Supporting Information:

Line-tension determination for a CO_2/water/Bentheimer system

Including the concept of line tension, which gives the dependency of the contact angle on bubble size [27, 29, 30, 34, 35], leads to the modified Young equation:

$$\gamma_{lv} \times cos\theta + \frac{\sigma}{R} = \gamma_{sv} - \gamma_{sl} \ , \qquad (S.1)$$

where σ is the line tension (J/m) and R is the radius (m) of the solid-liquid contact circle that is the cross-section of the bubble which is captured on the surface (Fig.1). For an axisymmetric bubble on a homogeneous, smooth and horizontal surface, the three-phase contact line is a circle. If the bubble is extremely large (i.e., $R \rightarrow \infty$), the modified Young equation reduces to the original Young equation:

$$\gamma_{lv} \times cos\theta_{\infty} = \gamma_{sv} - \gamma_{sl} \ , \qquad (S.2)$$

where θ_{∞} is the contact angle for the bubble with $R \rightarrow \infty$. Combining Eqs. S.1 and S.2 yields to an expression describing the contact angle for a bubble with a finite radius:

$$cos\theta = cos\theta_{\infty} - \frac{\sigma}{R\gamma_{lv}} \qquad (S.3)$$

The infinite contact angle and the line tension can be determined at a constant temperature and pressure from a plot of $cos\theta$ versus *1/R*.

For the system at hand, the line tension is determined using the contact-angle data as a function of bubble size and the length of the contact line (section 4.1.1). Due to the pressure dependency of the interfacial tension, the line tension is also a function of pressure. Therefore, to identify the dependency of the contact angle on the bubble size and line-tension determination, the pressure effect has been excluded by using only results of the contact angle at a constant pressure of either 1.04 or 4.97 MPa (Figs. S.1a and S.1b).

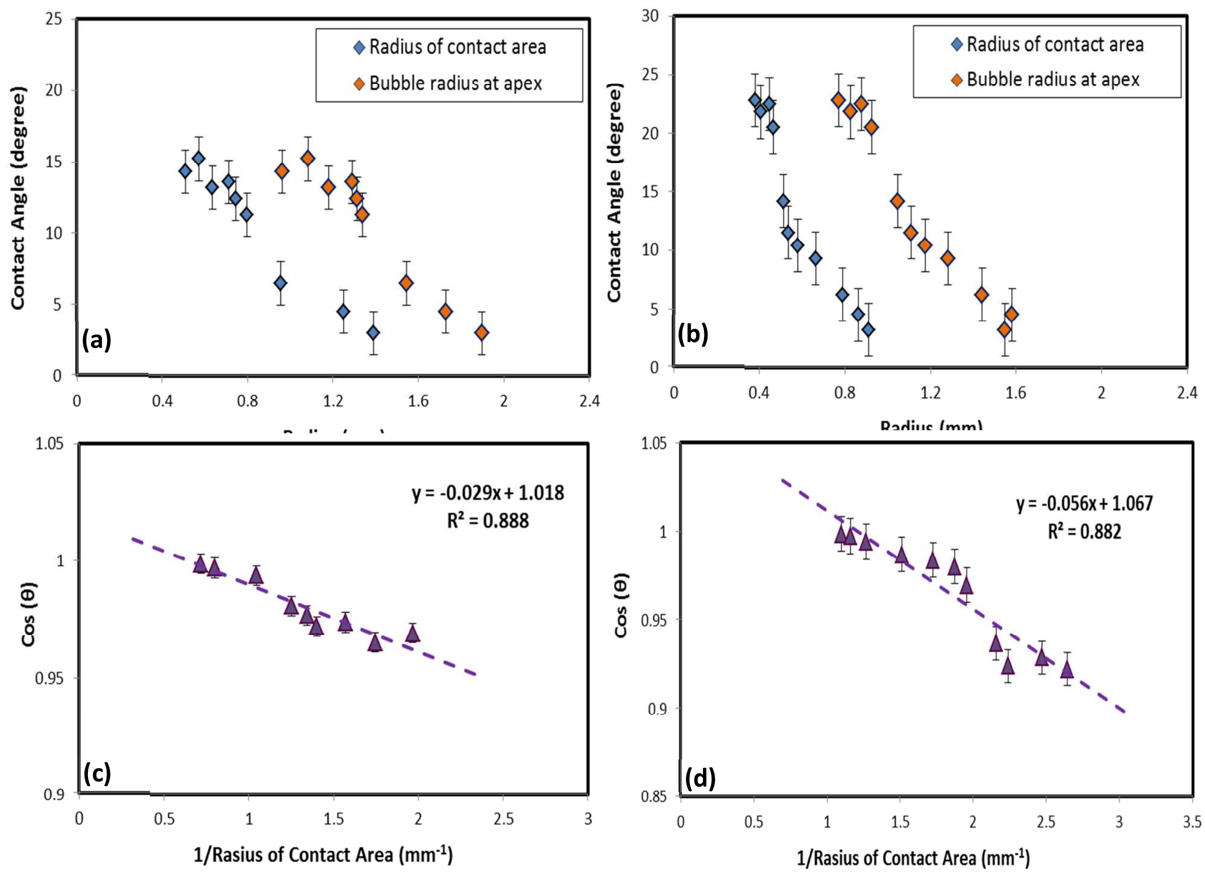

Figure S.1. Contact angle versus bubble radius or the inverse length of the contact line at two different pressures: (a, c) at 1.04 MPa and (b, d) at 4.97 MPa. Error bars are obtained based on the standard error calculation.

The line tension, σ, has been derived based on the data given in Figs. S.1c and S.1d and using Eq. S.3. At constant pressures of 1.04 MPa and 4.97 MPa, the line tensions were 1.921×10^{-6} (J/m) and 2.46×10^{-6} (J/m), respectively. However the intercepts of the linear fit between $cos\theta$ and *1/R,* which give the contact angle for the infinite large bubble ($R \rightarrow \infty$), were more than one, i.e. 1.018 and 1.067 for constant pressures of 1.04 MPa and 4.97 MPa, respectively. A value of $cos\theta$ larger than one is mathematically impossible. It originates from: 1) the fact that the contact angle of the very large CO_2 bubble on Bentheimer sandstone is zero, which means that the system is absolutely water-wet and there is no three phase contact line in this case, and/or 2) the uncertainty in the determined values of the parameters used for the calculation of the line tension such as contact angle and length of the contact line. Nevertheless, no data of line tension are found in the literature on Bentheimer sandstone to provide a comparison with previous studies.

In addition to the contradictory results in literature regarding line-tension determination, for such a strongly water-wet system like Bentheimer sandstone with small contact angles (<20°), the bubble profiles near the solid surface are sometimes blurry and indistinct. In this system, even a small error in the detection of the bubble contact with the solid surface may cause errors in the determined contact angles. This uncertainty may lead to overestimation in the infinite contact-angle determination. Following these results, it can be concluded that the line-tension concept may not be a proper method to describe the dependency of the contact angle on the bubble size for the system at hand, since surface non-ideality and roughness have a significant influence on the reliability of this method.