



## Nano-coated surfaces to mitigate fouling in thermal water services

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### ABSTRACT

In thermal water services, the overwhelming proportion of losses by fouling is compensated by additional consumption of fossil energy carriers. This comes, not surprisingly, with an enormous energy price-tag as well as considerable green-house gas emissions, acidification of water resources and release of fouling chemical inhibitors. In the present paper, the performances of innovative nano-modified surfaces are discussed when subjected to CaSO<sub>4</sub> scale deposition during convective heat transfer. Two types of non-structured and structured nano-modified surfaces were experimentally examined. The experimental results demonstrated that such coatings would increase the induction time before fouling starts and also reduce the subsequent fouling rate, in comparison with untreated stainless steel surfaces. After laboratory runs, the coatings that performed best in laboratory were utilized in a desalination plant in a plate heat exchanger. The field findings resulted in applying thinner thickness of the attempted coatings with no sign of fouling. Considering these promising results, the paper continues to assess the requirements for better thermal and mechanical stabilities that such coatings would have to satisfy.

*Keywords:* Fouling; Cleaning; Desalination; Heat exchanger; Modified surfaces

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### 1. Introduction

Fouling/scaling of heat exchangers is a common industrial problem which may also occur where aqueous solutions are involved, e.g. using natural water for evaporative desalination or cooling purposes. It refers to the deposition of mineral salts from solution either by crystallization on the heat transfer surface or

in the bulk liquid phase. The result can be a tenacious “hard scale” which may be difficult to remove, or a soft deposit that is sometimes called “sludge”. This, in turn, can lead to (i) increased thermal resistance due to the lower thermal conductivity of the deposit layer which would range from 0.5 to 3.0 W/m K [1], (ii) increased pressure drop across the heat exchanger and (iii) substantial efficiency and production losses. Hanlon [2] reported that the potential scale

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precipitation in thermal desalination plants is rather considerable. For instance, for a 1 million gallon/d desalination plant under normal concentration conditions, a maximum of about 1,400 kg of  $\text{CaCO}_3$  or 80 kg Mg  $(\text{OH})_2$  could be precipitated each day. In terms of scale thickness this could correspond to a build-up of 0.1 mm/d on the total heat exchanger surface within a typical thermal desalination plant.

Although tangible mechanical and chemical cleaning/mitigation techniques have been developed to minimize the detrimental effect of deposit formation on heat transfer surfaces, nearly all industries still suffer from heat exchanger fouling. Instead of cleaning techniques it would be more desirable to produce heat transfer surfaces which inherently do not suffer from fouling. Modern mitigation techniques are based on approaches to increase the duration of the induction period, hence decreasing the adhesive strength of the fouling precursors [3]. The principle of such approach is the fact that the formation of deposit on a heat transfer surface always results from the initial interactive forces between the deposit and the surface. If the surface is modified such that these forces are minimized, then less scaling is expected. Several investigators demonstrated experimentally the significant effects of surface properties on fouling process. In most cases, but certainly not all, lower surface energy resulted in a reduced propensity of the surface to foul [4,5]. The applications included dairy, crystallization and bio-fouling. While there have been some successful attempts at fouling mitigation with modified surfaces, their utilization in industrial application is still problematic due to degradation of the modified surface performance as a result of relatively poor thermal, mechanical and physicochemical stabilities of coatings.

Within the framework of the MEDESOL project, this study was mainly focused on the development of innovative, environmentally-friendly anti-fouling heat transfer surfaces that could substantially (i) reduce the design and operation penalties and (ii) increase the membrane distillation (MD) efficiency. Accordingly, a comprehensive and rigorous set of laboratory fouling runs was performed with aqueous solutions as working fluid. The operating conditions followed closely to those expected in the MEDESOL MD desalination unit.

