Review

Self-Driven Sustainable Oil Separation from Water Surfaces by Biomimetic Adsorbing and Transporting Materials

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Abstract: Oil films on water are an increasingly major contamination problem worldwide. In 2020, we published a novel adsorption and transportation technology for oil–water separation based on biological role models like the floating fern *Salvinia*. This application provides an unexpected ability for the fast and efficient removal of oil films, particularly in ecologically important freshwater biota. A single small Bionic Oil Adsorber (BOA) with 1 m^2 functional textile can collect up to 4 L of oil per hour, which equals about 100 m^2 of oil film from a water surface into a collecting vessel. This is a safe, fast, and sustainable solution for the ubiquitous contaminations of, e.g., fuel oil in freshwater environments. Here, we present updated, new experimental data, and a review of the literature published since.

1. Introduction

Over the last two decades, global oil production, oil consumption, and the risk of oil pollution have gradually increased [1]. In 2022, global oil production amounted to 4.4 billion tons [2]. During the extraction, transport, and use of oil, accidents occur frequently, causing severe and sometimes irreversible pollution of the environment and harm to humans [3,4].

There are various methods to remove oil spills from water surfaces. First, the pollution is usually contained by oil booms which prevent further spreading and the oil can then be burnt in situ. Furthermore, dispersants can be applied, which break up the oil film into fine droplets that are decomposed by bioremediation. It is also possible to use sorbents, by which the oil is absorbed and thus separated from the water. Various types of skimmers can be used to mechanically remove oil from the water surface. All methods have various deficiencies that make their use difficult and especially limit the removal of oil from inland waters.

For many technical applications, unexpected solutions come from the field of biology. Millions of years of evolution led to optimized surfaces of living organisms for their interaction with the environment [5,6]. Solutions-often rather unfamiliar to materials scientists and difficult to accept [7]. Our long-time routine examination of around 20,000 different species showed that there is an almost infinite variety of structures and functionalities. Some species in particular stand out for their excellent water repellency. Further tests showed that these often have very good oil adsorption properties [8,9]. It was shown that, e.g., leaves of the floating fern *Salvinia molesta* (Figures 1 and 2), adsorb oil and are able to transport it on their surfaces. It was shown that these structures are also able to release the

oil again, allowing it to be transported into a vessel and thus enable the separation and collection of oil [8].

Figure 1. Adsorption and transport of a crude oil droplet from a water surface by a leaf of *Salvinia molesta*. The oil is adsorbed completely within seconds [8]; see Video S1 in the Supplementary Materials.
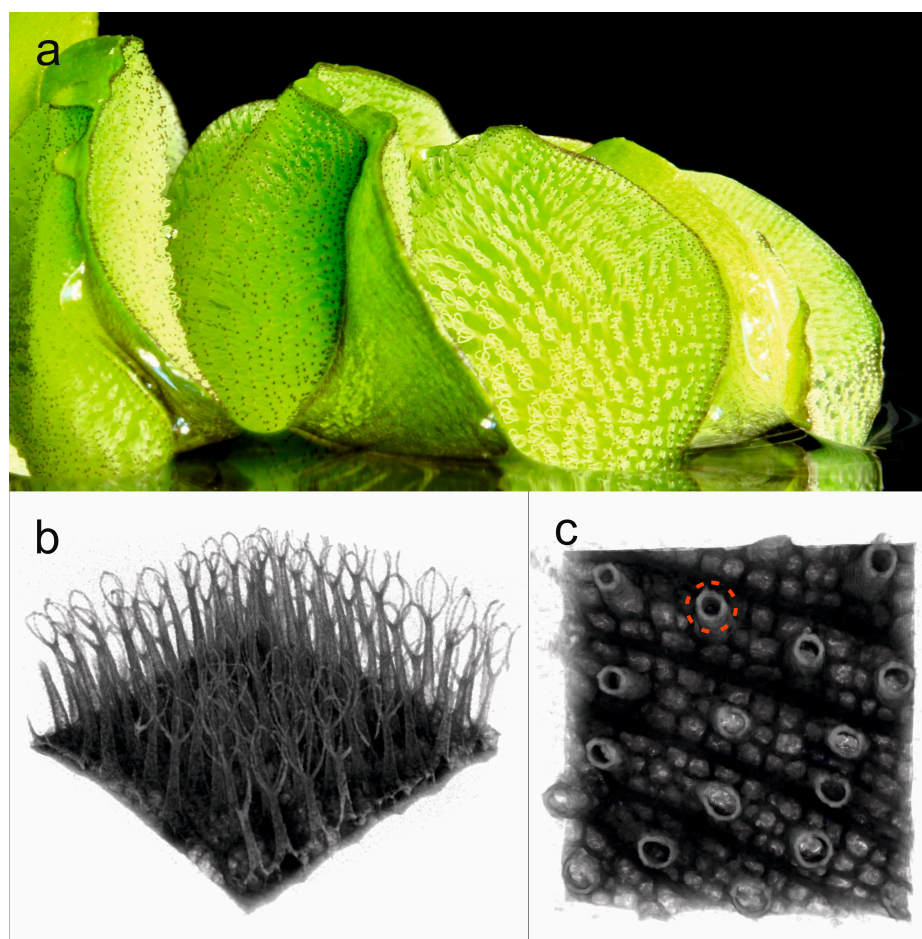

