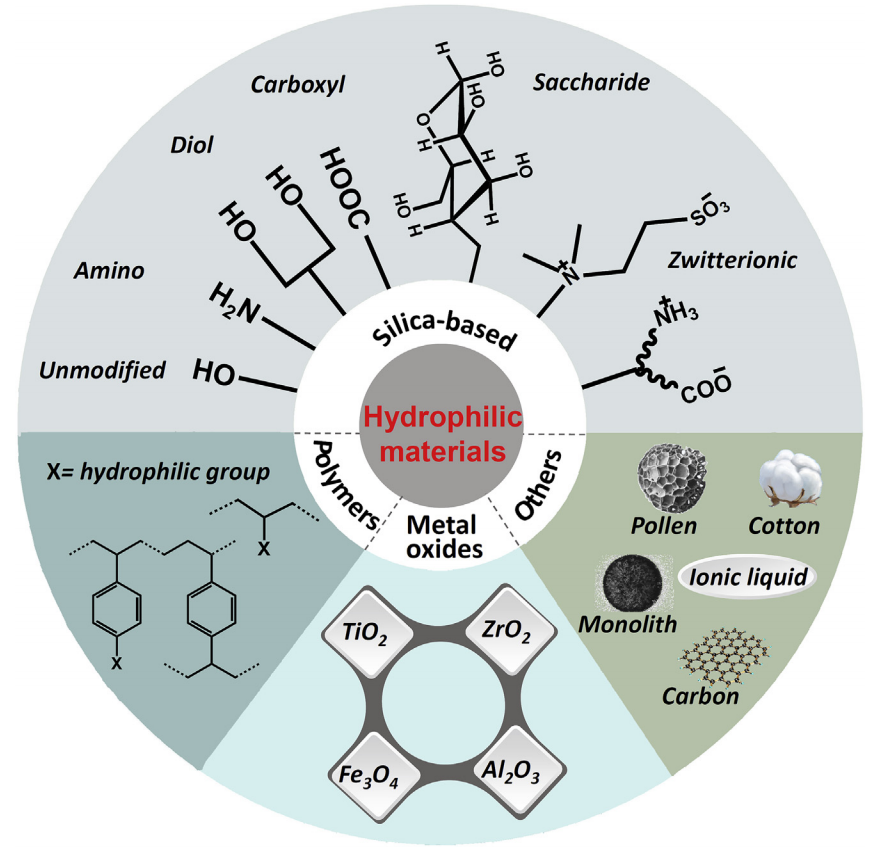


Figure 14. Hydrophilic materials used in sample pre-treatment [198].

The most frequently used hydrophilic materials in HILIC-based sample pre-treatment is silica based materials. Other hydrophilic based materials are shown in Figure 14. These materials, such as amino-, diol, zwitterionic, carboxyl and saccharide silica type materials, have proven their practical application for HILIC-based sample pre-treatment.

# 8. Applications of hydrophobic materials

Hydrophobic coatings can be applied to many materials and can be used in various situations. Since SHO coatings repel water, it can be useful in many everyday situations which will be discussed in this section.

## 8.1 Petroleum

Hydrophobic coatings could play an important role in the cleaning up of oil spills. Oils spills on freshwater can bring many problems environmentally and economically. Since the behaviour of oil in water is very hard to predict, cleaning up oils spills can be a hard task. The clean-up of oil spills depends on the type of oil that is spilt, the type of water, water turbulence and water temperature [55]. Petroleum consists of different hydrocarbons: aliphatic hydrocarbons (40-80%), aromatic hydrocarbons (15-50%) and resins and asphaltenes (0 – 20%) [199]. Petroleum also contains harmful polycyclic aromatic hydrocarbons (PAHs) and monoaromatic hydrocarbons (MAHs) which can be released into the environment where it pollutes the air, water and soil.

Hydrophobic materials can be applied to solve both problems. Research by *C. Filho* *et al.* [199]used mesoporous aerogels, made during a sol-gel process, to adsorb PAHs from an audacious solution. The adsorption by aerogels is found to contain both chemi- and physisorption mechanisms. The research found that the sorption remained the same for all solutions although the use of different starting concentrations.

