

New approaches to characterizing and understanding biofouling of spiral wound membrane systems

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ABSTRACT

Historically, biofouling research on spiral wound membrane systems is typically problem solving oriented. Membrane modules are studied as black box systems, investigated by autopsies. Biofouling is not a simple process. Many factors influence each other in a non-linear fashion. These features make biofouling a subject which is not easy to study using a fundamental scientific approach. Nevertheless to solve or minimize the negative impacts of biofouling, a clear understanding of the interacting basic principles is needed. Recent research into microbiological characterizing of biofouling, small scale test units, application of *in situ* visualization methods, and model approaches allow such an integrated study of biofouling.

Key words | biofouling, extracellular polymeric substances (EPS), microbiology, modelling, nanofiltration (NF), reverse osmosis (RO)

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INTRODUCTION

Surfaces in contact with water will inevitably be colonized by bacteria forming a biofilm layer. Membrane units are therefore intrinsically excellent systems for bacteria to colonize. Not surprisingly, biofouling is an important problem in the application of membrane filtration processes for water treatment. Biofouling has been, and still is, an important research topic which until now has not been solved. This is largely due to the very complex nature of the problem involving physical, chemical and biological aspects in a complex geometry. Partly because of the complexity of the problem, biofouling studies have a rather anecdotal nature

and the information obtained from pilot and full scale installations is rather anecdotal. Biofilm growth occurs on each surface in contact with water, even in ultrapure water systems (Chicote *et al.* 2004). In principle, for membrane filtration systems, biofilm growth is not equal to biofouling. Only when biofilm growth leads to a significantly decreased membrane flux, a much increased operational pressure of the system or a decrease in retention properties of the membrane is it called biofouling. In other words, while a biofilm has a clear definition, biofouling is related to operational practises. A further problem is caused by the fact that

water quality is not feasible for biofouling characterization. In addition, sampling and analysing membrane units is not easy. Membrane processes are typically black box systems that can only be studied effectively by an autopsy at the end of a filter run. Therefore there is a need for well defined tools allowing the assessment of the complex interactions occurring in such process units. Finally, biofouling in low pressure membranes (microfiltration (MF), ultrafiltration (UF)) and high pressure membranes (nanofiltration (NF), reverse osmosis (RO)) cannot be directly compared. This is due to the fact that different fouling mechanisms play a dominant role in these systems. One example is the concept of critical flux which is applicable for microfiltration and ultrafiltration (Bacchin *et al.* 1995; Field *et al.* 1995; Howell 1995) but is not relevant for NF and RO systems (Vrouwenvelder *et al.* 2009a,b). In this contribution we focus on NF and RO-membrane fouling.

BIOFOULING STUDIES

Biofouling control is pursued by: (i) biological pre-treatment removing nutrients (e.g. sand filtration), (ii) applying biocide dosage or UV irradiation (metabolic inactivation of bacteria), (iii) membrane and spacer modification; and (iv) chemical cleaning. Development of membranes materials or coatings which prevent adhesion is one general advocated method to prevent biofouling without the need of applying toxic chemicals. In view of the wide range of possibilities of microorganisms to anchor themselves to surfaces, these approaches to avoid this adhesion do not lead to success. Protection against overgrowth by microorganisms in higher organisms (animals and plants), is mainly obtained by continuous renewal of epithelium cells or, e.g. applying strong wax layers or structures (e.g. lotus leaves). The first is difficult in non-living systems, while the latter is usually not a realistic option. Lotus leaves have a very strong hydrophobic nanostructure which makes them effectively dry and not in contact with the water, protecting them against microbial colonization. This characteristic is sometimes suggested for membrane modification (Marmur 2006), however the hydrophobic nature makes the membranes unsuitable for water permeation (except as a vapour). Surface modification by hydrophilic groups or polymer brushes is also suggested for minimization of microbial adhesion. The effects of these modifications are usually investigated in short term studies. These studies focus on initial adhesion, which is not representative for biofilm formation (Gjaltema *et al.* 1997). In general, adhesion is

influenced by physico-chemical interactions (for instance electrostatic forces, hydrophobicity, etc.) while biofilm formation is dependent on physical interactions (biofilm strength, shear forces, attachment surface roughness). A low adhesion surface might delay biofilm formation, but not prevent it. Moreover in practical systems a surface gets quickly covered by a conditioning layer from organic molecules in the treated water, masking the surface modification effect.