Figure 2. (**a**) *Salvinia molesta*; the floating leaves of this floating fern show a most complex, hierarchically structured, superhydrophobic surface with excellent oil adsorption properties. (**b**) The section of a micro-CT scan of one of the *Salvinia* leaves shows the eggbeater-shaped surface structures in more detail. (**c**) A micro-CT cross-section parallel to the *Salvinia* leaf surface shows that the surface structures at the base can be approximated as circles (red dotted line), which was used for the 2D simulation of the oil adsorption and transport abilities on these surfaces in [10].

The investigation of these structured surfaces shows that the oil adsorption rate depends on various factors. For example, it depends on the viscosity of the oil, the

hierarchical micro-architecture (e.g., size, shape, and distance of the microstructures (hairs)), and the wetting behavior of the surface. The observations, particularly in *Salvinia* leaves, inspired to transfer the effect to technical surfaces for the separation of oil and water, which enables a non-toxic, easy-to-use, and versatile process for a sustainable oil spill clean-up without additional energy input and waste material. This enables a novel physical method to remove, e.g., oil films from water surfaces and allows the development of a device for bioinspired oil–water separation. We called the device that functions according to this principle a Bionic Oil Adsorber (BOA) [8]. Here, we present further new results of the unexpected fast and efficient oil adsorption and transportation based on our research project carried out in 2019–2022 [9], in which we investigated biological surfaces as well as different technical textiles including the result from a flow simulation project of the surfaces [10].

In the two-dimensional simulation, the surface structure of *Salvinia* was approximately modeled by cylinders of uniform diameter and spacing. This led to the conclusion that the material for the biomimetic textiles should be superhydrophobic and completely wettable with oil (contact angle to oil $\leq 20°$). Additionally, the cylinder diameter and the ratio of cylinder distance to cylinder diameter should be adapted to the oil viscosity. These data serve as the basis for the development of a biologically inspired textile by which the oil–water separation can be transferred to engineering [10]. We refer to it as BOA-Textile.

Apart from the BOA, there are various scientific approaches to developing an oil–water separation process by adsorption with superhydrophobic surfaces. In this publication, we present a technical implementation of the BOA and provide a brief overview of the structures and materials used in these processes.

2. Bionic Oil Adsorber

On the basis of the biological investigations and computer simulations described above, various textiles were selected with which the biological structure can be reproduced. A total of 54 different textiles were examined, with a selection of different two-dimensional (2D) and three-dimensional (3D) textiles, such as three spiral structures, two spacer textiles, and eight velours. Spiral structures have an open mesh structure with spirals of hydrolysis-resistant polyethylene terephthalate (PET) joined together on the left and right side. One spiral structure was finished with a nonwoven. The spacer textiles consist of two cover surfaces that are maintained at a distance by pile yarn. They are produced using the warp knitting process on a raschel machine. The velours include both woven structures and tufted materials. Beneath these core textiles, other textiles like nonwovens with needled fiber batts were investigated. Velour, in an optical manner, was comparable to the biological role model, but the density of the pile yarn was not as high as the density of hairs on the surface of *Salvinia*. The knitted spacer textiles with two main surfaces show a lot of fiber batts as contact surfaces.

