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Surfactants are ineffective for reducing imbibition of water-based fracturing fluids in deep gas reservoirs

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ABSTRACT: Minimizing loss of injected hydraulic fracturing fluids into shale along fracture-matrix boundaries is desired because imbibed water restricts gas production and wastes valuable water resources. This problem has motivated the addition of surfactants into water-based hydraulic fracturing fluids in order to reduce the capillary driving force for imbibition. Here, we show that reduction in interfacial tension and wettability alteration have negligible ability to reduce imbibition in deep gas reservoirs. The effectiveness of altering capillary forces acting at the wetting front also depends on the injection pressure acting at the fracture-matrix boundary. The pressure at the interface between the fracture and the shale matrix is constrained between the reservoir pore pressure and formation pressure (rock fracture pressure, also known as breakdown pressure of the rock) and increases with depth to magnitudes that greatly exceed that of capillary pressures. The analyses presented here show that even maximum alteration of interfacial properties that result in strongly hydrophobic interactions between the fracturing fluid and reservoir rock is incapable of significantly reducing imbibition in deep reservoirs. Instead of using surfactants, this analysis points to decreases in wellbore shut-in pressures and shut-in times as practical options for reducing imbibition losses of water-based fluids.

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1. INTRODUCTION

Hydraulic fracturing for stimulating production of low-permeability shale gas reservoirs uses large volumes of water, often exceeding 10^4 m^3 per well, while competing for valuable water resources and incurring high costs for water supply and treatment of flowback water[1-3]. In addition to these burdens, loss of water-based fracturing fluids into reservoir matrix rocks during shut-in impedes production through blocking pores and through structural damage within the wetted fracture-matrix interface zone [1, 4-6]. The adverse effects of water imbibition into shales have motivated investigations on ways to minimize water loss within reservoirs through manipulating interfacial properties and capillary forces. Indeed, a key strategy advanced in stimulating unconventional gas reservoirs is to use surfactants in hydraulic fracturing fluids in order to reduce the capillary pressure driving imbibition and thereby decrease thicknesses of water-blocked zones at the fracture-matrix interface[7-16]. Indeed, in enhanced oil recovery (EOR), the applications of surfactants to reduce surface tension, stabilize microemulsions, and alter reservoir wettability to facilitate oil mobilization are well recognized[17-22]. It is worth noting that surfactants are also added into hydraulic fracturing fluids for controlling rheological properties. As essential components of foams and gelling agents, surfactants can help enhance viscosity, thereby increase fracture width and improve proppant delivery [23-25].

Instead of revisiting the extensively studied and complex problem of surfactant applications in EOR, the analyses developed here reexamines the importance of changing capillary forces in the context of hydraulic fracturing in deep gas reservoirs. To the best of our knowledge, a critical limitation of all the previous studies on surfactant applications for controlling imbibition in shale gas reservoirs is that influences of high reservoir pressures and temperatures were not considered. For a typical depth of 2.5 km for a horizontal well[26], hydraulic

47 fracturing occurs at pressures and temperatures of about 35 MPa and 80 ˚C, and in the following
48 we show that such conditions severely limit the extent to which changing capillary forces can
49 influence frac fluid imbibition in gas reservoirs.

50 Before considering capillary effects, it is worth briefly noting that electric double layer
51 expansion and osmotic potential gradients can also influence imbibition in clay-rich, saline
52 sediments. Although correlations between clay content, salinity, and imbibition rates have been
53 obtained[27-29], and osmotic potentials associated with saline reservoir pore waters have been
54 determined[30], challenges persist for quantifying their actual impacts under reservoir conditions. A
55 limitation associated with interpreting how clay swelling impacts imbibition is that experiments
56 to date have largely been conducted on unloaded or only uniaxially loaded cores. Lack of total
57 confinement allows multidimensional sample expansion during hydration, producing highly
58 transmissive microfracture networks that artificially enhance imbibition relative to that actually
59 occurring under in-situ reservoir stresses. Indeed, such free swelling leads to artificial
60 microfracturing and even break up of unconfined samples[10, 28, 31]. The importance of applying
61 confining stresses during imbibition tests on shales is underscored by closure of microfractures
62 during hydration experiments conducted under isotropic confinement[32].