In order to study biofouling a 'scaled-down' monitor representative of actual membrane processes is needed in order to be able to do controlled experiments and analyse the system. In essence, for spiral wound NF and RO membrane systems this can be done by a system that represents the geometry of one feed channel with identical geometry and flow rates. The recently developed biofouling monitor (Vrouwenvelder *et al.* 2006) has such properties. This system makes the biofouling more experimentally accessible. Nevertheless in such experimental systems the complex interaction of processes in, for instance, a biofilm in a NF /RO module are still difficult to assess. It is difficult to change just one parameter at a time; an increased flow rate means more shear, more mass transfer and a different biofilm geometry, all affecting the impact of the biofilm on the filtration processes.

In-situ visualization methods such as nuclear magnetic resonance (NMR) imaging (Graf von der Schulenburg *et al.* 2008; Vrouwenvelder 2009c) or CT-scanning of full membrane modules allows us to assess how processes at the micrometre scale influence the macroscopic behaviour of the membrane processes. Mathematical simulation models can help to quantitatively assess the interaction of the many different processes (flow, diffusion, permeation, microbial growth, etc.), which individually are well known (Picioareanu *et al.* 2009; Radu *et al.* 2010). This combination of experimental and modelling approaches has been shown to be a fruitful approach leading to better understanding of biofouling, as discussed below.

The main items will be addressed in the following sections.

RECENT MICROBIOLOGICAL RESEARCH INTO CHARACTERIZING BIOFOULING

Using flow cells allows regular sampling and analysis of the microbial community that leads to biofouling. Bereschenko *et al.* (2010) indicated that in a fresh water RO module, first colonization likely occurs by sphingomonas-type bacteria, a bacterium hitherto not studied in this context. These

organisms are well adapted to oligotrophic environments and very rapidly form a gelatinous monolayer on a fresh membrane. These organisms have twitching and swarming motilities, which allow them to rapidly colonize a fresh membrane (Pang *et al.* 2005; Bereschenko *et al.* 2010). This thin layer does not directly influence the membrane performance in fresh water systems, but when further colonized by a thicker layer of other microorganisms, fouling occurs. Sphingomonads form in a developed fouling layer only a minor fraction of the microbial population and will not easily be recognized as important, but as they may form the connection between the membrane and the rest of the fouling layer they could form a good target for microbial control. In this respect it is also relevant to realize that these organisms generally make a different extracellular polymeric substances (EPS) than most other bacteria. Herzberg *et al.* (2010) concluded that the RO membrane selects for a specific microbial community. Indeed better characterization of the organisms involved might open the way to directed control of the specific bacteria and their EPS involved.

Chemical cleaning leads to a reduction of the pressure over the feed spacer in an RO module. However after chemical cleaning still a major fraction of the biofilm is present as dead cell material in a gel matrix (Figure 2). Likely pieces of biofilm subjected to large shear have been detached, resulting in less pressure drop, however the majority of the biofilm is still present (Bereschenko *et al.* 2011). Molecular genomic studies showed that after a chemical cleaning the sphingomonads (living in the deeper layer of the fouling layer and thereby more protected from cleaning) quickly regrow, (Figure 1) likely on the cell mass. Within a short time the fouling is established again.

FLOW CELL STUDIES

Biofilm morphology (and the related impact on pressure drop) is influenced not only by substrate load but also by linear velocity, substrate concentration and fluid shear on the biofilm (Vrouwenvelder *et al.* 2009a), showing the complexity in interpreting pressure drop measurements as signal for microbial growth in membrane modules.

In practice, applied cleanings usually inactivate but do not remove (most of) the accumulated biomass from the RO/NF membrane module, which is the reason why most cleanings are not effective for biofouling control. The effect of chemical cleaning on biofouled RO systems has been studied with the *in-situ* MRI technique (Creber *et al.*

2010). Chemical cleaning at an early stage of biofouling was more efficient in removing biomass than cleanings performed at a later stage. Preventive cleaning is probably preferred over curative cleaning.

