Royal Belgian Institute of Marine Engineers

Hydrogel-Based Third Generation Fouling Release Coatings

Peter THORLAKSEN[1], Diego M. YEBRA[1], Pere CATALÀ[2]

Summary

The so-called fouling release coatings typically rely on a dual non-toxic mode of action: "non-stick" properties, traditionally achieved through surface smoothness and a low surface tension (hydrophobicity), and a "fouling release" behaviour related to a low modulus of elasticity (Young's modulus). While the "fouling release" properties associated with the polydimethylsiloxane matrix have remained basically unchanged over the past years, a significant amount of research has been devoted to enhancing the "non-stick" properties. The invention behind the 3rd generation fouling release coatings is a unique blend of silicone polymers that maintains a more hydrophilic surface, clearly delaying diatomic slime settlement and, hence, subsequent fouling. This translates into a lower hull skin friction over longer periods of time which contributes to significantly lower fuel consumption, and, directly related, lower emissions of green house gases, combustion particles (PM) and other harmful gases such as SOx and NOx.

1 Fouling Control Denmark, Group R&D, Hempel A/S, Lundtoftegårdsvej 91, DK-2800
2 Fouling Control Spain, Group R&D, Pinturas Hempel S.A.U, Carretera de Sentmenat 108, E-08213 Polinya, Barcelona, Spain

INTRODUCTION

Marine biological fouling, usually termed marine biofouling, can be defined as the undesirable accumulation of microorganisms, plants and animals on artificial surfaces immersed in sea water (Yebra et al., 2004; Jones, 2009). For the specific case of ocean-going vessels, the recognized effects of biofouling are higher frictional resistance with subsequent speed reduction or higher fuel consumption (see Table 1), loss of maneuverability and higher frequency of dry-dockings. As a direct consequence of increased fuel consumption, fouling can be associated to significantly increased emissions of harmful gases to the atmosphere, such as CO2. NOx and SOx (Table 2).

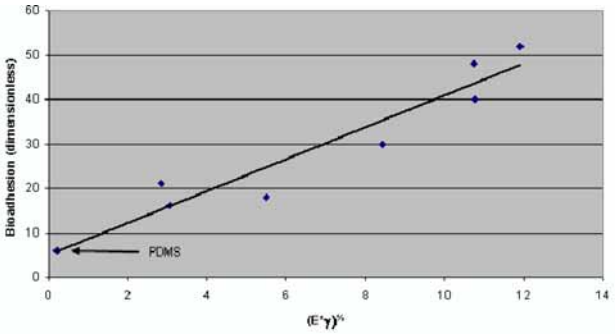

Table 1. Reported effect of different hull conditions on the total shaft power (ST) for the case of U.S. Navy Oliver Hazard Perry class frigate. Increased fouling severity involves higher frictional resistance and, consequently, higher powering demands to keep a speed of 15 knots (Schultz, 2007 and Munk et al., 2009).

	Relative adhesion	γ_c	E-modulus	(E * γc)½
PHFP	21	16,2	0,5	2,85
PTFE	16	18,6	0,5	3,05
PDMS	6	23	0,002	0,22
PVF	18	25	1,2	5,48
PE	30	33,7	2,1	8,41
PS	40	40	2,9	10,77
PMM	48	41,2	2,8	10,74
Nylon66	52	45,9	3,1	11,93

Table 2. Review of MARPOL Annex VI, International Maritime Organisation (IMO) , Sub-committee on Bulk Liquids and Gases (BLG), 2007. CO2 emissions from shipping constitute about 3.5% of the total CO2 emissions.

Additionally, marine biofouling has been demonstrated to be one of the main vectors for the introduction of invasive species into sensitive ecosystems (Piola et al., 2009), as already acknowledged by the International Maritime Organization (IMO; see e.g. BLG 13/9).

Back in 1970, Hempel filed one the first patents protecting the use of low surface energy polysiloxane surfaces as an alternative to biocidal paints (Krøyer, 1970). However, further development was drastically hindered by the overwhelming success of a quite different technology, stemming from a patent filed by Milne and Hails in 1971. The so-called selfpolishing copolymer (SPC) antifouling paints allowed a constant emission of a highly efficient, broad-spectrum toxin, tributyltin (TBT) offering ship operators cost-efficient 60+ months foul-free hulls and demonstrated reductions in fuel consumption (Yebra et al., 2004).

