Kinetic aspects of major and trace elements in olivines from variably cooled basaltic melts

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1- Introduction

Olivine is a common mineral phase in basaltic systems and represents an important information source about the origin, crystallization and differentiation of primitive magmas.

We have experimentally investigated the effects of both cooling rate and undercooling (-ΔT) on the textures and compositions of olivine crystals crystallizing in a tholeiitic melt.

We observe the formation of a diffusive boundary layer (chemical gradient) in the melt close to the crystal boundary. This layer is controlled by i) the growth rate of the crystal and ii) diffusivity of cations in the melt. The chemistry of the olivine crystals is in turn controlled by the chemical composition of the boundary layer itself, and by the partitioning of major and trace cations at the crystal-melt interface.

Furthermore, our results provide new constraints on the -ΔT threshold at which the crystal morphology shifts from interface-controlled (polyhedral shape) to diffusion-controlled (skeletal/dendritic shapes).

Our results introduce new important information for the comprehension and modelling of the transport, ascent, and emplacement mechanisms of magmas.

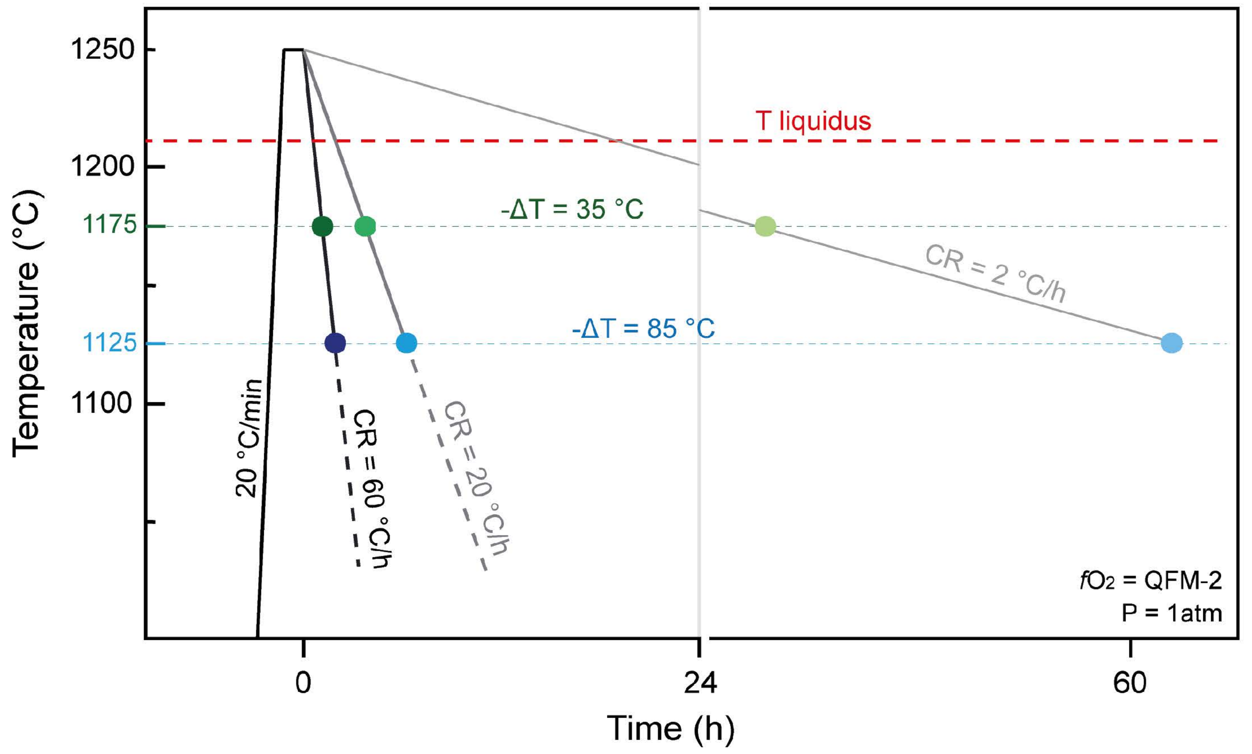
2- Methodology

Experimental method:

- Starting material: 'Golden pumice', undoped primitive Kilauea tholeiite

(Shea et al., 2019)

- Sample holder: Pt-loops of 2 µm diameter



Analytical method:

Transect olivine/glass:
- Jeol-JXA8200EDS-WDS combined electron microprobe.
- Elements analyzed: Fe, Mg, Si, Ca, Mn, Ni, Ti, Cr, P, Al, Na, K, Co
- Olivine: 20 keV, 300 nA
- Glass: 20 keV, 20 nA
- 2 µm spot size, step of 2 µm

Chemical maps:
- Mg, Fe, Ca, Al, P → microprobe (15 keV, 7.5 nA)
- Mg, Fe, Ca, Al, Mn, Ni, Co → SEM (25 keV, 2.6 nA)

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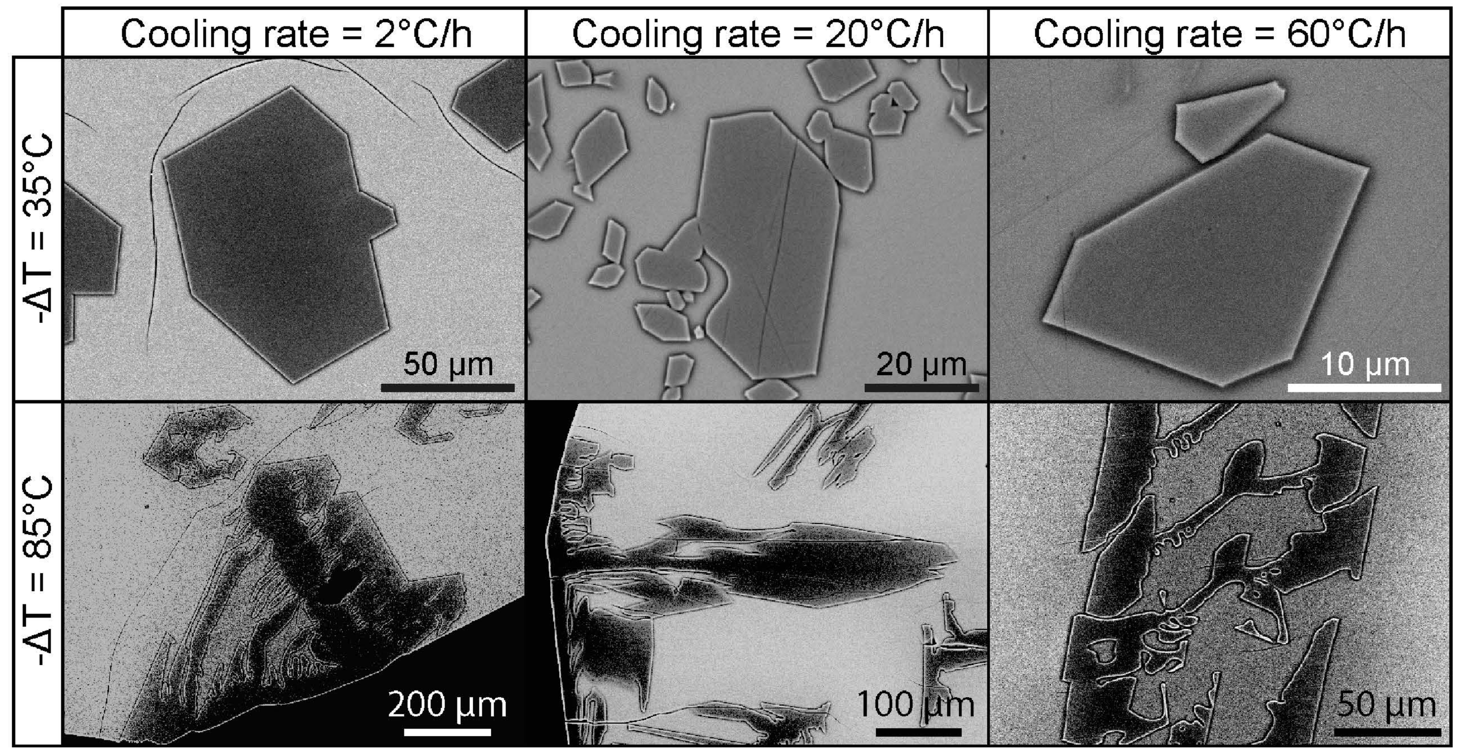
3- Textures



