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**Notes**

# Stylolites in limestones: Barriers to fluid flow?

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## ABSTRACT

**Stylolites—products of intergranular pressure-solution—are laterally extensive, planar features. They are a common strain localization feature in sedimentary rocks. Their potential impact on regional fluid flow has interested geoscientists for almost a century. Prevalent views are that they act as permeability barriers, although laboratory studies are extremely rare. Here we report on a systematic laboratory study of the influence of stylolites on permeability in limestone. Our data demonstrate that, contrary to conventional wisdom, the studied stylolites do not act as barriers to fluid flow. In detail, when a stylolite occurs perpendicular to the direction of flow, the permeability simply follows the same power law permeability-porosity trend as the stylolite-free material. We show, using a combination of high-resolution (4 μm) X-ray computed tomography, optical microscopy, and chemical analyses, that the stylolites of this study are not only perforated layers constructed from numerous discontinuous pressure solution seams, but comprise minerals of similar or lower density to the host rock. The stylolites are not continuous high-density layers. Our data affirm that stylolites may not impact regional fluid flow as much as previously anticipated.**

## INTRODUCTION

Stylolites are complex column-and-socket interdigitation features that form as a result of intergranular pressure solution (e.g., Nenna and Aydin, 2011; Croizé et al., 2013). They are a common product of strain localization in sedimentary rocks and can form laterally extensive, planar drapes (some stylolites are known to reach lengths of almost 1 km; Safaric and Davison, 2005). Knowledge of their impact on regional fluid flow is an important consideration in many facets of geoscience (e.g., geotechnical engineering, petroleum geoscience, structural geology). Widespread opinion, inferred from a variety of petrographic analyses and borehole logging data, suggests that either (1) stylolites and/or the reprecipitation associated with their formation decrease the porosity and permeability of the host rock, or (2) stylolites act as permeability barriers (e.g., Dunnington, 1967; Nelson, 1981; Burgess and Peter, 1985; Koepnick, 1987; Finkel and Wilkinson, 1990; Dutton and Willis, 1998; Alsharhan and Sadd, 2000). By contrast, several studies have observed that, during the formation of a stylolite, enhanced porosity zones can develop along the flanks and at the tip of the stylolite (Carozzi and von Bergen, 1987; Dawson, 1988; Raynaud and Carri-Schaffhauser, 1992; Van Geet et al., 2000; Gingras et al., 2002; Harris, 2006). These zones of enhanced porosity have been hypothesized to enhance the circulation of fluids (Carozzi and von Bergen, 1987; Van Geet et al., 2000). However, while recent laboratory investigations have shown that compaction bands (another type of strain localization feature) significantly decrease the permeability of sedimentary rock (by up to three orders of magnitude; Baud et al., 2012), complementary laboratory data on the impact of stylolites are extremely rare. An isolated study by Lind et al. (1994) found that the permeability of high porosity chalk samples containing stylolites did not differ from the adjacent stylolite-free material, unless the stylolite was associated with a fracture. However, no systematic study on the influence of stylolites on permeability has been published to date. Here therefore we present a systematic laboratory study on the porosity and permeability of limestones containing stylolites.

## MATERIALS AND METHODS

The materials of this study were taken from vertical boreholes drilled around the ANDRA Underground Research Laboratory near Bure, France. We selected four lithologies to sample (Fig. 1), three within the Oxfordian stage (Late Jurassic) and one within the Middle Jurassic “Dogger” series. We selected these lithologies based on their differences in porosity and texture. Macroscopically there is no systematic difference in stylolite density or thickness (50–200 μm) between the limestone units, although occasional thicker stylolites (1500–3000 μm) are observed in the Dogger limestone. In this study we chose to focus on bedding-parallel sedimentary stylolites; subvertical stylolites of tectonic origin are also present within these formations (Rolland et al., 2013), but only occur rarely in borehole cores due to the vertical drilling orientation.