## 2. Short description of the MEDESOL project

The present study was part of a European project entitled “seawater desalination by innovative solar-powered membrane-distillation system (MEDESOL)”. The project aimed at developing an environmentally friendly and innovative concept of multi-stage MD in order to minimize specific thermal energy input and

membrane area required. The required heat had to be provided by solar thermal collectors. Another objective was to discern the technical feasibility of producing 0.5–50 m<sup>3</sup>/d potable water from seawater by integrating several MD modules. A conceptual schematic diagram of the MEDESOL is presented in Fig. 1. The prototype consists of three main components, i.e. solar collector (1), heat exchanger (2) and the membrane module (3).

MD is a desalination technique where saline water is heated to increase its vapour pressure, which in turn generates the difference between the partial pressure at both sides of the membrane. Hot water evaporates through non-wetted pores of hydrophobic membranes. The passing vapour is then condensed on a cooler surface to produce fresh water. The overall functionality of the process is illustrated in Fig. 1. The seawater (A) at low temperature of (e.g. 25°C) is heated with the hot water coming from the solar collector to provide warm seawater (B, e.g. 90°C) to be fed into the evaporator channel of the membrane. The partial pressure difference caused by the temperature difference across both sides of the membrane is the driving force for the steam (fresh water) passing through the membrane. The seawater is cooled through the evaporator channel arriving to (C, e.g. 45°C). The efficiency of the MD module increases as the inlet temperature of seawater goes up. Thus, the unit should permit seawater to heat up as close as possible to its saturation temperature without the risk of deposit formation. For the sake of compactness and maximum life expectancy, it was decided to use a plate and frame heat exchanger (PHE) which has the following advantages over conventional shell & tube heat exchangers [6]:

- (i) typically only one-fourth to one-tenth of the space of that of shell & tube heat exchangers is required [6];
- (ii) it can be easily disassembled, inspected, maintained, cleaned if necessary and reassembled again; and
- (iii) high rate of heat transfer is expected due to high turbulence and low thermal resistance of the plates.

These advantages have increased the market share of plate and frame heat exchangers. Nevertheless, the formation of deposit on exchanger plates would deteriorate the performance as typically illustrated in Fig. 2.

## 3. Fouling mitigation techniques

Numerous mechanical, chemical and physical approaches have been developed for mitigating or

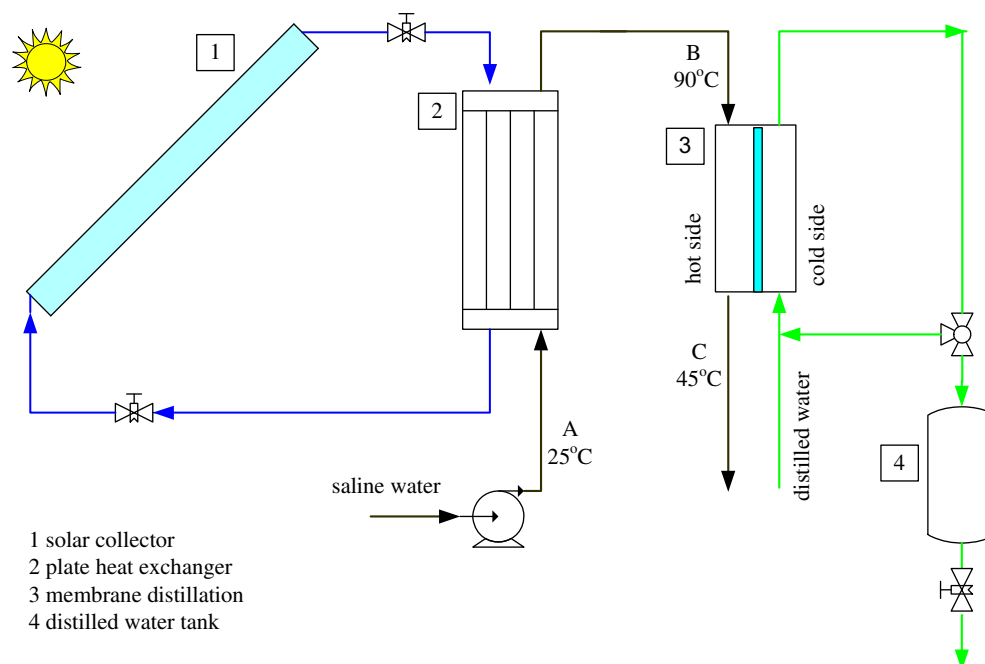


Fig. 1. Conceptual description of the MEDESOL prototype.

