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Potential and Challenges of Foam-Assisted CO_2 Sequestration

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Abstract

Foam is a promising means to assist in the permanent, safe subsurface sequestration of CO_2, whether in aquifers or as part of an enhanced-oil-recovery (EOR) process. Here we review the advantages demonstrated for foam that would assist CO_2 sequestration, in particular sweep efficiency and residual trapping, and the challenges yet to be overcome.

CO_2 is trapped in porous geological layers by an impermeable overburden layer and residual trapping, dissolution into resident brine, and conversion to minerals in the pore space. Over-filling of geological traps and gravity segregation of injected CO_2 can lead to excessive stress and cracking of the overburden. Maximizing storage while minimizing overburden stress in the near term depends on residual trapping in the swept zone. Therefore, we review the research and field-trial literature on CO_2 foam sweep efficiency and capillary gas trapping in foam. We also review issues involved in surfactant selection for CO_2 foam applications.

Foam increases both sweep efficiency and residual gas saturation in the region swept. Both properties reduce gravity segregation of CO_2. Among gases injected in EOR, CO_2 has advantages of easier foam generation, better injectivity, and better prospects for long-distance foam propagation at low pressure gradient. In CO_2 injection into aquifers, there is not the issue of destabilization of foam by contact with oil, as in EOR. In all reservoirs, surfactant-alternating-gas foam injection maximizes sweep efficiency while reducing injection pressure compared to direct foam injection. In heterogeneous formations, foam helps equalize injection over various layers. In addition, spontaneous foam generation at layer boundaries reduces gravity segregation of CO_2.

Challenges to foam-assisted CO_2 sequestration include the following: 1) verifying the advantages indicated by laboratory research at the field scale 2) optimizing surfactant performance, while further reducing cost and adsorption if possible 3) long-term chemical stability of surfactant, and dilution of surfactant in the foam bank by flow of water. Residual gas must reside in place for decades, even if surfactant degrades or is diluted. 4) verifying whether foam can block upward flow of CO_2 through overburden, either through pore pathways or microfractures. 5) optimizing injectivity and sweep efficiency in the field-design strategy.

We review foam field trials for EOR and the state of the art from laboratory and modeling research on CO_2 foam properties to present the prospects and challenges for foam-assisted CO_2 sequestration.

Introduction

Secure underground storage of anthropogenic carbon dioxide (CO_2) is an essential component in a strategy to reduce atmospheric CO_2 (IPCC, 2018; Michael et al., 2010) and avoiding catastrophic global warming. Such storage could be in aquifers, oil reservoirs, gas reservoirs, or karsts and salt domes. In this paper we focus on aquifers and hydrocarbon reservoirs.

As CO_2 enters the formation, it sweeps radially outward and segregates rapidly to the top of the formation because of the density differences between CO_2 and resident liquid(s). CO_2 is trapped in the formation by various mechanisms (Zhao et al., 2014; Krevor et al., 2015; Gershenzon et al., 2017; Dejam and Hassanizadeh, 2018). In the region CO_2 sweeps before segregation, it is trapped as a residual phase and dissolved into resident brine and oil. CO_2 in the override zone is trapped beneath the impermeable overburden. Beyond the region influenced by the viscous pressure gradient of the injection well (i.e., the swept zone), CO_2 will migrate up-structure under strata that are capillary barriers to gas entry. This migration path is referred to as the "override zone". This migration may be limited by hydrogeological traps (structural, stratagraphic, or fault traps) (Bjorlykke, 1989). The hydrogeological traps may be breached by overfilling past the spill point or the pressure exceeding the fracture initiation pressure, fault leakage pressure, or capillary entry pressure. (Gunter et al. 2004).

Over a period of many years, CO_2 in the override zone may be dissolved into the brine beneath it, helped by convection caused by the greater density of CO_2-saturated brine (Farajzadeh et al., 2009a, 2011; Khosrokhavar et al., 2014; Lyu et al., 2021). Also, over period of many years, some CO_2 reacts with components in the brine and formation minerals and is converted to mineral.

It is crucial to maximize the sweep of CO_2 before segregation, in order to reduce the size of the override zone in the first decades after sequestration. Mineral trapping and slow loss of CO_2 from the override zone into brine beneath it may come too late to prevent stress and fracturing of the overburden in the initial years. Expressed differently, increasing sweep maximizes the amount of CO_2 that can be safely stored in a given formation. Increased sweep in CO_2 injection into oil reservoirs for enhanced oil (gas) recovery of course both increases CO_2 storage and oil (or gas) production.

Foam can greatly increase gas sweep of the formation (Kovscek and Radke, 1994; Rossen, 1996; Enick et al., 2012; Huagen et al., 2014). Foam in a geological formation consists of bubbles at least as large as pores, separated by thin aqueous films, stabilized by surfactant and sometimes nanoparticles (Eftekhari et al., 2015). Foam not only increases the reservoir volume swept by gas; it also increases the residual gas saturation in the region swept. Both effects increase the volume of gas retained in the swept zone and reduce the gas entering the override zone (Vitoonkijvanich et al., 2015; Foren et al., 2020). Combination of foam with surfactants with ultra-low interfacial tension has been recently suggested as one of the EOR methods for carbonate reservoirs with low permeability, where injection of long-chain polymer molecules becomes problematic (Das et al., 2020; Farajzadeh et al., 2013, Li et al., 2010). So far the majority of the published experimental data on this process (referred to as surfactant foam or low-tension gas flooding) have utilized nitrogen or methane as the gas (Das et al., 2020). However, with designing a CO_2-compatible formulation, this process can be adapted to produce oil and store CO_2 simultaneously, especially in carbonate reservoirs.

Due to its relatively greater density and viscosity, CO_2 can be injected to enhance gas recovery in depleted gas reservoirs. This process also suffers from early breakthrough of injected CO_2 and excess recycling of CO_2 in addition to mixing of gases that degrades the value of produced methane (Oldenburg et al., 2001). Foam can be used to reduce the mobility of the injected CO_2 and to block the high-permeability layers. In the context of enhanced gas recovery, foam is also likely to reduce the mixing between CO_2 and natural gas (Zecca, 2018).