The first Oxfordian limestone, O1 (depth = 159 m), is an oomicrite (grain size = 0.1–0.5 mm) with a porosity of ~15.4 vol%. We also note the occasional microfossil (foraminifera) and shell fragment. The second, O3 (depth = 174 m), is an oomicrite (grain size = 0.2–1.0 mm) with a porosity of ~15.0 vol%. Finally, O6 (depth = 364 m) is a biomicrite with a porosity of ~6.7 vol%. O6 is very heterogeneous and contains large intraclasts (>1 mm in diameter) containing cemented shell debris and cement overgrowth, and large (>1 mm in length) shell fragments. The Dogger limestone (depth = 747 m) is a micrite with a porosity of 3.4 vol%. The Dogger limestone contains both thin (20–500 μm) and thick (1500–3000 μm) stylolites. More information on the materials of this study can be found in the GSA Data Repository<sup>1</sup>.

Cylindrical core samples were prepared to contain either (1) one stylolite perpendicular to the long axis of the core (i.e., perpendicular to the imposed flow direction), or (2) no stylolite (Fig. 1). We also prepared complementary samples containing a single stylolite parallel to the long axis of the core (i.e., parallel to the imposed flow direction). Our samples were carefully selected, after coring and grinding, to contain no obvious stylolite-associated fractures. Although stylolites may become fractured during partial uplift or unloading during mesodiagenesis, we did not want to provide false overestimates for permeability associated with any laboratory-induced fracturing (Nelson, 1981). We note that no microfractures were observed during optical microscopic, scanning electron microscopic, or X-ray computed tomographic (μCT) analyses of the stylolites. The gas permeability of each core sample was measured in a hydrostatic pressure vessel and porosities were measured using the triple-weight saturation technique. Details on the experimental methods can be found in the Data Repository.

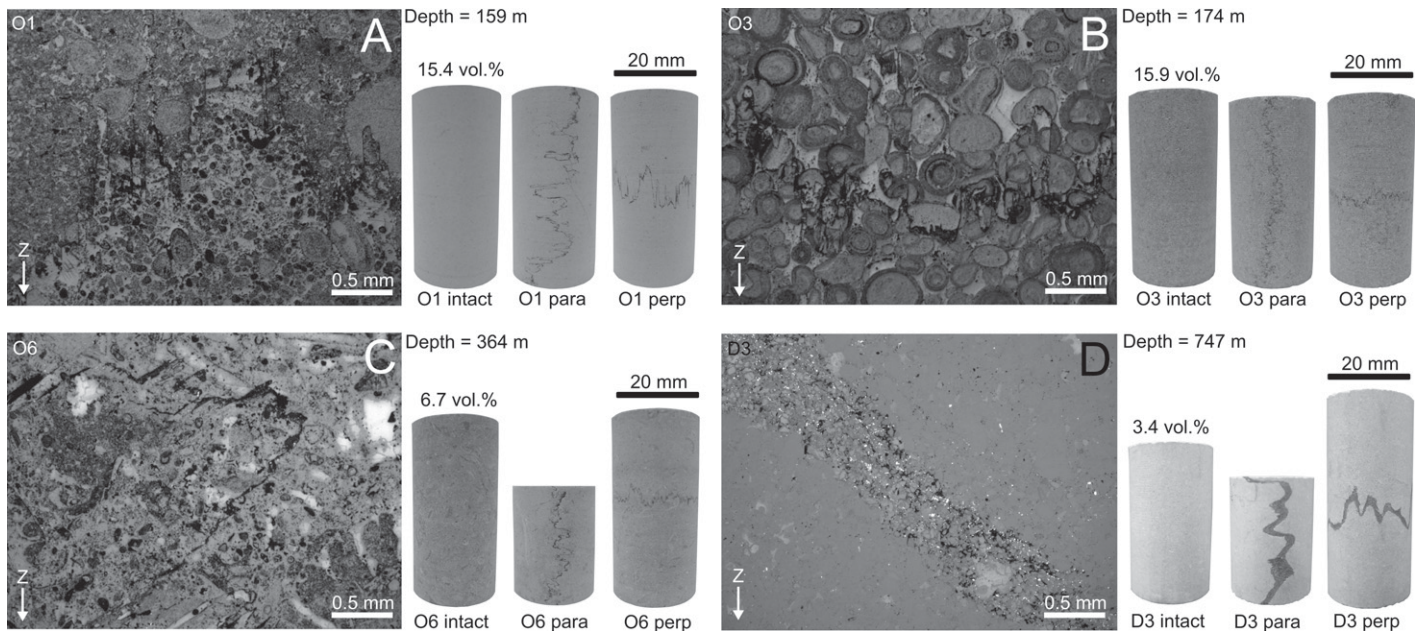
To interpret these data we employed a combination of optical microscopy, high-resolution (4 μm) μCT, and chemical analyses (details on the chemical analyses can be found in the Data Repository).

