

Understanding alkali-silica reaction:

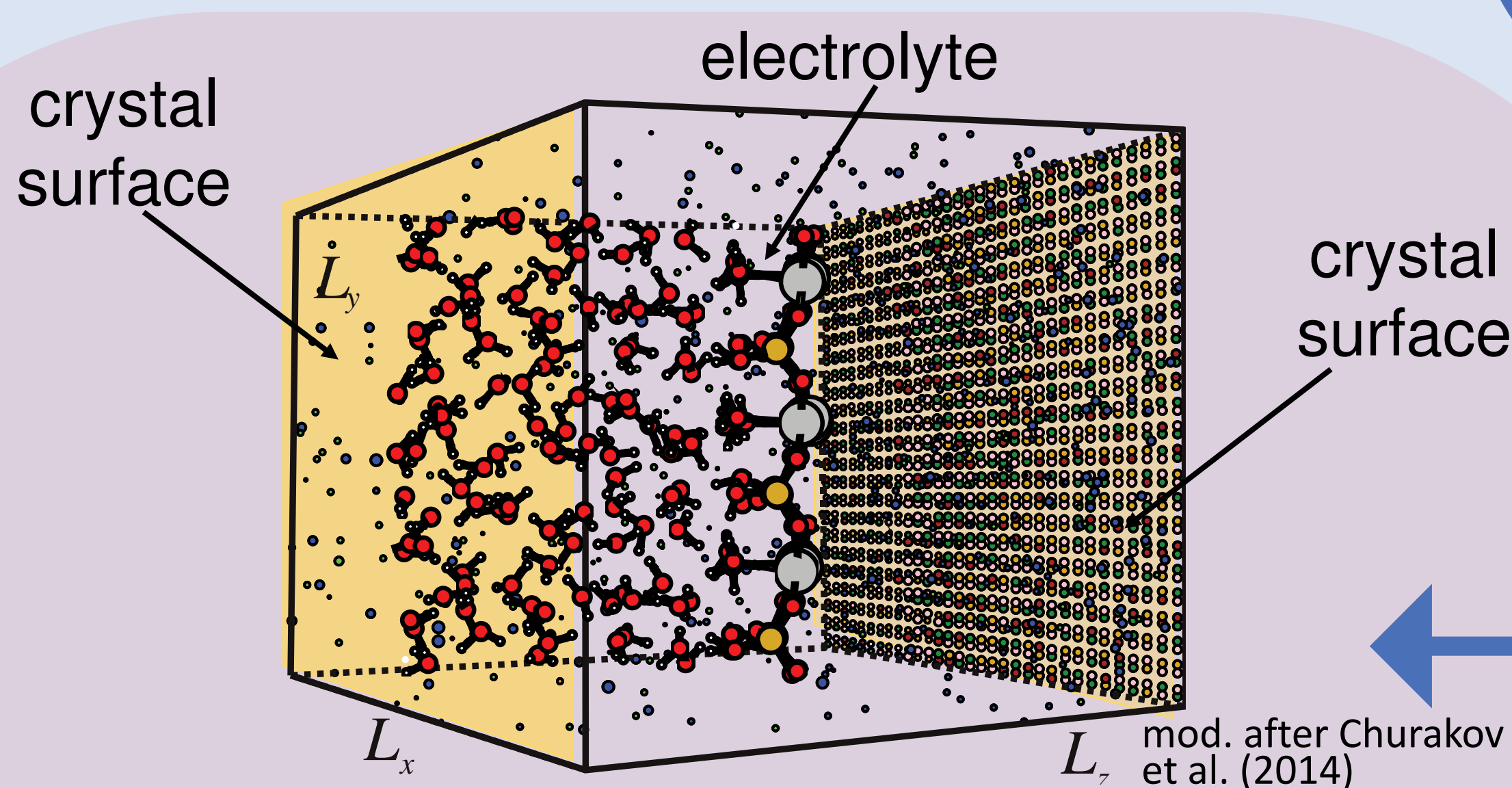
Atomistic modelling of chemo-mechanical processes responsible for concrete degradation