63 The extent to which imbibition can be facilitated by osmotic forces also requires better
64 understanding because shales are not ideal semi-permeable membranes. The osmotic pressure
65 gradient driving imbibition is moderated by the osmotic efficiency, σ, which ranges from 0 to 1,
66 and depends inversely on the product of characteristic pore-size times the square-root of salt
67 concentration[33]. The inverse dependence on the square-root of salinity diminishes σ to less than
68 0.01 as salinities increase above 1 mol L^{-1} in monovalent waters[33-37], and small fractions of
69 divalent cations lower σ even further[38]. Thus, measurements of σ for shale systems are needed in

70 order to evaluate the significance of osmotically driven imbibition[39], and confinement of shale
71 samples during such measurements is again warranted for preventing microfracturing.

72 The analyses developed here compare two basic driving forces for imbibition, capillary
73 forces acting at the imbibition front and the hydrostatic reservoir pressure acting at the fracture-
74 matrix boundary. Through comparing both of these forces, the effectiveness of manipulating
75 interfacial properties will be shown to have a strong dependence on both depth and permeability,
76 such that surfactant use in deep reservoirs has negligible ability to reduce imbibition.

77

78 2. METHODOLOGY

79 Because a one-dimensional analysis of transient imbibition will be used to compare influences of
80 matrix rock capillarity and pressures in fractures, we first provide justification for this
81 simplification of the geometry of fluids entering reservoirs through complex networks of
82 hydraulic fractures and natural fractures. A conceptual model of how fracturing fluids are
83 distributed shortly after hydraulic fracturing is shown in Figure 1, with (a) the km scale overview
84 of multiple hydraulic fractures within a single fracturing stage, (b) a m scale depiction of a
85 region including both a hydraulic fracture and numerous secondary fractures (natural and
86 stimulated) supplied with frac fluid via main hydraulic fractures, and (c) a mm scale close-up
87 view of water imbibing into shale matrix from a vertical hydraulic fracture and secondary
88 (natural or stimulated) horizontal fracture. In the days following fracturing and prior to gas
89 production from reservoirs with matrix permeability k in the range of 10^{-21} to 10^{-17} m^2 (n-Darcy to
90 10 μ-Darcy), imbibition fronts only propagate beyond the fracture-matrix interface to distances
91 in the range of mm to cm into the matrix[40]. When such short penetration distances are small
92 relative to characteristic distances between the fractures, water imbibition occurs locally as an

effectively one-dimensional process emanating from complex fracture networks. Therefore, in the analysis developed next, flow is represented as being effectively one-dimensional.

