RESERVOIR DIMINISHED PRODUCTIVITY CAUSED BY DOWNHOLE BIOFILM GROWTH IN SHALE PLAYS

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Palabras clave: biofilm, unconventional, biocide, protection

ABSTRACT

Las operaciones de perforación y terminación asociadas con la fractura hidráulica introducen contaminantes microbianos en el reservorio a través de numerosas vías, incluidos lodos de perforación, fluidos de terminaciones y propante. Una proporción de estos microbios introducidos se establecerá en el reservorio, lo que podría conducir a: a) Aumento de las concentraciones de sulfuro de hidrógeno (H2S) durante la producción; b) Bioincrustación y corrosión por influencia microbiana (MIC) de equipos en superficie y de fondo del pozo y; c) Reducción del caudal y producción de hidrocarburos debido a la acumulación de biofilm y taponamiento. Tradicionalmente, abordar las preocupaciones microbianas en la fractura hidráulica se ha centrado en control de agriamiento o MIC. En este trabajo, diseñamos una técnica novedosa para estudiar los efectos del crecimiento de biopelículas en el "Shale" y sus impactos en las tasas de producción. En la fractura hidráulica, los biocidas se inyectan en el reservorio como un componente del fluido de fractura durante la terminación del pozo. Dado que solo hay una oportunidad para inyectar el biocida en el reservorio durante este proceso, es fundamental seleccionar un biocida efectivo y una tasa de dosificación en función de las condiciones específicas del campo.
Como resultado de este estudio se pudo observar que el crecimiento incontrolado de bacterias condujo al crecimiento de biopelículas y a la producción de FeS, lo que resultó en una pérdida del 10-20% en la conducción que habilita el propante. Esto se correlaciona con una pérdida de petróleo de aproximadamente 6000 barriles durante los primeros 180 días de producción. Al final, este trabajo destaca la importancia de un programa de control microbiano para garantizar que la formación de biopelículas y las reacciones mineralógicas con H2S no impidan las vías de flujo en los reservorios de fracturación hidráulica e impacten negativamente en las tasas de producción.

INTRODUCTION

Hydraulic fracturing of shale in unconventional oil & gas recovery is accomplished through the addition of large volumes of water and biodegradable additives to the reservoirs, creating environments conducive for microbial contamination. Contamination of reservoirs by sulfate reducing organisms often leads to biogenic souring, the biological production of hydrogen sulfide (H2S), which devalues crude, corrodes metallic assets via microbially influenced corrosion (MIC), and creates dangerous

health and safety conditions for field personnel (Ferrar *et al.* (2021)). Often overlooked is the impact of this downhole contamination on formation damage and hydrocarbon recovery.

Microbes introduced into a hydraulically fractured reservoir form biofilms by attaching to the rock or proppant surfaces and secreting a lipopolysaccharide "gel" that protects the microbial colony. These biopolymers are dynamic and have the potential to affect reservoir permeability and conductivity in tight shales. Furthermore, biofouling can impact the reservoir pore mineral cementation via biomineralization and the production of iron sulfide scale. Bottero *et al.* (2010) modelled formation damage and the impact on gas flow caused by biofilms growing within proppant packs of hydraulically fractured reservoirs. They concluded that, in the presence of 10% biofilm volume, the gas production flow rates were reduced by half and that the biofilm growth on proppants affected the gas flow rate due to the differential change in the medium tortuosity and flow patterns.

The development of effective solutions for reservoir biosouring, bioclogging, and scaling requires a thorough understanding of in-situ microbe-rock-water-chemistry interactions, and mechanisms of biofilm formation, souring, and cementations under actual subsurface hydrocarbon reservoir conditions. Furthermore, it requires the practical understanding of biofouling impacts on conductivity in industry-accepted assays. It has been experimentally challenging to perform such studies under representative reservoir conditions in the laboratory or field. Resultantly, two projects were undertaken to develop these capabilities and to develop data sets that can be translated to oil and gas applications. The first is the capability development of the microfluidic device called GeoBioCell which is detailed in Fouke *et al.* (2021) and includes:

- Development of a GeoBioCell experimental system (silica and real rock) that operated under representative subsurface reservoir conditions to study interactions amongst microbial biofilms, reservoir rock, chemical treatments and oil.
- Understanding how interactions between microbial control technologies and reservoir porous media affect microbial biofilm control effectiveness.
- Understanding dynamic changes in microbial biofilm formation and dispersal in response to pore-scale geochemical condition changes.
- Collection of scientific evidence to support the hypothesis that biofilm development and associated biomineralization within subsurface reservoir lithologies, including near bore areas, can negatively impact reservoir biosouring control.
- Understanding how current antimicrobial technologies can maintain reservoir integrity.
- Initiate use of these results to better position current products, provide successful targeted directions for improved formulations and new technology development, as well as evaluate new technologies for reservoir integrity maintenance.

The second project involves the adaptation of an American Petroleum Institute (API) pressurized conductivity cell with two shale cores and proppant that has been anaerobically inoculated with field relevant microorganisms to evaluate biofilm impact on reservoir conductivity.

METHODS

Shale GeoBioCell Design

The design details of Shale GeoBioCell microfluidic channel test bed are illustrated in Figure 1 and are further detailed in Fouke *et al.* (2021). It is composed of a 2-cm-wide by 4-cm-long by 2-cm-thick rectangular piece of New Albany shale cut into four pieces. These included a triangular inlet piece, a rectangular outlet piece, and two longer pieces forming the sides of the inlet channels and the primary microfluidic channel. The faces along the length of the microchannel were polished smooth on a 600-grit lap wheel. The primary microfluidic channel was created with the two longer shale pieces (separated by two 250 µm thick sheets of wax-coated paper) and the triangular inlet piece epoxied atop two stacked and epoxied glass sides, and then cut down and polished to a thickness of 350 µm. The wax-coated paper was subsequently removed, leaving a 500-µm-wide by 350-µm-deep microfluidic channel that simulates subsurface shale fractures. One of the glass slides was notched into which polyetheretherketone tubing was attached to each of the two inlets and outlet. The entire assembly was then sealed from the top with an epoxied glass coverslip. The proppant grains introduced into the channel were 100 mesh moderately well-sorted quartz sand grains sieved through a 40-mesh sieve. Proppants were suspended in 100% ethanol, loaded into a 10-ml syringe, and injected into the GeoBioCell channel through the inlet. The GeoBioCell was gently tapped while injecting the slurry to assist in packing of proppants with minimum space gaps in the channel. The GeoBioCell assembly was then inserted into a custom-made aluminum

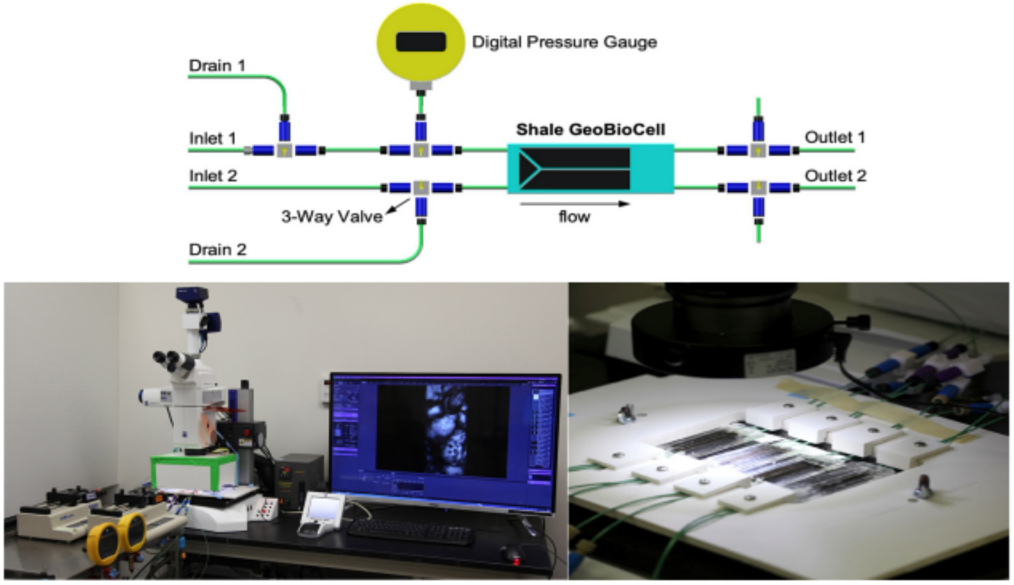

Figure 1. (Top Image) Diagram of the GeoBioCell highlighting flow through the system and fracture channels. (Bottom Left Image) Zeiss AxioZoom V16 microscope on which the shale GeoBioCell experiments were run. (Bottom Right Image) Expanded manifold that holds four shale GeoBioCell testbeds which permits four experiments to be conducted in parallel.