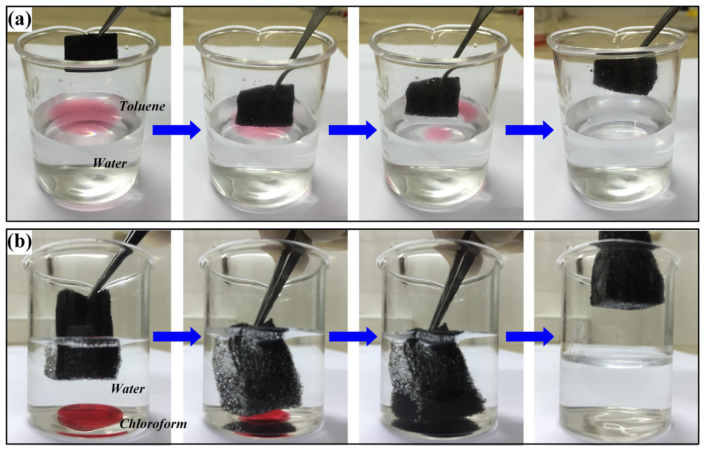
Other researchers [200–205] also made SHO sponges and gels that were capable of adsorbing oil from aqueous solutions. The research by L. Zhang et al. [205] shows the fabrication of a hydrophobic sponge. The experiment has shown that the sponge was successfully capable to adsorb toluene and chloroform from an aqueous water solution. Figure 15 shows pictures from the experiment, where it is clear to see that the sponge adsorbed the toluene completely.

Figure 15. Experiment from L. Zhang et al. [205] showing that the SH sponge fully adsorbed toluene.

Since many research studies have already shown the capability of fabricating (academic size) sponges and gels that are capable of adsorbing alkenes, the application of SHO materials for bigger problems is very interesting and promising.

## 8.2 Plastic

Plastic materials are heavily used in both everyday household items, as well as functional industrial materials. Plastic can be used in packaging, bottles, containers, buckets, dishware, glasses, cups, electrical circuit boards, cushions and more. The application of plastic in everyday household items is very common and the use of hydrophobic and SHO coatings can improve the quality of these items [206].

As there are many different types of plastic, they can be applied in various ways. Polyurethane (PU) sponges can be applied to adsorb organic solvents from water surfaces due to their high porosity and low density, resulting in a high adsorption ability at low costs. PU is naturally hydrophilic, but modifying the PU sponges with TiO2 increases the PU sponges’ roughness which increases the hydrophobicity [154,203,204]. Sponges as such can be applied as a selective filter to separate oil from water. Polyethylene (PE) can be applied as a hydrophobic coating to paper and paperboard packages, ensuring that the porous cellulose networks of paper are more water-resistant. One of the major applications of this technology are the single-use paper cups [207–210]. Since the hydrophobic coating normally is petroleum derived, the usage of the coatings is not environmentally friendly. Current research is in progress for single-use paper cups with bio-derived PE [211–213]. Other applications of hydrophobic plastic coatings could be their self-cleaning property. This property can be applied in both the biotechnology and the household sector:

1. The biotechnology sector can apply plastic hydrophobic coatings to control droplets that contain biologically relevant molecules such as DNA and proteins while minimizing the chance of contamination.
2. The household sector can apply plastic hydrophobic coatings to cutting boards, cups, cushions and food containers to make them self-cleaning. The water repelling effect will result in fewer stains and makes the cleaning of kitchen items easier [154,214].

As hydrophobic coatings are heavily applied in packaging, everyday household items and technology industries, it is important that bio-derived plastic coatings replace the traditional petroleum based-coatings. This will ensure a more sustainable future.

## 8.3 Heat pipes

Heat pipes are widely applied in various heat exchange systems such as air-conditioners and heat pumps. In air conditioners, moisture condenses and accumulates on the heat transfer surface of either an evaporator or cooling coil when the surface temperature is below the dew point of the conditioned air [18,215]. Air-conditioners often have a water hold-up, which is caused by remaining water droplets on the air-conditioners’ surface. Water hold-up can lead to unwanted conditions: it can increase the air-side pressure drop, reduces the air-side heat transfer coefficient, degrades the cooling capacity, leads to corrosion and provides an attractive environment for biological activity [216,217]. Frost layers can occur on the heat transfer’s surface during colder days, which increases the heat transfer resistance, which can lead to the shutdown of the heat pump [218,219]. Hydrophobic and SHO coatings could be applied to heat pipes to increase their performance.