In this paper we review the potential advantages of foam for CO_2 sequestration and remaining challenges to realize this potential.

Brief Review of CO_2 Foam Trials

The oil industry has used foams to modify the in-depth conformance of the injected gas and to potentially reduce the gas mobility deep inside the reservoir to improve sweep efficiency. The first foam pilots were designed to block the high-permeability thief zones near the injector and divert fluids to otherwise unswept regions. Foam injection improved the water-alternating-gas (WAG) process in the Rangely Weber Sand Unit in Colorado (Jonas et al., 1990). Foam was generated by simultaneous injection of CO_2 and surfactant solution after emplacement of an initial surfactant slug. CO_2 production in one of the producers declined significantly while oil production improved to some extent. Additionally, the injectivity remained at lower values for two months after implementation of a chase CO_2 flood (Jonas et al., 1990). In the North Ward-Estes field in Texas, four surfactant-alternating-gas (SAG) cycles were injected within two years to treat a near-wellbore area (Chou et al., 1992). Foam was applied through a fractured injection well to alleviate the fast CO_2 breakthrough towards the production well. Foam generation was reported based on a 40-85% decline in injectivity, accompanied by a significant Gas-Oil-Ratio (GOR) reduction in the "problem" producer (Chou et al., 1992). In Wasson field in Texas, foam was utilized to reduce the CO_2 cycling rate (Henry et al., 1996). CO_2 injection and production rates in the target wells declined after the foam treatment, but post-treatment profiles did not show any improvement in the gas sweep efficiency. The poor design of the surfactant pretreatment and the limited injected foam volume were reported to explain the poor sweep efficient at the end of the pilot (Henry et al., 1996).

Supercritical CO_2 foam was injected into a tight sandstone reservoir in the Rock Creek field in Virginia by co-injecting surfactant solution and gas (Heller et al., 1985). Despite some incremental oil detection at an observation well, the foam-flood process had to be halted due to injectivity issues. Improving immiscible CO_2 sweep efficiency by foam was investigated in the tar zone of Fault Block V in the Wilmington field (Holm et al., 1988). Foam was generated after injecting one surfactant slug, followed by a CO_2 slug. The generated foam resulted in fluid diversion and reduction in the gas production rates; however, a subsequent waterflood diluted the foam and led to its collapse. The necessity of adding surfactant to the chase-water slugs was emphasized in order to keep the pre-generated foam stable (Holm et al., 1988). The results of four different CO_2 foam pilots in San Andres (West Texas) and platform carbonate (Southeast Utah) reservoirs showed that while foam reduced the injectivity in all the four studied cases, the decline in gas-production rate was observed in only two of the pilots. It was asserted that surfactant-gas co-injection resulted in lower injectivities compared to the SAG approach and it was operationally more challenging (Hoefner et al., 1995).

The benefit of foam mobility control in gas injection in a dipping formation is illustrated by the foam-assisted water-alternating-gas (FAWAG) application in the Snorre field (Blaker, et al., 2002). Since transportation was not available for the produced gas, gas injection was alternated with water injection (WAG). In some cases, gas breakthrough in up-dip wells was observed after less than one month of gas injection, even though the well spacing was greater than one km. Alpha olefin sulfonate with a 14-to-16-atom carbon chain (AOS 14-16), at concentrations of 0.4% - 0.7%, was used to generate foam for this sandstone reservoir. The surfactant solution could either be co-injected with gas or alternated with gas (SAG). Co-injection resulted in the fracture-initiation pressure being exceeded. Thus, the remaining injection was in SAG mode. The benefit of foam mobility control was apparent from the amount of injected gas produced in the updip production well comparing WAG and SAG injection. After multiple WAG cycles, 65% to 100% of the injected gas was produced in the updip producer. The back-production of gas with

SAG injection averaged 33%. This clearly demonstrated the benefit of foam mobility control in storing the injected gas.

The increasing attention toward CO_2 sequestration, in parallel to oil recovery, and the lessons learned from the past pilots have further motivated energy companies to apply CO_2 foam technology in the last ten years. The SACROC field in West Texas was the site of a foam test with supercritical CO_2 in 2012 (Sanders, et al., 2012). The pilot objective was to improve both the conformance and mobility of the ongoing WAG process at the time by utilizing a CO_2-soluble surfactant, called ELEVATE. The initial stage was applied on a 4-spot pattern with a single injector. After completing the first stage, a injectivity reduction of about 50% and a 30% improvement in the oil-production rate from an offset-well was reported. However, scaling up the pilot from one pattern to a larger 4-injector pattern was unsuccessful due to limited understanding of the reservoir and sub-optimum injection strategy (Sanders, et al., 2012). Most recently, the same CO_2-soluble surfactant (ELEVATE) was tested in a supercritical-CO_2 foam pilot in West Texas (Mirzaei et al., 2020). The selected pilot covered four adjacent patterns suffering from conformance issues during the WAG process. Despite the conformance improvements and injectivity reduction near two of the injectors, the expected increase in oil recovery was not met. This was attributed to the interplay between the increase in volumetric sweep efficiency and the decrease in the throughput rate. While the reduction in throughput rate dominated in the early stages of the pilot, some improvements in the volumetric sweep efficiency increased the oil-production rate slightly at the later stages. The importance of reservoir simulation and the optimization between the effects of volumetric sweep efficiency vs throughput rate were emphasized for achieving successful CO_2 foam pilots in the future (Mirzaei et al., 2020).

Several recent pilots have focused on a single pattern, rather than a multiple-pattern area. An example is a supercritical CO_2 foam pilot in a sandstone formation in Salt Creek Filed in Wyoming (Mukherjee et al., 2014; Norris et al., 2014; Mukherjee et al., 2016; Patil et al., 2018). The pilot design was an inverted 5-spot pattern with a central single injector and 4 surrounding producers, which suffered from high GOR and CO_2 processing rate. The objective was to improve the overall CO_2 sweep efficiency compared to the baseline WAG process. The pilot started with four SAG cycles of short length, and then proceeded to 25 cycles in total. The cycle length was gradually increased by extending the CO_2-injection period while keeping the water/surfactant-injection period constant. The injection well was operated at a constant well-head pressure, and fluid injection rate was monitored with time. An initial 40% reduction in injectivity was attributed to foam generation and propagation in the reservoir. The increase in CO_2 injection rate took place less than 24 hours prior to foam injection but the period of increased injection rate increased to two weeks after several SAG cycles. Gas-tracer tests supported the finding that foam was generated deep in the reservoir (Norris et al., 2014). Lower tracer concentration, delayed tracer breakthrough, and less oscillation in tracer concentration were observed after foam injection, compared to the prior WAG process. A considerable volume of incremental cumulative oil production due to foam injection was reported at the end of the pilot (Mukherjee et al., 2016).