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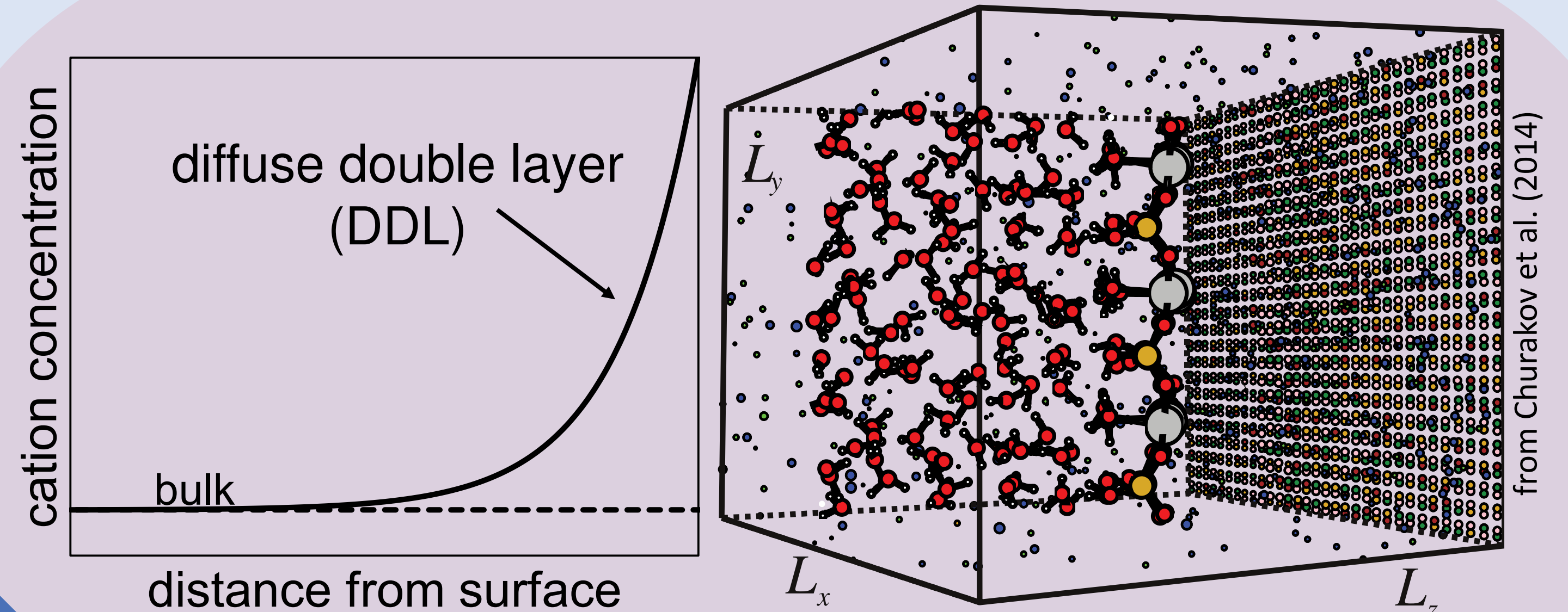
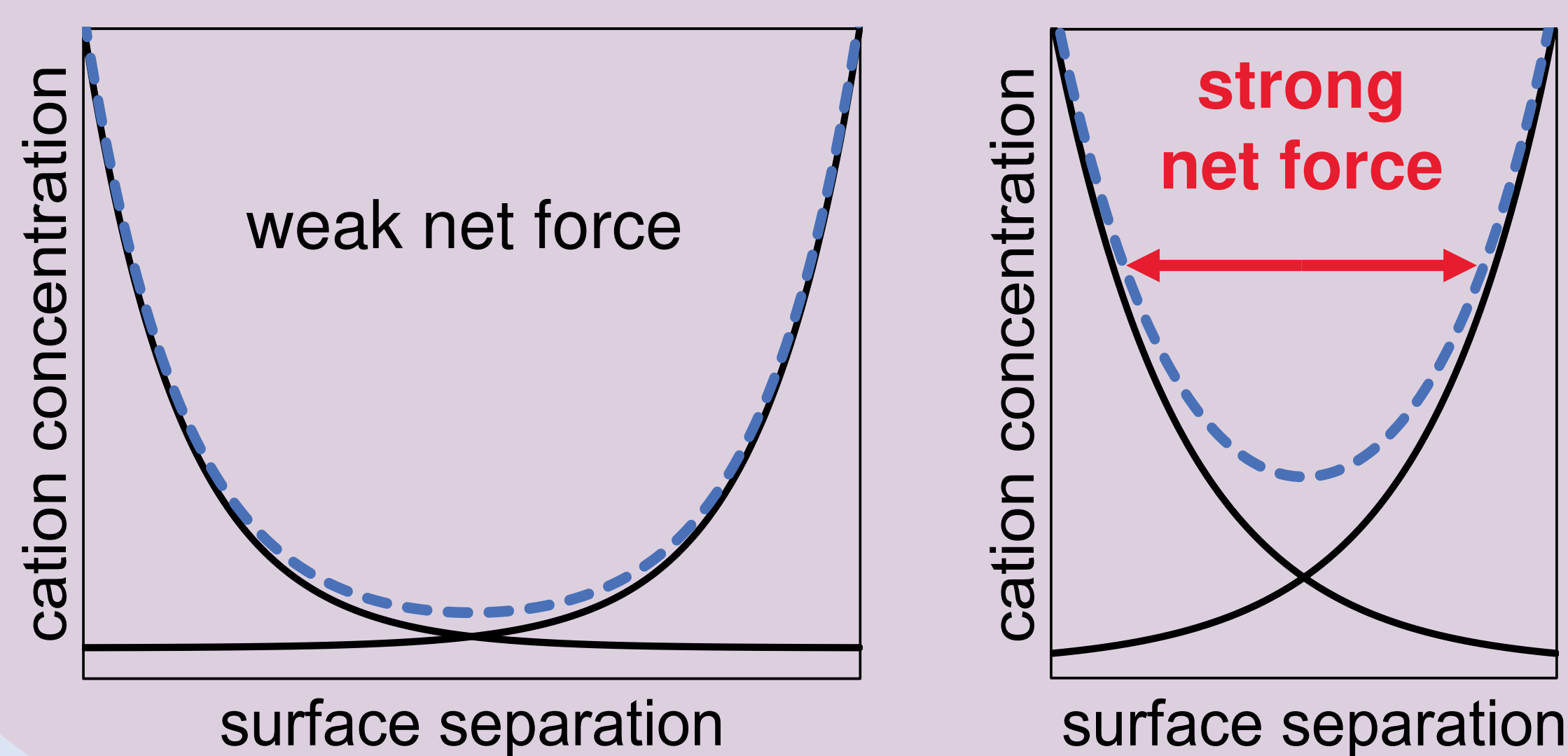
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Motivation