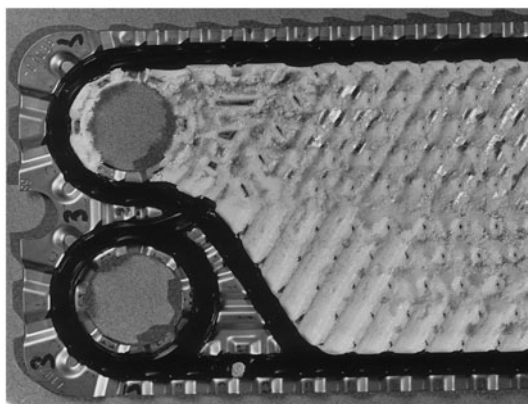


Fig. 2. Deposit build-up of  $\text{CaSO}_4$  in a plate heat exchanger [24].

controlling mineral scale formation including sponge balls, enhanced heat exchanger surfaces and chemical inhibitors such as dispersing and chelating agents. In addition, the mechanical mitigation techniques must be done carefully and cautiously to avoid damage of the heat transfer surfaces, thus increasing cost for labour and down-time. Problems associated with chemical cleaning of heat exchangers are due to the danger of handling (burn, toxic) as well as chemical attack on the heat exchanger material (overheating, corrosion) and finally disposal problems [3].

When a particle such as a crystal or bacterium reaches the surface, first it contacts the surface and if

the adhesion energies are strong enough then will stick to the surface. The interfacial properties, particularly surface roughness, topography, wettability and surface energy would play a decisive role in the adhesion process [7]. In recent years, technological advancements in surface treatment have given new impetus for developing innovative coating technologies and materials for a wide range of fouling problems under various modes of heat transfer such as convection and boiling [8,9]. The present study highlights the impact of surface treatment technology as an environmentally friendly technique on fouling mitigation that would occur in thermal desalination units.

#### 4. Surface treatment

The physical concept for the surface treatment is to produce a surface with lower surface energy which shows weak deposit adhesion or even non-sticking properties. The adhesive strength is reduced between deposits and the surface by weakening the interactive forces [10]. Two main coating procedures are currently available: (i) outer surface coatings and (ii) embedded coatings. Outer surface coatings generally preferable over the embedded ones for reasons given below:

- coating reaches normally inaccessible areas;
- they are less labour intensive;
- coating can be performed *in situ*; and
- suitable for large size of heat transfer surfaces.

Among outer surface coating techniques, this study endeavoured to investigate the influence of the nano-coatings on the fouling mitigation. Nano-coating refers to the act of covering a material with a layer thickness of nano scale and/or using nano-produced materials to coat the target surface. Generally speaking, for enhanced heat transfer characteristics and fouling mitigation, the aim is to have nano-coatings with high degree of hydrophobicity or lower surface energy. In most cases, but not certainly all, lower surface energy materials reduce fouling adhesion compared with untreated surfaces [10]. The structure nano-modified surfaces are as follows:

- (1) Unstructured coatings which involve a mixture of various materials with high repellence tendency such as wax or polymeric layers which can be fortified with metal atoms to enhance stickiness between substrate and coating.
- (2) Nano-structured surfaces which include surfaces having high degree of repellency. They could be obtained by controlling the topography of hydrophobic surface. The coating, also known as *lotus-like surfaces* or *self-cleaning surfaces*, are characterized by combination of low surface energy materials and a specific surface topography. Micro and nano-scale rippled structures is a vital way in generating the self-cleaning super-hydrophobic property of a surface.