Foam was also piloted in the East Vacuum Grayburg San Andres Unit (EVGSAU), which showed poor vertical communication between its upper zones and the more-permeable lower zones with out-of-zone (OOZ) injection of the fluids. Supercritical CO_2 foam was generated around a central injector, surrounded by 8 producers that suffered from early gas breakthrough, low sweep efficiency, and low run time of production wells due to excessive gas-production rates. Injection included more than 15 alternating cycles of water, CO_2 plus surfactant, and CO_2-only slugs in about two years. The injection-profile logs showed an improvement in vertical conformance upon foam generation. In particular, the Upper/Lower gas distribution ratio was changed from ~30/70 (prior to foam injection) to 47/53 at the end of the 11th cycle. In accordance with this conformance control, oil production was increased by 20% and the gas utilization ratio was improved by 16% with respect to the baseline. Furthermore, the OOZ was eliminated and the run time of the high GOR producers was increased (Katiyar et al., 2020).

A conventional water-soluble surfactant slug was used in the most recent pilot in the East Seminole field of West Texas. Foam flooding was via SAG injection strategy in an inverted 5-spot pattern. The objective was to reduce CO_2 mobility and thereby increase CO_2 storage potential and oil recovery. Preliminary observations indicated about 70% reduction in CO_2 injectivity, compared to the baseline value. However, the reported results at the time were not conclusive, according to the report, because the reduction in CO_2 relative permeability (due to the presence of water) was not distinguished from that of from foam generation (Alcorn et al., 2020). Nonetheless, CO_2 mobility was reduced during CO_2 injection after each surfactant flood.

Potential of CO_2 Foam for Sequestration

In the field, foam is usually placed in the reservoir through alternate injection of aqueous surfactant solution and gas (SAG) rather than direct co-injection of gas and surfactant solution. Reasons include operational safety, reduced corrosion, and improved injectivity. Imbibition and drainage cycling around the well promotes foam generation (Chen et al., 1990; Blaker et al., 2002).

In heterogeneous formations, foam helps even-out the injection profile between high- and low-permeability layers (Al Ayesh et al., 2017). Foam is more resistant to flow in higher-permeability layers (Vassenden and Holt, 2000; Bertin and Kovscek, 2003; Farajzadeh et al., 2015; Kapetas et al., 2017). In both homogeneous and heterogeneous formations, foam reduces gravity segregation and increases the formation volume swept by gas before segregation. Here we review the mechanisms by which foam increases the volume swept before segregation and increases capillary gas trapping within the swept zone.

Improved sweep in a combined EOR or EGR and sequestration process increases oil or gas recovery, increasing the economic incentive for the process, and also increases the extent of CO_2 storage at the end of the process. It also reduces the carbon footprint of oil produced by CO_2 EOR (Farajzadeh et al., 2020).

Injection Profile

As noted, foam helps even-out the injection profile (Al Ayesh et al., 2017; Kapetas, 2017). In the context of gravity segregation, in a formation with higher-permeability layers near the top, foam can divert flow into lower-permeability layers lower in the formation. Gas injected into those layers would travel further before segregating upward, and thereby sweep more reservoir volume and leave more trapped CO_2 below the override zone. Periodic near-well treatments might suffice to alter the profile, without the need for foam to sweep a large radial area, which can entail issues described in the next section.

Gravity Segregation

Homogeneous Formations. The equation of Stone (1982) is a useful tool for understanding ways to reduce gravity segregation (Rossen and van Duijn, 2004). The model makes simplifying assumptions: Newtonian mobilities, incompressible flow, horizontal homogeneous reservoir. It also assumes co-injection of gas and liquid. Stone argues that, if WAG slug sizes are not too large, then away from the injection well conditions approximate co-injection. The mixed zone is at the uniform saturation corresponding to the injected foam quality (gas fractional flow); the override zone is at irreducible water and the underride zone has no gas. Rossen and van Duijn (2004) verified Stone's equation and showed it applies to foam with the same simplifying assumptions; see also Shi and Rossen (1998).

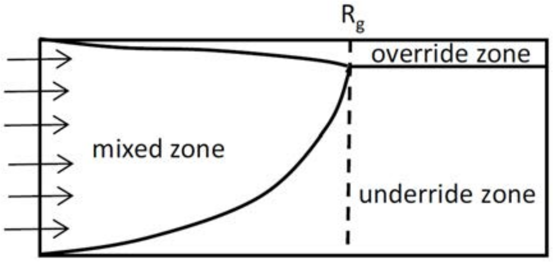

Figure 1—Schematic of gravity-segregation model of Stone (1982)

At fixed injection rate the model predicts that the position of complete segregation of gas and water occurs at a radius R_g given by

$$R_g = \sqrt{\frac{Q}{k_z(\rho_w - \rho_g)g\lambda_{rt}^m}} \tag{1}$$

with

$$\lambda_{rt^m} = [(k_{rg}/\mu_g) + (k_{rw}/\mu_w)] \tag{2}$$

where Q is total volumetric injection rate at field conditions, k_z vertical permeability, ρ_w and ρ_g the densities of the aqueous and gas phases, g the gravitational constant, λ_{rt}^m the total relative mobility in the mixed (swept) zone around the well, before gravity segregation, k_{rw} and k_{rg} the relative permeabilities of water and gas corresponding to the injected foam quality, and μ_w and μ_g the viscosities of water and gas in foam at that quality. Equation (1) applies to foam as long as it approximates incompressible flow and Newtonian rheology. Equation (1) indicates two ways to increase radius of the zone swept by gas before segregation: increase injection rate Q, or reduce mobility in the swept zone λ_{rt}^m. Unfortunately, both strategies increase injection well pressure, which is often limited to avoid fracturing. With some additional assumptions (Rossen et al., 2010), one can show that at fixed injection pressure $p(R_w)$, R_g is given by

$$p(R_w) - p(R_g) = \frac{k_z(\rho_w - \rho_g)g}{2Hk_h} R_g^2[\ln\left(\frac{R_g}{R_w}\right) - \frac{1}{2}\left(1 - \left(\frac{R_w}{R_g}\right)^2\right)] \tag{3}$$

where k_h is horizontal permeability. Reducing the mobility of gas and water in the mixed zone does not help, as long as mobility is uniform in the mixed zone, as is assumed in the model.

