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## *Appendix A*

# DERIVATION OF SURFACE MODEL EQUATIONS

Dove et al. (2005) found that the same equations originally developed to describe crystal growth (Burton et al., 1951; Chernov, 1984; Malkin et al., 1989) could also be used to describe dissolution. Although developed for a single component crystal, we step through the model below as it may still provide useful insight into the behavior of $CaCO_3$. According to the surface model, dissolution occurs via the consecutive removal of crystal layers, where each dissolving layer has a defined thickness, $h$ (step height, nm), and retreats along the face of the crystal with a velocity, $v$ (cm/s). There can be several, simultaneous dissolution fronts, and the average spacing between them, $\lambda$ (nm), influences the overall rate. Closer spacings (smaller $\lambda$) allow for more dissolution fronts and a faster rate, whereas farther spacings (larger $\lambda$) can only support slower rates. Conceptualized this way, the normalized dissolution rate (length/time) is given by

$$R = \frac{hv}{\lambda} \tag{A.1}$$

The generalized form of Eq. (A.1) holds true for dissolution mechanisms that are not limited by the rate of transport to/from the mineral surface.

The retreat velocity, $v$, is linearly dependent on the step kinetic coefficient for the solid, the thermodynamic driving force, and the volume element being dissolved. It is classically formulated as (Chernov, 1984; Malkin et al., 1989)

$$v = \omega\beta C_e(1 - \Omega) \tag{A.2}$$

where $\beta$ is the step kinetic coefficient (cm/s), $\omega$ is the molecular volume ($cm^3$), and $C_e$ is the equilibrium concentration of dissolved species in solution (molecules/$cm^3$).

The step spacing is the only term in Eq. (A.1) that changes depending on the dissolution mechanism, and it is therefore what sets the functional form of the rate equation. At low driving forces near $\Omega \approx 1$, dissolution occurs primarily via the retreat of pre-existing steps at edges and/or screw dislocations. The step spacing is then derived assuming spiral retreat around a dislocation exceeding a "critical radius," $r_c$, that is set by the local bonding environment of the crystal and the solution driving force. It is given by (Chernov, 1984; Chernov et al., 1986)

$$\lambda = \frac{8r_c + P}{m} \text{ with } r_c = -\frac{\omega\alpha}{k_b T|\sigma|};\ |\sigma| = \ln(\Omega) \qquad \text{(A.3)}$$

where $m$ is the number of elementary steps (order 1), $P$ is the perimeter of the core of the dislocation (proportional to $2\pi mh$), $k_b$ is Boltzmann's constant, $T$ is the temperature (Kelvin), $|\sigma|=\ln(\Omega)$ is a measure of the solution driving force, and $\alpha$ is the free energy of step formation per unit step height (mJ/m$^2$). A larger $\alpha$ implies a slower dissolution rate, as the formation/retreat of a step produces a greater increase in the local surface energy of the crystal. The step edge free energy varies depending upon the local bonding environment of the material, where the bonding environment is affected by solid-solid interactions (whether dissolution is at a kink, step, dislocation, flat surface, etc.) and solid-solution interactions (changes in the chemical speciation of the surface). $\alpha$ is therefore distinct from the average surface energy of a perfect crystal (Burton et al., 1951; Burton and Cabrera, 1949; Cabrera et al., 1954; Cabrera and Levine, 1956; Chernov, 1984).

As the solution undersaturation increases, 2D etch pits begin to form first at crystal defects, and then homogenously across the mineral surface. Although step-retreat continues, it is limited to a single direction (the direction of the step), so its contribution to the overall rate is small once the production and radial spread of etch pits is activated. 2D dissolution therefore changes the dominant shape of the dissolving front from a spiral (Eq. A.3) to a spreading area, and in doing so changes the relevant step spacing to use in Eq. (A.1).

The spacing of etch pits of average area, $\lambda^2$, is related to the speed of propagation ($v$ from Eq. A.2) and the steady-state rate of pit opening, $J$ (cm$^{-2}$s$^{-1}$). The lifetime of a pit is given by (Chernov, 1984).

$$\frac{\lambda}{v} = \frac{1}{J}\frac{1}{\lambda^2} \tag{A.4}$$

Substituting (A.4) into (A.1) gives a new equation that describes the rate of 2D dissolution, either at defects or homogenously across the surface.

$$R_{2D} = h(v^2 J)^{\frac{1}{3}} \tag{A.5}$$

Eq. (A.5) may be further expanded to account for the solution's effect on the steady-state rate of pit opening, $J$. The rate of pit opening depends on two things: the frequency of new site formation (sites/time) and the probability of surpassing some critical free energy barrier, $\Delta G_{Crit}^{2D}$. The overall energy barrier is, in turn, a function of the *local* step edge free energy $\alpha$, and the driving force of the solution, $\sigma$. It is given by (Malkin et al., 1989)

$$\Delta G_{Crit}^{2D} = -\frac{\pi \alpha^2 \omega h}{k_b T |\sigma|} \tag{A.6}$$