However, the IMO process to ban the underwater use of TBT-based paint products, finalized in September 2008, caused a major change in the antifouling paint industry. The definitive step of this process took place in 2003, when the major paint manufacturers decided to replace the tin-based assortment by a new generation of TBT-free products. Such environmentally friendlier products, used by the majority of the world-wide fleet, are still based upon mixtures of cuprous oxide (Cu2O) and organic co-biocides embedded into a controlled-release matrix.

These compounds are progressively being subject to close scrutiny by legislators (see e.g. Environmental Protection Agency in USA and the Biocidal Product Directive in the EU: Pereira and Ankjærgård, 2009) which may result in new restrictions and/or prohibitions. Over the last years, biocide-free technologies based on "non-stick" and "fouling release" approaches have recovered focus and attention from paint makers and customers. This paper presents the latest commercial development in this field.

LOW SURFACE ENERGY, NON-STICK, FOULING RELEASE TECHNOLOGY

Non-stick Fouling Release coatings are based on a technology which initially prevents the adhesion of fouling organisms by providing a low-friction, ultra-smooth surface on which organisms have great difficulties in adhering. Most, if not all, sessile species rely on attaching mechanism involving some kind of glue, with the adhesion strength highly dependent on the ability of this glue to spread over the surface and bind physicochemically to it. For a given amount of glue (e.g. the case of Ulva spores), the higher the surface tension of the substrate the better the glue can wet

the surface and the higher numbering of "anchoring points" translating into better adhesion of the organism (Callow et al., 2005). Some organisms, such as the blue mussel (Callow et al., 2005), seem to compensate the latter by secreting larger amounts of glue when in contact to low surface energy surfaces.

The first trials for non-stick surfaces were based on polytetraflourethylene (PTFE; e.g. TEFLON®) which is known for a very low surface tension (16 - 18 mN/m; Figure 1). However a stronger bio adhesion was seen on PTFE surfaces. One explanation is that the micro porosity of the PTFE surface allows the organisms a mechanical anchoring (Brady and Singer, 2000).

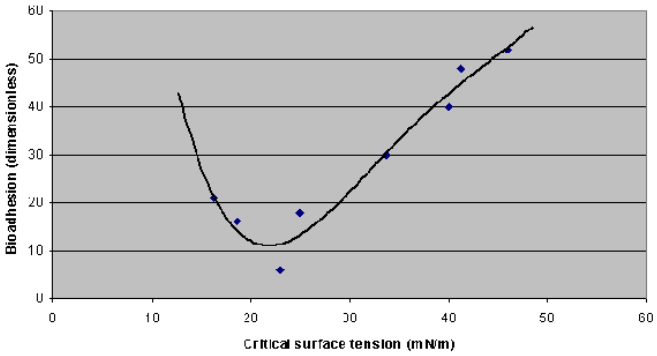

Figure 1.- Empirical correlation between relative bio adhesion and critical surface tension - the Baier curve. Lowest adhesion strength is found at 23-24 mJ/m2, and not at the lowest surface tension

Polydimethylsiloxane binders usually have a surface tension of 23-25 mN/m combined with a great smoothness which leads to the minimum bio adhesion. There is, however, one more parameter which comes from fracture mechanic studies (Townsin and Anderson, 2009). Data from practical measurements, where a disc is being glued onto a substrate, shows that the adhesion, P, is proportional with the square root of the product of the Elasticity modulus, E, and the critical surface tension, γ_c of the polymer (the glue) and inversely proportional with the dry film thickness of the polymer (glue) film, t, see equation 1 (Brady and Singer, 2000).

$$P = K \cdot \left(\frac{E \cdot \gamma_c}{t} \right)^{1/2} \qquad (1)$$

Compared to Figure 1, Figure 2 shows the relative bio adhesion as a function of $(E \cdot \gamma_c)^{1/2}$ and shows that the elasticity modulus is equally important as the surface tension when it comes to fouling release performance (see data in Table 3). Summarizing, silicone elastomers combine all known factors associated to high resistance to surface colonization, namely surface energy or surface tension, smoothness and elasticity.

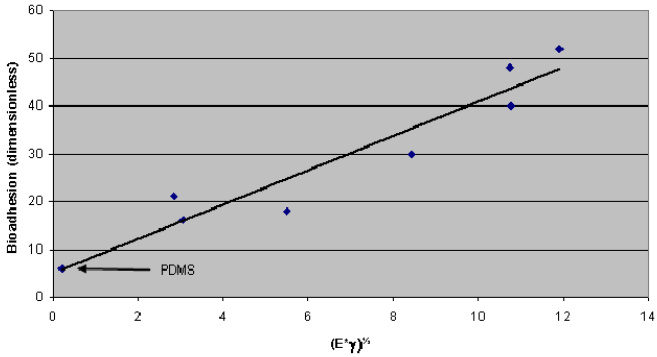

Figure 2. From above data it is clear that PDMS binders shows good fouling release properties. Reason being the low elasticity modulus of this polymer, combined with the relative low surface tension.