Foam mobility is not uniform in the mixed zone, in at least two ways. First, foam is usually shear-thinning, especially at relatively low quality (Osterloh and Jante, 1992; Alvarez et al., 2001). This means that injection rate can be greater than assumed in Eqs. (1) and (3), without raising injection pressure. Jamshidnezhad et al. (2010) show that a power-law exponent of 0.4 (similar to that observed for foam at low quality) can result in an increase in R_g by about 60 to 90%: the swept volume increases by a factor of between 2.5 and 4.9.

Second, SAG injection gives much better injectivity than the co-injection assumed in Eq. (2), because of much greater mobility near the well, especially during gas injection, as foam dries out and collapses there.

A very-idealized model of Shan and Rossen (2004), in which one large slug of liquid is followed by constant-pressure injection of gas, indicates that gas could travel long distances before gravity segregation in a homogeneous reservoir. Grassia et al. (2014) show that in principle the distance is unlimited, but of course there would be practical difficulties in such a process, including injection of sufficient surfactant solution. This model also takes no account of possible limits to propagation at low ∇p as discussed below.

Gong et al. (2020a, b, c) investigated phenomena expected near the injection well that would greatly affect injectivity of both gas and liquid. In those experiments, during injection of the gas slug, a collapsed-foam zone propagated at a dimensionless velocity of (1/400). If the injected gas slug represents a pore volume out to a radius of 100 m, this implies collapsed foam to a distance of 5 m. For a well radius of 10 cm, half the injection pressure between the well and 100 m would be expected to be dissipated within about the first 3 m of the well. This collapsed-foam region greatly increases the injectivity of gas.

In those experiments, the next surfactant slug filled the collapsed-foam region with good mobility, though with a significant gas saturation. In the region beyond, most gas in the foam remained trapped, giving very low liquid mobility. However, unsaturated liquid formed fingers through the trapped gas and then dissolved the gas within those fingers, giving relatively high mobility within that fraction of pore space. As a result, mobility again increases greatly in a region moving outward from the well. Gas around the fingers remained trapped in place. Nguyen et al. (2009) first observed this fingering behavior and showed that it applies whether the next slug is surfactant solution or surfactant-free brine. The original experiments were performed with N_2 gas. With CO_2, much more soluble than N_2 in water, the high-mobility front advanced about 10 times faster in Gong's coreflood (Gong and Rossen, 2022).

Heterogeneous Formations. Foam offers an additional advantage in suppressing gravity override in heterogeneous formations. In flow across a sharp boundary from a water-wet low-permeability to a high-permeability layer, gas flow is blocked by an effect like the capillary end effect in laboratory corefloods: water accumulates at the boundary (Falls et al.,1988; Tanzil et al., 2002; Shah et al., 2019). If surfactant is present in the water, foam generation results as gas flows through this barrier. The foam may propagate from the barrier, remain in place, or show cycles of generation, dry-out, collapse and regeneration. In any event, upward migration of gas is reduced. In effect, the vertical permeability k_z in Eqs. (1) and (3) is reduced, increasing the radius of the swept zone attainable at a given injection pressure.

Gas Trapping by Foam

Within the swept region, CO_2 bubbles are trapped and retained by foam (Falls et al., 1989; Kovscek and Radke, 1994; Bertin and Kovscek, 2003; Nguyen et al., 2009b; Kil et al., 2011). A fraction is also dissolved in resident water and, over time, may react with formation salts and minerals and be retained in mineral form. The residual saturation of CO_2 in foam is much greater than in gas-liquid flow without foam. Gong et al. (2020a,c) found that even in the region of "collapsed foam" created during gas injection very near the well, much gas was retained upon subsequent imbibition of surfactant solution or water: a gas saturation greater than 50% in their corefloods. In the portion of the core corresponding to formation beyond the immediate vicinity of the wellbore, where foam would not have collapsed, residual saturation was greater than 70%. Thus foam both increases sweep efficiency and the saturation of gas trapped by capillary forces within that region.

Foam for Reducing CO_2 Footprint of Operations

It has been shown that the amount of the emitted CO_2 per barrel of oil depends on the (net and gross) mass-CO_2 utilization factor, i.e., the volume of oil produced per mass of CO_2 injected, usually expressed in bbl/tCO_2. Figure 2 shows the total CO_2 intensity of net oil (production and burning of oil in later applications) for EOR with CO_2 captured from oil-fueled power plants. It has been assumed that oxidation of oil leads to emission of 422 kg-CO_2/bbl. The details of the calculation can be found in Farajzadeh et al. (2020). The net mass-CO_2 utilization factor of the published field data on CO_2 EOR are in the range of 1.8–4.2 bbl oil/tCO_2, with an average value of ~2 bbl oil/tCO_2. In fact, a common drawback in most of the field applications of CO_2 EOR is poor CO_2 utilization factor. This results in circulation of excess amounts of gas in the producers and hence increases the CO_2 emissions resulting from the operations. Foam can be used to improve the utilization factor of gas in CO_2 EOR and EGR processes. For example, improving the utilization factor from 2 bbl/tCO_2 to 4 bbl/tCO_2 can reduce the CO_2 intensity of the produced oil from 120 kgCO_2/bbl to 80 kgCO_2/bbl (33% reduction). In some cases, application of foam can also extend the lifetime of high-GOR fields, if CO_2 intensity cut-offs are applied to oil production (Farajzadeh et al., 2022)

