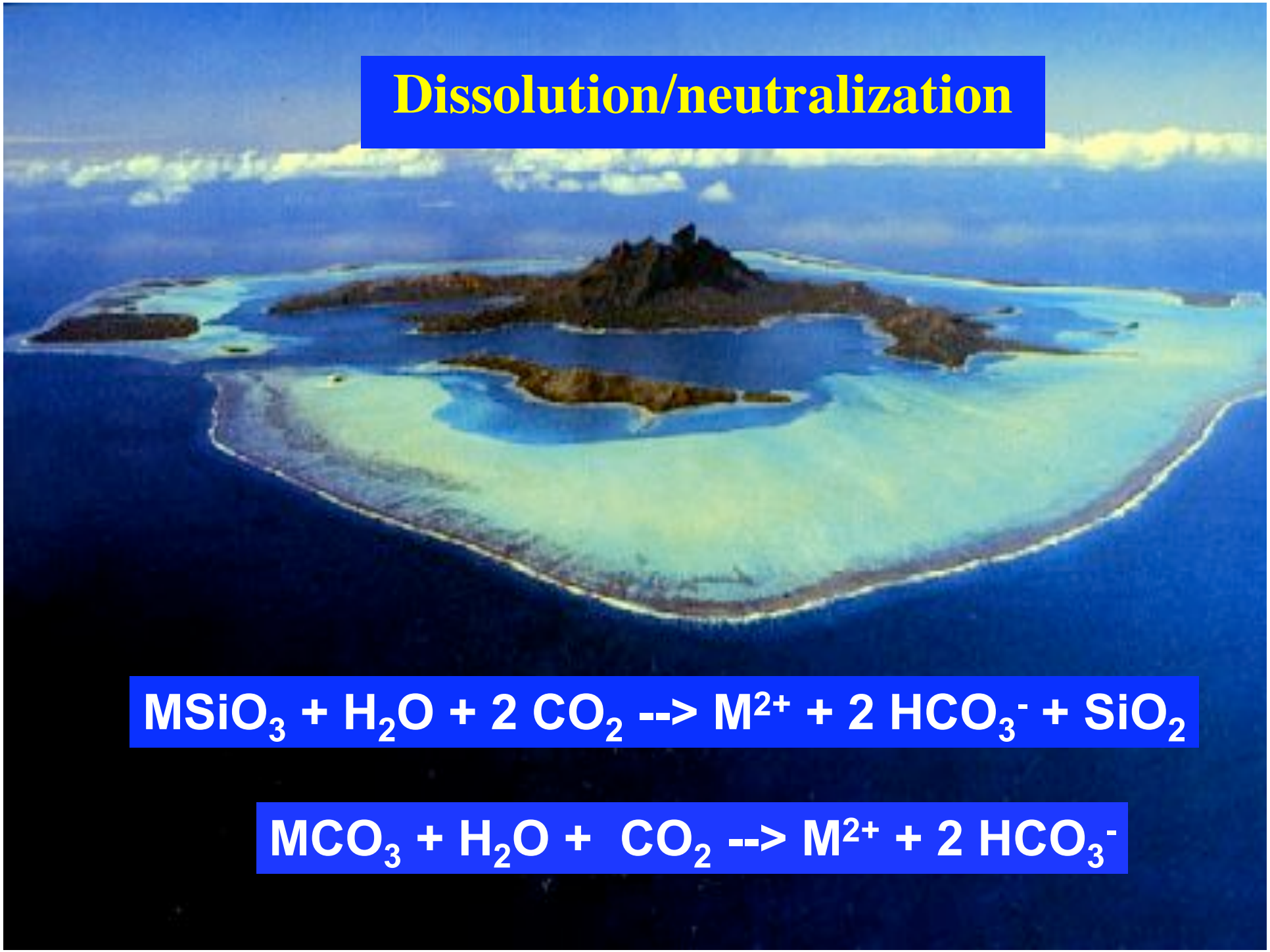

Dissolution/neutralization
$MSiO_3 + H_2O + 2\ CO_2 \rightarrow M^{2+} + 2\ HCO_3^- + SiO_2$
$MCO_3 + H_2O + CO_2 \rightarrow M^{2+} + 2\ HCO_3^-$

Dissolution rates

$$r = k_0 . A_{\min} . \exp(-E_a/RT) . g(I) . a_{H+}{}^{n} . \prod a_i{}^{ni} . f(\Delta_r G)$$