	Relative adhesion	γ_c	E-modulus	$(E * \gamma c)^{1/2}$
PHFP	21	16,2	0,5	2,85
PTFE	16	18,6	0,5	3,05
PDMS	6	23	0,002	0,22
PVF	18	25	1,2	5,48
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PS	40	40	2,9	10,77
PMM	48	41,2	2,8	10,74
Nylon66	52	45,9	3,1	11,93

Table 3. Shows the data used in Figures 1 and 2. Polydimethylsiloxane (PDMS) has an Elasticity modulus much lower than i.e. PTFE.

TRADITIONAL FOULING RELEASE COATINGS

The first commercial breakthrough of silicone coatings into the market took place thanks to "pure silicone" formulations (Anderson et al., 2003 and Townsin and Anderson, 2009). The PDMS matrix was reinforced through the addition of low molecular weight silicone polymers ('oils') to enhance foul-release properties of PDMS polymers. This is thought to be mainly due to the surface tension and hydrophobicity changes deriving from the self-stratifying of these oils to the surface due to immiscibility with the cured PDMS. These coatings provided self cleanability when sailing at a certain speed at a certain activity, typically above 15kn during minimum 75% of the time.

Traditional fouling release coatings showed very good initial performance against shell fouling and macroalgae, while slime fouling could be rapidly observed after a few months sailing (Townsin and Anderson, 2009). Multiple studies demonstrate that the ideal substrate for one fouling type may not be the same for another (Finlay et al. 2002). As an example, the green algae Ulva's zoospore was shown to have a higher affinity to hydrophobic substrates, but with very weak adhesion to such, while diatoms showed strong adhesion to hydrophobic surfaces. The same can actually happen between different species of the same family. Clare and Aldred (2009) hypothesize that the barnacle Balanus Improvisus could prefer more hydrophilic surfaces, just opposite as its relative the Balanus Amphitrite.

As mentioned before, it was soon realized that slime did manage to stick tenaciously to the early hydrophobic PDMS-based fouling release coatings, "masking" the non-stick surface therefore facilitating subsequent colonization by superior organisms such as macro algae and shell fouling. These may or may not be released upon sailing depending on mainly 1) vessel speed, 2) length of idle periods, 3) sailing time at high speed, 4) adhesion strength and 5) foul release capabilities of the coating. The end result of the above is that the now broadly

accepted fuel saving properties of fouling release coatings (e.g. Schultz, 2004) start to decrease, eventually to a lower level than top-quality self-polishing copolymer paints, largely fouling free during their service life.

It seems clear that the latest developments in the field of fouling release are related to the optimization of these non stick properties through the addition of polymeric additives. This is demonstrated, for example, in patent WO02074870 dealing with the development of PDMS matrixes modified with low molecular weight fluorinated polymers and which has led to one of the most recent commercial launches.

HYDROGEL SILICONE

Hydrogels consist of a network of polymer chains that are water-insoluble but highlyabsorbent so that they can contain over 99% water. Hydrogels also possess a degree of

flexibility very similar to natural tissue, due to their significant water content. The use of hydrogels is common in medical applications (Henriques et al. 2005) due to their well-known capability to minimize protein and bacterial adhesion. Nevertheless, they are still typically combined with some sort of antibiotic or biocidal agent for this purpose (Wirtanen et al. 1998, Ahearn et al. 2000). Several studies point at the potential use of hydrogels for fouling control purposes. As an example, Rasmussen and Østgaard (2001) tested various gels against marine bacterial adhesion finding best results with a modified polyvinyl alcohol gel. Similarly, Ekblad et al. (2008) report about the testing of a protein-resistant poly(ethylene glycol) (PEG) based hydrogel coating for antifouling applications. Despite the promising results, one could argue that even if a practical coating based on this approach was developed, there could be doubts about the long term stability, mechanical properties and performance of the hydrogel. Gateholm's et al. (1995) approach was slightly different, since their idea was based on immobilizing viable marine bacteria secreting compounds with antifouling properties.

