Kinetic Monte Carlo simulations of crystal dissolution using HTC

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Introduction

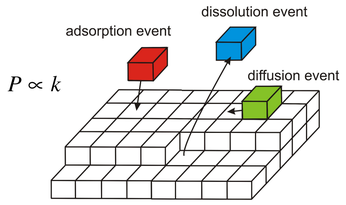
What are the areas, where crystal dissolution is important?

- •metal corrosion and synthetic material stability
- •transport of metals and toxic substances in rivers
- •weathering of rocks
- •formation of pore space in oil resourvoirs
- •geologic radioactive waste utilization: long-term stability of hosting rocks
- •drug dissolution in human's body

This work is focused on massive Kinetic Monte Carlo simulations of crystal dissolution. There are two main problems in the field:
-system size and time trajectory length are limited (this problem can be solved using parallel programming techiques)

-large number of unknown parameters (activation energies, dislocation network) which has to be predefined or studied by systematically varying their values. (this problem can be solved using High Troughput Computing techniques)

Kinetic Monte Carlo method



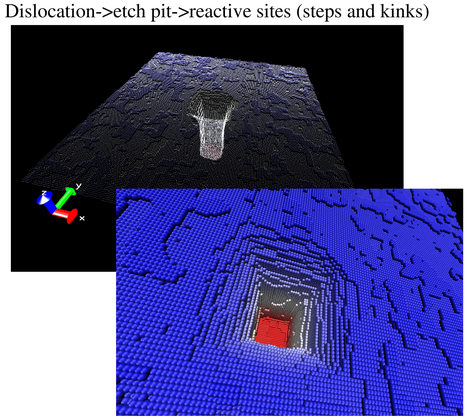
$$k_n^- = v \cdot \exp\left(\frac{-n\phi}{kT}\right) \qquad P_{event} = \frac{k_{event}}{v}$$

$$k^+ = k_{eq} \cdot \exp\left(\frac{\Delta\mu}{kT}\right) = v \cdot \exp\left(\frac{-3\varphi}{kT}\right) \cdot \exp\left(\frac{\Delta\mu}{kT}\right)$$

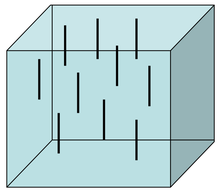
$$k_{diff} = v \cdot \exp\left(\frac{-E_a^{diff}}{kT}\right) \cdot \exp\left(\frac{-(n-1)\varphi}{kT}\right)$$

Role of dislocations

Dislocation->etch pit->reactive sites (steps and kinks)



Methods



Input parameters

•Activation energy of single reaction Ea
Range: 1-30 kcal/mol
Number of possible reactions: 3n for n types of bonds (dissolution, diffusion, precipitation)

•dislocation density
Range: 10^2 – 10^{16} dislocations/cm^2

•Depth of the dislocation core (defined by Burger's vectors and lattice properties)
Range: 1 nm-1µm

Starting model

•cubic crystal of size LxLyLz
•(L=100-5000 atoms)
•Screw dislocations are randomly seeded in the crystal

How dissolution rate of a simplest cubic crystal would respond to change in..
-dislocation parameters
-environmental conditions?

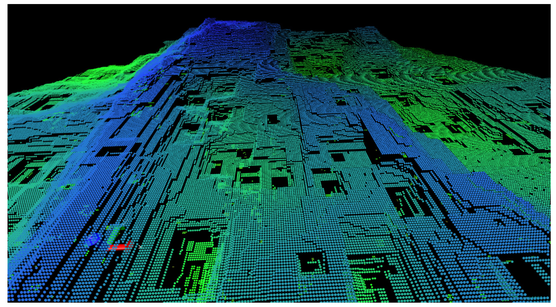
Computational time

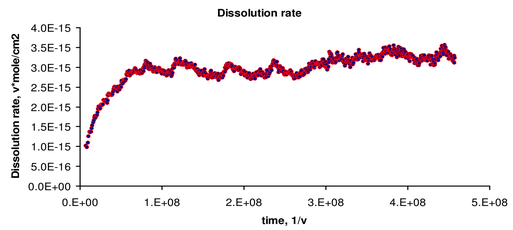
CPU time for a one job (the simplest system): 2-5 hours
Coplex system: up to 7 days (depending on system size and algorithm used)

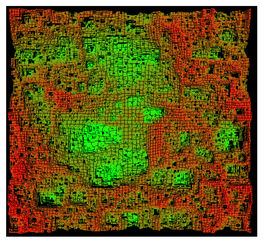
Output parameters

•dissolution rates
•density of reactive sites
•surface roughness
} 100-500 kB

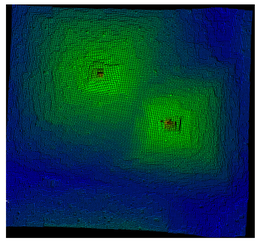
•Surface toporgaphy data (optional, important for visualization)- 5-50 MB