The effect of substrate concentration, linear flow velocity, substrate load and flow direction on pressure drop development and biofilm accumulation was investigated in membrane fouling simulators (Vrouwenvelder *et al.* 2009a). The pressure drop increase was related to the amount of accumulated biomass and linear flow velocity. Biomass accumulation was related to the substrate load.

In RO and NF membrane elements, at linear flow velocities applied in practice, voluminous and filamentous biofilm structures were developed in the feed spacer channel, causing a significant increase in feed channel pressure drop. The amount of accumulated biomass was independent of the applied shear, but was depending on the substrate load. A high shear force resulted in a more compact and less filamentous biofilm structure compared with a low shear force, causing a lower feed channel pressure drop increase. This is in accordance with observation in other biofilm systems. A biofilm grown at low shear is however easier to remove by water flushing compared with a biofilm grown at high shear. The flow regime can be used to manipulate the biofilm morphology, enabling new approaches to control biofouling.

Operating membranes at low flow velocity reduces the effect of biomass accumulation on feed channel pressure drop/increase (Vrouwenvelder *et al.* 2009a). Membrane systems could be operated at an optimal feed channel pressure, tolerating biomass accumulation. In other words: a high biomass concentration causing a low feed channel pressure drop increase only is not a problem. Therefore, from a practical point of view the focus of research should be restricting the feed channel pressure drop and the feed channel pressure drop/increase rather than biomass accumulation. The Optiflux concept (Van der Meer 2003; Van Paassen *et al.* 2005) reduces the operational costs of membrane filtration by applying fewer membrane modules in a pressure vessel than commonly applied (i.e. three modules instead of up to eight). The operational cost reduction is based on lower energy consumption. In addition, a lower flow velocity will impact the effect of the biomass amount on the feed channel pressure drop, so the Optiflux concept may reduce the feed channel pressure drop increase caused by biofouling, reducing the membrane operational costs even further. Biomass accumulation is reduced at the location where impact on the feed channel pressure drop is highest and because of the lower flow, the impact of biomass on pressure drop increase is lower.

Reducing the impact of biofouling by reducing the linear flow velocity implies that the impact biofouling will be balanced with extra concentration polarization. Therefore, studies combining concentration polarization and biofouling are important. Digital Holographic Interferometry has proved to be a valid technique to visualize concentration polarization *in-situ* in a cross flow RO system. Experimental results showed that the hydrodynamics has great influence on the polarization layer (Fernández-Sempere *et al.* 2010).

From these flow cell studies it becomes clear that the interaction between hydraulics, load, substrate and membrane operational parameters is so complex that only detailed analysis can give sufficient insight into the mechanisms.

There is a substantial need for novel measurement techniques that enable non-invasive spatially resolved observation of biofouling in NF and RO membrane modules. Such measurements will enhance our understanding of the key design and operational parameters influencing biofilm fouling. In our research we have demonstrated the first application of NMR microscopy to a spiral wound RO membrane module. Despite the opaque nature of membrane design, NMR microscopy is shown to be able to provide a non-invasive quantitative measurement of RO membrane biofouling and its impact on hydrodynamics and mass transport. The developed NMR protocols allow the evolution of biofouling to be followed. Minimal biofilm growth is shown to have a substantial impact on: (i) flow field homogeneity, (ii) the spatially resolved velocity field, and (iii) displacement propagators, which are distributions of molecular displacement of a passive tracer (in our case, water) in the membrane. From these measurements, the effective membrane surface area (subjected to flow) is quantified. The observed channeling is in agreement with observation from practice, the above described monitor studies and the below mentioned numerical modelling studies.

Biofouling activity quantification in membrane filtration installations addressed assessment of oxygen consumption rates in membrane installations by measuring the oxygen concentration of the water before and after passing through the installation (feed and concentrate), but this measurement was not suitable to assess the biofouling activity during membrane operation (Kappelhof *et al.* 2003). Studies making a carbon or chemical oxygen demand (COD) balance of biological processes in membrane systems by analysing relevant parameters in feed water, concentrate and permeate have not resulted in a closed balance. Thus,

no suitable method has been found yet for non-destructive assessment of biological activity in membrane systems.