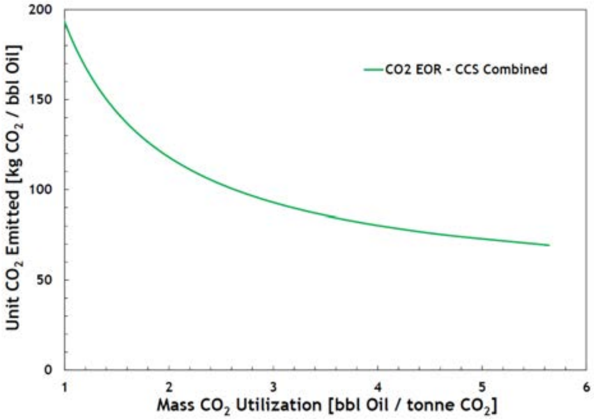

Figure 2—CO_2 intensity of oil production (including its burning in later application e.g. in a power plant) for a CO_2 EOR process combined with CCS, as a function of mass CO_2 utilization factor (reproduced from Farajzadeh et al. 2020).

For a CCS-only process, the major CO_2-intensive (or energy intensive) components include capture, compression, and transportation of CO_2:

$$\dot{e}_{CCS} = \dot{e}_{capture} + \dot{e}_{comp.} + \dot{e}_{transp.} \tag{4}$$

where $\dot{e}_{CCS}$ is the CO_2 intensity (kg-CO_2/kg-CO_2 stored) of the process and its magnitude depends on the source of the energy (mainly electricity) and the efficiency of the applied technology. For simplicity, this equation excludes other possible CO_2-leakage sources. With the values reported in Farajzadeh et al., ~5.5 MJ (1.5 kWh) energy is required to store 1 kg of CO_2. For CCS to result in low CO_2 emission from the operation, the source of this electricity should be low-carbon. For foam application the CO_2 emissions from surfactant manufacturing and shipping should also be considered in Eq. (4), which has been estimated to be 2.5-3.5 kg-CO_2/kg-surfactant (Farajzadeh et al., 2021). The storage efficiency of an aquifer (CO_2 volume in the reservoir) is often very small, in the range of 1-6% (van der Meer, 1995). This means that storage of significant volumes of CO_2 will require multiple aquifers, which increase the overall operational footprint and consequently CO_2 emissions. In this case, application of foam can create huge storage volumes by efficient displacement of the brine. This will lead to operations in smaller number of storage sites. In other words, the overall CO_2 emission of stored CO_2 can be considerably reduced by foam.

Foam-aided Cold CO_2 Injection

CO_2 storage in depleted gas reservoirs is a potential near-term solution to reduce CO_2 concentrations in the atmosphere, even though compared to aquifers they provide smaller storage volumes. The risks and uncertainties associated with depleted fields are lower due to their extended production history, proved sealing capacity, and availability of infrastructure required to store the gas. However, expansion of CO_2 from pipeline pressure (around 100bar) to low pressures of the depleted reservoirs under (quasi) adiabatic conditions can result in temperature drop (so-called Joule-Thomson effect), that can potentially cause injectivity issues because of formation of hydrates and ice (Oldenburg, 2007). To increase the mass rate of stored CO_2, injection of CO_2 in dense form is preferred. This could be achieved by heating CO_2 before injection, which is very costly and energy (carbon) intensive. Foam can potentially help overcome this issue by creating a pressure gradient around the wellbore such the pressure drop is not abrupt. Using foam, the pressure at the sandface increases and depending on the strength and propagation of foam the pressure in the foamed zone will be above the reservoir pressure. Therefore, we expect foam to prevent rapid expansion of CO_2 and distribute the expansion over a greater distance. This permits more heat conduction with the reservoir and consequently severe temperature drops will not be experienced. Injection of foam can reduce the injectivity, therefore, the trade-off between injectivity and this advantage as well as the duration of this effect should be investigated further.

Blocking CO_2 Leakage Through Overburden or Well Casing

Some studies suggest that foam can reduce leakage from CO_2 storage reservoirs (Castaneda-Herrera et al., 2018; Zhu et al., 2021)

Surfactant Selection for Foam Applications

Foam is a dispersion of a gas in a continuous liquid phase. A foaming agent is needed to stabilize the thin films of liquid separating the gas bubbles to mitigate or avoid their coalescence. Foaming agents are generally surfactants (surface active agents) but nanoparticles can also be foaming agents or act as a surfactant enhancement. Surfactants are amphiphilic molecules. The hydrophilic (water-loving) part prefers to reside in the water phase while the hydrophobic (water-hating) part, or hydrophobe, prefers to reside in the non-aqueous phase. The hydrophilic property may result from an electrically charged group (i.e., ionic), a dipole (e.g., zwitterion), and/or from the tendency to hydrogen bond with water (i.e., non-ionic). The hydrophobic property of the molecule is the result of the hydrocarbon part. The tendency of surfactants to adsorb at an interface has beneficial as well as detrimental effects. Adsorption of surfactants at a fluid-fluid interface reduces the surface tension (gas/liquid interface) or interfacial tension (liquid/liquid interface). Adsorption of surfactant at the interface also contributes to the shear-and-dilatational surface viscosity of the interface. Most crucially for foam, adsorption of surfactant on both surfaces of a foam film contributes to stabilizing the thinning and rupture of the foam film. The force resisting (and aiding) the thinning of a foam film is called the "disjoining pressure". Surfactant adsorbed on the interfaces contributes to these forces by electrostatic repulsion, dipole-dipole interaction, and steric forces (e.g., the space–occupying effect of an ethoxylate chain). Surfactant micelles and nanoparticles contribute to the disjoining pressure by the ordering of charged particles in the foam film (Wasan and Nikolov, 2007). Surfactants also adsorb at liquid/solid interfaces, resulting in adsorption losses or retention on the rock.

Types of surfactants

The types of surfactants considered here are categorized as: (1) anionic, (2) nonionic, (3) ethoxylated anionic, (4) cationic, (5) zwitterionic, and (6) CO_2-soluble. The choice of surfactant is dependent on the formation material (e.g., sandstone or carbonate), temperature, brine salinity and hardness, and mode of injection of surfactant (Enick, et al., 2012).