stage insert, attached to inlet pumps, valves, and a pressure transducer, and placed on a Zeiss AxioZoom.V16 microscope stage.

Shale GeoBioCell Operation

A field culture, dominated by sulfate-reducing bacteria (SRB), was isolated from a North American oil and gas producing well and grown on a previously described media (Sandford and Tiedje (1996)) amended with a volatile fatty acid (VFA) mixture containing acetate (13 mM), formate (15 mM), butyrate (1.2 mM), and propionate (1.4 mM) and incubated at 45oC. The proppant-filled channel in each GeoBioCell was saturated with growth medium media, and mature field culture was injected into inlet 1 with a fresh VFA mixture injected into inlet 2 to prevent growth and clogging of the injection tubing. After a 12 hr inoculation period, injection rates were maintained at a flow rate of 100 ml/min and were operated for six to ten days until the pressure drop across the device became too high to sustain flow. All experiments were performed in duplicate at 45oC.

Imaging of biofilm growth, iron sulfide mineral precipitation, injected proppants, and shale within the GeoBioCell microfluidic channels was done on a Zeiss Axio Zoom.V16 fluorescence microscope. For environmental scanning electron microscope (ESEM), a Thermo Fisher Quanta 450 FEG ESEM was used with samples prepared and sputter-coated according to procedures outlined by Fouke *et al.* (2021).

API Conductivity Cell Design

The American Petroleum Institute (API) conductivity cell that accommodates two core slabs 7 inches long by 1.5 inches wide and about 0.4 inch or more thick is shown in Figure 2. The baseline cell (no frac fluid) differs from the Figure 2 diagram by not having the leak-off capabilities, and the entrance and exit flow ends are the same design as the pressure monitoring or dP (delta pressure) ports are illustrated at the bottom of the upper diagram. Initially, one core is placed in the conductivity cell and the desired amount of proppant to be evaluated placed over the core surface and leveled. As demonstrated in the image, the second core is then placed on top of the proppant to complete the proppant/core sandwich or fracture. The individual cell is then stacked together and placed in a hydraulic press. Heat bands are applied to the cells. The differential pressure is measured across the internal 5 inches of the pack and used with the flow rate and fluid properties to calculate the fracture conductivity.