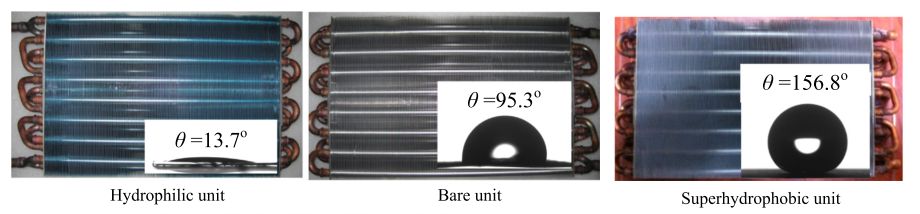
Many researchers have investigated the application of hydrophobic and SHO coatings of heat exchangers and heat pipes to make the system more (energy) efficient. The application of these layers has been done through chemical etching. Many research studies [17,18,73,147,217,219] found that the heat transfer capability does not change after applying hydrophobic or SHO coatings. They also found that frost layers immediately shed from the SHO coatings at the beginning of frost melting. Hydrophobic surfaces contained water after the frost melting but there was no water retention seen on the SHO surface [220–222].

Figure 16. Experimental set-ups of each hydrophilic, normal and superhydrophobic units. It can clearly be seen that the SHO unit repels the water where the hydrophilic attracts the water droplet [220].

Hydrophobic and SHO coatings can be applied to heat pipes to improve the anti-freezing behaviour. Figure 16 shows three heat pipe systems, where the hydrophilic unit strongly attracts the water droplets and the SHO unit strongly repels the water droplets. Applying SHO coatings on heat pipe systems not only improves the performance of the system, it also prevents the unwanted build-up of water hold-up [182,223–225].

## 8.4 Other metal applications

Hydrophobic and SHO can be applied to various types of metal to improve their performance. Polytetrafluoroethylene (TEFLON) is often applied to aluminium alloys to increase the surface hydrophobicity. A very common application of TEFLON is pan coatings. The TEFLON coating is highly hydrophobic (θe ≈ 120˚), which results in a non-stick coating which prevents food to stick to the pan [59,223,226,227].

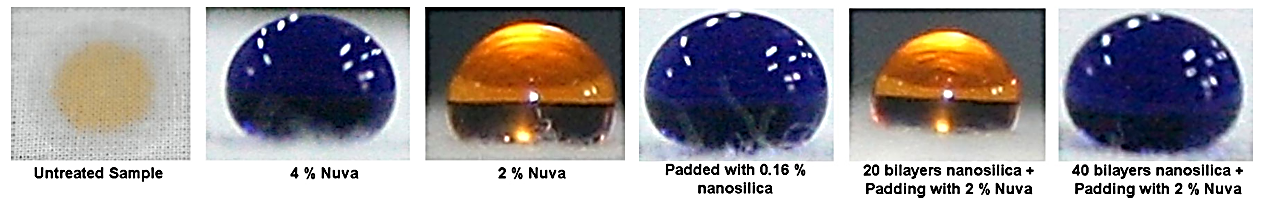
TEFLON and other hydrophobic and SHO coatings are also applied to metals for other purposes than cooking. As hydrophobic and SHO coatings have the characteristic that water drops roll off the surface with the slightest tilt which makes the application of these coatings interesting in other areas such as microfluidic devices and anti-icing applications [227,228]. SHO coatings can also be applied to prevent corrosion of metals that are vulnerable to corrosion. As SHO coatings repel water, it results in a high corrosion resistance property and stability. These conditions contribute to the chemical stability of a metal complex [229–231]. Another application of hydrophobic and SHO coatings can be found in nanotechnology. Research of *Cho et al.* [232] has found a method to produce memory devices based on multilayer stacks of densely packed hydrophobic coated metal arrays. The memory of these devices can be significantly enhanced by increasing the charge trap elementals with hydrophobic properties. Hydrophobic and SHO surfaces can also be applied in the capture of harmful (volatile) organic compounds. The surfaces can be applied to remove oils from surface water, but can also be applied to capture organic compounds in other moist environments such as steam [233,234].

There seems to be a large interest in metal hydrophobic coated materials and research studies, such as *Cho et al.* [232]are currently applying this technology to improve current technology.

## 8.5 Textiles

The use of hydrophobic and SHO coatings on textiles has increased drastically the last couple of years. The interest of self-cleaning clothes or clothes, that do not get dirty at all, has always been an interesting topic [235]. This technology developed rapidly the last decades and products as such are now widely available for the customers. An example is the hydrophobic shoe spray that has been on the market for some years now which has been loved and used by many customers as it makes their everyday life more convenient [236].