Two-dimensional textiles were mainly considered to better understand the transport processes. With 3D textiles, the biological models can be reproduced better. Primarily polymers were used as raw materials, with a focus on polyesters and polyamides. The textiles were hydrophobized with a layered silicate in a dip-coating process. By investigating the adsorption speed and the possible volume flow for oils with viscosities of 4.5 cP to 196.7 cP (at 20 °C), it was found that the highest transport speeds and volume flows were achieved with knitted spacer fabrics. The maximum volume flow is 4 L per hour by use of 1 m^2 functional textile and diesel as test medium. This textile can be produced industrially and is therefore easily scalable.

Subsequently, a device was developed with which the textile can be applied on bodies of water. For this purpose, the principle presented was analyzed and transferred to a floating oil collector [8].

The textile adsorbs oil from the water surface and transports it into the oil container which can be emptied manually in certain time intervals. Depending on the size and type of the oil contamination, the BOA can be designed in different sizes so different collection volumes can be realized. The whole BOA apparatus consists of various components, the

most important of which is the BOA textile (Figure 3). It adsorbs the oil on the water surface and transports it within the plane of the textile. This allows the oil to flow into the internal part of the BOA, where it desorbs from the textile and is collected in a collection container. This container is removable and can be emptied manually if necessary.
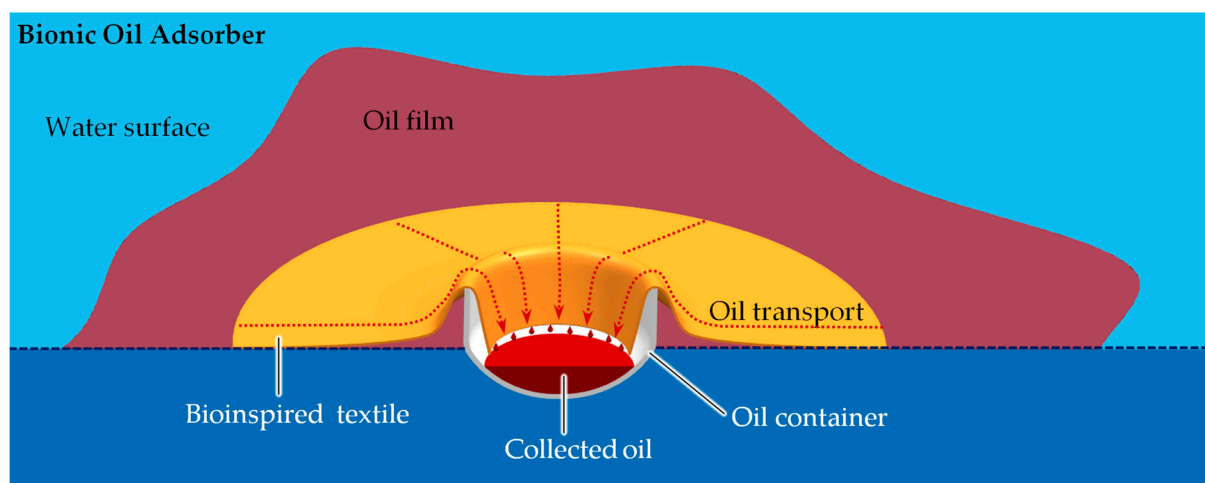

Figure 3. Cross-section of computer-aided (CAD) model of the Bionic Oil Adsorber. The scheme shows an oil film (red) on a water surface (light blue). In the floating container (gray), the textile (orange) is fixed so that it is in contact with the oil film and the end protrudes into the container. The oil is adsorbed and transported by the BOA textile. As shown in the cross-section, it enters the container, where it is released again and accumulates at the bottom of the container. See Video S3 in the Supplementary Materials.

