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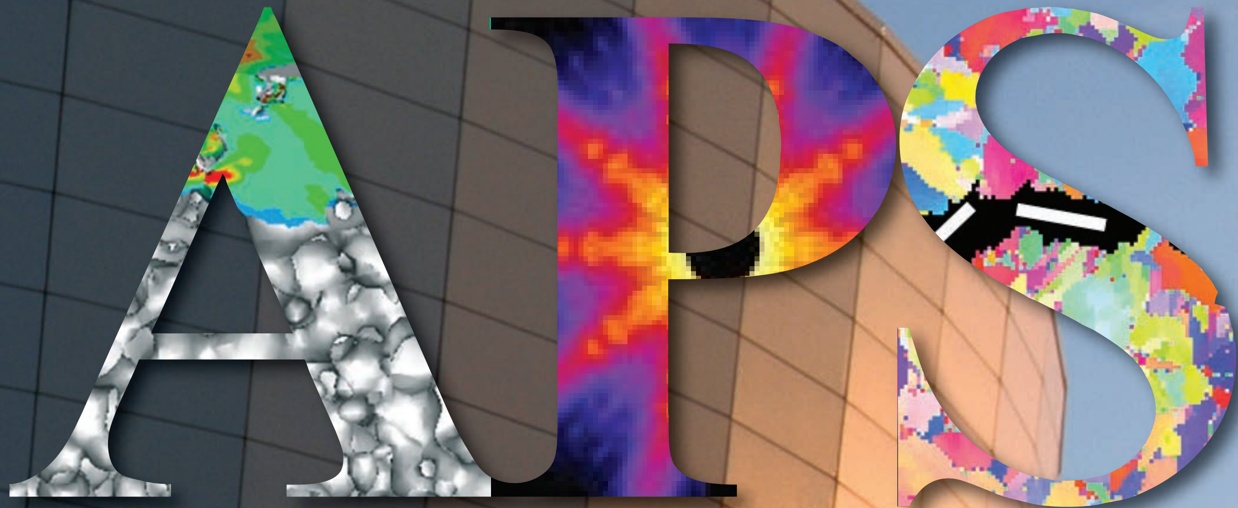

Research and Engineering Highlights from the Advanced Photon Source at Argonne National Laboratory

Includes APS research into the novel coronavirus

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On the cover: Background: The APS auditorium exterior at dusk.

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Argonne is a U.S. Department of Energy (DOE) laboratory managed by UChicago Argonne, LLC.
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Oil Blob Imagery for Assessing Surfactant-Enhanced Aquifer Remediation

Oil pipelines and fracking operations frequently make headlines because of their potential for, and past history of, groundwater contamination, an enormous public safety hazard. Remediation processes exist but aren't necessarily well-understood. One such process, surfactant-enhanced aquifer remediation, injects surfactants – substances that lower the surface tension between liquids – into the contaminated soil or aquifer and then relies on flushing to displace and extract the hopefully modified, more-mobile oil-phase contaminant from the system. But how exactly do oil drops, wetting agents, and soil make-up interact on the scale at which this technique works? Recent research at the APS provides images of the oil distribution and how it changes throughout the flushing process, continuing to characterize the circumstances under which this technique is most effective.

In many cases, significant effort is required to remove crude oil from soil and groundwater because it is a non-aqueous phase liquid resistant to mixing with or dissolving in water. As such, these high-viscosity, low-solubility oil phases can essentially become trapped within pore spaces, making it difficult to mobilize when flushing with water alone. Surfactant-enhanced aquifer remediation techniques assume the surfactant will sufficiently lower the interfacial/surface tension of the oil (between aqueous solution and solid media), allowing it to more easily mobilize through the pore networks and/or mix with water (during flushing events), thereby enabling the oil phase to be transported through the sand/soil and removed from the aquifer more expeditiously. A multinational team of researchers assessed the surfactant-enhanced aquifer remediation process on the pore scale to quantify how the distribution of oil trapped in soil is dependent on the size of the physical characteristics of the oil-water-sand system, and how it changes with each flushing event.

To study the effect of these characteristics, the team constructed three experimental setups, added heavy crude oil to them, and flushed them with water several times, taking measurements after each flushing. The setups differ by the amount of unconsolidated sand-size fraction used to define three distinct pore (permeability) configurations: a homogeneous sand (homogeneous), a heterogeneous sand (mildly heterogeneous), an extremely heterogeneous sand (highly heterogeneous). The team's experimental process was to pack columns with the appropriate sand, saturate the column with liquid, add the heavy oil, flush the system with the surfactant, and image the resulting sand and oil network using novel synchrotron x-ray microtomography (SXM) performed at the GSE-CARS 13-BM-D beamline at the APS. The three-dimensional microtomography images allowed the team to measure oil distribution, blob volume, and the number of blobs before and after each surfactant flushing event.

The team's image results showed that the size of an individual oil blob, blob distribution, and the total volume of oil present in the columns were different due to both the specific sand (porous media) configuration in the experimental set-up and the flushing events (Fig. 1).

The oil distribution within the homogenous sand-packed column was predominantly comprised of a single large, interconnected ganglion of oil and progressive surfactant flushing did not do much to fragment distribution (or impart enough interfacial tension reduction effect for removal), so a negligible fraction of the oil was removed during the flushing process.

