



(BIO)FOULING AND ANTIFOULING MEASURES

Fouling: an overall issue

The term fouling refers to the accumulation of unwanted material on a surface, with the result of reducing the efficiency and functionality of the surface and/or of the device it belongs to. Fouling affects many more fields than one would expect – medical, marine and industrial – always creating severe losses of money. Here some examples are presented, as well as some methods adopted as fouling countermeasures, also mimicking ingenious strategies derived from nature

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Introduction

In its broadest sense, fouling is any accumulation of unwanted material on a surface, which causes a side effect or impairs the functionality and efficiency of the surface and/or of the device it belongs to.

Several types of fouling and their combinations may occur: 1) crystalline or precipitation fouling, 2) corrosion fouling, 3) particulate fouling, 4) chemical reaction fouling, and 5) biological fouling or biofouling. Biological fouling results from a) development of a biofilm consisting of microorganisms and their products (microbial fouling), b) deposition and growth of macroorganisms (macrobial fouling), and c) assorted detritus. Microbial fouling usually precedes colonization of the surface by macroorganisms.

The importance given to fouling phenomena is ultimately due to the fact that they result in severe energy losses, either if the deposits increase the fluid frictional resistance at a surface or impede the flow of heat across surfaces, or of a fluid across membranes, or increase the rate of corrosion at a surface [1].

A remarkable number of papers in scientific literature deal with the problem of fouling, reflecting the fact that

many are the fields in which this phenomenon creates concern. Just to mention the most remarkable, fouling affects the long-term functionality of implantable bioelectronics and malfunction of biosensors in the medical field, while in industrial applications it can give unwanted effects in power plants (e.g., geothermal), water-treatment systems (e.g., for desalination or wastewater reclamation), heating exchangers, sensors (and other devices) used for river and marine monitoring and even in the food processing industry.

This paper summarizes the principal aspects related to fouling in various fields, with the aim to give an overview of the problem and of the methods adopted as countermeasures.

Devices for environmental applications

Sensors for environmental monitoring

Typically, water quality is assessed by monitoring parameters such as pH, conductivity, dissolved oxygen, temperature, turbidity, nitrate and phosphate concentrations.

A variety of sensors is available for these purposes and a wide range of antifouling measures must be developed to ensure that sensor performance is not impeded by biofouling (e.g., biofilm formation on the glass membrane - a specially formulated, ultrathin glass - of the proton-selective electrode used for *in situ* pH measurement).

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Generally, for marine and riverine sensors, biofouling decreases the operating lifetime and increases the cost of maintenance of the sensor, since the latter must be removed from the sampling location to be cleaned. Biofouling will also introduce a degree of error into the collected data, e.g., if a fluorimeter is used to quantitate the chlorophyll concentration in water, accumulation of other absorbing species on the sensor will reduce the amount of light which can be absorbed by the analyte. Biofouling also poses problems for the platforms on which the sensors are deployed.

Sensors employed for marine and riverine monitoring primarily undergo aquatic biofouling, which comprises four stages: i) adsorption of a conditioning layer, ii) adhesion of bacteria, iii) growth of a biofilm and iv) macrofouling.

Among the methods used in the past to combat fouling there is the mechanical cleaning by high pressure water jets, but it is not suitable for delicate sensor components; chlorination has also been used, but it has been shown that byproducts of chlorine in water include carcinogenic compounds such as trihalomethanes.

As a consequence of Tributyltin banning, due to its extreme toxicity, research on antifouling coatings has focussed on two different types of materials. The first type, non-stick coatings, resists adhesion by fouling organisms, thus preventing the growth of biofilms at a surface; they are materials with low surface energy, usually silicones and fluorinated polymers.

The second type of materials is prepared by incorporating a compound, which is biologically active against those organisms settling on the surface (antimicrobial activity).

Mechanical antifouling methods are a more benign approach to antifouling than leaching of biocides from surface coatings into the water. The U.S. Navy patented an oceanographic sensor, which vibrates upon excitation by an electric potential, thus removing fouling material from the surface [2]. However, the power required is quite high and this makes it unsuitable for use in battery-powered remote sensors; moreover, the sensitivity of the sensor can be decreased as a result of the coating.