$$f(\Delta_r G) = 1 - \exp(\Delta_r G/RT) \; ?$$

Scientific issues

Rate-Affinity relationships

Effective reaction surface during dissolution

Coupling with mechanical properties

Thermodynamics in supercritical CO_2-H_2O

Scientific issues

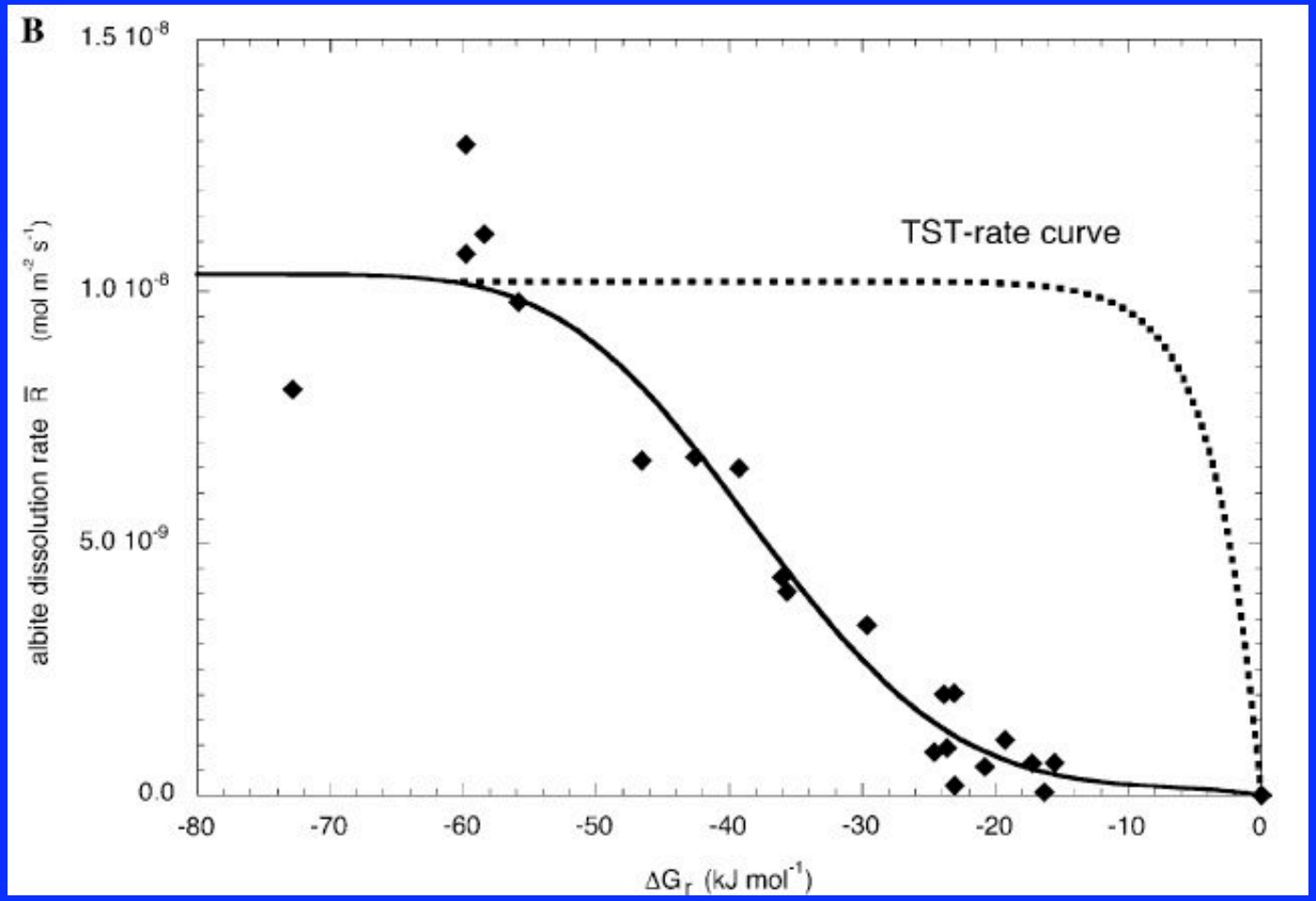
Rate-Affinity relationships

Effective reaction surface during dissolution

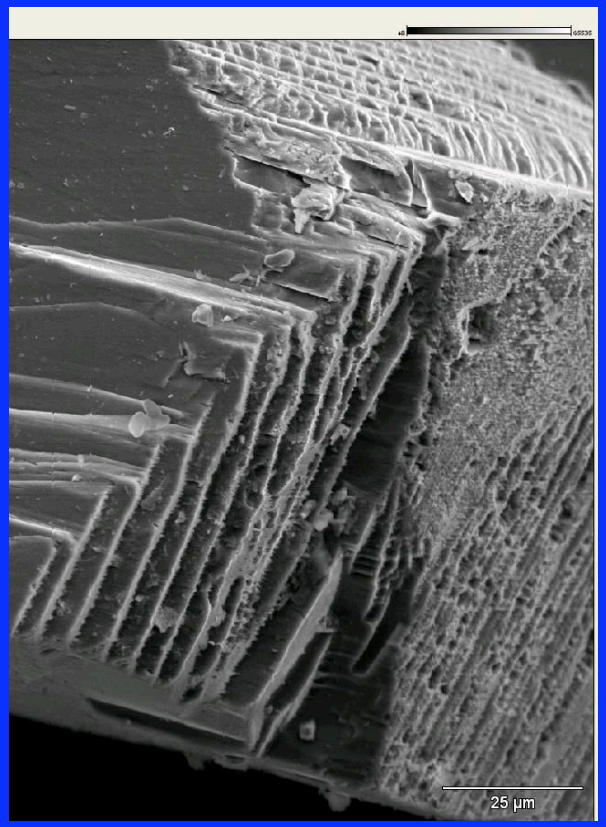
Coupling with mechanical properties

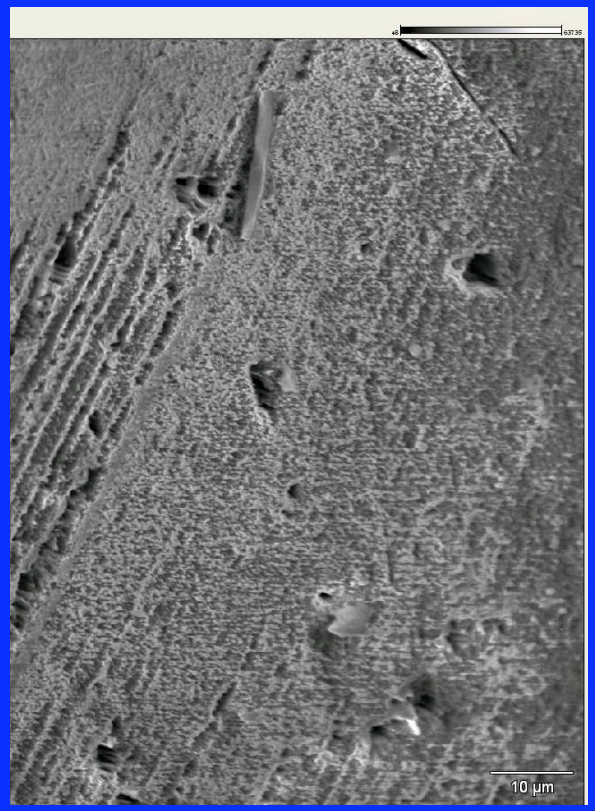
Thermodynamics in supercritical CO_2-H_2O

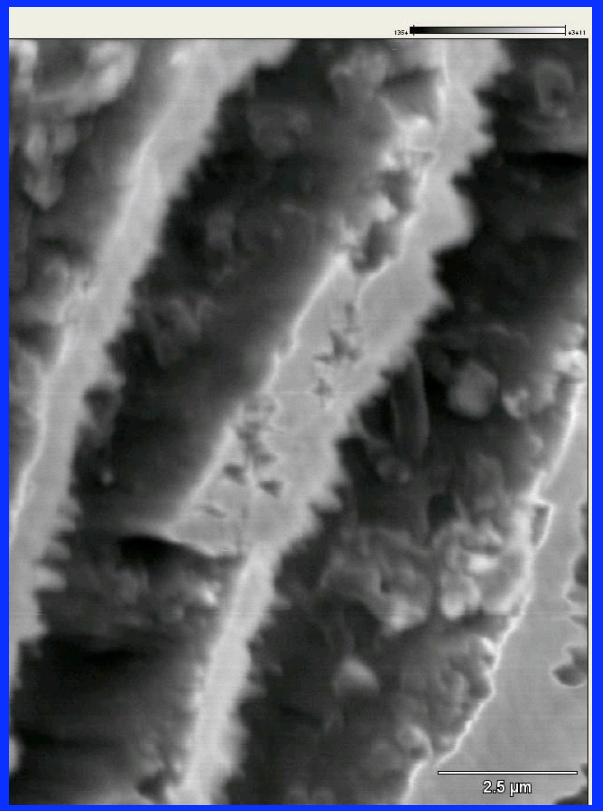
TST : $f(\Delta_r G) = 1 - \exp(\Delta_r G/RT)$



