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**The influence of rock heterogeneity on the scaling properties of simulated
and natural stylolites**

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22 **Abstract**

23
24 Stylolites are among the most prominent deformation patterns in sedimentary rocks that
25 document localized pressure solution. Recent studies revealed that stylolite roughness is
26 characterized by two distinct scaling regimes. The main goal of the present study is to
27 decipher whether this complex scaling behavior of stylolites is caused by the composition of
28 the host rock, *i.e.* heterogeneities in the material, or is governed by inherent processes on
29 respective scales, namely the transition from a surface energy to an elastic energy dominated
30 regime, as theoretically predicted. For this purpose we have developed a discrete numerical
31 technique, based on a lattice spring model, to simulate the competition between stress, strain,
32 and dissolution during stylolite roughening. We varied systematically the quenched noise,
33 initially present in the material, which controls the roughening. We also changed the size,
34 amount, and dissolution rate of the heterogeneities introduced in our model and evaluated the
35 influence on the scaling exponents. Our findings demonstrate that the roughness and growth
36 exponents are independent of the exact nature of the heterogeneities. We discovered two
37 coinciding crossover phenomena in space and time that separate length and timescales for
38 which the roughening process is either balanced by surface and elastic energies. Our
39 observations are coherent with analytical predictions and with investigations quantifying
40 precisely the scaling laws in the morphology of natural stylolites. The findings presented here
41 can further be used to refine volume loss estimates from the finite strain pattern of stylolites.

42

43 **1. Introduction**

44 Pressure solution in sedimentary rocks results in either intergranular or localized dissolution
45 of material (e.g. Tada & Siever, 1989). The latter is responsible for the formation of stylolites,
46 a frequent deformation pattern in sedimentary rocks (e. g. Stockdale, 1922; Dunnington 1954,
47 Heald, 1955, Park & Schot, 1968, Buxton & Sibley 1981, Rutter, 1983; Railsback, 1993).
48 Stylolites are rough interfaces that frequently contain insoluble material (Fig. 1), which is

49 considered to be the residuum of the dissolved rock (Railsback, 1993; and references cited
50 therein). Stylolite initiation is still highly debated (e.g. Tada & Siever, 1989) but several
51 mechanisms have been proposed that are in agreement with field observations: Formation (I)
52 along preexisting anisotropies (Bathurst, 1987) (II) as anticracks (Fletcher and Pollard, 1981)
53 that propagate due to stress concentrations at anticrack tips (even though this idea was
54 challenged recently by Katsman et al. 2006) and (III) by stress induced self-organization
55 (Merino, 1992, Railsback, 1998; Merino et al., 2006).

56 In the present study we focus on a quantitative description and characterization of the
57 roughness of simulated stylolites and study their dynamic development independent of the
58 process leading to the initial development of the localization of dissolution along a plane.
59 Based on recent quantitative methods of stylolite roughness characterization (Renard et al.,
60 2004; Schmittbuhl et al., 2004, Koehn et al., 2007, Ebner et al., submitted) we will use
61 statistical tools to compare simulated and natural stylolites. In particular we study the
62 influence of initial heterogeneity concentration in the host-rock on a) stylolite roughness, b)
63 dynamic roughness growth and c) the correlation of crossover phenomena in space and time.
64 To integrate the results of our study in the context of quantitative characterization we will first
65 review some of the major findings and basic principles, used in recent studies, necessary for
66 the understanding of our approach.

67 The exact classification of stylolites in the field is a difficult task because there is a wide
68 range of geometries (e.g. Park and Schott, 1968) that are often transitional even within a
69 single outcrop. Many previous studies (Park & Schot, 1968; Buxton & Sibley, 1981,
70 Guzzetta, 1984; Tada & Siver, 1989; Railsback, 1993) used classification schemes that were
71 based on visual descriptions of macroscopic features of stylolites. These classification
72 schemes are however not quantitative and hard to compare since these studies focused on a
73 variety of different aspects of stylolite formation. Recent studies, however (Drummond &
74 Sexton, 1998, Karcz & Scholz, 2003) took a more quantitative approach using fractal

75 concepts to describe the stylolite roughness in a statistical sense. They could describe stylolite
76 roughness with a fractal scaling over several orders of magnitude, which means that their
77 roughness is not dominated by a certain wavelength.

78 Renard et al. (2004) and Schmittbuhl et al. (2004) went one step further and revealed that
79 bedding parallel stylolite surfaces show a self-affine scaling invariance with characteristic
80 Hurst exponents (also called roughness exponents). A self-affine rough surface is
81 characterized statistically by the fact that points along the surface separated by a distance Δx
82 from each other are typically distant in the direction transverse to the surface by $\Delta h = \Delta x^\alpha$,
83 where α is the roughness exponent. It was further noticed that two distinct scaling regimes
84 exist that were characterized by two different Hurst or roughness exponents separated by a
85 crossover-length (L), around the millimeter scale for the analyzed natural stylolites. Above
86 this crossover, all investigated stylolites exhibit a Hurst exponent of about 0.5 meaning that
87 they change relatively fast from being flat features on larger scale to being rough features on
88 the smaller scale. Below the crossover-length the Hurst exponent is about 1.0, which means
89 that the slopes, or aspect ratio $\Delta z / \Delta x$, stays more or less constant. Schmittbuhl et al. (2004)
90 and Renard et al. (2004) established from first principles of mechanics and chemistry a model
91 for stylolite growth under the form of a stochastic partial differential equation (called in this
92 case a generalized Langevin equation). This equation simulates the roughening of a stylolite
93 surface as a competition between stabilizing forces (that keep the surface flat), which are
94 controlled by long range elastic and local surface tension effects, and destabilizing forces (that
95 roughen the interface) that are induced by pinning effects of material heterogeneities. The
96 analytical solution of Schmittbuhl et al. (2004) reproduced the observed scaling behavior of
97 natural stylolites and demonstrated that the two scaling regimes (characterized by the two
98 different Hurst exponents) correspond to two thermodynamic regimes that are dominated by
99 either surface or elastic energies on small and large scales, respectively (Renard et al., 2004;
100 Schmittbuhl et al., 2004, Gratier et al., 2005). Based on the work of Schmittbuhl et al., (2004)

101 it was demonstrated for the first time by Ebner et al., (submitted) that the crossover-length of
102 natural stylolites, which should be a function of the stress during stylolite growth, can be used
103 to determine stress magnitudes and burial depth in sedimentary basins. The discrete numerical
104 simulation technique of Koehn et al. (2007) enabled to study the dynamics of the roughening
105 process through time revealing that the stylolite interface width w (defined in detail below)
106 grows as a power law with time ($w \sim t^\beta$) with a growth exponent β of 0.5 in the surface energy
107 dominated regime and a growth exponent of 0.8 in the elastic energy dominated regime. In
108 addition the roughness growth may saturate so that the stylolites lose their memory for
109 compaction or finite strain. It is important to notice that the roughness of simulated stylolites
110 in this contribution is produced by heterogeneities in the material that pin the stylolitic
111 interface due to slower dissolution rate constants, which are in competition with the surface
112 and elastic energies which tend to flatten the surface (Koehn et al., 2007). Therefore the
113 obvious question to ask is whether a variation of the quenched noise changes the scaling
114 properties of the stylolitic interface?

115 Thus, in the present contribution we investigate the influence of different heterogeneities
116 (namely the percentage of pinning particles, their pinning factor (defined below), and their
117 size) on the scaling behavior, dynamic growth, and determined crossover length of simulated
118 stylolites.

119

120 **2. Setup numerical model**

121 The numerical technique that we use to simulate stylolite roughening is based on a lattice-
122 spring model coupled with a dissolution routine (Koehn et al., 2004, 2006, 2007). The model
123 itself is embedded as a module in the “*Elle*” modeling-platform (Bons et al., 2008).

124 For computational reasons, to access large systems and analyze scaling laws over a large ratio
125 between the system size and the resolution, we will consider situations spatially invariant
126 along one of the directions tangential to the stylolite – and effectively treat systems with two

127 spatial dimensions. The initial configuration of this 2D model, as shown in Fig. 2a, contains a
 128 predefined flat interface that is considered to be filled with a confined fluid. Two blocks of
 129 particles are separated by a fluid pocket. Such an approximate configuration would be
 130 expected for example, for a fluid pocket embedded between two lowly permeable
 131 sedimentary layers. This model system represents two solids or rocks that are pressed together
 132 by inward moving top and bottom boundaries, whereas the side boundaries remain fixed
 133 (uniaxial strain). A quenched noise (denoted by darker particles in Fig 2a & b) is introduced
 134 by a change of the dissolution rate constant of a certain fraction of the particles (= pinning
 135 particles) and represents material heterogeneities initially present in the host rock of natural
 136 stylolites.