-ΔT= 35 °C:
→ polyhedral shape only
CR= 2 °C/h: 60-100 µm diameter
CR= 20 °C/h: 40-60 µm diameter
CR= 60 °C/h: 10-20 µm diameter

-ΔT= 85 °C:
→ dendritic shape only
Crystal size: 100-1000 µm

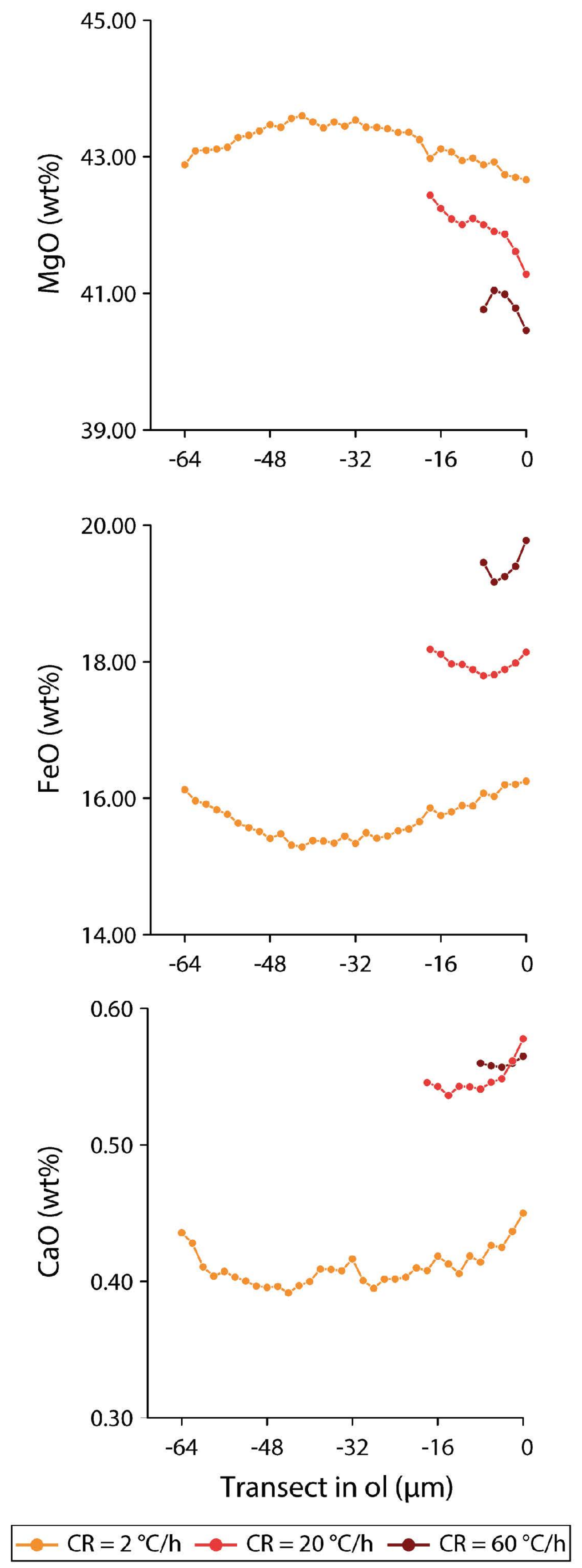
wt% cristalization (-ΔT= 35 °C)
(using the average of olivine and glass composition)