Dissolution microstructures







Challenge : continuous monitoring during reactions

Scientific issues

Rate-Affinity relationships

Effective reaction surface during dissolution

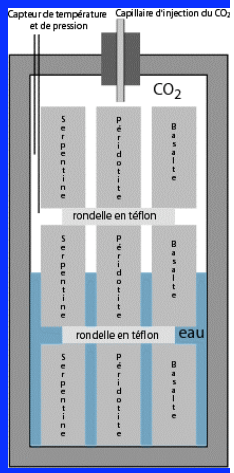
Coupling with mechanical properties

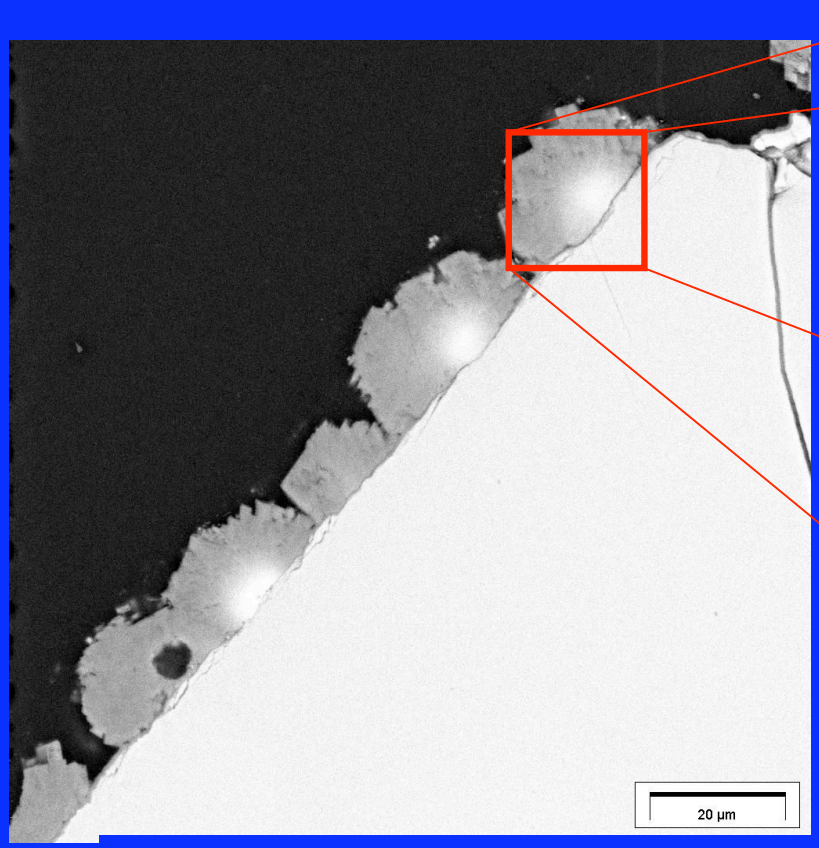
Thermodynamics in supercritical CO_2-H_2O

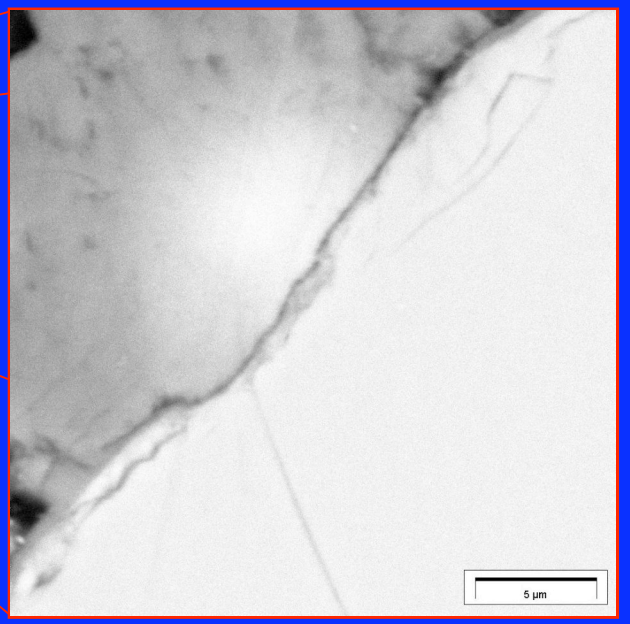
Partial passivation of dissolution surfaces

Olivine : Mg_2SiO_4

280 bar CO_2. 90°C







Dufaud et al., 2007

$$Mg_2SiO_4 + 2\ H_2O + 4\ CO_2 \rightarrow 2\ Mg^{2+} + 4\ HCO_3^- + SiO_2$$

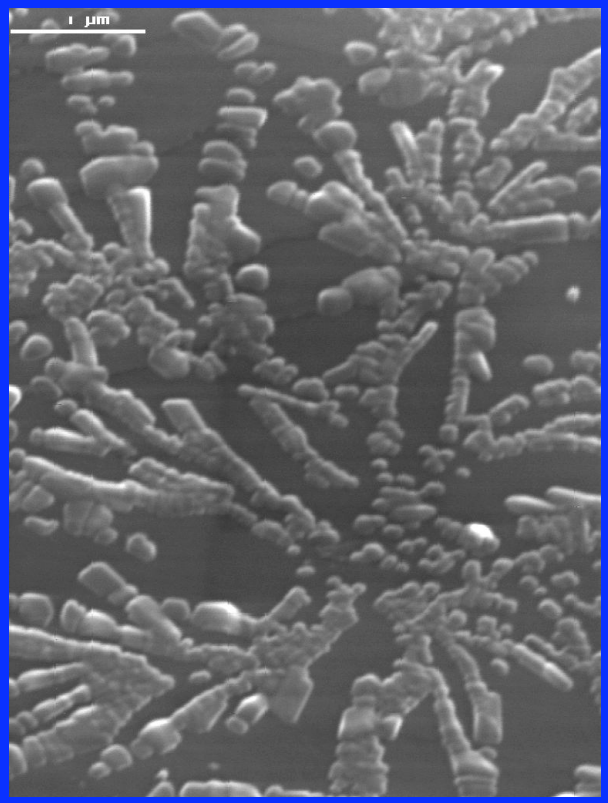
$$2\ Mg^{2+} + 4\ HCO_3^- \rightarrow 2\ MgCO_3 + 2\ H_2O + 2\ CO_2$$