Many researchers have proven that it is possible to apply hydrophobic and SHO coatings onto textiles such as cotton, polyamide and polyester textiles [237–240]. All studies succeed in applying the hydrophobic coating, where most studies even reached SHO textile characteristics. Figure 17 gives an overview of the research by *Joshi et al.* [241]where it can clearly be seen that all treated coatings show a hydrophobic effect and the untreated sample is absorbed into the fabric. The research showed that the hydrophobic coatings successfully repelled water while retaining the air permeability of the fabric which maintains the comfortable characteristics of the fabric.

Figure 17. Research by Johsi et al. [241] showing that the untreated textile adsorbed the droplet, where all the treated samples showed the hydrophobic effect as they repel the liquid droplet.

Although the results of the application of hydrophobic and SHO coatings to textiles are very positive, it still has its downsides. As the application of the coating is a very pricey process, and it is likely that this technology will be applied to clothes. Another downside is that the hydrophobic/SHO coating slowly wears off. Research by *Su et al.* [242]found that applying friction to the material changes the structure of the coating. Even though the research showed that the hydrophobic characteristics were still present after 1000 times of friction, it is mandatory to understand that the hydrophobic layer slowly changes which can change the hydrophobic effect.

Applying hydrophobic and SHO coatings to both commercial as industrial textiles is an interesting concept, as it contains the self-cleaning characteristic. Applying these coatings to industrial textiles such as aprons, lab coats and uniforms ensures the material to remain stain-free or be safer as it does not absorb any liquid [240,243–245]. The application of these coatings is an interesting concept, yet, more research is required before it can be applied on a large scale.

## 8.6 Glass

Glass can be applied with hydrophobic and SHO coatings for various reasons. One of these reasons can, again, be the self-cleaning effect which simplifies the day to day life. The coating can be applied to outdoor glass and solar panel glass. During the application of hydrophobic coatings to glass, it is important that a good transparency and transmittance remains. Multiple research studies have shown that these characteristics remain after the application of the hydrophobic layer. The transparency remained higher than 80% and the optimal transmittance improved by 7%, this is caused by the anti-reflection effect of the coating [246,247].

Hydrophobic glass coatings can also be applied in various other ways, especially in the biotechnology sector. Various research studies have applied hydrophobic glass in their research:

1. *Zhang et al.* [248]were successful to adsorb chromo-bacterium lipase by using hydrophobic glass beads.
2. *Garg et al.* [249] used glass with different hydrophobic coatings and found out that SHO coatings had the least microbiological growth.
3. *Yayapour et al.* [250] used hydrophobic glass surfaces to investigate the kinetics of cell adhesion between blood and the glass. The research showed that leukocytes adhere to hydrophobic surfaces, which can be applied to separate the leukocytes from the lymphocytes.

The technology sector also found ways to imply the hydrophobic layer on glass. The coatings can be used for anti-icing methods. The research of *Hu et al.* [251]investigated the effect of frozen water droplets on hydrophilic and SHO surfaces. One of the results is shown in Figure 18, where it clearly shows that the water density of the droplets on the SHO surface is decreased. The ice structure contains air chambers that are created in between the SHO droplets. These air chambers are the interfaces between the ice layer and the glass insulator, as these chambers are filled with air, the water density on the surface decreases. As the water droplets slightly melt, the droplets at the SHO surface easily slide off, where the droplets on the hydrophilic surface stay on [251,252].

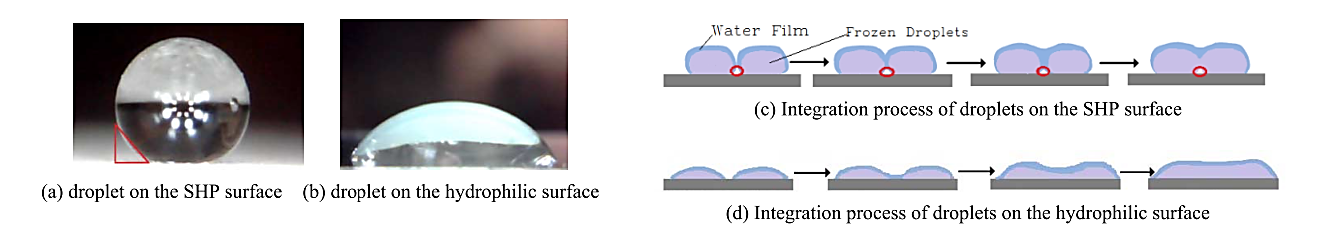


Figure 18. The shape of frozen droplets on both SH and hydrophilic surfaces. Due to the air chambers at the SHO surface, the over-all density is lower and as the ice droplets melt they will slide of the SH surface, while they will stay at the hydrophilic surface [251].