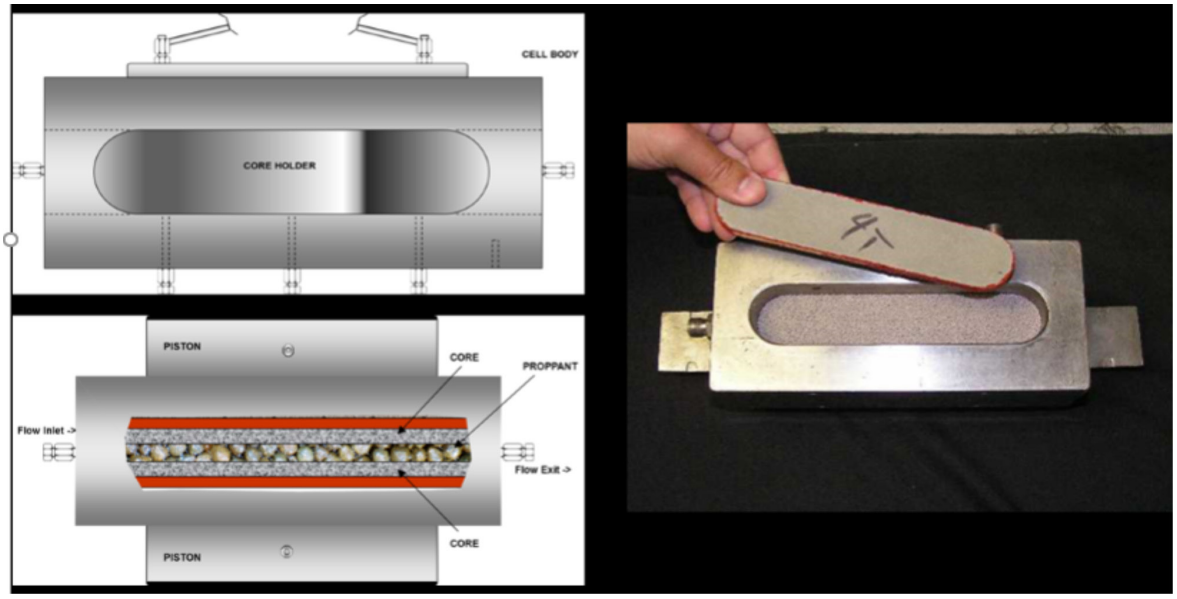

Figure 2. (Left images) Diagram of the key components of the API Conductivity cell highlighting the core slabs, core holder and proppant placement. (Right) Image of the core holder and placement of core slabs.

API Conductivity Cell Operation

A field culture isolated from produced water in a North American shale play was used in this assay. This culture was maintained in marine broth amended with thiosulfate at 60oC and was dominated by Thermoanaerobacter, a spore forming, thiosulfate-reducing bacteria that can produce copious amounts of H2S. Thermoanaerobacter, was grown in marine broth amended with the VFA mixture described above along with thiosulfate and incubated at 60°C. This culture was grown for 72 hr prior to injection followed by a 7-day shut-in period after injection into the conductivity cells.

Conductivity was evaluated at 2000 psi and 60°C with 2% KCl for the pre-injection baseline conductivity evaluation and post-field culture fluid regain and conductivity analysis. The pre–injection conductivity allowed for direct comparison of cleanup of the injected field culture with a proppant concentration of 1 lb/ft². The proppant used for this study was a 40/70 mesh Premium Northern White Sand, and the shale cores were from the Niobrara field. The field culture was injected into each test cell entrance port while maintaining the deoxygenated 2% KCl in the conductivity flow lines. A bypass flow backpressure regulator was placed on the entrance to void the fluid between the valve on the cylinder and the injection valve on the entrance of the 2% KCl injection lines used to evaluate the baseline conductivity. The differential pressure lines remained on the proppant pack (valves shut), and only the proppant pack was flowed with field culture. The cells were then shut-in to a back-pressure regulator set at 400 psi. The cells remained shut-in for 7 days to allow growth of anaerobic field culture at 60°C and 2000 psi before initiating flow and obtaining regain conductivity for plugging growth analysis. Data tables reflect closure stress vs.

conductivity and permeability results. All tests were performed in duplicate and in Niobrara Shale core slabs used for generic shale testing.

RESULTS AND DISCUSSION

GeoBioCell

Microbial growth within subsurface pores and fractures has both positive and negative impacts on oil and gas extraction, recovery, and storage. As detailed in Fouke *et al.* (2021), this work primarily targeted the enhancement of subsurface reservoir flow by spatially selective occlusion of porosity and permeability by microbial biofilm (Gerlach and Cunningham, 2010). However, little has previously been known regarding the mechanisms and processes by which microbial biofilms impact the permeability of shale fractures. Resultantly, the Shale GeoBioCell was constructed to track in real-time these microbe-mineral-water interactions within proppant filled microchannels constructed within core samples of New Albany Shale.

The proppant-filled shale fracture was imaged at time points before, during, and after experimentation at a location in the microchannel within millimeters of the GeoBioCell inlet and