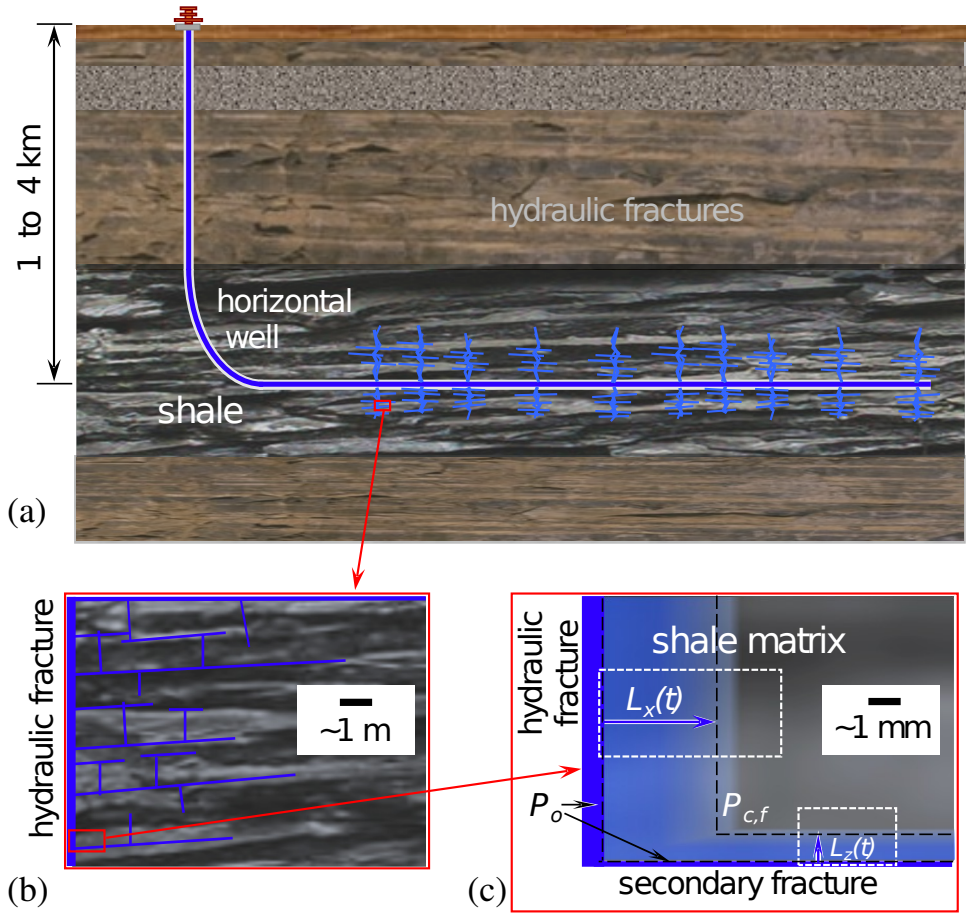

Figure 1. Conceptual model of hydraulic fracturing fluid injection into shale gas reservoir. (a) Cross-section along horizontal well showing water distribution via nominally vertically oriented hydraulic fractures. (b) Local region adjacent to a hydraulic fracture, showing water injected into network of horizontal and vertical fractures (both natural and stimulated). (c) Injected water imbibing into a portion of shale matrix bounded by a vertical hydraulic fracture and a secondary horizontal fracture, both with fluid pressure P_o. This close-up shows the orientation-dependent imbibition front distances $L_i(t)$ at which the capillary pressure is $P_{c,f}$.

At the local scale of shale matrix adjacent to a fracture (microfracture) supplied with hydraulic fracturing fluid, the pressure difference acting across the advancing wet matrix of thickness $L(t)$ is the sum of the pressure at the fracture-matrix interface P_o and the capillary pressure at the advancing wetting front, $P_{c,f}$. Because of strongly anisotropic k in shales[41], $L(t)$ generally exhibits orientation-dependence as indicated in Figure 1c. We will show later that anisotropy can be included through use of k components perpendicular and parallel to bedding planes, but does not impact conclusions concerning alteration of $P_{c,f}$. Thus for simplicity, we consider isotropic k, and a pressure gradient acting across the expanding imbibition zone given by $(P_{c,f} + P_o)/L$. The transient Darcy equation for 1-dimensional flow in the step-function wetting zone approximation is

$$J = \frac{\Delta\theta dL}{dt} = \frac{k}{\mu}\frac{P_o + P_{c,f}}{L} \tag{1}$$

where J is the volumetric flow of water per unit area, $\Delta\theta$ is the volumetric water content increase within the wetted zone, t is time, and μ is the viscosity of the fracturing fluid. Rearrangement and integration of eq 1 gives the time-dependent thickness of the wet zone

$$L(t) = \sqrt{\frac{2k}{\mu\Delta\theta}(P_o + P_{c,f})}\sqrt{t} \tag{2}$$

Because the cumulative volumetric water flux per unit area $I(t) = \Delta\theta L(t)$, the local volumetric imbibition rate per unit area of fracture-matrix contact area is expressed in the Green-Ampt model[40, 42, 43] as

$$I(t) = \sqrt{\frac{2\triangle\theta k}{\mu}(P_{c,f} + P_0)}\sqrt{t} \tag{3}$$

The Green-Ampt equation results directly from Darcy's law applied to the migration of an imbibition front characterized by constant $P_{c,f}$, and precedes the practically identical Lucas-