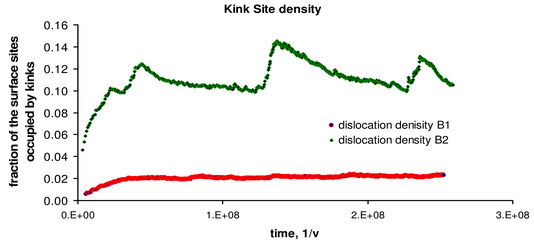




Dislocation density B1



Dislocation density B2



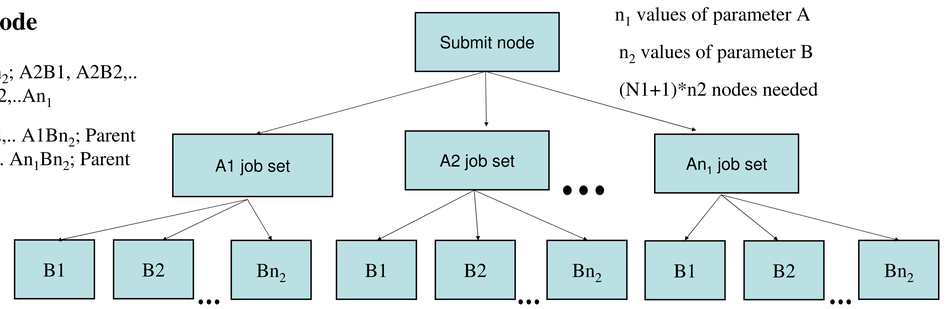
Control of the workflow using Condor's Dagman

Since we have a large amount of jobs with the different parameters to run, we need to automatically control the workflow and analyze the data. Condor's Dagman manager can solve this problem, describing the ierarhical relationships between the jobs. Fox example, if we would like to solve a 2-parametric problem: how activation energy (A) and dislocation density (B) influences dissolution patterns and rates, we can create a 2-layer network as shown below.
A1..An$_1$ nodes collect and analyze data from B nodes. Each An$_1$ node collects n$_2$ folders with output data;

Dagman's code

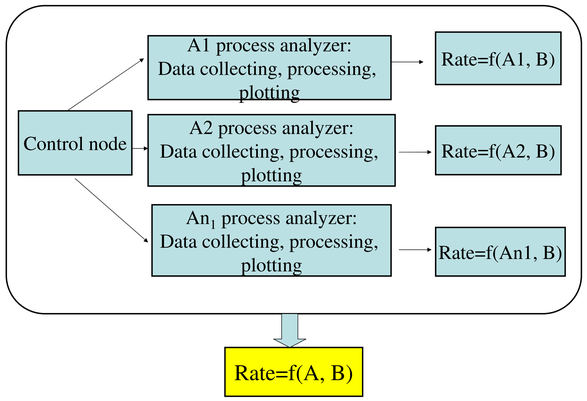
Job list: A1B1, A1B2, ..A1Bn$_2$; A2B1, A2B2,.. A2Bn$_2$,…;…; An$_1$Bn$_2$; A1, A2,..An$_1$

Relations: Child A1B1, A1B2,.. A1Bn$_2$; Parent A1; … Child An$_1$B1, An$_1$B2,.. An$_1$Bn$_2$; Parent An$_1$



Expected outcome

The large amount of output data needs to be automatically analyzed. Python or Shell scripting may help to group the output data depending on the parameter values, find "steady-state" intervals, calculate accociated rates and their variance, plot rate vs. parameter value dependensies. If we study how dislocation density affects dissolution rates at different activation energies, we need to find what is the functional rate dependence from the disclocation density and then compare the dependences found for different activation energies.



Resources needed

The amount of the resources required depend on the particular problem to solve. A single job for the simplest cubic crystal (our starting model) of the size 500x500x500 nm would run 2-3 hours (if we want to get meaningful time trajectory). If we would like to take 10 values of activation energies, 100 values of dislocation density and 10 values of Burger's vectors, we will need 2.3 years of computational time.If we need to solve more complicated problem, e.g. to vary activation energies for a complex crystalline system, we will need about 30 years of CPU time.
Therefore, having access to the 100 of nodes at Rice University SUGAR grid or OSG grid, we could solve the problem in weeks or month.

More problems to solve

-Let's make more complex system. How it would respond to the change of environmental conditions written in activation energy sets?

-Most of values of activation energies are unknown. However, we have experimental data about dissolution patterns and rates. Which sets of activation energies would reproduce experimental data?

Acknowledgements

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