1. Anionic: Many household surfactants are anionic. Soap is a carboxylate. Carboxylates are salts of a weak acid, and its degree of dissociation is a function of pH. Carboxylates are problematic because they are prone to precipitate in the presence of divalent cations (e.g., calcium and magnesium). Synthetic detergents often have a sulfate or sulfonate ionic group. These surfactants are anionic or negatively charged. The most common anionic surfactant tested in EOR applications with foam is alpha olefin sulfonate with 14 to 16 carbons (AOS 14-16) (Svorstol, et al., 1995; Svorstol, et al., 1996; Blaker, et al., 2002; Farajzadeh, et al., 2008).
2. Nonionic: Nonionic surfactants may have ethylene oxide ($–CH_2-CH_2O-$) (EO), or sugar-based hydrophilic groups. These groups interact with water via hydrogen bonding (Akins, et al., 2010; Alcorn, et al., 2020; Chen, et al., 2015; Jian, et al., 2016; Jian, et al., 2020; Xing, et al., 2012).
3. Ethoxylated anionic: These surfactants have several EO groups terminating in an anionic group. The EO groups extend the distance that the charged group can reside in the aqueous phase. The resulting reduced charge density makes these surfactants much more tolerant to high salinity and hardness from divalent cations. The ethoxylated sulfates are subject to hydrolysis at low pH and elevated temperatures. The ethoxylated sulfonates are stable but expensive.

4. Cationic: Quaternary ammonium surfactants are common but expensive. Amine, substituted amine, or diamine surfactants are more economical compared to the quaternary ammonium surfactants. They become more strongly cationic with reduced pH as in water with dissolved CO_2. These surfactants are sometime called switchable surfactants (Elhag et al., 2014; Jian, et al., 2019).
5. Zwitterionic: Zwitterionic surfactants, such as betaines, have both an anionic and cationic group, thus they have a dipole. This tends to make them less sensitive to salinity. Sometimes they are added as a "foam booster" (Li, et al., 2012; Da et al., 2018).
6. CO_2-soluble: Short-chain length, branched, nonionic surfactants are potentially soluble in high-pressure CO_2. These surfactants can be dissolved in the injected CO_2 (Akins, et al., 2010; McLendon, et al., 2012; Xing, et al., 2012; Ren et al., 2018).

Surface Activity

There are a number of measures of surface activity, but the focus here is the ability to stabilize foam films against rupture due to film thinning. For the foaming agents soluble only in the aqueous phase, the carbon number of the hydrophobe is the most important parameter for providing surface activity. A carbon number of 12 (e.g., SDS) is the minimum in most foam applications. Increasing the carbon number increases the surface activity, but if it is equal to or greater than 18 (and linear) an ionic surfactant may precipitate (rather than form micelles), unless the temperature is above its Krafft temperature. Even if an ionic surfactant is above the Krafft temperature, a surfactant-rich "liquid crystal" phase may form unless the temperature is high enough (Abe, et al., 1986). The liquid-crystal phase may be beneficial in stabilizing foam and emulsions, but it can lead to high surfactant retention in flow through porous media. For aliphatic hydrophobes in anionic surfactants, a linear carbon chain (compared to a branched hydrophobe of same molecular weight) increases foam stability as well as the tendency to form liquid crystals. Branched aliphatic hydrophobes in anionic surfactants are less prone to formation of liquid crystals and viscous emulsions, but form weaker foam for the same carbon number (Abe, et al. 1986; Carty, et al., 2004). An example of this is the weak foaming behavior of a blend of internal olefin sulfonate and a branched alcohol-ethoxylate sulfate. In such cases, it may be necessary to add a zwitterionic surfactant as a "foam booster" to have adequate foam strength (Li, et al., 2012).

In the case of nonionic surfactants, increasing the carbon number increases the surface activity, but the EO number should also be adequately increased. Nevertheless, the "cloud point" may be reached, resulting in phase separation, high surfactant retention in flow through porous media, and poor foaming. The ethoxylated nonionic surfactants are a class of surfactants where hydrophobe types, linear, branched or methylated, have been studied for supercritical CO_2-foams (Adkins, et al., 2010; Xing, et al., 2012; Chen, et al., 2015; Alcorn, et al., 2020; Jian, et al., 2020).

A common measure of surface activity is the reduction in surface tension due to adsorption of surfactant at the gas/liquid interface. The degree of reduction in the surface tension is a function of the surface concentration of the surfactant at the gas/liquid interface. If a foam film is suddenly stretched, the film thins but the surfactant surface concentration also decreases; therefore, the surface tension increases, and the increase in surface tension resists the stretching. This phenomenon is known as "Gibbs' elasticity". A gradient in the surface tension can result in stresses that induce or retard the flow of the interface and the surrounding fluids (Hirasaki and Lawson, 1985). This is known as the "Marangoni effect" (Scriven and Sternling, 1960). These effects can be visualized by light reflected from foam films (Sheludko, 1967; Suja, et al., 2020).

Adsorption of surfactant at the gas/water interface can alter the rheology of the interface by resulting in an interfacial shear and dilatational viscosity. This interfacial viscosity tends to dampen or even inhibit the Marangoni effect. This can be demonstrated by adding a small amount of dodecanol to a foam film made with sodium dodecyl sulfate (SDS). When two bubbles approach each other, a nearly flat, circular foam film forms between the two bubbles. If the interfacial viscosity is large enough, the flow is radial, and the film is thinner next to the edge because of the capillary suction due to the curved interface of the Plateau border (meniscus). This results in slow drainage. If the interfacial viscosity is low enough, the surface-tension gradients produce outward flow in some places and inward flow in other places. This results in rapid film thinning to the equilibrium thickness or to film rupture (Joye, et al., 1994).