The alkali-silica reaction (ASR, «concrete cancer») is one of the most important concrete degradation mechanisms. The fundamental mechanisms leading to cracking upon ASR are still poorly understood.¹ Some studies suggest that crystallisation pressure is the main driving force, while others suggest swelling pressure. What is the underlying mechanism leading to the pressure build-up?



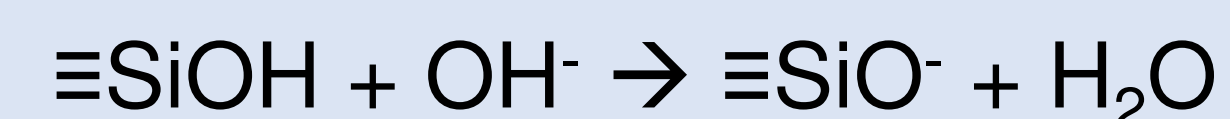
ASR products grow inside pores of an aggregate and the separation between mineral surfaces decreases. DDLs of two neighboring crystal surfaces start to overlap. According to Poisson-Boltzmann (PB) theory, a repulsive force between the crystal surfaces is generated if DDLs overlap. If the force exceeds a certain threshold value, the aggregate will crack.



Surfaces of cement particles and ASR products are negatively charged. Cations in the pore solution adsorb at the surface and form a diffuse double layer (DDL). The lateral extent of the double layer depends on the surface charge and the ionic strength of the solution.

Methods

We use grand canonical Monte-Carlo (GCMC) simulations to investigate interaction between parallel charged planes in different electrolytes in wide range of pH and plane separations. Planes are represented by lattice of point sites, each point is a surface $\equiv\text{SiOH}$ group of the mineral shlykovite (a proxy for ASR product). Surface groups can deprotonate:



At high pH, negative surface charge density is higher.² Since PB theory neglects atomic scale ion-ion correlations, it always predicts repulsive forces between charged surfaces. However, the neglected correlations can result in attractive interaction and a net attractive force between charged surfaces in some cases.³ The modelling approach applied in this study takes these interactions into account.