#### 4.1. Application of nano-modified surfaces in various deposition processes

##### 4.1.1. Low temperature applications

Biofouling defines as undesirable growth of living organisms on a surface. It is a major fouling occurrence on surfaces having temperatures typically less than 60°C. Small changes in temperature are likely to make a significant variation of biofilm growth as microbial activity is very sensitive to temperature. For instance, it was shown that biofilm thickness increased by 80% by raising the temperature from 30 to 35°C [11]. It has been found that the incorporation of silicon into paints has helped to mitigate biofouling on ship hulls. Such paints can replace less environmentally compatible paints with biocides (Ambio Project). The Ni-P coating causes a significant reduction in bacterial adhesion. It reduced *E. coli* attachment by 66% compared with untreated stainless steel surface [12]. Therasa et al. [13] also found that forming copper nano-films on the titanium substrate by using

electroless plating technique provides an excellent condenser material to resist biofouling and corrosion. They also showed that the biofilm formation has found to be increased with decrease in copper nano-particle size.

Another biofouling type is the proteinaceous food soil (baked tomato paste). Modification of stainless steel 316L by the nano-coating (sol-gel) reduced the total surface energy significantly (15.5 mN/m) in comparison to that of the stainless steel (54.1 mN/m). Reduced surface energy of such magnitude causes a lower adhesion force between the deposit (food soil) and the surface. The deposits were then readily removed by the lowest practicable value of fluid dynamic gauge pressure [14].

##### 4.1.2. High temperature applications

The majority of heat transfer surfaces like those in thermal desalination plants are utilized at the elevated surface temperatures ( $T_s > 100^\circ\text{C}$ ) as for subcooled flow boiling, pool boiling and forced convective heat transfer. In the course of the past decade, more efforts have been made to utilize the non-sticking property of the nano-coatings for fouling mitigation. For instance, the electroless Ni-P coatings can significantly reduce the deposition of the calcium carbonate ( $\text{CaCO}_3$ ) during the boiling process of the tap water [15]. However, the corrosion rate of the Ni-P coating increases with the salt concentration [12]. Georgieva and Armyanov [16] also reported that the addition of Cu and Sn co-deposition in electroless Ni-P plating causes a positive suppression of the corrosion potential of Ni-Cu-P and Ni-Sn-P in the presence of NaCl and  $\text{H}_2\text{SO}_4$ .

Titanium dioxide thin film ( $\text{TiO}_2$ ) is of industrial interest because of their outstanding chemical resistance. The film thickness of  $\text{TiO}_2$  coating layers in nanometre scale was used to mitigate fouling in pool boiling of the  $\text{CaCO}_3$  solutions. The results indicate that (i) the  $\text{TiO}_2$  coating surfaces can inhibit  $\text{CaCO}_3$  deposition, (ii) the fouling induction time on  $\text{TiO}_2$  coating surface in pool boiling with film thickness of  $8 \times 10^{-8}$  m is about 50 times longer than that of untreated or polished surfaces and (iii) the  $\text{CaCO}_3$  crystal fouling has a weak adhesion which can easily be removed from the surface [17].

The utilization of nano-coatings under forced convective heat transfer shows promising results as in the case of using the *Fluorinated Ethylene Propylene* and *Perfluoroalkoxy* coatings. Both of them cause more than 40% reduction in fouling resistance of the  $\text{CaSO}_4$  solution in comparison to that of the uncoated stainless steel surface (AISI 304 BA) [18]. Such reduction in

fouling rate was not occurred by using the sol-gel coating with high degree of hydrophobicity. Instead the induction period prolongs 4 times than that of the stainless steel surface [4]. Nano-coatings have also been used to decrease fouling inside plate heat exchanger for milk production [19]. They found that certain polyurethane-coated plates formed thinner deposit layers compared to untreated surfaces.

## 5. Nano-modified surfaces for MEDESOL project

The stainless steel substrates (AISI 304 BA) were modified by the following coating materials for the laboratory fouling runs:

- **Unstructured solvent based coatings (SB1–SB4):** which consist of hexagonal boron nitride (BN), aluminium silicate as filler and fluorinated polyethylene wax.
- **Unstructured water based coating (WB):** It is produced by directly dispersing the components (hexagonal boron nitride (BN), titanium dioxide (TiO<sub>2</sub>) as filler, fluorinated polyethylene wax) in water.
- **Unstructured electroless Ni–P–BN coatings:** consists of two layers. The first layer consists of 40 μm of Ni–P with 7–9% P and the second layer on top is made from 10 μm of BN (Ni–P with 12–15% boron nitride).
- **Nano-structured coatings (KS1 and KS2):** The coatings include silicon nano-particles that are spread on the substrate to increase hydrophobicity: The thicknesses of the coatings are 1.5 and 2.5 μm, respectively.