Chemical stability

Chemical stability is important for either EOR or CO_2-sequestration processes because the duration of the displacement process may be months and the temperature may be elevated significantly compared to ambient conditions. Trace amount of dissolved oxygen result in free-radical degradation of EO groups in nonionic surfactants or ethoxylated anionic surfactants and unsaturated carbon-carbon bonds. Thus, addition of oxygen scavenger is essential to the formulation. The sulfate group in sulfate surfactants is slowly hydrolyzed at low pH and high temperature (Talley, 1988). Thus, sulfates are unsuitable for long-term CO_2 applications. The amide group in some amido-zwitterionic surfactants will hydrolyze at low or high pH and elevated temperature. Degradation of amide is slower at neutral pH (Cui, L., 2014).

Physical stability

Carboxylate surfactants (soaps) are well known for their tendency to precipitate or form liquid crystal in the presence of divalent (calcium and magnesium) cations and/or large amount of sodium ions. Sulfonate and sulfate surfactants are much less sensitive to divalent cations (compared to sodium ions) than the carboxylate surfactants. Ethoxylated sulfate and sulfonate surfactants can be designed to be tolerant to divalent cations and high salinity. Ethoxylated nonionic surfactants will phase-separate above the cloud point temperature. Amine surfactants are fully ionic at the pH of water saturated with high-pressure CO_2 and is tolerant to high temperatures. (Jian, et al., 2019)

Surfactant Adsorption

Surfactant adsorption is a function of the surfactant structure and formation minerals (Mannhardt et al., 1994; Ma, et al., 2013). Sedimentary formations can be distinguished as sandstone or carbonate. Sandstones are predominately negatively charged and tend to hydrogen-bond. Clays in sandstone have negative charges on their faces but positive charges on the edges. Anionic surfactants, since they are negatively charged, are favored in sandstone formations because of lower adsorption. Carbonate formations tend to be predominately positively charged and have insignificant hydrogen bonding. Thus, nonionic surfactants are preferred for carbonate formations provided the temperature is well below the cloud-point temperature. If a carbonate formation has temperature near or above the cloud-point temperature, a cationic surfactant could be utilized (Jian, et al., 2019). If the carbonate formation has significant silica or clays, the adsorption of cationic surfactants may be excessive (Cui, et al., 2015).

Nanoparticles

Recently there has been much interest in testing nanoparticles to stabilize foam. Nanoparticles can stabilize foam by two mechanisms. (1) The nanoparticle can be amphiphilic, i.e., one part favoring water and another part disfavoring water, like surfactants. This is analogous to Pickering emulsions stabilized by solid particles that adsorb onto the interface between the two phases. (2) The nanoparticles can be completely hydrophilic with an electrically charged and/or polymer-coated surface. In this case the mutual repulsion of the particles results in structural ordering that can contribute to foam stability (Wasan and Nikolov, 2007). A combination of both is possible. A limitation of foam generation with only nanoparticles is that more energy is required to generate the foam. A combination of surfactant and nanoparticles may achieve the favorable attributes of both (Eftekhari et al., 2015; Issakhov, et al., 2021; Nazari, et al., 2020; Singh and Mohanty, 2014; Singh and Mohanty, 2017; Worthen, et al., 2015; Xue, et al., 2016).

Unknowns and Challenges for Foam CO_2 Sequestration

Minimizing Chemical Cost

Chemical costs are a significant component of the economics of applying foam to CO_2 sequestration. The surfactants described above were designed with the economics of an EOR process in mind. Further reductions in chemical cost (including reductions in surfactant adsorption) would reduce the cost of a sequestration process. It is important to note that foam required for brine displacement in aquifers should not be necessarily strong, as the viscosity of the brine is typically less 1 cP. For diversion purposes stronger foam might be required to distribute the injected CO_2 more evenly. In the absence of oil, generation and stability of foam is also easier to maintain in aquifers, which makes the choice of surfactant and injection concentrations less expensive than for EOR.

Possible limits to Long-Distance Foam Propagation

Propagation of foam radially outward to large distances from an injection well is limited by gravity segregation, injection of sufficient surfactant, and possible chemical degradation. Foam in bulk (in a column or a beaker much larger than the bubbles) degrades over time by diffusion, but in porous media this process stops when bubbles reach roughly pore size. There may be an additional fundamental limit to long-distance foam propagation at low superficial velocity u_t and pressure gradient ∇p far from an injection well. There is evidence for this in a steam-foam field test (Friedmann et a., 1994) (questioned by Patzek (1996)), and laboratory studies (Friedmann et al., 1994, Yu et al., 2020) find limits to propagation at low u_t and ∇p. Modeling (Ashoori et al., 2012) indicates that this is a result of a minimum u_t or ∇p for foam (re-) generation at the leading edge of the foam bank. This in turn depends on the ease of mobilizing lamellae and the stability of lamellae once created (Rossen and Gauglitz, 1990). Mobilization depends on gas-liquid surface tension, which can be 10 times or more lower for supercritical CO_2 foam than for N_2 foam at room temperature (Lee et all, 1991; Rossen, 1996). The experimental conditions of the propagation experiments were ideal for strong foam, and stability would be much more problematic at field conditions (temperature, salinity, wettability, presence of oil (in EOR)), which would make propagation more difficult. Cui et al. (2016) found a minimum ∇p for generation of a CO_2 foam in dolomite core of 10 psi/ft (2.26×10^5 Pa/m) at a salinity of 22% total dissolved solids (TDS), using Ethomeen C12 surfactant, with 12-14 carbons in the hydrocarbon chain. Jian et al. (2019) found no minimum ∇p for CO_2 generation in a limestone core at the same high temperature/high salinity using Duomeen TTM surfactant with 16-18 carbons in the chain.

Direct experimental data on propagation at field pressure gradients are few, and most involve N_2 foam. The ∇p for N_2 foam propagation in these experiments was impractical for field implementation. If the limitation depends on the ∇p required for foam generation, then the 10-100x lower ∇p threshold for generation found by Gauglitz et al. (2002) suggests much less of an issue for CO_2 foam. CO_2 foam shows lower apparent viscosity and ∇p than N_2 foam at the same injection rates (Chou, 1991; Zeng, et al., 2016). This is sometimes interpreted as reduced stability of CO_2 foam, but it could also be a reflection of lower surface tension, resulting in less capillary resistance to flow and less capillary gas trapping as foam flows.