Alternatively, the sensor can be exposed for the minimum time required to sample, and then the sensor is

removed from the fouling environment [3].

Electrochemistry can also be used to kill fouling organisms, e.g., the generation of chlorine and hypochlorous acid by electrolysis of seawater has been proposed as a method for preventing marine sensors from fouling [4]. Otherwise, electrochemistry can be used to kill microorganisms by direct transfer of electrons from the electrode to the fouling organisms [5].

Another method used as antifouling is the irradiation of surfaces, e.g., ultraviolet light has been used on marine sensors, but also on filtration membranes, valves, intake gratings and also for wastewater disinfection [6]. However, this method is not practical to use with remote sensors due to demanding power requirements. Instead, no energy supply is needed when coating surface with a photocatalytic material, for the photocatalytic inhibition, e.g., of algal growth [7]. Photolysis of water in the presence of the zinc oxide photoactive material leads to the formation of hydrogen peroxide, a known toxicant [8].

Laser irradiation was also investigated as a means of preventing biofouling by barnacles and diatoms [9] and ultrasonic irradiation for control of biofilm formation on glass tubing [10], and low frequency sound, too, has been tested to prevent zebra mussel fouling.

As far as sensors are concerned, a strategy is to render the membrane more hydrophilic, e.g., by polymerization of the surfaces. The interaction of the cationic polymer chains with negatively charged areas on the bacterial cell membrane is claimed to explain the efficacy of the treatment.

Also hybrid organic/inorganic reverse osmosis membranes, containing aromatic polyamide thin films underneath titanium dioxide nanoparticles, have been tested to inhibit membrane fouling [11]. TiO_2 photocatalysis is known to generate various active oxygen species, such as hydroxyl radicals and hydrogen peroxide that kill bacteria by destruction of the bacterial cell membrane.

The ideal antifouling strategy for sensors would provide a low cost, easily implemented, environmentally benign solution to fouling, which would allow sensors to operate unattended for a sufficient time span, but at present the methods described above cannot satisfy all of these criteria. Further research is needed in deter-

mining the long-term environmental effects of substances tested to this aim, and to completely understand the mechanism of action of many naturally antifouling compounds [12].

Permeable reactive barriers

The permeable reactive barrier (PRB) is a passive treatment technology used to treat contaminated groundwater. PRBs are generally used for long-term treatment (decades) and during their lifetime fouling caused by mineral precipitation is a major concern. Fouling causes loss of pore space and reactive surface area of the reactive medium, consequently flow paths and residence time can be altered, thus influencing the effectiveness of the barrier. Changes in residence time are particularly important, as contaminants must remain within the reactive medium long enough to ensure that the treatment will effectively react with contaminants [13].

Most PRBs use granular zero-valent iron (ZVI) to create redox conditions, resulting in degradation or immobilization of chlorinated solvents and herbicides, heavy metals, and radionuclides. The involved reactions also cause the precipitation of secondary minerals, such as iron oxides, (oxy)hydroxides and carbonates [14]. Accumulation of minerals in ZVI reduces the porosity and hydraulic conductivity, affects the surface area for reactivity, and alters flowpaths, resulting in preferential flow and/or blockage of flow [15]. The rate of porosity reduction is a function of the ground water chemistry and flow rate, with greater amounts of minerals accumulating when the inflowing ground water has higher concentrations of dissolved mineral-forming ions [16]. Simulations of ground water flow and reactive transport have been used to evaluate how mineral fouling may affect the hydraulic behaviour of PRBs over decades of continuous flow in carbonate-rich alluvial aquifers. Results of the simulations show that a little change in hydraulic behaviour occurs within 10 years from the time of installation, which is consistent with field experience to date. Significant changes in hydraulic behaviour should be expected after ~30 years due to larger reductions in porosity and hydraulic conductivity. After 50 years, large regions of PRBs may become clogged and the PRB is likely to become less permeable than

the aquifer, resulting in appreciable bypassing of the barrier by groundwater.