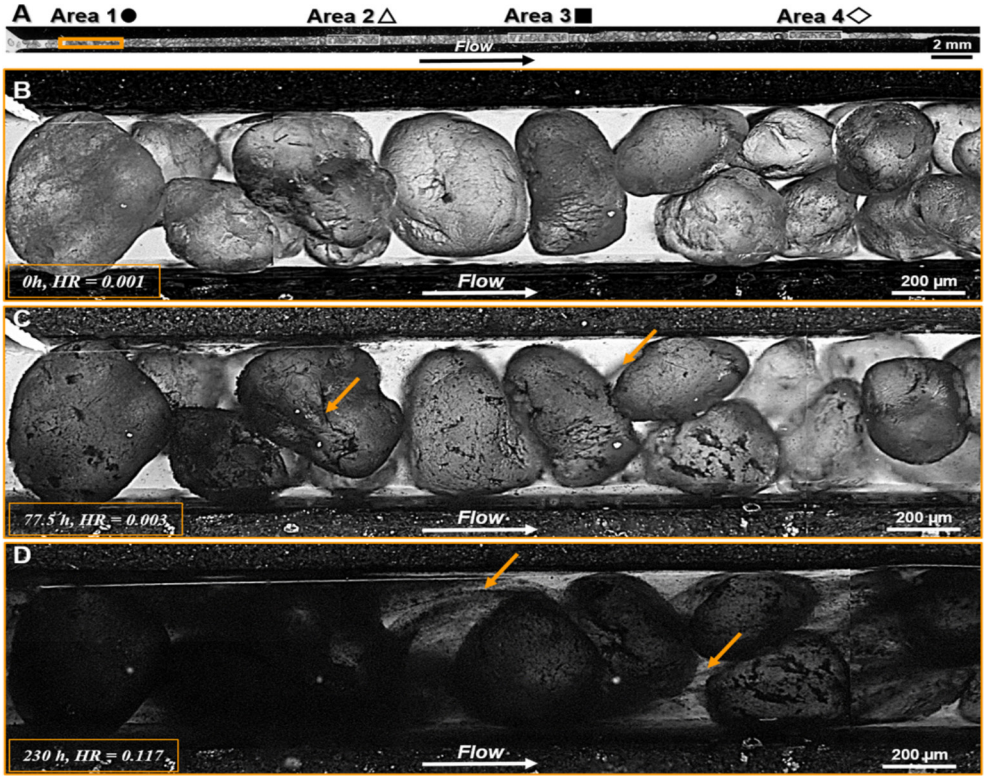

Figure 3. Brightfield imaging and image intensity graphs of sulfate-reducing bacteria (SRB) biofilm growth, iron sulfide (FeS) mineral precipitation, and proppants within the shale GeoBioCell. Image B is before inoculation of the field culture and is the enlargement of the orange box in Image A. Image C is the GeoBioCell after 78 hr and Image D after 230 hr.

outlet (Figure 3). Imaging before inoculation (Figure 3B) indicates that the original clean shale channel walls and proppant grain surfaces are clearly visible at both upstream and downstream locations, and no biomass or mineral precipitates are apparent. Approximately 78 hr after inoculation at the upstream location (Figure 3C), SRB biofilm coatings and streamers become visible and spherical amorphous iron sulfide minerals turn the biomass dark and opaque. As experimentation progresses (230 hr), pore spaces become darker and sometimes opaque, rendering the shale walls, proppants, and pore spaces virtually indistinguishable (Figure 3D). Visible in some pores are SRB streamers that span pore spaces and connect adjacent proppants. The results suggest SRB biofilm coatings, streamers, and associated iron sulfide precipitation increase with time at the upstream location until pore spaces appear almost occluded (Figure 3D). ESEM imaging (Figure 4) further demonstrates biofilm growth in the GeoBioCell channels connecting the sand grains with the shale surface and obstructing flow through the pore spaces.