The advantage of the principle is that no additional energy has to be applied to operate the BOA. The oil is separated from the surrounding water by the surface properties of the textile and transported through the textile driven solely by capillary forces, even against gravity. When it reaches the end of the textile in the collecting container, the oil desorbs without any further external influence due to the gravitational forces. At certain points of the textile, the transport takes place at different velocities. This is related to which forces counteract the capillary force F_C. These are mainly the frictional force F_F and the gravitational force F_g, the latter leading to faster transport at the point of desorption. Due to the changing effects of the forces the different force balances F_1 until F_4 develop at different points of the textile (Figure 4). It can be observed that the oil does not immediately drip out of the textile when it reaches the textile edge inside the oil collecting container. Initially, an oil volume builds up inside the textile so that a liquid boundary can be seen above the textile edge (point 4 in Figure 4). This creates a fluid static pressure. If this pressure is greater than the surface tension of the transported oil, a drop forms at the terminal edge of the textile inside the BOA device and the oil desorbs. From this point on, a steady dripping can be observed. During the use of the device, the BOA can float autonomously on any water surface like a buoy and only needs to be maintained at certain intervals, depending on the oil volume and the final dimensioning of the unit, i.e., the capacity.

The different force balances have to be considered in the construction of the final BOA. It is particularly relevant to keep the slope and rise height of the textile from the outside of the BOA into the internal container as low as possible (point 2 in Figure 4).
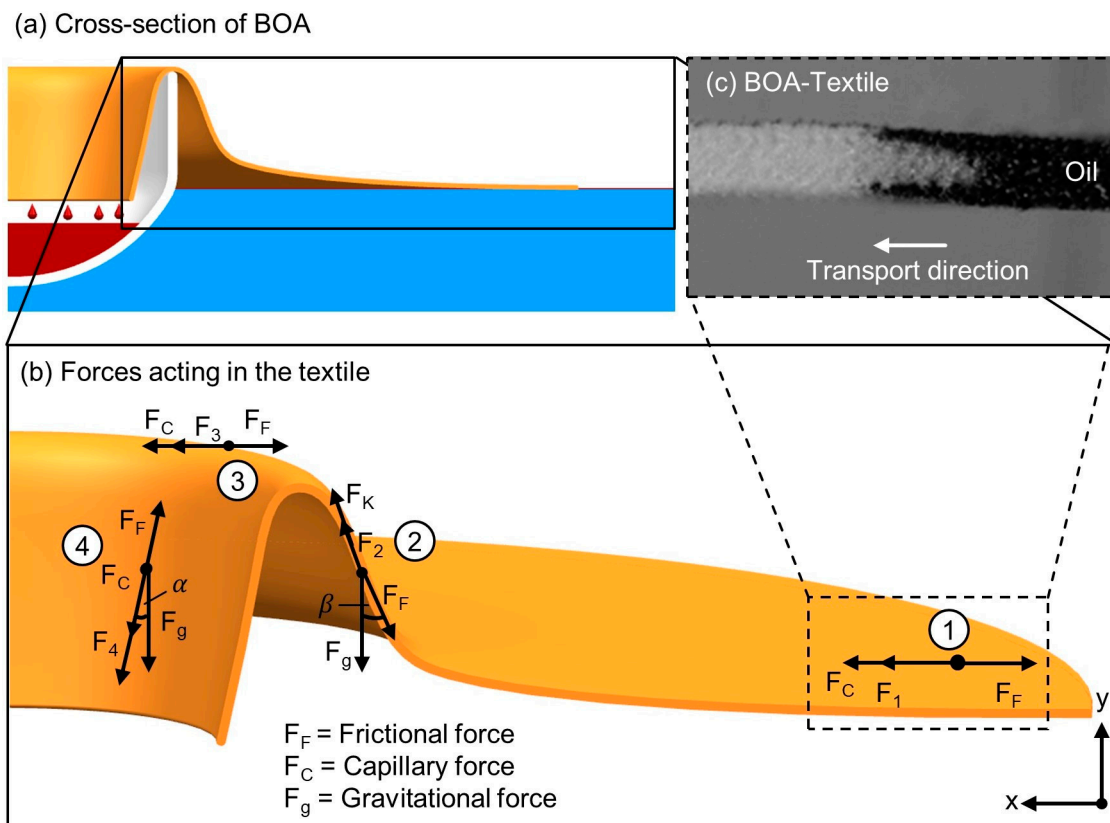
Figure 4. Cross-section of the BOA–CAD model (**a**) with acting forces in the cross-section of the textile at different positions (**b**). The main acting forces are frictional force (F_F), capillary force (F_C), and gravitational force (F_G). The picture above shows the cross-section of the BOA-textile transporting waste oil in horizontal direction (**c**).