137

138 *2.1. Theory*

139 This section provides only a cursory review on the governing equations of the dissolution
 140 process used in the model, for a detailed description and implementation the reader is referred
 141 to Koehn et al., (2007) and Bons et al., (2008).

142 The pressure solution process is discretized in steps of dissolution of entire particles,
 143 following a linear rate law (Koehn et al., 2007 and references cited therein) according to

$$144 \quad D = kV \left[1 - \exp\left(-\frac{\{\Delta\psi + \Delta\sigma_n\}V}{RT}\right) \right], \quad (1)$$

145 where D is the dissolution velocity of the interface (m s^{-1}), k a dissolution kinetics rate
 146 constant ($\text{mol m}^{-2} \text{s}^{-1}$), V the molecular volume of the solid ($\text{m}^3 \text{mol}^{-1}$), R the universal gas
 147 constant ($8.314 \text{ J mol}^{-1} \text{ }^\circ\text{K}^{-1}$), T the temperature ($^\circ\text{K}$), $\Delta\psi$ (Pa) the changes in Helmholtz free
 148 energy density (which accounts for the variations in elastic and surface energies) of the solid
 149 during dissolution of a solid element, and $\Delta\sigma_n$ (Pa) the differences between the average
 150 normal stress along the interface and the local normal stress at a specific location which is due
 151 to the repulsion of the solids (Koehn et al., 2007).

152 Surface energies (E^S) of particles are calculated from the local curvature of the interface
153 around each particle, which can be expressed as

154
$$E^s = \frac{\gamma}{\rho}, \quad (2)$$

155 where γ is the surface free energy and ρ is the local radius of curvature of the interface. We
156 consider a plane strain situation, i.e. an invariance along the third spatial dimension, so one
157 radius along the 2D plane investigated entirely characterizes the curvature of the interface –
158 the radius of curvature along the direction of invariance is infinite, and no surface energy is
159 associated to this direction. The surface energies of individual particles are averaged over
160 their neighbors to avoid artifacts from the discreteness of the model (for details see Koehn et
161 al., 2007).

162 In the lattice spring model every particle (i) is connected to its neighbors (j) via a triangular
163 linear elastic spring network. The elastic energy (E^{el}) of a single element is given by

164
$$E^{el} = \frac{1}{4} \sum_{(j)} \kappa (|x_i - x_j| - l)^2, \quad (3)$$

165 where the sum is over all neighbors (j), κ is a spring constant, l is the equilibrium distance
166 between elements i and j .

167

168 *2.2. Basic numerical step*

169 The constitutive equations stated above are implemented as follows:

170 • Top and bottom walls are moved inwards simultaneously at a given time/deformation
171 step.

172 • For every deformation step the rate law (Eq. 1) is used to calculate if individual
173 particles at the interface can dissolve in the given time as soon as the two solids meet.

174 • When elements dissolve they are removed completely and the system can relax.

175 Relaxation is accomplished by an over-relaxation algorithm that finds the new

176 equilibrium configuration for the lattice. Dissolution of particles can take place as long
177 as the given time for the individual deformation step is not consumed.

178 • If the deformation time is used up or no particles can dissolve within the given time
179 the system is stressed again by a deformation step.

180

181 *2.3. Parameters, boundary conditions, limitations*

182 The material parameters we use resemble those of a limestone, these values are analog to
183 those found in reference works (e.g. Clark, 1966) and in line with values used for the
184 analytical solutions of Renard et al. (2004) and Schmittbuhl et al. (2004): a molar volume of
185 $0.00004 \text{ m}^3/\text{mol}$, a Young's Modulus of 80 GPa, a Poisson's ratio of 0.33 (this number is
186 given by the triangular lattice configuration), a surface free energy of 0.27 J/m^2 , a temperature
187 of 300 K and a dissolution rate constant of $0.0001 \text{ mol}/(\text{m}^2\text{-s})$. In addition, the displacement
188 rate of the upper and lower boundaries are fixed at a constant value corresponding to
189 velocities between 10^{-10} and $10^{-12} \text{ m s}^{-1}$.

190 The boundary condition can be seen as equivalent to a constant load boundary condition since
191 the dissolution process is fast enough to relax the stresses that build up during a single
192 deformation step. The sidewalls remain fixed during the model runs and there is no wrapping
193 of particles in the x-direction of the model.

194 We use three basic model setups for which we systematically vary the heterogeneities in the
195 structure. All boxes used have the same number of particles (400 particles) in the x-direction
196 but three different particles-sizes were used 0.01 mm, 0.1 mm and 1mm, which corresponds
197 to absolute box-sizes of 4, 40 and 400 mm.

198 To introduce the quenched noise in the simulations a pseudorandom algorithm is used to
199 create a spatial Gaussian distribution of particles that dissolve slower (pinning particles). We
200 varied three parameters of the quenched noise in this study: (i) number of pinning particles in
201 a range from 1-20%, (ii) dissolution rate constant of pinning particles (from 0.1-0.99

202 normalized to the dissolution rate constant of the matrix, which is 1), which determines the
203 pinning factor and (iii) the absolute size of the heterogeneities which varies with the particle
204 size in the range of 0.01-1 mm.

205

206 **3. Data analysis & results**

207 The individual model runs are grouped with respect to the particle-size of the model and are
208 termed *surface*, *intermediate* and *elastic class* according to the dominance of the energy
209 regime during the roughening process. The *surface class* has a particle size of 0.01 mm (box
210 size of 4 mm), the *intermediate class* a particle size of 0.1 mm (box size of 4 cm) and the
211 *elastic class* a particle size of 1mm (box size of 40 cm). In nature the particles may resemble
212 actual grains so that the grain size of the rock varies between the different classes.

213 Unfortunately we are restricted by the resolution of the numerical model to 400 particles in
214 the x-direction, with this resolution a single simulation may run from 10 to 15 days on 4 cores
215 of a recent workstation. Figure 3 shows the roughening of stylolites of these three classes,
216 each with identical quenched noise (5 % pinning particles with half the dissolution rate of the
217 matrix) to demonstrate the influence of the absolute box/particle size. The differences in the
218 roughness and the roughness growth can easily be seen when individual steps of different
219 classes are compared but also by following the growth of individual stylolite peaks along the
220 time axis. In the *elastic class* individual peaks are growing very persistently whereas in the
221 *surface class* the growth is often disrupted, due to dissolution of pinning particles as a result
222 of high surface energies along pronounced peaks.

223 In the following sections we concentrate on the influence of the noise (amount and pinning
224 factor of the noise particles) on 1) the roughness exponents, 2) the growth exponents and 3)
225 the crossover length.

226

227 *3.1. Roughness exponents*

228 To quantitatively characterize the roughness of an individual 1D profile of a stylolite we used
 229 concepts from statistical physics (Barabasi & Stanley, 1995), which are briefly introduced in
 230 the first part of this section. The methods used here are the same as those of previous studies
 231 of natural stylolites (Renard et al., 2004; Schmittbuhl et al., 2004; Gratier et al., 2005; Ebner
 232 et al., submitted), which facilitates comparison.

233 The prerequisite for the application of these scaling methods is that the 1D signal of the
 234 numerical stylolite obeys a self-affine scaling invariance, which is given by (e.g. Barabasi &
 235 Stanley, 1995)

$$236 \quad f(bx) \sim b^\alpha f(x), \quad (4)$$

237

238 where $f(x)$ is a single valued function and the power-law exponent α is called *roughness* or
 239 *Hurst* exponent and provides a quantitative measurement of the roughness of the signal. A
 240 self-affine function must be rescaled differently in x and y directions to obtain a scaling
 241 invariance i.e. horizontal rescaling of the form $x \rightarrow bx$, b being a dilation factor, has to be
 242 rescaled in the vertical direction by $y \rightarrow b^{-\alpha}y$ to obtain a scaling invariance. Different statistical
 243 methods can be used to evaluate the self-affine character of a signal and to determine the
 244 associated roughness exponent. We apply two independent methods in this contribution, the
 245 *Fourier method* and the *Average Wavelet Coefficient method*.