Li and Benson [17] proposed some strategies to limit the impact of fouling in PRBs. Residence times are less affected by mineral precipitation when a pre-treatment zone is employed. pH adjustment limits the total amount of hydroxide ions in groundwater to reduce porosity reduction and to retain larger residence times. Larger ZVI particles reduce porosity reduction as a result of the smaller iron surface area for iron corrosion, and retain longer residence time. Mechanical treatment redistributes the porosity uniformly throughout the PRB over time, which is effective in maintaining the residence time. These findings are predicted with numerical models, additional research and monitoring are necessary to confirm that the performances anticipated can be used in practical in situ application.

Membranes fouling

Reverse osmosis for desalination

Problems with water are expected to grow worse in the coming decades, therefore, many researchers have focused on methods suitable to obtain freshwater by saltwater desalination and water reuse to sustain future generations. The reverse osmosis (RO) technology is considered as a promising solution and is gaining worldwide acceptance at present [18]. RO is a pressure-driven process whereby a semi-permeable membrane (i.e., RO membrane) rejects dissolved constituents in the feeding water while allowing water to pass through. The progress in RO technology is greatly dependent on the development of RO membranes, which has become both possible and practical after the invention of the thin-film composite (TFC) aromatic polyamide membrane.

Despite its many advantages, one of the obstacles to the widespread use of TFC polyamide RO membrane is the proneness to fouling [19]. Fouling is a process where solutes or particles in feeding water deposit onto RO membrane surface in a way that causes flux decline and affects the quality of the water produced. This will inevitably make the operation difficult and decrease the membrane lifetime, which will be translated into higher costs.

To prevent RO membrane fouling, a number of methods

for antifouling RO membranes have been developed, including the selection of new starting monomers, the improvement of interfacial polymerization process, surface modification of conventional RO membrane and the incorporation of inorganic particles [20].

There are mainly four types of foulants in RO membrane fouling: inorganic (salt precipitations such as metal hydroxides and carbonates), organic (natural organic matters such as humic acid), colloidal (suspended particles such as silica) and biological (such as bacteria and fungi). Physicochemical properties of RO membrane surface, such as hydrophilicity, roughness and electrostatic charge, are major factors influencing the membrane fouling [21].

The development of fouling-resistant RO membranes takes these major factors into account.

Increase in hydrophilicity offers better fouling resistance since many foulants, such as protein, are hydrophobic in nature [22].

A smoother surface is commonly expected to experience less fouling, presumably because foulant particles are more likely to be entrained by rougher topologies than by smoother membrane surfaces [23].

Finally, surface-bound long-chain hydrophilic molecules (e.g., polyethyleneglycol) are very effective in preventing the adsorption of macromolecules, such as protein onto membrane surface, due to the steric repulsion mechanism [24].

Most research is aimed to face the factors listed above, e.g., by the introduction of hydrophilic layer, the reduction of surface roughness, the improvement of charge property and the utilization of the steric repulsion effect. Nonetheless, fouling cannot be thoroughly prevented, since there are no membranes that are free from fouling under any circumstances [22].

Reverse osmosis and nanofiltration for effluent reclamation

RO is also increasingly used, together with nanofiltration (NF), in the advanced treatment of municipal secondary effluents for the production of high-quality reuse water [25]. However, membrane fouling is a major obstacle in the development of membrane technology in this field.

These systems undergo fouling occurrences similar

than the RO membranes described above, but their nature is linked to the particular media treated. Thus the main fouling agents are: effluent organic matter (EfOM), microbial and inorganic membrane fouling.

EfOM represents a large group of structurally complex, heterogeneous, and poorly defined organic compounds [26].

Biofouling originates from the following processes: microorganisms irreversibly attach on the membrane surface and then grow, reproduce, and secrete substances by utilizing the nutrients in wastewater before a biofilm is finally formed [27]; this biofilm decreases the membrane flux, increases the transmembrane pressure, and causes the membrane biodegradation and salt flux increase [28].