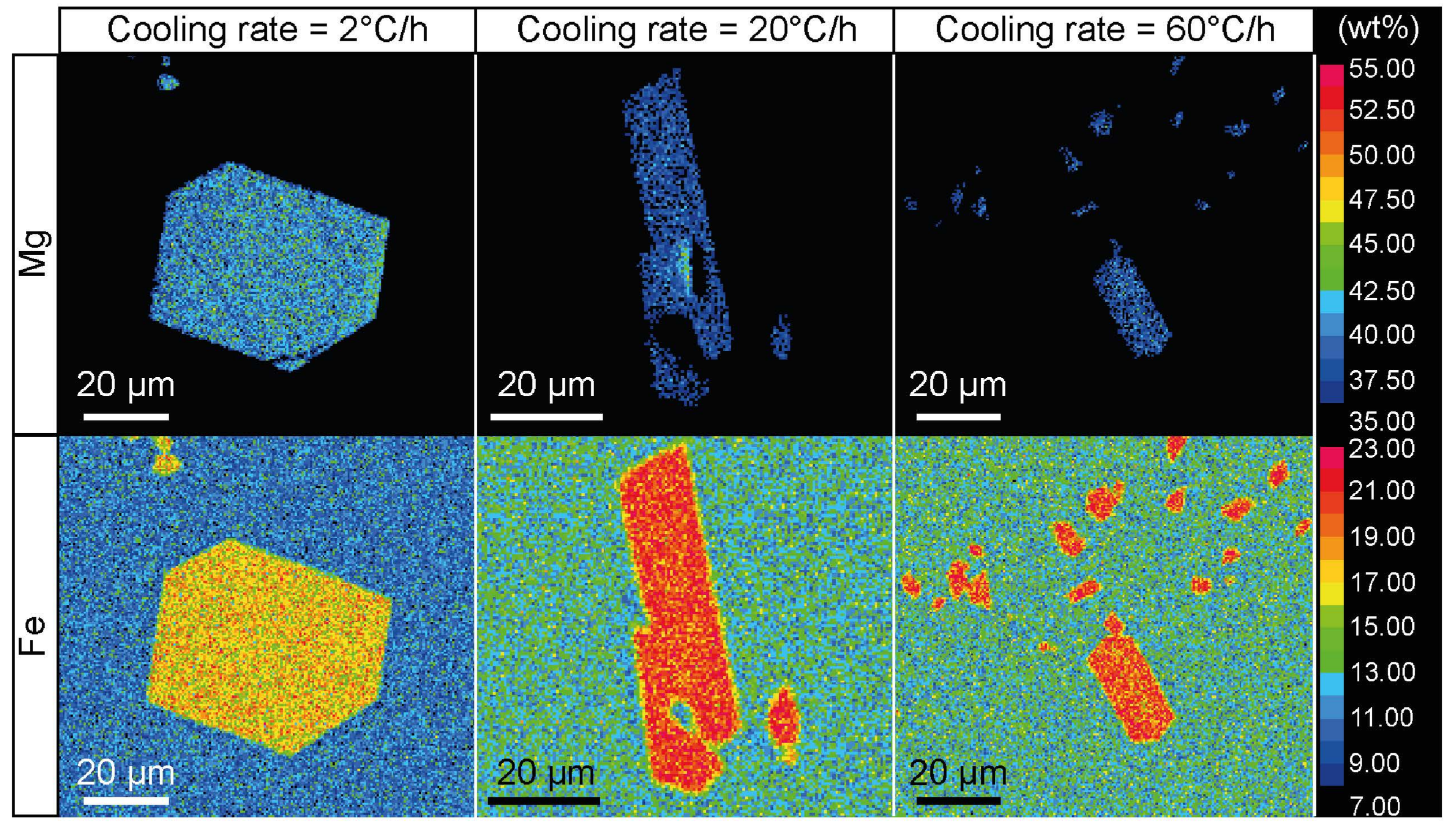
$$X_{ol,\ Mean\ (\%)} = \sum \left(\frac{C_{MgO}^{initial} - C_{MgO}^{final}}{C_{MgO}^{ol} - C_{MgO}^{final}} \times 100 \right) / N$$

(Mourey and Shea 2019)

CR= 2° C/h	CR= 20 °C/h	CR= 60 °C/h
6.8 wt%	7.1 wt%	7.3 wt%

4- Olivine composition (-ΔT= 35 °C)