246 The *Fourier method* (e.g. Barabasi & Stanley, 1995; Schmittbuhl et al., 1995) is based on a
 247 Fourier transform of the original 1D signal (Fig. 4a). For every 1D signal (every deformation
 248 step) the Fourier power spectrum $P(k)$ i.e., the square of the modulus of the Fourier transform,
 249 was calculated as a function of the wave-number k . Plotting $P(k)$ as a function of k in log-log
 250 space reveals a linear trend for a self-affine function (Fig. 4b), and the slope is a function of
 251 the Hurst exponent through (Renard et al., 2004; Schmittbuhl et al., 2004):

$$252 \quad P(k) \sim k^{-1-2\alpha} . \quad (5)$$

253 The *Average wavelet coefficient method* (*AWC*) was used as a second independent method to
 254 confirm the scaling results (Simonsen et al., 1998; Hansen et al., 2000). This method is again
 255 based on a decomposition of the 1D signal into wavelets, whose amplitude depends on scale
 256 and the position. The wavelet transform is defined after Simonsen et al. (1998) by

$$257 \quad W_{a,b} = \frac{1}{\sqrt{a}} \int_{-\infty}^{\infty} \psi\left(\frac{x-b}{a}\right) f(x) dx, \quad (6)$$

258 where ψ is the wavelet basis (Daubechies wavelet of order 12) which is parameterized by a
 259 scale parameter a and a translation parameter b , and f is the single-valued original function.
 260 Finally the wavelet coefficients are averaged over the translation parameter b for every a to
 261 obtain the average wavelet coefficient $W(a)$. If the input signal is self-affine, the wavelet
 262 transform verifies that the average wavelet coefficient $W(a)$ scales as (Simonsen et al., 1998)

$$263 \quad W(a) \sim a^{\alpha+1/2}. \quad (7)$$

264 Plotting the average wavelet coefficients as a function of the scale parameter a in log-log
 265 space (Fig. 4c), the slope of the linear regression through the data is again a function of the
 266 Hurst exponent.

267 Using these two statistical methods, we first study the dynamics of the roughness exponents
 268 through time during stylolite growth, and then concentrate on their stability with respect to
 269 variations of the noise. The roughness exponents increase relatively quickly in the course of a
 270 simulation run (Fig. 4d) and become stable after model step 3000 with only minor
 271 fluctuations. The *Fourier-* as well as the *AWC-method* show consistent evolutions and similar
 272 values of the roughness exponents. Averages of the plateau values reached (after step 3000)
 273 for individual model runs are used as a characteristic value for the roughness exponent for a
 274 specific setup. Error bars underline the standard deviation around this average (Fig. 5).

275 The *surface class* is characterized by consistently high values for the Hurst exponent i.e. $\alpha \sim 1$ -
 276 1.1, independent of the pinning factor (i.e. the dissolution rate constant k in Eq 1; cp Fig. 5a)
 277 or the amount of pinning particles (Fig. 5b).

278 The exponents only decrease when the pinning factor of particles is very low (dissolution rate
279 constant > 0.9) and the rock becomes very homogeneous. The most stable roughness
280 exponents for the surface class are reached in the range 0.1 to 0.8 for the pinning factor and 1
281 to 20 % of pinning particles.

282 The *elastic class* reveals lower Hurst exponents ($\alpha \sim 0.6$ to 0.9) than the *surface class*. If the
283 pinning factor of particles is very strong (below the value 0.4 for the relative dissolution rate
284 constant in Fig. 5a) stress concentrations are locally too high once two pinning particles meet
285 and artifacts develop (usually anticracks that grow laterally emerge from these concentrations,
286 hence modifying the surface topography) in the numerical model within the *elastic class*.
287 Therefore we did not include values below 0.4 from elastic simulations in Fig. 5a. Generally
288 the roughness exponents in the elastic class show stronger fluctuations than those of the
289 surface class. They are relatively stable within a pinning factor range of 0.5 to 0.8 (Fig. 5a)
290 and 1 to 20 % of pinning particles.

291 The *surface* and *elastic classes* correspond well to the two scaling regimes found in natural
292 stylolites (Renard et al., 2004; Schmittbuhl et al., 2004) that are separated by a crossover-
293 length at the millimeter scale. The roughness exponents of the surface class ($\alpha \sim 1.1$) are in
294 very good agreement with analytical predictions and experimental observations (e.g. Gratier
295 et al., 2005). The *elastic class* displays values for the Hurst exponent ($\alpha \sim 0.6-0.9$) that are
296 higher than exponents from natural examples (Renard et al., 2004; Schmittbuhl et al., 2004;
297 Ebner et al., submitted) or analytical predictions, which are generally around 0.5. These
298 analytical predictions are usually based on linear approximations, which are strictly speaking
299 valid as long as the surface morphology is not too developed. The present model does not
300 present any such limitations, and the fully developed situation can thus present a different
301 Hurst exponent from the initial one. The discrepancy with natural data may arise from the
302 large particle (or grain) size that we use in the setup for the elastic class. In nature the grain
303 size is much smaller and corresponds to the values that we use in the surface class.

304

305 3.2. Interface growth

306 In addition to the dynamic development of the roughness exponents the simulations allow us
307 to study how fast the amplitude of the stylolite roughness grows through (model-) time or as a
308 function of the finite strain. First we concentrate on different growth regimes of stylolites, the
309 associated growth exponents and prefactors of scaling functions and then study the variation
310 of these factors as a function of host-rock heterogeneities. In order to quantify the amplitude
311 of the roughness we use the *interface width* (w) that is defined as the root-mean-square
312 fluctuation of the height of the interface for a given time-step (Barabasi & Stanley, 1995)

$$313 \quad w(L,t) \equiv \sqrt{\frac{1}{L} \sum_{i=1}^L [h(i,t) - \bar{h}(t)]^2}, \quad (8)$$

314 where w is the interface width as a function of system size L and time t , h is the height of
315 point i on the interface at time t and \bar{h} the average height of the interface at time t given by

$$316 \quad \bar{h}(t) \equiv \frac{1}{L} \sum_{i=1}^L h(i,t). \quad (9)$$

317 In our simulations, the system size L is defined as the number of elements in the x-direction,
318 which is constant for all simulations, i. e. 400 particles. Roughening processes of interfaces in
319 a wide range of fields have been demonstrated to follow a power law in time (e.g. Barabasi &
320 Stanley, 1995) defined by a growth-exponent β (given by, $w(L,t) \sim t^\beta$). This initial phase of
321 interface growth is usually followed by a second regime during which the interface width
322 reaches a saturation value, w_{sat} , which is directly related to the system-size.

323 Both, the growth and saturation regimes can be seen in Fig. 6a for an experiment of the
324 surface class with a characteristic growth exponent of $\beta \sim 0.5$. The arrow in Figure 6a marks
325 the transition from the power law growth regime to the regime where the interface width
326 saturates and stays constant. The *intermediate class* simulations show a similar growth
327 exponent (around 0.5) but do not saturate in the given deformation time. The *elastic class*

328 (Fig. 6b) shows two successive growth regimes, the first being defined by a growth exponent
 329 of $\beta \sim 0.5$ up to a crossover interface width followed by a second regime with $\beta \sim 0.8$ without
 330 reaching the saturation regime. We suggest the following schematic growth regimes (Fig 6c)
 331 for stylolites: (i) growth in the surface energy dominated regime with an exponential growth
 332 defined by $\beta \sim 0.5$ followed by (ii) growth in the elastic energy dominated regime with $\beta \sim 0.8$
 333 and finally reaching (iii) a saturation regime where the interface width stays constant.

334 This strict non-linearity of the interface growth in our simulations suggests that estimated
 335 amounts of compaction (here used as synonymous with volume/area loss due to pressure
 336 solution) from stylolite amplitude heights (e.g. Tada & Siever, 1989 and references cited
 337 therein) only capture a small part of the actual compaction. To cope with this problem, Koehn
 338 et al. (2007) demonstrated that the actual displacement can be expressed for the elastic or
 339 surface energy dominated growth regimes as a function of the interface width and the growth
 340 exponent (as long as the critical saturation time is not reached), given by

$$341 \quad A \sim (w/l)^{1/\beta} l \quad (10)$$

342 where, A is the compaction displacement, w the interface width, β the growth exponent (for a
 343 certain class) and l the particle size. The slope of this function gives a prefactor for the scaling
 344 relation which should remain constant until the saturation time is reached. We call the slope
 345 of this relation here and in the subsequent sections *compaction prefactor* because it relates the
 346 interface width to the total compaction (Fig. 7). As soon as the saturation time is reached the
 347 relation does not hold any more and the function deviates from the linear trend (Fig. 7a). This
 348 effect can be observed in Figure 7a where the arrow marks the onset of the saturation of the
 349 interface width, compaction goes on but the interface width (x-axis) remains constant.
 350 However, if the saturation is not attained (Fig. 7b) the actual compaction can be calculated
 351 accurately from the interface width and the growth exponent using Eq. 9.