$$Mg_2SiO_4 + 2\ CO_2 \rightarrow 2\ MgCO_3 + SiO_2$$

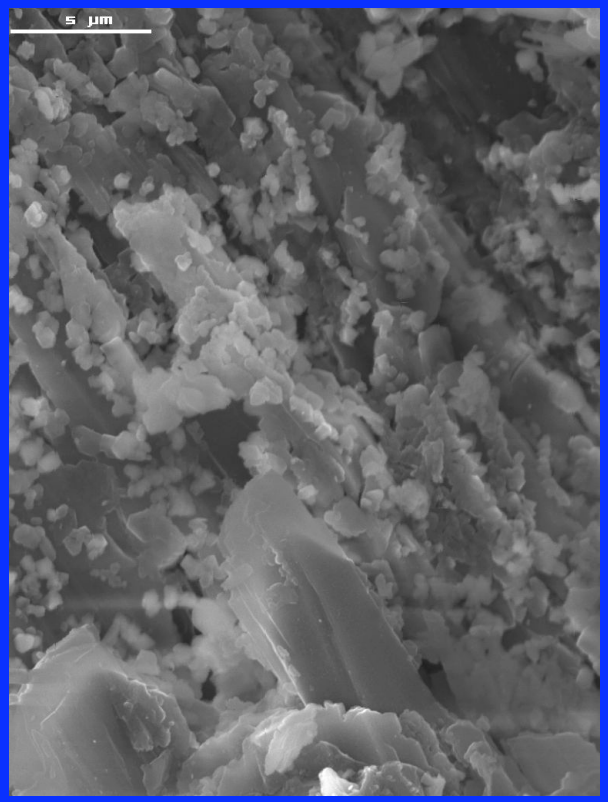
Effective reactive surface vs time

supercritical CO_2

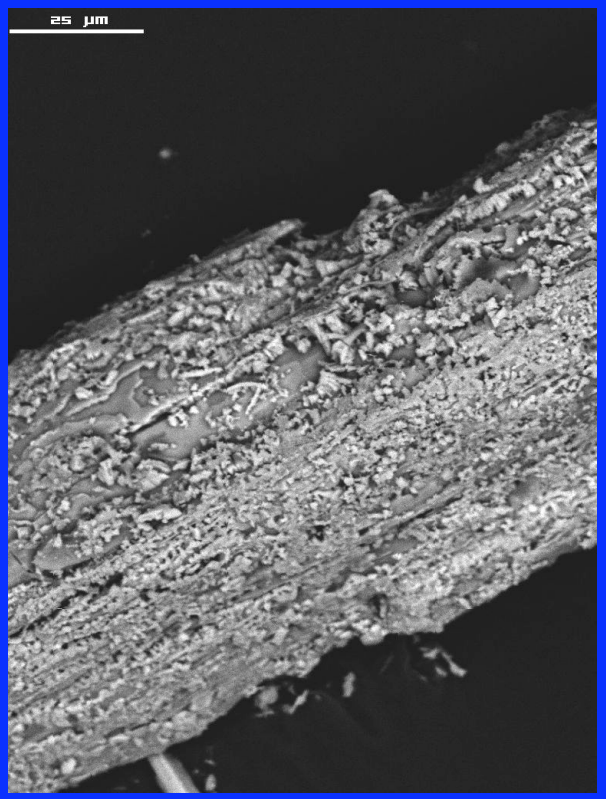
8 hrs



1 day

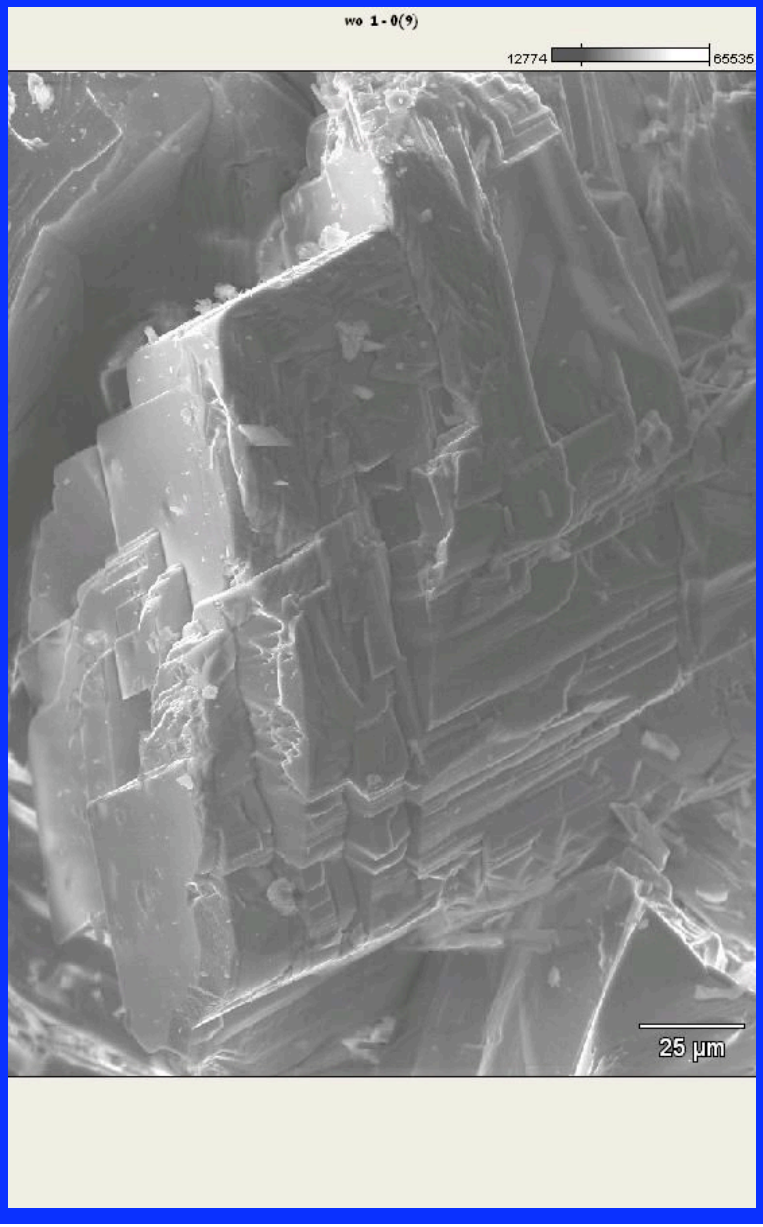


11 days

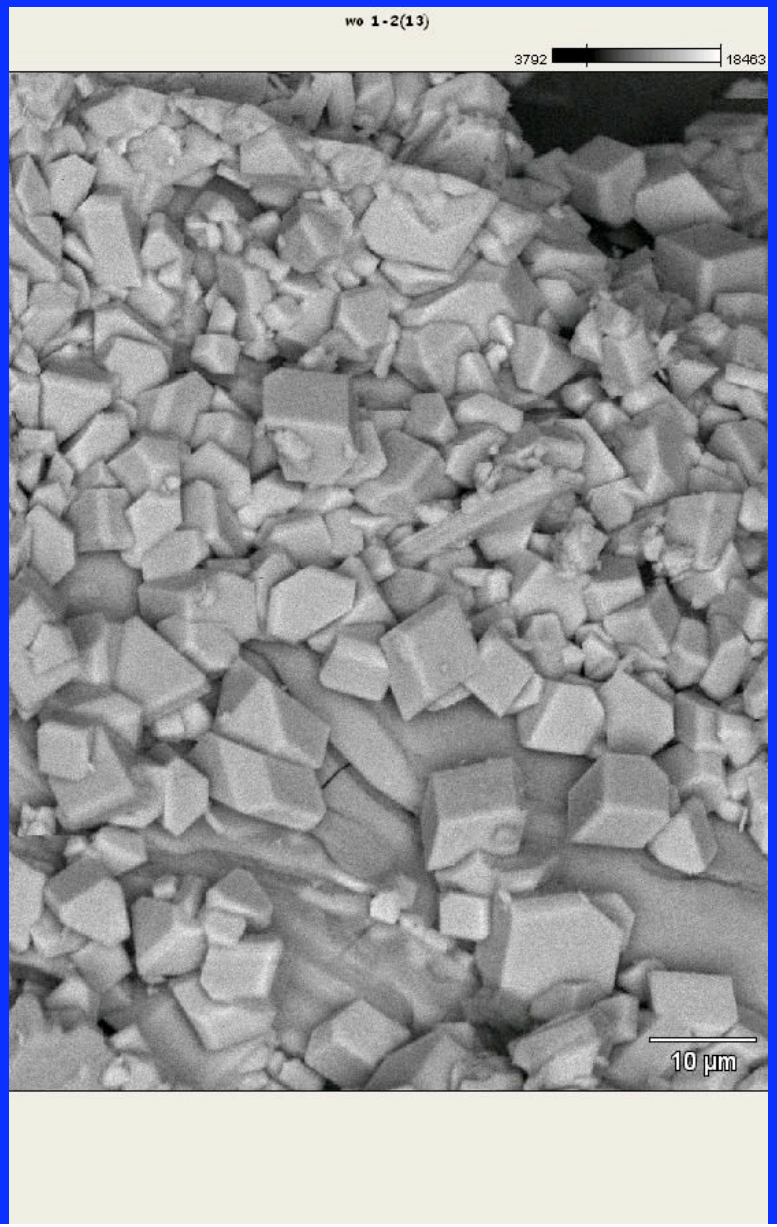


Partial passivation

Initial water



Initial alkaline



Scientific issues

Rate-Affinity relationships