His et al. (1999), chose to load the hydrogel with benzalkonium chloride. Potentially, enzymes could be also immobilized in a similar manner way (Olsen et al., 2009) even though how to get the hydrogel from the can to the hull keeping activity might prove cumbersome. Hempel A/S has taken the traditional Fouling Release coatings one step further by making use of the hydrogel technology. Recent findings have shown that a PDMS coating with a hydrogel introduced at the water interface of the coating provides an improved resistance towards algae and slime fouling, while the intrinsic properties of pure PDMS, such as release mechanisms and drag reduction, remain. Due to a certain, controlled immiscibility, the hydrogel polymers have a tendency to slowly phase-separate to the PDMS-water interface. Once in contact with water they become hydrated and impart a certain hydrophilic character to the otherwise hydrophobic matrix and additives. The PDMS matrix now also serves as reservoir for hydrogel precursors which self-regenerate the hydrogel surface layer in case that it is damaged through e.g. mechanical abrasion.

Composition	Contact angle (deg)		
	Advancing	Receding	Static
Traditional Fouling Release Composition	105±1	54±1	104±1
Hydrogel Silicone	96±1	23±1	96±1

Table 4. Contact angles measured using distilled water, temperature kept at 22.5 °C

EXPERIMENTAL RESULTS

Contact angle measurements

Contact angles with distilled water were measured on different compositions. Table 4 shows the contact angles of a traditional fouling release coating and the Hydrogel Silicone.

Substrates prepared by drawdown method.

Results show, that the Hydrogel Silicone has a lower contact angle with water, especially the receding contact angle is less than 50% the value of a non-hydrogel commercial fouling release coating. The lower receding contact angle measured on the Hydrogel Silicone indicates that the water has a larger affinity towards this surface than towards a traditional FR surface. It is believed that this is due to the presence of highly hydrated hydrogel layers combined with hydrophobic PDMS surface functionalities.

High-throughput screening results

Webster et al. (2009) describe the use of high throughput screening methods as a useful tool to design and optimise fouling release coatings based on non-toxic mechanisms. In Figure 3 the experimental workflow is described. Candidate coatings are first pre-leached to evaluate their potential toxicity. Subsequently, the coatings are exposed to fouling organisms, namely marine bacteria (Cellulophaga lytica), marine diatom (Navicula incerta) and barnacles (Balanus amphitrite) as described in Webster et al. (2009)

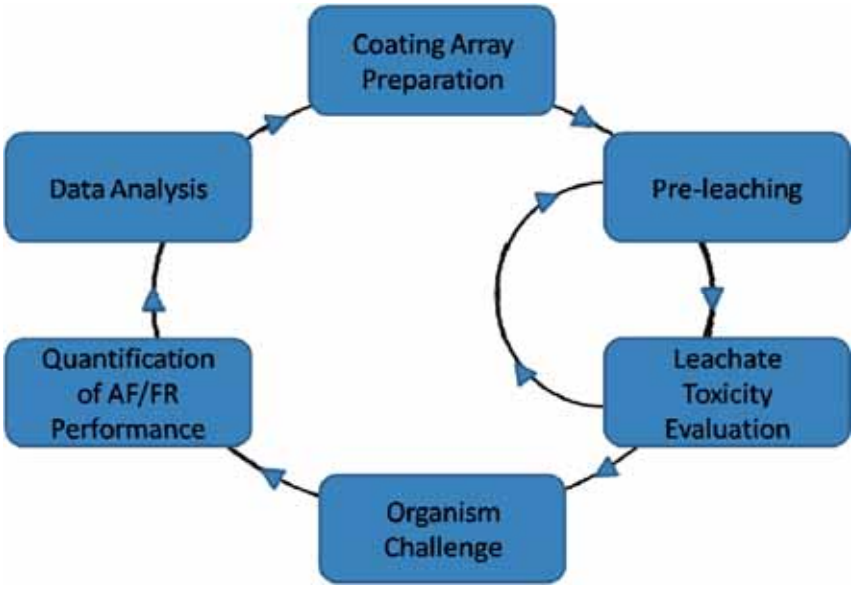

Figure 3. High-throughput biological workflow for the evaluation of antifouling marine coatings.