### 5.1. Influence of nano-coating on the performance of the MEDSOL heat exchanger

Before any attempt to coat the plates of the PHE, firstly the coatings were comprehensively examined in laboratory; refer to Al-Janabi [10] for more details

about the experimental set-up and procedure. The accumulation of CaSO<sub>4</sub> deposits on modified surfaces presented in the preceding section has continuously been monitored by measuring the variation in surface temperature with time. Fouling rate and induction time were used to evaluate the impact of various modified surfaces on mitigating deposit formation.

*Induction period* is defined as the elapsed time before a substantial increase in fouling resistance takes place and the *fouling rate* which is the slope of the curve where the fouling resistance increases continuously during the fouling period. The laboratory results showed that the coatings under conditions similar to those in MEDESOL desalination unit causes a significant reduction in fouling rate and increased induction time in comparison to that of the stainless steel surfaces as shown in Table 1.

For nano-structured modified surfaces (KS1 and KS2) as time went on the fouling resistance increased followed by an almost drastic drop implying that the deposit layer has been spalled off the surface. This implies the low stickability of the deposit layer to the surface in tandem with exerted shear forces of fluid that can help deposit removal as shown in Fig. 3(a). The same behaviour has been observed for the Ni–P–BN coated surface as shown in Fig. 3(b). However, some spots of oxidization were observed for the Ni–P–BN and the KS2 coated surfaces. The spots were in red colour which is the hallmark of corrosion and occurred only at the interface between the damaged part of the coating and the stainless steel substrate [8].

Fig. 4 summarizes the percentage reduction of fouling resistance and heat transfer performance ( $U_t/U_o$ ) for all coated surfaces after the first 5 h of the experiment. The comparison provides a tool to assess the impact of the coated surfaces on the fouling propensity. Obviously using the nano-coatings for altering the surface properties of the stainless steel surfaces causes in most cases a significant decrease in fouling resistance and, as a consequence, better heat transfer performance.

Table 1

Fouling rate and induction time for the attempted untreated and modified surfaces for the laboratory fouling runs

Type of surface		Fouling rate ( $10^{-6} \times \text{m}^2 \text{K/W min}$ )	Induction time (min)
Untreated stainless steel surface	AISI 304 BA	4.3	43
Unstructured solvent based coated surfaces	SB1	4.1	89
	SB2	1.171	181
	SB3	3.9	58
	SB4	3.93	72
Unstructured electroless Ni–P coated surfaces	Ni–P–BN	0.961	580
Nano-structured coated surfaces	KS1	2.96	130