Effective reaction surface during dissolution

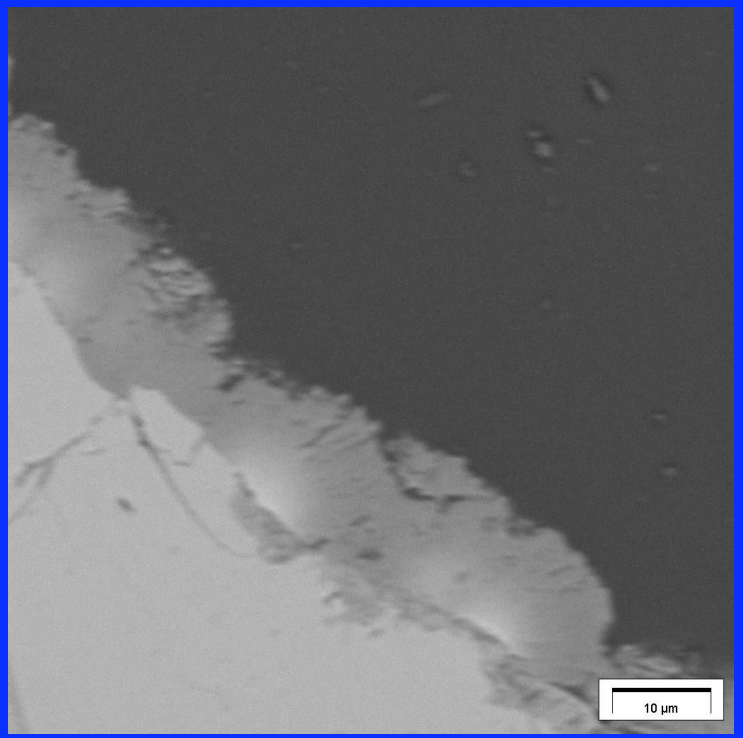
Coupling with mechanical properties

Thermodynamics in supercritical CO_2-H_2O

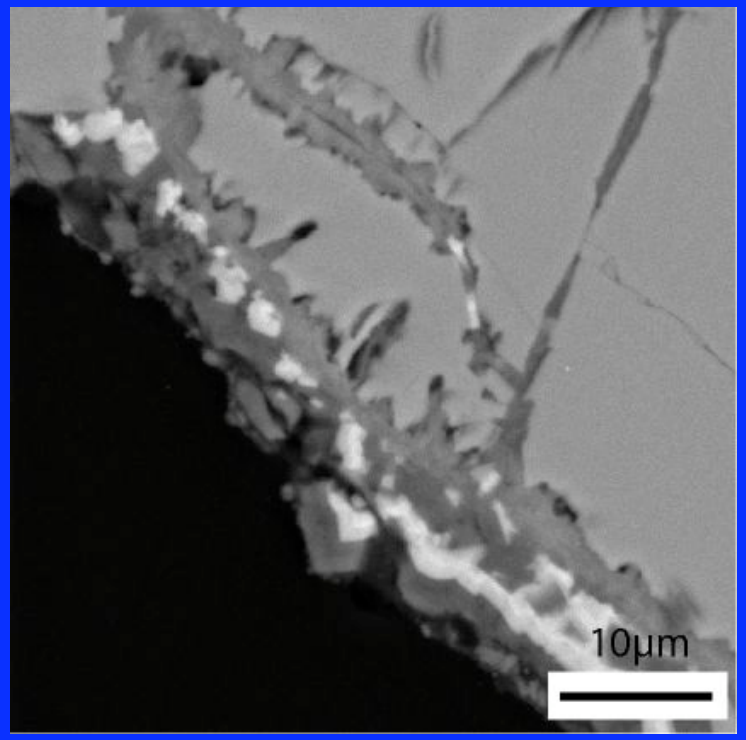
Réactivités comparées dans la phase aqueuse et dans le CO_2 supercritique saturé en eau

$Mg_3Si_2O_5(OH)_4 + 3CO_2 \rightarrow 3MgCO_3 + 2SiO_2 + 2H_2O$

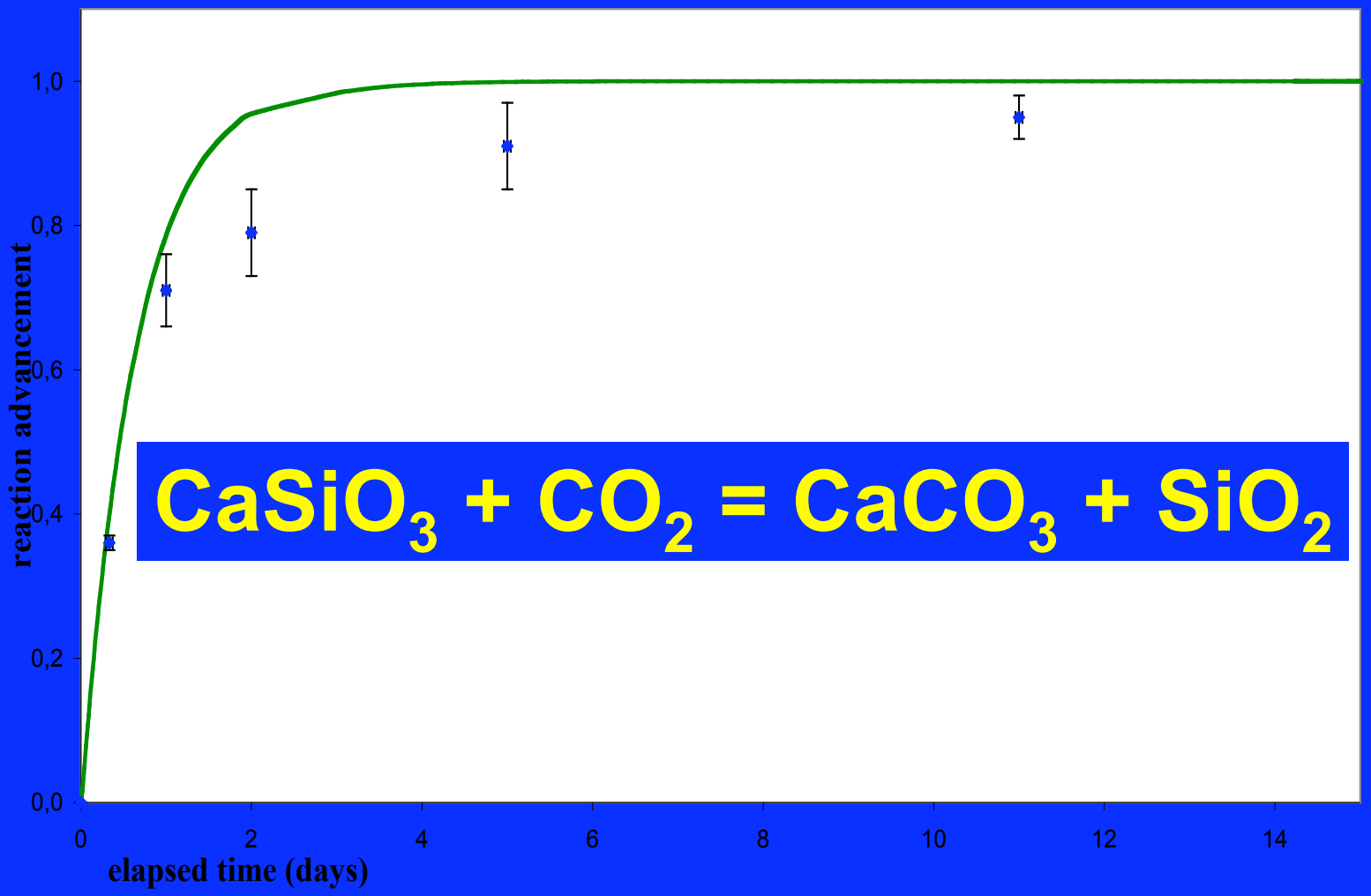
H_2O- CO_2.



CO_2 -H_2O.