The results are shown in Figure 4. As references, 2 commercial fouling release coatings, one commercial silicone elastomer and one polyurethane coating (negative reference) are used.
Compared to these references, the Hydrogel Silicone coating experienced clearly lower settlement of C. lytica and N. incerta. This can be seen in the markedly lower Crystal Violet
Absorbance and Fluorescence Intensity values reported in Figure 4, which are directly related to presence of these organisms on the coating surface (a) and b) left plots. In addition to lower C. lytica settlement, the Hydrogel Silicone coating featured virtually 100% removal of those cells when the coating was exposed to a 10 psi water jet. For the case of N. incerta, the percentage removed after 10 psi washing was approximately 20% lower than one of the commercial references, but the settlement of this diatom on the Hydrogel Silicone was approximately 45% lower compared to the mentioned commercial coating. Finally, plots c) show how the Hydrogel Silicone coating was effective in inhibiting the settlement of B.
Amphitrite barnacles which, furthermore, were very weakly attached to the surface (low force was needed to detach them).

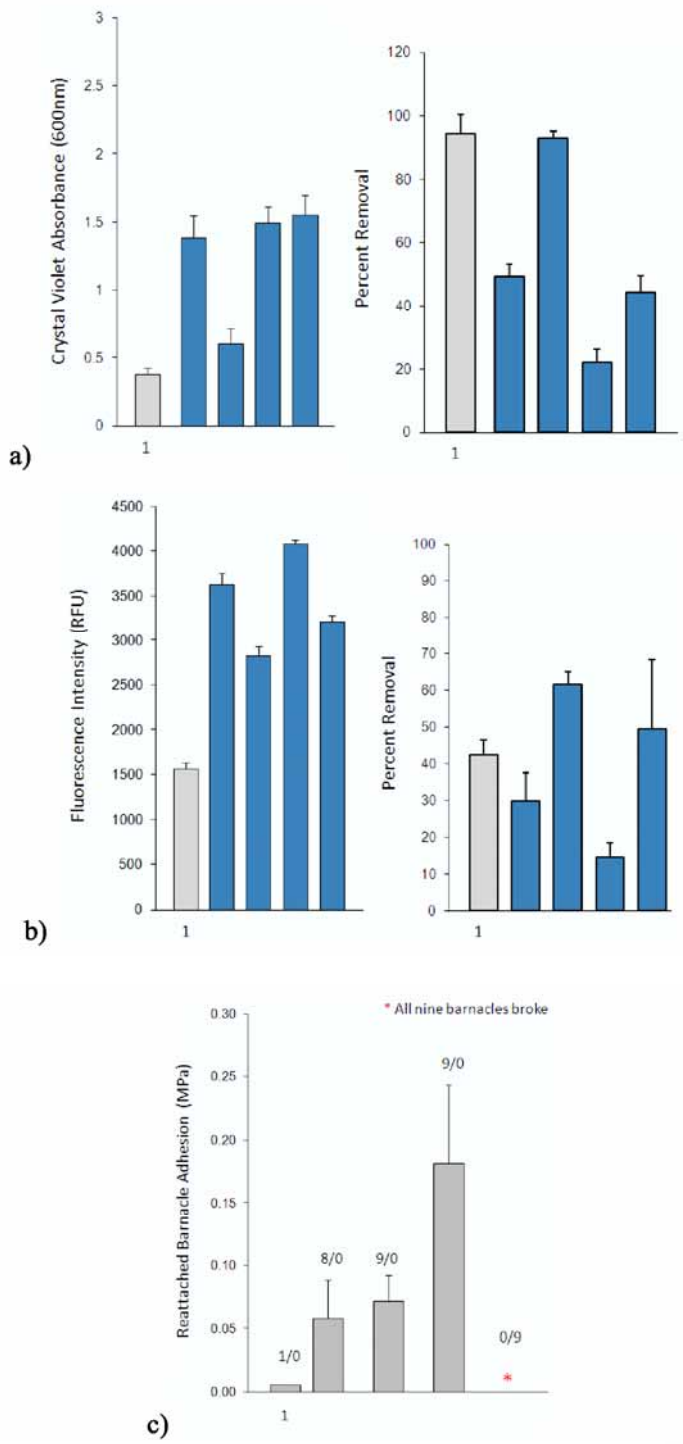

Figure 4. Results obtained from fast screening assays performed to early Hydrogel Silicone commercial coating compared to the standards. a) Settlement (left) and removal after 10 psi water jet exposure (right) of C. lytica. b) Settlement (left) and removal after 10 psi water jet exposure (right) of N. incerta. c) 2 week reattached barnacle adhesion test with B .amphitrite (measured/broken).