352 Looking at the growth exponent as a function of the quenched noise it can be clearly
 353 demonstrated that neither a variation in the pinning factor of particles (Fig. 8a) nor in the

354 amount of pinning particles (Fig. 8b) has a significant influence. The values for the growth
 355 exponent cluster around $\beta \sim 0.5$ for the *surface* and *intermediate classes* and around $\beta \sim 0.8$ for
 356 the *elastic class*. The compaction prefactors display a higher variability than the growth
 357 exponents but no systematic trend can be seen that relates this variation to the pinning factor
 358 or amount of pinning particles in the host-rock. Figures 8c & 8d show that there is no
 359 significant difference between the three classes of particle sizes used with values for the
 360 compaction prefactor in a range between ~ 12 and ~ 25 . As a third quantity we compare the
 361 maximum interface width normalized by the particle size (w_{max}) that develops during
 362 simulations with different heterogeneities (Fig. 8e & f). The largest interface widths are
 363 achieved in the *elastic class* with $w_{max} \sim 20$ in contrast to $w_{max} \sim 10$ reached in the *intermediate*
 364 and *surface class*. Hence the interface growth displays twice the displacement in the *elastic*
 365 *class* in the given simulation time due to the larger growth exponents than those of the
 366 *intermediate* or *surface class*, respectively. For the *surface*, *intermediate* and *elastic classes*
 367 the variation of the pinning factor of particles (Fig. 8e) have no considerable influence on the
 368 maximum interface width. However the amount of pinning particles has a significant
 369 influence on the *surface* and *intermediate class* (Fig. 8f). Both classes show an evident
 370 decrease in the maximum interface width with increasing amount of pinning particles. This
 371 trend cannot be observed in the *elastic class* (Fig. 8f).

372 We also tested the influence of the initial shape of the predefined interface separating the two
 373 blocks that are pressed together during an experimental run (compare Fig. 2a), which is flat in
 374 all the simulation data shown in the preceding sections. To investigate the dynamic roughness
 375 evolution of an already rough interface we arbitrarily choose a time/deformation step (t_n) of a
 376 simulation run (Fig 9a) and subtracted the topography (h) of this step from the subsequent
 377 time steps similar to $h(t) = h(t_n + m \cdot \Delta t) - h(t_n)$. This procedure allows to investigate the dynamic
 378 evolution of a rough interface but statistically evaluating the difference of the evolving
 379 roughness from time t_n onwards (Fig 9b). Departing from an already rough interface does not

380 change the scaling parameters (growth and roughness exponent) as depicted in Fig 9. Hence
 381 the model setup we choose in this work (i.e. a flat initial interface) can also account for
 382 complex initial topographies.

383

384 3.3. Crossover length scales

385 The crossover-length of the stylolite roughness that separates the surface energy regime from
 386 the elastic energy dominated regime is a function of the stress during stylolite growth and can
 387 be used as paleo-stress gauge. It is of fundamental importance to know if this crossover is
 388 constant when the heterogeneities in the host-rock vary. In the following section we want to
 389 explore how sensitive the crossover-length scale is on variations in the amount and strength of
 390 pinning particles. The *surface* and *elastic classes* of our simulations reveal roughness
 391 exponents that are characteristic for the two regimes where surface or elastic energy are
 392 dominant. The crossover-length between the two regimes can be found in the *intermediate*
 393 *class* of our simulations that reveals two different roughness exponents and hence the
 394 transition between the two scaling regimes (Fig. 10). The Fourier power spectrum of the 1D
 395 signal of a stylolite in the intermediate class (Fig. 10b) shows a change from a shallow to a
 396 steep slope, i.e. small and large roughness exponents on large and small scales, respectively.
 397 To avoid bias due to improper fitting of the crossover-length we used a nonlinear least square
 398 curve fitting algorithm in logarithmic space to model our scaling function (Ebner et al.,
 399 submitted)

$$400 f(x) = (a_L x + m_L)(1 - w(x)) + (a_S x + m_S)w(x) \quad (11a)$$

401 and

$$402 w(x) = \frac{(\tanh(x + L) + 1)}{2}, \quad (11b)$$

403 where $a_{L,S}$ are the exponents of the scaling function for large and small scales, $m_{L,S}$ the
 404 corresponding intercepts with the ordinate and $w(x)$ the weighting function. During this

405 procedure the roughness exponents of our nonlinear model function were fixed according to
406 the roughness exponents ($\alpha_S \sim 1.1$; $\alpha_L \sim 0.5$) reported from natural stylolites (Renard et al., 2004;
407 Schmittbuhl et al., 2004, Ebner et al., submitted).

408 The crossover-lengths obtained for all experiments of the intermediate class are in a range of
409 $L \sim 1.33 \pm 0.09$ mm, the crossover length usually develops simultaneously with the achievement
410 of the plateau values (compare Fig. 4d). Neither of the quenched noise parameters varied in
411 the experiments influences the crossover-length (Fig. 11).

412 The time evolution of the roughness presented in the previous section showed that the surface
413 energy dominated regime is characterized by a growth exponent of 0.5 whereas the elastic
414 energy dominated regime is characterized by a growth exponent of 0.8. Looking at the
415 roughness growth in the elastic class one could still see the slow surface energy dominated
416 growth in the beginning of the roughness evolution (Fig. 6). Therefore the growth exponents
417 also show a transition (that we term the crossover interface width) between growth in the
418 surface energy dominated regime and growth in the elastic energy dominated regime, similar
419 to the two roughness exponents that are characteristic for these two regimes. The crossover
420 interface width is very consistent for all experiments with $w \sim 1.23 \pm 0.04$ mm, independent of
421 the quenched noise introduced in the system. We did not observe a crossover in the interface
422 growth of the *intermediate* and *surface class* because the interface width in these classes is
423 simply not large enough to reach the elastic growth regime. Due to the very good correlation
424 between the magnitudes of the crossover-length and the crossover interface width (Fig. 11)
425 we argue that both crossovers arise from the same process, namely the transition from a
426 surface energy to an elastic energy dominated regime.

427

428 **4. Discussion**

429 In the following section we first discuss the influence of the quenched noise on the scaling
430 parameters in our numerical simulations, deal with the relevance of the noise and compare the

431 results to natural stylolites. Secondly, we will focus on the crossover phenomena and their
432 significance for the estimate of volume loss along a finite natural stylolite.

433 The influence of the exact nature of the heterogeneities (i.e. pinning factor, amount and size
434 of pinning particles) on the scaling exponents can be directly investigated by a close
435 examination of Figures 5, 8 and 11. Doing so, it is immediately evident that the pinning factor
436 has the least influence on the roughness and growth exponents as well as on the crossover
437 scaling (L and w_{cross}), with resulting values that are very consistent over wide ranges of the
438 parameter space. The amount of pinning particles only shows an influence on the maximum
439 interface width w_{max} , which decreases with increasing amount of heterogeneities (Fig. 8f).

440 This fact is in good agreement with observations made on natural stylolites that stylolite
441 amplitudes decrease with the amount of heterogeneities i.e. clay particles (e.g. Tada & Siever,
442 1989, and references cited therein). The scaling exponents themselves are independent of the
443 amount of pinning particles. The biggest influence exists between the different experiment
444 classes i.e. particle sizes, which reveal different roughness and growth exponents that are on
445 the other hand very consistent within an individual class. But it has been demonstrated
446 (Renard et al., 2004; Schmittbuhl et al., 2004, Koehn et al., 2007), that these differences arise
447 from a transition from a surface energy dominated regime (where surface energies smoothen
448 the interface) to an elastic energy dominated regime (where elastic energies smoothen the
449 interface). The differences in the scaling exponents between the experimental classes are
450 therefore not caused by the heterogeneities themselves but by the processes that govern the
451 roughening on respective scales. We are thus convinced that the influence of the exact nature
452 of the heterogeneities plays a minor role and that roughening is dominated by an inherent
453 process that depends on the length-scale.

454 The quenched noise we introduced in the different simulations, i.e. changes in the dissolution
455 rate constant that influence the dissolution velocity of a particle (cp. Eq. 1), is a simple
456 chemical noise as pointed out by Koehn et al. (2007). We are aware that along natural

457 stylolitic interfaces the elastic parameters, surface energies, crystallographic orientations are
458 very likely to change, in addition to chemical variations. However the effects of a change of
459 these other parameters in our model also result in a change of the dissolution velocity. We
460 therefore argue that for the developing structure it should make no difference what the exact
461 nature of the noise is, since any particle with a slower dissolution velocity will pin the surface
462 and therefore cause a roughening of the interface.