127 Washburn equation describing flow in capillaries[44, 45], and an equivalent later model by Handy[46]
128 that is commonly used in petroleum recovery research[10, 47-50]. The Green-Ampt model also
129 predates other studies on soils that have highlighted the square-root of time dependence of
130 imbibition, where the first grouped square-root term on the right-hand side of eq 3 is the
131 sorptivity S [51], a parameter now also coming into use for characterizing imbibition in
132 hydrocarbon reservoir rocks[52-54]. In both the Green-Ampt and Handy models, the relative
133 permeability function is not needed because early stages of imbibition under positive pressure
134 (and especially at high pressure) at the Darcy continuum scale proceeds at rates close to that
135 resulting from single phase flow. Recently, it was shown that imbibition measurements used to
136 determine S also provide reasonable estimates of k over a range of measured k that spanned 10
137 orders of magnitude[40]. The fact that the database used to develop the S–k correlation included
138 shales suggests that osmotic effects did not significantly enhance imbibition rates into those very
139 low k samples.

140 Given that k and $\Delta\theta$ of a given reservoir are relatively fixed, the controllable variables
141 available for reducing imbibition are μ , $P_{c,f}$, P_o , and t. While eq 3 clearly shows that thickeners
142 used to increase μ for proppant transport cause decreases in fluid imbibition, rheological
143 properties of fracturing fluids are largely designed with fracture generation and proppant
144 transport in mind[55]. Therefore, our focus here will be on examining the significance of potential
145 changes to $P_{c,f}$, particularly in comparison to P_o. From eq 3, $P_{c,f}$ and P_o act additively within the
146 square-root term to drive imbibition. Thus, the magnitude of $P_{c,f}$ relative to P_o determines the
147 relative importance of surfactant additions on reducing imbibition. It is worth noting that the
148 equivalent effects that $P_{c,f}$ and P_o have in driving imbibition are evident from their additive
149 influences in eq 3, and have been supported by experimental tests conducted over ranges of

150 elevated injection pressures[56, 57]. For the purpose of this study, we focus on the relative
151 magnitudes of $P_{c,f}$ and P_o under commonly employed laboratory spontaneous imbibition
152 experiments and under field reservoir conditions.

153 Newly generated fractures experience elevated fracturing pressures that are depth-
154 dependent. Following shut-in, P_o begins to decline back towards the reservoir pressure[58]. Thus,
155 values of P_o are depth-dependent, being greater than the reservoir pressure and less than the rock
156 fracture pressure. For purposes of this study, the hydrostatic gradient for reservoir pressure was
157 taken as 10 MPa km^{-1}, the fracture gradient was taken as 18 MPa km^{-1} [59], and the wellbore shut-in
158 P_o was assigned values midway between these limiting pressures. Thus, a hypothetical shut-in
159 gradient of 14 MPa km^{-1} was assumed, with P_o declining towards the reservoir pressure in the
160 limit of very long shut-in times (Figure 2). It is important to note that reservoir pressures are very
161 high in deep reservoirs, hence P_o remains high prior to the production phase. Also included in
162 Figure 2 are a lithostatic pressure gradient of 24.5 MPa km^{-1} and geothermal gradient of 25 ˚C
163 km^{-1}.

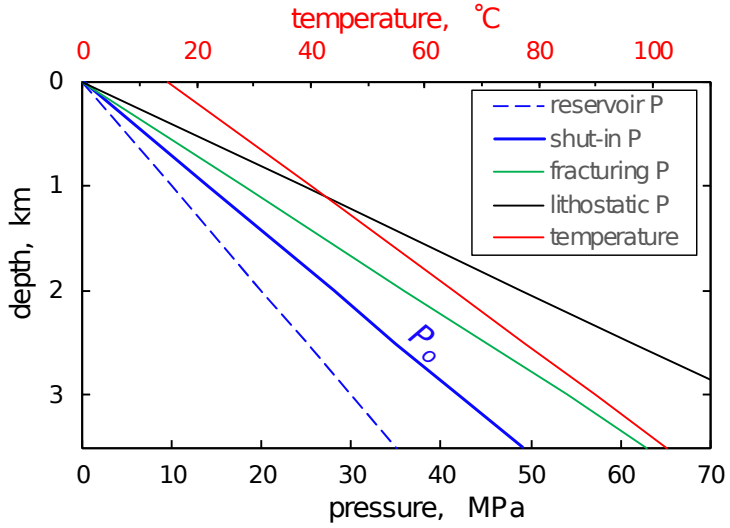

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Figure 2. Depth profiles of reservoir pressure, bottom hole shut-in pressure P_o, formation fracture pressure, lithostatic pressure, and temperature. Surface pressures and temperature are 0 MPa and 15 ˚C, respectively.