Exposure tests

The improved resistance to fouling of this innovative combination of a hydrogel surface in an elastic PDMS matrix has been evaluated over time in several worldwide locations (see Sanchez and Yebra, 2009). Panels coated with experimental paint compositions have been placed in near vicinity of each other and evaluated at regular intervals. The various locations cover a broad spectrum of fouling scenarios, which means that paints are exposed to both soft (algae- and slime) and hard (e.g. barnacles, tubeworms) fouling species. The first overwhelming results were obtained back in 2005 where the Hydrogel Silicone was tested alongside similar compositions with hydrophobic oils (first and second generation commercial coatings). Figure 5 shows the results after almost 4 years of static exposure in the Mediterranean Sea.

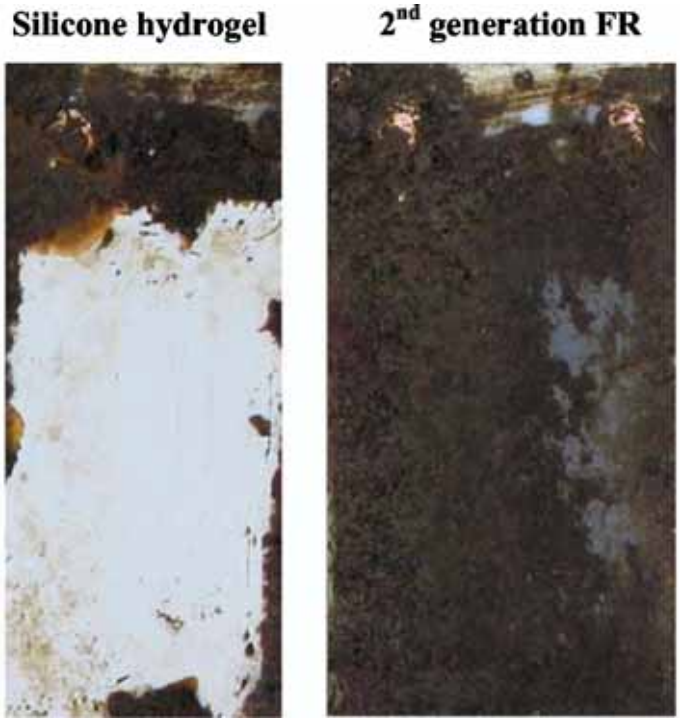

Figure 5. Effect of the addition of the Hydrogel Silicone onto a pure PDMS-based paint at a Mediterranean immersion site. The right half of the panels is subjected to periodic "cleanability" tests, to assess the release properties of the topcoat with time. Over the testing interval, the ability of the hydrogel surface layer to regenerate after such cleaning operations has become evident.

One of the differences in the panels above was also the shade, so one could argue that such a difference could have an effect, as hinted by Swain et al., (2006). Hence, subsequent tests have made sure that such an uncertainty is eliminated.