463 It was demonstrated that individual natural stylolites from different outcrop localities and
464 lithologies, i.e. different host-rock compositions, reveal the same scaling behavior (e.g.
465 Renard et al., 2004; Schmittbuhl et al., 2004, Ebner et al., submitted). Consequently the
466 investigation of natural stylolites corroborates the fact that a common underlying mechanism
467 for stylolite roughening can be assumed rather than a roughening that is dominated by the
468 composition of the host rock.

469 Nevertheless we do not claim that knowledge of the exact nature or distribution of the
470 material heterogeneities is unimportant. Brouste et al., (2007) have shown that a changing
471 amount of heterogeneities might cause a stylolite to become a non-stationary signal with
472 alternating wavy and flat portions along the interface. We have not investigated the effects of
473 irregularly distributed noise since the heterogeneities are distributed equally in our model
474 setup.

475 The roughness data of simulated stylolites presented in this study reveal two self-affine
476 scaling regimes that are separated by a distinct crossover-length of $L \sim 1.3$ mm, which is well
477 in line with investigations of natural stylolites (Renard et al., 2004; Schmittbuhl et al., 2004,
478 Ebner et al., submitted). Additionally we have detected a crossover in the growth of the
479 interface during which the initial growth exponent of $\beta \sim 0.5$ up to a crossover interface width
480 of $w \sim 1.23$ mm is replaced by a growth regime with an exponent of $\beta \sim 0.8$. Due to the very
481 good correlation between the magnitudes of the crossover-length and the crossover interface
482 width we argue that both crossovers arise from the same process, namely the transitions from

483 a surface energy to an elastic energy dominated regime. The knowledge of the crossover
484 length L which can be derived from the finite pattern of a natural stylolite with the above
485 methods is thus equivalent to knowledge of the crossover interface width. This fact has
486 important consequences regarding the assessment of the amount of total compaction of
487 individual stylolites. Substituting the growth exponents and the compaction prefactors found
488 for the two growth regimes in combination with the crossover-length, which separates the two
489 growth regimes (cp. Fig. 10c), into Eq. 10 should allow an exact reconstruction of the amount
490 of total compaction from finite pattern of a natural stylolites.

491

492 **5. Conclusions**

493 In the course of this contribution we evaluated the scaling properties of simulated stylolites,
494 which facilitate a quantitative comparison with natural examples, exactly reproducing their
495 scaling. We observed only minor correlation between the exact nature of the noise introduced
496 in the model or the topography of the predefined interface and the scaling parameters
497 investigated, concluding that inherent processes i.e. the transition from a surface to an elastic
498 energy dominated regime control the roughening process.

499 Nevertheless the amount of heterogeneities has a negative effect on the maximum interface
500 width (w_{max}) achieved during deformation revealing increasing interface width with
501 decreasing amount of quenched noise. The absolute particle/noise size influences the
502 roughness and growth exponents, which in turn is caused by the transition from a surface to
503 an elastic energy dominated regime. Therefore it is important to know how large the noise or
504 pinning particles are in natural systems. The transition from surface energy as the dominant
505 stabilizing force of the interface to the dominance of elastic energies is the causation for the
506 most significant scaling transitions: (i) the roughness is characterized by two distinct spatial
507 scaling regimes on small and large length scales, respectively; (ii) the interface growth reveals
508 two growth regimes with a growth exponent of $\beta \sim 0.5$ up to a crossover interface width that

509 coincides with the crossover-length L followed by a growth regime with an exponent of $\beta \sim 0.8$
510 that eventually saturates due to finite size effects; (iii) the crossover interface width w
511 coincides with the crossover-length L and thus allows to accurately reconstruct the
512 compaction history of finite stylolite patterns.

513 Our study corroborates the evidence that the simple mechanisms summarized above support
514 analytical predictions and natural observations given in previous studies, and are a convincing
515 causation for the formation of stylolite roughness. However a detailed study on the exact
516 nature and distribution of quenched noise in the host rocks of natural stylolites would shed
517 light on the origin and initiation of these complex structures.

518

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523

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596

597 **Figure captions**

598

599 **Figure 1:** Plane section of a bedding parallel stylolite in a jurassic limestone from Cirque de
600 Navacelles (southern France). The rough interface is accentuated by a thin clay layer that is
601 considered to be the residuum of the dissolved rock mass.

602

603 **Figure 2:** Simplified sketch of the setup of the numerical model (modified after Koehn et al.,
604 2007). The top and bottom walls of the box are moved inwards simultaneously to stress the
605 system and initiate dissolution along the interface. **a)** Initial configuration of the setup
606 showing a flat interface (dashed line). **b)** Configuration after a certain amount of compaction.
607 The interface (dashed line) has developed a distinct roughness, note that the heterogeneities
608 (darker spheres) accumulate along the interface.

609

610 **Figure 3:** Growth of three stylolites with similar heterogeneities but different lengths and
611 discretization resolution. Left panels show 3D plots of the stylolite growth from a flat
612 interface until the end of the experiment. Right panels show 3 individual deformation steps
613 (step 400, 2000 & 4000) corresponding to the solid lines in the 3D plot **a)** surface class (box
614 size=0.4 cm) **b)** intermediate class (box size =4 cm) **c)** elastic class (box size =40 cm); Notice
615 the disrupted growth of the surface class whereas the elastic class exhibits very continuous
616 growth and pronounced peaks and teeth.

617

618 **Figure 4:** Roughness characterization of single deformation steps by means of the Hurst
619 exponent. **a)** 1D profile (deformation step 3000) of the surface class with 5% pinning particles
620 and a dissolution rate constant of $k=0.4$; **b)** Fourier power spectrum $P(k)$ of the signal from

621 Fig. 4a plotted as a function of the wavenumber k . Linear regression (solid line) is calculated
622 from the binned spectra (crosses), the slope of the regression is a function of the Hurst
623 exponent, see section 3.1.1. for detailed description. **c)** Wavelet spectra $W(a)$ plotted (crosses)
624 as a function of the scaling parameter a (see average wavelet coefficient-method in section
625 3.1.1.). The slope of the linear regression (solid line) is again a function of the Hurst
626 exponent. **d)** Evolution of the Hurst exponent in the course of an entire experiment for the
627 Fourier and AWC methods. Stars indicate the Hurst exponent of the 1D signal shown in Fig.
628 4a for the two methods used. Notice the leveling off at a specific Hurst exponent (plateau
629 value) which is characteristic for all simulations.

630

631

632 **Figure 5:** Composite plots showing the roughness data (Hurst exponents) for the surface and
633 elastic classes. **a)** Hurst exponent plotted versus dissolution rate constants k with a fixed
634 amount of pinning particles of 5% (where $k=1$ is the dissolution rate of the matrix). **b)** Hurst
635 exponent plotted versus amount of pinning particles with a dissolution rate constant of $k=0.5$
636 for all experiments. The error bars correspond to the standard deviation given by the
637 fluctuation around the plateau values, compare Fig 4d.

638

639 **Figure 6:** Interface growth depicted by means of the interface width (Eq. 8). **a)** Log-log plot
640 of the interface width as a function of time in the surface class (5% pinning particles &
641 dissolution rate constant of 0.4). Growth exponent $\beta=0.5$ is given by the slope of the linear
642 regression (solid line). Notice the saturation point (indicated by arrow) i.e. interface width
643 remains constant during ongoing deformation. **b)** Log-log plot of the interface width as a
644 function of time in the elastic class (5% pinning particles & dissolution rate constant of 0.4).
645 Notice the two successive growth regimes characterized by an initial growth exponent of
646 $\beta=0.5$ up to a crossover width (indicated by arrow) followed by an exponent of $\beta=0.8$; no

647 saturation can be observed within the given simulation time. **c)** Proposed composite graph of
648 the interface growth of simulated stylolites. Two successive growth regimes separated by a
649 narrow crossover width that are dominated either by surface or elastic energies are followed
650 by a saturation of the interface growth due correlation introduced by finite size effects.

651

652 **Figure 7:** Total compaction displacement expressed as a linear function (solid line) of the
653 interface width and the compaction prefactor (compare Eq. 10). **a)** Surface class experiment
654 (5% pinning particles & dissolution rate constant of $k=0.4$) demonstrates that the relationship
655 is only valid as long as the interface is not saturated i.e. strong deviation from linear trend
656 (compare Fig 6a). **b)** Elastic class experiment (5% pinning particles & dissolution rate
657 constant of 0.4) reveals an accurate reproduction of the linear relationship (solid line) stated in
658 Eq. 10. No saturation was observed in the given simulation time.