MATHEMATICAL MODELLING

Recently, biofilm models have been integrated with computational fluid dynamics (CFD) to investigate biofouling in RO and NF systems (Picioareanu *et al.* 2009; Pintelon *et al.* 2010; Radu *et al.* 2010). These mathematical models are useful tools to analyse complex phenomena occurring when a biofilm is formed in RO or NF feed channels. These models are based on basic physical, chemical and biological processes involved in biofouling. One of the challenges in modelling is to maintain a good compromise between the model complexity and insight gained.

Flow cell studies as well as modelling studies showed that fouling of the spacer very rapidly leads to flow channeling and large areas of the membrane being not accessible any more for fluid flow (Figures 3 and 4, Picioareanu *et al.* 2009; Vrouwenvelder *et al.* 2009d). These dead zones decrease the effective membrane area and influence solute mass transport. Biofouling is therefore indicated as a spacer problem. For salt water systems however concentration polarization is more important and a thin biofilm will already lead to an increased concentration polarization. Expectedly, the formation of preferential flow channels due to biofilm growth in brackish and seawater systems will lead to a severe degradation of plant performance.

Mathematical modelling allows us to assess the relative impact of different fouling mechanisms as a function of process conditions. Biofilm enhanced concentration



Figure 1 | Rapid recolonization of a dead 'cleaned' fouling layer by saphingomonads.