Summary & outlook

We showed that mono- and divalent cations have very different effects on the forces between surfaces of ASR products.

Three further questions remain open and are addressed in current and subsequent simulations:

- What pressures are expected for realistic pore water, i.e. mix of mono- and divalent electrolyte?
- What are the differences in osmotic pressures in ASR product and C-S-H, the main hydrate phase in cement?
- How realistic are predictions of cracking of aggregate due to modelled pressures at meso- and macroscale?

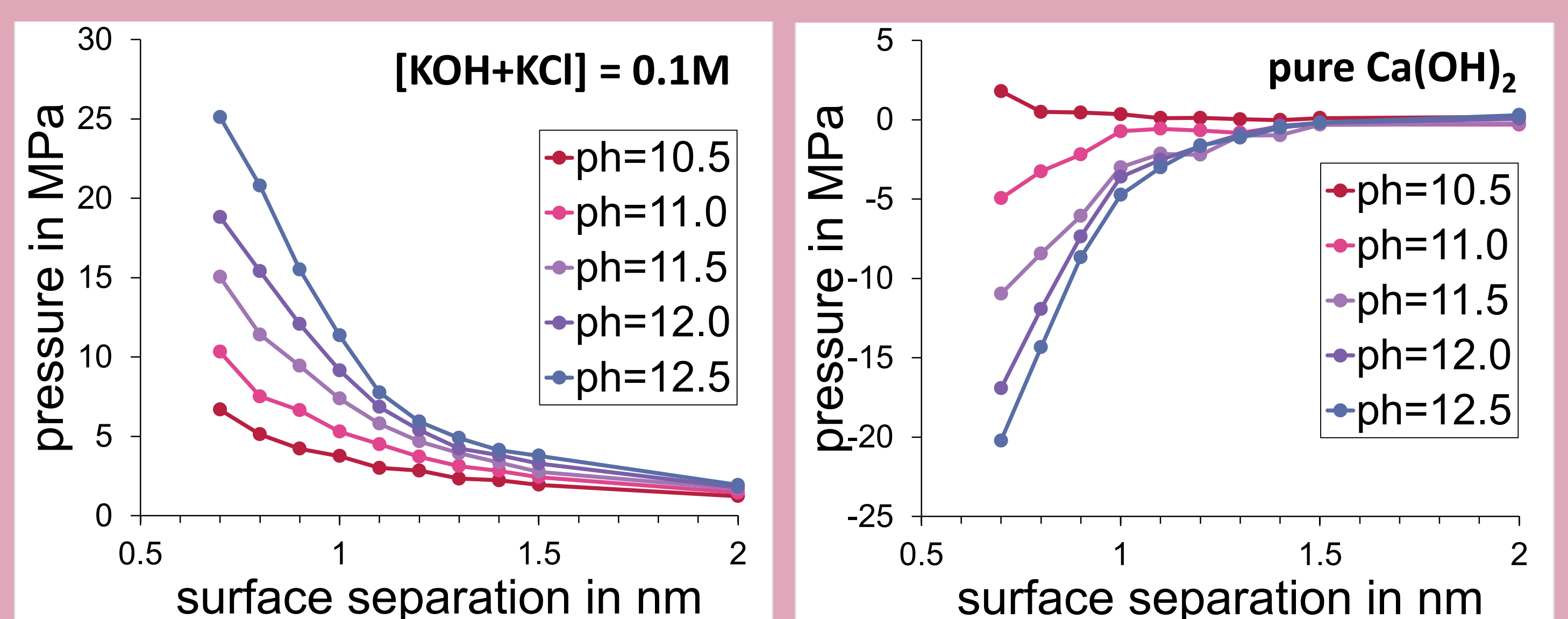
Acknowledgements

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References:

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Results & discussion



- **1:1 electrolytes (KOH) lead to repulsive forces**, as predicted by PB theory. With increasing pH, surfaces become more negative and more cations are adsorbed. This leads to stronger overall and repulsion forces.
- **Attractive forces dominate for 2:1 electrolytes (Ca(OH)₂)**. This explains why some colloidal systems can generate strong cohesion, e.g. calcium-silicate hydrates (C-S-H) in cement. Attraction becomes stronger with increasing pH.

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