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Laboratory testing of enhanced biocide mitigation of an oilfield biofilm and its microbiologically influenced corrosion of carbon steel in the presence of oilfield chemicals

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ABSTRACT

Microbiologically influenced corrosion (MIC) is prevalent in the oil and gas industry. Problematic biofilms cause MIC and reservoir souring. A high biocide concentration is usually required to mitigate biofilms compared with planktonic cells. This causes economic and environmental concerns. A biocide enhancer can make a biocide more effective using the same or lower biocide dosage. In this work, an equimolar mixture of 100 ppm (w/w) of four D-amino acids (D-methionine, D-tyrosine, D-tryptophan, and D-leucine) labeled as D-mix enhanced 100 ppm tetrakis (hydroxymethyl) phosphonium sulfate (THPS) against a field biofilm consortium on C1018 carbon steel coupons. In order to test chemical compatibilities, D-amino acids were added together with THPS and enhanced oil recovery chemicals (a polymer, a surfactant, a corrosion inhibitor, and a scale inhibitor) to treat the mature biofilm consortium. After a 7-day biofilm removal test in 125 ml anaerobic vials, the cocktail of 100 ppm THPS +100 ppm D-mix achieved extra logs of reduction in sessile cell counts compared with the 100 ppm THPS alone treatment. The combination also achieved lower weight loss and smaller maximum pit depths. Electrochemical tests corroborated the weight loss and pitting data.

1. Introduction

Enhanced oil recovery (EOR) is practiced more often because of aging reservoirs with depleting pressures (Mandal, 2015). Chemicals are usually added in water flooding (Sheng, 2014). Seawater is widely chosen as the injection fluid (Zhang et al., 2007). However, seawater injection brings nutrients and oxidants (most notably sulfate) to a reservoir, allowing microbes to flourish (Voordouw, 2011). In natural environments, most microbes live in biofilm communities which protect sessile cells (Jia et al., 2017b). Biofilms cause corrosion of pipes in many industries including marine environments, the oil and gas industry, and water systems (Lou et al., 2016; Wikieł et al., 2014; Jia et al., 2017a). This corrosion was found more than one hundred years ago, known as biocorrosion or microbiologically influenced corrosion (MIC) (Gaines, 1910). More than 20% of the total cost of corrosion was caused by MIC (Oguzie et al., 2013). It was the main suspect for the 2006 Trans-Alaska Pipeline leak which caused large economic losses and raised awareness of MIC (Jacobson, 2007).

The reservoir environment is anaerobic since geological times due to the abundance of organic carbon that consumed all the oxygen (Aitken et al., 2004). Injection water is typically deoxygenated to prevent oxygen corrosion. In such an environment, anaerobic microbes, especially sulfate reducing bacteria (SRB) flourish in the presence of sulfate (Bhagobaty, 2014). SRB are ubiquitous in anaerobic environments with sulfate. They are often associated with MIC pitting and biogenic reservoir souring (Li et al., 2016a, 2017). Although some corrosion resistant metals have been used for downhole tubing (Fu et al., 2016), carbon steels are still in use due to their low costs (Fu et al., 2016; Migahed et al., 2012). In anaerobic MIC, SRB catalyze sulfate reduction (Jia et al., 2017d). In this kind of MIC, biofilms shuttle electrons from extracellular iron oxidation across the cell wall to SRB's cytoplasm for the intracellular sulfate reduction (Jia et al., 2017f). Another type of MIC is

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caused by secreted corrosive metabolites such as organic acids which lead to much lower local pH underneath a biofilm than in the bulk fluid (Xu and Gu, 2014). Biofilms are responsible for these two types of common anaerobic MIC. In addition to MIC, biofilms also cause biofouling in many industries (Johnson et al., 2016; Zhang et al., 2015a), including reservoir souring. Thus, the MIC mitigation is about the biofilm mitigation.

In the field, microbes live in communities to form synergistic biofilm consortia (Li et al., 2016b). Biofilms defend sessile cells against antimicrobials with several defense mechanisms (Li et al., 2016b). This makes it more difficult to treat sessile cells than planktonic cells, requiring much higher biocide concentrations (Mi et al., 2014). THPS (tetrakis hydroxymethyl phosphonium sulfate) is one of the most popular biocides for large-scale applications because this biodegradable chemical has excellent broad-spectrum efficacies. THPS can disrupt the disulfide bond in microbial enzymes (Russell, 2002). Unfortunately, repeated uses of the same biocide over time may lead to a promotion of resistant microbes thus causing dosage escalation (Lalitha et al., 2017). In some oilfields, the high THPS dosage introduced so much sulfate that it precipitates Ba^{2+} in the drilling fluid, leading to the formation of problematic scales downhole (Li et al., 2016b). A high THPS dosage is costly and raises environmental concerns when discharged. Since the choices of biocides for large-scale field applications are very limited and new desirable biocides are not easy to come by, more effective application of an existing biocide such as THPS is an attractive practice to combat problematic biofilms (Xu et al., 2017).