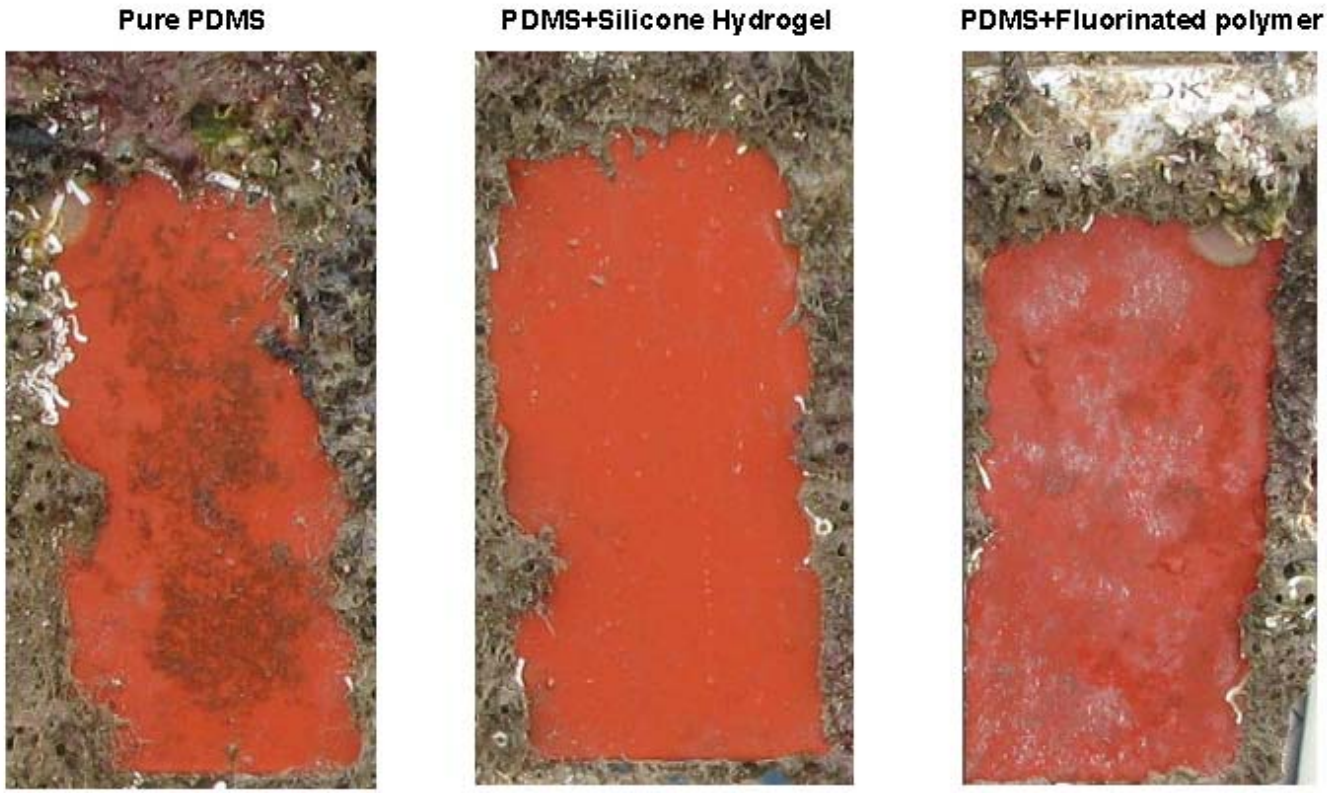

Figure 6. Performance of Hydrogel Silicone compared to a second generation hydrophobic fouling release paint and a PDMS modified with fluorinated oils. The static exposure tests were conducted at a Mediterranean immersion site (90 weeks). Note the intense fouling pressure as seen on the uncoated area of the panels.

Similar results are obtained when the paints are exposed to alternating dynamic (8 knots) and static periods (60% activity) for 90 weeks.

Figure 7. Performance of Hydrogel Silicone compared to a second generation hydrophobic fouling release paint and a PDMS modified with fluorinated oils. The dynamic/static exposure tests were conducted at a Mediterranean immersion site (90 weeks)

In tropical waters, the animal fouling pressure is much higher and barnacles, tubeworms, hydroids and bryozoans settle and grow very rapidly on blank panels (Sanchez and Yebra, 2009). The fouling control properties of fouling release coatings under these conditions are clearly shortened compared to temperate environments, especially under static testing.
In Figure 8, it is possible to observe that the Hydrogel Silicone coating clearly overperforms other technologies after roughly 4 months of static exposure, and on level/slightly better after roughly 6 months. These are very good results taking into account that these coatings are today never specified for vessels below 60% activity unless the vessel sails at really high speeds