The dependence of $P_{c,f}$ on k and depth was determined in three steps, for water-wet rock with negligible variation in contact angle. The impact of wettability will be examined later in this analysis. First, a correlation between k and the capillary pressure for air entry into hydrophilic water-saturated porous materials $P_{c,a}$ was determined using data from the literature on room temperature water-air systems (where interfacial tension $\gamma \approx 72$ mN/m)[40]. The power-law correlation given by

$$P_{c,a}(0.1\ MPa,\ 20\ ^\circ C) = (0.106\ Pa\ m^{0.72})k^{-0.36} \quad (4)$$

fits data on consolidated and unconsolidated media with a root mean-square deviation in $log\ P_{c,a}$ of 0.71, for measurements spanning twelve orders of magnitude in k. In previous work, the same data set was scaled with interfacial tension to estimate the methane gas-entry capillary pressure $P_{c,g}$ for a reservoir at 70 ˚C and 20 MPa[40]. Equation 4 was next multiplied by the previously determined linear correlation[40] $P_{c,f} = 1.21P_{c,a}$ to obtain the dependence of $P_{c,f}$ on k at the ground surface

$$P_{c,f}(z=0,0.1\ MPa,\ 20\ ^\circ C) = \alpha k^{-0.36} \quad (5)$$

where $\alpha = 1.21 \times 0.106$ Pa $m^{0.72} = 0.128$ Pa $m^{0.72}$.

The final step needed to predict how $P_{c,f}$ varies with both depth and k requires determining how the methane-water interfacial tension γ varies with depth z below the ground surface. This relation was obtained by interpolating values of methane-water γ from the

literature[60, 61] over a range of pressure-temperature combinations that are compatible with the P_o and T depth profiles illustrated in Figure 2.

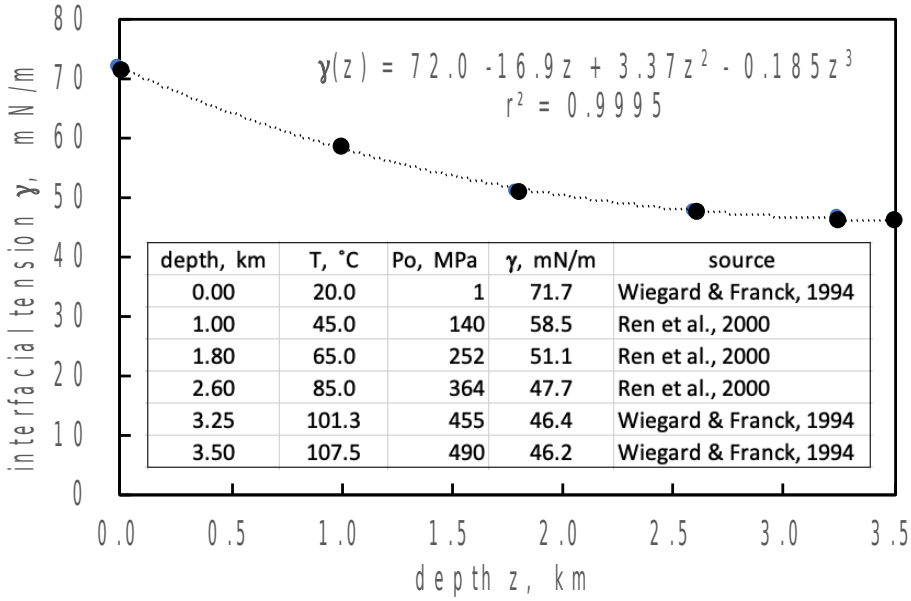


Figure 3. Depth-dependence of methane-water γ, given the estimated temperature and wellbore shut-in pressure profiles.