## RESULTS AND DISCUSSION

### Porosity of Stylolitic Samples

We note that the porosity of the samples containing stylolites is nearly always higher than their stylolite-free counterparts (Table DR1

<sup>1</sup>GSA Data Repository item 2014013, additional materials, methods, and analyses, is available online at [www.geosociety.org/pubs/ft2014.htm](http://www.geosociety.org/pubs/ft2014.htm), or on request from [editing@geosociety.org](mailto:editing@geosociety.org) or Documents Secretary, GSA, P.O. Box 9140, Boulder, CO 80301, USA.



**Figure 1.** Optical photomicrographs showing the stylolites of this study, together with photographs showing examples of the three types of core sample for O1 (A), O3 (B), O6 (C), and D3 (D). The porosities of the stylolite-free core samples are given above the relevant photograph, and the borehole depths are given next to the photomicrographs. All photomicrographs are of polished thin sections viewed under reflected light. The Z-direction indicates the direction of the borehole drilling (i.e., vertical and perpendicular to bedding).

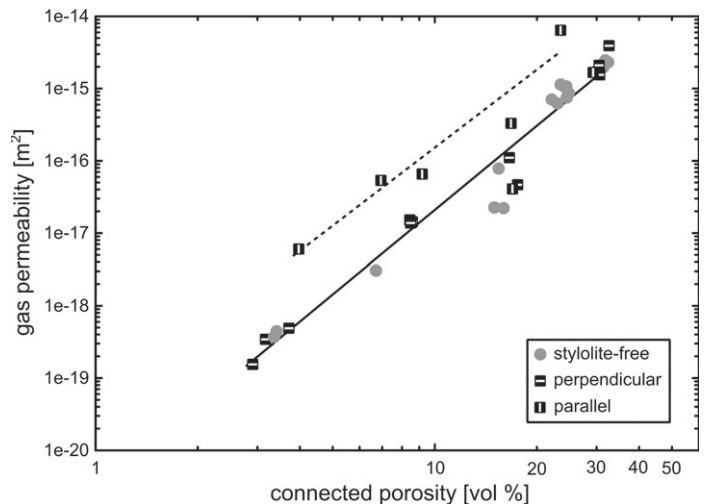
in the Data Repository). The presence of a higher porosity zone surrounding a stylolite has been observed previously, and its origin has been intensely debated. The higher porosity zone has been postulated to be a consequence of (1) post-stylolitization marginal dissolution, where stylolite seams have served as conduits for calcium carbonate-undersaturated basal fluids (Dawson, 1988), (2) solute mass transfer from a “process zone” on either side of the stylolite to a “cementation zone” during stylolite formation (Raynaud and Carrio-Schaffhauser, 1992), (3) their sutured, medium- to high-amplitude nature, encouraging the generation of fabric-selective secondary porosity (Carozzi and von Bergen, 1987), or (4) that they form preferentially in zones of higher porosity and permeability (Braithwaite, 1989), an argument also suggested for compaction bands in limestones (Vajdova et al., 2012). While the origin of this porosity is not the focus of this study, we reiterate that care was taken during sample selection to exclude the possibility of stylolite-associated fracture porosity.