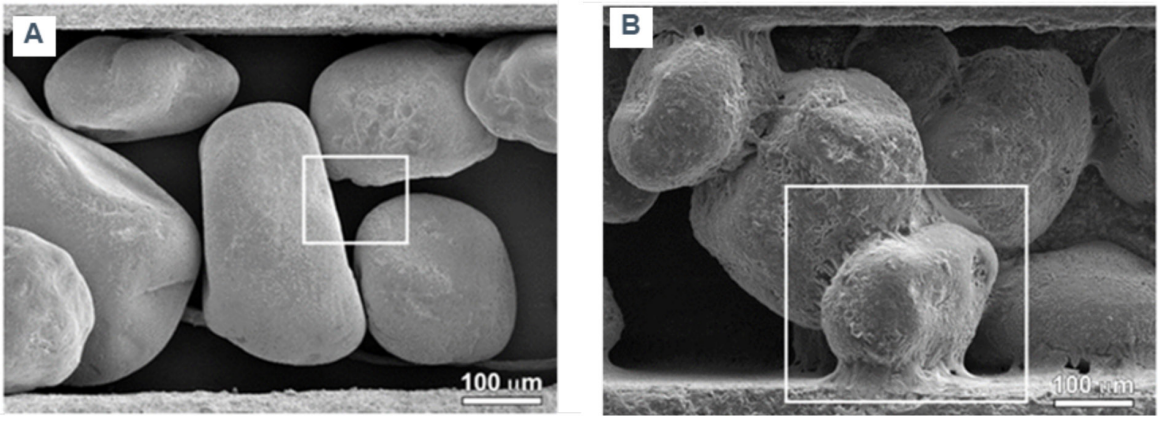

Figure 4. ESEM imaging of uninoculated (Image A) proppants within the GeoBioCell and 230 hr (Image B) after field culture inoculation. The white boxes highlight open pore space in Image A and biofilm occlusion in Image B

As detailed in Fouke *et al.* (2021), a fundamentally important relationship required to predict oil and gas recovery from fractured shale reservoirs is the association between increasing biomass growth and increasing hydraulic resistance (HR) measured as function of pressure drop during hydrocarbon production. Pressure changes were measured for each shale GeoBioCell microchannel every ~12 hours in replicate experiments which were used to calculate HR (Figure 5). During the first three days (~80 h), HR increased only marginally in both replicates. However, on the third day for shale GeoBioCell replicate 2, and on the seventh day for shale GeoBioCell replicate 1, HR started to increase very rapidly, going from less than 0.01 to 0.12 psi/min in less than 3 days in both cases. Once HR exceeded 0.12 psi/min, the experiments were stopped because higher HR would break the cover glass encasing the shale GeoBioCell microchannel. These results, in combination with the real-time imaging and ESEM analyses at each critical time point, indicate that HR increase in the shale GeoBioCell microchannel is due to porosity and permeability occlusion by SRB biofilm coatings and streamers and FeS precipitation.

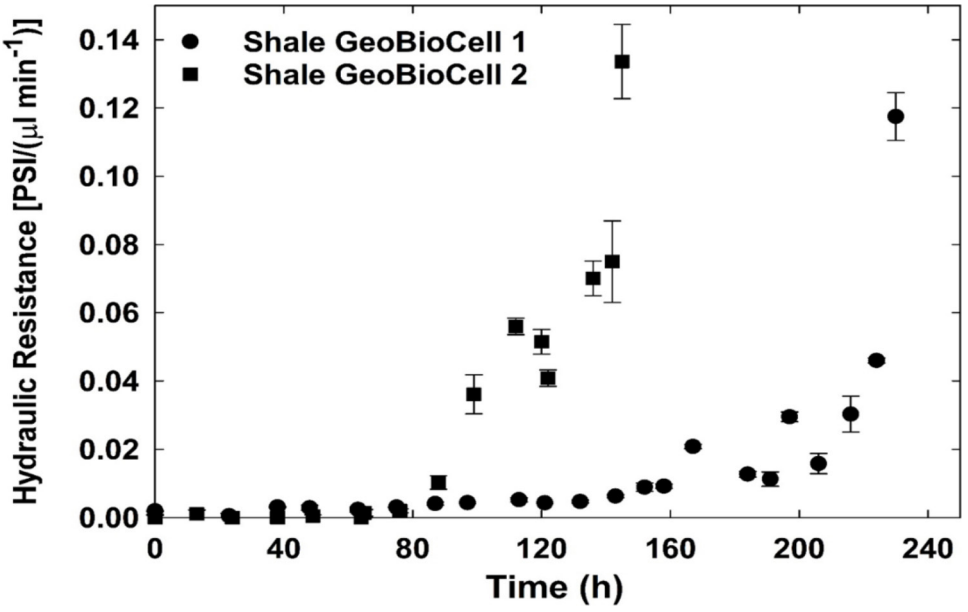

Figure 5. Graph of duration of experiment on x-axis versus hydraulic resistance on y-axis for the two proppant-filled, replicate shale GeoBioCells.