659

660 **Figure 8:** Cumulative data for the surface, intermediate and elastic classes for: **a)** Growth
661 exponent versus dissolution rate constant (pinning particles fixed at 5%); **b)** Growth exponent
662 versus amount of pinning particles (dissolution rate constant fixed at $k= 0.5$); **c)** Compaction
663 prefactor versus dissolution rate constant (pinning particles fixed at 5%); **d)** Compaction
664 prefactor versus amount of pinning particles (dissolution rate constant fixed at $k= 0.5$).
665 Maximum interface width normalized by the particle size attained during experimental runs
666 for the *surface*, *intermediate* and *elastic* class **e)** with changing dissolution rate constant (with
667 5% pinning particles); **f)** with changing amount of pinning particles (dissolution rate constant
668 is fixed at 0.5).

669

670 **Figure 9:** The influence of the topography of the initial predefined interface on the dynamic
671 roughness evolution. **a)** Rough initial interface used as starting point to evaluate the interface
672 width evolution on top of this roughness for a *surface class* simulation with 5% pinning

673 particles and a pinning factor of 0.5 (for details see text). **b)** Comparison of the interface
674 width evolution of an initially flat and rough (see Fig 9a) interface. Both interfaces show a
675 similar evolution with growth exponents of 0.54 and 0.51 for the flat and rough initial
676 interface respectively. Notice that both growth and the roughness exponents (not shown) are
677 independent of the initial topography of the predefined interface.

678

679 **Figure 10:** Crossover phenomena in the roughness scaling and interface growth. **a)** 1D signal
680 of an experiment from the intermediate class (5% pinning particles; dissolution rate constant
681 $k=0.4$); **b)** Fourier power spectrum (inset) of the signal and binned spectra (crosses). A
682 nonlinear model function (for explanation see text) used to minimize the original data (solid
683 line) is used to accurately locate the position of the crossover length (triangle) $L=1.27$ mm; **c)**
684 Log-log plot of the interface width versus time of the elastic class (5% pinning particles &
685 dissolution rate constant $k=0.4$). Notice the two successive growth regimes characterized by
686 an initial growth exponent of $\beta=0.5$ up to a crossover width $w=1.24$ mm followed by an
687 exponent of $\beta=0.8$. Notice that both crossover scales correspond to the transition from a
688 surface to an elastic energy dominated regime.

689

690 **Figure 11:** Crossover length L and crossover interface width w plotted as a function of the
691 quenched noise. **a)** Crossover length (circles) calculated from the Fourier power spectrum (cp.
692 Fig. 10b) and crossover interface width (diamonds) for simulations with different dissolution
693 rate constants. **b)** Crossover length (circles) and crossover interface width (diamonds) for
694 simulations with different amounts of pinning particles.