D-amino acids exist in microbes, plants, food products, animals and even humans (Konno et al., 2009). In recent years, they were found to disperse bacterial biofilms. D-methionine (D-met), D-leucine (D-leu), D-tyrosine (D-tyr), and D-tryptophan (D-trp) were found to disperse *Staphylococcus aureus*, *Bacillus subtilis*, and *Pseudomonas aeruginosa* biofilms (Kolodkin-Gal et al., 2010). However, Kao et al. (2017) reported that D-amino acids alone failed to inhibit the *P. aeruginosa* biofilm formation. Therefore, a biocidal stress is recommended together with D-amino acids for recalcitrant biofilms (Xu et al., 2012). One ppm (w/w) D-tyr and 100 ppm D-met individually were found to enhance 50 ppm THPS against the *Desulfovibrio vulgaris* (an SRB) biofilm on carbon steel coupons (Xu et al., 2012, 2014). One ppm D-tyr and 50 ppm D-met individually were also found to enhance 10 ppm alkyldimethylbenzylammonium chloride against the *D. vulgaris* biofilm on carbon steel (Jia et al., 2017c). D-tyr at 2 ppm enhanced 30 ppm ciprofloxacin against a *P. aeruginosa* biofilm on carbon steel in a 7-day biofilm prevention test (Jia et al., 2017e). Zilm et al. (2017) reported that antimicrobials alone did not significantly reduce the biofilm, while the cocktail of antimicrobial + D-amino acids were able to reduce *Enterococcus faecalis* biofilms significantly. Li et al. (2016b) found that individual D-amino acids had a limited ability to enhance THPS against field biofilm consortia. However, several D-amino acids combined together worked effectively with THPS. It was possible that different microbes in a biofilm community responded to different D-amino acids.

EOR implements various chemicals such as polymers and surfactants to increase oil production (Mandal, 2015). These EOR chemicals are typically applied in a single batch application in the oilfield with other chemicals including scale inhibitors, corrosion inhibitors and biocides. Thus, D-amino acids and THPS should be chemically compatible with EOR chemicals, corrosion inhibitors and scale removers. In this work, an equimolar D-amino acid mixture (D-mix) of four D-amino acids containing D-met, D-trp, D-tyr, and D-leu was evaluated to enhance THPS against a field biofilm consortium on C1018 carbon steel which is similar to X65 pipeline steel in MIC behavior (Xu et al., 2014; Zhang, 2014).

2. Materials and methods

2.1. Microbes and chemicals

A corrosive field biofilm consortium from an oilfield labeled as Consortium II was used in this work. The metagenomics results showed that it contained SRB, fermentative microbes and biodegradation microbes (Li et al., 2016b). Enriched artificial seawater was used to culture this biofilm. Its composition (g/l) was: Na_2SO_4 3.917, NaCl 23.476, $NaHCO_3$ 0.192, KBr 0.096, KCl 0.664, H_3BO_3 0.026, $SrCl_2 \cdot 6H_2O$ 0.040, $MgCl_2$ $6H_2O$ 10.610, $CaCl_2 \cdot 2H_2O$ 1.469, yeast extract 1, tri-sodium citrate 0.5, sodium lactate 3.5, $CaSO_4$ 0.1, NH_4Cl 0.1, $MgSO_4 \cdot H_2O$ 0.4, $Fe(NH_4)_2(SO_4)_2$ 0.5, K_2HPO_4 0.05. One hundred ppm L-cysteine was added to the culture medium to eliminate any possible oxygen ingress. The initial pH of the enriched artificial seawater was adjusted to 7.0 before inoculation. Microbes were grown in 125 ml anaerobic vials (Wheaton Industries Inc., Millville, NJ, USA). The EOR polymer (partially hydrolyzed polyacrylamide), the corrosion inhibitor (imidazoline salt), the surfactant (alpha olefin sulfonate), and the scale inhibitor (diethylene triamine penta (methylene phosphonic acid) (DTPMPA)) were provided by our sponsor. All the other chemicals were purchased from either Fisher Scientific (Pittsburgh, PA, USA) or Sigma-Aldrich (St. Louis, MO, USA). The anaerobic vials together with their septa and aluminum caps, and culture medium liquid were sterilized at 121 °C for 20 min in an autoclave. Solutions of THPS and D-amino acids were sterilized with a 0.22 μm Stericup (Millipore, Bedford, MA, USA). Dissolved oxygen in liquid solutions was removed by sparging with filter-sterilized N_2 for at least 1 h.

2.2. Biofilm removal test

Biofilms were grown on C1018 carbon steel (UNS G10180) coupons in anaerobic vials. Each coupon had a top exposed surface area of 1 cm^2. Inert Teflon paint was used to protect the other coupon surfaces. The composition of carbon steel was (w/w): Mn 0.60–0.90, C 0.14–0.20, P 0.04, Si 0.15–0.30, S 0.05, and Fe balance (Jia et al., 2017b). Coupons were abraded sequentially with 180, 400 and 600 grit abrasive sheets. Each vial contained 5 coupons in 100 ml culture medium. Each vial was inoculated using 1 ml seed culture in an anaerobic chamber to yield an initial planktonic cell count of 10^6 cells/ml. All the vials were capped and incubated without shaking at 37 °C for 4 days. It was found that in the lab test, 4 days were enough to achieve biofilm maturity. Mature biofilms were used for different chemical treatments in this work to simulate established biofilms. After that, the vials were opened in the anaerobic chamber. One thousand ppm partially hydrolyzed polyacrylamide (polymer), 5000 ppm alpha olefin sulfonate (surfactant), 200 ppm imidazoline salt (corrosion inhibitor), and 15 ppm DTPMPA (scale inhibitor) were added to all treatments. The biocide alone treatment contained 100 ppm THPS. The cocktail treatment contained 100 ppm THPS and 100 ppm D-mix. All chemicals were injected into the vials at the same time to treat the biofilms in a new 7-day treatment period at 37 °C. After the additional 7-day treatment, coupons were taken out for assays. The abiotic control contained coupons, the culture medium, and treatment chemicals without inoculation. This biofilm removal test was performed at least 3 times for reproducibility.

2.3. Sessile cell count and biofilm examination

Coupons were retrieved from the vials after the biofilm removal test to enumerate sessile cells. The most probable number (MPN) method was adopted using three different liquid culture media (Biotechnology Solutions, Houston, TX, USA). They are standard