#### Influence of Stylolites on Fluid Flow

Our experimental data, together with those of Lind et al. (1994), are shown in Figure 2 (and provided in Table DR2). The compiled data show that, over the entire range of porosity (3–33 vol%), gas permeability ranges from  $\sim 10^{-19}$  to  $\sim 10^{-14}$  m<sup>2</sup>. Firstly, we note that we found no evidence of permeability anisotropy within the stylolite-free material (Table DR2). The data for stylolite-free samples and for samples containing stylolites perpendicular to fluid flow can be adequately described by the same power law permeability- porosity relationship defined by the black solid line in Figure 2. In other words, the studied stylolites do not act as barriers to fluid flow. Our data also allow us to consider whether the stylolites (and/or any associated features, such as “process zones” along their flanks) can act as conduits for fluid flow. We note that five of the samples, all with stylolites parallel to the direction of flow, exhibit permeabilities that are about an order of magnitude higher than the aforementioned power law permeability- porosity relationship (black dashed line in Fig. 2). These latter data suggest that, in some cases, the stylolites (and/or any associated features) can act as conduits for fluid flow.

#### Optical Microscopic, $\mu$ CT, and Chemical Analyses

Stylolites are thought to be the product of the horizontal linkage and vertical coalescence of numerous pressure-solution seams (Nenna and Aydin, 2011). The hierarchical nature of stylolite formation, combined with the impact of grain-scale heterogeneities (Ebner et al., 2010) and the inhomogeneous stress distribution surrounding geometric asperities (Zhou and Aydin, 2010), results in (1) a complex internal structure (inset