These depth-dependent methane-water γ(z) values were fit to $\gamma(z) = 72.0 - 16.9z + 3.37z^2 - 0.185z^3$ shown in Figure 3, then used to scale Equation 5 in order to estimate how $P_{c,f}$ depends on both k and z

$$P_{c,f}(z,k) = [\gamma(z)/\gamma(0)]\alpha k^{-0.36} \quad (6)$$

where $\gamma(0)$ = 71.7 mN m^{-1}, the methane-water γ at the ground surface. Substituting the depth-dependent wellbore shut-in $P_o(z)$ and eq 6 into eq 3 gives:

$$I(t)=\sqrt{\frac{2\triangle\theta k}{\mu}\left(\frac{\gamma(z)}{\gamma(0)}\alpha k^{-0.36}+Bz\right)}\sqrt{t} \tag{7a}$$

where B = 14.0 MPa km^{-1}, the wellbore shut-in gradient. The impact of surfactants on $I(t)$ can then be expressed as

$$I(t)=\sqrt{\frac{2\triangle\theta k}{\mu}\left(\frac{f\gamma(z)}{\gamma(0)}\alpha k^{-0.36}+Bz\right)}\sqrt{t} \tag{7b}$$

where f represents the fractional reduction in $\gamma(z)$ imparted by addition of surfactants. For example, reducing γ from 50 mN m^{-1} to 15 mN m^{-1} corresponds to f = 0.3. Thus, the impact of surfactant addition in reducing fracturing fluid imbibition relative to the unaltered fluid (with f = 1) is given by

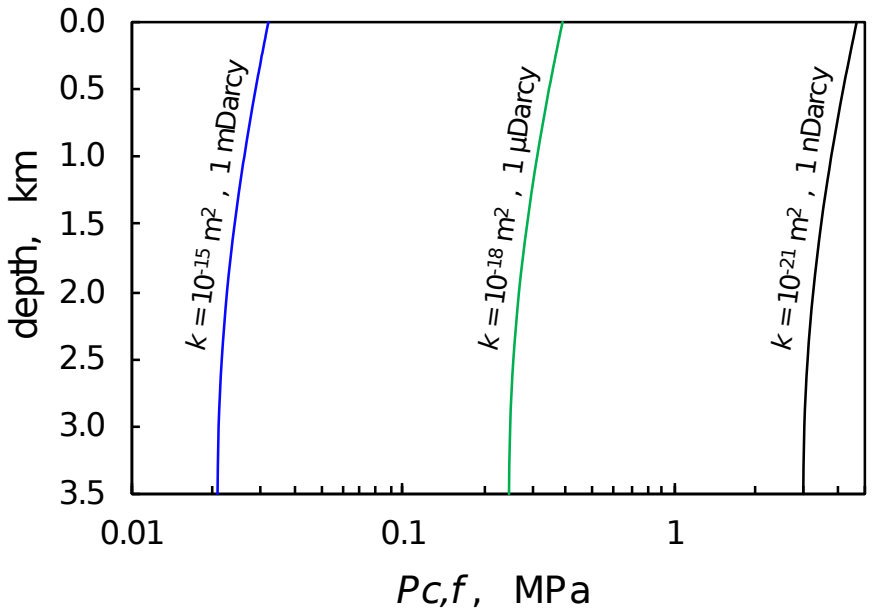
$$1-\frac{I(f,z)}{I_0(z)}=1-\left[\frac{\frac{f\gamma(z)}{\gamma(0)}\alpha k^{-0.36}+Bz}{\frac{\gamma(z)}{\gamma(0)}\alpha k^{-0.36}+Bz}\right]^{0.5} \tag{8}$$

which we term the capillary alteration effectiveness (CAE). The CAE can range from 0 where the imbibed fluid volume is unaffected by the addition of surfactant, up to 1 where surfactant addition prevents any fluid imbibition into the reservoir due to extremely strong hydrophobic interaction. Note that anisotropy can be accommodated for imbibition perpendicular to and parallel to shale bedding planes by using suitable directional components of k in eq 8. Here, it is also convenient to consider how changing wettability can be incorporated into eq 8. Recall that P_c in a straight capillary tube of radius R is given by the Young–Laplace equation

$$P_c=\frac{2\gamma\cos\theta}{R} \tag{9}$$

where θ is the contact angle. Although scaling of P_c by $\cos\theta$, in porous media is not generally accurate[62], it serves as a useful approximation[63]. By scaling P_c by both γ and $\cos\theta$, f can be