695

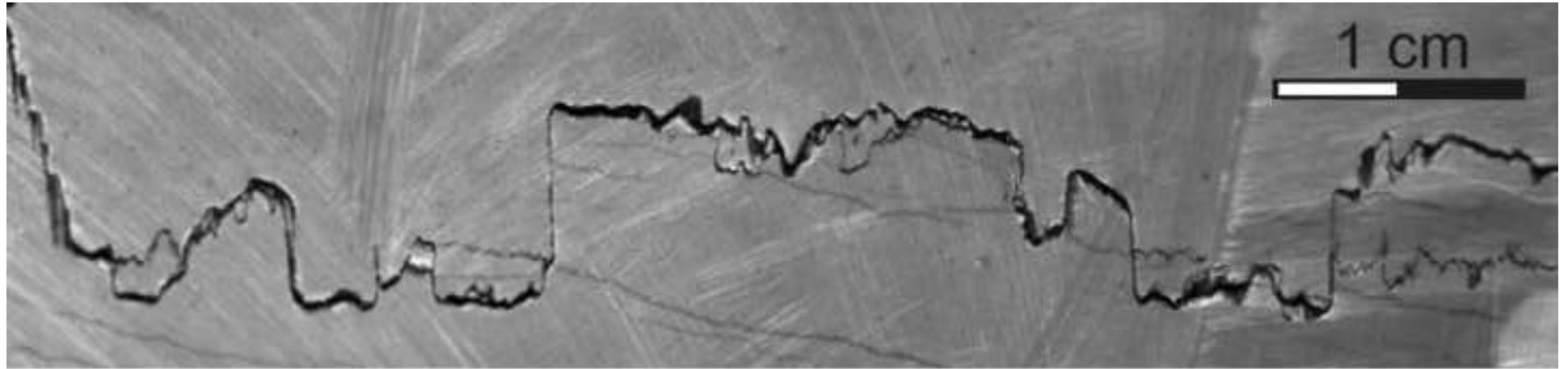


Figure 1

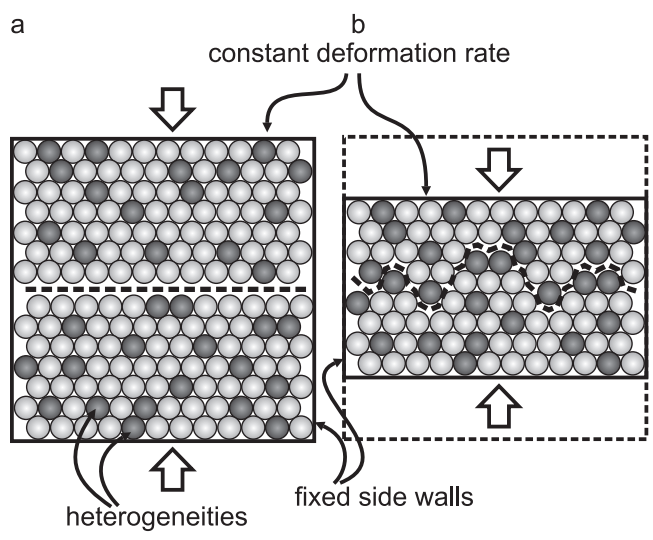


Figure 2

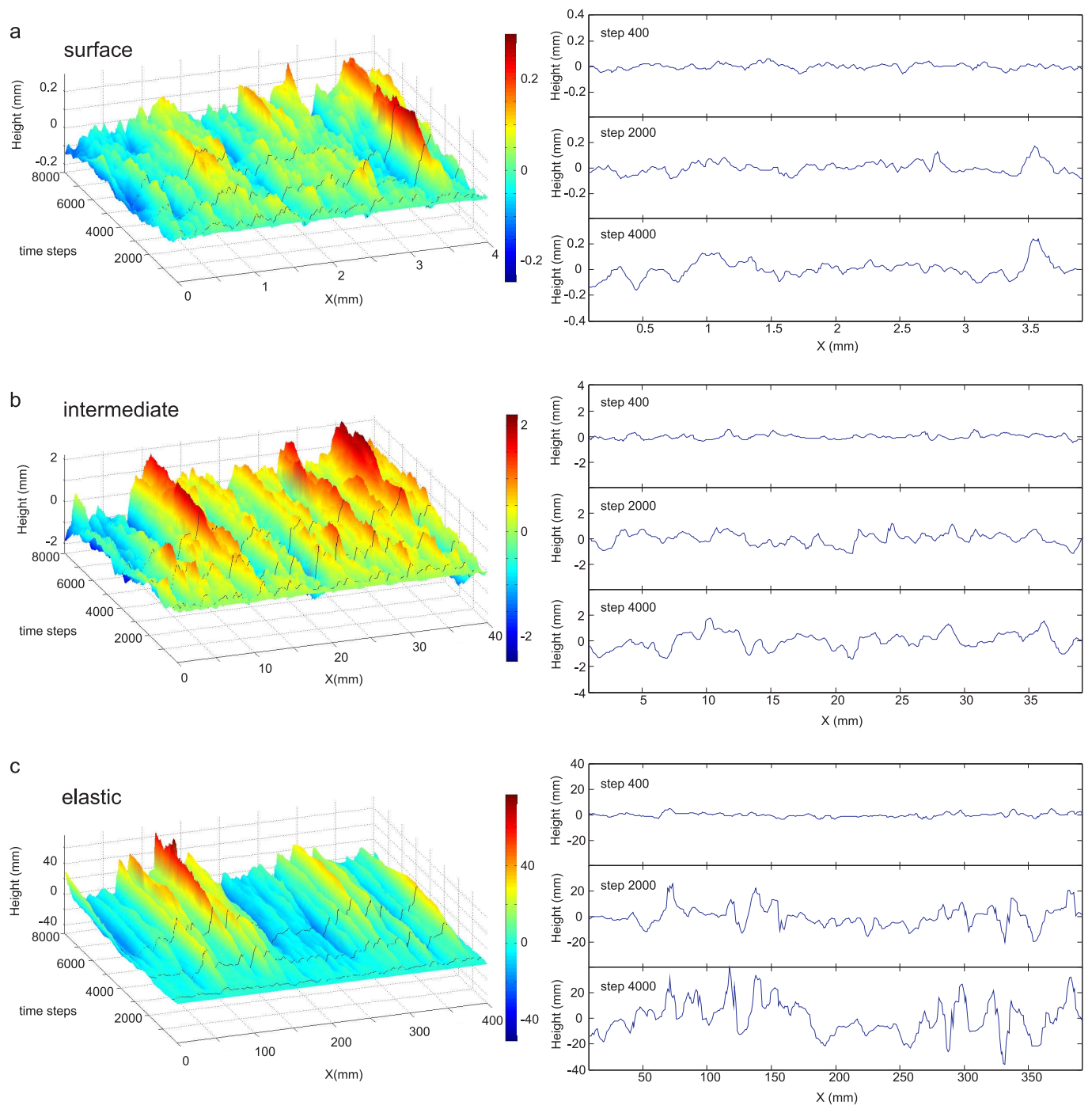


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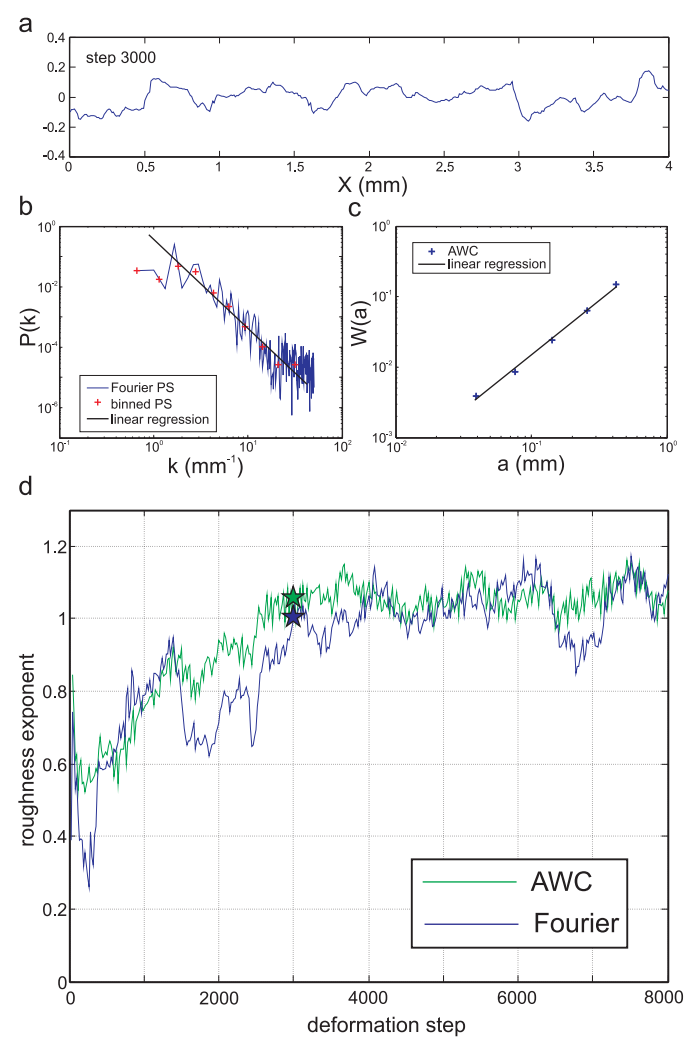


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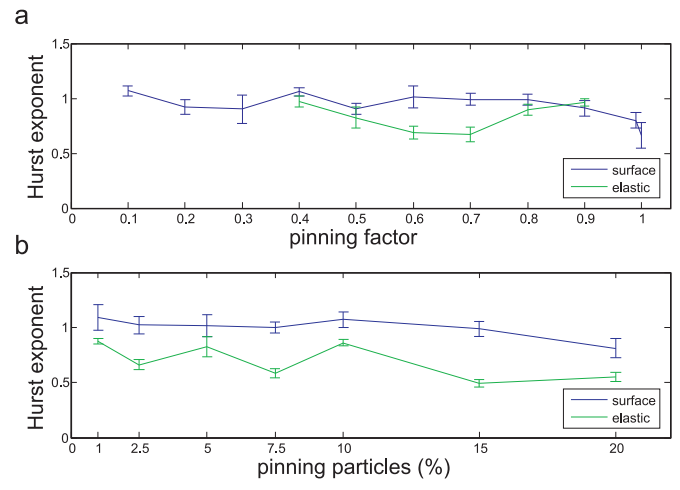


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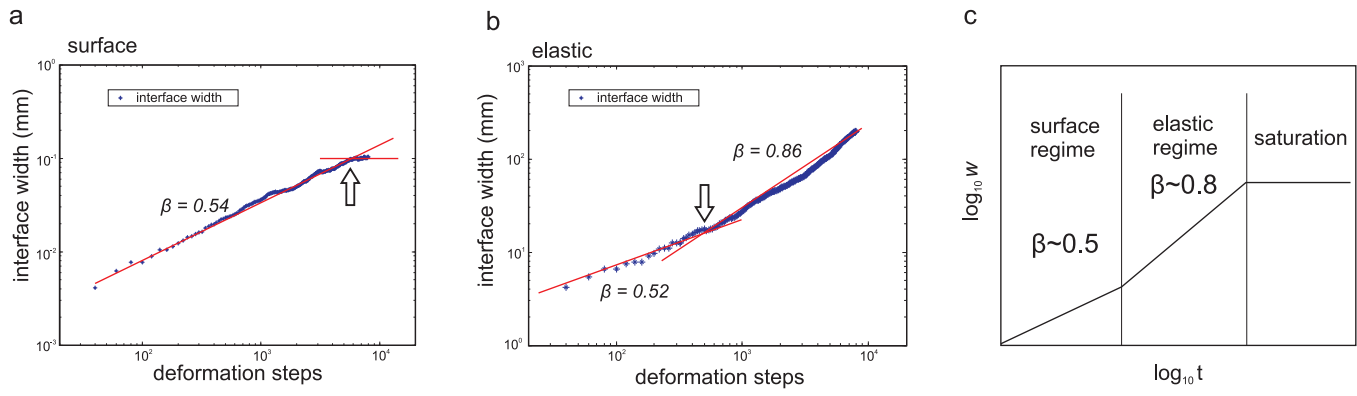


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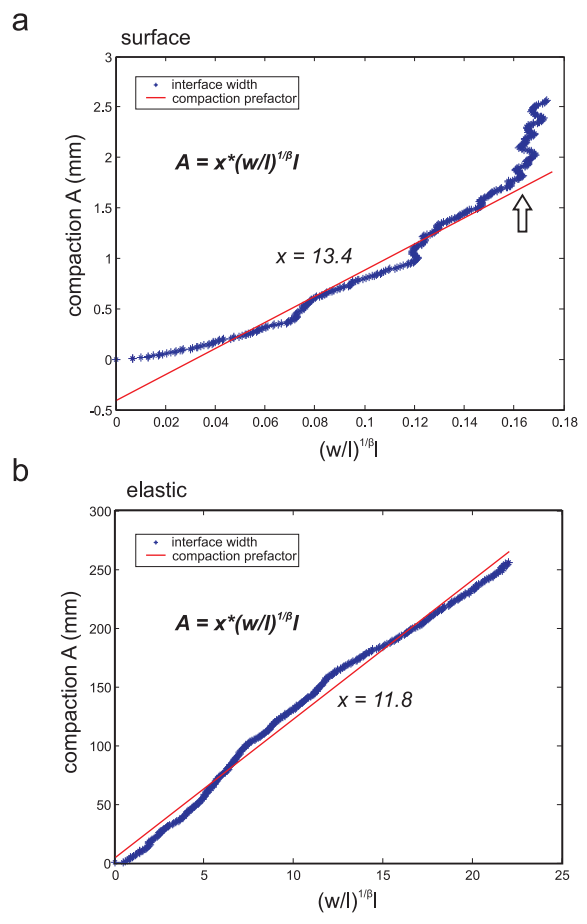