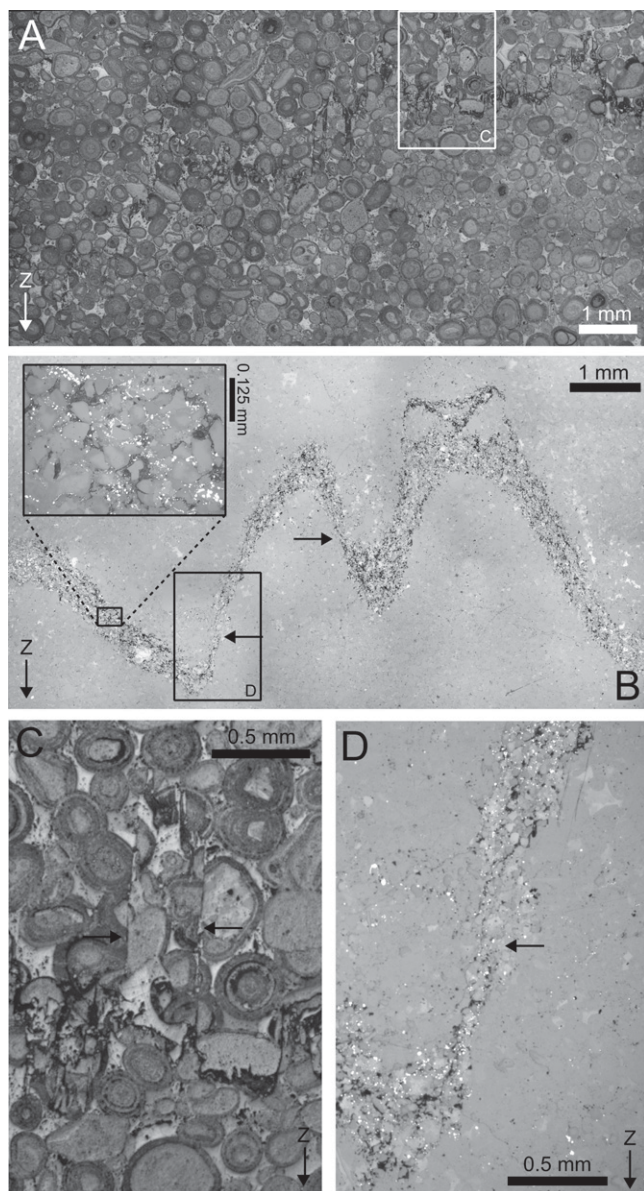


**Figure 2.** Log porosity against log gas permeability for all of the samples measured in this study (measured under a confining pressure of 2 MPa, the data are provided in Table DR2 in the Data Repository [see footnote 1]), together with the gas permeability data from Lind et al. (1994) on high-porosity chalk (the data above 20 vol% porosity). The data of Lind et al. (1994) were collected under a confining pressure of 1.8 MPa using air as a pore fluid. Black solid line corresponds to the power law fit through the “stylolite-free” and “stylolite perpendicular to flow” data. The black dashed line corresponds to the power law fit through the “stylolite parallel to flow” data (excluding two data points that follow the previous trend).

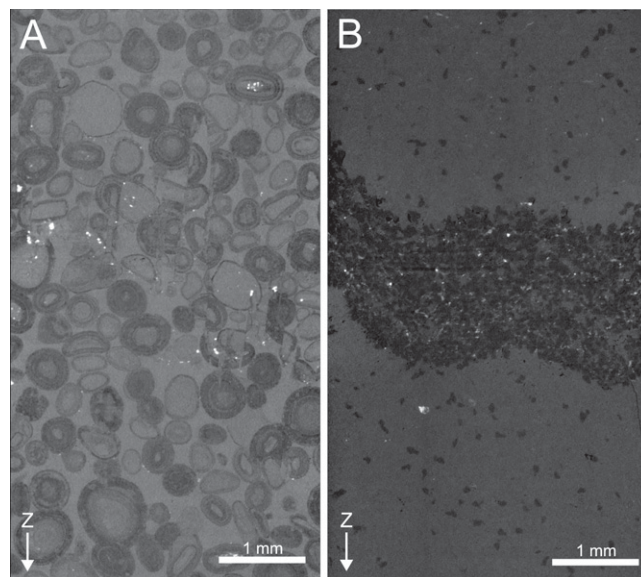
in Fig. 3B), and (2) an inconsistent stylolite thickness. Importantly, we observe that, in some areas, the thickness of the stylolite (i.e., the insoluble residue) is at or close to zero (Figs. 3C and 3D). We can conclude that not only could the stylolites be viewed as “perforated” layers that provide many pathways for fluid flow, but their discontinuous internal structure may not construct an effective barrier.

In the past decade, CT has become a popular and powerful tool in the geosciences. The technique provides tomographic images of the sample where the gray level is a proxy for density; darker regions being