220 interpreted more generally to be the product of $\cos\theta$ times the relative reduction in ɣ. Such a
221 generalization allows examining the implications of a hypothetical extremely hydrophobic
222 interaction between the reservoir and fracturing fluid. Such a case, with $\theta = \pi$ and $\cos\theta$ = -1 can
223 then be examined with f = -1, and captures the theoretical maximum impact achievable for
224 capillary alteration of imbibition. The full range of wetting behavior, including that of complex
225 mixed-wet systems[64, 65], is encompassed by varying f from 1 (strongly hydrophilic) to -1 (strongly
226 hydrophobic). In order to focus on the potential impacts that weakening the capillary driving
227 force can have on imbibition, values of f that range from 0.3 to -1 will later be examined.
228
229 3. RESULTS AND DISCUSSION
230 The capillary pressure acting at the imbibition front, $P_{c,f}$, depends on k, and decreases with depth
231 as approximated by eq 4. The strong k-dependence of $P_{c,f}$ shown in Figure 4 reflects the power-
232 law correlation with $k^{-0.36}$ (eq 5), which differs from $k^{-0.5}$ predicted from Leverett scaling[40, 66].
233 Decreases in ɣ with depth (Figure 3) account for the depth-dependence of $P_{c,f}$.
234



235

236 **Figure 4.** Depth-dependence of imbibition front $P_{c,f}$ for different k, showing that $P_{c,f}$ decreases
237 with depth and is larger for lower k.

238

239 The CAE is both k- and depth-dependent, as illustrated in Figure 5, where depth profiles
240 are plotted for $k = 10^{-15}$ (1 mDarcy), 10^{-18} (1 μDarcy), and 10^{-21} m^2 (1 nDarcy), with interfacial
241 tensions reduced by $f = 0.3$, 0.01, and -1. These three f values represent modest, very strong, and
242 maximum possible decreases in the capillary driving force for imbibition, respectively.
243 Influences of surfactant alteration can be more important in lower k reservoirs, e.g. shales,
244 because their higher $P_{c,f}$ can be more effectively reduced with surfactant addition. However,
245 capillary alteration effectiveness decreases with depth because the pressure at the fracture-matrix
246 boundary P_o becomes large enough to be the dominant driving force, even in very low k
247 reservoirs. The depths by which capillary alteration effectiveness diminishes to only 10% are
248 reached where the profiles cross the vertical red line in Figure 5. Thus, even with interfacial
249 tension reduced by $f = 0.01$, capillary alteration effectiveness is diminished by only 10% at

depths of 0.11 and 1.1 km, for 1 μDarcy and 1 nDarcy reservoirs, respectively. At the much greater depths commonly stimulated with hydraulic fracturing, the effectiveness in reducing fluid imbibition with surfactants will be negligible. Figure 5 also shows that even if a 1 nDarcy reservoir was stimulated with a fluid that imparted extremely hydrophobic interactions ($\theta = \pi, f =$ -1), the effectiveness in reducing imbibition would be limited to less than 10% at depths greater than 2.2 km. Because k is commonly larger parallel to shale bedding planes, Figure 5 shows that the CAE for imbibition from vertical fractures is far less than that for imbibition from horizontal fractures. Given that μ is generally optimized for proppant delivery, only decreases in P_o and t remain as controllable variables that have the capacity to significantly reduce imbibition during reservoir stimulation.