Glass has many applications, both industrial and domestic [248,253,254]. Although there are already so many ways to employ this technology, further improvement and development is needed to optimize them.

## 8.7 Ceramics

Ceramic materials become more popular in the application for water filtration. 70% of the earth’s surface consists of water which essentially comes from seawater and icebergs [255]. Only a small amount of this surface water is potable (3%) which has become a major concern as new fresh water producing techniques are required to offer a sustainable amount of fresh water [256–258]. Different techniques such as reverse osmosis (RO), electrodialysis (ED), membrane distillation (MD) and multi-stage flash distillation can be applied for the desalination of brackish and seawater. Especially MD has gotten an increased attention in the last couple of years for being a simple yet highly efficient technique. MD has shown salt rejection rates around 99-100% [259–262]. MD usually uses membranes made from different polymers but these do have disadvantages as they have poor thermal, mechanical and chemical stabilities, making it challenging to commercially apply them as membranes [260,263,264].

Ceramics can be applied as MD membranes, as their thermal, mechanical and chemical stabilities are more stable. In addition, their chemical inertness, porosity structure and good flux make them interesting for the application as MD membranes [265–267]. Ceramics are naturally hydrophilic, as they contain multiple metal oxides which are hydrophobic due to the presence of hydroxyl (OH-) groups on the surface. Surface modification, usually done by either immersion or grafting processes, reduces the pore size and forms the hydroxyl groups into organosilane compounds, which makes the surface hydrophobic [268–270]. Figure 19 shows the grafting process, where the hydrophilic hydroxyl groups are treated with perfluoroalkylsilane to form a hydrophobic coating [262]. Figure 20 shows the affinity of water droplets on a ceramic surface before and after the grafting process [263].

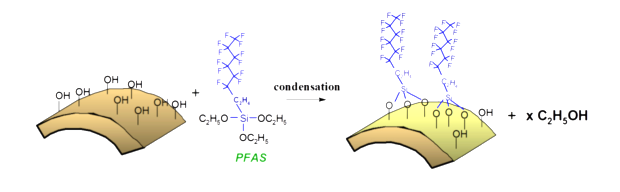
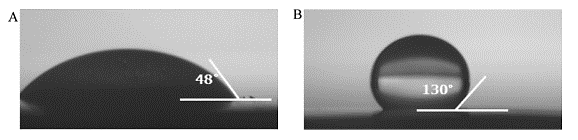


Figure 19. The grafting process where the hydrophilic hydroxyl groups are treated with perfluoroalkylsilane to form a hydrophobic coating [262].

Figure 20. The water droplet affinity with a ceramic surface before (a) and after (b) the grafting process [263].

The hydrophobic ceramic membranes formed can be applied in different separation processes:

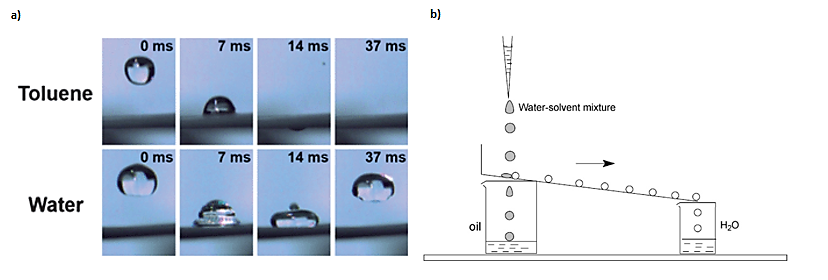
1. The hydrophobic ceramic membrane can be applied in the production of fresh water from brackish and seawater. As the water condensates on one side of the membrane and goes through the pores, that are not permeable for salt ions and the water condensates on the other side of the membrane, the water is cleared from the salt ions [260,263,264].
2. Hydrophobic and SH ceramic membranes can also be applied for the removal of volatile organic compounds from water by vacuum pervaporation. As the liquid streams encounter the membrane, the volatile organic compounds are absorbed into the membrane and permeate through the permeate side. This successfully removes volatile organic compounds from the liquid stream [261,262,267,268].

Hydrophobic ceramic coatings show great potential to be applied in many industries, however, it requires improvement before they can be applied in industries, as the techniques do not always show the highest efficiency.