of lower density, and vice-versa. We performed  $\mu$ CT on an example of a thin (50–200  $\mu$ m) and a thick (1500–3000  $\mu$ m) stylolite (Fig. 4). We note that both rocks are chiefly microporous. The stylolites should be observable in the  $\mu$ CT images if any significant density contrast exists on the microscale (the thin stylolite should be 12–40 pixels in width). However, the only visual indications of the thin stylolite are (1) a tortuous, sporadic line of high-density pyrite particles, and (2) truncated oolites (Fig. 4A). There is no high-density layer. By contrast, the thick stylolite is clearly visible, but as a low-density layer (Fig. 4B). Previous CT data (Raynaud et al., 1990; Van Geet et al., 2000, 2001), albeit at lower resolution, and magnetic resonance imaging (Gingras et al., 2002) analysis of stylolite-bearing limestone samples have also shown that stylolites do not appear as high-density layers. The reason for this is likely due to the mineralogy of the original host rock. As calcite is dissolved and is precipitated elsewhere, any insoluble minerals (i.e., minerals that have a significantly lower dissolution rate than calcite) present in the host rock will accumulate along the pressure-solution interfaces. Our chemical analyses show that the stylolites of this study are enriched with quartz, dolomite, and gypsum (with minor pyrite). Quartz (density = 2650 kg/m<sup>3</sup>) and gypsum (2320 kg/m<sup>3</sup>) are less dense than the predominantly calcitic host rock (2710 kg/m<sup>3</sup>), and dolomite is not significantly denser (2850 kg/m<sup>3</sup>). There is therefore little wonder that we did not see a significant density contrast for the thin stylolite. Three possible reasons exist for the thick stylolite to appear as a low-density layer: (1) that the stylolite represents a high porosity layer, (2) that the stylolite contains minerals of a lower density, or (3) a combination of the two. However, while we can confirm that the thick stylolite contains an abundance of quartz (Table DR1), we cannot draw any firm conclusions as to whether it represents a zone of elevated porosity. Although, we note that the thick stylolite has a rather complex internal structure (inset in Fig. 3B), in contrast to the organized, micritic host rock, and therefore has a higher potential to contain intergranular porosity. This may offer an explanation as to why some stylolites act as a conduit when they occur parallel to the flow direction. Although, if stylolites form preferentially in zones of



**Figure 3.** A: Optical photomicrograph map of a sample of O3 containing a thin (50–200  $\mu$ m) stylolite. B: Optical photomicrograph map of a sample of D3 containing a thick (1500–3000  $\mu$ m) stylolite. Black arrows point to positions where the stylolite is at or close to zero thickness. Inset shows the discontinuous nature of the pressure-solution seams forming the macroscopic stylolite seam. C and D: Optical photomicrographs of a sample of O3 (C) and D3 (D) containing a thin and thick stylolite, respectively. Black arrows point to positions where the stylolite is at or close to zero thickness. Locations of these images are shown on maps in A and B. All photomicrographs are of polished thin sections viewed under reflected light. Z-direction indicates direction of borehole drilling (i.e., vertical and perpendicular to bedding).



**Figure 4.** High-resolution (4  $\mu$ m) X-ray computed tomographic images of a thin stylolite (50–200  $\mu$ m; sample O3) (A) and, a thick stylolite (1500–3000  $\mu$ m; sample D3) (B). Dark gray concentric bands within the ooids in A represent bands with a higher porosity (microporosity); light gray is calcite that contains significantly less microporosity. Dark gray minerals in B are quartz grains; light gray is calcite. White dots in A and B are pyrite grains. Z-direction indicates direction of borehole drilling (i.e., vertical and perpendicular to bedding).

higher porosity and permeability (e.g., Braithwaite, 1989) or they contain a “process zone” of higher porosity (Raynaud and Carrio-Schaffhauser, 1992), the higher permeability could simply be explained by flow through a locally more porous and permeable host rock.

Although the experimental data of our study suggest that stylolites in limestones do not act as barriers to fluid flow, caution is perhaps still required. Some stylolites may have the potential to become barriers, and is likely to depend on the abundance of insoluble minerals with a “permeability reducing potential” in the host rock. For example, if the stylolite becomes passively enriched with platy clays or phyllosilicates then the subsequent compaction of these minerals may create a low porosity and permeability layer. However, as explained above, it is still questionable as to whether stylolites can form a consistent and continuous layer. We also note that, while our study does not support the assumption that stylolites are barriers to flow, if the dissolved calcite is reprecipitated locally, the reduction in host rock porosity could reduce its permeability, thus providing an obstacle for regional fluid migration. Local precipitation may be encouraged by (1) a low fluid flux, (2) a locally low partial pressure of CO<sub>2</sub> or high pH, and/or (3) a locally high temperature. Calcite precipitation can also be influenced by mineral coatings (e.g., Zuddas et al., 2003). Indeed, stylolite density has previously been measured to be inversely proportional to porosity in certain limestone reservoirs (e.g., Alsharhan and Sadd, 2000). The data of our study suggest that, under these circumstances, it is likely that the local precipitation, rather than the stylolite, is influencing fluid migration.