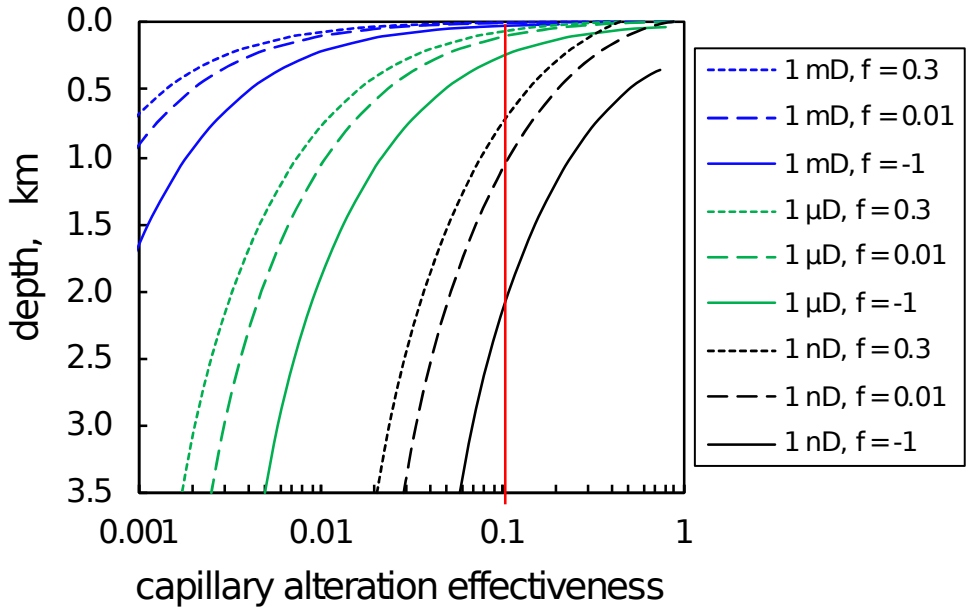


Figure 5. The effectiveness of altering interfacial forces in order to minimize fracturing fluid imbibition depends on reservoir k (1 mDarcy, 1 μDarcy, 1 nDarcy) and depth. The parameter f in the legend represents the fractional reduction in water-gas surface tension achieved by addition

265 of surfactant. The cases with f = -1 represent the maximum reduction in capillary uptake
266 achievable with extremely hydrophobic interactions.
267
268 The shallow limit of z = 0 km in Figure 5 represents typical laboratory imbibition tests,
269 where water or fracturing fluids are commonly introduced with negligible pressure head, i.e. P_o
270 $\approx$ 0 MPa. Under negligible P_o, alteration of $P_{c,f}$ with surfactants results in clear decreases to
271 imbibition rates shown in recent studies[9-12]. It is noteworthy that the contrasting effects of high
272 P_o (24 MPa) versus negligible P_o was recognized in comparisons of imbibition without and with
273 surfactants in a previous laboratory study on imbibition of fracturing fluids[67]. Our analyses
274 generalizes such findings by showing how the depth-dependent P_o and k-dependent $P_{c,f}$ limit the
275 effectiveness that alterations of interfacial tension and wettability can have on imbibition.
276 CONCLUSION
277 Thicknesses of water-blocked regions along fracture-matrix interfaces grow by interfacial
278 tension-dependent capillarity acting at the advancing wetting front $P_{c,f}$, and by the fracturing fluid
279 pressure acting at the fracture-matrix boundary, P_o. Therefore, it is important to understand the
280 relative magnitudes of these driving forces in order to determine the effectiveness of surfactant
281 additions aimed at reducing the capillary contributions to imbibition. Our analyses show the
282 following.
283 • The extent to which surfactants can reduce imbibition is limited by the k-dependent
284 magnitude of $P_{c,f}$, and has greater potential effect in lower k reservoirs.
285 • Magnitudes of P_o in deep reservoirs are in the range of tens of MPa, and are greater than
286 $P_{c,f}$. values of even the lowest k reservoirs.

- Therefore, alteration of water-gas interfacial tension and wettability have negligible ability to reduce imbibition of water-based fracturing fluids in deep, unconventional reservoirs.
- Given that rheological properties of fracturing fluids are largely prescribed for the purpose of delivering proppants into fractures, the remaining operational options for minimizing imbibition and water block thicknesses are reducing shut-in times and reducing the magnitude of P_o.

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