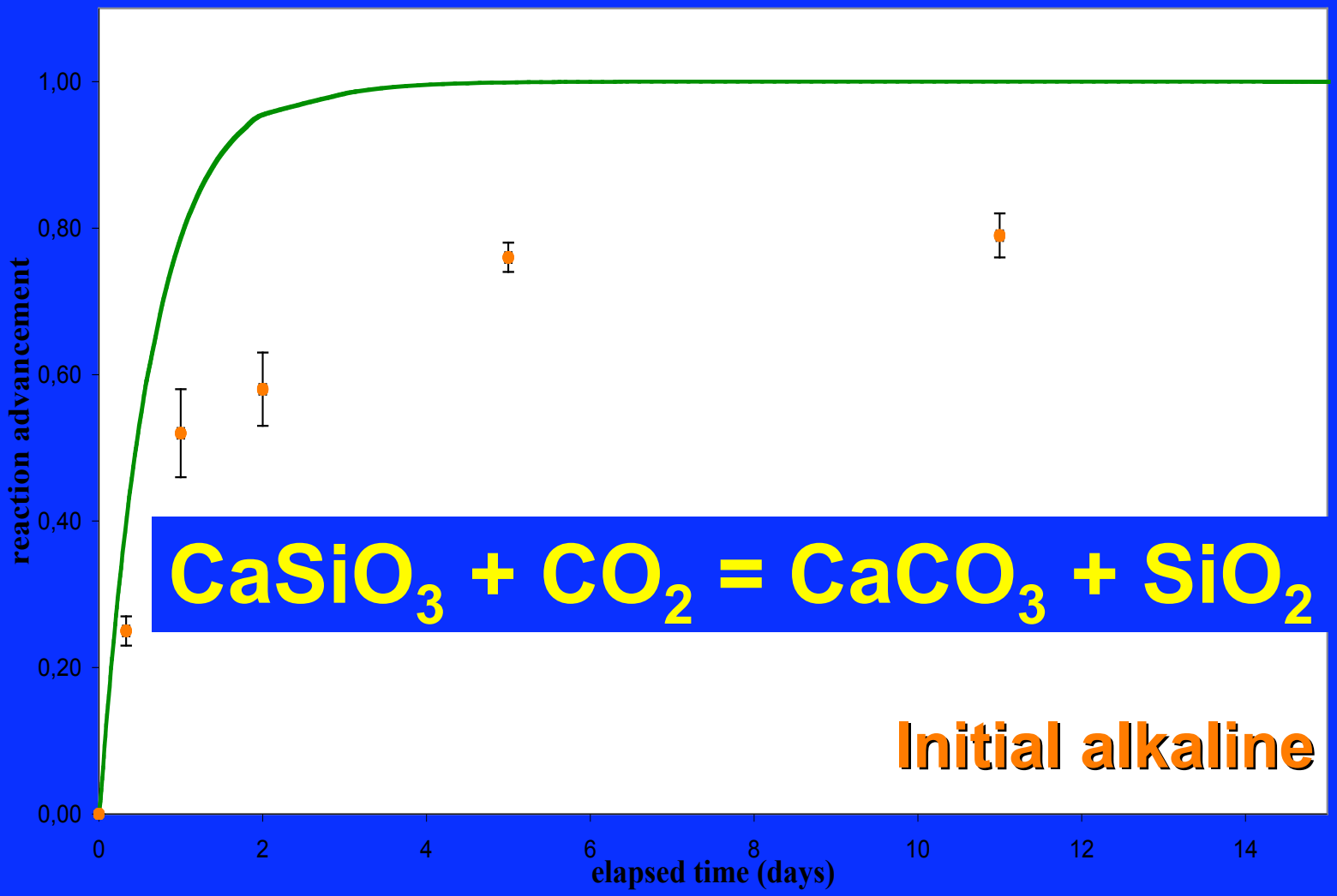


Geochemical modeling

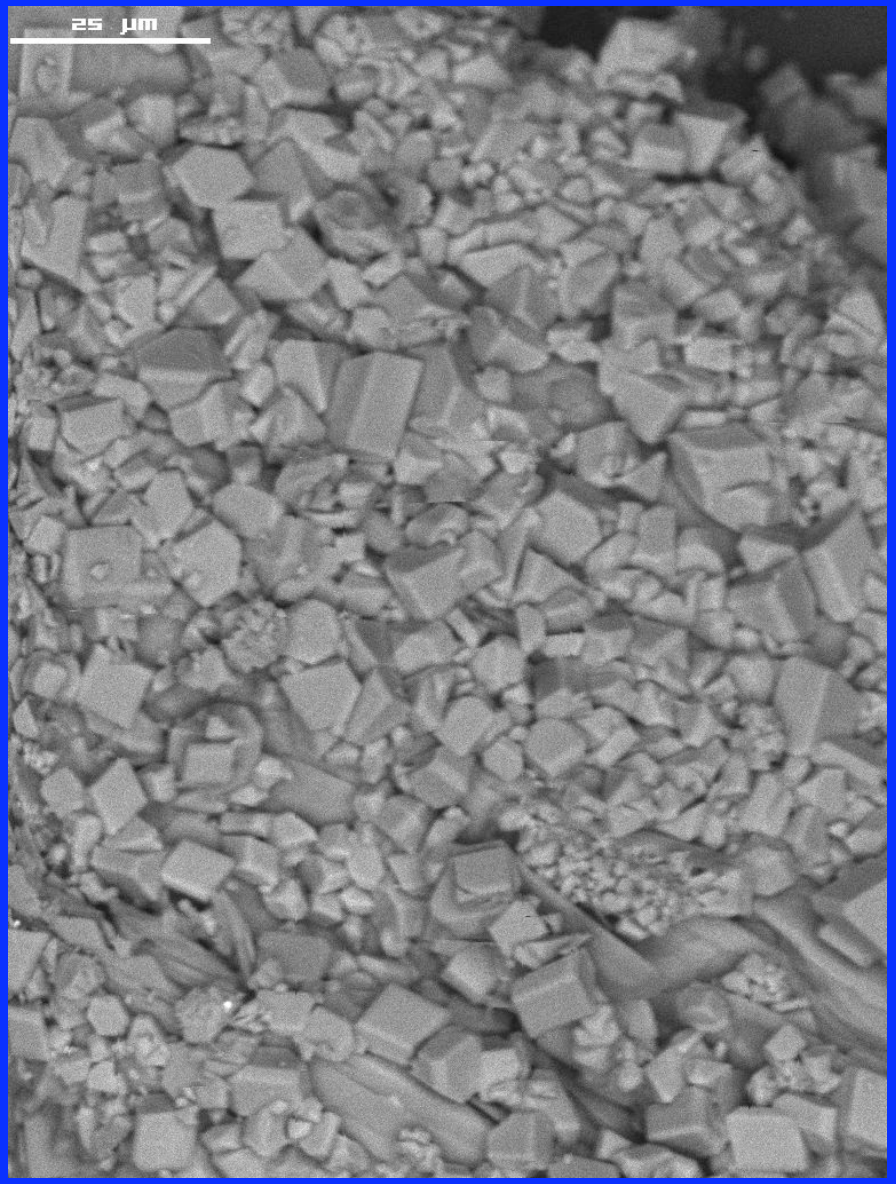


$$CaSiO_3 + CO_2 = CaCO_3 + SiO_2$$

Geochemical modeling

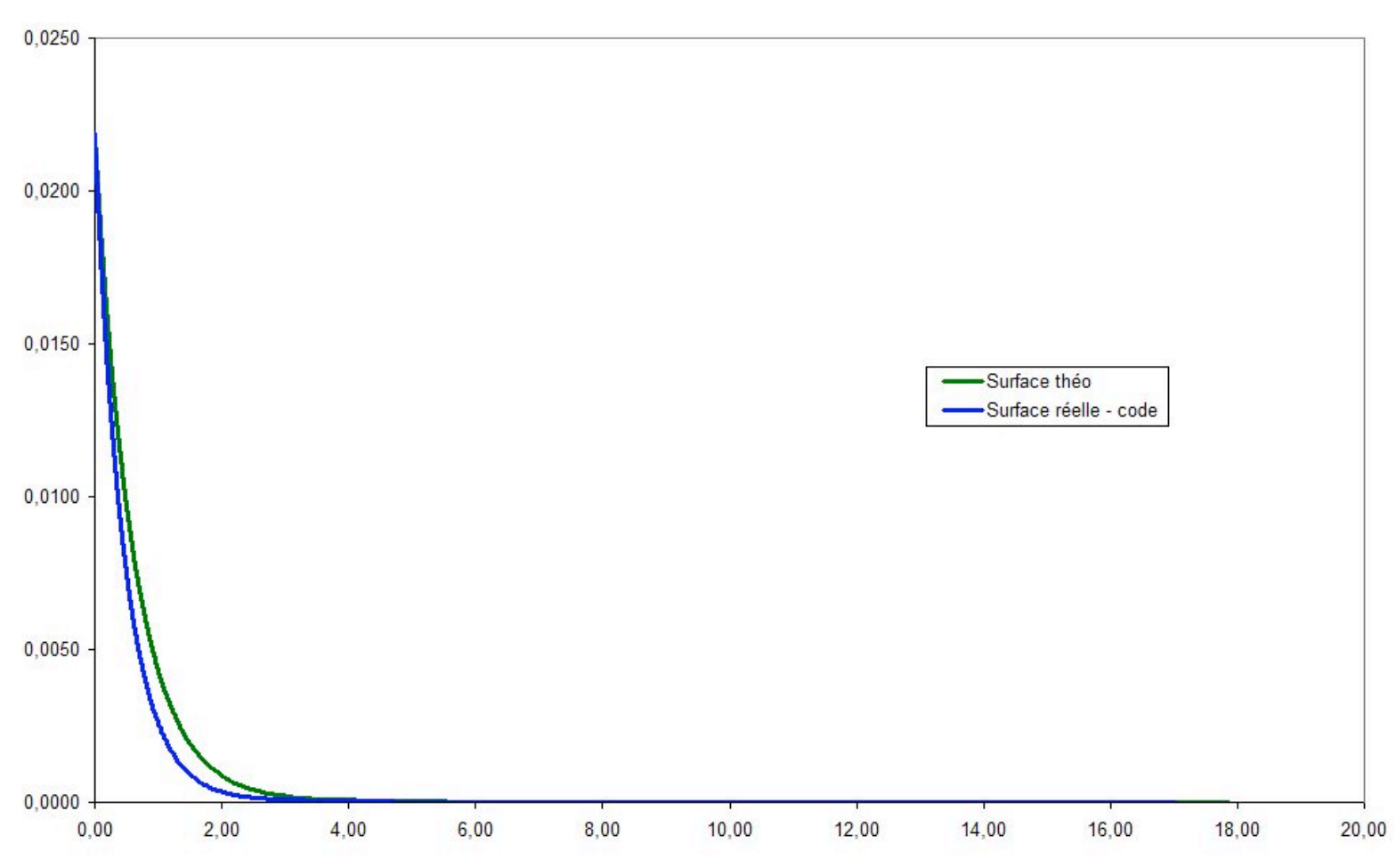


Deviations due to surface effects