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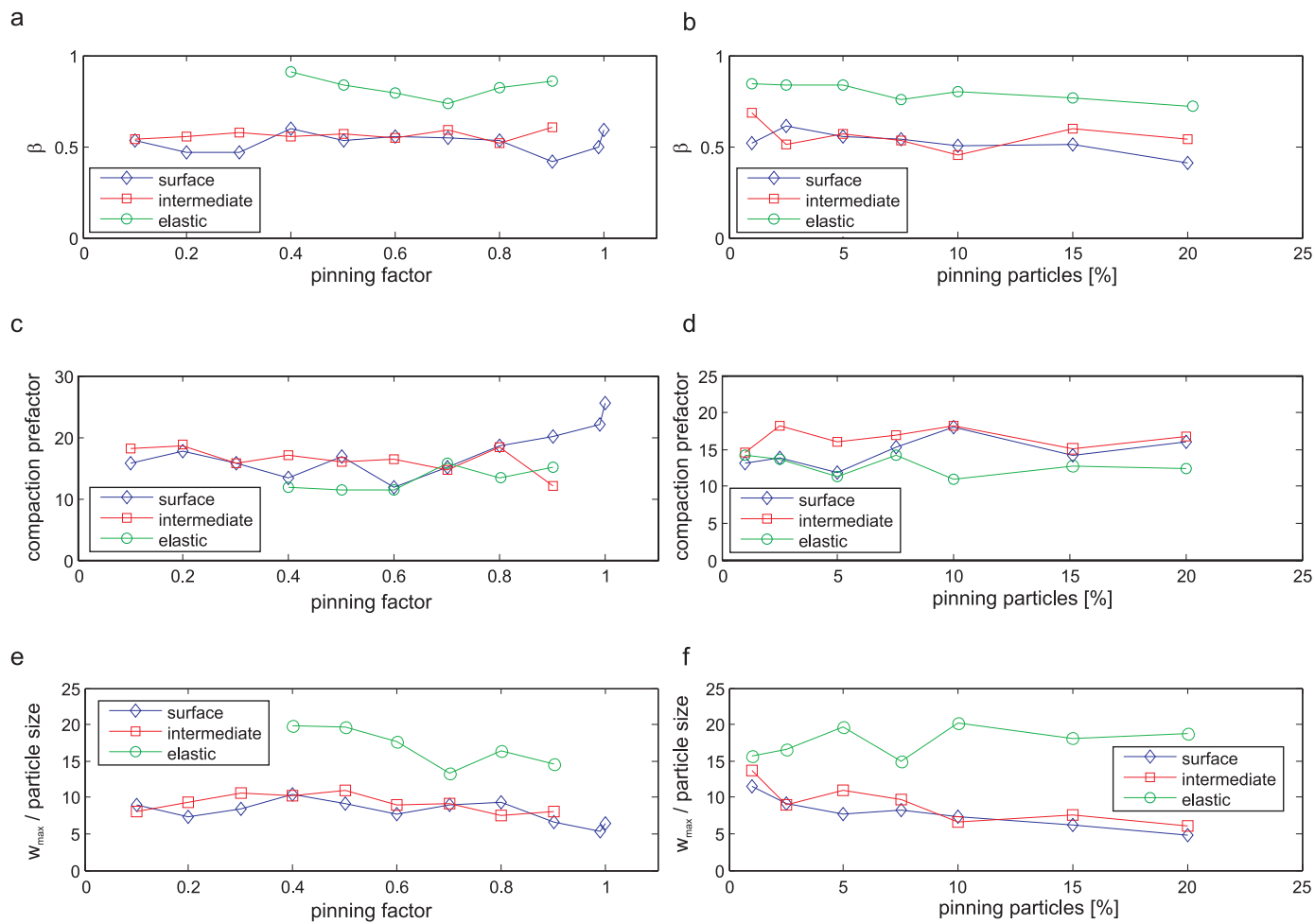


Figure 8

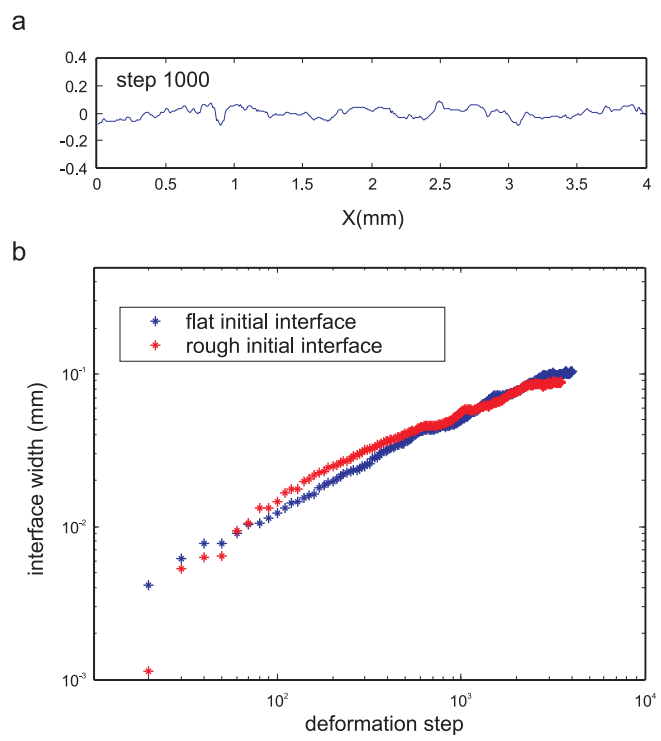


Figure 9

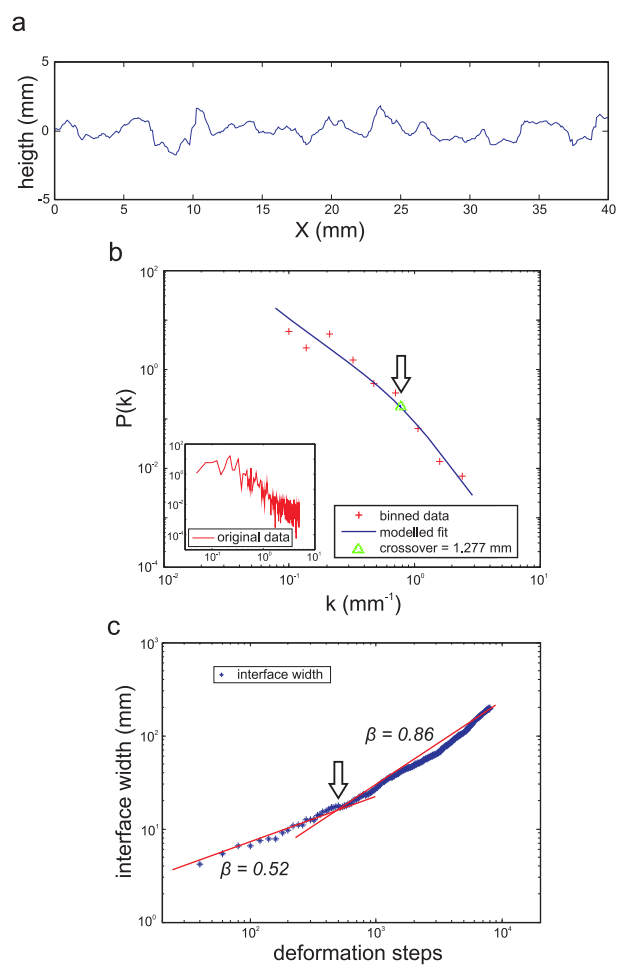


Figure 10

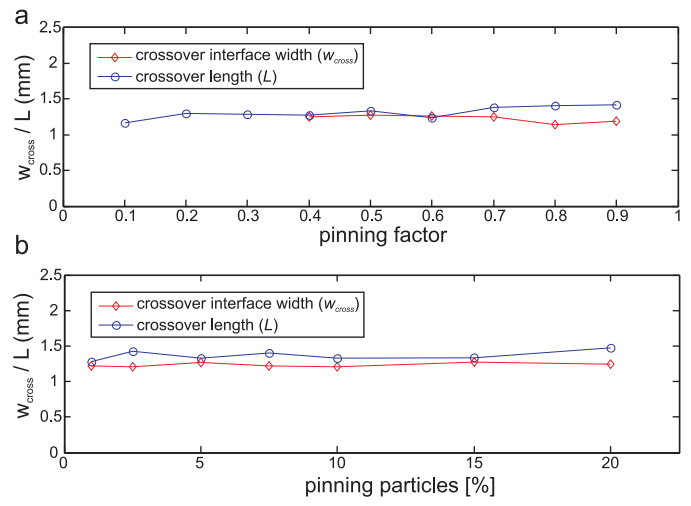


Figure 11