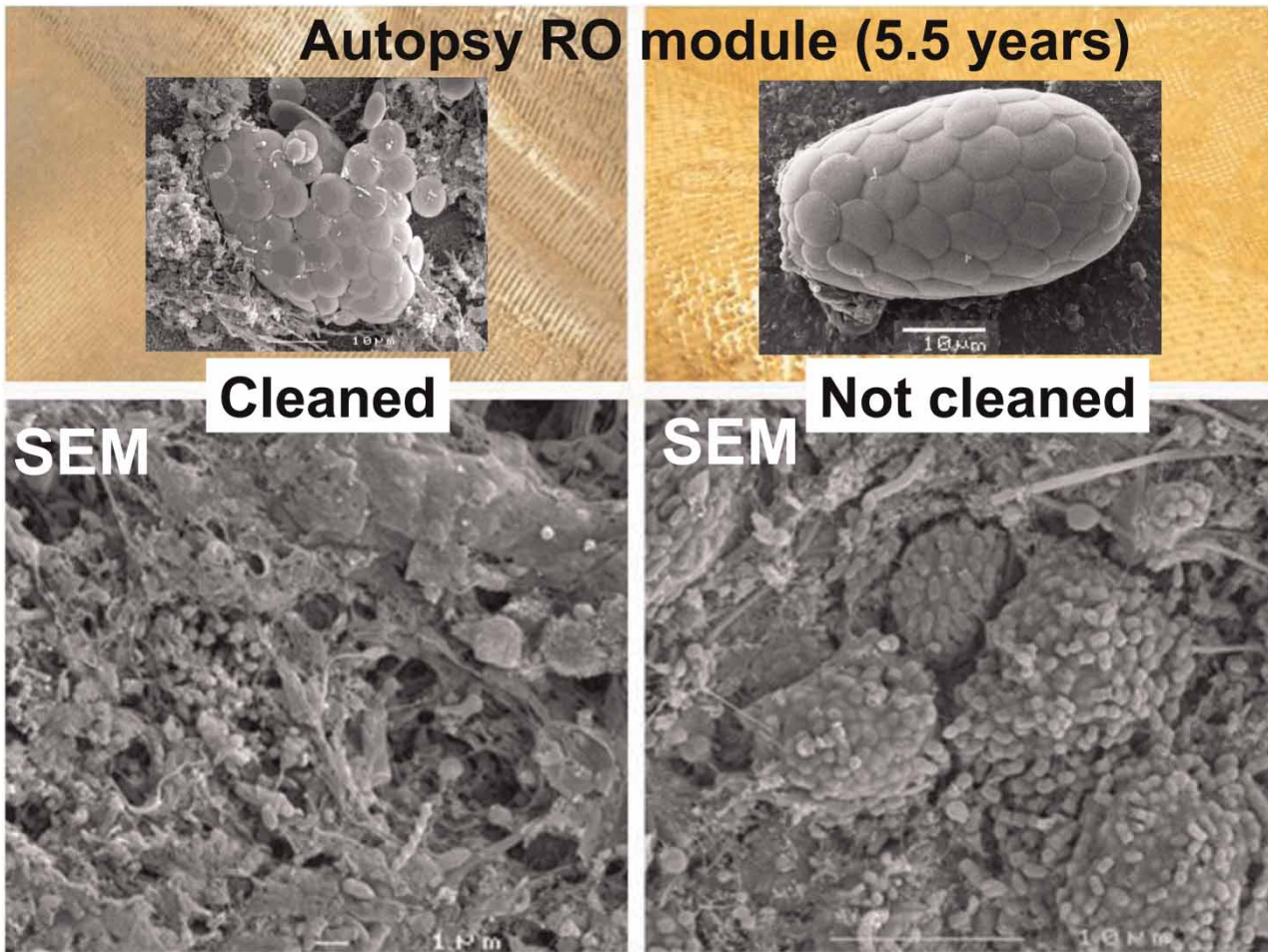


Figure 2 | EM pictures from full scale RO membranes after 5 years of operation. Left side was cleaned, right membrane module was not cleaned. Both show copious amounts of biomass, although the left side of the biomass shows a more deteriorated image.

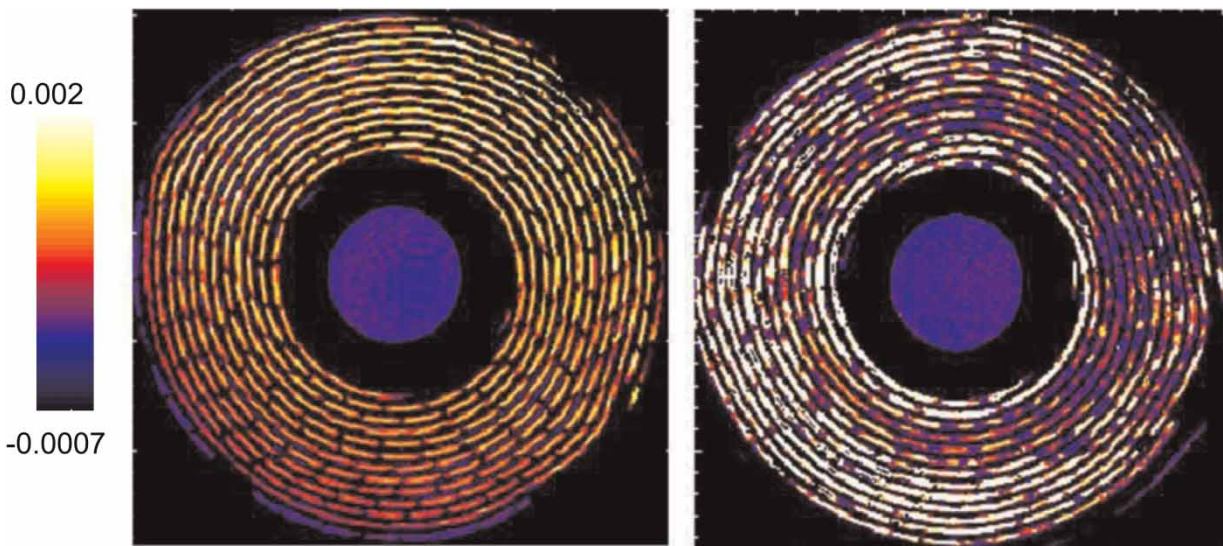


Figure 3 | NMR velocity imaging of a RO membrane module. Left: Clean module, flow evenly distributed, right: fouled membrane with strongly heterogenous flow. Colour scale: Liquid velocity in m/s. Adapted from Graf von der Schulenburg *et al.* (2008).