Most transport processes of liquids in textiles are based on capillary forces. The flow of liquids through a porous system is considered as a network of many capillaries and can be described by the Lucas–Washburn equation (Equation (1)). The velocity is determined as the derivative of the rise height h from the surface tension γ_{LG}, the capillary radius r, the contact angle θ, the viscosity of the liquid η, the density of the liquid ρ_L, and the gravity g:

$$\frac{dh}{dt} = \frac{\gamma_{LG}\, r\, \cos(\theta)}{4\,\eta\,h} - \frac{r^2\,\rho_L\,g}{8\,\eta} \tag{1}$$

The rise height can also be described as a function of time by solving the differential equation (Equation (2)). Gravity can be neglected if the transport is horizontal, the height of climb h, and the radius r is very small:

$$h(t) = \sqrt{\frac{r_f\,\gamma_{LG}\,\cos(\theta)\,t}{2\,\eta}} \tag{2}$$

If Equation (2) is used to calculate the transport velocity, it corresponds to the velocity of the visually visible flow edge of the oil in the BOA textile in horizontal orientation (point 1 in Figure 4b and picture in Figure 4c).

Preliminary tests have shown that oil can be successfully separated from water surfaces using a prototype of the device. Experiments in an application-related environment, like small harbors or ponds, are pending.

If the oil volume flows measured so far are scaled to the circumference of the textile of 80 cm provided in the application so far, approximately 3 L of diesel can be separated from water by one device of the Bionic Oil Adsorber per hour.

In total, the BOA device offers high added value in contrast to conventional cleaning methods for oil on inland waters. No additional energy requirements, such as with oil skimmers, are necessary, there is no secondary toxicity, such as with oil dispersants, there is high reusability of textiles and equipment, "no waste remains", and it is inexpensive in terms of the amount of oil removed.

3. Other Technologies for Oil–Water Separation

The results regarding the novel bionic oil-adsorbing method presented here are very promising, especially since no additional energy is required and the method is not toxic to the environment. Beyond the BOA, there are other approaches, which were identified in a literature search. For this purpose, we considered the references that cite [8] and, in addition, those found using the search term "oil water separation" on Google scholar (publication year < 2021). The presented and discussed adsorbing materials are foams, sponges, meshes, textiles, and nanoscaled structures.

3.1. Foams and Sponges

A frequently chosen approaches for the production of adsorbers are sponges and foams. For example, conventional foams are equipped with novel coatings to create a superhydrophobic surface within the sponge. For this purpose, Mu et al. coat a melamine foam with a siloxane coating, as this provides alkyl silane on the surface. This results in a water contact angle of 156° [11].

Another approach is based on the use of sponges made of polyurethane (PU), whereby the surface is coated with a mixture of different nanoparticles (e.g., graphite powder, manganese, and iron oxide). Due to the special surface characteristics of individual pores, it is possible to absorb oil from the oil–water interface. After application, oil can be recovered and recycled. Due to the porosity of the sponge, a large volume of oil can be absorbed and stored. As a result, thirty times its own weight can be taken up due the cleaning method is environmentally friendly, the same as the BOA [12]. PU foams can also be functionalized with silicon dioxide and activated carbon in such a way that oils of different viscosities can be adsorbed by water surfaces [13].

By using perfluorotriethoxysilanes, Pang et al. were able to find another approach with which polyurethane foams can be hydrophobized. In a sol-gel process, a mechanically stable coating was produced, which even after 100 compressions showed a contact angle of 144°. The foam had an adsorption capacity of 20–40 times its own weight and the separation rate was 98.72% [14].