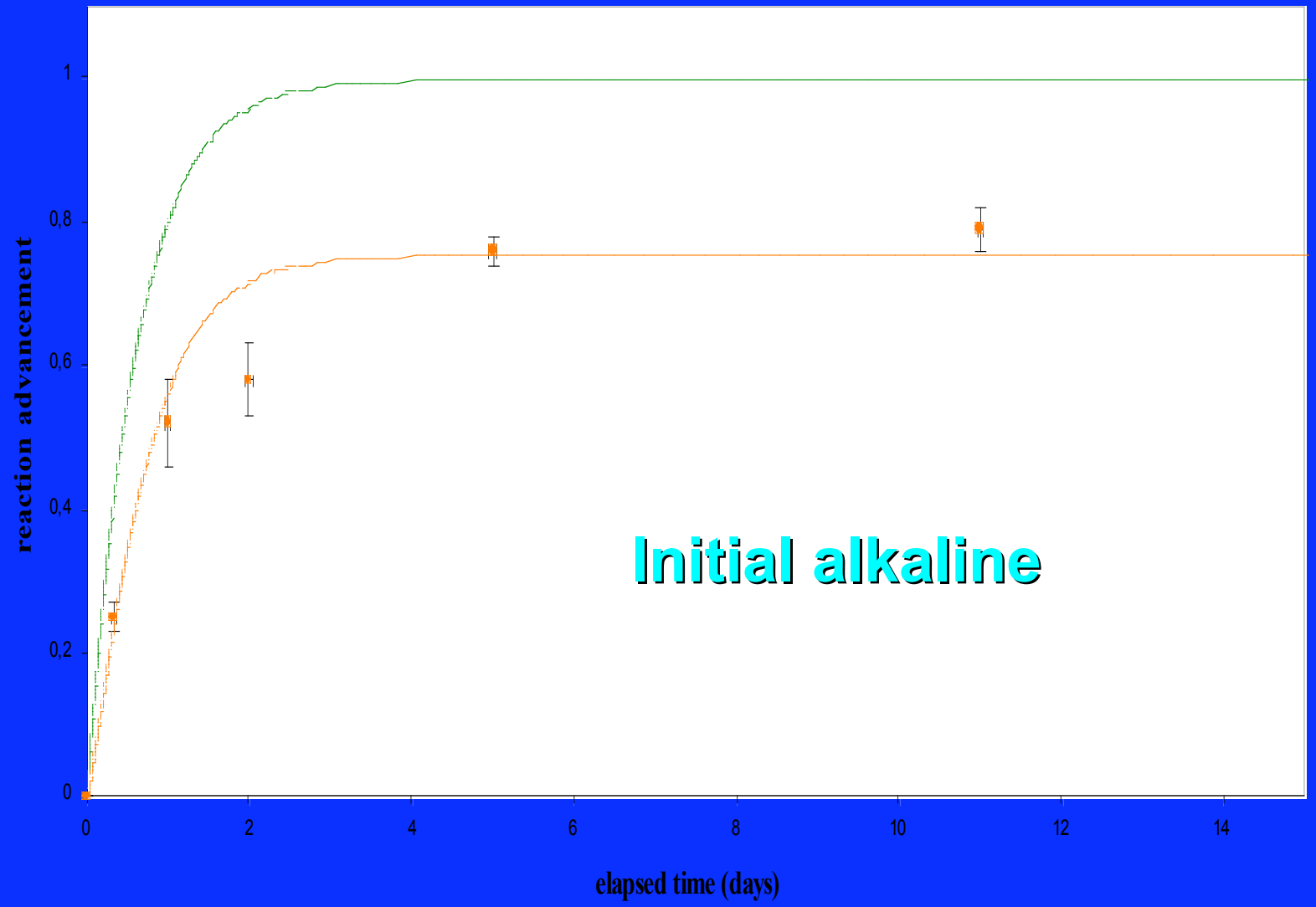


Initial Alkaline:
Larger supersaturation
Small crystals

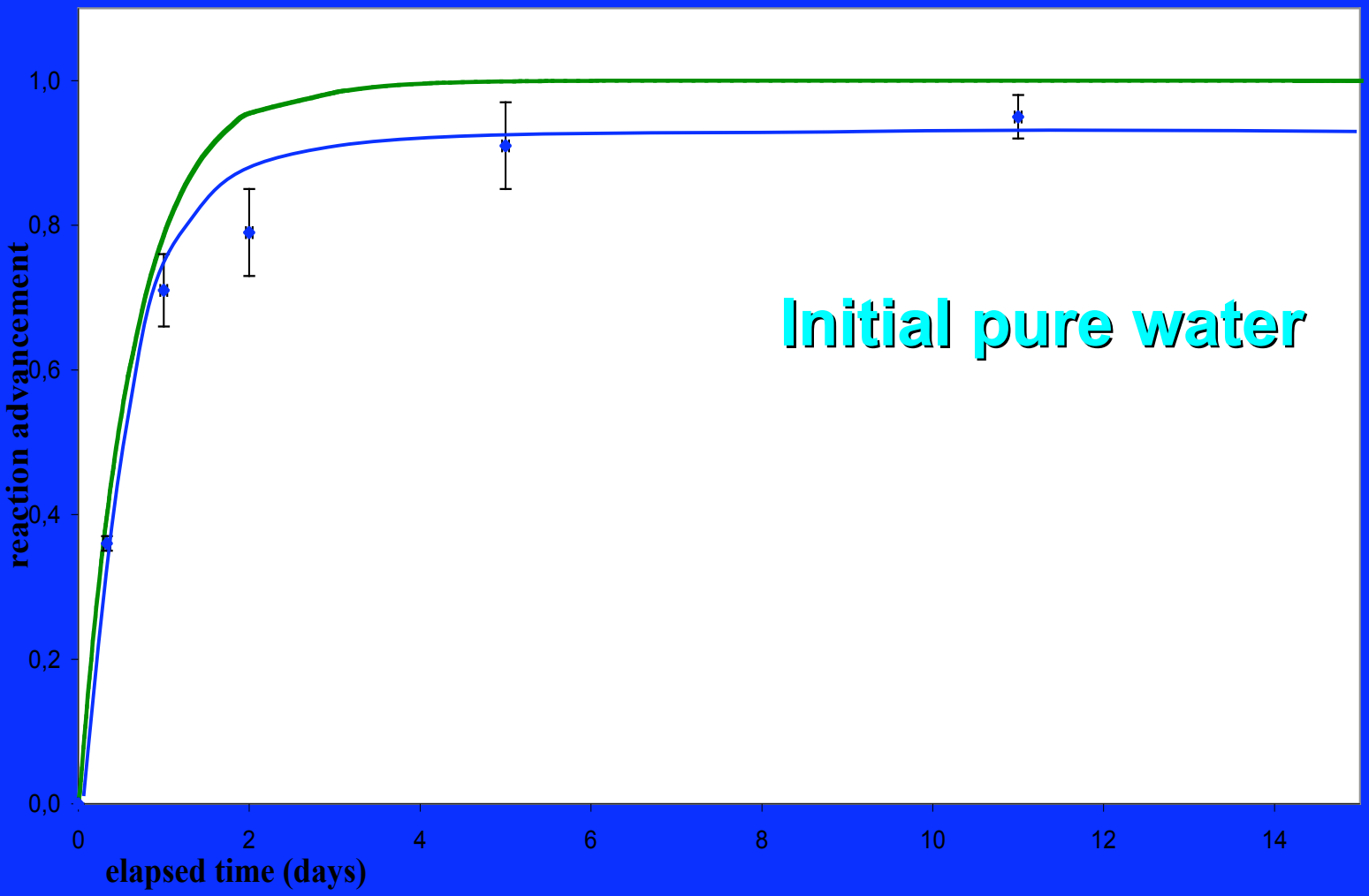
Surface evolution



Modeling taking into account surface evolution



Modeling taking into account surface evolution