API Conductivity Cell

The API conductivity cell has been used for years by the hydraulic fracturing industry to determine conductivity and permeability at fracture closing stress and to evaluate proppant optimization and damage mechanisms. Furthermore, these datasets can allow operators to understand the impact of completions fluids on proppant performance at reservoir and downhole conditions which can be incorporated into hydraulic fracture simulators available to industry (Duenckel *et al.* (2016)). Our experiments were performed to test the influence of uncontrolled biofilm growth, originated from the completion fluids, on reservoir conductivity and hydrocarbon recovery.

Figure 6 compares the conductivity of API cells that have been inoculated with shale-relevant microbes for 7 days prior to testing versus uninoculated controls, indicating that there is a ~17% loss of conductivity in the treated vs. untreated cells. Treated cells were inoculated with a field culture dominated by Thermoanerobacter that presumably was able to attach to the proppant and shale during the 7-day incubation period, produce a layer of protective biopolymers forming a biofilm, metabolize the introduced VFAs, and reduce thiosulfate to H_2S. As demonstrated in Figure 7, the biogenically produced H_2S reacted with iron in the shale, producing iron sulfide (FeS). The lower photo of Figure 7 illustrates a close-up of the proppant pack showing the shiny, translucent biofilm on the individual sand grains surrounded by black FeS scale.

The growth of biofilm and precipitation of FeS scale in the treated API cell negatively impacted conductivity. To translate the ~17% conductivity loss to hydrocarbon recovery, the

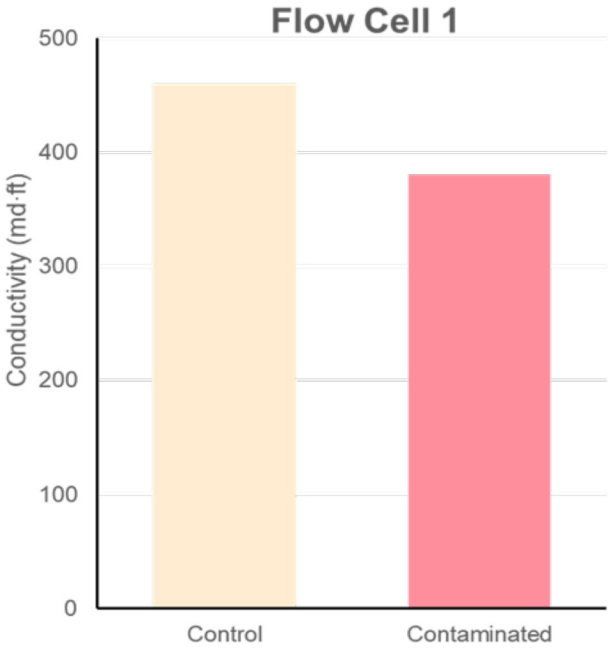

Figure 6. Graph measuring conductivity data obtained for the 1 lb/ft² 40/70 Premium Northern White Sand both before and after being inoculated with shale-relevant microbes (72 hr. incubation) and shutting in for growth period of 7 days. Conductivity is measured in millidarcy per foot (md/ft)