The authors of another study [15] also focused on the use of fluorine-based polymers for the production of a superhydrophobic foam. They used a thermally impacted nonsolvent-induced phase separation method to produce a three-dimensional porous monolith on polyvinylidene fluoride (PVDF). This had a water contact angle of 154° and an oil contact angle of 0°. The material has an absorption capacity of 21.3 g/g and can be reused up to five times. It is proposed as a material for large-scale oil spill cleanup.

Venkatesan et al. [16] use bio-based materials by processing leather cuttings into activated carbon at 400 °C and coating a melamine sponge with it. The water contact angle of this material is approximately 158°, whereas without the coating it is approximately 152°. Thus, an adsorption capacity of 75.77 to 141.24 times its own weight could be achieved.

Particles can also be used to functionalize foams. For example, titanium dioxide and graphene oxide are processed into a foam in a hydrothermal treatment and freeze-drying method. In this way, a separation and adsorption efficiency of 98% was achieved [17].

3.2. Meshes and Textiles

Another structure for oil–water separation is meshes and textile materials. These materials can be used to separate oil from water by pressing the mixture by means of pressure through the structures. For the production of superhydrophobic nets, copper, stainless steel, nickel, and titanium are often used as metallic materials [18]. A copper mesh

can be functionalized in a two-step soaking process so as to create a micro-nanostructure on the surface. This allows water contact angles of 158.7° to be achieved [19]. In a further process, nanoparticles were bound to the mesh surface by etching with an ammonia-base titanium dioxide, which resulted in nanostructuring. The surface was then further functionalized with n-hexadecyltrimethoxysilanes (HDTMS) to achieve superhydrophobicity and superoleophilicity The application is planned for use with heavy oil [20].

In addition to the hydrophobic surface chemistry, the production of porosity is also highly relevant for adsorption. For this purpose, a back-propagation neural network could be developed with which the porosity can be adapted to the application of stainless steel meshes [21].

By functionalizing such stainless steel meshes with multi-walled carbon nanotubes through thermal chemical vapor deposition, a three-dimensional porous structure can be created. The water contact angles are between 140 and 150°, whereby oil completely wets the surface [22].

Apart from synthetic materials, natural materials also show a pronounced hydrophobicity and oleophilicity. Hollow fibers of *Chorosia*-fruits are cellulosic materials that naturally exhibit oil adsorption when untreated. Treatment with sodium chlorite ($NaClO_2$) can further increase oil absorption and achieve oil–water separation [23].

3.3. Nanoscaled Structures

Nanofibers are also used to achieve a surface structured at the nanolevel. In the solution blow process, hydrophobic nanofiber membranes made of polybutylene succinate (PBS) can be produced with relatively high productivity (27.0 g/h per nozzle). With these membranes, an oil adsorption capacity of up to 38.5 g/g could be achieved. Since PBS is biodegradable, the membrane can be returned to the biodegradation process by hydrolysis [24].