## CONCLUDING REMARKS

Our study affirms that the abundant stylolites within the Jurassic limestones at Bure (France) are not barriers to fluid flow. The stylolites of this study are not only perforated layers constructed from numerous discontinuous pressure solution seams, but comprise minerals of similar or lower density to the host rock. Our study also suggests that these stylolites can act as conduits for fluid flow. This may be due to enhanced flow through a more porous and permeable stylolite, or a locally more porous and permeable host rock. This would imply that, if one were to consider sedimentary stylolites only, the movement of fluids is unimpeded vertically and enhanced laterally. We therefore suggest that similar (in terms of composition, maturity, etc.) stylolites in limestones worldwide may impact fluid flow in a different manner than previously thought. However, we note that stylolites could provide a barrier to flow if they are enriched with minerals with a “permeability reducing potential” and that the impact of stylolitization can result in host rock porosity reduction which may lower formation permeability and provide an obstacle for regional fluid migration.

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## REFERENCES CITED

- Alsharhan, A., and Sadd, J.L., 2000, Stylolites in Lower Cretaceous carbonate reservoirs, U.A.E: Society for Sedimentary Geology Special Publication, v. 69, p. 185–207.
- Baud, P., Meredith, P.G., and Townend, E., 2012, Permeability evolution during triaxial compaction of an anisotropic porous sandstone: *Journal of Geophysical Research*, v. 117, B05203, doi:10.1029/2012JB009176.
- Braithwaite, C.J.R., 1989, Stylolites as open fluid conduits: *Marine and Petroleum Geology*, v. 6, p. 93–96, doi:10.1016/0264-8172(89)90078-0.
- Burgess, C. J. and Peter, C. K., 1985, Formation, distribution, and prediction of stylolites as permeability barriers in the Thamama Group, Abu Dhabi: Middle East Oil Technical Conference and Exhibition, 11–14 March 1985, Bahrain, doi:10.2118/13698-MS.
- Carozzi, A.V., and von Bergen, D., 1987, Stylolitic porosity in carbonates: A critical factor for deep hydrocarbon production: *Journal of Petroleum Geology*, v. 10, p. 267–282, doi:10.1111/j.1747-5457.1987.tb00946.x.
- Croizé, D., Renard, F., and Gratier, J.-P., 2013, Chapter 3—Compaction and porosity reduction in carbonates: A review of observations, theory, and experiments: *Advances in Geophysics*, v. 54, p. 181–238, doi:10.1016/B978-0-12-380940-7.00003-2.
- Dawson, W.C., 1988, Stylolite porosity in carbonate reservoirs: American Association of Petroleum Geologists Search and Discovery Article, American Association of Petroleum Geologists Annual Convention, Houston, Texas, 20–23 March 1988, Article #91030.
- Dunnington, H.V., 1967, Aspects of diagenesis and shape change in stylolitic limestone reservoirs: 7<sup>th</sup> World Petroleum Congress, 2–9 April 1967, Mexico City, Mexico, v. 2, 12129, p. 339–352.
- Dutton, S.P., and Willis, B.J., 1998, Comparison of outcrop and subsurface sandstone permeability distribution, Lower Cretaceous Fall River Formation, South Dakota and Wyoming: *Journal of Sedimentary Research*, v. 68, p. 890–900, doi:10.2110/jsr.68.890.
- Ebner, M., Piazolo, S., Renard, F., and Koehn, D., 2010, Stylolite interfaces and surrounding matrix material: Nature and role of heterogeneities in roughness and microstructural development: *Journal of Structural Geology*, v. 32, p. 1070–1084, doi:10.1016/j.jsg.2010.06.014.