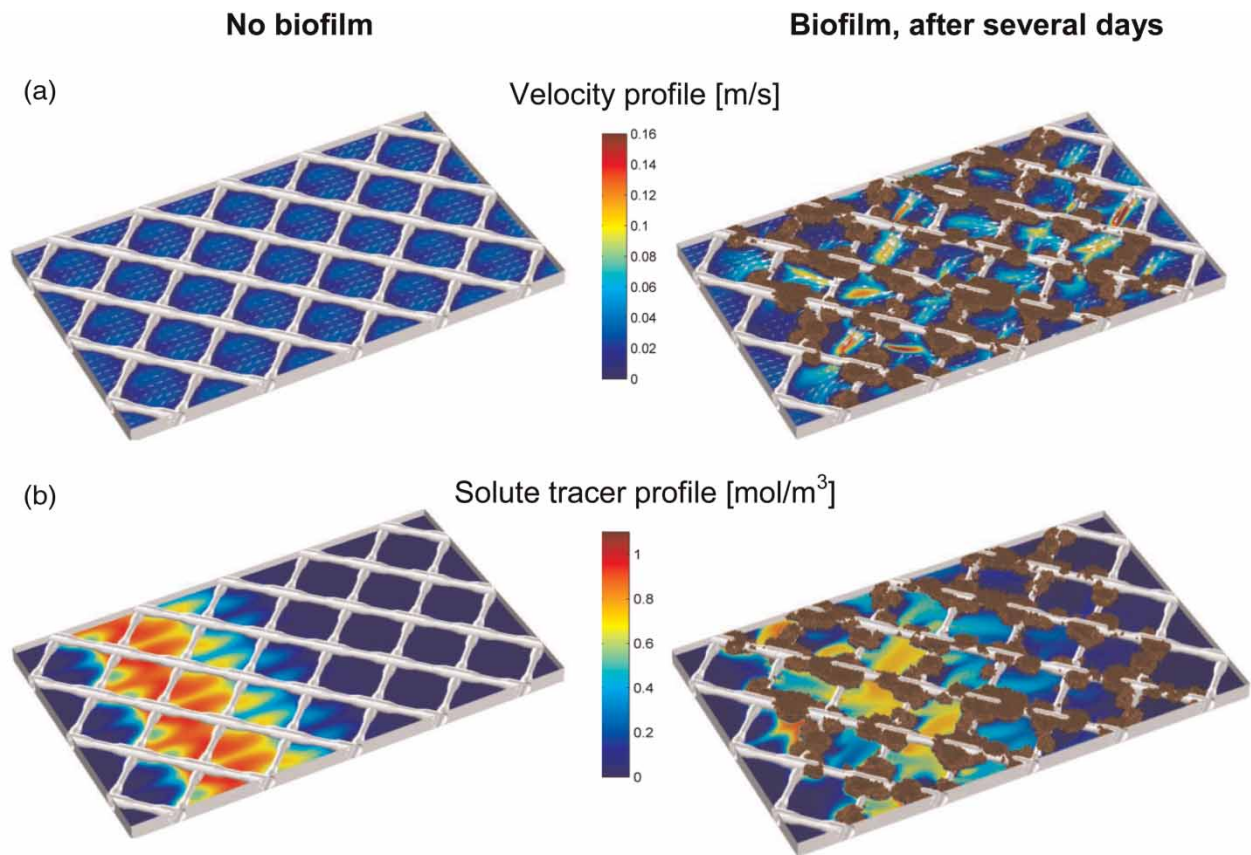


Figure 4 | Three dimensional simulation of flow and solute mass transport: (a) Simulated flow velocity in a two-dimensional section along the feed channel without biofilm (left) and with a developed biofilm (right). The initial regular flow pattern is changed into a preferential flow with narrow channels. (b) Two dimensional solute tracer distribution. Flow is from left to right.

polarization of nutrients is regularly mentioned to enable even faster biofilm growth. Mathematical modelling makes clear that only in very specific cases is this true, but that for normal operation such a mechanism is quantitatively marginal. Numerical tools have been developed to a stage where the interaction between biofilms and other potential foulants can be studied in a virtual RO system.

Given that micro-scale models provide a good understanding of local effects of biofilms (flow channelling, concentration polarization, salt passage, hydraulic resistance), and their relation to global performance parameters of RO systems (flux, pressure drop, salt passage), the modelling approach can be used to complement experimental studies.

CONCLUSION

In general, control of biofouling should be based on a good pre-treatment of the water before NF and/or RO to minimize

microbial growth. However, as inevitably some microbial growth will occur, systems should be designed in such a way that limited growth will not lead to biofouling and cleaning can be easily carried out (both by better spacer and module design). A good understanding for the design of such systems is only provided by a combination of an experimental scale-down approach and mathematical models supported by critical observation of full scale membrane processes.

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