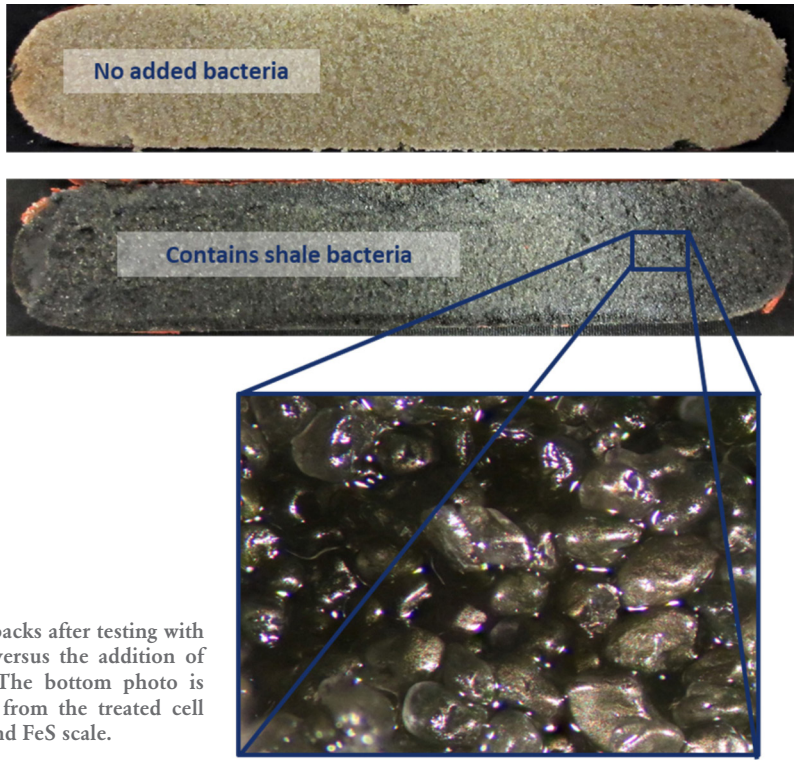

Figure 7. Images of the proppant packs after testing with no added microbes (top photo) versus the addition of shale microbes (middle photo). The bottom photo is a close-up of the proppant pack from the treated cell showing the presence of biofilm and FeS scale.

Predict-K software package (Barree *et al.* (2003)) was used to correlate the measured conductivity impacts to accumulated barrels of oil. Under the standard software conditions and a reservoir permeability of 0.1 md, 6,010 standard barrels of oil (STB) were not recovered over the first 180

days of production. If the permeability was increased to 0.2 md, 3,250 STB were not recovered over the first 180 days of production. These results, in combination with the proppant pack imaging also indicate that conductivity loss and reduced hydrocarbon recovery is due to porosity and permeability occlusion by biofilm coatings and FeS precipitation.

CONCLUSIONS AND RECOMMENDATIONS

A novel, flow-through shale microfluidic testbed, called the shale GeoBioCell, was used to determine the combined influence of sulfate reducing microbes biofilms and FeS biomineralization on porosity occlusion, decreased permeability, and increased hydraulic resistance (HR) in shale fractures (Fouke *et al.* 2021). Similarly, results from the API Conductivity Cell study also concluded that the presence of Thermoanerobacter dominated biofilms and FeS biomineralization in proppant packs negatively impacted porosity, permeability, reservoir conductivity and hydrocarbon recovery. These two studies provide convincing evidence that uncontrolled microbial growth of completion fluids can lead to formation damage and the loss of hydrocarbon recovery during the first six months of production.

Demonstrating that a microbial control program can provide long term biofilm and FeS control in a hydraulically fractured reservoir has challenged the industry for years. Our labs recently developed a suite of High Pressure, High Temperature (HPHT) bioreactors specifically designed to simulate the downhole environment of a hydraulically fractured reservoir (Ferrar *et al.* (2020)). These reactors can evaluate the extent of microbial contamination in a reservoir by simulating extended shut-ins at reservoir-relevant temperatures and pressures in the presence of formation solids and fracturing fluid additives (anionic friction reducer and proppant). Subsequently, these HPHT reactors were used to evaluate the performance of numerous preservative biocides for extended downhole protection from microbial contamination (Ferrar, *et al.* (2021)). This work highlighted the performance of 4,4-dimethyloxazolidine (DMO) for control of biogenic souring

Preservative Treatment	Glutaraldehyde (ppm a.i.)*	DMO (ppm a.i.)**	THNM (ppm a.i.)	Relative Strength of Downhole Protection	Residual Biocide Stability and Activity
Option 1: Complete		100 – 200 ppm	100 – 200 ppm	+++	> 8 weeks
Option 2: Moderate		40 – 100 ppm	75 – 100 ppm	++	2 – 8 weeks
Option 3: Basic	100 – 200 ppm			+	1 – 2 weeks

Table 1. Recommended preservative concentrations for the long-term control of reservoir microbial contamination. a.i. refers to the active ingredient of the biocide formulation.

and reservoir contamination for weeks-to-months at a time, depending on biocide dosing level. This performance was partially due to the favorable chemical compatibility of this active with the numerous anionic species and surfaces encountered during completion, shut-in, and production. Ultimately, field recommendations derived from these studies for preservatives and glutaraldehyde are summarized in Table 1.

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