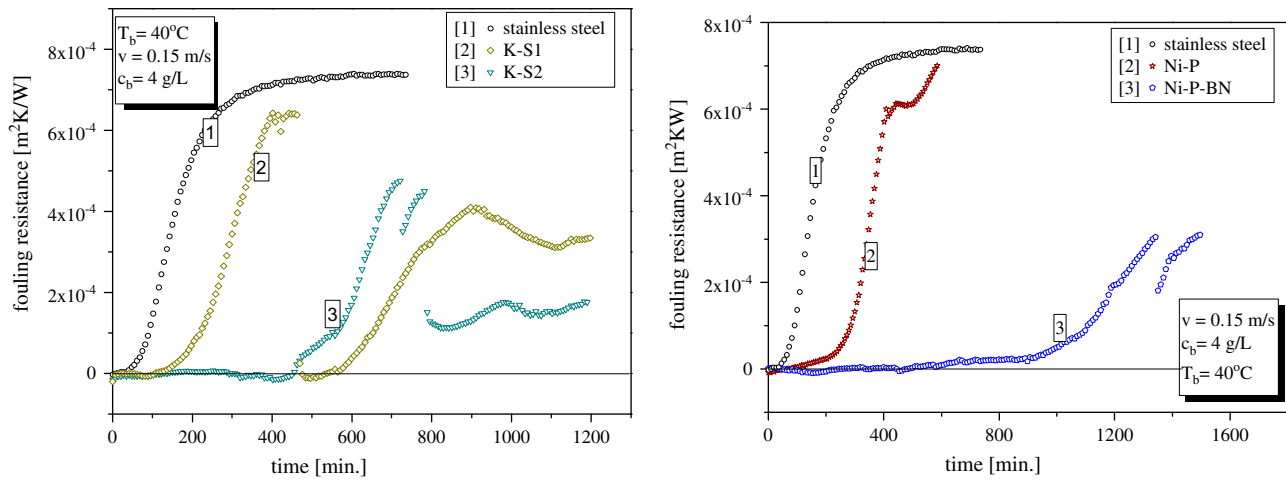


Fig. 3. Fouling resistance as a function of time for: (a) the nano-structured and (b) the electroless Ni-P surfaces.

The coated surfaces with promising laboratory results were also investigated in the desalination plant of the MEDESOL project, described before. The plates were coated by the SB materials and subsequently examined under harsh fouling and thermal conditions such that the surface temperature varied from 40 to 90°C in daily basis. The field results showed no sign of fouling as shown in Fig. 5 which validated the suitability of the selected coatings for this application.

## 6. Challenges in the utilization of nano-technology for fouling mitigation

Despite the substantial mitigation in fouling process in terms of longer induction time, lower fouling rate and weaker stickiness of the fouling layer to the nano-modified surfaces, many formidable challenges still lay ahead before (i) such technologies can widely be commercialized and (ii) achieve their full potential in mitigating fouling.

A commercialized application of the nano-coatings must sustain two distinctive but interrelated conditions of fouling mitigation as well as mechanical and chemical stabilities of coatings as shown in Fig. 6. These conditions must be addressed in tandem for a successful application of nano-coatings. The desired nano-coatings should be:

- anti-corrosive;
- anti-abrasive;
- economically viable;
- environmentally acceptable; and
- long time operation.

One of the main challenges for the nano-coating technology is the poor adhesion of the coating to the

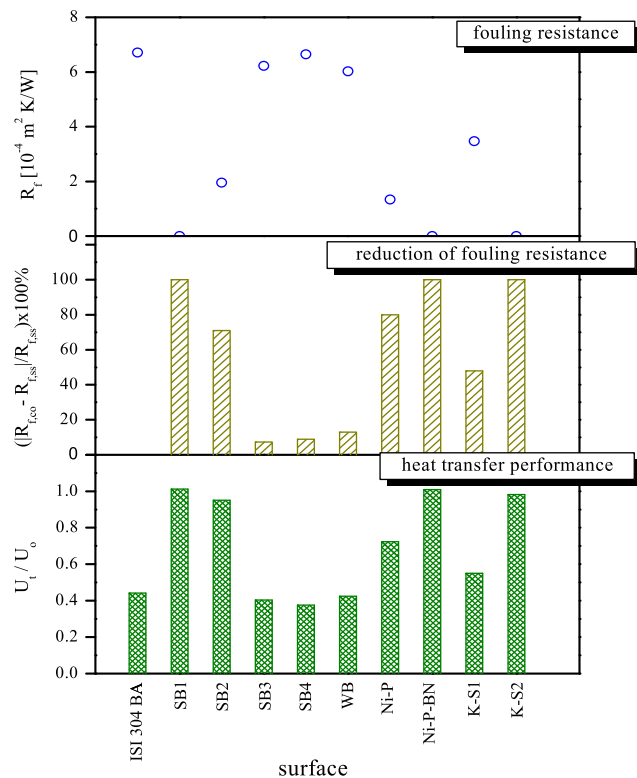


Fig. 4. Extent of fouling resistance and heat transfer performance for untreated and coated surfaces at  $T_b = 40^\circ\text{C}$ ,  $v = 0.15\text{ m/s}$  and  $c_b = 4\text{ g/L}$  at  $t = 5\text{ h}$ .