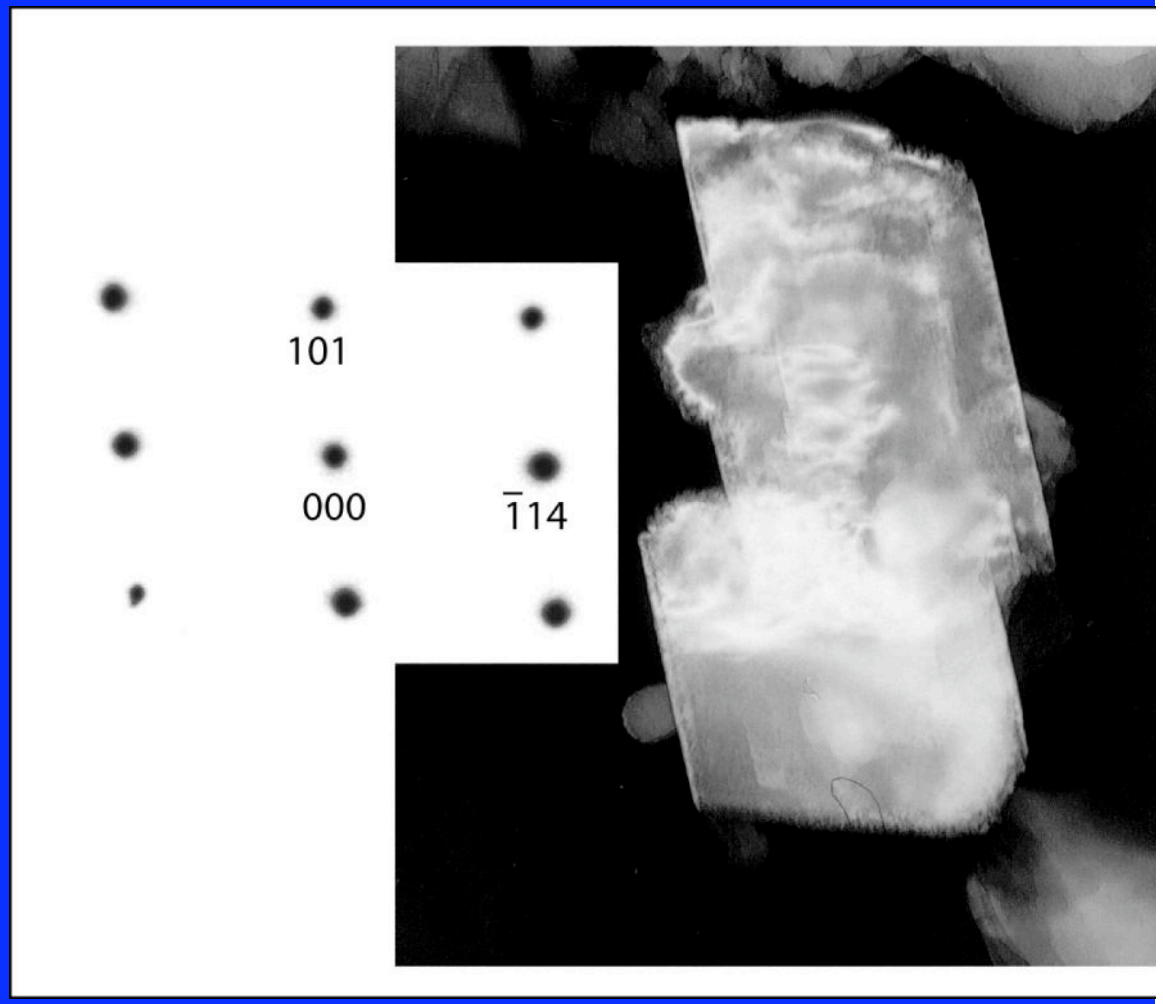


Recognition and analysis of post-injection newly formed phases

TRANSMISSION ELECTRON MICROSCOPY

$Mg_2SiO_4 + 2CO_2 \rightarrow 2\ MgCO_3 + SiO_2$

STABLE ISOTOPES



Dufaud, Martinez, Shilobreva,2007