Supplementary Information

Simulating carbon mineralization at pore scale in capillary networks of digital rock

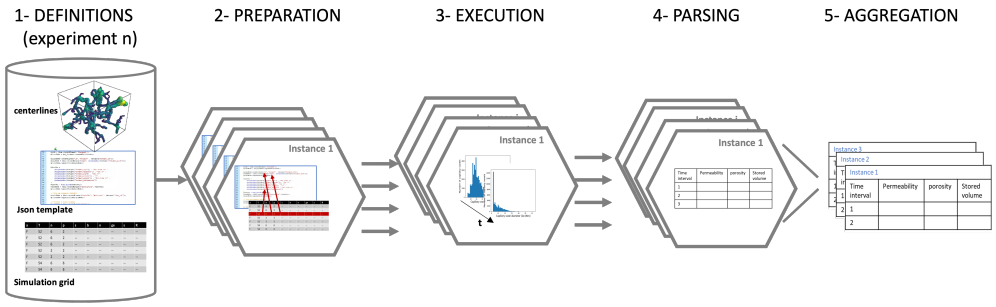
David A. Lazo Vasquez[1], Jaione Tirapu Azpiroz[2], Rodrigo Neumann Barros Ferreira[2], Ronaldo Giro[3], Manuela Fernandes Blanco Rodriguez[2], Matheus Esteves Ferreira[2], Mathias B. Steiner[2]

[1]IBM Research, Rua Tutóia, 1157, São Paulo, Sao Paulo, 04007-005, SP, Brazil.
[2]IBM Research, Av. República do Chile, 330, CEP 20031-170, Rio de Janeiro, RJ, Brazil.
[3]IBM Research, Rd J Fco Aguirre Proenca Km 9 Sp101, Hortolandia, Sao Paulo, 13186-900, SP, Brazil.

S1 Automated simulation workflow

The large number of scenarios with different simulation parameters lead to a parameter space that is intractable, but by leveraging high-throughput automated simulation workflows. Given the large parameter space that needs to be swept to fully assess the mineralization potential in a given geological storage scenario, we employed the Simulation Toolkit for Scientific Discovery (ST4SD) [56] to automate the execution of long simulation campaigns with several chained steps. By doing so, we drastically reduce the human labor involved in preparing the simulation inputs, submitting the jobs, and post-processing the results. The use of such a workflow scheduler also ensures the reproducibility of our results and enables efficiency gains by optimizing the use of computing resources. The underlying code implementation for this automation has been shared for repeatability (see "Code Availability" section).

Figure S1 shows the sequence of steps executed by each ST4SD experiment comprising 1) an experiment *definition*, needed for the 2) *preparation* of inputs for the 3) *execution* of the simulation, followed by the output 4) *parsing*, leading tot he final 5) *aggregation* of all the results. The user input at the *definition* step comprises three parts: the rock sample CNM in which the simulations are performed; a template of the simulation configuration file where to enter all fluid, rock, and reaction properties (e.g., viscosity, reaction rates), as well as the flow simulation conditions (e.g., pressures, temperatures) necessary for each execution; and a simulation grid where each line defines the values of all properties and conditions, including the types of reactive transport phenomena considered, that are applied to each simulation instance. The next step is the *preparation*, for each simulation instance, of the configuration files and inputs based on the defined user inputs. Each combination of rock and fluid samples, flow simulation conditions, and reactive transport parameters extracted from the



Supplementary Figure S1 Automated computational methodology showing, from left to right, experiment definition, preparation of inputs, simulation execution, output parsing, and aggregation of results.

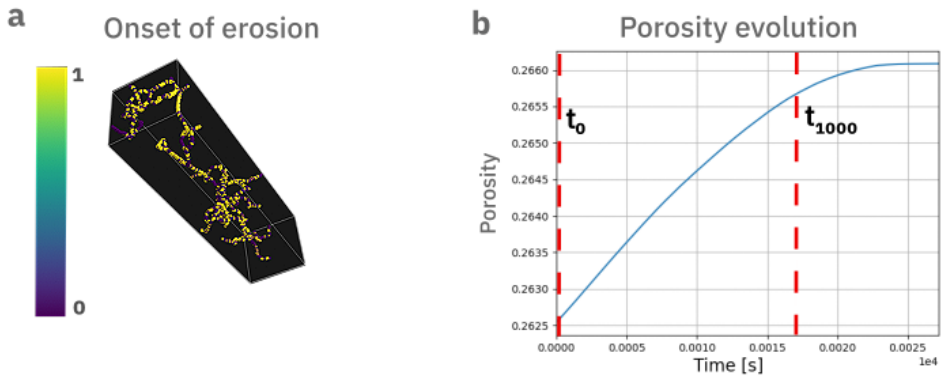
simulation grid leads to a different simulation scenario or instance. Many simulation conditions are not explicitly provided by the user, so default values are used instead.

The simulations are automatically scheduled and executed by ST4SD workflow, running all scenarios in parallel during the *execution* step. Per instance, this step corresponds to the execution of the coupled reactive-transport simulation, as illustrated in Fig 1, consisting of the iterative workflow where single-phase flow simulations are intercalated with a geometry modification routine that updates the capillary network based on the reactive phenomena. The *execution* step runs until reaching a convergence criteria and is then followed by the *parsing* of the simulation results to extract the relevant figures-of-merit from each parallel execution. Values of the sample porosity or the accumulated precipitated volume, for instance, are computed after each iteration and then saved into a single document during *parsing*, thus allowing eliminating memory-consuming interim files. A final post-processing of results is performed during the *aggregation* step, where all relevant figures-of-merit from all parallel simulations are collected into a single table format to facilitate user consumption.