Colloidal natural organic matter, colloidal calcium phosphate, and sometimes colloidal silicates are the main components of the inorganic foulant, all of which have great affinity towards aggregation with one another.

These fouling processes and their interrelations are still poorly understood, so further studies are necessary to examine their mechanism, identify their properties, and take the relevant control measures.

Energy production and delivery

Geothermal plants

Geothermal energy is one of most promising energy supply source and many geothermal power stations have been set up and operated in several countries, furnishing houses and industries with energy.

The present challenge is to continue to lower production costs without compromising safety, in order to remain competitive with other power sources. Among the factors involved in lowering the cost of geothermal utilization, significant fouling and corrosion are two control issues that have not been satisfactorily settled [29]. Scaling (term used to indicate mineral fouling) and corrosion of highly saline and corrosive geothermal water are often observed within plants or in reservoirs in which the cooled fluid is reinjected into formations, thereby decreasing the fluid flow by clogging the pipes of the plant and the pores of the rock. Fouling simultaneously results in an increase in fluid resistance, as well as extra energy consumption and wastewater dischar-



ge; furthermore, an incomplete fouling layer can lead to local corrosion [30].

The most corrosion - and scaling-relevant compounds in geothermal fluids are scales of carbonates, silica, sulfides, oxides and also soluble salt minerals (halite) originating from, e.g., evaporite formations [31].

Among them, the main contributors to geothermal fouling are the scalings of silica and calcium carbonate, since they are primary components of the earth's crust. Calcium scaling in geothermal plants is largely driven by pressure reduction through fluid transmission devices, thus the geothermal hot water scale deposits onto heat transfer surfaces of heat exchangers and onto the surfaces of the flowing conduits. When the pressure of the brine solution decreases rapidly, CO_2 gas is evolved from the brine due to its decrease in solubility. This increases the pH of the brine and causes the deposition and crystal growth of calcium carbonate. The kinetics of this reaction is very fast, causing scale formation immediately downstream of such pressure drops and the plugging of, e.g., valves, pressure taps and flow instruments. Calcium carbonate is also found on heated surfaces (see next paragraph), since its solubility decreases as temperature increases (retrograde solubility). For silica scale, the deposition mechanism is more complicated than that of carbonate. Silica solubility increases as brine temperature increases (prograde solubility), and is saturated in geothermal brines in the downhole environment. Consequently it can become supersaturated as the brine is cooled through the heat exchange, or when part of the brine is flashed into steam. Supersaturation causes the precipitation of silica in an amorphous form on heat exchanger surfaces, separators, well lines and discharged lines. The scale formed by silica is hard and not easily removed by mechanical or chemical methods.

The scale so deposited deteriorates the heat transfer capability dramatically and, at the same time, it remarkably increases the pumping power needed to flow geothermal hot water, to the detriment of the stable and long operation of the system.

Several technologies for inhibiting fouling have been developed over the past decades based on the fouling and corrosion categories and severity, including crystallizer-clarifiers scale inhibitors [29], plant and

fitting material selections, electrical submersible pumps [29], steam cleaning and various coatings, such as polyphenylenesulfide-based, or epoxy resin [32], or SiO_2 on copper substrate [30].

Even if many improvements have been achieved so far, further work is still needed to protect the plant components against corrosion, oxidation, and scaling in the harsh, hostile geothermal environment, and to develop a system for the effective use of this natural "high density" energy.

Mineral fouling in heat exchangers

As mentioned for the geothermal plants, mineral fouling (scaling) is also experienced in heat exchangers, especially with the use of cooling water systems. It is the deposition of precipitated mineral salt crystals on a heat transfer surface. The formed fouling layer decreases the thermal efficiency of the heat exchanger, increasing the operating cost. Fouling demands billions of dollars annually for cleaning and maintaining the equipment: studies show that 1 mm limestone deposit could double the energy consumption in a heat power plant [33 and references therein].