substrate [20]. Weak adhesion indicates that they cannot be easily scaled-up to a practical system. However, increasing the roughness profile of the substrate gives rise to increase the stickability of the coating layer to the substrate [10] as shown in Fig. 7. This is primarily

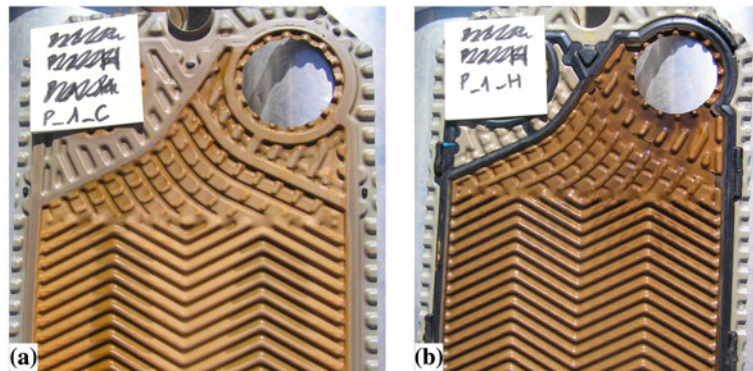


Fig. 5. MEDESOL heat transfer surfaces under severe fouling conditions after 45 d for both: (a) cold fluid side and (b) hot fluid side.

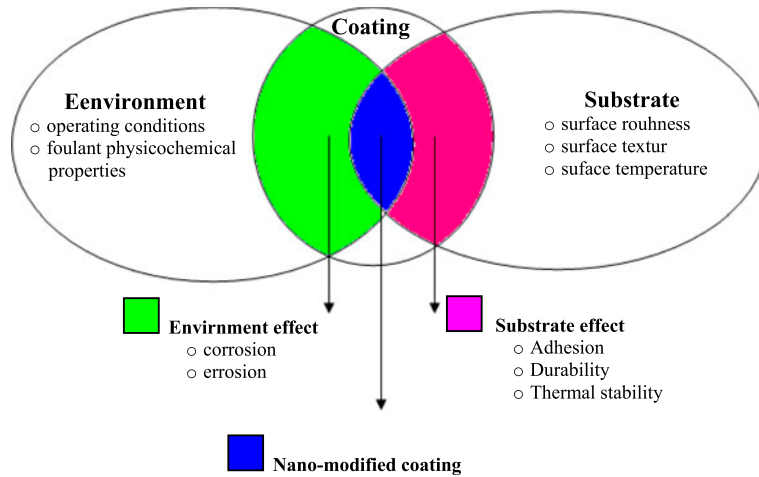


Fig. 6. Criteria for the selection of an optimum nano-coating.

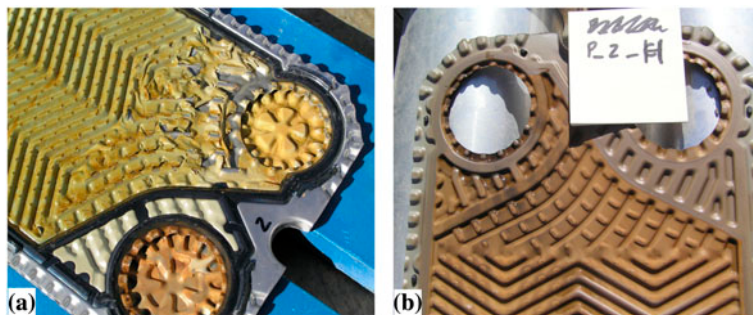


Fig. 7. SB coating layer of: (a) a smooth substrate and (b) a rough substrate.

due to the stronger adhesive joints between the substrate and coating [21,22]. The change in increasing the surface roughness should carefully be done that it does not affect the performance of the coating under fouling conditions.