Currently, nanofibers are produced more frequently by electrospinning than by the solution blow process. This makes it possible, for example, to produce membranes based on polyacrylonitrile (PAN) for oil–water separation. However, Sun et al. [25] use superhydrophilicity, not superhydrophobicity, for this purpose.Other authors were inspired by spider silk, which enabled them to produce a membrane with nanoscale pores (mean pore size 0.634 μm) on the base of PAN. This allows nanoemulsions to be separated by sieving. The membrane consists of two layers, with a nanofiber layer serving as a support layer and a layer of spindle-knotted structures [26]. Multilayer membranes are also made of PVDF. For example, a functional layer of sodium alginate, chitosan, and iron is applied to a PVDF membrane by grafting with polyacrylic acid [27].
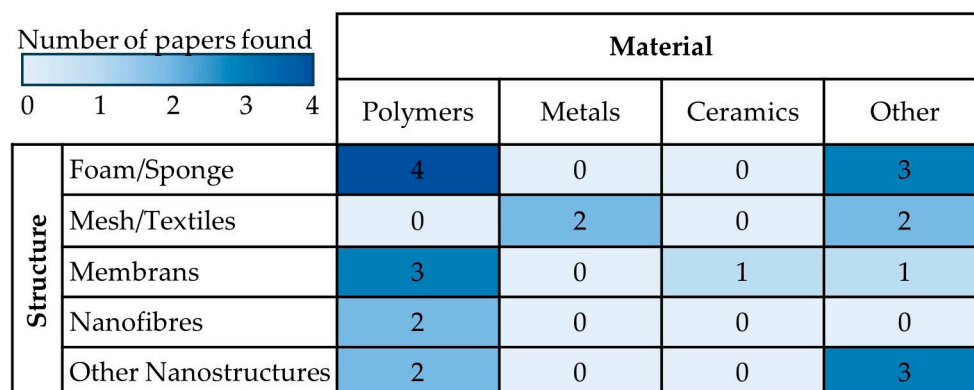
Other methods and materials are also used to achieve structuring at the nanofiber level. For example, nanoparticles can be used to functionalize microfiltration by polyvinylidene fluoride membranes [28]. Other research groups are trying to use fluorine-free materials: Gao et al. [29] use an epoxidized natural rubber latex with polyvinyl alcohol to make a three-dimensional porous material with a nanostructure; Jha et al. [30] prepare superhydrophobic nanotubes from polypyrrole; and Ubah et al. use *Moringa oleifera* to functionalize zerovalent iron particles [31].

4. Discussion

Research is currently being conducted on sponges, foams, nets, textiles, nanofibers, and nanoporous structures as solutions for adsorbing oil from water. In these projects, metals (e.g., stainless steel), polymers, ceramics, and natural fibers are used as adsorbing media (Figure 5) [11–32].

Most research work is currently concerned with the development of oil-adsorbing sponges and foams. This may be due to the fact that these can be produced economically on an industrial scale if development is sufficiently advanced. Currently, many nanostructured surfaces can only be produced on a laboratory scale, which is why further material-specific process development is necessary. Due to this difficulty, other materials

such as textiles still offer an attractive alternative for the production of adsorption materials for oil–water separation.

Number of papers found	Material			
0 1 2 3 4	Polymers	Metals	Ceramics	Other
Foam/Sponge	4	0	0	3
Mesh/Textiles	0	2	0	2
Membrans	3	0	1	1
Nanofibres	2	0	0	0
Other Nanostructures	2	0	0	3

(Rows grouped under "Structure")

Results including relevant research that cited (Barthlott et al. 2020) and papers found in Google scholar for „oil water separation", publication year < 2021

Figure 5. The graph shows the number of publications found, divided into structures and materials, using the Google scholar search engine for the search term "oil water separation" since 2021 and additionally those sources that cite the paper [8].

Fundamentally, it can be observed that all methods are based on a hierarchical structuring of the adsorption materials on the micro and nano levels. This is an attempt to create a Cassie–Baxter stage to achieve a stable and distinctive hydrophobicity.

Concerning materials, mainly metallic and polymeric materials are used, as this is an application area with high requirements, such as resistance to oils, salt water, and UV radiation. Due to these requirements, it is obvious to use perfluorinated materials. These are still widely used in industry for hydrophobization, but they have a bioaccumulative effect and are heavily regulated in the EU, for example [33]. Therefore, research should pay attention to the use of other resistant materials. Considering bioeconomy and circular value creation, it is reasonable to increasingly use bio-based raw materials. There are already some attempts to use adsorption materials made of wood or biodegradable polymers [34].

The wetting property of the adsorbent material is essential for the separation of oil and water. Almost all approaches found are based on superhydrophobicity. The highest water contact angles (WCA) are reported for melamine foams and copper meshes (approximately 158°). In comparison, the WCA of *Salvinia molesta* is even higher at 164°, which can be explained by its hierarchical structure. The WCA of the BOA textile coated with layered silicate is also slightly lower (approximated 152°). This indicates that the biological model has not yet been fully transferred and other technical solutions, such as those presented in the literature, can still be improved by bionic approaches. It may be worth optimizing the wetting properties to improve the separation rate.