We can see from Eq. (A.6) that the required $\Delta G_{Crit}^{2D}$ to stabilize and open a pit decreases for constant $\alpha$ as the solution becomes more undersaturated (greater $|\sigma|$). The frequency of new site formation is related to the density of active nucleation sites ($n_s$, sites/cm$^2$), the lattice spacing ($a$, nm), and the spreading rate constant for the material ($\beta$).

The probability of opening a pit is set by $\Delta G_{Crit}^{2D}$ via an Arrhenius-style relation, where the pre-exponential factor contains the steady-state frequency of new site formation, $n_s a \beta$.

$$J = J_0 \exp\left(-\frac{\Delta G_{Crit}^{2D}}{k_b T}\right) \text{ with } J_0 = |\sigma|^{\frac{1}{2}} n_s a h C_e \beta \tag{A.7}$$

The step height, *h*, and the lattice spacing, *a,* are physical properties of the mineral that do not change with temperature or solution undersaturation. $C_e$ exhibits a temperature dependence, but this effect is well documented in seawater (Dickson and Millero, 1987; Mehrbach et al., 1973) and simple to account for in the model. Therefore, the crystal parameters in $J_0$ that set the overall, steady-state nucleation rate are the density of active pit nucleation sites, $n_s$, and the step kinetic coefficient, *β*.

*β* is expected to exhibit temperature dependence according to (Chernov, 1984; Malkin et al., 1989; Xu et al., 2010; Zhang and Nancollas, 1992):

$$\beta = \beta_0 \exp\left(-\frac{\epsilon_{step}}{k_b T}\right) \tag{A.8}$$

Here, the interpretation of $\epsilon_{step}$ changes depending upon which of two potential dissolution pathways is occurring. In the first pathway, the dissolving species directly detaches from a kink/step and enters the solution. In the second pathway, the species does not directly detach, but instead diffuses away from a kink/step to become an adatom that can subsequently desorb from the surface. $\epsilon_{step}$ in the first case is the energy of direct detachment from a kink/step, and $\epsilon_{step}$ in the second case is the energy of surface diffusion. Surface diffusion is hindered on calcite by water (Liang and Baer, 1997) and dipoles on the calcite surface (Gratz et al., 1993), so $\epsilon_{step}$ in our system is the energy of direct detachment from kinks/steps.

$n_s$ exhibits a temperature dependence according to (Chernov, 1984; Chernov et al., 1986; Dove et al., 2005):

$$n_s = n_{s0} \exp\left(-\frac{\epsilon_{init}}{k_b T}\right) \tag{A.9}$$

Here, $\epsilon_{init}$ is the kinetic energy barrier for removing a species from the surface to initiate a new etch pit. $\epsilon_{init}$ is distinct from Eq. (A.6) because, while etch pits can initiate on the surface, they will not be stable and propagate across the mineral face unless $\Delta G^{2D}_{Crit}$ has also been surpassed. The pre-exponential factors in Eq. (A.8) and (A.9) contain entropy terms (Burton et al., 1951).

Substituting Eq. (A.7), (A.6) and (A.2) into (A.5) and rearranging yields an equation describing dissolution by either homogenous or defect-assisted 2D dissolution:

$$\ln\left(\frac{R_{2D}}{(1-\Omega)^{\frac{2}{3}}|\sigma|^{\frac{1}{6}}}\right) = \ln\left(h\beta C_e(\omega^2 h n_s a)^{\frac{1}{3}}\right) - \frac{\pi\alpha^2\omega h}{3(k_bT)^2}\left|\frac{1}{\sigma}\right| \tag{A.10}$$

We arrive at a similar equation for dissolution by step retreat by substituting Eq.'s (A.3) and (A.2) into (A.1) and rearranging:

$$\begin{aligned}\ln\left(\frac{R_{step}}{(1-\Omega)^{\frac{2}{3}}|\sigma|^{\frac{1}{6}}}\right)&\\ &= \ln\left(\frac{\omega\beta C_e m h}{P}\right) + \ln\left((1-\Omega)^{\frac{1}{3}}\left|\frac{1}{\sigma}\right|^{\frac{1}{6}}\right)\\ &- \ln\left(1+8\left(\frac{\omega\alpha}{Pk_bT}\right)\left|\frac{1}{\sigma}\right|\right)\end{aligned} \tag{A.11}$$