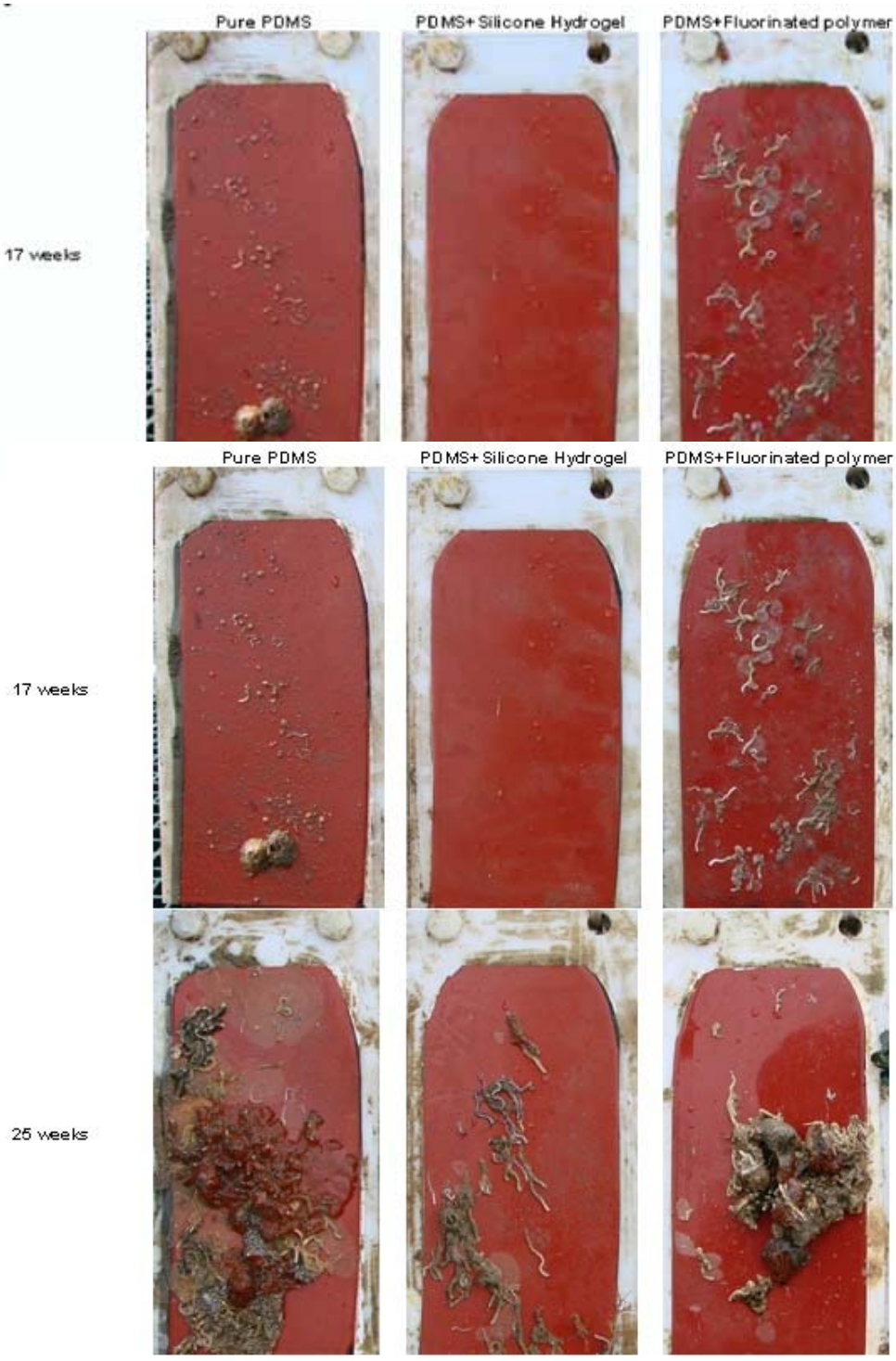

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Figure 8. Performance of Hydrogel Silicone compared to a second generation hydrophobic fouling release paint and a PDMS modified with fluorinated oils. The static exposure tests were conducted in Singapore.

Real life examples

Numerous ship trials over the past years have demonstrated the potential of this technology.

The vessel in Figure 9 has been in service for 25 months trading worldwide with 60% activity (i.e. 40% idle) and an average cruising speed of up to 13 knots with idle periods of up to one month. Both the Hydrogel Silicone test-patch and the self-polishing biocide based paint were applied during the last dry-docking of the vessel.

Figure 9. Hydrogel Silicone test patch (left) observed completely free from fouling. Algae and slime observed on self-polishing AF.

Similarly, the condition of the 380 metre long Ultra Large Crude Carrier TI Asia after 13 months of exposure is shown in Figure 10. The presence of the silicone hydrogel coating on her hull was reported to save 12 tons of fuel a day, roughly equivalent to 36 tons of CO2 and no less than 3600 USD per day.

Figure 10. TI Asia newly refurbished with 3rd generation Hydrogel Silicone fouling release technology (left) and after 13 months of sailing between the Arabic Gulf and California at an average speed of 17 knots and estimated activity of 80% (right).

CONCLUSION

Surface modification of commercial PDMS matrixes with self-stratifying hydrogel-promoting polymers markedly improves the resistance towards the settlement of fouling on traditional Fouling Release coatings, particularly slime and algae fouling. The Hydrogel Silicone technology has also shown superior performance compared to other commercial alternatives
by reinforcing the fouling release properties of the PDMS films. Static, dynamic, static/dynamic and real life tests have proved that this new technology is able to keep a fouling free surface even at conditions of low speed and low activity. On a large container vessel, the latter can potentially translate into annual savings of up to 7775 tonnes of fuel a, equivalent to 24550 tonnes of CO2, 490 tonnes of SOx and 780 tonnes of NOx (FORCE Technology, 2008).

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