Chemical maps

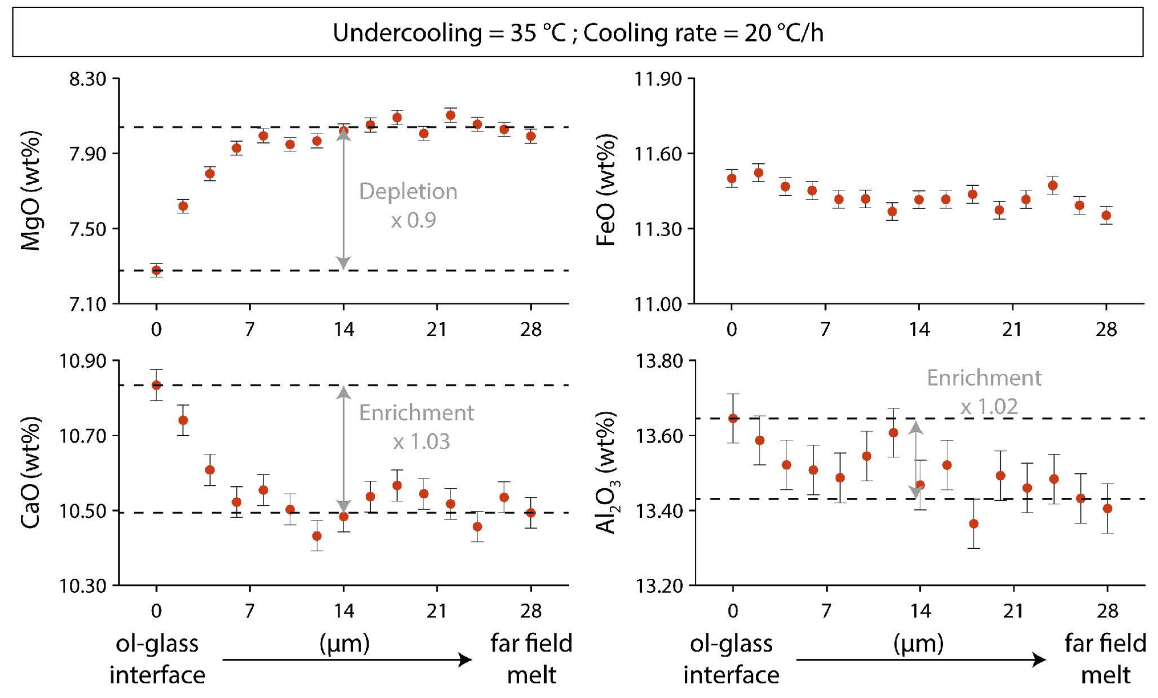
From low to high cooling rate:
- Olivine MgO content decreases from 43.2 to 40.8 wt%
- Olivine FeO content increases from 15.7 to 19.4 wt%

Mg#	CR= 2 °C/h	CR= 20 °C/h	CR= 60 °C/h
Core	83.6	82.6	80.8
Rim	80.2	79.2	78.5

Chemical transect

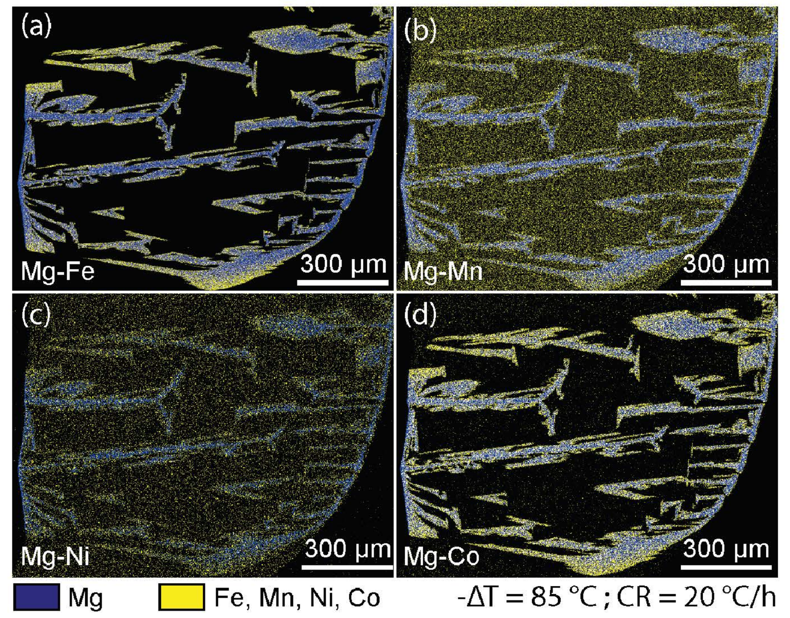
→ Core to rim zonation:
- Core enriched in MgO / depleted in FeO
- Rim depleated in MgO / enriched in FeO
- CaO increases with the degree of cooling rate
- CaO is preferentially incorporated at the rim