## 8.8 Mesh

The increasing amount of industrial oily wastewater and oil spill accidents creates environmental, economic and social issues. Due to these issues, there is a constant search for effective and inexpensive clean-up methods [271]. Hydrophobic and SHO mesh has been widely applied for the separation of oil and water. Where other materials are often only coated with a hydrophobic or SHO coating, the coating of most mesh also contains super-oleophilic (SO) properties, resulting in an even higher affinity for oils. Wet chemistry or acid etch methods can be applied to the mesh to fabricate SHO and SO coatings. Most research studies focus on surface roughness or the application of hydrophobic agents, where often they are expensive, harmful and oleophobic chemicals were used, making it difficult to scale-up for industrial applications. In addition, during the application of polymer-based coatings, it is difficult to control both the surface structure and the coatings are not thermostable [272,273].

Materials with both SHO and SO characteristics are optimal for heavy oil/water separation as they exhibit stable water-resisting, anti-chemical erosion and anti-hot ageing properties. Successful oil removal requires a material with high separation capacity, with resistance to oil fouling that is easily recyclable. If the oil separation capacity is not high enough, the removing material can easily be fouled or blocked up by oils due to their oleophilic property. Especially high-viscosity oils tend to adhere to the material, which affects the separation efficiency, are hard to remove and results in secondary pollution during post-treatment processes which waste both the oils and oleophilic material [274–276].

Figure 21.(a) Behaviour of a water droplet on SH and SO mesh and (b) a successful set-up to separate water and oil.

Many research studies have proven that an SHO and SO mesh is able to successfully separate oil from water. Figure 21 gives an overview of two of these experiments. Figure 21a shows the behaviour of a water droplet on an SHO and SO surface. It can be seen from the figure that the water droplet is strongly repelled, as it bounces back from the surface where toluene is strongly attracted and goes through the mesh. Figure 21b shows an experimental set-up for the separation of an oil/water solution where the SHO and SO mesh strongly attracts the oil, which permeates through the mesh and is collected in a beaker. The water is repelled and is collected in another beaker. This technology can, when further improved, be applied on a bigger scale as it has been proven to be highly effective [277–279].

## 8.9 Paint

Paint is initially hydrophilic and tends to be contaminated by pollutants in water. As paint is hydrophilic, it attracts water with dirt which make the visible appearance of the paint old and dirty. Therefore, cleaning is required and manpower is expensive. Due to this, the attention for self-cleaning paints have grown. The application of hydrophobic and SHO coatings is quite common, however, the production of a hydrophobic paint is still very uncommon. In addition, the processes to fabricate hydrophobic materials often requires expensive and environmentally-harmful chemicals, which is not suitable for large-scale applications [280].

Researchers have an interest in the application of hydrophobic and SHO paints in various domains:

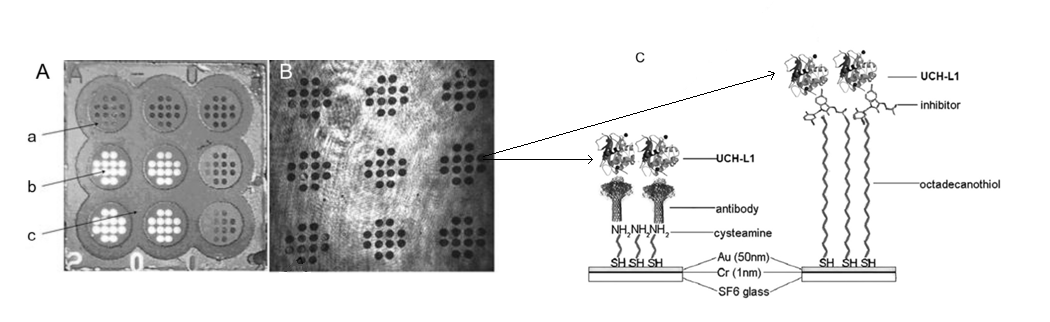
1. Hydrophobic and SHO paints can be applied to marine ships. The hydrophobic characteristic of the paint carries many advantages for ships: it is self-cleaning, repels water and other sea-creatures, and has anticorrosive protection and minimizes drag resistance which reduces the fuel consumption. The application of these paints is still in development, as earlier antifouling paints slowly degraded which spread paint particles through natural waters [281,282].
2. Hydrophobic and SHO paints can be applied for a self-cleaning or protecting effect on various materials. The self-cleaning effect is desirable for building materials, as it improves the aesthetic appearance and reduces maintenance costs. The hydrophobic paint coat can also be applied as a protection layer, e.g. applying the paint to reduce the amount of tritium (3H) in concrete walls [280,283,284].
3. The paint can be applied in the biotechnology sector so can chips with hydrophobic paint layers be used to measure different samples without mixing the tested solutions. The chip can test 9 different solutions and has 12 slots per solution, making them useful e.g. blood tests and DNA tests [285,286]. Figure 22 shows an overview of the chip and gives a schematic overview of the sensor’s active part.
4. Hydrophobic and SHO paints can also be applied to other sensors. *Sakaue et al.* researched the AA-PSP sensor, which is a fast responding global pressure sensor that consists of a pressure sensitive probe on a porous supporting matrix of anodized aluminium. As the sensor adsorbs water, the measurements have a higher uncertainty. Applying the hydrophobic/SHO paint can repel water, which will prevent the measurement uncertainty of the sensor [287].