When a heat pump is used as an air-conditioning system, the outside heat exchanger is used as a condenser, where heat has to be rejected to the surroundings. The mineral ions contained in circulating water are accumulated, and their concentration increases with time, creating fouling problems. The precipitated solids form both soft and hard scale deposits on the heat transfer surfaces, increasing the resistance to heat transfer and subsequently decreasing the thermal efficiency of the equipment.

The concentration of fouling materials (foulants), temperature, pH, pressure, time, flow velocity, mechanical motions, radiation, and impurities are factors affecting nucleation and subsequent crystal formation.

Fouling can be "soft" and "hard": the former is due to particulate accumulation, prevalently particulate matter, bacteria, corrosion products and so on [34], the latter is due to mineral crystallization, mostly calcium carbonate.

To date, chemical treatments have been the most effective approach for scaling prevention, however water pollution may derive from the chemicals employed.

Alternative methods have been tested and proposed, e.g., the use of oscillating electric field and of devices such as permanent magnets, solenoid coil device, high-voltage electrode [35], electro-flocculation mechanisms [36] and [33].

Medical devices affected by fouling

Medical biofouling occurs in areas such as prosthetic implants, biosensors, catheters, dental implants and medical equipment, and can cause problems such as implant rejection, malfunction of biosensors and spread of infectious diseases. As far as medical implants are concerned, more than 45% of hospital-contracted infections are linked to biofilm-infected medical devices. For instance, catheters are the most commonly used medical device and the second highest cause of infection [37].

Biofouling in these cases is due to the adhesion of proteins or microorganisms (biofilm) to the device and begins soon after implantation. Treating biofilms on infected medical devices often requires surgical replacement, which increases the risk of mortality and antibody resistance.

The affected medical devices can be permanent (implanted and intended for long-term use) or temporary (intended for short-term use). Permanent implant devices include biosensors, heart valves, bone plates, fasteners, orthopaedic implants, dental implants, pacemakers, drug-delivery devices and ventilation tubes [38]. Immediately after surgery, the permanent implant is flooded with blood followed by the adsorption of proteins onto the surface [39]. Such adsorption on a biosensor may lead to sensor 'blindness', reduced lifespan and increased power consumption. Mechanical heart valve biofilms can lead to tissue inflammation from microorganisms, which can also enter the bloodstream by the surrounding skin or other devices. A severe trauma often requires bone plates and fastener implants, that are susceptible to biofilm formation because of the high concentration of microorganisms in the contaminated wound area, and once infected they generally require removal [40].

Temporary implant devices include biosensors, ca-

theters, drug-delivery devices, bone plates, fasteners, needleless connectors and ventilator tubes [38]. The most common biosensor is the single-use blood glucose monitoring device for diabetic patients, this device operates through a membrane, where biofouling starts upon bodily contact when micro-organisms, proteins and other components adhere to the surface, impeding the sensor's diffusion ability. Failures of this biosensor can be also caused by fibrous encapsulation, electrode passivation and biodegradation [41].

Furthermore, urinary catheter calcification from bacterial colonization may cause bladder stone formation and urinary tract infections [42]. Pulmonary, transdermal, intravenous and subcutaneous drug delivery implanted devices are limited, owing to biofouling of electrode surfaces or membranes.

Needless to mention how important are the effects of fouling in the medical field, since, in addition to huge losses of money, in this case risks are posed for human health.

Conclusions

Although the most widely known form of fouling is found in the marine environment - where biofouling colonizes ships, buoys, offshore structures, oil installations, cables, etc. - a large number of other fields are affected by this phenomenon. Fouling is recognised as a most critical factor affecting natural aquatic systems, water distribution systems, wastewater treatment systems, heat exchangers, fuel consumption by ships, and even human health.

The development of antifouling methods is an important research path and has attracted wide attention in recent years. To achieve effective solutions, fouling has to be tackled in terms of the fundamental physical, chemical, and biological processes involved, as well as by analysing its influence on energy losses and stimulating fundamental investigations on the relevant topics. Continued work in this research field is expected to deliver cheaper, more reliable solutions to this age-old problem, also drawing inspiration from nature, where flora and fauna demonstrate a multitude of antifouling lessons that can be mimicked for engineering purposes. ●

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