- Finkel, E.A., and Wilkinson, B.H., 1990, Stylolitization as source of cement in Mississippian Salem Limestone, west-central Indiana: *The American Association of Petroleum Geologists Bulletin*, v. 74, p. 174–186.
- Gingras, M.K., MacMillan, B., and Balcom, B.J., 2002, Visualizing the internal physical characteristics of carbonate sediments with magnetic resonance imaging and petrography: *Bulletin of Canadian Petroleum Geology*, v. 50, p. 363–369, doi:10.2113/50.3.363.
- Harris, N.B., 2006, Low-porosity haloes at stylolites in the feldspathic Upper Jurassic Ula sandstone, Norwegian North Sea: An integrated petrographic and chemical mass-balance approach: *Journal of Sedimentary Research*, v. 76, p. 444–459, doi:10.2110/jsr.2006.040.
- Koepnick, R.B., 1987, Distribution and permeability of stylolite-bearing horizons within a Lower Cretaceous carbonate reservoir in the Middle East: *Society of Petroleum Engineers Formation Evaluation*, v. 2, doi:10.2118/14173-PA.
- Lind, I., Nykjaer, O., Priisholm, S., and Springer, N., 1994, Permeability of stylolite-bearing chalk: *Journal of Petroleum Technology*, v. 46, p. 986–993, doi:10.2118/26019-PA.
- Nelson, R.A., 1981, Significance of fracture sets associated with stylolite zones: *The American Association of Petroleum Geologists Bulletin*, v. 65, p. 2417–2425.
- Nenna, F., and Aydin, A., 2011, Formation and growth of pressure solution seams in clastic rocks: A field and analytical study: *Journal of Structural Geology*, v. 33, p. 633–643, doi:10.1016/j.jsg.2011.01.014.
- Raynaud, S., and Carrio-Schaffhauser, E., 1992, Rock matrix structures in a zone influenced by a stylolite: *Journal of Structural Geology*, v. 14, p. 973–980, doi:10.1016/0191-8141(92)90028-U.
- Raynaud, S., Carrio-Schaffhauser, E., Mazerolle, F., and Latière, H.-J., 1990, Computed tomography a tool for measuring porosity in millimetric rock volumes around a stylolite joint: *Comptes Rendus de l'Académie des Sciences Paris*, v. 310, p. 271–276.
- Rolland, A., Toussaint, R., Baud, P., Conil, N., and Landrein, P., 2013, Morphological analysis of stylolites for paleostress estimation in limestones: *International Journal of Rock Mechanics and Mining Sciences*, doi:10.1016/j.ijrmms.2013.08.021.
- Safaric, M., and Davison, I., 2005, Pressure solution in chalk: *The American Association of Petroleum Geologists Bulletin*, v. 89, p. 383–401, doi:10.1306/10250404015.
- Vajdova, V., Baud, P., Wu, L., and Wong, T.-f., 2012, Micromechanics of inelastic compaction in two allochemical limestones: *Journal of Structural Geology*, v. 43, p. 100–117, doi:10.1016/j.jsg.2012.07.006.
- Van Geet, M., Swennen, R., and Wevers, M., 2000, Quantitative analysis of reservoir rocks by microfocus X-ray computerized tomography: *Sedimentary Geology*, v. 132, p. 25–36, doi:10.1016/S0037-0738(99)00127-X.
- Van Geet, M., Swennen, R., and Wevers, M., 2001, Towards 3-D petrography: Application of microfocus computer tomography in geological science: *Computers & Geosciences*, v. 27, p. 1091–1099, doi:10.1016/S0098-3004(00)00154-0.
- Zhou, X., and Aydin, A., 2010, Mechanics of pressure solution seam growth and evolution: *Journal of Geophysical Research*, v. 115, B12207, doi:10.1029/2010JB007614.
- Zuddas, P., Pachana, K., and Faivre, D., 2003, The influence of dissolved humic acids on the kinetics of calcite precipitation from seawater solutions: *Chemical Geology*, v. 201, p. 91–101, doi:10.1016/S0009-2541(03)00230-4.

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