Figure 22. The figure shows (A) a picture of the chip with (a) a photopolymer, (b) free gold surface and (c) the hydrophobic paint, (B) an image of the chip obtained by a charge-coupled device camera and (C) a schematic overview of the sensor active part [286].

## 8.10 Electronics

Hydrophobic and SHO coatings are slowly becoming more popular in the electronics industry as more handheld devices are protected against water and dust by applying these coatings. Not much research into the use of hydrophobic coatings on electronics has been done before, and most research is stand-alone research rather than follow up research. Some applications of hydrophobic and SHO coatings in the electronics industry that are currently in development are:

1. *Quesada-Gonzáles et al.* [288]used hydrophobic printer material to print different hydrophobic bar channels made of alkyl ketene dimer where each channel can be used for another sample. This application is similar to the (III) paint application. This technology can be applied in the biotechnology sector and is much cheaper than the chip shown in Figure 22.
2. The use of hydrophobic and SHO coatings to protect handheld devices is becoming more popular and advanced. Current high-end mobile devices present such coatings to protect the device to both water and dust. The hydrophobic layer often ensures the use of the mobile device to be underwater for 30 minutes with a maximal depth of 1.5 m [289–291].
3. *Bonfante et al.* [292]and *Luo et al.* [293]researched hydrophobic and SHO coatings used in electro-wetting applications. The coatings were successfully applied and repelled and can be applied to various applications such as adjustable lenses, electronic (outdoor) displays and switches.

## 8.11 Other applications

Hydrophobic and SHO coatings have great potential and can be applied to various types of materials to improve their lifetime or make them more efficient. Even though many materials have already been covered, there are still more applications for hydrophobic and SHO coatings.

A common application of hydrophobic and SHO coatings is on wood. Wood can be applied in many fields and initially has a hydrophilic characteristic caused by natural pores and hydroxyl groups on the surface. This hydrophilic behaviour adsorbed water which can cause the wood to crack, rot and degrade [207,294]. Hydrophobic and SHO coatings could prevent the degradation process and potentially create more applications for the wood. Hydrophobic and SHO coatings are often made during the sol-gen process, where the wood is placed in a ɣ-aminopropyltriethoxysilane bath where the silination of present hydroxyl groups occurs. After the bath, the wood surface is modified with an SHO material such as ethanolic lauryl aldehyde/acid. The surface modification process of wood can also be seen in Figure 23 [72,76,152]. Wood with hydrophobic and SHO coatings can be applied in many fields as: (I) it shows great durability when exposed to acid rain, (II) it reduces the speed of moisture absorption and the amount of moisture that is absorbed is decreases, showing that it is much more water resistant, (III) the coating shows a strong self-cleaning effect which contributes to anti-contamination coatings and (IV) producing the hydrophobic and SHO coatings is a low-cost process, making it easy to apply this technology to many products [295–299]. *Lourençon et al.* [294]used bio-oil, a by-product of pyrolysis (waste-to-energy conversion method), to apply an SHO coating on wood. This coating has increased the overall stability of the wood, resulting in the increase of the lifetime of wooden products. Pyrolysis results in no net carbon emission, making the application of bio-oil interesting as it is not as (environmentally) harmful as other chemicals [300,301].