S2 Reaction Model Library

Figure S2a displays a distribution of capillaries under the effect of erosion based on the calculation of reaction onset thresholds[7] with an erodibility coefficient $\kappa_{\text{er}} = 0.071 s \cdot m^{-1}$, 1000 secs of simulation time on a small sandstone rock sample of $225\mu m \times 225\mu m \times 900\mu m$ in size. The pore-scale processes involved are geometry and flow dependent, which determine the reaction rate as well as the potential existence of a process within each capillary as determined by Equation 1, and thus the number of eroded capillaries in the network may vary after each simulation time step. Moreover, the geometrical variation of each capillary within the network can be different given the varying flow characteristics (i.e., flow rate, pressure gradient) that may lead to either erosion, deposition, or none of those processes. When no process is present, the diameter remains constant. In the simulation results displayed in Fig. S3a, only the portion of the capillaries satisfying the criteria are affected by erosion and thus a non-uniform evolution of the geometrical changes is expected between the time

steps t_0,t_{1000}, and t_{5500}, corresponding to instants 0s, 17s and 93.5s, respectively. Fig. S3b shows the distribution of capillary diameters at those same time steps. As a consequence, the porosity of the small rock sample displayed in Fig S2a increases until reaching a plateau as shown in Fig S2b, where the geometry and flow conditions are not sufficient to meet the erosion and deposition law thresholds and, therefore no more variation in the diameter of the capillaries in the network occurs due to erosion.



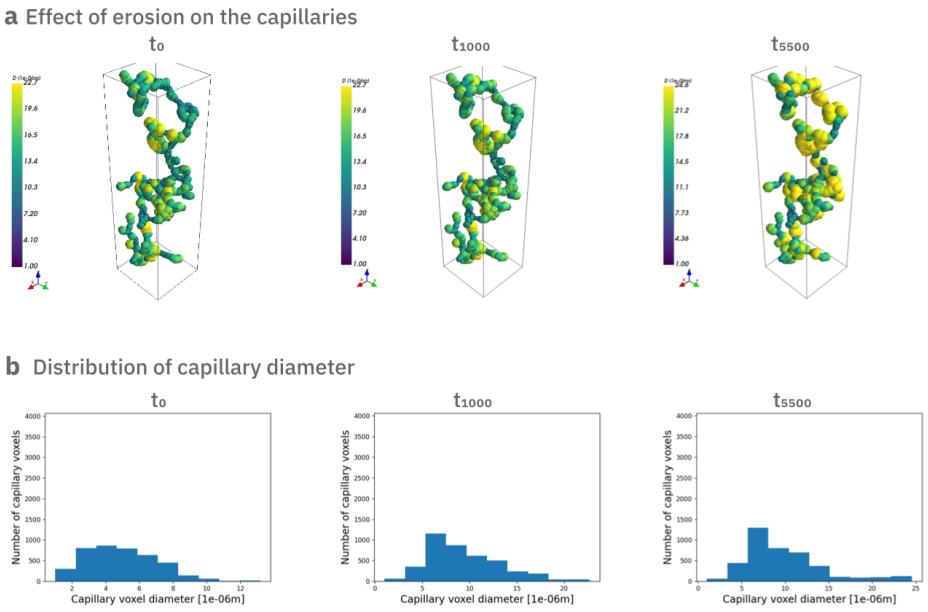
Supplementary Figure S2 a) Distribution of capillaries under the effect of erosion (label 1) based on the calculation of reaction onset thresholds[7] with an erodibility coefficient $\kappa_{\text{er}} = 0.071s \cdot m^{-1}$, 1000 secs of simulation time on a small sandstone sample of size $225\mu m \times 225\mu m \times 900\mu m$. b) Evolution of porosity as a result of erosion with time.

S2.1 Mineral precipitation

Noiriel et al.[10] proposed an integrated experimental and modeling approach for studying an induced calcium carbonate growth in cylindrical cores packed with glass beads and calcium carbonate crystals injected over 28 days with a supersaturated mixture of $CaCl_2$ and $NaHCO_3$. A general equation to describe the rate of calcium carbonate precipitation [44] ($\dot{r}_{ppt}$ $\left[\text{mol/m}^2\text{s}\right]$) reads:

$$\dot{r}_{ppt} = k_{ppt}\left[\exp\left(\frac{m\Delta G}{R^* \cdot T}\right) - 1\right]^n = k_{ppt}\left[\exp\left(\frac{IAP}{K_{sp}}\right)^m - 1\right]^n = k_{ppt}\left(\Omega^m - 1\right)^n \tag{S13}$$

where k_{ppt} represent the precipitation rate constant (mol m^{-2} s^{-1}), ΔG the Gibbs free energy change of the overall reaction (J mol^{-1}), Ω the saturation index $\Omega = IAP/K_{sp}$, K_{sp} the solubility of calcium carbonate, IAP the ion activity product defined by $a_{Ca^{2+}}a_{CO_3^{2-}}$, with $a_{Ca^{2+}}$ and $a_{CO_3^{2-}}$ the activities of Ca^{2+} and CO_3^{2-}, respectively. R^* represents the gas constant (J K^{-1} mol^{-1}) and T the absolute temperature (K). The values of n and m are semi-empirical constants that depend on the kinetic behavior involved in the chemical reaction.



Supplementary Figure S3 a) Evolution of the capillaries meeting the criteria for erosion at three simulation time steps t_0, t_{1000}, and t_{5500}, corresponding to instants 0s, 17s and 93.5s, respectively. b) Distribution of capillaries diameters as a result of erosion at those same time steps.

We consider two approaches for simulating mineral precipitation (i.e., uniform and flow-dependent crystallization) of calcium carbonate rock samples in brine and the consequent clogging in time using the correlations of Lasaga [47] and Noiriel et al.[10].