5- Glass composition (-ΔT= 35 °C)

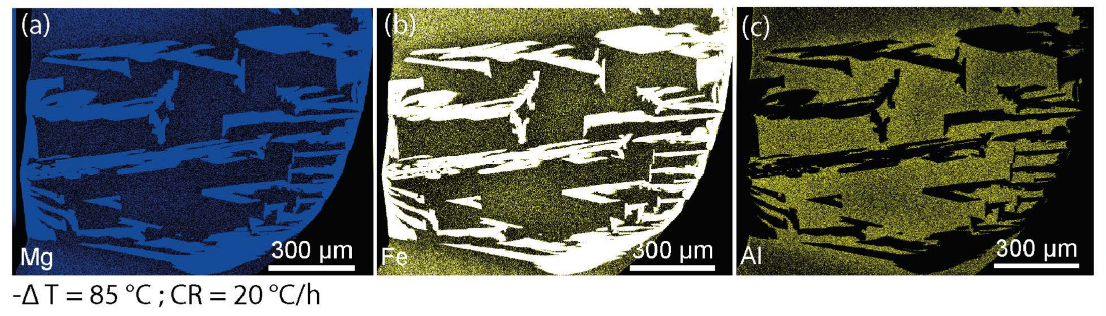


- Boundary layer: ~10-12 µm
- MgO depletion registered at the interface points to slow diffusivity of Mg in the melt with respect to the apparent crystal growth rate:
  - CR= 2 °C/h → 6.29×10^{-10} ms^{-1}
  - CR= 20 °C/h → 1.29×10^{-9} ms^{-1}
  - CR= 60 °C/h → 2.08×10^{-9} ms^{-1}
- CaO and Al_2O_3 are enriched in the boundary layer by up to x1.03 and x1.02 respectively.
  → these element are less favourably incorporated within the crystal, their concentrations in the melt progressively increases with increasing cooling rate.

6- Olivine zoning and melt gradients at -ΔT= 85 °C



→ Mn, Ni, Co are mainly enriched in the rim rather than the core, as for the case of Fe.



- Thickness of the boundary layer: ~ 28-57 µm
- MgO and FeO → depletion
- Al_2O_3 → enrichment
- Depletion of FeO is characteristic of -ΔT= 85 °C
  → This suggests that the far field melt does not have enough time to supply fresh Fe cations to the advancing crystal interface.

7- Summary

- Dendritic crystals → -ΔT = 85 °C
  Polyhedral shapes → -ΔT = 35 °C.
- The crystal size decreases with increasing cooling rate.
- Mg-content in olivine decreases at increasing cooling rates, conversely, Fe and Ca increase.
- The crystal core is mainly enriched in Mg, while the crystal rim is enriched in Fe and transition elements.
- The boundary layer is enriched in Ca and Al, but is depleted in Mg.
- Fe is affected by -ΔT. At low -ΔT, Fe-diffusivity is enough to readily supply Fe cations; at high -ΔT, the crystal growth rate is faster than the diffusion of Fe in the melt.

References:

Shea, T., Hammer, J.E., Hellebrand, E. et al. Phosphorus and aluminum zoning in olivine: contrasting behavior of two nominally incompatible trace elements. Contrib Mineral Petrol 174, 85 (2019). https://doi.org/10.1007/s00410-019-1618-y