Weak abrasion resistance of the coating layer can lead to coating flake off once the foulant started to deposit. As a consequence, the damaged surface provides additional nucleation sites which result in increased fouling rates [10]. Fig. 8 presents a picture

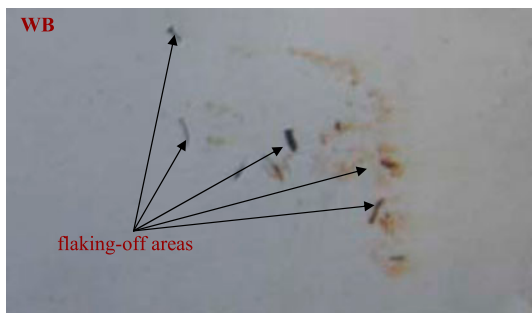


Fig. 8. Unstructured WB coated surface after fouling test.

of the water based (WB) coated surface which was taken after terminating the experiment.

In most cases, the thermal conductivity of the coating is lower than that of the metal substrate. It represents a major obstacle in the deployment of nano-coatings for modifying the heat transfer surfaces. To overcome this issue, a minimal thickness of the coating is required. However, the requirement of a minimal layer thickness results in, in most cases, an inhomogeneous texture of the surface coating [18].

The impact of foulant on the coating can be represented by, for example, corrosion and erosion. Anti-corrosion is a must property for an anti-fouling coating. Shen et al. [23] investigated the corrosion resistance characteristics of  $\text{TiO}_2$  nanoparticle coating with film thickness of from  $1.96 \times 10^{-7}$  m to  $6.40 \times 10^{-7}$  m on 316 L stainless steel substrate. They found that the sample with thickness of  $4.64 \times 10^{-7}$  m has an excellent anticorrosion behaviour in the NaCl solution with concentration of less than  $1,000 \text{ M m}^{-3}$  and pH higher than 2. With high concentration or acidic solution environment, the  $\text{TiO}_2$  films will dissolve gradually and the anti-corrosive ability profoundly decreases. Table 2 shows the ability of nano-coating material to corrosion.

As it can be seen, most of the base materials for nano-coatings, e.g. Ni-P, Ni-P-BN have a poor corro-

sion resistance compared with silicon or polymeric layers fortified with metal atoms. This can be related to electron charge difference that may exist between the metal substrate and the coating which contains metal atoms such as nickel, boron-nitride and phosphor.

## 7. Concluding remarks

Unstructured and nano-structured coatings were used as an approach to minimize deposit adhesion energies and to provide environmentally-friendly modified surfaces for utilization in the plate heat exchanger of the thermal desalination unit of the MEDESOL project. Both the laboratory and field findings showed a significant reduction in fouling rate and increased induction period compared to that of untreated stainless steel surfaces. In addition, the drastic drop of the fouling layer refers to the low stickability of the deposit layer to the coated surfaces.

The mechanical stability in terms of weak abrasion resistance of the coating layer has been treated by increasing the surface roughness of the base material before applying the coating for the field trials. However, some spots of oxidization were observed on the unstructured electroless Ni-P-BN and nano-structured K-S2 coated surfaces. These spots may be due to an electron charge difference that may exist between the metal substrate and the coating.

## Nomenclature

$R_f$	—	fouling resistance ( $\text{m}^2 \text{K/W}$ )
$t$	—	time (min)
$T$	—	temperature (K)
$U$	—	overall heat transfer coefficient ( $\text{W/m}^2 \text{K}$ )
$v$	—	velocity (m/s)

## Subscripts

$b$	—	bulk
$f$	—	foulant
$s$	—	surface

## Abbreviations

AISI	—	American Iron and Steel Institute
BN	—	boron nitride
MD	—	membrane distillation
Ni-P	—	nickel-phosphor
PFA	—	Perfluoroalkoxy
SB	—	solvent based coated surface
SS	—	stainless steel
WB	—	water based coated surface

Table 2  
Corrosion resistance of nano-coated surfaces

Coating	Corrosive	Anti-corrosive
SB1–SB4		+
WB		+
Sol-gel		+
$\text{TiO}_2$		+
Polyurethane		+
Ni-P	+	
Ni-P-BN	+	
KS1	–	
KS2	+	



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