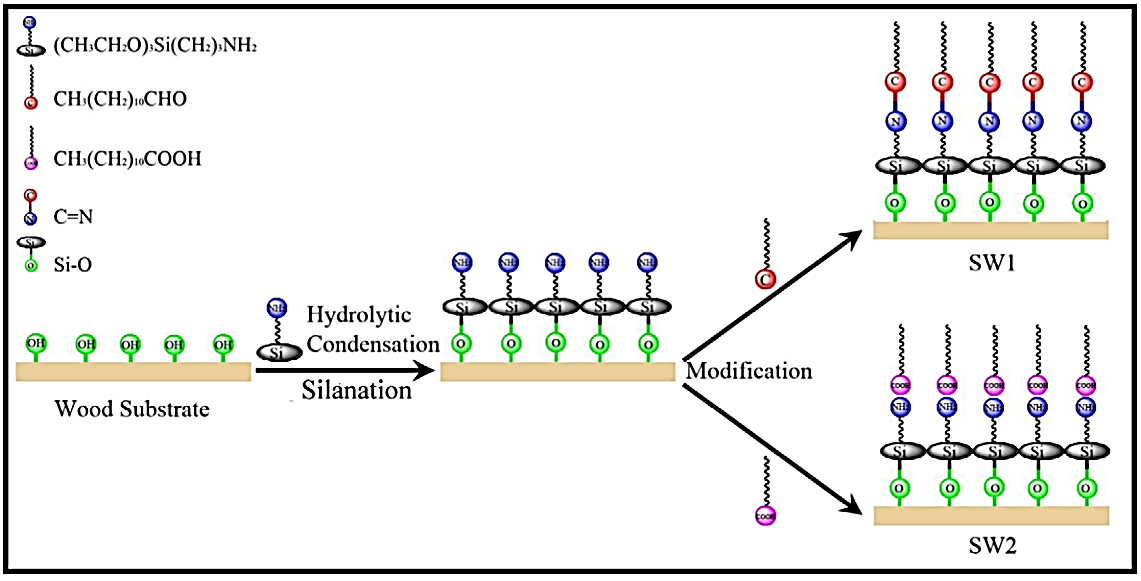


Figure 23. An overview of the production of hydrophobic and SH coatings on wood [76].

Hydrophobic characteristics are also observed in coal. Coal can be specified in different rankings, where especially high-ranked coals show hydrophobic characteristics and low-ranked coal normally shows more hydrophilic characteristics. The difference between the water repelling characteristics is mainly caused by the high amount of oxygen-rich groups at low-ranked coal which are not as much present in the high-ranked coal [302–304]. The lower number of oxygen-rich groups resulted in higher contact angle values in the high-ranked coals, making them more hydrophobic. The increased number of aliphatic compounds in high-ranked charcoal has also increased the coal hydrophobicity. The hydrophobicity is often applied to remove hydrophobic organic compounds out of liquids. Biochar, another pyrolysis by-product, has been proven to successfully absorb organic compounds out of aqueous solutions [305–308].

In addition to these two common applications, more research is currently executed on many other applications such as (I) the application of nano-silver coatings for antifouling and drag reduction applications [309], (II) the application of palm oil to create improved SHO coatings for windows, optical devices and solar panels [310], (III) the cell capture and antimicrobial effect of hydrophobic chitosan on *E. Coli* [311] and (IV) other miscellaneous applications [312–315].

# 9. Conclusion

Hydrophilic and superhydrophilic are properties that are present in nature. These are used as survival mechanisms for many plant and animal species. Close observation of how these species utilise such properties have enabled scientists to mimic this behaviour and allow the development of many novel products that have significant impact on our daily lives.

The two main methods of hydrophilic fabrication can be altered depending on the intended use. The applications of such property are applied across many disciplines. These vary from biomedical devices to paint used in road marking.

Hydrophobic and SHO coatings consist of strong water repelling characteristics that can be applied to various materials to enhance the product with self-cleaning, anti-fogging, anti-reflecting and pollutant adsorbing characteristic. Amongst all applications, the use of hydrophobic and SHO coatings are very much focussed on the purification of water, as there is a severe freshwater scarcity and industrial oil spills are common. The application of hydrophobic and SHO coatings can drastically change our current water treatment methods, as it is found to be very effective in the removal of both oil and salt.

Most of the current materials are being tested on academic scale, it has been observed that the application of hydrophobic and SHO coatings greatly increased the materials’ functionality. As the coatings show great potential, the attention for scale-up is still to be further investigated. However, most hydrophobic production methods are expensive and use environmentally hazardous chemicals, making the scale-up process very challenging. Research into inexpensive, sustainable production methods is needed for feasible scale-up.

Wetting and non-wetting behaviour can be further investigated in terms of membranes to identify their potential use in filtration of water, aqueous solutions, and oil/water separation. This could potentially result in separation step that allows the processes to occur at lower energy consumption, and thus lower costs.

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