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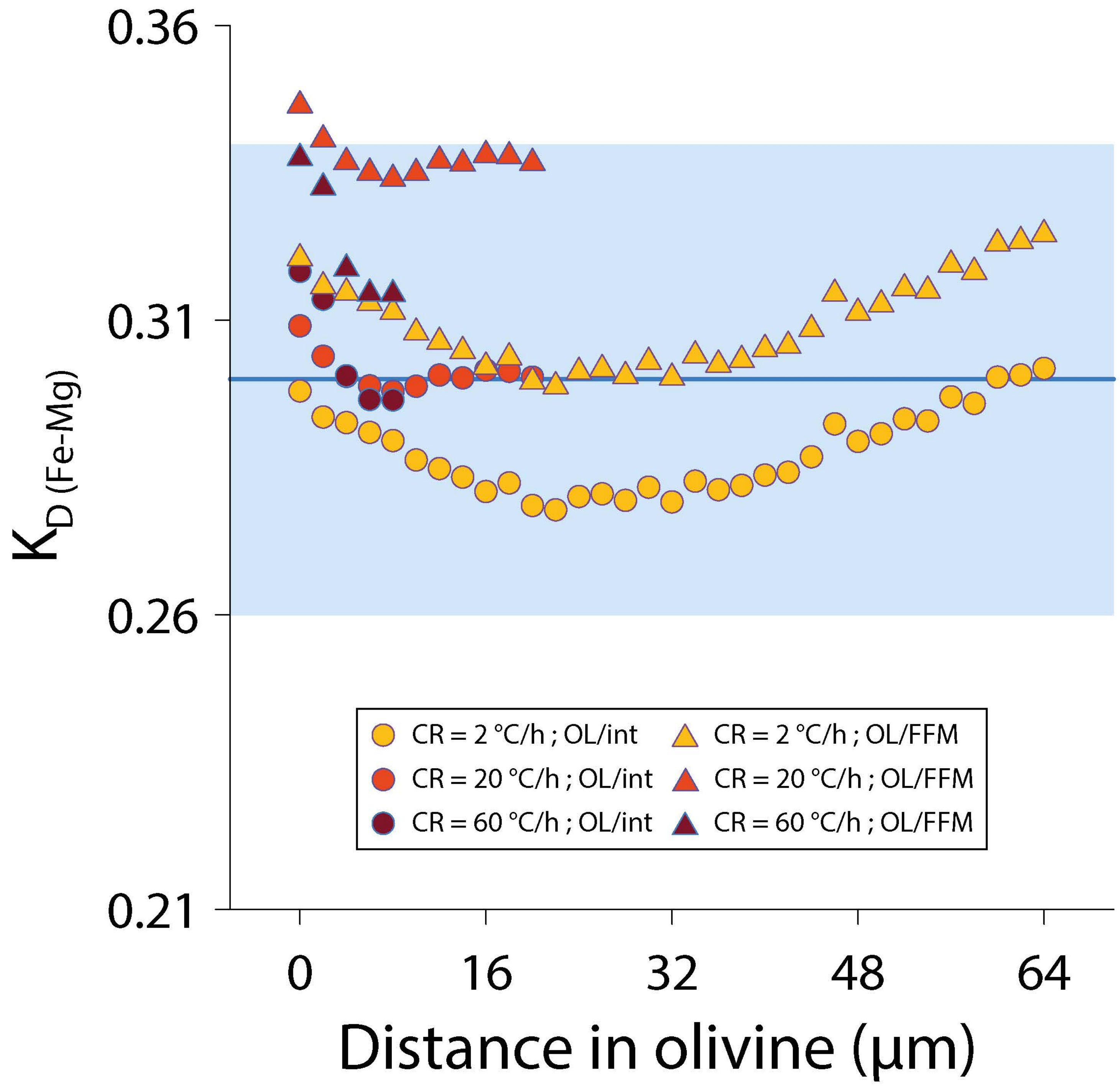

Rim to rim Fe-Mg exchange in olivine crystals

- Circle = $K_{D(Fe\text{-}Mg)}$ calculated with the ***olivine composition*** and the composition of the ***melt next to the crystal rim***.

- Triangle = $K_{D(Fe\text{-}Mg)}$ calculated with the ***olivine composition*** and the composition of the ***far field melt***.

- Yellow = experiment at -ΔT= 35 °C and CR= 2 °C/h
- Light red = experiment at -ΔT= 35 °C and CR= 20 °C/h
- Dark red = experiment at -ΔT= 35 °C and CR= 60 °C/h

- Blue area = equilibrium range (0.30 ± 0.04)
- Blue line = equilibrium (0.30)

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