The oil distribution in the mildly heterogeneous sand-packed column was comprised of many discrete blobs of oil; the percentage of oil saturation was an order of magnitude smaller than in the homogeneous column under the same preparation and experimental conditions. The team found that although progressive surfactant flushing events caused some blob fragmentation and re-distribution, only a negligible relative fraction of heavy oil phase was removed from the system.

The oil distribution in the highly heterogeneous sand-packed column also was initially comprised of many discrete blobs prior to surfactant flushing, under a low oil saturation condition. However, a more pronounced enhanced

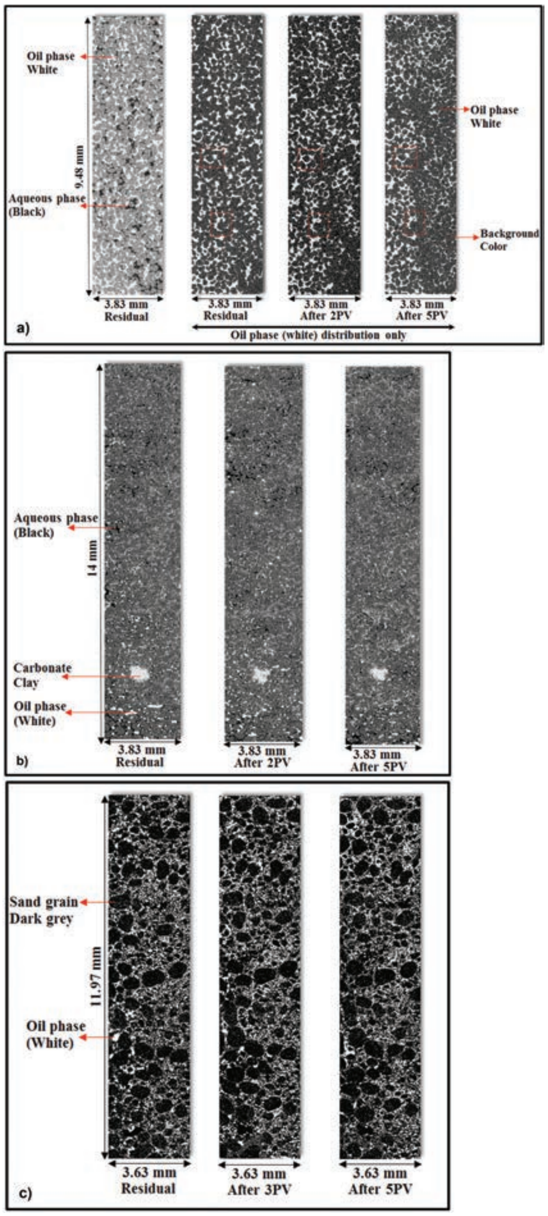

Fig. 1. SXM images of vertical sections of columns along the X-Z direction showing the distribution/saturation of heavy crude oil pre- (initial residual) and post-surfactant flushing: a) homogeneous (H-sand) porous medium (40/50 Accusand, $C_U = 1$) showing trapping of a continuous oil phase. Note, that the first image shows both the aqueous and NAPL (oil) phases for the residual saturation condition and second image shows only NAPL (oil) phase for residual saturation condition. The equilibrium pH over the duration of surfactant flushing ranged between 3.6 and 3.9. b) mildly heterogeneous (MH-sand) porous medium (Accusand/carbonate-fine mix, $C_U = 5.8$) showing discrete fragmentation of oil blobs. The equilibrium pH over the duration of surfactant flushing was approximately 8.13; c) highly heterogeneous (HH-sand) porous medium (Accusand/carbonate-fine mix, $C_U = 10.6$) showing discrete fragmentation of oil blobs. The equilibrium pH over the duration of surfactant flushing was approximately 8.31. The aqueous and oil (NAPL) phases were separated as binary images and then overlain as gray scale images. From J. Ghosh et al., J. Contam. Hydro. **223**, 103471 (2019).

blob fragmentation resulted after progressive flushing events. In this particular experiment, the flushing events acted first on large blobs, having a greater control on blob fragmentation into smaller blobs and enhanced interfacial reduction effect, leading to the greatest relative oil recovery (percentage of oil removed) for all the flushing experiments.

The team concluded that the heavy crude oil removal efficiency observed in the highly heterogeneous sand-packed column may have occurred due to the higher pH value of the system (a greater percentage of carbonate sand fraction that may have contributed to greater reduction of interfacial effects, i.e., interfacial tension, and more effective fragmentation and relative removal of oil blobs from the system. Conversely, the high permeability of the homogeneous column allowed the oil droplets to stay in contact with one another, thus resisting wetting that may exist under a more oil-wet condition (whereby surface cohesion with solid grains would be greater) and a larger connected oil ganglion that would require exceedingly (unrealistic) high viscous forces to mobilize as a single unit through the porous system, leading to negligibly relative oil removal during progressive surfactant flushing events.

The team was able to evaluate the relative importance of characteristics of the sand-oil system. Their effort demonstrated that an imaging procedure and ability for fluid-fluid quantification, using novel synchrotron x-ray microtomography, can be used to assess how amenable a particular physical environment or condition (such as porosity, permeability, porous media type, pH value, and contaminant type/properties, etc.) would be to surfactant-enhanced aquifer remediation. – Mary Alexandra Agner

See: Jaydeep Ghosh[1,2], Geoffrey R. Tick[1*], Nihat Hakan Akyol[3], and Yong Zhang[1], "A pore-scale investigation of heavy crude oil trapping and removal during surfactant-enhanced remediation," J. Contam. Hydro. **223**, 103471 (2019).
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