The described materials for oil–water separation can be used for different separations depending on their ability to transport oils of different viscosities. For the BOA, oil viscosities from 4.5 cP to 196.7 cP (at 20 °C) were investigated and the BOA textile proved to be functional in this range. Other materials found in the literature are mostly tested with gasoline, diesel oil, kerosene, and soybean and other vegetable oils. These normally show a low viscosity around 5 cP at room temperature. It appears to be particularly challenging to separate highly viscous oils. High viscosities are found particularly in accidents involving crude and heavy oils.

5. Conclusions

As mentioned above, oil spill contamination on water are a major environmental problem worldwide. The technologies previously cited and described deal with the solution to this problem, and as shown, the approaches are very diverse. All methods based on

adsorbent materials have in common that these materials achieve separation by wetting properties and achieve adsorption by a porous surface. The latter creates a capillary system that is filled by the oil. For hydrophobicity and porosity, it is favorable to use a multi-hierarchical surface. This is mainly achieved by a macrostructure, such as a mesh or foam, and a complementary coating, such as nanoparticles.

The novel method of the Bionic Oil Adsorber presented here is also based on a hierarchical structure, the BOA textile, which has been hydrophobized by a coating. But it fundamentally differs from all published approaches: no external energy needs to be supplied for operation, it can be used multiple times, and no toxic substances are introduced into the water body and the oil is sustainably collected in a removable collector. This makes it cost-effective and sustainable. In this publication, we were able to show quantitatively for the first time that the adsorbing and collecting potential is larger than expected, and that the BOA is an important addition to the existing approaches and a very promising technology for removing oil from waters in an environmentally friendly and cost-effective way in the future.

The BOA device can be used preventively and in acute emergencies. Its design can be adapted to the size of the oil spills. It is easy to handle, can be used independent, and does not need any energy supply. Thus, it offers an interesting alternative to existing oil–water separation processes.

Self-driven oil separation from water surfaces by biomimetic adsorbing and transporting materials probably differs from all the existing technologies by its fast, self-driven, automatic, and completely sustainable removal of the contaminant from a water surface.

6. Future Directions

All presented materials should not only be produced efficiently but also be used efficiently. To this end, it is desirable to make them reusable on the one hand and to use digital developments such as artificial intelligence on the other, in order to be able to use oil removal methods in a targeted manner [35].

The BOA and other adsorbing materials contribute to the achievement of various Sustainable Development Goals (SDGs) of the UN. These are primarily Clean Water and Sanitation (Goal 6) and Life Below Water (Goal 14). Indirectly, it also contributes to Zero Hunger (Goal 2) and Life on Land (Goal 15), as clean water is a highly important component for food security and a healthy life on land for people and animals. However, in the production of the textile and the entire product, all other SDGs should also be considered. Especially in the textile process chain, there is often exploitation of workers, which should be avoided at all costs.

In addition, there are some research groups working on smart materials for oil–water separation. These are divided into stimulus-responsive and prewetting-induced types. The former can switch their wetting properties between superhydrophobic and superhydrophilic, whereas the latter can separate either oil or water from mixtures without external stimulus. Similar structures and materials are used for this as for the other developments presented [36].

With regard to this future work, inspiring role models can be found in nature that enable the rapid development of new materials and processes. One of the best examples continues to be *Salvinia* [37]. Moreover, as early evolutionary structures, terrestrial cyanobacteria like *Hassallia byssoidea* can offer exciting impulses for the design of surfaces that are fundamentally amenable to oil–water separation due to superhydrophobicity [6].

Adsorbent textiles can also be used in other areas for oil–water separation. For example, emulsions in industrial applications or in the automotive sector could be purified so that they can either be reused or properly disposed of. As this is a very large field of application apart from oil spills in marine environments, the application of the developed adsorption materials should be tested in this field as well.

Oil films on water are a major global environmental threat. We could prove, that the novel biomimetic BOA technology for a self-driven separation and automatic collection

of oil films including their complete removal from water is surprisingly highly efficient and sustainable. Moreover, it can possibly be applied in different related separation processes. The technology differs from all existing approaches that have been proposed to date. However, further work is necessary to adapt and scale up the collectors for different applications and scales: a promising task for future applied engineering projects.

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