There is a need for further studies to determine whether u_t and ∇p limits to propagation identified for N_2 foam apply to CO_2 foam, and under more realistic field conditions.

Even if foam cannot propagate directly from an injection well, there are other ways foam could be created far from an injection well. As noted above, foam can be created as gas migrates upward across a sharp layer boundary from lower to higher permeability, with no requirement on ∇p. This depends on placing surfactant far from the well. Also, with sufficiently large slugs of gas and surfactant solution, repeated imbibition/drainage cycles could help in foam generation (Chen et al., 1990). As noted in the section above on CO_2 foam field trials, foam has reduced CO_2 production substantially in field application.

Wormholing

CO_2 injection into brine-saturated carbonates results in acid formation, carbonate dissolution, and wormhole formation. Foam injection would require injection of additional water with the gas, which could allow greater acid formation and wormholing. Foam is used as a diverter in acid well stimulation. In coreflood experiments, foam led to narrower, deeper wormholes than acid without foam (Bernardiner et al., 1992; Al-Nakhli et al., 2021). These results depend on the competition between convection and acid transport to the wormhole wall quantified in the Damkohler number (Hoefner and Fogler, 1987), and of course reflect a greater dissolution capacity of HCl acid than CO_2-saturated brine.

Further research is needed to quantify the significance of this issue for foam-assisted CO_2 storage in carbonates, and strategies to overcome it.

Long-Term Stability of Foam: Chemical Stability and Dilution of Surfactant

CO_2 sequestration must be permanent. As noted, it can be decades or more before CO_2 dissolution into brine significantly reduces the override zone. Therefore, it is important that CO_2 trapped by capillary forces within the foam-swept zone remain trapped for decades. During that time surfactant may degrade chemically or be displaced out of the swept zone by slow convection in the aquifer.

It is important therefore to verify, if possible, the long-term chemical and biological stability of the surfactant under reservoir conditions. Regarding displacement of the surfactant slug by aquifer water, lab coreflood experiments (Nguyen et al., 2009a; Gong et al., 2020a,c) suggest that water does not displace foam uniformly, but fingers through, leaving most trapped gas (and surfactant) in place. Eventually, large volumes of water dissolve the gas within and surrounding the fingers.

The instability leading to this fingering may depend on superficial velocity, as in other fingering phenomena (Lake et al., 2014). If so, then it is important to verify the extent of capillary gas trapping under conditions of imbibition as surfactant concentration is reduced. Imbibition, by reducing capillary pressure, may help foam to remain stable and keep gas trapped (Stegemeier, 1977). On the other hand, as noted above, Holm et al. (1988) observed foam collapse during waterflood following foam.

It is likely that capillary gas trapping under conditions of imbibition in the foam-swept zone is much greater than without foam. The region of the formation swept by foam generally has a large initial gas saturation. As noted above, Gong et al. (202a,c) found gas saturation of 50% even in the "collapsed foam" zone where mobility was large during imbibition following the collapse. Thus the "foam collapse" noted by Holm, which presumably meant restored liquid mobility, does not mean complete loss of gas trapping. Foam should increase the volume of CO_2 retained in the swept region as residual gas saturation and dissolved in water even if surfactant degrades or is displaced. Determining the residual gas saturation in the swept zone if surfactant does disappear remains an important unknown.

Foam in Fractured Formations

Gravity segregation is much more rapid in fractured formations. If fractures are more than a small fraction of 1 mm wide, capillary forces do not trap foam, and segregation is rapid. A fracture's aperture varies along its length, and fracture walls may touch; these could provide locations to regenerate foam, if surfactant solution is transported to those locations (Fernø et al., 2016). Ren et al. (2018) found foam generation during CO_2 and surfactant-solution injection into a 30-cm core fractured along its axis. Some applications of foam have used polymer gel with surfactant in the aqueous phase to prevent or at least postpone segregation (Hughes et al., 1997). Foam was effective in near-well diversion in fractured field (Ocampo-Florez et al., 2014). Investigations of foam behavior in fractured cores can be found in Ren et al. (2018), Haugen et al. (2014) and Dong et al. (2019). If foam remains stable, it can also divert CO_2 from fractures into the matrix by creating a horizontal pressure drop between the fracture and the matrix (Farajzadeh et al., 2012). If there is no foam in the fracture, the only driving force for the injected CO_2 to enter the matrix will be the gravity force (determined by density difference between CO_2 and water), which is very slow but also insufficient when CO_2 is injected as a dense phase.

Accurate, Predictive Modeling and Simulation

Once the mechanisms and effectiveness of sweep improvement and long-term gas trapping are verified, they should be incorporated into a reservoir simulator for design, prediction and optimization of field application. Such a simulator must account not only for the mobility of foam under reservoir conditions as a function of reservoir properties, but also explicitly account for capillary gas trapping and hysteresis in behavior between imbibition and drainage. It must also account for reduced upward migration of gas across layer boundaries, either explicitly modeling foam generation there, or empirically with a reduced vertical permeability in the presence of surfactant and gas. If CO_2 foam propagation at practical field pressure gradients is in question, then mechanisms of foam generation and propagation must be incorporated into the simulator (Yu, 2021). Similarly, if wormholing, long-term chemical stability of the surfactant formulation, or foam stability in fractured reservoirs is involved in a given field application, the model must accurately represent that phenomenon.

Conclusions

Foam can increase the amount of CO_2 that can be safely stored in a porous subsurface formation by increasing the volume swept before gravity segregation, and thereby increasing trapping within that zone and reducing the stress on the override zone. Foam also reduces CO_2 footprint of a sequestration process. Surfactant formulations have been developed for challenging environments and field applications show promise.

Remaining challenges include the following:

- confirming the effectiveness of foam properties seen in laboratory studies for field application
- verifying stability of foam over decades, given possible dilution and degradation of surfactant
- testing the effectiveness of foam in fractured formations
- minimizing chemical costs
- checking for possible limits to long-distance foam propagation
- determining the effects of foam on wormholing in carbonate formations and effectiveness of foam in wormhole-penetrated formations
- developing accurate and predictive simulation models that can guide process design and include the key mechanisms of foam effectiveness, sweep efficiency and gas trapping.

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